<table>
<thead>
<tr>
<th>Items</th>
<th>( {\text{U}} )</th>
<th>( {\text{U}}^{235} )</th>
<th>Pu</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Beginning inventory</strong></td>
<td>4,575,091</td>
<td>117,680</td>
<td>404</td>
</tr>
<tr>
<td><strong>Increase during the term concerned</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Receipt</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Production of Pu</td>
<td>-</td>
<td>-</td>
<td>1,463</td>
</tr>
<tr>
<td>Other increases</td>
<td>62</td>
<td>15</td>
<td>16</td>
</tr>
<tr>
<td>Adjustment (for rounding off)</td>
<td>0</td>
<td>0</td>
<td>1</td>
</tr>
<tr>
<td><strong>Total</strong></td>
<td>4,575,153</td>
<td>117,695</td>
<td>1,884</td>
</tr>
<tr>
<td><strong>Decrease during the term concerned</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Shipment</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Nuclear loss</td>
<td>4,851</td>
<td>3,970</td>
<td>0</td>
</tr>
<tr>
<td>Disposal as waste</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Accidental loss</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Other decreases</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td><strong>Term-end inventory</strong></td>
<td>4,570,309</td>
<td>113,726</td>
<td>1,884</td>
</tr>
<tr>
<td>Adjustment (for rounding off)</td>
<td>0</td>
<td>-1</td>
<td>0</td>
</tr>
<tr>
<td><strong>Total</strong></td>
<td>4,575,160</td>
<td>117,695</td>
<td>1,884</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Detail of term-end inventory ( {\text{U}}^{235} )</th>
<th><strong>Loading</strong></th>
<th><strong>New</strong></th>
<th><strong>Storage</strong></th>
<th><strong>Spent</strong></th>
</tr>
</thead>
<tbody>
<tr>
<td>( 4,166,130 ) (103,262)</td>
<td>230,154 (5,972)</td>
<td>174,025 (4,491)</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>( {\text{U}}^{235} )</th>
<th><strong>Month</strong></th>
<th><strong>Charge</strong></th>
<th><strong>Discharge</strong></th>
<th><strong>Nuclear loss</strong></th>
<th><strong>Burn-up (MWd)</strong></th>
<th><strong>Pu-production</strong></th>
</tr>
</thead>
<tbody>
<tr>
<td>January</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>300 (247)</td>
<td>195</td>
<td>94</td>
</tr>
<tr>
<td>February</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>306 (232)</td>
<td>202</td>
<td>95</td>
</tr>
<tr>
<td>March</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>632 (519)</td>
<td>425</td>
<td>196</td>
</tr>
<tr>
<td>April</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>878 (720)</td>
<td>610</td>
<td>268</td>
</tr>
<tr>
<td>May</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>1,116 (913)</td>
<td>812</td>
<td>335</td>
</tr>
<tr>
<td>June</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>1,610 (1,319)</td>
<td>1,223</td>
<td>475</td>
</tr>
<tr>
<td><strong>Total</strong></td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>4,851 (3,920)</td>
<td>3,467</td>
<td>1,463</td>
</tr>
</tbody>
</table>

Load at beginning of term: \( 4,166,130 \) (103,262) Load at end of term: \( 4,166,130 \) (103,262)
NUCLEAR MATERIALS MANAGEMENT
The following States are Members of the International Atomic Energy Agency:

AFGHANISTAN  FEDERAL REPUBLIC OF GERMANY  NIGERIA
ALBANIA  GABON  NORWAY
ALGERIA  GHANA  PAKISTAN
ARGENTINA  GREECE  PARAGUAY
AUSTRALIA  GUATEMALA  PERU
AUSTRIA  HAITI  PHILIPPINES
BELGIUM  HOLY SEE  POLAND
BOLIVIA  HONDURAS  PORTUGAL
BRAZIL  HUNGARY  ROMANIA
BULGARIA  ICELAND  SAUDI ARABIA
BURMA  INDIA  SENEGAL
BYELORUSSIAN SOVIET SOCIALIST REPUBLIC  INDONESIA  SOUTH AFRICA
CAMBODIA  IRAN  SPAIN
CAMEROON  IRAQ  SUDAN
CANADA  ISRAEL  SWEDEN
CEYLON  ITALY  SWITZERLAND
CHILE  IVORY COAST  SYRIA
CHINA  JAPAN  THAILAND
COLOMBIA  KENYA  TUNISIA
CONGO, DEMOCRATIC  REPUBLIC OF KOREA  TURKEY
COSTA RICA  KUWAIT  UKRAINIAN SOVIET SOCIALIST REPUBLIC
CUBA  LEBANON  UNION OF SOVIET SOCIALIST REPUBLICS
CYPRUS  LIBERIA  UNITED ARAB REPUBLIC
CZECHOSLOVAK SOCIALIST REPUBLIC  LIBYA  UNITED KINGDOM OF GREAT BRITAIN AND NORTHERN IRELAND
DENMARK  LUXEMBOURG  UNITED STATES OF AMERICA
DOMINICAN REPUBLIC  MADAGASCAR  URUGUAY
ECUADOR  MALI  VENEZUELA
EL SALVADOR  MEXICO  VIET-NAM
ETHIOPIA  MONACO  YUGOSLAVIA
FINLAND  NETHERLANDS
FRANCE  NEW ZEALAND

The Agency's Statute was approved on 23 October 1956 by the Conference on the Statute of the IAEA held at United Nations Headquarters, New York; it entered into force on 29 July 1957. The Headquarters of the Agency are situated in Vienna. Its principal objective is "to accelerate and enlarge the contribution of atomic energy to peace, health and prosperity throughout the world".

Printed by the IAEA in Austria
February 1966
Symposium on Nuclear Materials Management, Vienna, 30 August - 3 September 1965. Proceedings ... held by the International Atomic Energy Agency ... Vienna, the Agency, 1966. 888 p. (IAEA, Proceedings series)

621.039.54:658.78
658.78:621.039.54

NUCLEAR MATERIALS MANAGEMENT
IAEA, VIENNA, 1966
STI/PUB/110
FOREWORD

An increasing number of countries are using nuclear materials which, because of their high value and the potential hazards involved, require special methods of handling. To discuss these and to provide a forum at which different systems for achieving the necessary economy and safety could be compared, the International Atomic Energy Agency held a Symposium at Vienna on Nuclear Materials Management from 30 August to 3 September, 1965. It was attended by 115 participants from 19 Member States and two international organizations.

Nuclear materials are already being used on an industrial scale and their high cost demands close and continuous control to ensure that they are delivered precisely on time and that they are used to the fullest possible extent before they are withdrawn from service. Routine industrial methods of material control and verification are widely used to ensure safe and economical operation and handling in nuclear power stations, in fuel-element fabrication and reprocessing plants, and in storage facilities. In addition special refinements are needed to take account of the value and the degree of purity required of nuclear materials. Quality as well as quantity has to be checked thoroughly and the utmost economy in processing is necessary.

The radioactivity of the material poses special problems of handling and storage and creates a potential hazard to health. A further problem is that of criticality. These dangers and the means of averting them are well understood, as is evidenced by the outstandingly good safety record of the atomic energy industry. But besides accommodating all these special problems, day-to-day procedures must be simple enough to fit in with industrial conditions.

Many of the 58 papers presented at the Symposium emphasized that records, checks, measurements and handling precautions, if suitably devised, provide the control vital to efficient operation, serve as checks against loss or waste of valuable materials and help meet the demands for safety. The topics discussed included material control systems, recording and reporting techniques, sampling, analysis and physical measurement, determination of production and consumption of fissile materials in reactors and various considerations of safety and economic management.
EDITORIAL NOTE

The papers and discussions incorporated in the proceedings published by the International Atomic Energy Agency are edited by the Agency's editorial staff to the extent considered necessary for the reader's assistance. The views expressed and the general style adopted remain, however, the responsibility of the named authors or participants.

For the sake of speed of publication the present Proceedings have been printed by composition typing and photo-offset lithography. Within the limitations imposed by this method, every effort has been made to maintain a high editorial standard; in particular, the units and symbols employed are to the fullest practicable extent those standardized or recommended by the competent international scientific bodies.

The affiliations of authors are those given at the time of nomination.

The use in these Proceedings of particular designations of countries or territories does not imply any judgement by the Agency as to the legal status of such countries or territories, of their authorities and institutions or of the delimitation of their boundaries.

The mention of specific companies or of their products or brand-names does not imply any endorsement or recommendation on the part of the International Atomic Energy Agency.
CONTENTS

MATERIAL CONTROL SYSTEMS (Sessions I, II and III)

Contrôle des matières dans les centrales nucléaires et les installations de recherche italiennes (SM-67/1) ..................... 3
E. Santarelli (Italy)

Recherche d'une amélioration de la centralisation des données comptables, notamment par l'emploi de calculatrices électroniques (SM-67/18) ...................... 19
F. Ronteix (France)
Discussion ................................................................. 30

Criteria for special nuclear materials inventory and control procedures (SM-67/19) .................................................. 31
E. M. Kinderman and R. R. Tarrice (United States of America)

Case study in internal audit of nuclear material (SM-67/27) ........ 43
S. Kops (United States of America)
Discussion ................................................................. 49

Organisation de la comptabilité des matières de base dans une usine de fabrication d'éléments combustibles (SM-67/9) ........... 51
J. Nadal et J. Pinochet (France)

Materials control in the fabrication of enriched uranium fuels (SM-67/25) ............................................................... 65
R. G. Cardwell Jr. (United States of America)
Discussion ................................................................. 86

A nuclear materials management control system for uranium reactor fuel element fabrication (a hypothetical case) (SM-67/28) ......................... 89
L. D. Hazelton (United States of America)
Discussion ................................................................. 121

Management and control of EBR-II fuel (SM-67/21) ................ 123
G. K. Whitham, T. R. Spalding and M. J. Feldman (United States of America)

Special nuclear material control by the power reactor operator (SM-67/40) ............................................................... 141
R. A. Cordin (United States of America)
Discussion ................................................................. 151

Professional nuclear materials management (SM-67/60) .......... 153
A. A. Forcella and W. J. O'Leary (United States of America)

Contrôles possibles sur la base du bilan matières (SM-67/11) ...... 167
A. Gilles (France)
Discussion ................................................................. 175
Gestion des matières nucléaires à l'atelier de traitement
de l'uranothorianite de l'usine du Bouchet (SM-67/13) .................. 177

C. Lorrain (France)

Nuclear materials management in a recovery facility for
unirradiated enriched uranium (SM-67/26) ................................. 189

G.R. Jasny (United States of America)

Discussion .................................................................................. 201

The system for controlling source and special nuclear material
at the Eurochemic reprocessing plant (SM-67/3) ............................. 205

W. Frenzel and W. Schüller (Belgium)

Nuclear materials management at the Japan Atomic Energy
Research Institute (SM-67/4) .......................................................... 213

C. Machida and I. Hiramatsu (Japan)

The management of nuclear materials in a research
establishment (SM-67/56) ............................................................... 229

W.J. Wright and D. R. Hocking (Australia)

Discussion .................................................................................. 235

Accounting systems for heavy water and fissionable
materials (SM-67/57) ................................................................. 237

G.W. Fletcher, H.B. Reid and W.G. Jenkinson (Canada)

Discussion .................................................................................. 257

Determination of the uranium content in scrap (SM-67/5) ............... 259

K. Nakajima and N. Saito (Japan)

A method of identification and inspection for inventory control
of irradiated fuel elements (SM-67/20) ........................................... 275

E.M. Kinderman and J.S. Mills (United States of America)

Discussion .................................................................................. 292

Materials control system for enriched uranium chemical
processing facility for irradiated nuclear materials
(SM-67/23) ............................................................................... 293

F.H. Tingey and B.S. Lewis (United States of America)

Discussion .................................................................................. 311

RECORDING, REPORTING AND GENERATION OF QUANTITATIVE
DATA (Session IV)

Méthodes d'inventaire dans un établissement de transformation
(SM-67/14) ............................................................................... 315

G. Billy (France)

Discussion .................................................................................. 323

Plant measurement, sampling and analysis for accountancy
purposes with particular reference to separation plants
at Windscale (SM-67/53) ............................................................. 325

A.S. Davidson, F. Elliott, R. Powell and K.A. Swinburn
(United Kingdom)
Sampling uranium hexafluoride (SM-67/31) .......................... 341
W. D. McCluen (United States of America)

Opérations d'échantillonnage entre fournisseurs et clients
(SM-67/8) ........................................ 353
J. Artaud, R. Bodu et C. Lorrain (France)
Discussion ....................................... 373

Uranium ore and concentrate sampling (SM-67/30) .......................... 375
F. E. McGinley, D. L. Brown and R. W. Langridge
(United States of America)
Discussion ....................................... 391

Generation of quantitative data while sampling ores and
concentrates (SM-67/39) .................................. 393
W. J. Shelley and W. A. Ziegler (United States of America)
Discussion ....................................... 411

Método de muestreo doble para el control de calidad de un
proceso de fabricación (SM-67/61) .................................. 413
J. A. Cerrolaza, A. Lago y Rosa M. Montojo (Spain)

A multi-scaler recording system and its application to radiometric
"off-line" analysis (SM-67/49) .................................. 433
H. Bisby (United Kingdom)

EVALUATION OF MEASUREMENT METHODS;
NUCLEAR SAFETY AND CRITICALITY CONTROL (Session V)

Handling and measurement of plutonium shipments (SM-67/29) ............ 447
J. T. Byrne (United States of America)
Discussion ....................................... 462

Statistical control of measurement quality (SM-67/35) .......................... 465
C. A. Bennett (United States of America)
Discussion ....................................... 480

La gestion des matières nucléaires dans ses rapports avec la
sécurité et le contrôle de la criticité; étude des systèmes
à sécurité non inhérente (SM-67/15) .................................. 481
Y. Fruchard (France)
Discussion ....................................... 489

Nuclear materials management and its relationship to
nuclear safety and criticality control (SM-67/22) .................................. 491
F. H. Tingey (United States of America)

Application of reactor physics calculation methods to
criticality control of non-inherently safe systems
in fuel processing plants (SM-67/43) .................................. 499
M. Räber (United States of America)
Discussion ....................................... 514

Обеспечение ядерной безопасности при работе
с делящимися веществами (SM-67/62) .................................. 515
A. V. Kamaev, F. M. Kuznetsov, G. M. Vladimirov и
E. G. Dubovskii (CCCP)
ECONOMIC CONSIDERATIONS, GOVERNMENT ACTIVITIES
(Session VI)

The economic basis for establishing the number of measurements according to the value of the material (SM-67/2) .................................................. 527
H. H. Ph. Moeken and H. Bokelund (Belgium)

Economic considerations in the measurement of nuclear materials (SM-67/24) .......................................................... 537
F. H. Tingey (United States of America)

Economic criteria applied to nuclear materials management (SM-67/38) ................................................................. 547
W. J. Shelley and M. N. Kuehn (United States of America)

Discussion .............................................................................. 557

Gestion économique d'un centre de traitement de résidus radioactifs (SM-67/58) ................................................................. 559
J. M. Van Lint (Belgium)

Discussion .............................................................................. 568

Aspects économiques comparés du traitement et du stockage des produits de fission issus des usines de traitement de l'uranium naturel irradié (SM-67/17) .................................................. 571
L. Thiriet, P. Lesur, R. Giraud et J. Wanlin (France)

Technical support activities of a nuclear materials management programme (SM-67/37) ......................................................... 585
S. C. T. McDowell (United States of America)

Administrative co-ordination of fissile material management and accounting in the UKAEA (SM-67/55) ......................................................... 597
St. C. C. Hood (United Kingdom)

Discussion .............................................................................. 611

The United States Atomic Energy Commission programme of nuclear materials management (SM-67/59) ......................................................... 613
D. E. George (United States of America)

Discussion .............................................................................. 625

CHEMICAL AND ISOTOPIC ANALYSIS (Session VII)

Méthodes d'analyse du plutonium (SM-67/10) ................................................. 629
F. Regnaud (France)

Analytical methods for uranium concentration measurements (SM-67/32) ................................................................. 639
J. P. Highfill and C. J. Rodden (United States of America)

Discussion .............................................................................. 656

Methods for the determination of plutonium in spent reactor fuels, plutonium metal, alloys and compounds (SM-67/33) ........................................... 657
C. F. Metz and G. R. Waterbury (United States of America)

Discussion .............................................................................. 676
Practical gamma counting of unirradiated uranium-235

(PM-67/47) 

P. Colsmann (United States of America)

Discussion

An appraisal of analytical methods for plutonium and their applications to the analysis of nuclear materials (SM-67/51) 

G.W.C. Milner and G. Phillips (United Kingdom)

Discussion

Méthodes utilisées pour l'analyse isotopique de l'uranium et du plutonium (SM-67/16) 

R. Bir, J. Chenouard et Monique Lucas (France)

Isotopic measurement of uranium by mass spectrometry

(SM-67/34) 

G.F. Kaufman and C.D. Tabor (United States of America)

The evolution of the accuracy of isotopic analysis by thermal ionization from 2% to 0.02% (SM-67/41) 

W. R. Shields (United States of America)

Discussion

BURN-UP AND PRODUCTION (Sessions VIII and IX)

Plutonium production and burn-up calculation methods for light-water moderated reactors (SM-67/42) 

G.A. Sofer, A.H. Kazi, J.R. Tomonto and P. Buck (United States of America)

Discussion

Investigation of methods of calculating heavy-isotope content of operating fuel elements (SM-67/44) 

M.F. Valerino and Z.R. Rosztoczy (United States of America)

Discussion

Burn-up calculations for the Brookhaven graphite research reactor fuel elements (SM-67/36) 

P. Colsmann (United States of America)

Determination of plutonium-239 content in nuclear fuel by neutron transmission technique (SM-67/6) 

F. Putz, T. Al-Khafaji and P. Weinzierl (Austria)

Discussion

Operating experience with the semiconductor gamma-Compton spectrometer to determine the burn-up and burn-up history of nuclear fuels (SM-67/7) 

M.J. Higatsberger, H. Hick, K. Rumpold, P. Weinzierl and A. Burtscher (Austria)

Discussion

The non-destructive measurement of burn-up by gamma-ray spectroscopy (SM-67/45) 

N.C. Rasmussen, J.A. Sovka and S.A. Mayman (United States of America)

Discussion
Non-destructive methods for determining burn-up in
nuclear fuel (SM-67/16) ........................................ 851
W. J. McGonnagle (United States of America)
The non-destructive examination of irradiated nuclear fuel
for burn-up by gamma spectrometry with mechanical scanning
(SM-67/50) ............................................................ 867
A. J. Fudge, E. Foster and L. Murphy (United Kingdom)
Discussion .......................................................... 878

Chairmen of Sessions and Secretariat of the Symposium .......... 879
List of Participants ................................................ 880
Author Index ...................................................... 887
MATERIAL CONTROL SYSTEMS
(Sessions I, II and III)
CONTROL OF MATERIALS IN ITALIAN NUCLEAR POWER STATIONS AND RESEARCH INSTITUTIONS.

The paper deals with the management of nuclear materials at a facility with a gas-graphite power reactor using natural uranium, at another with a boiling-water reactor using enriched uranium, at an industrial installation with a research reactor and at a nuclear research laboratory.

For each of these installations the control of radioactive materials during fabrication and transport operations, while the materials are being used in reactors or in laboratories, and at the time when the receiver takes delivery of the materials is discussed.

The first part of the paper deals, from the management point of view, with all the recording operations and the methods employed for the internal auditing of data on losses, production and transfer of nuclear materials.

The second part assesses the shortcomings of the systems at present used in Italy and the problems of official materials management and control services, insofar as they affect safety.
reactor de agua hirviente y uranio enriquecido, en un establecimiento industrial con un reactor de investigación y, por último, en un laboratorio de investigaciones nucleares.

Por cada una de estas instalaciones el autor estudia la cuestión del control de los materiales radiactivos durante las operaciones de fabricación y transporte, en el momento en que llegan al establecimiento destinario y durante su empleo en los reactores y los laboratorios.

La primera parte de la memoria trata de los aspectos administrativos de las operaciones de registro y de los métodos empleados para la verificación interna de los datos relativos a las pérdidas, la producción y las transferencias de los materiales nucleares.

La segunda parte se refiere a las deficiencias de los sistemas adoptados en Italia y a los problemas que los servicios oficiales de control administrativo de los materiales tienen planteados desde el punto de vista de la seguridad.

1. INTRODUCTION

Selon la loi italienne n° 1860, art. 3, du 31 décembre 1962, il incombe à tous les exploitants de faire la déclaration de leur entrée en possession de matières fissiles spéciales et de matières brutes à l'administration compétente (Ministère de l'industrie), dans les cinq jours à compter de la réception du matériel dans l'établissement ou au lieu de stockage. Selon la même loi, tout détenteur des matières ci-dessus est tenu de présenter une mise à jour annuelle (au 31 décembre de chaque année) de la comptabilité des matières. Tous les contrôles concernant ces matières sont dévolus au CNEN (Comitato Nazionale per l'Energia Nucleare).

Une réglementation (prévue par le décret n° 185, art. 30, du 13 février 1964) est actuellement en cours d'élaboration, pour définir complètement les modalités de déclaration et de tenue des comptes. En attendant que ce règlement établisse des normes définitives, les services techniques et d'inspection de sûreté du CNEN ont procédé aux contrôles préliminaires et provisoires de toutes les matières nucléaires importées ainsi que de tout le matériel en cours de fabrication ou produit en Italie.

Le présent mémoire a pour but d'exposer certains des contrôles effectués dans deux grandes centrales nucléaires de puissance, une installation dotée d'un réacteur de recherche et un centre d'études et d'expériences. Pour chacune de ces installations, on décrit les systèmes de contrôle et d'enregistrement ainsi que les inconvenients éventuels.

On mentionne enfin les normes italiennes actuellement en cours d'élaboration concernant la comptabilité des matières et les contrôles.

2. PROBLEMES DE CONTROLE DES MATIERES NUCLEAIRES

Le contrôle des matières nucléaires soulève de nombreux problèmes de nature politique, technique, économique, et de sûreté et de protection sanitaire. Les problèmes politiques et économiques présentent, à leur tour, des aspects importants au niveau international aussi bien que des exigences de caractère national pour tout pays qui possède ou utilise de telles matières, de quelque façon que ce soit.

Le présent mémoire ne traite pas les problèmes qui se posent sur le plan international et se limite à décrire certaines enquêtes préliminaires
effectuées à l'échelle nationale, en Italie, en tenant compte essentiellement de la législation fondamentale existante. Toutefois, comme des conventions et des normes sont à l'étude tant sur le plan international que dans les organismes italiens compétents, il nous semble opportun que les autorités compétentes fixent d'abord les buts et les limites des organismes internationaux et les procédures de contrôle, ces normes et conventions dépendant naturellement des buts et limites fixés.

Si le but du contrôle est essentiellement d'empêcher le détournement de quantités dangereuses d'uranium ou de plutonium des utilisations pacifiques auxquelles ils sont destinés à l'origine, les procédures pourront être assez semblables aux méthodes adoptées pour le contrôle des matériaux conventionnels et aux systèmes fiscaux en vigueur pour le contrôle des importations, de la fabrication, de la vente et de l'utilisation des substances spéciales (pétrole, essence, alcool, etc.). Dans ce cas, les normes de contrôle seraient très onéreuses pour l'exploitant en ce sens qu'un grand nombre d'appareils de mesure et d'analyse et de moyens de vérification et d'essai adoptés dans les secteurs conventionnels devraient s'appliquer également aux activités nucléaires, ce qui exigerait des transformations et adaptations très coûteuses (automatiques, électriques, électroniques, etc.).

Si, au contraire, le but du contrôle est essentiellement de nature technico-économique, les frais de procédure dépendront du degré de précision désiré dans les constats et les vérifications de comptabilité. Dans ce cas, en effet, les données les plus importantes seront la consommation et la production de matières fissiles. Mais nous savons, d'autre part, que ces données, même si l'on adopte des méthodes de calcul complexes et laborieuses, comportent un facteur d'incertitude de l'ordre de 10% environ, ce qui enlève sa signification à tout contrôle de ce genre touchant les combustibles utilisés dans les grands réacteurs de puissance.

Par conséquent, il serait plus opportun de s'orienter vers un système de contrôle qui prévoit plutôt l'inaliénabilité du combustible au cours de son utilisation par l'exploitant, un contrôle quantitatif rigoureux des récipients servant au transport du combustible irradié et, également, la présence de l'organisme de contrôle (ou, à la rigueur, une comptabilité fiscale précise) au moment de l'introduction des matériaux dans le cycle de retraitement; ce n'est qu'au cours de cette phase, en effet, et grâce à des analyses après dissolution, que l'on peut obtenir des données valables sur la production nucléaire et sur les consommations de combustible.

3. CONTROLE DES MATIERES DANS LES CENTRALES DE PUISSANCE

3.1. Gestion des matières dans une centrale du type Latina

On examine ici la gestion du combustible nucléaire dans une centrale nucléaire de puissance, du type gaz-graphite, telle celle de Latina.

Le combustible est constitué par de l'uranium naturel, c'est-à-dire par de l'uranium-238 ayant une teneur en uranium-235 ne dépassant pas 0,72% en poids. Chaque élément de combustible est constitué par une barre cylindrique d'uranium recouverte d'une gaine de magnox (alliages légers à base de magnésium); la barre d'uranium a un diamètre de 30 mm environ,
une longueur de 925 mm et un poids moyen de 11,5 kg. L'élément de combustible complet (uranium plus magnox) a un diamètre de 100 mm environ et une longueur de 1023 mm.

En cours de fabrication, les éléments de combustible sont soumis à des contrôles rigoureux tels que:

a) analyse des échantillons de barres d'uranium, pour en vérifier la composition chimique;
b) contrôle des dimensions et de la distorsion éventuelle de la barre;
c) contrôle des défauts superficiels, effectué au moyen de liquides pénétrants fluorescents ou par le système des courants parasites;
d) vérification du matériau destiné à la fabrication des gaines de magnox;
e) contrôle des gaines après fabrication;
f) contrôle de la fabrication des bouchons d'obturation.

Une fois terminée la fabrication des barres d'uranium, des gaines et des bouchons d'obturation, on procède aux essais de porosité de chaque gaine avant l'introduction des barres de combustible. Ces essais sont effectués, en général, selon des méthodes fondées sur les gaz révélateurs. Les éléments de combustible achevés sont vérifiés à l'aide du spectromètre de masse.

Dans le cas étudié, le nombre d'éléments de combustible fournis à la centrale italienne pour sa première période d'activité a dépassé 35 000. Le cœur du réacteur de la centrale comprend plus de 2900 canaux et chaque canal contient huit éléments de combustible, c'est-à-dire qu'en cours de fonctionnement, le cœur du réacteur contient normalement environ 23 420 éléments de combustible à uranium naturel.

Le transport de ce nombre considérable d'éléments du pays producteur à l'installation d'utilisation s'est effectué exclusivement par voie ferrée. À leur arrivée, les caisses métalliques sont emmagasinées dans le dépôt de la centrale, à côté de la salle du réacteur. Ces caisses scellées ne sont ouvertes qu'au moment du chargement dans le réacteur.

Dans chaque caisse le fabricant introduit la liste des éléments qu'elle contient; une liste identique est adressée par poste à la direction de la centrale, au moment du transport. Chaque élément est identifié grâce à un numéro matricule gravé sur une des quatre ailettes externes verticales.

Pour l'enregistrement des divers éléments on utilise des «fiches combustible», qui sont compilées et tenues à jour au cours de l'utilisation de l'élément; sur la fiche on note, dans des colonnes appropriées et selon des codes précis, les mouvements de chaque élément, les irrégularités ou avaries enregistrées en cours d'activité, ainsi que d'autres données fondamentales d'utilisation.

On donne ci-dessous un exemple de code de mouvement du combustible:

Exemple de code de mouvement

01 Prise en charge de l'élément dans le dépôt
11 Manipulation du dépôt au réacteur
12 Positionnement dans le réacteur
13 Déplacement temporaire hors du canal
14 Manipulation d'un autre endroit au réacteur
21 Manipulation du réacteur au bac de désactivation  
22 Positionnement dans le bac  
31 Manipulation du dépôt à l'expédition  
32 Manipulation du réacteur à l'expédition  
33 Manipulation du bac à l'expédition  

e tc...  

Les avaries et les irrégularités enregistrées sont indiquées conformément au « code de garantie ».

Exemple de code de garantie  
A.1. Enregistrement d'un défaut avant le 90ème jour d'activité  
A.2. La température de la gaine a dépassé les limites de garantie  
A.3. La température de sortie du gaz a dépassé les limites de garantie  
A.4. La vitesse de variation de la température de la gaine a dépassé les limites de garantie  
A.5. Fréquence de changement de puissance excessive hors garantie  
A.6. Puissance de canal hors garantie  
A.7. Vitesse de variation de la puissance du réacteur hors garantie  
A.8. Humidité ou impuretés dans le gaz réfrigérant hors garantie...  

La « fiche combustible » porte aussi le numéro d'ordre du chargement dans le réacteur, tiré du registre d'activité de l'installation.  
On note enfin sur la fiche  
a) l'irradiation de l'élément combustible  
b) la zone du réacteur  
c) le canal du réacteur  
d) la position verticale dans le canal  
e) le mouvement de décharge dans le réacteur  
f) le récipient contenant le combustible irradié  
g) les données concernant les expéditions de renvoi.  

Le calcul de l'irradiation, de la consommation de l'uranium-235 et de la production nucléaire est assez compliqué en raison du grand nombre d'éléments en présence et des vastes dimensions du cœur. Ce calcul est néanmoins confié à un centre qui, périodiquement, élabore, enregistre et contrôle les données reçues.  

Les éléments déchargés d'un canal du réacteur sont transférés dans le bac de désactivation à l'aide d'un appareil approprié muni d'un système d'écrans. Etant donné leur forte radioactivité et les dommages qu'ils ont pu subir, il n'est pas possible de s'approcher des éléments irradiés pour en contrôler - par exemple - le numéro matricule. Sur le fond du bac de désactivation on dépose les containers métalliques ouverts, dont le numéro d'identification est visible à travers l'eau; dans ces containers on dépose mécaniquement jusqu'à 140 éléments, dont on découpe ensuite les ailettes externes. Le découpage des ailettes exclut toute possibilité ultérieure d'identifier les barres de combustible (le numéro matricule est, avons-nous précisé, gravé sur une des ailettes); d'autre part, les ailettes constituent
également des résidus actifs et doivent être enfouies dans des fosses appro-
priées à l'intérieur de l'établissement.
Après désactivation, les barres sont placées dans de plus grands con-
tainers (contenant jusqu'à 200 barres), puis enfermées dans des récipients
spéciaux pour être transportées au retraitement.

3.2. Gestion du combustible dans une installation de puissance dotée d'un
réacteur à eau bouillante

La gestion du combustible dans une installation dotée d'un réacteur à
eau bouillante, c'est-à-dire utilisant de l'uranium enrichi, se présente très
différemment. Le combustible utilisé par l'installation italienne à eau bouil-
lante actuellement en activité est constitué par de l'uranium sous forme
d'oxyde enrichi à 2% en uranium-235. Chaque élément est constitué par
81 barres d'uranium et contient au total environ 220 kg d'uranium-238 et
4,5 kg d'uranium-235. Pour la première période d'activité de la centrale,
l'approvisionnement a été de 230 éléments environ (208 pour le cœur du
réacteur, le reste comme stock).
On note l'énorme différence existant entre le nombre limité d'éléments
nécessaires à ce second type d'installation et les 20 000 ou 30 000 éléments
nécessaires dans une installation du type gaz-graphite à uranium naturel.
Il en découle, bien entendu, une différence substantielle dans la façon de
concevoir les systèmes de contrôle et de comptabilité.
L'élément de combustible se présente sous la forme d'un parallélépipède
à section quadrangulaire dont les parois sont en zircaloy (épaisseur 2 mm)
et en acier inoxydable (épaisseur 1,5 mm). À l'intérieur de cette enveloppe
sont disposées les 81 barres cylindriques d'uranium recouvertes d'une gaine
dezircaloy (épaisseur 0,75 mm, diamètre extérieur 13,5 mm).
La barre de combustible proprement dite est constituée par des pas-
tilles de bioxyde d'uranium d'environ 12 mm de diamètre et 25 mm de ha-
teur, empliées dans la gaine de zircaloy. Chaque barre a une longueur de
2,75 m environ (joint inclus); la longueur totale de l'élément est supérieure
t à 3,20 m. L'élément de combustible forme donc un véritable ensemble, un
tout de dimensions considérables et facile à identifier dans chaque phase
de son utilisation.
Les principaux contrôles de fabrication effectués sur les pastilles d'ura-
nium sont:
a) échantillonnage du matériau (UO₂) en poudre et analyse chimique;
b) essais de frittage sur échantillons pour chaque lot de matériau en poudre
(contrôle de la densité minimale après frittage);
c) vérification de la composition chimique des pastilles frittées, sur des
échantillons prélevés tous les deux lots;
d) contrôle de la composition isotopique (enrichissement) des pastilles
frittées, sur des échantillons prélevés tous les deux lots;
e) contrôle de la réactivité minimale et maximale du combustible;
f) contrôle de la densité du matériau fritté;
g) contrôle visuel et contrôle des dimensions des pastilles.
Les tubes de zircaloy sont soumis aux contrôles suivants:
a) contrôle externe et contrôle des dimensions;
b) contrôles par ultrasons;
c) essai sous pression hydraulique.

En outre, sur des échantillons représentant en général deux lots de tubes, on procède aux vérifications suivantes:
a) analyse de la composition chimique du matériau;
b) vérification des caractéristiques mécaniques;
c) essai d'éclatement;
d) vérification de la résistance à la corrosion;
e) contrôle métallographique.

Après introduction du combustible dans les gaines, chaque barre est soumise aux contrôles suivants:
a) contrôle externe et contrôle du poids;
b) essai de porosité, avec hélium.

En outre on effectue des contrôles radiographiques et de dimensions sur des échantillons représentant deux lots ou plus.

Enfin, une fois achevée la fabrication des éléments contenant chacun 81 barres de combustible, on procède sur chaque élément à un essai de porosité; les vérifications finales en usine consistent en des contrôles de dimensions et une inspection externe avant l'emballage et l'expédition.

A leur arrivée à la centrale, les éléments combustibles sont stockés dans un dépôt spécial qui, dans le cas cité, est un local isolé par des parois en béton armé dont l'unique accès est une porte en acier résistant au feu. Les éléments sont déposés, au moyen de crochets, dans des râteliers qui garantissent un espacement sûr entre les éléments. Des précautions particulières sont prises pour éviter la criticité au cours du déplacement et de l'emmagasinage du combustible. Les éléments de combustibles doivent être en permanence, soit dans les containers, soit dans les râteliers, soit fixés aux crochets spéciaux. Ces instruments de stockage sont conçus de telle façon que la criticité ne saurait en aucun cas être atteinte. Pour éviter, par ailleurs, que la criticité ne soit atteinte accidentellement, au cas où interviendrait une géométrie non contrôlée, il n'est admis que deux éléments à la fois dans une zone donnée, compte tenu de ce que la masse de deux éléments est toujours inférieure au tiers de la masse critique.

Une enquête sur les possibilités d'inondation de l'installation par débordement des fleuves situés à proximité de la centrale a montré que le dénivellement entre les niveaux maximaux des crues de ces fleuves et les cotes des locaux affectés au dépôt et des salles de manipulation des combustibles exclut toute possibilité d'inondation des aires de stockage et de manipulation du combustible. Ces aires ne sont pas munies d'installations hydrauliques.

On a également effectué une enquête sur les risques d'incendie pouvant affecter les mêmes zones et on a conclu que, même en cas d'incendie grave et de non-fonctionnement des moyens d'extinction, la criticité ne saurait être atteinte et qu'il n'y aurait pas de risque d'irradiation. Les plus hautes températures pouvant être atteintes ne sauraient provoquer de modifications considérables de la géométrie sous-critique d'emmagasinage, ni faire fondre les pastilles de bioxyde d'uranium en quantité notable.

On a enfin procédé à des calculs du facteur de multiplication infini en cas d'incidents exceptionnels susceptibles de déterminer une géométrie non contrôlée des éléments ou d'annuler l'effet d'empoisonnement des parois
des râteliers (celles-ci sont en effet construites en tôle d'acier contenant du carbone, et plaquées au cadmium); on a conclu que, même dans ces cas-là, et compte tenu de l'absence d'un modérateur, il ne pourra jamais se produire d'incidents dus à la criticité, quels que soient le nombre et la disposition géométrique des éléments à l'intérieur des dépôts.

Chaque élément est désigné par un numéro matricule composé d'une lettre (A) et d'un numéro de fabrication progressif (A.1...A.208). A l'enregistrement des matériaux on mentionne les zones suivantes de l'installation:

a) dépôt de combustible frais;
b) cœur du réacteur;
c) piscine de stockage du combustible irradié.

Après vérification de l'intégrité de chaque élément à la suite du transport, le combustible est introduit dans le cœur et son positionnement est mentionné sur le registre d'activité du réacteur et sur un graphique approprié (section horizontale du cœur). Les variations éventuelles de la géométrie du cœur, l'évacuation des combustibles irradiés vers la piscine et leur substitution figurent également sur les documents.

Provisoirement et en attendant le règlement prévu, la direction de la centrale transmet chaque mois aux autorités compétentes un état des matières fissiles spéciales disponibles où la quantité totale d'uranium est répartie entre ses isotopes 235 et 238 (système EURATOM); à cet état est annexé un inventaire des éléments combustibles où figurent les numéros matricules de tous les éléments, répartis entre les trois zones mentionnées (dépôt frais, réacteur, piscine). Le poids de l'uranium et de ses isotopes n'est pas indiqué sur l'inventaire.

La centrale est dotée d'un certain nombre de chaînes de chambres d'ionisation pour la mesure du flux neutronique dans le réacteur; chaque chambre contient environ 0,04 g d'uranium-235; dans le cas cité la quantité totale d'uranium-235 contenue dans ces instruments ne dépasse pas 5 g. Toutefois, étant donné que, tant dans le système EURATOM que dans le système italien, les matières fissiles spéciales doivent être déclarées et comptabilisées quelle que soit leur quantité, l'exploitant doit ajouter (sur l'état mensuel) ces quantités insignifiantes aux 50 tonnes environ d'uranium enrichi à 2% qui constituent le combustible disponible.

La composition isotopique initiale du matériau figure sur les états mensuels jusqu'au jour où ces éléments sont envoyés au retraitement; en conséquence, ni la consommation d'uranium-235, ni la production nucléaire de plutonium ne sont communiquées en cours d'activité.

Etant donné le nombre limité d'éléments faisant l'objet de la gestion des matières d'une centrale du type étudié, il est possible d'établir une «fiche historique» pour chaque élément utilisé. Sur cette fiche, on note les périodes d'irradiation de l'élément en relation avec son positionnement dans le cœur, les consommations partielles correspondantes d'uranium-235 et la formation de plutonium. Il n'est pas difficile, au moment de l'expédition du combustible irradié, de reconstituer les phases d'utilisation et de calculer avec une certaine précision les variations de poids de l'uranium-235 et la production de plutonium.
3.3. Contrôle des matières dans une installation dotée d'un réacteur de recherche

La comptabilité des matières, dans le cas d'installations dotées d'un réacteur de recherche, est plus complexe que celle qui concerne les installations dotées d'un réacteur de puissance.

En effet, tandis que, dans une centrale de puissance, les matières fissiles spéciales et brutes ne sont constituées en général que par du combustible et du matériel (d'un poids toujours insignifiant) faisant partie des instruments ou sources, dans le cas d'un réacteur de recherche on a en outre diverses quantités de matières utilisées comme échantillons à irradier et servant à des études ou à des expériences. On peut même avoir une production de radioéléments ou de radioisotopes artificiels qui, naturellement, exigent une comptabilité et des contrôles séparés.

Dans le cas étudié, l'installation est constituée par un réacteur à piscine utilisant des éléments à plaques du type MTR, d'une puissance de 5 à 7 MW.

Pour une certaine période d'activité, la charge de combustible a consisté en deux lots d'éléments, l'un enrichi à 20%, l'autre à 90% en uranium-235. On sait que l'élément de type MTR est constitué par un groupe de plaques en alliage uranium-aluminium; dans le cas étudié, les plaques ont 7,6 cm environ de large, 62,5 cm de haut et une épaisseur de l'ordre de 1,3 mm. L'élément entier a environ 87 cm de long. La première charge a consisté en 30 éléments normaux environ, 10 éléments de contrôle et quelques éléments partiels. Les éléments normaux du lot enrichi à 20% comportaient 12 plaques d'alliage combustible, tandis que les éléments partiels comprenaient quelques plaques inertes constituées seulement d'aluminium. Les éléments de contrôle n'avaient que six plaques du fait de l'existence de la cavité centrale réservée à la barre de contrôle. Les éléments étaient de fabrication américaine.

En cours de fabrication, de nombreux contrôles sont effectués, entre autres:

a) l'échantillonnage et l'analyse des lingots;
b) le contrôle de l'uranium (poids et teneur);
c) le contrôle radiographique des plaques finies;
d) le contrôle de soufflure et formation de bulles;
e) le contrôle par ultrasons;
f) le contrôle des dimensions.

Le transport du pays producteur à l'installation d'utilisation est effectué par voie aérienne; des containers métalliques sont utilisés.

La comptabilité de gestion des éléments combustibles, compte tenu de leur nombre limité, a été conçue de façon relativement simple, par enregistrement des mouvements du magasin au réacteur et de celui-ci au dépôt des éléments irradiés. Le positionnement des divers éléments figure sur le registre d'activité du réacteur.

On a veillé particulièrement au calcul de l'irradiation de chaque élément, de la consommation d'uranium-235 et de la production de plutonium; cela, afin de pouvoir obtenir - après la phase initiale d'activité - une con-
SANTARELLI

sommation uniforme d'uranium-235 dans les divers éléments. Pour évaluer la consommation de matériau fissile, on a tenu compte:

a) de la distribution du flux neutronique pour chaque géométrie du cœur;
b) des données et des conditions de fonctionnement du réacteur;
c) de la position des barres de contrôle;
d) de la présence de réflecteurs et de modérateurs;
e) de la puissance thermique.

Une fois connue l'énergie produite par chaque géométrie, on en déduit l'énergie moyenne par élément. Étant donné que cette énergie moyenne n'aurait de signification que dans le cas d'une distribution uniforme du flux neutronique thermique (condition qui ne se vérifie pas dans la pratique), on a cherché une grandeur empirique (facteur de l'élément) proportionnelle au flux thermique moyen de chaque élément de combustible.

A l'aide de méthodes de calcul appropriées, on a déterminé les valeurs du «facteur» pour chaque élément dans les diverses géométries du noyau du réacteur. Pour chaque élément et pour chaque configuration du noyau, le produit du «facteur» par l'énergie moyenne a donné la valeur de l'énergie produite par l'élément considéré. On a calculé enfin la consommation d'uranium-235 et évalué la production nucléaire. Ces résultats ont été portés séparément pour chaque élément sur un «fascicule historique» ou figurent les données suivantes:

Ième partie

1. Numéro d'identification de l'élément
2. Fabricant
3. Année de fabrication
4. Caractéristiques de construction
5. Teneur initiale
   - en uranium (teneur totale)
   - en uranium-235 (et/ou 233)
   - en uranium-238
   - en thorium
6. Installation (réacteur d'utilisation)
7. Date d'arrivée à l'installation
8. Provenance (fournisseur)
9. Mise en route de l'utilisation

IIème partie

10. Utilisation de l'élément:
    - transfert d'une installation à une autre
    - transformations et modifications
    - utilisation pour des études et expériences
    - utilisation pour des mesures
    - avaries et ruptures, etc.
Illème partie

11. Résumé de l'utilisation (combustible irradié)
12. Etat final:
   - périodes d'irradiation
   - énergie totale
   - consommation d'uranium-235
   - production de plutonium
   - autres matières radioactives.

Un tel document – que le responsable de l'installation a rédigé pour chacun des éléments de combustible – s'est révélé particulièrement utile lorsque, au terme de leur utilisation dans le réacteur, les éléments constituant la première charge ont été transférés temporairement dans une installation de stockage avant d'être retraités.

Le contrôle de la comptabilité de ces combustibles a été pleinement satisfaisant, en ce sens que le système d'enregistrement adopté par l'utilisateur aussi bien que les méthodes de calcul des consommations et des productions se sont révélés conformes aux normes fondamentales actuellement en vigueur en Italie dans ce domaine.

En ce qui concerne les autres matières nucléaires disponibles pour des études, des expériences ou des travaux divers, la comptabilité de l'installation couvre actuellement quatre lots séparés de matériaux d'uranium naturel en plaques, d'uranium enrichi à 20% sous forme de pastilles en alliage aluminium, d'uranium métallique appauvri et de pellicules de $\text{U}_3\text{O}_8$ dans des compteurs à chambre de fission. On n'a pas encore fixé le système d'enregistrement et de contrôle de ces matières; pour l'instant, on a adopté la méthode d'enregistrement prescrite par l'EURATOM.

3.4. Gestion des matières dans un grand centre d'études nucléaires

Il peut être intéressant d'exposer brièvement l'organisation – sur la base d'un règlement – de la gestion des matières spéciales au Centre d'études nucléaires de la Casaccia.

La direction du Centre a estimé opportun (en raison d'exigences internes) d'étendre l'appellation «matières spéciales» à des matières non fissiles, tout en appliquant à ces dernières tous les contrôles prescrits par les traités internationaux et par la législation italienne.

Le règlement de gestion intéresse donc les quatre groupes suivants:
Groupe I : matières fissile spéciales
Groupe II : matières brutes
Groupe III: autres matières radioactives
Groupe IV: matières non actives, d'intérêt nucléaire.

Dans le groupe I figurent l'uranium-233, l'uranium-235, l'uranium enrichi, le plutonium. Dans le groupe II,l'uranium naturel et appauvri, le thorium. Dans le groupe III, tous les autres matériaux radioactifs prévus par la liste de la CEEA. Dans le groupe IV, les matériaux mentionnés par la direction du Centre, tels que l'eau lourde, le béryllium, etc.
L'organisation de la gestion des matériaux du Centre comprend
- un bureau central (bureau de coordination),
- les laboratoires qui détiennent les matières.

La première partie du règlement prévoit des normes générales pour
l'approvisionnement et le transport du matériel arrivant au Centre.

Un premier contrôle est effectué - sur la base des documents de trans-
port - à l'entrée du Centre, où l'on prend note de la date d'arrivée et des
caractéristiques principales du lot de matières.

Les matières faisant partie des groupes I, II et IV sont dirigées, dans
leur emballage, vers le dépôt du laboratoire intéressé, à l'intérieur même
du Centre; elles sont alors prises en charge par le personnel qualifié. Les
matériaux faisant partie du groupe III - qui sont, en général, des radio-
éléments ou des radioisotopes incorporés à des sources - passent par le
laboratoire de contrôle du Service de physique sanitaire, avant d'être re-
mis aux laboratoires intéressés.

Il incombe au directeur de laboratoire de transmettre à temps voulu
au bureau de coordination toutes les données nécessaires pour permettre à
la direction du Centre d'établir
a) les déclaration prescrites par la loi italienne;
b) les communications prévues par le traité de l'EURATOM et les autres
conventions internationales.

Les déclarations aux autorités nationales sont actuellement établies sur
un formulaire expérimental. Le nombre considérable d'exemplaires pré-
parés pour les communications destinées à l'EURATOM est également va-
lable pour l'enregistrement du bureau de coordination et pour le laboratoire
qui détient les matières.

En ce qui concerne les matières des groupes III et IV, qui ne sont pas
assujetties au système de contrôle de la CEEA, la direction du Centre a
prévus des formulaires spéciaux d'enregistrement et de contrôle.

Toutes les matières des groupes I, III et IV disponibles dans un labora-
toire sont réparties en lots homogènes; un registre à fiches doit être ouvert
pour chacun de ces lots.

En ce qui concerne les matières radioactives du groupe III, on ne tient
pas de registre; le contrôle est effectué sur la base des fiches d'entrée com-
pilées par le service de physique sanitaire.

Pour ce qui est des combustibles nucléaires, le règlement précise, à
titre d'exemple, qu'au même lot homogène appartiennent les éléments qui
sont identiques en raison
- du type de fabrication;
- de la composition chimique et du titre;
- de l'état physique;
- du projet de construction et de fabrication;
- de l'enrichissement;
- du poids de matière fissile spéciale contenu.

Pour chaque élément de combustible, il est nécessaire d'établir une
fiche chronologique permettant le calcul de l'irradiation moyenne, de la
consommation du combustible, et de l'éventuelle production nucléaire. Le
règlement indique que les matériaux qui pénètrent dans un laboratoire du
Centre doivent être soumis à un contrôle pour vérifier, autant que possible,
s'ils répondent aux spécifications d'achat et de fourniture. Les responsables du laboratoire doivent garantir que les matières sont utilisées conformément aux normes de projet et de contrat, et dans le respect absolu des normes de sûreté et protection sanitaire.

Les laboratoires (qu'ils soient ou non dotés d'un réacteur) classés comme installations selon le système de sûreté de l'EURATOM, doivent procéder à la compilation des états et des inventaires mensuels sur les formulaires fournis par la CEEA. Le bureau de coordination recueille les renseignements mensuels fournis par tous les laboratoires du Centre et les transmet à l'EURATOM à Bruxelles.

Lorsque la charge active d'un lot homogène de matériel devient nulle, par suite de consommation ou de transfert, le laboratoire restitue le registre correspondant au bureau de coordination qui le conserve dans des archives spéciales du Centre.

Etant donné qu'un lot homogène de matériel assigné à un laboratoire peut, à l'intérieur même de celui-ci, être réparti entre divers dépôts, départements ou secteurs, on a prévu des fiches de répartition interne établies en double exemplaire, dont l'un reste au responsable du laboratoire et l'autre va au consignataire du matériel. Le règlement contient, en outre, des normes générales et des recommandations concernant l'enregistrement des consommations et des pertes normales de matériaux.

On a prévu en particulier la restitution au service de physique sanitaire des sources scellées devenues insuffisantes pour les besoins du laboratoire qui en a la charge. On a également prescrit l'inventaire des disponibilités des divers laboratoires en fin d'année.

Enfin le règlement prévoit certaines procédures concernant les matériaux appauvris et les résidus (actifs ou non actifs), afin que la direction du Centre puisse les diriger vers leur destination finale (retraitement ou élimination définitive). À leur sortie du Centre, les matériaux sont soumis à des contrôles appropriés dont les résultats sont enregistrés et adressés à l'EURATOM et aux autorités nationales.

C'est le bureau de coordination qui se charge d'obtenir les autorisations éventuellement nécessaires pour le transport de sortie et le déchargement.

4. CONCLUSIONS

Le caractère général et panoramique de ce mémoire ne permet pas d'entrer dans le détail des nombreux problèmes posés par la comptabilité et les contrôles techniques des matières. On peut dire toutefois que la brève expérience italienne dans ce domaine confirme certains principes généraux qu'il faudra retenir dans l'élaboration des normes nationales et internationales.

Le premier point, qui paraît évident, est que la comptabilité et le contrôle des combustibles nucléaires (qu'ils soient constitués par des matières fissiles spéciales ou par des matières brutes), doivent faire l'objet d'une réglementation spéciale, nettement séparée de celle qui est prévue pour les mêmes matières lorsqu'elles sont utilisées comme échantillons ou incorporées dans des instruments ou dans des sources scellées.
Chaque fois que cela est possible et opportun, l'élément de combustible (surtout s'il contient de l'uranium enrichi ou du plutonium) doit être considéré comme un tout; de ce fait, dès sa fabrication, il devrait être muni d'un numéro (ou sigle) d'identité officiel, homologué par le service de contrôle compétent, et accompagné, lors de son transfert à l'installation d'utilisation et pour toute la durée de son utilisation, d'un document technique («fascicule historique») sur lequel le fabricant, les utilisateurs et les inspecteurs pourraient enregistrer les essais, les vérifications et contrôles, ainsi que les résultats des calculs concernant l'irradiation, la consommation et la production nucléaire éventuelle. En fin d'utilisation, le «fascicule historique» complet devrait être transmis à l'installation de retraitement, en même temps que l'élément correspondant.

Dans le cas de gros éléments destinés à des réacteurs de puissance, les contrôles officiels devraient commencer chez le fabricant; un représentant de l'organisme de contrôle pourrait assister, à la fabrique, à certains des contrôles essentiels en cours même de fabrication, ainsi qu'aux essais finaux. Dans le cas d'éléments petits et nombreux (comme ceux du réacteur à uranium naturel et ceux à plaques des réacteurs de recherche), les contrôles en fabrique devraient être effectués sur des éléments échantillons représentant des lots de 100 à 500 éléments.

Les éléments contrôlés et acceptés par le service compétent à la fabrique devraient être considérés comme inaliénables jusqu'à leur retraitement, c'est-à-dire que l'utilisateur ne saurait les endommager ou les modifier sans l'autorisation préalable du service de contrôle. Après utilisation il faudrait prévoir des contrôles officiels qualitatifs et quantitatifs dans l'installation de retraitement.

Pour les combustibles à uranium naturel destinés à des réacteurs de puissance, les normes d'un éventuel règlement devraient tenir compte de ce que:
- en général, une livraison est constituée par des milliers d'éléments, difficilement contrôlables après leur introduction dans le cœur du réacteur;
- le poids d'uranium présent dans le réacteur atteint des centaines de tonnes;
- la production de plutonium est, de ce fait, considérable.

Les combustibles à uranium naturel présentent, en outre, une caractéristique juridique particulière selon les lois italiennes en vigueur; en effet, avant d'être introduits dans le réacteur, ils sont considérés comme des «matières brutes ou d'origine», tandis que, après irradiation, ils deviennent des «matières fissiles spéciales» du fait qu'ils contiennent du plutonium.

Pour les combustibles à uranium enrichi, l'importance du contrôle effectué en fabrique sur le degré exact d'enrichissement est considérable, car il doit permettre de prévenir des incidents possibles de criticité au cours du transport et de l'emmagasinage dans la centrale nucléaire.

Pour les combustibles à plaques et, en particulier, pour ceux à plaques mobiles, c'est le contrôle en fabrique du laminage, des systèmes de traitement thermique et des méthodes destinées à détecter et à réparer les défauts superficiels ou la distribution non uniforme du «noyau» actif qui est important, car ce genre de défauts peut présenter des inconvénients considérables en cours de fonctionnement du réacteur de recherche, particulièrement dans le cas d'expériences de mesures physiques, ou autres.
Il faut ensuite établir des normes spéciales pour la comptabilité et pour le contrôle des combustibles irradiés; et cela, également, en raison des caractéristiques variables des lieux de dépôt et des moyens de transfert.

Si un ou plusieurs éléments de combustible sont destinés à être utilisés comme échantillons et à être soumis à des essais en vue d'études et d'expériences, leur destination devra figurer officiellement sur le «fascicule historique» et sur les certificats de fabrication; toute autre utilisation devra être autorisée par le service de contrôle compétent. Dans le cas des réacteurs de puissance, les inspecteurs devraient assister au chargement initial du combustible et à l'enregistrement correspondant. Les déplacements successifs ou les chargements partiels d'éléments dans le réacteur devraient apparaître sur le registre d'activité; les inspecteurs pourraient contrôler ce registre avec une certaine régularité (deux fois par an pour les réacteurs à uranium enrichi et une fois par mois pour les réacteurs à uranium naturel) et vérifier les quantités se trouvant dans les dépôts et les bacs de désactivation.

Il faut également résoudre, du point de vue des normes, le problème du contrôle visuel des combustibles irradiés; ce problème est assez grave dans le cas, par exemple, des milliers de petits éléments à uranium naturel qui finissent par s'amonceler dans les bacs de désactivation.

En second lieu, la comptabilité des matières fissiles spéciales et des matières brutes non destinées à être utilisées comme «combustibles» doit être simplifiée autant que possible. On atteindra plus facilement ce but si les normes fixées par les règlements tiennent compte des principes suivants:

a) les matières incorporées aux instruments ou sources ne peuvent être déplacées ou enlevées par l'utilisateur sans l'autorisation du service compétent;

b) les matières destinées aux études et expériences doivent être utilisées conformément à des programmes techniques précis, préalablement communiqués au service de contrôle; les changements éventuels doivent être approuvés par les autorités compétentes;

c) les matières acquises par une installation de fabrication doivent être travaillées selon des cycles et des méthodes approuvés et contrôlables; toute modification doit être préalablement approuvée par l'autorité compétente.

d) les matières acquises par une installation de retraitement du combustible doivent également être utilisées selon des cycles et des méthodes approuvés et contrôlables.

Tout changement apporté à l'installation et susceptible d'entraîner des modifications dans l'utilisation des matières doit être préalablement approuvé. L'expérience italienne a également démontré que la comptabilité et les contrôles doivent se référer à des lots «homogènes» de matériaux. La notion de lots homogènes, dans le cas des matériaux destinés aux réacteurs de puissance, est assez simple; en général, un réacteur de puissance utilise deux à trois types d'éléments ayant des caractéristiques de construction à peu près identiques et qui peuvent différer par l'enrichissement et par le poids d'uranium contenu (éléments spéciaux). On aura donc deux à trois lots homogènes de combustibles à enregistrer. Les chambres de fission ou, mieux encore, les matières qu'elles contiennent, constituent en elles-
mêmes des lots homogènes. Il pourra éventuellement y avoir d'autres lots: par exemple, les matières incorporées dans les sources, les échantillons, etc.

La notion de lots homogènes est moins précise pour les matériaux irradiés destinés au retraitement et pour les matériaux fissiles nécessaires à la fabrication des éléments de combustible. Dans ces cas-là, il semble plus approprié de parler de «lots d'utilisation» ou de «lots de destination» englobant les matériaux résultant du traitement ou contenus dans les produits finis.

On a noté enfin que la gestion des matières dans les centres d'études et de recherches nucléaires constitués par plusieurs laboratoires annexes exige une organisation interne capillaire, compte tenu des fréquents déplacements entre les divers services de chaque laboratoire et les transferts d'un laboratoire à l'autre.

Le problème qui reste maintenant à résoudre est celui de la compétence du service officiel de contrôle dans les divers déplacements à l'intérieur d'un laboratoire ou d'un centre et de l'opportunité de concentrer dans un bureau unique toute la comptabilité des matières se trouvant dans les divers laboratoires.
RECHERCHE D'UNE AMÉLIORATION DE LA CENTRALISATION DES DONNÉES COMPTABLES, NOTAMMENT PAR L'EMPLOI DE CALCULATRICES ELECTRONIQUES

F. RONTEIX
COMMISSARIAT A L'ÉNERGIE ATOMIQUE,
FONTENAY-AUX-ROSES, FRANCE

Abstract — Résumé — Аннотация — Resumen

IMPROVEMENT OF AUDITING DATA CENTRALIZATION, IN PARTICULAR BY USING ELECTRONIC COMPUTERS. Nuclear materials in France are distributed among numerous installations of all types: mines, factories, reactors, research centres, storage depots. One of the problems to be solved in the management of these materials is that of centralizing the auditing data produced by all these installations.

The first part of the paper gives a brief account of the French system of nuclear materials management and of the problem of auditing data centralization as it arises in this system. The main difficulty in recent years has been the time required for the monthly closing and centralization of accounts for all the installations. The steps taken to cope with these difficulties are described.

The second part describes the main method adopted, namely, the use of office machinery for centralization of data, the preparation of balances and the publication of a number of periodical documents. The paper explains why and under what conditions the use of office machinery was considered advantageous, and in particular it describes the coding system employed, the document circuits, and the time involved, mention also being made of the modifications which had to be made to the existing auditing system to meet the requirements imposed by the office machinery.

The third part deals with various conclusions, resulting from the introduction of machine accounting, in respect of the cost in manpower and money of this centralization of auditing data and, more generally, the cost of nuclear materials management.

RECHERCHE D'UNE AMÉLIORATION DE LA CENTRALISATION DES DONNÉES COMPTABLES, NOTAMMENT PAR L'EMPLOI DE CALCULATRICES ELECTRONIQUES. Les matières nucléaires sont réparties en France entre de nombreuses installations de toutes natures: mines, usines, réacteurs, centres de recherche, magasins de stockage. Un des problèmes à résoudre pour la gestion de ces matières est celui de la centralisation des données comptables émanant de toutes ces installations.

La première partie du mémoire consiste en un exposé rapide du système français de gestion des matières nucléaires et du problème de la centralisation des données comptables tel qu'il se pose dans ce système. La principale difficulté à laquelle on s'est heurté ces dernières années était celle des délais nécessaires pour arrêter manuellement les comptes de l'ensemble des installations et assurer leur regroupement. L'auteur décrit les mesures prises pour y remédier.

La deuxième partie a pour objet de décrire la principale mesure prise, qui est l'utilisation de la mécanographie pour assurer à la fois la centralisation des données, l'établissement des bilans et la publication d'un certain nombre de documents périodiques.

L'auteur explique pourquoi et dans quelles conditions on a estimé que l'utilisation de la mécanographie présenterait un intérêt: il décrit notamment le système de codification utilisé, les circuits de documents, les délais, en indiquant au passage les retouches qu'on a dû apporter au système de comptabilité existant pour répondre aux exigences de la mécanographie.

Enfin dans la troisième partie, l'auteur présente quelques conclusions et commentaires à l'occasion de la mise en œuvre de cette comptabilité mécanographique, sur le coût en hommes et en argent de cette centralisation des données comptables, et plus généralement sur le coût de la gestion des matières nucléaires.

УЛИЧЕНИЕ ЦЕНТРАЛИЗАЦИИ УЧЕТНЫХ ДАННЫХ, ОСОБЕННО НА ОСНОВЕ ПРИМЕНЕНИЯ ЭЛЕКТРОННЫХ ВЫЧИСЛИТЕЛЬНЫХ МАШИН. Во Франции ядерные материалы распределяются между многими различными установками на рудниках, заводах, реакторах,
в исследовательских центрах, хранилищах. Одной из проблем, которую надлежит решить в интересах обращения с этими материалами, является проблема централизации учетных данных, поступающих со всех этих установок.

В первой части дается краткое изложение французской системы обращения с ядерными материалами и соответствующей проблемы централизации учетных данных в том виде, как она ставится в этой системе. Основная трудность, с которой сталкивались в последние годы, заключалась в сроках, необходимых для ежемесячного составления отчетов по всем установкам и для обеспечения перерегистрации таких отчетов. Приводятся описание мер, принимаемых для исправления положения.

Вторая часть посвящена описанию основной принятой меры, которая заключается в использовании механографии для одновременного обеспечения централизации данных, составления балансов и публикации определенного количества периодических документов.

Говорится о том, почему и в каких условиях пришли к тому, что использование механографии представляет известный интерес. Дается описание, в частности, используемой системы кодификации, контуров документов, сроков. Одновременно упоминается на поправки, которые следует внести в существующую систему отчетности для удовлетворения потребностей механографии.

Наконец, в третьей части указывается на некоторые соображения, к которым пришли в связи с разработкой этой механографической отчетности в отношении стоимости сил и денежных расходов, связанных с такой централизацией учетных данных, и в более общей форме указывается стоимость обращения с ядерными материалами.

MEJORAMIENTO DE LA CENTRALIZACIÓN DE LOS DATOS CONTABLES, EN PARTICULAR MEDIANTE EL EMPLEO DE CALCULADORAS ELECTRONICAS. En Francia los materiales nucleares se hallan distribuidos entre un gran número de instalaciones de todas clases: minas, fábricas, reactores, centros de investigación y almacenes. Uno de los problemas que hay que resolver para la buena administración de estos materiales es el de la centralización de los datos contables procedentes de todas esas instalaciones.

En la primera parte de la memoria se expone sucintamente el sistema francés de administración de los materiales nucleares y el problema de la centralización de los datos contables que en él se plantea. La principal dificultad con que se tropezó durante los últimos años fue la de los plazos necesarios para cerrar mensualmente las cuentas del conjunto de las instalaciones y lograr su reagrupación. En la memoria se describen las medidas tomadas para remediarla.

En la segunda parte se describe la principal medida adoptada: el empleo de la mecanografía, que asegura a la vez la centralización de los datos, el establecimiento de los balances y la publicación de un cierto número de documentos periódicos.

El autor expone por qué motivos y en qué condiciones se consideró que el empleo de la mecanografía podía dar buenos resultados: describe, en particular, el sistema de clave empleado, los circuitos de documentos y los plazos, e indica de paso las modificaciones que ha sido necesario introducir en el sistema de contabilidad utilizado para ajustarlo a las exigencias de la mecanografía.

Por último, en la tercera parte se trata de exponer algunas de las consideraciones sobre el costo en personal y en dinero de esta centralización de los datos contables y, en general, sobre el costo de la administración de los materiales nucleares, a que ha dado lugar el empleo de la contabilidad mecanográfica.

1. LE SYSTEME FRANCAIS DE GESTION DES MATIERES NUCLEAIRES

1.1. Activités de la France dans le domaine de l'énergie nucléaire

La France poursuit un programme nucléaire dont les grandes lignes sont connues. Le programme est centralisé et coordonné par le Commissariat à l'énergie atomique (CEA) créé en 1945, en liaison avec l'Electricité de France, entreprise publique chargée en France de la production et de la distribution d'énergie électrique et qui notamment construit et exploite les centrales nucléaires.
1.1.1. Les principaux points de ce programme sont les suivants :
- la construction de centrales atteignant une puissance de 1500 MW(e) installés en 1967 et 4000 à 5000 MW(e) en 1975, et l'achèvement de la mise en place d'un circuit complet d'alimentation en combustible de ces centrales et de retraitement de leurs combustibles irradiés ;
- un programme de recherche fondamentale et appliquée poursuivi soit isolément, soit en collaboration avec les partenaires de la France au sein de l'EURATOM ou de l'Agence européenne de l'énergie atomique ;
- l'étude et la construction de sous-marins à propulsion nucléaire.

1.1.2. Les activités françaises dans le domaine nucléaire ont déjà entraîné la mise en œuvre de plusieurs milliers de tonnes d'uranium naturel, plusieurs centaines de kilos de plutonium, plusieurs centaines de kilos d'uranium-235.

1.1.3. Ces matières, qui sont soit produites en France, soit importées, sont réparties entre
- seize sièges miniers, dont neuf appartenant au Commissariat à l'énergie atomique et sept à des entreprises privées ;
- huit usines de transformation de l'uranium aux différents stades : quatre pour la concentration des minerais, deux pour le raffinage, deux pour la fabrication en série des éléments de combustible ; deux d'entre elles sont exploitées par le Commissariat à l'énergie atomique et six par des entreprises privées ;
- quatre centres de recherche civils appartenant au Commissariat à l'énergie atomique dans lesquels fonctionnent ou sont en construction vingt-deux réacteurs de recherche ou prototypes ou expériences critiques ;
- plusieurs laboratoires ou centres de recherches, spécialisés dans l'énergie nucléaire, appartenant à des entreprises privées ;
- un centre de production de plutonium dans lequel fonctionnent trois réacteurs ;
- deux usines de retraitement des combustibles irradiés ;
- quatre établissements de l'Electricité de France dans lesquels fonctionnent ou sont en construction six réacteurs de puissance ;
- une usine de séparation des isotopes de l'uranium.

Par ailleurs, un certain nombre d'industriels ou de laboratoires privés ou publics détiennent des matières nucléaires dans le cadre de contrats de recherche passés avec le Commissariat à l'énergie atomique.

1.2. Principe généraux de gestion des matières nucléaires

La gestion des matières nucléaires en France est fondée sur quatre grands principes :

1.2.1. Premier principe

Tout au long du cycle des combustibles en amont et en aval des réacteurs, le Commissariat à l'énergie atomique est seul responsable de l'affectation des matières aux différents objectifs poursuivis, de leur gestion et de leur contrôle.
Ce principe s'applique aussi bien aux matières se trouvant dans les établissements du Commissariat à l'énergie atomique qu'à celles se trouvant dans d'autres établissements. Il s'applique évidemment sous réserve du droit de propriété général sur les matières fissiles spéciales conféré aux membres de l'EURATOM par le traité de Rome de 1957, et des limitations d'emploi auxquelles sont soumises certaines matières importées, des Etats-Unis notamment.

En ce qui concerne les réacteurs de puissance construits et exploités par l'Electricité de France, le Commissariat à l'énergie atomique reste propriétaire du combustible qu'il met à la disposition de l'Electricité de France pendant toute la durée de l'irradiation moyennant une redevance financière et il en reprend la disposition après la sortie du combustible du réacteur.

1.2.2. Deuxième principe

Chacun des établissements énumérés ci-dessus, ou éventuellement une partie de l'un d'eux, constitue une «unité de gestion des matières nucléaires». Chacune de ces unités est responsable des matières qu'elle détient; elle en organise la gestion avec une certaine liberté à condition de respecter certaines prescriptions minimales contenues dans un «code des matières nucléaires». Elle effectue un inventaire au moins une fois par an sous sa propre responsabilité.

1.2.3. Troisième principe

L'ensemble des données comptables émanant des unités de gestion est regroupé par un organisme central. Chaque unité de gestion est responsable devant lui de la gestion des matières qui lui sont confiées. Elle lui adresse des relevés comptables de la situation de ces matières (mouvements internes, inventaires). Tout mouvement de matière entre deux unités de gestion fait l'objet d'un bon de livraison dont un exemplaire lui est adressé. Cet organisme central est en outre chargé d'une façon générale de veiller au bon fonctionnement de l'ensemble du système de gestion des matières nucléaires et de proposer et de faire appliquer les nouvelles règles qui peuvent se révéler nécessaires.

1.2.4. Quatrième principe

Le système de comptabilité matière est unique. Il est conçu de façon à pouvoir être utilisé aussi bien pour l'établissement des programmes et le contrôle de leur réalisation que pour les besoins de la gestion financière, ou pour les nécessités des contrôles extérieurs auxquels la France a accepté de soumettre un certain nombre de ces matières.

1 Unité de gestion des matières nucléaires: Ensemble du personnel et des installations affectés à la gestion des matières nucléaires placées dans des limites topographiques déterminées sous une responsabilité unique et affectées à un même objectif. Une unité de gestion est définie comme telle par son inscription sur une liste appropriée.
L’institution d’un tel système centralisé de gestion des matières nucléaires n’a pas répondu initialement à un souci de contrôle de sécurité. Il était surtout destiné à servir de base à l’établissement des programmes d’approvisionnement, de production, de transformation et d’utilisation des matières nucléaires, et à fournir une information rapide et automatique sur leur réalisation. Lorsque le Commissariat à l’énergie atomique a dû faire face à des contrôles extérieurs, le système de gestion préexistant a été tout naturellement utilisé pour faire face à ces obligations nouvelles. Ces contrôles extérieurs sont, d’une part, celui que nos partenaires et nous-mêmes avons volontairement institué par le traité de l’EURATOM et qui a été appliqué à partir de 1959 sur toutes les matières affectées au programme civil français et, d’autre part, celui que l’USAEC impose sur les matières fissiles spéciales qu’elle met à la disposition de tiers.

C’est actuellement la Direction des productions du Commissariat à l’énergie atomique (plus précisément la Section des programmes du Département technique de cette Direction) qui est chargée
a) d’organiser, dans le cadre d’un groupe de travail permanent réunissant les principaux services intéressés, la gestion des matières nucléaires en France.
b) de centraliser toutes les données techniques, administratives, ou de sécurité relatives à leur gestion.

1.3. Fonctionnement du système

1.3.1. On distingue à l’intérieur des unités de gestion deux parties:
- les lieux de stockage, dans lesquels une matière demeure entreposée sous une forme déterminée,
- les lieux de transformation (atelier, réacteur) dans lesquels la matière subit une transformation physique ou chimique.

1.3.2. Le système de comptabilité des matières nucléaires prévoit
a) l’enregistrement des mouvements entre des unités de gestion ou entre l’une d’elles et l’extérieur; des mouvements à l’intérieur d’une unité de gestion, entre un lieu de stockage et un lieu de transformation; des variations de stock: pertes, différences d’inventaire, réévaluation, productions nucléaires, etc.
b) l’établissement des balances pour chaque unité de gestion, matière par matière et produit par produit, pour calculer, compte tenu des stocks du début du mois, la situation des stocks à la fin de chaque mois et être à même de la comparer à tout moment avec la situation réelle résultant de l’inventaire de ces services.

2 Une «matière» est définie par un élément chimique, une composition isotopique, un régime de contrôle et d’approvisionnement, par exemple: uranium enrichi à 20% fourni sur la base de l’accord bilatéral franco-américain.
3 Un «produit» est la forme chimique ou géométrique sous laquelle peut se présenter une matière donnée, par exemple: pour la matière «uranium naturel produit en France» les formes «uranate de soude», «UF₆», «lingot de tel type», «élément combustible de tel type».
1.3.3. Les documents de base utilisés par cette comptabilité sont
a) Les bons de mouvements externes: bon de livraison émis par l'unité
de gestion qui expédie vers une autre unité de gestion ou vers l'extérieur
du système; bon d'entrée émis par une unité de gestion qui reçoit de l'ex-
térieur (et non d'une autre unité de gestion).

b) Les relevés de mouvements internes qui, pour chaque unité de ges-
tion, à l'exception de celles constituées par les laboratoires des centres
de recherche, retracent tous les mouvements qui se produisent à l'intérieur
de l'unité de gestion, entre lieux de stockage et lieux de transformation.

c) Les bons de variation de stock émis par les unités de gestion pour
toute opération autre qu'un mouvement externe qui a pour effet de modifier
le stock total d'une matière donnée.

Ces divers documents se présentent sous forme de liasses de plusieurs
exemplaires qui permettent à l'unité de gestion émettrice d'informer si-
multanément les divers services intéressés et, dans le cas de mouvement
externe, de se donner mutuellement décharge.

1.3.4. Les responsabilités concernant ces documents sont établies par
un «Code des matières nucléaires». Une liste nominative des personnes
pouvant émettre ou recevoir les bons est tenue à jour.

Lorsque les matières se trouvent, comme c'est le cas pour la plus
grande quantité, en possession d'un organisme extérieur au Commissariat
da l'énergie atomique, le département du Commissariat à l'énergie atomique
chargé des rapports techniques et contractuels avec cet organisme est res-
ponsable des matières. Pour les installations les plus importantes, le Com-
missariat à l'énergie atomique dispose sur place d'un contrôleur permanent.

1.3.5. Les unités employées sont celles du système métrique et décimal.
Toutes les matières, quelle que soit la forme sous laquelle elles se pré-
sentent et leur composition isotopique, sont comptabilisées en éléments con-
tenus, ce qui présente l'avantage que tout au cours du cycle de combustible
une même matière est comptabilisée dans la même unité. Les quantités ne
sont donc jamais exprimées par exemple en livres d'U₃O₈ ou en grammes
d'uranium-235.

La précision des données numériques, le nombre de chiffres signifi-
catifs, les erreurs admissibles, sont fixés par le «Code des matières nu-
cléaires» dont il a déjà été parlé.

1.3.6. Les matières soumises à ces dispositions particulières de gestion
sont principalement:
- l'uranium,
- le thorium,
- le plutonium,

mais les règles de gestion qui avaient donné satisfaction dans leur cas ont
aussi été étendues à d'autres matières:
- les autres transuraniens,
- l'eau lourde et le deutérium,
- le lithium et le tritium,
- et, en partie, le zirconium et le béryllium.
2. UTILISATION DE MOYENS MECANOGRAPHIQUES

2.1. Considérations générales

La croissance rapide des quantités de matières nucléaires, du nombre des installations dans lesquelles elles sont produites, transformées et utilisées (unités de gestion), la diversité des formes physiques, chimiques, géométriques et isotopiques sous lesquelles elles sont présentées (matières et produits) et enfin l'augmentation du nombre des utilisateurs de données établies par la comptabilité matièr...
2.2. Description du système actuellement expérimenté en France

On se limite évidemment dans ce mémoire à une description assez générale du système, sans entrer dans le détail de la technique mécanographique pour laquelle le problème à traiter était relativement classique.

2.2.1. Les documents élémentaires utilisés sont les mêmes que par le passé (voir 1.3.3.).

Ces documents élémentaires émis par les unités de gestion sont codés et transmis chaque mois à l'atelier de mécanographie, qui dispose déjà de la situation des stocks à la fin du mois précédent. Celui-ci établit, pour chaque unité de gestion, les balances par matière et par produit et en tire les nouveaux stocks à la fin du mois considéré. Ces balances et ces stocks sont directement présentés sous les différentes formes correspondant aux besoins des divers utilisateurs.

2.2.2. La comptabilité établie par l'atelier de mécanographie étant constituée par des balances entre des mouvements de différentes matières intéressant différentes unités de gestion, une partie importante du travail de préparation a consisté à établir et à codifier une nomenclature détaillée et rigoureuse - des matières et des formes chimiques, physiques, géométriques et isotopiques sous lesquelles elles peuvent se présenter, - des unités de gestion et des sous-unités distinguées à l'intérieur de celles-ci, - des types de mouvements ou d'opérations à comptabiliser, - des régimes juridiques auxquels sont soumises les différentes matières, - des fournisseurs ou clients extérieurs qui livrent ou reçoivent ces matières.

2.2.3. Les documents établis par l'atelier de mécanographie sont:

a) Pour chaque unité de gestion et pour chacune des matières qu'elle détient:

L'état A:

C'est la balance des stocks et des mouvements internes et externes, produit par produit. L'état A comporte, pour chaque produit ayant été présent au cours du mois dans un lieu de stockage, autant de lignes que de bons de livraisons ou de mouvements internes enregistrés, plus les lignes «ancien stock» et «nouveau stock». Il comporte pour finir, sous la même forme, une balance des quantités de matières ayant été présentes au cours du mois dans un lieu de transformation.

L'état B:

C'est une balance générale et unique pour une matière donnée (à la différence de celles de l'état A établies produit par produit) des mouvements
externes, de l'ancien, et du nouveau stock. Elle comporte une ligne par mouvement externe plus les lignes «ancien stock» et «nouveau stock».

L'état C:

Il donne l'inventaire de l'unité de gestion et comporte, pour une matière donnée, autant de lignes que de produits, plus une ligne «en cours de transformation». C'est, autrement dit, la récapitulation des lignes «nouveau stock» de l'état A. Le total de l'état C redonne évidemment le chiffre de la ligne «nouveau stock» de l'état B.

b) Pour l'ensemble des unités de gestion produisant ou utilisant un produit donné:

L'état D:

Il donne pour chaque produit une récapitulation des productions (assimilées aux livraisons des lieux de transformation aux lieux de stockage) et des consommations (assimilées aux livraisons des lieux de stockage aux lieux de transformation) pour l'ensemble des unités de gestion productrices et consommatrices d'un produit donné. Cet état est surtout destiné à permettre de surveiller la réalisation des programmes et le niveau des stocks.

c) Pour l'ensemble des unités de gestion et pour chaque matière:

L'état E:

C'est une récapitulation matière par matière des lignes «nouveau stock» de l'état B. Il donne donc une répartition, au dernier jour du mois, d'une matière donnée entre l'ensemble des installations françaises.

2.3. Mise en œuvre et premiers enseignements

La décision de principe de recourir à la mécanographie pour la tenue de la comptabilité physique des matières de base a été prise au printemps de 1964. Les grandes lignes du système ont été tracées au cours du mois de juillet, les détails en ont été mis au point au cours du deuxième semestre de 1964; le système a été appliqué à partir du 1er janvier 1965.

La comptabilité du mois de janvier a été adressée par l'atelier de mécanographie le 5 mai 1965, les mois suivants ont suivi à une cadence moyenne d'une série de comptes tous les 15 jours selon les possibilités du planning de l'atelier, et le régime normal a été atteint à partir du mois de juin (comptes du mois de mai). Si ce régime normal est maintenu jusqu'à la fin de l'année 1965, et on a toute raison de penser qu'il en sera bien ainsi, on pourra considérer que le système est satisfaisant du point de vue des délais.

La principale difficulté rencontrée tient à des imprécisions de terminologie dans la rédaction des documents de base. Ces imprécisions peuvent être relatives à la nature des produits qui fait, par exemple, qu'on a comptabilisé un produit sous une rubrique donnée à l'entrée dans un magasin...
(document rédigé par l’expéditeur) et sous une autre à la sortie du magasin (document rédigé par le gestionnaire du magasin). Il est arrivé qu’une telle erreur fasse même apparaître un stock négatif.

Une autre imperfection du système tient au fait qu’il semble bien que, malgré le caractère aussi universel que possible qu’on a voulu lui donner, celui-ci ne semble pas pouvoir remplacer la totalité des comptabilités tenues localement. On s’attachera dans les mois qui viennent, et en vue de la mise en œuvre d’un système définitif, à tenir mieux compte des besoins particuliers de telle ou telle installation ou de tel ou tel destinataire des documents comptables. C’est ainsi que des dispositions ont déjà été prises pour doubler, dans le cas de certains produits, la comptabilité en élément contenu - seule utilisée officiellement jusqu’ici - par une comptabilité en quantité de produits (tonnes de minerais, quantités d’éléments combustibles). Mais un tel souci comporte évidemment des limites et il faudra probablement beaucoup d’efforts de persuasion pour convaincre certains services de s’adapter au nouveau système.

3. COUT DE LA GESTION DES MATIERES NUCLEAIRES

L’organisation d’une comptabilité centralisée à l’échelle d’un ensemble comme celui constitué par les établissements du Commissariat à l’énergie atomique, les différentes entreprises industrielles françaises détenant de telles matières, et l’Electricité de France, se justifie par un certain nombre de considérations:

a) D’abord la nécessité de s’assurer, à chaque instant, une connaissance aussi précise et aussi exacte que possible de la localisation de ces matières et de la forme sous laquelle elles se trouvent, de façon à adapter les décisions de production et d’approvisionnement aux besoins réels.

b) Ensuite le souci de déceler les parties du cycle de combustible génératrices de pertes afin de pouvoir agir sur les causes de ces pertes.

c) Enfin le souci de faire apparaître, lorsqu’elles se produisent, les immobilisations inutiles de ces matières coûteuses, de façon à essayer de les réduire.

Il ne faut pas perdre de vue qu’en tout état de cause une telle comptabilité centralisée repose sur l’exploitation de comptabilités de base qui comportent en elles-mêmes leur propre justification: toute la gamme des opérations d’échantillonnage, d’analyse, d’établissement de bilans et d’inventaires qu’elles nécessitent présente en effet un caractère absolument indispensable, qu’il s’agisse de contrôle de fabrication ou de contrôles liés à la santé et la sécurité (criticalité, rejets d’effluents...). Les seuls coûts que nous prendrons ici en considération sont donc ceux du collationnement des données et de leur exploitation en vue des objectifs propres de la comptabilité centralisée.

Or cela ne demande pas, en principe, de personnel supplémentaire au niveau des unités de gestion. Par contre, il faut disposer à l’échelon central d’un personnel qualifié assurant un fonctionnement «vivant» de cette comptabilité, c’est-à-dire capable en permanence de remettre en question l’existence de chaque document comptable, de chaque état, de chaque circuit de document, de chaque degré de précision dans la comptabilité de façon
que celle-ci demeure toujours adaptée aux besoins - très changeants - du moment.

A la Direction des productions du Commissariat à l'énergie atomique, trois agents ont suffi jusqu'ici à assurer la gestion des matières nucléaires à l'échelon central. Le coût d'un agent est de l'ordre de 30 000 francs, montant qui comprend ses émoluments, les charges sociales qui s'y rapportent, les fournitures et l'amortissement du matériel qu'il utilise, ainsi que sa prise en charge par l'ensemble des services généraux. Le coût de l'échelon central de gestion à l'échelle des programmes français actuels représente une part infime de la valeur du stock géré et un pourcentage de l'ordre de $10^{-4}$ du flux annuel contrôlé. Il s'agit donc là de dépenses minimes tant en valeur absolue qu'en valeur relative.

Devant une telle conclusion, on pourrait se demander si l'introduction de la mécanographie est justifiée. De toute façon, on ne peut en espérer une diminution d'un personnel déjà limité au strict minimum. L'ensemble du travail de comptabilité des matières nucléaires occupe 10% de l'activité de l'atelier de mécanographie auquel il a été confié. Les dépenses mensuelles de cet atelier sont de l'ordre de 50 000 francs. Les opérations affectées à la gestion des matières nucléaires reviendraient donc à environ 50 000 francs par mois soit l'équivalent du coût de deux comptables. En fait, il serait plus juste de raisonner en coût marginal, cette activité venant s'intercaler dans le mois à une période de sous emploi pour l'atelier.

Les avantages de la mécanographie sont la réduction des délais et la simplification des tâches des divers services intéressés qui disposent désormais de comptes uniques et parfaitement cohérents entre eux, qui n'auraient pu être obtenus que par une multiplication des effectifs jusqu'ici affectés à cette tâche. La rentabilité de l'opération (peu onéreuse en valeur absolue) tient à ce qu'elle rend la comptabilité des matières nucléaires apte à satisfaire les besoins de tous les intéressés qui l'utilisent à des fins souvent assez différentes.

4. CONCLUSION

De l'exposé qui précède il serait vain de vouloir tirer des conclusions de caractère définitif. La « vulgarisation » des matières nucléaires, qui accompagnera inévitablement le développement des programmes d'utilisation pacifique, ramènera-t-elle la gestion des matières nucléaires à une simple comptabilité matière qui ne sera pas différente de celle qu'on tient dans toute industrie? Ou bien leur caractère dangereux et leur importance stratégique conduiront-ils à ce que soit exercé sur elles un contrôle de plus en plus rigoureux, que celui-ci soit national ou international? Il est bien difficile de le prévoir. En tout état de cause on ne dispose pas encore d'un recul suffisant pour apprécier si ceux qui ont actuellement la charge de la gestion des matières nucléaires pèchent par légèreté ou par excès de rigueur, ou bien s'ils ont déjà su s'éloigner de ces deux extrêmes et se maintenir à un juste milieu.
D. E. GEORGE (Chairman): What are the criteria for establishing a management unit, i.e. how do you define such a unit?

F. RONTEIX: The main criteria are geographical unity (propinquity), unity of responsibility (sole command) and unity of purpose. Thus we have "management units" at the level of a thousand tons of natural uranium and others at the level of a few hundred kilograms.

D. E. GEORGE: How do you manage to keep materials from different origins separate from each other?

F. RONTEIX: By definition, we regard materials subjected to different conditions of supply and control as different materials calling for different accounts. Actually such distinctions have not yet been necessary except for enriched uranium and plutonium, for which differences in origin have always coincided with differences in isotopic composition or differences in time (the two types of material not being present in the same installation at the same moment).

However, cases where this coincidence does not occur are now beginning to arise. We try to maintain some separation according to origin, but it sometimes becomes quite artificial. Where for example, enriched-uranium waste from two different sources is reprocessed at the same time, yield and losses are apportioned between the two sources in proportion to the incoming quantities.

J. SORNEIN: In connection with Mr. George's question as to whether the management of special fissile materials from different sources subject to different controls can give rise to major difficulties, I should like to add a comment. I have frequently found that this momentary complication even has a certain advantage: it obliges the various management units to take more care in drawing up the balances.

J. SCHMETS: What type of data do you record, Mr. Ronteix, for fertile and fissile materials destroyed or formed in reactors? Do you rely on provisional estimates or do you wait for results of analysis?

F. RONTEIX: We use provisional figures based on burn-up and build-up calculations. However, irradiated fuels do not constitute the only case in which we record provisional figures in our accounting. We do this also for waste prior to reprocessing or for concentrates for which the delay in analysis is relatively long. For irradiated fuels from power reactors, too, until reprocessing we prefer to use the provisional weight and isotopic composition as determined from the irradiation level, which is usually expressed in MWd/t.
CRITERIA FOR SPECIAL NUCLEAR MATERIALS INVENTORY AND CONTROL PROCEDURES*

E.M. KINDERMAN AND R.R. TARRICE
STANFORD RESEARCH INSTITUTE, MENLO PARK, CALIF., UNITED STATES OF AMERICA

Abstract — Résumé — Аннотация — Resumen

CRITERIA FOR SPECIAL NUCLEAR MATERIALS INVENTORY AND CONTROL PROCEDURES. One of the most significant problems that will face investors, managers and operators in nuclear activities and especially in the field of commercial nuclear power, will be the proper control of a nuclear materials inventory that will exceed US $5000 million in value by 1980.

Special nuclear materials are expensive when compared to most materials of commerce, e.g. US prices for 90% enriched uranium and 3% enriched uranium as hexafluoride, and for heavy water are $10808, $254 and $61.60 per kg, respectively. Moreover, in many cases these materials are subjected because of health and safety requirements to special governmental controls not directly related to their monetary value. Despite the high monetary values assigned to these materials, they are destined to be used in large quantity, e.g. some 50-75 t of 3% enriched material will be used in 500-MW light-water-moderated reactor, and perhaps the equivalent of 200 to 300 reactors of such size will be in operation throughout the world by 1980.

Past experience has resulted in the development of special procedures and practice for the commercial control of the large quantity, lower-value materials such as coal or iron ore and for the small quantity, higher-value materials such as the precious metals. While they have like prices, special nuclear materials are different in kind and will be handled in quantities much greater than the precious metals. However, while special techniques or special adaptations of old techniques may be necessary, proper use of various established inventory control practices should be sufficient in most cases to protect adequately the investment of nations and individuals in these expensive materials.

This paper establishes criteria for materials control. It specifically considers the appropriateness of various techniques of inventory control ranging from annual balancing of book records of receipts and shipments through detailed daily physical inventory in the light of the specific value and the total inventory of these special nuclear materials. A matrix of solutions to the management and inventory control of special nuclear materials will be presented. The multiplicity and relative effectiveness of varied techniques at key stages of the materials supply, utilization and recovery are assessed.

* This paper arises in part from past study of special nuclear material management and inventory control performed as part of Project Agreement 49, Contract AT(04-3)-115 between Stanford Research Institute and the US Atomic Energy Commission.
пети"ес quantifs de matières de grande valeur comme les métaux précieux. Tout en ayant des prix comparables à ces derniers, les matières nucléaires spéciales sont de nature différente et seront utilisées en quantités beaucoup plus importantes que les métaux précieux. Bien qu'il puisse être nécessaire de prévoir des méthodes spéciales ou d'adapter des techniques anciennes, il devrait toutefois suffire dans la plupart des cas d'utiliser de façon appropriée les différentes méthodes d'inventaire bien connues pour assurer la protection des investissements des gouvernements et des particuliers dans l'exploitation de ces matières coûteuses.

Les auteurs du mémoire définissent les critères à utiliser pour le contrôle des matières nucléaires. Ils étudient notamment l'appréciabilité de différentes méthodes d'inventaire: pointage annuel des registres des matières reçues et expédiées, inventaire quotidien et détaillé des matières elles-mêmes en tenant compte de leur valeur spécifique, et inventaire général de ces matières nucléaires spéciales. Les auteurs présentent des solutions types permettant de gérer les matières nucléaires spéciales et de procéder à leur inventaire. Ils procèdent à une évaluation de l'efficacité relative des multiples méthodes applicables aux stades importants de la fourniture, de l'utilisation et de la récupération des matières nucléaires.

**КРИТЕРИИ ДЛЯ ИНВЕНТАРИЗАЦИИ СПЕЦИАЛЬНЫХ ЯДЕРНЫХ МАТЕРИАЛОВ И МЕТОДЫ УЧЕТА.** Одной из самых важных проблем, с которой столкнутся предприниматели, руководители и операторы в области атомной энергии, и особенно в области промышленной ядерной энергетики, является установление надлежащего контроля инвентаризации ядерных материалов, стоимость которых к 1980 году превысит 5 млрд. долл. США.

Специальные ядерные материалы дороги по сравнению с большинством промышленных материалов, например в США цены на уран 90%-го обогащения и на уран 3%-го обогащения в виде шестифтористого урана и на тяжелую воду составляют соответственно 10 808, 254 и 61,60 долл. США за 1 кг. Более того, во многих случаях эти материалы, как тому требует охрана здоровья и техника безопасности, находятся под специальным правительственным контролем, не связанной непосредственно с их денежной стоимостью. Несмотря на большую стоимость этих материалов, предусматривается применять их в большом количестве; например, в реакторе с водным замедлителем мощностью 500 мгвт будет использовано приблизительно 50 – 75 т материала 3%-го обогащения, и, вероятно, во всем мире к 1980 году мощность реакторов, находящихся в эксплуатации, будет эквивалентно равна мощности приблизительно 200 – 300 реакторов такого размера.

На основании прошлого опыта разработаны специальные методы и налажена практика промышленного учета недорогостоящих материалов в большом количестве, например уголь или железная руда, и дорогостоящих материалов в небольшом количестве, например драгоценные металлы. При почти одинаковых ценах специальные ядерные материалы различаются по виду и будут использоваться в количествах, значительно больших по сравнению с драгоценными металлами. Хотя, вероятно, потребуются специальные методы или соответствующее изменение старых методов, надлежащее использование многообразной установленной практики проверки и учёта должно оказаться достаточным в большинстве случаев для должности защиты капиталовложений стран и отдельных лиц в производство этих дорогостоящих материалов.

Устанавливаются критерии для учета материалов. Специально рассматривается вопрос о соответствии различных методов инвентарного контроля, начиная от составления ежедневных балансов учета поступлений и отправок до подробной ежедневной физической инвентарной проверки в отношении определенной стоимости и общего запаса этих специальных ядерных материалов. Дается сводка решений для руководства и порядок осуществления инвентарного контроля за специальными ядерными материалами. Дается оценка множественности и относительной эффективности различных методов на ключевых этапах поставки, использования и восстановления материалов.

**CRITERIOS A QUE DEBEN AJUSTARSE LOS PROCEDIMIENTOS DE INVENTARIO Y CONTROL DE LOS MATERIALES NUCLEARES ESPECIALES.** Uno de los problemas más importantes que deberán resolver las personas que finanzen, administren o desarrollan actividades relacionadas con los procesos nucleares, especialmente en la esfera de la producción comercial de energía nucleoeléctrica, ser el que plantea el control adecuado de los materiales nucleares, cuyo valor en 1980 excederá de 5000 millones de dólares de los Estados Unidos.

Los materiales nucleares especiales son caros: en los Estados Unidos, por ejemplo, el uranio enriquecido al 90%, el hexafluoruro de uranio enriquecido al 3% y el agua pesada cuesta, respectivamente, 10 808, 254 y 61,60 dólares el kg. Además, por motivos sanitarios y de seguridad, esos materiales están en muchos casos sometidos por parte de las autoridades a un control especial sin relación directa con su valor monetario. Pese a su elevado precio, los materiales nucleares se emplearán en grandes cantidades: basta señalar que un reactor
de 500 MW, moderado por agua ligera, necesitará de 50 a 75 toneladas de material enriquecido al 3% y que es probable que en 1980 funcionen en el mundo de 200 a 300 reactores de esta potencia.

La experiencia ha permitido elaborar procedimientos y prácticas especiales para el control comercial de grandes cantidades de materiales baratos, como el carbón o el mineral de hierro, y de pequeñas cantidades de materiales caros, como los metales preciosos. Aunque sus precios son similares a los de estos últimos, los materiales nucleares especiales son de distinta clase y se utilizarán en cantidades mucho mayores. De todas formas, aunque quizá sea necesario recurrir a técnicas especiales o adaptar técnicas antiguas, en la mayoría de los casos bastará utilizar convenientemente los procedimientos ya conocidos de control de las existencias para proteger en forma adecuada las inversiones nacionales o individuales en esos materiales tan caros.

En la memoria se exponen algunos criterios para el control de los materiales. Se estudian especialmente las técnicas de control de las existencias (desde el balance contable anual hasta el inventario físico efectuado a diario) que resultan más apropiadas dado el valor específico y la cantidad total de los materiales nucleares especiales. Se presenta una matriz de soluciones para la administración y el control de esas existencias. Se evalúa la multiplicidad y la eficacia relativa de diversas técnicas en momentos decisivos del suministro, el empleo y la regeneración de los materiales.

1. INTRODUCTION

The nuclear industry – especially the electric power portion of it – is beginning a long period of rapid growth. This growth involves several features which make the industry one of unusual interest.

In the first place, in several of the larger nations of the world, with the transfer of large numbers of nuclear technologists to peaceful uses of nuclear energy such as electric power, large-scale peaceful operations have been undertaken in a time that is short compared with that which would be required for normal industrial development. Also, the increase in the need for electrical power, brought about through rapid industrialization in almost every nation, has placed new and increasing demands on the producers of electrical power and on the conventional energy sources for that power – coal, oil, gas, and water. These new requirements, coupled with our ability to bring the needed technologies rapidly into large-scale use, make likely, for the nuclear industry, a rate of growth and eventual level of operation much greater than usual in a new industry.

A second factor also tends to make this new industry unique. Nuclear methods of power production are inherently more intensive; the specific energy derived from nuclear transformation is enormous compared with that from chemical transformations (combustion). The quantities of fuel material that will be required for nuclear power generation, while large enough to be measured in thousands of tons, are small compared to the millions of tons of coal and oil required today for commercial power production. The new nuclear fuels are correspondingly expensive. Current United States prices for 90% enriched uranium and 3% enriched uranium as hexafluoride are $10 808 and $254 per kg, respectively. United States prices for heavy water have been $61.60 per kg though new Canadian contracts indicate a lower price will soon be reached. Uranium concentrates are priced, depending on circumstances, at $11 to $17 per kg of U₃O₈. These prices should be compared to prices for platinum ($2500 per kg), gold ($1000 per kg), silver ($33 per kg) and zirconium ($17 per kg).

The monetary value of the reactor fuel inventory will not be prohibitively high, but the enriched uranium in the core of a 500-MW(e) water-moderated reactor has an input inventory value of approximately $20 million.
By comparison a typical United States 500-MW(e) coal-fired plant might consume that value of coal over a three- to four-year period and at any time have a coal inventory on hand valued at one-tenth to one-twentieth of the reactor core value. We believe it is reasonable to assume an in-process inventory for the nuclear-fired portion of the United States power industry of about $5 billion by 1980. This inventory will enable the utilities to supply 10% of the electrical power capacity and 15% of the power used in the United States. Equivalent or even larger nuclear power production and process inventories will exist in the other industrialized countries of the world.

In addition to the technical and economic factors mentioned above, one must consider that the nuclear power industry is developing at a time when scientific and engineering technology is subject to widespread public discussion and that the impact of technical developments on the health of the community is considered carefully by thoughtful citizens. The introduction of intense power sources from these nuclear programmes has received careful scrutiny by private citizens and by local and national governments. For example, proper regulation of the material to ensure that critical incidents or uncontrolled dispersal of small quantities of radioactive material do not occur has been the topic of symposia sponsored by the International Atomic Energy Agency.

High specific material value, large inventory investment, and potential hazard to health and safety each require good inventory control; the combination of them makes it mandatory on the managers and owners of the facilities involved to direct special attention to their fiscal and social responsibilities.

2. CONTROL OF NUCLEAR MATERIALS

One might ask what techniques are available to ensure this needed control. Analysis and sampling, record keeping, audit, and other standard inventory control procedures certainly can and should be used. Suitable chemical and physical methods of analysis are available for measurement of inventories of simple materials in the reactor cycle such as ore, uranium dioxide, and uncontaminated uranyl nitrate solutions. Analyses of more complex materials or difficult-to-separate components such as process scrap, fuel elements containing both fuel and poison, and the highly radioactive "head-end" solutions of an irradiated fuel reprocessing plant are less precise and more difficult. Sampling also varies in difficulty, accuracy, and precision. Most analytical methods used within the nuclear industry produce results that compare favourably with the best methods used in other industry. With the high specific value of nuclear products is this enough to protect adequately the financial resources invested? How can the analyses be supplemented to ensure greater protection?

Obvious technical approaches to improved control include careful review of the technical processes used and physical manipulations involved to ensure that all probable (and perhaps improbable) sources of loss or diversion from measured channels are covered by physical barriers and/or rigorous administrative controls. Other useful techniques to ensure proper
control over inventory are maintenance of proper and adequate records of material transfers, design of scale and balance and analytical standard programmes to avoid bias, proper application of statistical analysis to permit evaluation of the inventory uncertainties arising from measurement error, frequent physical inventory, audit of physical measurement as well as of record systems, and combinations of these techniques. Other comparable accounting procedures could be listed.

Complete and thorough application of these procedures requires extensive commitment of a highly trained staff. The expense of this commitment must be considered carefully. This leads to another question. Should all the techniques described be used at all facilities that have stocks of uranium, plutonium, heavy water, or other valuable materials of the nuclear industry? While an easy answer is, "Of course, use all of the control techniques available," our answer is, "Not necessarily." We believe that economic factors must be considered as well. Are there criteria for selection of the proper techniques? We believe that there are, but how should one select the techniques to be used? This paper offers some criteria for that selection process for consideration by those who will have the responsibility to make decisions on the control of nuclear materials inventories.

3. CRITERIA FOR CONTROL

Two sets of criteria can be assembled for our purpose. The first set is related to monetary value while the second defines the relative degree of control offered by various control techniques. For illustration of the second set, we might point out that precise and accurate measurement of materials received offers the possibility of good control, while estimates alone of materials received or acceptance of quantities stated by the shipper without measurement introduce the possibility of poor control and error.

Application of monetary criteria defines in economic terms the degree of control required. The subsequent qualitative evaluation of available control techniques can be matched to this to define the degree of effort required for any particular situation.

For examination of the first set of criteria, let us arrange unit value and total inventory value in a matrix as shown in Table I.

One would expect intuitively that inventory control would be more important to the operations and inventory represented by A and D, than to operations B and C. For equivalent monetary control of inventory, the high specific value of the material in operation A requires that measurement, and other techniques of control, be more precise for that operation than for operation D.

The qualitative values represented by high, etc. can be used to establish the matrix. The definition of large, medium, and small total inventory can best be established by the individual industrial organization. The degree of risk to the financial operations of various companies can be equivalent over a very wide range of total nuclear materials inventories. Only the management of a given company is in a position to ascertain the proper degree of control relative to its responsibilities. Comparison of fuel inventory
value with total inventory or with total investment might be appropriate. For example, a major electric power producer might designate an in-reactor inventory ($10-25 million) as one of medium inventory value, while the total inventory for several reactors, and their related in-process fuel stockpiles ($>100 million) would fall in the major category. On the other hand, a small independent scrap reprocessor with only a $2 million inventory on hand might well consider this a major or high inventory investment.

It is convenient to draw comparisons with precious metals to exhibit the significance of high, intermediate, and low unit values. High specific value materials include highly enriched uranium, plutonium, or any other material whose value exceeds that of gold or platinum ($>1000/kg).

Intermediate value materials include slightly enriched uranium, suitable for use in the power reactors of the light-water moderated type ($>200/kg), heavy water ($30 to $50/kg), and similar materials whose values compare somewhat with silver. Low specific value materials include uranium and thorium ore concentrates ($<20/kg), depleted uranium, and other materials whose specific values compare generally with zirconium and other specialty metals.

The second set of criteria relates to techniques of control. Typical techniques are: physical and chemical measurements of shipments, receipts, and inventories; documentation of transfers; internal material balance or inventory control areas; established organizational responsibility for inventory control; announced internal audit at regular intervals; unannounced external audit and inspection; statistical and/or analytical control programmes, including scale and balance control programmes; total material balance reports.

While all these techniques are desirable, the degree to which they should be applied will be dictated by the matrix described above. We believe that the techniques of Table II should be applied to control all inventories, resulting in the degrees of control shown. The degree of control indicated is the result of our judgments. Those applying the methods must consider their individual requirements and arrive at their own individual judgments.
TABLE II
CONTROL TECHNIQUES
APPLIED UNIFORMLY TO ALL INVENTORIES

<table>
<thead>
<tr>
<th>Generally applicable techniques</th>
<th>Resulting degree of control</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>High</td>
</tr>
<tr>
<td>Physical and chemical measurements of shipments, receipts and inventories</td>
<td>.</td>
</tr>
<tr>
<td>Documentation of all transfers</td>
<td>.</td>
</tr>
<tr>
<td>Established responsibility for inventory control</td>
<td>.</td>
</tr>
<tr>
<td>Unannounced audit</td>
<td>.</td>
</tr>
</tbody>
</table>

The measures which can be applied to varying degrees are shown in Table III. Again, we have indicated our evaluation of the degree of control offered by the various techniques.

The case of a large nuclear complex involving several plants with differing composition of inventory may serve to demonstrate the applicability of the considered approach to materials control that we have postulated. The matrix in Table IV presents such an approach.

The techniques for control in each case are supplementary to those of regular measurement, record keeping, assigned responsibility, and unannounced audit which are assumed to be generally applicable in all cases.

The concepts we are presenting can be illustrated through a hypothetical case study. This case could represent a national effort on a relatively small scale or the operations of a large vertically-integrated industrial organization.

The complex could conceivably be composed of the activities shown in Table V, each of which has been coded for identification in a later evaluation. Each of the activities indicated will have characteristic mixes of materials and values of inventory. Assuming logical ranges of operations in these activities, it is possible to postulate the probable variations among them that would lead to adoption of specific techniques and degrees of control (Table VI).

Here for the illustration we have used the limits of 0.5 million and 10 million to define the separation between small, medium, and large inventory. We repeat that individual organizations must set their own limits according to their financial responsibilities. The limits used here are arbitrary ones chosen for example. Several activities may appear in several
KINDERMAN and TARRICE

TABLE III

CONTROL TECHNIQUES
APPLIED IN VARYING DEGREES TO INVENTORIES

<table>
<thead>
<tr>
<th>Techniques of varying applicability</th>
<th>Resulting degree of control</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>High</td>
</tr>
<tr>
<td>Audit</td>
<td></td>
</tr>
<tr>
<td>Resident</td>
<td></td>
</tr>
<tr>
<td>Annual</td>
<td></td>
</tr>
<tr>
<td>Biannual</td>
<td></td>
</tr>
<tr>
<td>Monthly</td>
<td></td>
</tr>
<tr>
<td>Material balance reports</td>
<td></td>
</tr>
<tr>
<td>Quarterly</td>
<td></td>
</tr>
<tr>
<td>Annual</td>
<td></td>
</tr>
<tr>
<td>Statistical and/or analytical control programmes</td>
<td></td>
</tr>
<tr>
<td>Internal material balance areas where possible</td>
<td></td>
</tr>
</tbody>
</table>

of the columns and rows. For example, a research and development laboratory (RD) engaged in development of fast reactor fuels will have a moderate inventory of high value material. Another laboratory investigating the separation of thorium from its ores would have a small inventory of low unit value material.

A conversion and fuel fabrication plant (CF) may be of varying size. A small process inventory for a partial core of a research reactor, which uses medium to high enrichment uranium, would result in a high unit value in the medium inventory classification. The same processor holding an inventory for a large power reactor core of low enrichment uranium would fall into the large inventory, intermediate value category.

What control system is appropriate for an activity with varying inventory and unit values? It would seem reasonable to apply the control system which would protect the largest inventory and the highest unit value materials expected to be processed. In the particular example of the conversion fuel fabrication plant described, the previously presented control matrix calls for the same techniques for high value-medium inventory cases as for intermediate value-large inventory cases. These include, as always, generally applicable techniques of regular measurements of receipts, shipments and inventory, planned record keeping, assigned responsibility, and unannounced audit as well as the specific techniques of annual audit, a statistical and analytical control programme, and monthly inventory.

Similar reasoning can be applied to any activity, be it a plant or separate portion of a plant, where several materials of differing unit values are
### TABLE IV

**MATRIX DEMONSTRATING APPLICATION OF VARYING CONTROL TECHNIQUES TO INVENTORIES OF VARYING SIZE AND KIND**

<table>
<thead>
<tr>
<th>Total inventory</th>
<th>Unit value</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td><strong>High</strong></td>
</tr>
<tr>
<td>Large</td>
<td>Resident auditor</td>
</tr>
<tr>
<td></td>
<td>Statistical and/or analytical control</td>
</tr>
<tr>
<td></td>
<td>Monthly inventory</td>
</tr>
<tr>
<td>Medium</td>
<td>Annual audit</td>
</tr>
<tr>
<td></td>
<td>Statistical and/or analytical control</td>
</tr>
<tr>
<td></td>
<td>Monthly inventory</td>
</tr>
<tr>
<td>Small</td>
<td>Biannual audit</td>
</tr>
<tr>
<td></td>
<td>Quarterly inventory</td>
</tr>
</tbody>
</table>

### TABLE V

**ACTIVITIES TYPICAL OF A LARGE, VERTICALLY INTEGRATED INDUSTRIAL ORGANIZATION**

<table>
<thead>
<tr>
<th>Activity</th>
<th>Code</th>
</tr>
</thead>
<tbody>
<tr>
<td>Research and development laboratory</td>
<td>RD</td>
</tr>
<tr>
<td>Mining and milling operations</td>
<td>MM</td>
</tr>
<tr>
<td>Refining</td>
<td>RE</td>
</tr>
<tr>
<td>Enrichment</td>
<td>EN</td>
</tr>
<tr>
<td>Conversion and fuel fabrication</td>
<td>CF</td>
</tr>
<tr>
<td>Scrap reprocessing</td>
<td>SR</td>
</tr>
<tr>
<td>Power production</td>
<td>PP</td>
</tr>
<tr>
<td>Fuel reprocessing</td>
<td>FR</td>
</tr>
</tbody>
</table>
found together. Uranium of varying enrichment, for example, cannot be readily distinguished by simple inspection. To protect the most valuable material, all the uranium must be controlled in the manner appropriate to that of the most valuable material.

The principal responsibility for given materials inventories will rest with individual organizations. The organizations' managements will in turn dictate the specific system(s) of control appropriate to the materials involved. Adequate internal control of materials can therefore be anticipated within a plant or corporate structure. Although not considered separately in this paper, the inter-organization problems that can arise in extensive transfers of the materials considered here can be measurably decreased when co-ordinated programmes of control are established on an interorganization basis as well as intra-organization. Co-ordination can take many forms, including, as a first step, establishment of standards for materials content and methods of measurement for the entire industry. Thousands of individual transfers of materials, representing commerce of billions of dollars annually between hundreds of organizations, will create the need for close and careful co-ordination among the countries and industrial firms involved. Such co-ordination would appear to offer such economies and increased control potential as to make it most worthy of further detailed consideration.
4. CONCLUSION

An outline of a general system by which material control procedures can be evaluated has been presented. This system suggests specific evaluations of the many individual problems faced by the management of organizations using and processing nuclear materials. The system uses monetary value of individual units and of total inventory to set criteria for control procedures. We have offered examples to illustrate the method, selecting arbitrarily the monetary levels which divide categories of control. The monetary levels, particularly those relating to total inventory, used as dividing lines between categories must be selected by the individual organization.
CASE STUDY IN INTERNAL AUDIT OF NUCLEAR MATERIAL

S. KOPS
UNITED STATES ATOMIC ENERGY COMMISSION, ARGONNE, ILL., UNITED STATES OF AMERICA

Abstract — Résumé — Аннотация — Resumen

CASE STUDY IN INTERNAL AUDIT OF NUCLEAR MATERIAL. The general need for the internal audit function and the basic principles of management underlying this need are briefly discussed. The size, complexity and degree of centralization, or decentralization, are usually the main factors in determining the need for a conventional internal audit. Here, the more specialized needs and characteristics of an internal audit peculiar to an organization handling source or special fissionable material (nuclear material) are discussed and contrasted with an audit of the standard type. The necessity of a special audit, because of the high monetary and strategic value, is demonstrated.

Planning the internal audit within a production plant is discussed in detail. This includes the rationale of timing the review together with the records and physical inventory to be tested and verified. Quality and types of professional skills of the personnel to be used in internal audit are also discussed. A qualified statistical plan in choosing the items to be tested is discussed.

The application of the plans in the actual performance of the internal audit are described in detail. Possible variances from a recognized norm and possible approaches to audit resolution are discussed. Although in many other manufacturing situations there are variations in the methods of verifying inventories without physically doing so, the necessity for physically testing inventories of nuclear material is demonstrated.

The various means of reporting the results of the internal audit are presented. The needs of individual groups within the organization are considered in determining the format of the report, as well as the basic content. Distribution of the report and its variations are discussed.

ETUDE D’UN SYSTEME DE VERIFICATION COMPTABLE INTERNE DES MATIERES NUCLEAIRES. L’auteur étudie brièvement la nécessité de la vérification comptable interne en général, et les principes essentiels de gestion auxquels répond cette nécessité. La taille de l’installation, la complexité et le degré de centralisation ou de décentralisation sont généralement les principaux facteurs à considérer lorsque l’on veut déterminer s’il y a lieu d’établir un système classique de vérification comptable interne. L’auteur analyse les besoins et les caractéristiques d’un système de vérification comptable interne dans le cas particulier d’une organisation qui manipule des matières brutes ou des produits fissiles spéciaux (matières nucléaires), en les confrontant à ceux d’un système de vérification classique. Il montre comment, étant donné le prix élevé et la valeur stratégique des matières, il est nécessaire d’établir un système spécial de vérification comptable.

Le mémoire étudie en détail l’organisation du système de vérification comptable interne dans une installation produisant des matières nucléaires; il indique notamment comment fixer la date des contrôles et comment procéder à la vérification des livres et inventaires. Il indique également la compétence et les qualifications professionnelles que doit avoir le personnel chargé de la vérification comptable, ainsi que les considérations d’ordre statistique à appliquer pour déterminer les données qui devront faire l’objet d’une vérification.

Le mémoire décrit de façon détaillée la manière dont ces dispositions sont appliquées dans la pratique de la vérification comptable interne. Il indique les modifications qui peuvent être apportées aux normes établies ainsi que les différentes manières d’envisager le problème de la vérification des comptes et montre que, s’il est possible dans le cas d’autres fabrications de procéder simplement à une vérification comptable des stocks, il est nécessaire dans l’industrie nucléaire de procéder effectivement à l’inventaire matériel du stock.

Le mémoire présente les divers moyens utilisés pour communiquer les résultats de la vérification interne. La présentation du rapport comptable et sa teneur sont déterminées en fonction des besoins des différents services. Le mémoire indique les diverses manières dont le rapport comptable peut être distribué.
ИЗУЧЕНИЕ ПРИМЕРА ОРГАНИЗАЦИИ ВНУТРЕННЕГО УЧЕТА ЯДЕРНЫХ МАТЕРИАЛОВ НА ПРЕДПРИЯТИИ. Кратко обсуждаются общая потребность в организации внутренней проверки наличия ядерных материалов на предприятии и основные принципы административно-хозяйственного учета. При определении потребности в организации обычного внутреннего учета главными факторами являются: размер предприятия, его сложность и степень централизации или децентрализации. Обсуждаются и противопоставляются внутреннему учету обычного типа более специальные требования и особенности внутреннего учета, присущие предприятию, располагающему радиоактивными источниками или специальными расщепляющимися материалами (ядерными материалами). Ввиду высокой денежной стоимости и стратегической важности материалов доказывается необходимость организации особой внутренней проверки.

Подробно обсуждается планирование организации внутренней проверки на производственном предприятии. Помимо ведения учетных документов и фактической проверки наличия материалов, подлежащих проверке и выявлению, объясняются соображения, лежащие в основе установления сроков обследования. Обсуждаются также квалификация и категории специалистов, которые должны быть привлечены к выполнению функций внутренней проверки. Обсуждается конкретный статистический план выбора объектов для испытаний и проб.

При определении форм представления отчетов о результатах внутренней проверки. Обсуждаются различные способы представления отчетов. При определении формы представления отчетов и их содержания учитываются потребности разных отделов предприятия. Обсуждается схема рассылки отчетов и их возможные варианты.

Management has the responsibility for adopting sound accounting policies for maintaining an adequate and effective system of internal control for the safeguarding of assets. The effectiveness of internal control, in the final analysis, can best be determined by periodic inspections or audits by knowledgeable people.

In most industries, as a business is organized, management is usually composed of a small body of closely associated single-purpose individuals. They have a common interest of moulding a successful business out of li-
terally nothing. There are very few problems of internal control in the beginning since the people involved are of such a single-minded purpose and the production and administration are being carried on in a single establishment. As the organization grows, however, problems are introduced which were not present before. As production is carried on in numerous locations, administration becomes more difficult. In addition, specialists are now hired to fulfill the functions which were previously carried on by the owners. Communication problems become more difficult. The problem of internal control grows larger.

At this point in the growth of any organization, this need for an internal audit programme is justified. If, however, the organization had grown but did not decentralize, another set of factors would have to be considered before going into an internal audit programme. With all production and administration in one location, the problem of internal control is greatly reduced.

"Any system of internal control, however, regardless of its fundamental soundness, may deteriorate if not reviewed periodically. The system of internal control should be under continuing supervision to determine whether (1) prescribed policies are being interpreted properly and are being carried out, (2) changes in operating conditions have made the procedures cumbersome, obsolete or inadequate, and (3) effective corrective measures are taken promptly where breakdowns in the system appear. An internal audit... is a strong factor in a system of internal control, since it provides a means of surveying the effectiveness of and adherence to the prescribed procedures."  

As has been indicated above, an internal audit is an important part of internal control. As has also been pointed out, the need for internal audit normally increases with production expansion and decentralization. With an organization handling source or special fissionable material (nuclear material), the need for internal audit is evidenced even though this organization is small and centralized. The high monetary value of the material is usually so out of proportion to the contract price that management is encouraged by economics alone to institute an internal audit programme to police adherence to the internal control procedures.

As an example, using government-furnished highly enriched uranium (93%) with a monetary value of approximately $11,000.00 per kilogram of uranium as UF₆, it is probable that this contingent liability for losing a portion of the material would exceed the processing contract price. In a specific case, a prime contractor of the USAEC had subcontracted a fabrication of highly enriched uranium plates for the total fabrication price of $350,000.00. The subcontract article provided that the Commission would supply 1280 kg of uranium as UF₆ at 93% enrichment. Based upon the published value of 93% enriched uranium, the total value of the uranium processed was $14 million or four times the contract price. At this level of possible liability of loss internal audit becomes imperative, even though this organization is neither large nor decentralized. The possible mone-

---

1 Committee on Auditing Procedure of the American Institute of Certified Public Accountants, Statements on Auditing Procedure No. 33 (1963) 31.
tary loss alone becomes the major factor in deciding whether or not to institute an internal audit programme. In fact, the material unaccounted for in this specific case was 16 kg or $176,000 which represents 50% of the total contract price. Although the internal audit programme would probably have not reduced the loss at all, the administrative personnel were obviously unaware of the possible loss points which would have been highlighted in previous internal audits. This would then have played a large part in price estimating of future contracts.

In planning the internal audit programme for a fuel element production plant, the following major items should be considered:

1. Professional skills needed for review
2. Timing of the review
3. Examination of internal procedures
4. A statistical plan for this review
5. Review of actual procedures against the written ones
6. Records and inventory items to be tested.

Basically, an internal audit of a nuclear material facility should be conducted by professionally capable personnel. The underlined phrase is defined as having adequate technical training and proficiency. Obviously, the internal audit function in this area should be performed by professionals of varying skills. An ideal team would consist of an auditor, a chemist and a statistician. The complexity of such an inspection should encourage management to employ the ideal team as much as possible.

Once a decision has been reached as to the make-up of the internal audit team, the next factor is when should this audit be performed. Aside from the melodrama of having the bank auditor walk in unannounced and subsequently discovering a defalcation of funds, it is not practical to plan a nuclear material internal audit on an unannounced basis. On the other hand, it should be timed so that a complete processing chain can be observed to test the stated procedures with the actual. If only an inventory was to be performed, it would be preferable to do this while the plant was in as close to a shut-down condition as possible. However, since the goal of an internal audit is broader, the actual control procedures should be observable.

Before attempting to perform this audit, the audit team should examine the internal procedures which the organization alleges to be in effect for control purposes. These are usually set down in what is known as a "procedures manual." This procedures manual should be detailed enough to identify all control points in the processing of material and for storage of new material as well as finished product.

The following is an outline indicating the basic records and procedures present in the USAEC Nuclear Company fuel element fabrication area.

One of the prime functions of the Nuclear Materials Control Group is to establish a double entry accounting system for recording receipts, removals, transfers, inventories losses of nuclear material. The following records are maintained by the group: (1) Nuclear Material General Ledger, (2) Nuclear Material Subsidiary Ledger, (3) Receipts Journal, (4) Shipping Journal and (5) Material Unaccounted For Journal. The forms used to post
these records are: (1) Off-Site Transfer Documents, (2) Material Transfer Voucher (Internal Movement), and (3) Journal Voucher.

The procedures for the fuel element manufacturing area then detail the motions necessary to control and safeguard the material in process or in storage, and the method of controlling receipts and removals.

In comparing the written procedures with the actual, the first audit step would be to use an internal control questionnaire which would contain questions such as the following:

1. Has the Nuclear Material Management Group a material flow chart?
2. Has the Nuclear Material Management Group a material records flow chart?
3. Has the Nuclear Material Management Group an organization chart?
4. Is the Nuclear Accounting Group organizationally independent of the operating and storage departments?
5. Does the group include (or have available) personnel qualified in:
   (a) Accounting? (b) Chemistry? (c) Statistics?
6. Can the group cross organizational lines to enforce policy?
7. Is the subdivision into internal material balance areas commensurate with the need for control of nuclear material?
8. Are double entry records maintained?
9. Are subsidiary accounts periodically reconciled to control accounts?
10. Are perpetual inventory records maintained?
11. Are properly designed and approved forms used for recording adjustments to prior recorded quantities?
12. Is there a system (e.g. prenumbering of forms) to account for all internal transfer forms?
13. Is receiving shipping of nuclear materials centralized?
14. Is material sampled and analyzed upon receipt or before shipment?
15. Are physical inventories performed?

After reviewing the various procedures of internal control, the next step would be to test these procedures physically. Since the documentation used in receiving and shipping nuclear materials as well as transferring within a plant offers the best measurement of internal control, a statistical test of these transfer documents would be the next step of the internal audit. In examining these documents, acceptance and rejection is based upon the absolute minimum information necessary to identify adequately the material, stage of production, location, date of shipment, and acceptance of the material. Point values can be assigned to these individual characteristics such as:

Consecutiveness of document number
Date of transfer
Indication of the material balance areas involved
Material description
Quantity of material
Signatures of both shipper and receiver
Proper posting of document

---

2 These questions are a sampling taken from the Internal Control questionnaire in use within the USAEC field office nuclear materials management groups.
Using established point values, we can now choose the minimum number for an acceptable document.

Once the internal control procedures have been tested, the next step would be a physical inventory. In inventorying the nuclear material in the fabrication area, we must consider the various stages of production. The feed material may be in the form of oxides, carbides, metal, or metal alloy. In most of these forms, physical inventorying would consist of weighing and analysing isotopically as well as for the total uranium or plutonium. The alloy would present some problems since destructive analysis is not usually practical or economical. This same problem would exist on the in process or finished element material. Non-destructive analyses (gamma-scanning) would have to be performed where applicable. This is rather limited but it certainly has an application where we are fabricating plates or tubes. As was discussed earlier in this paper, the monetary value of this material is so great that a physical inventory is an absolute necessity. In most manufacturing situations, it is possible to verify total inventories without physically verifying the component parts. For example, in inventorying finished goods in clothing or furniture, it is not necessary to analyse the component parts completely. However, since we are interested in the contained uranium or plutonium, it becomes a primary function to attempt to analyse for these elements in the finished fuel. Gamma-scanning offers some promise in this case rather than destructively analysing a fuel element. The latter, of course, is uneconomical and not a practical solution.

After the internal audit has been completed, the means of communicating the results to managers looms as one of the largest problems. There are two basic approaches to reporting the findings. The first is the short-form report which merely states that the internal control is adequate or inadequate. Although this approach appears simple and to the point, it presupposes that management is only interested in results. This has been the subject of a most recent controversy in management groups. Managing "by exception" eliminates the need for long dissertations on items which are relatively unimportant. Therefore, the short-form report fits in with the management concept.

However, some management groups are not satisfied with a mere presentation of the final conclusion of the internal auditor. They are more interested in the facts behind the conclusions. In these cases, the internal audit group should furnish a long-form report. This report should then contain the reasons for the conclusions reached by the audit team. Basically the long-form report should include discussions on the major items of internal control (i.e.) the appropriateness of actual controls as compared to the stated; the results of the physical inventory; and evaluation of the material unaccounted for quantities.

Although top management is primarily concerned with this report, other groups within the organization have a strong interest and a need for this report. The transmittal of information provided in this report may not always get to the action group in any reasonable form or time if top management were to be completely responsible for transmitting the information downward.
If the operating groups are to be completely aware of all their responsibilities, they should be provided with a copy of the internal audit report at the same time it is provided to top management.

In conclusion, the internal audit function is more than just a necessary evil in the nuclear material industry. It is an absolute necessity in order to keep management informed of the status of their extremely high monetary investment.

**DISCUSSION**

J. SORNEIN: You said that the ideal team for the management of basic materials should consist of three professionals of varying skills, namely: an auditor, a chemist and a statistician. There is sometimes a certain ambiguity in regard to the precise qualifications of a "statistician". A statistician may be an accountant capable of keeping "statistical" records or he may be an engineer familiar with probability calculations; alternatively, his qualifications may lie anywhere between these two extremes. What exactly are the qualifications of the statistician to whom you refer?

S. KOPS: I define a "statistician" as a person specialized in the mathematical statistics field.

R. D. WALTON: In your paper you illustrate the importance of nuclear materials management by speaking of a contractor who got into difficulties because he was not fully aware of the financial value of the nuclear materials he was handling. On the other hand, the information you say should be given for nuclear materials transfers does not include monetary value. I wonder if you find this consistent?

S. KOPS: Since in the United States all transfers are recorded, regardless of monetary value, my discussion of monetary value may not necessarily be consistent, but it does correspond to the facts. We cannot always handle things in an ideal manner.

C. L. A. BUECHLER: Do you think that unannounced audits are impractical for all types of plant using nuclear materials? Might they not be practical, or even desirable, for plants which operate in a regular fashion, for example nuclear power plants in routine operation?

S. KOPS: Although I have an aversion to unannounced inspection, I believe it is practical in the case of continuous operations such as reactor operation.
ORGANISATION DE LA COMPTABILITE DES MATIERES DE BASE DANS UNE USINE DE FABRICATION D'ELEMENTS COMBUSTIBLES

J. NADAL, SOCIETE INDUSTRIELLE DES COMBUSTIBLES NUCLEAIRES, ANNECY, ET
J. PINOCHET, COMMISSARIAT A L'ENERGIE ATOMIQUE, SACLAY, FRANCE

Abstract — Résumé — Аннотация — Resumen

ORGANIZATION OF BASIC MATERIALS AUDITING IN A FUEL-ELEMENT FABRICATION PLANT. The authors describe the organization of auditing at the Annecy plant of the Société industrielle des combustibles nucléaires which produces, principally from natural uranium provided by the CEA, fuel elements for reactors of the graphite-gas type (in particular, G1, G2, G3, EDF1, EDF2, EDF3).

The plant may be schematically divided into five large sections: the melting shop and its annexes; machine shop; cladding shop; turnings recovery shop; warehouse. The auditing records used at Annecy may be classified in three categories: "The "follow-through" slips, the stock slips and the despatch notes.

The balance sheet established each month for uranium consists of two parts: one part showing stocks existing at the beginning and end of the month and internal and external movements during the month, and a recapitulation balance, which is so to say an account of the use of uranium, showing in parallel the amounts of uranium used and the production. The debit balance of this account corresponds to real or apparent losses.

ORGANISATION DE LA COMPTABILITE DES MATIERES DE BASE DANS UNE USINE DE FABRICATION D'ELEMENTS COMBUSTIBLES. Les auteurs décrivent l'organisation comptable de l'usine d'Annecy, qui appartient à la Société industrielle des combustibles nucléaires et fabrique essentiellement, à partir d'uranium naturel fourni par le CEA, des éléments combustibles destinés aux réacteurs de la filière graphite-gaz (G1, G2, G3, EDF1, EDF2, EDF3 notamment).

L'usine peut être divisée de façon schématique en cinq grands secteurs: l'atelier de fusion et ses annexes; l'atelier d'usinage; l'atelier de gainage; l'atelier de récupération des copeaux; le magasin. Les documents comptables utilisés à Annecy peuvent être classés en trois catégories: les fiches suiveuses, les fiches de stock et les bordereaux d'expédition.

Le bilan uranium établi chaque mois comporte deux parties: une partie où sont indiqués les stocks existant au début et à la fin du mois et les mouvements internes et externes intervenus au cours du mois, et un bilan récapitulatif, qui est en quelque sorte un compte d'exploitation uranium mettant en parallèle les quantités d'uranium consommé d'une part, les productions d'autre part. Le solde débiteur de ce compte correspond aux pertes réelles ou fictives.

ОРГАНИЗАЦИЯ УЧЕТА ОСНОВНЫХ МАТЕРИАЛОВ НА ЗАВОДЕ ТОПЛИВНЫХ ЭЛЕМЕНТОВ. Авторы описывают организацию учета на заводе топливных элементов в Аннеси, где из природного урана, поставляемого КАЭ, изготавливаются главным образом топливные элементы для реакторов системы графит-газ (в частности, G1, G2, G3, EDF1, EDF2, EDF3).

Схематично этот завод можно разделить на пять больших секторов: установка для синтеза и её подсобные помещения; цех механической обработки; цех покрытия; цех регенерации стружки; хранение.

Учетные документы, применяемые в Аннеси, можно разделить на три категории: сопроводительные карточки, карточки хранения и накладные.

Баланс, составляемый каждый месяц для урана, состоит из двух частей. В первой части указывается количество запаса, существующего в начале и конце месяца, а также внутренние и внешние передвижения в течение данного месяца. Вторая часть, именуемая суммирующим балансом, служит как бы учетом использования урана, причем параллельно указываются объем использованного урана и объем продукции. Дебетовое сальдо этого учета соответствует реальным или кажущимся потерям.
ORGANIZACION DE LA CONTABILIDAD DE LAS MATERIAS BASICAS EN UNA FABRICA DE ELEMENTOS COMBUSTIBLES. Los autores describen la organización contable de la fábrica de Annecy, que pertenece a la Sociedad Industrial de Combustibles Nucleares y produce principalmente, a partir de uranio natural suministrado por la Comisión de Energía Atómica, elementos combustibles destinados a los reactores de la serie grafito-gas (G1, G2, G3, EDF1, EDF2, EDF3 en particular).

En forma esquemática, la fábrica puede dividirse en cinco grandes sectores: el taller de fusión y sus anexos, el taller de fabricación, el taller de envainado, el taller de recuperación de virutas y el almacén. Los documentos contables utilizados en Annecy se pueden clasificar en las tres categorías siguientes: las fichas de acompañamiento, las fichas de existencias y las notas de expedición.

El balance mensual de uranio consta de dos partes: la primera, donde se indican las existencias a principio y a fin de mes, así como los movimientos internos y externos realizados en el curso del mes, y un balance de recapitulación, que es en cierto modo una cuenta de explotación del uranio en la que se computan paralelamente las cantidades de uranio consumido, por un lado, y las producciones, por otro. El saldo deudor de esta cuenta expresa las pérdidas reales o ficticias.

INTRODUCTION

A. Généralités

L'organisation comptable qui va être décrite est celle de l'usine d'Annecy, appartenant à la Société industrielle des combustibles nucléaires (SICN). Elle fabrique essentiellement, à partir d'uranium naturel fourni par le CEA, des éléments combustibles destinés aux réacteurs de la filière graphite-gaz (G1, G2, G3, EDF1, EDF2, EDF3 notamment).

L'usine peut être divisée, de façon schématique, en cinq grands secteurs (voir figure 1):
- l'atelier de fusion et ses annexes, où sont effectués les coulées, le redressage, le traitement thermique et le contrôle par gammagraphie des tubes et barreaux d'uranium;
- l'atelier d'usinage, où les tubes et barreaux sont mis à la dimension voulue, usinés, dégraissés, sablés, étuvés;
- l'atelier de gainage, où sont effectués la mise en gaine, le soudage des bouchons de la gaine, enfin le gainage proprement dit, gainage hydraulique puis gainage thermo-pneumatique à chaud;
- l'atelier de récupération des copeaux;
- le magasin.

Pour le comptable des matières de base, le secteur le plus intéressant est sans aucun doute le magasin. D'une part parce que c'est là que sont tenus la plupart des documents comptables utilisés, mais surtout parce qu'une visite du magasin permet de découvrir rapidement quelles sont les différentes formes sous lesquelles peut apparaître l'uranium existant dans une usine de fabrication d'éléments combustibles. Le magasin est en effet divisé en plusieurs cellules dont chacune est utilisée pour le stockage de l'uranium apparaissant sous une forme déterminée. Il y a une cellule pour les lingots d'uranium arrivant des usines de raffinage, une cellule pour les cartouches, une cellule pour les produits semi-finis (barreaux ou tubes), enfin plusieurs cellules affectées au stockage des déchets.
FIG. 1. Schéma de la chaîne de fabrication
B. Classification des déchets

La fabrication des éléments combustibles entraîne l'apparition de déchets que l'on peut classer en quatre catégories:

- les déchets dits «directement récupérables», qui peuvent être utilisés, après un simple dégraissage, pour la constitution d'une autre charge de fusion (il s'agit essentiellement des têtes de barreaux ou de tubes, et des barreaux ou tubes rebutés en cours de fabrication);

- les copeaux qui apparaissent lors de l'usinage des tubes ou barreaux. Ils ne peuvent être réutilisés qu'après avoir été transformés en billettes de compactage qui sont elles-mêmes refondues pour obtenir des billettes de compléments copeaux. Seules ces dernières sont réutilisées pour la constitution de nouvelles charges de fusion destinées à obtenir un produit fini;

- des déchets dits «à retraiter», qui ne peuvent être récupérés sur place et sont envoyés dans une usine de retraitement. Ces déchets comprennent:
  - les copeaux qui n'ont pu, exceptionnellement, être récupérés à l'atelier de compactage,
  - l'uranium pollué, parce qu'il adhère à d'autres matières (graphite notamment) et ne peut en être séparé que par traitement chimique,
  - les déchets contenus dans les creusets. Ces déchets apparaissent sous la forme de déchets actifs. Les creusets sont, en effet, désactivés pendant cinq mois puis réusinés. Les déchets qu'ils contiennent sont alors récupérés sous forme de galettes et de poussières qui sont envoyées aux usines de retraitement;

- les déchets non récupérables définitivement perdus et qui n'ont aucune valeur, telles les boues de sablage et les poussières d'uranium contenues dans les filtres (leur teneur en uranium est au demeurant très faible, moins de 1‰). Ils sont envoyés sur les lieux de stockage définitifs rigoureusement étanches.

Cette description rapide des différentes formes sous lesquelles peut apparaître l'uranium dans une usine d'éléments combustibles permet de mesurer les difficultés qui ont dû être surmontées lors de la mise en place d'un système comptable ayant pour but d'enregistrer les mouvements de l'uranium entre les différents secteurs de l'usine. Les techniciens qui ont imaginé ce système ont également voulu que soit réalisé, grâce à lui, un enregistrement des conditions techniques dans lesquelles se déroulent les opérations concourant à l'élaboration des éléments combustibles.

Pour mettre à exécution ce double objet, il a été nécessaire de rattacher tous les documents comptables correspondant aux opérations réalisées à l'intérieur de l'usine à l'opération qui marque le début de la fabrication: la coulée. Un numéro d'ordre est attribué à chaque coulée et ce numéro est reproduit sur tous les documents établis lors de l'exécution des opérations.

Après avoir étudié les différents documents comptables utilisés, on précisera comment ils sont exploités pour l'établissement du bilan.
I. ETUDE DES DIFFERENTS DOCUMENTS COMPTABLES UTILISES

Les documents comptables utilisés à Annecy peuvent être classés en trois catégories:
- les fiches suivues, qui accompagnent l'uranium dans ses mouvements entre les secteurs de l'usine. Elles sont très variées et il sera, dès lors, nécessaire de décrire chacune d'entre elles. Leur circuit est représenté sur la figure 2;
- les fiches de stock, qui sont très nombreuses mais toutes conçues de la même manière;
- les bordereaux d'expédition, qui sont établis lorsqu'il y a transfert d'uranium entre l'usine d'Annecy et une entreprise extérieure.

1. La fiche de fusion

Elle est établie par le magasin lors de la préparation de la charge. Les indications suivantes y sont alors inscrites:
- numéro de la coulée en préparation,
- numéro et poids des lingots composant cette charge,
- poids des déchets récupérables composant cette charge (le numéro de la coulée d'origine de ces déchets et leur teneur en éléments d'addition sont également indiqués).

La fiche de fusion est envoyée au bureau technique de contrôle pour le calcul des éléments d'addition. Elle accompagne ensuite la charge à l'atelier de fusion. Après démoulage, la fiche de fusion est complétée par les indications suivantes:
- nombre et poids des barreaux obtenus,
- poids des déchets directement récupérables obtenus,
- poids des déchets pollués obtenus,
- poids des déchets actifs obtenus (c'est-à-dire différence entre le poids des creusets avant et après fusion),
- pertes résiduelles (le poids indiqué correspond à la différence entre le poids de la charge avant fusion et le poids total des produits obtenus).

La fiche de fusion est envoyée à la comptabilité qui enregistre les résultats de l'opération fusion. Elle est transmise ensuite au bureau technique de contrôle où elle est exploitée et archivée.

2. La fiche de résultats de fusion

Elle est établie par l'atelier de fusion. Les indications suivantes y sont inscrites:
- numéro de la coulée,
- nombre et poids des barreaux démoulés obtenus,
- poids des déchets directement récupérables obtenus,
- poids des déchets pollués obtenus,
- poids des déchets actifs.
Elle accompagne les déchets lors de leur transfert au magasin.
FIG. 2. Circuit des pièces comptables
3. La fiche d'usinage

Elle est établie par l'atelier de fusion. Les indications suivantes y sont inscrites:
- numéro de la coulée,
- poids total et nombre des barreaux bruts obtenus.
Elle suit les barreaux bruts au magasin puis à l'atelier d'usinage, où elle est complétée par les indications suivantes:
- poids des déchets directement récupérables obtenus par sciage (têtes ou barreaux mauvais),
- poids des barreaux rebutés non sciés,
- poids de chacun des barreaux bons (ou, plus exactement, écart pour chacun de ces barreaux par rapport au poids théorique);
- après étuvage, la fiche est complétée par l'indication du poids des barreaux rebutés lors de cette opération.
Elle est ensuite envoyée au bureau de contrôle technique.

4. La fiche de déchets d'usinage

Elle est établie par l'atelier d'usinage lors de l'apparition des déchets directement récupérables. Il peut donc y avoir plusieurs fiches de déchets d'usinage pour une même coulée. Il y en a généralement au moins deux: une qui est établie lors de l'usinage proprement dit, une autre, après étuvage, pour les barreaux qui sont rebutés à la suite de cette opération.
La fiche de déchets d'usinage accompagne les déchets récupérables au dégraissage d'abord, au magasin ensuite. Elle comporte les indications suivantes:
- numéro de la coulée,
- poids des déchets.

5. La fiche de copeaux et bon de mouvement des copeaux

Ils sont établis tous deux par l'atelier d'usinage:
- la fiche de copeaux, lorsque les copeaux sont envoyés au magasin puis dans une usine de retraitement (Le Bouchet ou Malvési),
- le bon de mouvement des copeaux, lorsque les copeaux sont envoyés à l'atelier de récupération des copeaux.
L'une et l'autre comportent les indications suivantes:
- nature de l'uranium,
- poids des copeaux (imprégnés d'huile).
La fiche de copeaux accompagne ces derniers au magasin. Le bon de mouvement accompagne les copeaux à l'atelier de récupération des copeaux.

6. La fiche de compactage des copeaux

Elle est établie par l'atelier de compactage et comporte les indications suivantes:
- poids des copeaux mouillés utilisés,
- poids de la billette compactée obtenue.
7. La fiche de refonte des copeaux et fiche de résultats de la refonte des copeaux

La fiche de refonte des copeaux est établie par l’atelier des copeaux. Elle comporte les indications suivantes:
- poids des billettes de compactage,
- numéro de la coulée.
Elle suit les billettes de compactage à l’atelier de fusion qui la complète et, avant de l’adresser au bureau de contrôle technique, y inscrit les résultats de l’opération de refusion:
- poids des billettes de complément copeaux obtenues,
- poids des déchets directement récupérables obtenus,
- poids des déchets pollués obtenus,
- poids des déchets actifs obtenus,
- pertes résiduelles.

La fiche de résultats de la refonte des copeaux établie par l’atelier de fusion est l’homologue de la fiche de résultats de fusion. Elle accompagne au magasin les billettes de complément copeaux et les déchets obtenus.

8. La fiche individuelle du barreau

Elle est établie par l’atelier de fusion, et comporte les indications suivantes:
- numéro de la coulée,
- numéro du barreau.
Elle comporte également divers renseignements permettant d’identifier le combustible (réacteur, nature de l’uranium, alliage, forme géométrique etc.); elle suit le barreau à l’atelier d’usinage.
Si le barreau est reconnu bon après usinage, l’atelier d’usinage indique l’écart du poids du barreau par rapport au poids théorique et le signe de cet écart. Si le barreau est rebuté, on indique le motif du rebut.
La fiche individuelle du barreau accompagne le barreau jusqu’au terme du circuit de production; elle est ensuite envoyée au bureau de contrôle technique. Les fiches des barreaux rebutés sont envoyées directement au bureau de contrôle technique.

9. La fiche individuelle de la cartouche

Elle est établie par l’atelier de gainage. Outre le numéro de la cartouche, elle comporte essentiellement des indications techniques permettant de retrouver:
- quelles ont été les conditions de réalisation des opérations de soudage et de gainage,
- quels ont été les matériaux (gaines et bouchons) utilisés,
- quelles sont les causes du rebut éventuel.
Elle est envoyée avec la fiche individuelle du barreau au bureau de contrôle technique.
10. Les fiches de stock

Elles sont établies par le magasin. Pour chaque produit il est créé une fiche « entrée » et une fiche « sortie ». Le stock existant est obtenu par différence.

Un produit est identifié par :
- sa nature : lingot, déchets récupérables ou non, barreaux, tubes, cartouches,
- la teneur isotopique de l'uranium,
- le poids (pour les lingots),
- le réacteur (pour les cartouches, les barreaux et tubes),
- l'alliage,
- la géométrie.

11. Les bordereaux d'expédition

Ils sont établis, soit par l'expéditeur du produit reçu par la SICN, soit par le magasin lorsque l'expédition est faite par l'usine d'Annecy. Ces bordereaux indiquent de façon précise :
- la nature du produit expédié (lingot, tube, barreaux, cartouches)
- le poids brut et le poids d'uranium contenu dans le produit expédié.

II. LE BILAN

Le bilan uranium établi chaque mois comporte deux parties :
- une première partie où sont indiqués les stocks existant au début et à la fin du mois, les mouvements internes et externes intervenus au cours du mois. Une distinction y est opérée entre les postes suivants : lingots, cartouches, volant de fabrication, billettes de compactage et billettes de complément copeaux, déchets récupérables par fusion directe, déchets à retraiter, copeaux à compacter,
- une deuxième partie, dite bilan récapitulatif, qui est en quelque sorte un compte d'exploitation uranium mettant en parallèle les quantités d'uranium consommé d'une part, les productions d'autre part. Le solde débité de ce compte correspond aux pertes réelles ou fictives. Un état annexe donne une analyse détaillée de ces pertes. Un autre état annexe précise les résultats des opérations de récupération de copeaux réalisées au cours du mois.

A. Les différents postes du bilan

1. Le poste « lingots »

a) Le stock au 1er du mois. Le chiffre indiqué pour chaque type de lingot est obtenu en dépouillement des fiches de stock, détenues par le magasin.

b) Les entrées. Les chiffres indiqués peuvent être obtenus, soit par le dépouillement des fiches de stock du magasin, soit en totalisant les bordereaux d'expédition accompagnant les quantités reçues au cours du mois.
c) Les «utilisés en fabrications». Les chiffres indiqués sont obtenus, soit en totalisant les renseignements correspondants indiqués sur les fiches de fusion lors de la constitution de la charge, soit par dépouillement des fiches de stock «sorties» tenues au magasin.

d) Les sorties. Les chiffres indiqués peuvent être obtenus, soit par le dépouillement des fiches de stock du magasin, soit en totalisant les bordereaux d'expédition établis par l'usine lors de l'envoi.

e) Le stock en fin de mois. Les chiffres indiqués sont obtenus en dépouillant les fiches de stock du magasin. Ils doivent normalement correspondre pour chaque catégorie de lingot au résultat de \((a + b - c - d)\).

2. Le poste «cartouches»

a) Le stock au 1er jour du mois. Le chiffre indiqué, pour chaque type de cartouche, est obtenu par dépouillement des fiches de stock détenues par le magasin.

b) Les entrées. Le chiffre indiqué, pour chaque type de cartouche, est obtenu, soit par dépouillement des bordereaux d'expédition accompagnant les cartouches reçues au cours du mois, soit par dépouillement des fiches de stock «entrées» tenues au magasin.

c) Les productions. Ne sont considérées comme produites que les cartouches acceptées par le CEA. Le chiffre indiqué au bilan à ce titre est obtenu par dépouillement des procès-verbaux de recette établis par le représentant du CEA. Les fiches de stock tenues en magasin permettent de vérifier l'exactitude des résultats obtenus.

d) Les «utilisés en fabrication». Le chiffre indiqué est obtenu par dépouillement des fiches de stock «sorties» tenues au magasin.

e) Les sorties. Le chiffre indiqué est obtenu soit par dépouillement des bordereaux d'expédition établis par la SICN, soit par dépouillement des fiches de stock «sorties».

f) Le stock en fin de mois. Le chiffre indiqué est obtenu par dépouillement des fiches de stock tenues au magasin.

3. Le volant de fabrication

a) Le stock au 1er jour du mois (ou à la fin du mois.) Il ne peut être obtenu que par un inventaire physique qui est fait dans chaque atelier le dernier jour du mois. Cet inventaire permet de déterminer le poids des produits semi-finis se trouvant dans les ateliers. Il convient d'y ajouter le poids des produits semi-finis se trouvant en magasin (poids obtenu par dépouillement des fiches de stock).

b) Les entrées. Le chiffre indiqué est obtenu par dépouillement des bordereaux d'expédition qui accompagnent les envois de produits semi-finis (barreau ou tube).

c) Les sorties. Le chiffre indiqué est également obtenu par dépouillement des bordereaux d'expédition établis par la SICN lors de l'envoi des produits semi-finis.
4. Les déchets directement récupérables

a) Stocks en début ou en fin de mois. Le dépouillement des fiches de stock tenues au magasin permet de déterminer le stock en magasin. A ce stock est ajouté celui se trouvant dans les ateliers, qui est déterminé lors de l'inventaire physique fait en fin de mois.

b) Les entrées. Le chiffre indiqué est obtenu, soit par dépouillement des bordereaux d'expédition qui accompagnent les déchets, soit par dépouillement des fiches de stock «entrées» tenues au magasin.

c) Les sorties. Le chiffre indiqué est obtenu, soit par dépouillement des bordereaux d'expédition établis par la SICN lors de l'envoi des déchets, soit par dépouillement des fiches de stock «sorties» tenues au magasin.

d) Les productions. Le chiffre indiqué est obtenu par dépouillement des fiches de résultats de fusion, déchets d'usinage. A ce chiffre il convient d'ajouter le poids des déchets se trouvant dans les ateliers en fin de mois, qui est déterminé lors de l'inventaire.

e) Les «utilisés en fabrication». Le chiffre indiqué est obtenu, soit par dépouillement de la fiche de fusion, soit par dépouillement des fiches de stock «sorties» tenues au magasin.

5. Les déchets à retraiter

Ce poste du bilan ne comprend, parmi les déchets à retraiter, que les déchets pollués et les copeaux à retraiter. Les déchets actifs sont considérés provisoirement comme perdus.

a) Les stocks au début et à la fin du mois. Le chiffre indiqué, tant pour les copeaux à retraiter dans une usine extérieure que pour l'uranium pollué, est obtenu par dépouillement des fiches de stock.

b) Les sorties. Les chiffres indiqués sont obtenus, soit par dépouillement des bordereaux d'expédition établis par la SICN lors de l'envoi des déchets vers les usines de retraitement, soit par dépouillement des fiches de stock.

c) Les productions
   - Uranium pollué. Le chiffre indiqué est obtenu par dépouillement des fiches résultats de fusion.
   - Copeaux à retraiter. Le chiffre indiqué est obtenu, soit par différence entre le stock en fin de mois et les sorties d'une part, le stock en début de mois d'autre part, soit par dépouillement des fiches de copeaux établies au cours du mois.

6. Les copeaux à compacter

a) Les stocks au début et en fin de mois. Ils sont déterminés par un inventaire physique fait à l'atelier de compactage le dernier jour du mois.

b) Les productions. Le chiffre indiqué est obtenu par dépouillement des bons de mouvement des copeaux.

c) Les «utilisés en fabrication». Le chiffre indiqué est obtenu par dépouillement des fiches de compactage des copeaux.
7. Les billettes de compactage

Le dépouillement des fiches de refonte des copeaux permet de déterminer à la fois les productions et les «utilisés en fabrication». Les billettes de compactage sont, en effet, refondues immédiatement.

8. Les billettes de compléments copeaux

a) Les stocks en début et en fin de mois. Les chiffres indiqués sont obtenus par dépouillement des fiches de stock tenues au magasin.

b) Les productions. Le chiffre indiqué est obtenu par dépouillement des fiches de refonte des copeaux.

c) Les «utilisés en fabrication». Le chiffre indiqué est obtenu par dépouillement des fiches de contrôle de fusion ou des fiches de stock «sorties» tenues au magasin.

B. Le bilan récapitulatif

1. Le compte d'exploitation uranium

Au débit de ce compte on inscrit:
- le poids total des lingots utilisés en fabrication,
- le poids total des déchets directement récupérables utilisés en fabrication,
- le poids total des billettes de complément copeaux utilisées en fabrication,
- le poids total des cartouches utilisées en fabrication,
- la différence entre le poids total du volant de fabrication au début du mois et à la fin du mois. Cette différence doit être, soit ajoutée (s'il y a diminution du stock), soit retranchée (s'il y a augmentation du stock).

La somme de ces chiffres correspond au poids total de l'uranium consommé.

Au crédit de ce compte on inscrit:
- le poids total des cartouches produites,
- le poids total des déchets directement récupérables produits,
- le poids total des déchets pollués produits,
- le poids des copeaux à retraiter produits,
- le poids total des billettes de compactage produites.

La somme de ces chiffres correspond au poids total des produits obtenus.

2. L'état détaillé des pertes

La différence entre le poids de l'uranium consommé et le poids des produits obtenus correspond:
- aux pertes résiduelles dont le montant est déterminé par dépouillement des fiches de contrôle de fusion,
- à la différence constatée sur le poids des lingots1 également déterminée par dépouillement des fiches de contrôle de fusion.

1 Les lingots sont pesés lors de la constitution de chaque charge, mais le poids retenu comme utilisé en fabrication est celui qui figure sur les fiches de stock du magasin.
- à la production de déchets actifs également déterminée par dépouillement des fiches de contrôle de fusion,
- aux pertes d'usinage, aux pertes fictives dues à l'évaluation des déchets, aux pertes fictives dues à l'évaluation des cartouches, aux pertes dues au compactage des copeaux.

Le total des pertes indiqué par le solde débiteur du compte d'exploitation uranium est un chiffre provisoire. Il doit être rectifié lorsque sont connues:
- les quantités d'uranium contenu dans les déchets actifs,
- les quantités d'uranium obtenues après retraitement des déchets à retraiter.

Cette correction ne peut être faite qu'avec un certain retard et d'une façon globale pour une période d'un an.

3. Le bilan de récupération des copeaux

Le bilan copeaux est en quelque sorte un tableau qui récapitule tous les renseignements inscrits sur les bons de mouvement des copeaux, les fiches de compactage des copeaux et les fiches de refonte des copeaux:
- poids des copeaux envoyés à l'atelier de compactage,
- poids des billettes de compactage obtenues,
- poids des billettes de complément copeaux obtenues,
- poids des déchets directement récupérables obtenus,
- poids des déchets obtenus (uranium pollué),
- poids des déchets actifs obtenus,
- pertes résiduelles.

CONCLUSION

Le système comptable qui vient d'être décrit et qui fonctionne depuis plusieurs années présente toutefois certains inconvénients, tant sur le plan comptable proprement dit que sur le plan technique.

D'une part, en effet, un inventaire physique est nécessaire pour déterminer le poids des copeaux à compacter et du volant de fabrication en stock à la fin du mois. Cet inventaire devrait avoir pour seule raison d'être le contrôle de l'exactitude des renseignements qui devraient être fournis par des documents comptables.

De plus, ces documents sont très nombreux et leur dépouillement ne peut être fait qu'avec un certain délai. Le système en vigueur ne permet pas, d'autre part, de disposer des informations techniques recherchées avant l'achèvement du processus de fabrication. Il ne permet pas non plus

---

2 Les copeaux envoyés au magasin sont pesés alors qu'ils sont imprégnés d'huile. Le poids d'uranium contenu est évalué à 86% du poids des copeaux mouillés. Ce taux a été déterminé à partir de renseignements statistiques établis par les usines de retraitement.

3 Le poids total des cartouches produites est déterminé à partir du poids théorique de chaque type de combustible (nombre de cartouches x poids théorique), qui a été déterminé à la suite d'études statistiques.
de situer rapidement les rebuts prononcés aux différents stades de fabrication.

Pour remédier à ces inconvénients et aussi pour faciliter la gestion de la production des éléments combustibles, on a étudié un autre système dont les caractéristiques essentielles seraient les suivantes:

- division, sur le plan comptable, de l'usine en plusieurs secteurs correspondant chacun à un stade de la fabrication,
- enregistrement journalier des mouvements entre chaque secteur, un bon distinct étant établi pour chaque produit,
- exploitation mécanographique des renseignements indiqués sur les bons.
MATERIALS CONTROL IN THE FABRICATION OF ENRICHED URANIUM FUELS

R.G. CARDWELL, Jr.
OAK RIDGE NATIONAL LABORATORY, OAK RIDGE, TENN., UNITED STATES OF AMERICA

Abstract — Résumé — Аннотация — Resumen

MATERIALS CONTROL IN THE FABRICATION OF ENRICHED URANIUM FUELS. Intense activity in the field of fuel element technology at Oak Ridge National Laboratory during the past 15 years has led to the establishment of sound process and enriched material control procedures that find wide applicability in the commercial fabrication of fuel elements today. Reliable techniques for handling enriched fuel in alloy, dispersion and bulk oxide form were developed and adopted as standards in the course of design and fabrication of prototype fuel elements for start-up operation of the MTR, Bulk Shielding or "Swimming Pool" Reactor, Army Package Power Reactor, Tower Shielding Reactor, Geneva Conference Display Reactor, High Flux Isotope Reactor, and the EGCR. The experience gained serves as background for this paper, which will stress material control problems and their solution during the fabrication of various types of enriched uranium fuel components.

The basic objective to be met in the design of a good materials control system are: (1) minimizing the number of material units to be accounted for; (2) designing separate records for each major fabrication step and linking these in a manner that permits isolation of differences with a minimum of effort; (3) integrating the maximum number of controls into the minimum number of records to eliminate duplication; and (4) introducing a sufficient number of cross-checks into the system to ensure reliability. In every fabrication programme, successful control was achieved by establishing a unit procedure in the following areas: (1) starting materials in the as-received form; (2) fabrication of components; (3) component processing; and (4) scrap handling. Consolidation of control records into a master summary was helpful in confirming the materials inventory, evaluating the fabrication process, and preparing management reports.

Establishment of sampling methods and examination of results indicated that multiple control is necessary to ensure proper fuel content. Mechanical adjustment and density measurement were successfully used where alloy fuel content was critical. Scrap handling had an important effect on the materials balance, by which fuel content was confirmed and good accountability was assured. Records and handling procedures, including batching and physical marking methods, were formulated in a manner that assisted the fabricator in criticality control.

CONTROLE DES MATIERES AU COURS DE LA FABRICATION DES COMBUSTIBLES A BASE D'URANIUM ENRICHI. Grâce aux efforts intenses qui ont été accomplis au cours des 15 dernières années dans le domaine de la technologie des éléments de combustible par le Laboratoire national d'Oak Ridge, il a été possible d'établir des méthodes rationnelles de fabrication et de contrôle des combustibles enrichis, qui trouvent une large application dans la fabrication industrielle des éléments de combustible à l'heure actuelle. Des techniques éprouvées de manipulation du combustible enrichi en alliages, en dispersion et sous forme d'oxyde en vrac ont été mises au point et appliquées à l'étude et à la fabrication des prototypes d'éléments combustibles utilisés pour le démarrage du réacteur d'essai de matériaux, du réacteur à protection constitué par la masse du ralentisseur ou réacteur piscine, du réacteur de puissance transportable construit par l'armée, du réacteur à protection en tour, du réacteur exposé à la Conférence de Genève, du réacteur à haut flux pour la production de radioisotopes et du réacteur expérimental refroidi par un gaz. L'expérience acquise est la base du présent mémoire qui traite essentiellement des problèmes de contrôle des matières qui se posent au cours de la fabrication de différents types d'éléments de combustible à base d'uranium enrichi et montre comment ils ont été résolus.

Les objectifs principaux d'un système rationnel de contrôle des matières sont les suivants: 1. réduire le plus possible le nombre des portes matière à contrôler; 2. établir des relevés distincts pour chacune des

* Research sponsored by the US Atomic Energy Commission under contract with the Union Carbide Corporation.
фазы основных операций и выработать режимы, чтобы в минимальной степени влиять на результаты. 2. упростить учеты и обеспечить правильность технологических процессов. 3. применять методы контроля, которые позволяют обнаруживать недостатки в минимальной степени.

Основными задачами при разработке системы учета материалов являются:
1) исходные материалы в том виде, как они получаются; 2) изготовление компонентов; 3) обращение с ломом. Сведения об учете в общую главную сводку сводя в систему достаточного числа проверок для обеспечения надежности. В каждой программе изготовления успех обеспечивал введение единовременной процедуры на следующих стадиях:
1) исходные материалы в том виде, как они получаются; 2) изготовление компонентов; 3) обращение с ломом. Сведения об учете в общую главную сводку сводя в систему достаточного числа проверок для обеспечения надежности. В каждой программе изготовления успех обеспечивал введение единовременной процедуры на следующих стадиях:
1) исходные материалы в том виде, как они получаются; 2) изготовление компонентов; 3) обращение с ломом. Сведения об учете в общую главную сводку сводя в систему достаточного числа проверок для обеспечения надежности. В каждой программе изготовления успех обеспечивал введение единовременной процедуры на следующих стадиях:
1) исходные материалы в том виде, как они получаются; 2) изготовление компонентов; 3) обращение с ломом. Сведения об учете в общую главную сводку сводя в систему достаточного числа проверок для обеспечения надежности. В каждой программе изготовления успех обеспечивал введение единовременной процедуры на следующих стадиях:
1) исходные материалы в том виде, как они получаются; 2) изготовление компонентов; 3) обращение с ломом. Сведения об учете в общую главную сводку сводя в систему достаточного числа проверок для обеспечения надежности. В каждой программе изготовления успех обеспечивал введение единовременной процедуры на следующих стадиях:
1) исходные материалы в том виде, как они получаются; 2) изготовление компонентов; 3) обращение с ломом. Сведения об учете в общую главную сводку сводя в систему достаточного числа проверок для обеспечения надежности. В каждой программе изготовления успех обеспечивал введение единовременной процедуры на следующих стадиях:
1) исходные материалы в том виде, как они получаются; 2) изготовление компонентов; 3) обращение с ломом. Сведения об учете в общую главную сводку сводя в систему достаточного числа проверок для обеспечения надежности. В каждой программе изготовления успех обеспечивал введение единовременной процедуры на следующих стадиях:
1) исходные материалы в том виде, как они получаются; 2) изготовление компонентов; 3) обращение с ломом. Сведения об учете в общую главную сводку сводя в систему достаточного числа проверок для обеспечения надежности. В каждой программе изготовления успех обеспечивал введение единовременной процедуры на следующих стадиях:
1) исходные материалы в том виде, как они получаются; 2) изготовление компонентов; 3) обращение с ломом. Сведения об учете в общую главную сводку сводя в систему достаточного числа проверок для обеспечения надежности. В каждой программе изготовления успех обеспечивал введение единовременной процедуры на следующих стадиях:
1) исходные материалы в том виде, как они получаются; 2) изготовление компонентов; 3) обращение с ломом. Сведения об учете в общую главную сводку сводя в систему достаточного числа проверок для обеспечения надежности. В каждой программе изготовления успех обеспечивал введение единовременной процедуры на следующих стадиях:
1) исходные материалы в том виде, как они получаются; 2) изготовление компонентов; 3) обращение с ломом. Сведения об учете в общую главную сводку сводя в систему достаточного числа проверок для обеспечения надежности. В каждой программе изготовления успех обеспечивал введение единовременной процедуры на следующих стадиях:
1) исходные материалы в том виде, как они получаются; 2) изготовление компонентов; 3) обращение с ломом. Сведения об учете в общую главную сводку сводя в систему достаточного числа проверок для обеспечения надежности. В каждой программе изготовления успех обеспечивал введение единовременной процедуры на следующих стадиях:
1) исходные материалы в том виде, как они получаются; 2) изготовление компонентов; 3) обращение с ломом. Сведения об учете в общую главную сводку сводя в систему достаточного числа проверок для обеспечения надежности. В каждой программе изготовления успех обеспечивал введение единовременной процедуры на следующих стадиях:
1) исходные материалы в том виде, как они получаются; 2) изготовление компонентов; 3) обращение с ломом. Сведения об учете в общую главную сводку сводя в систему достаточного числа проверок для обеспечения надежности. В каждой программе изготовления успех обеспечивал введение единовременной процедуры на следующих стадиях:
1) исходные материалы в том виде, как они получаются; 2) изготовление компонентов; 3) обращение с ломом. Сведения об учете в общую главную сводку сводя в систему достаточного числа проверок для обеспечения надежности. В каждой программе изготовления успех обеспечивал введение единовременной процедуры на следующих стадиях:
1) исходные материалы в том виде, как они получаются; 2) изготовление компонентов; 3) обращение с ломом. Сведения об учете в общую главную сводку сводя в систему достаточного числа проверок для обеспечения надежности. В каждой программе изготовления успех обеспечивал введение единовременной процедуры на следующих стадиях:
1) исходные материалы в том виде, как они получаются; 2) изготовление компонентов; 3) обращение с ломом. Сведения об учете в общую главную сводку сводя в систему достаточного числа проверок для обеспечения надежности. В каждой программе изготовления успех обеспечивал введение единовременной процедуры на следующих стадиях:
1) исходные материалы в том виде, как они получаются; 2) изготовление компонентов; 3) обращение с ломом. Сведения об учете в общую главную сводку сводя в систему достаточного числа проверок для обеспечения надежности. В каждой программе изготовления успех обеспечивал введение единовременной процедуры на следующих стадиях:
1) исходные материалы в том виде, как они получаются; 2) изготовление компонентов; 3) обращение с ломом. Сведения об учете в общую главную сводку сводя в систему достаточного числа проверок для обеспечения надежности. В каждой программе изготовления успех обеспечивал введение единовременной процедуры на следующих стадиях:
1) исходные материалы в том виде, как они получаются; 2) изготовление компонентов; 3) обращение с ломом. Сведения об учете в общую главную сводку сводя в систему достаточного числа проверок для обеспечения надежности. В каждой программе изготовления успех обеспечивал введение единовременной процедуры на следующих стадиях:
1) исходные материалы в том виде, как они получаются; 2) изготовление компонентов; 3) обращение с ломом. Сведения об учете в общую главную сводку сводя в систему достаточного числа проверок для обеспечения надежности. В каждой программе изготовления успех обеспечивал введение единовременной процедуры на следующих стадиях:
1) исходные материалы в том виде, как они получаются; 2) изготовление компонентов; 3) обращение с ломом. Сведения об учете в общую главную сводку сводя в систему достаточного числа проверок для обеспечения надежности. В каждой программе изготовления успех обеспечивал введение единовременной процедуры на следующих стадиях:
1) исходные материалы в том виде, как они получаются; 2) изготовление компонентов; 3) обращение с ломом. Сведения об учете в общую главную сводку сводя в систему достаточного числа проверок для обеспечения надежности. В каждой программе изготовления успех обеспечивал введение единовременной процедуры на следующих стадиях:
1) исходные материалы в том виде, как они получаются; 2) изготовление компонентов; 3) обращение с ломом. Сведения об учете в общую главную сводку сводя в систему достаточного числа проверок для обеспечения надежности. В каждой программе изготовления успех обеспечивал введение единовременной процедуры на следующих стадиях:
1) исходные материалы в том виде, как они получаются; 2) изготовление компонентов; 3) обращение с ломом. Сведения об учете в общую главную сводку сводя в систему достаточного числа проверок для обеспечения надежности. В каждой программе изготовления успех обеспечивал введение единовременной процедуры на следующих стадиях:
1) исходные материалы в том виде, как они получаются; 2) изготовление компонентов; 3) обращение с ломом. Сведения об учете в общую главную сводку сводя в систему достаточного числа проверок для обеспечения надежности. В каждой программе изготовления успех обеспечивал введение единовременной процедуры на следующих стадиях:
en marcha del reactor de ensayo de materiales (MTR), del BSR de tipo piscina, del reactor móvil de potencia del ejército, del reactor de torre de blindaje, del reactor de demostración para la Conferencia de Ginebra, del reactor de flujo elevado para la producción de isótopos y del reactor experimental refrigerado por gas. La experiencia adquirida sirve de base a esta memoria en la que se destacan los problemas de control de materiales y su solución durante la fabricación de varios tipos de componentes de combustible de uranio enriquecido.

Un buen sistema de control de materiales debe perseguir los objetivos básicos siguientes: a) reducir al mínimo el número de unidades de material que haya que contabilizar, b) establecer registros distintos para cada operación importante y relacionarlos de manera que resulte fácil encontrar las diferencias, c) integrar el mayor número posible de controles en el número mínimo de registros a fin de evitar la duplicación y d) introducir en el sistema un número de verificaciones suficiente para garantizar su seguridad. En todos los programas de fabricación se logró un control eficaz estableciendo un procedimiento unitario en las siguientes etapas: a) entrada de las materias primas, b) fabricación de componentes, c) tratamiento de los componentes y d) manipulación de la chatarra. La reunión de los registros de control en un resumen general ayudó a confirmar las existencias, a evaluar el proceso de fabricación y a preparar los informes administrativos.

El establecimiento de métodos de muestreo y el examen de los resultados pusieron de manifiesto la necesidad de un control múltiple para asegurar un contenido adecuado de combustible. Cuando el contenido de combustible en aleación revestía importancia crítica se recurrió con buenos resultados al ajuste mecánico y a la medición de la densidad. La manipulación de la chatarra tuvo una importante influencia en el balance de los materiales que confirmó el contenido de combustible y aseguró una buena contabilización. Los procedimientos de manipulación y registro, y en particular los métodos de determinación de las partidas y de marcaje físico, se formularon de tal manera que constituían una buena ayuda para el fabricante en lo que respecta al control de la criticidad.

INTRODUCTION

The scarcity, expense, and hazards of nuclear materials have often been cited as reasons for their need of proper control and management. Equally important, however, is that technical success in the manufacture of fuel elements, particularly those containing enriched uranium, depends largely upon how well these controls are fitted to each operation.

The purpose of this paper is to discuss the major control problems in the fabrication of enriched uranium fuels and their solution. The material presented is based on successful controls applied during the fabrication of a variety of fuel elements at the Oak Ridge National Laboratory, from the receipt and verification of starting materials to the process control summary performed upon completion of the fabrication effort.

Special problems in the handling of nuclear material, as opposed to conventional material, will be considered before the basic objectives in systems design are stated. After a brief description of fabricated product types, thorough discussion of enriched-uranium control procedures applicable to a general fabrication effort will be presented.

SPECIAL CONSIDERATIONS

Since enriched fuel element fabrication utilizes standard metallurgical processes and equipment, the process controls are similar to those found in any metals or ceramics industry. There are end-product specifications regarding chemical make-up, dimensions, and strength and process concerns such as weld integrity and grain size. These and other factors must be controlled within close tolerances.
In the area of materials control, however, there are few similarities between conventional and nuclear operations. Enriched uranium presents unique control problems in handling and processing. In addition to the regular health and safety hazards, the enriched-uranium fabricator handles a material that can, if not properly controlled, seriously damage his personnel and equipment. If the uranium is highly enriched, he also risks loss of a very expensive material.

**Health hazards**

During enrichment of $^{235}$U by the gaseous diffusion process, the very active $^{234}$U is also enriched. Although external damage to the human body resulting from this alpha activity is minimal, when this material is ingested or inhaled the hazard becomes acute. Therefore, we need to prevent it from entering the body.

**Criticality control**

The extreme property damage and loss of life that could result from a critical accumulation of $^{235}$U in the fabrication process area demand strong measures to ensure nuclear safety. Geometric control provides the best means of preventing a critical incident, but it must be reduced to practical terms and limits understood by the process technician. In addition, administrative practices must provide for a continuous surveillance to make certain that these limits are being adhered to. Yet, the limits must be flexible enough to permit some alteration for changes that occur in the process. Therefore the criticality expert should evaluate both the equipment and the process in setting limits.

**In-process losses**

The value of highly enriched uranium requires that careful attention be given to the smallest chances for losses, unnecessary accumulations of in-process materials, and the possibility of mixing with materials of lower enrichment. The design of process methods and equipment is very important in this respect.

Such procedures as filtering and sampling all exhausts, glove-box containment of powdered materials, and periodic burn recovery of filters should be established. There should be no open drains in the primary fabrication area, in which the uranium is first incorporated in non-fissile materials, and all spills should be cleaned up with dry, absorbent materials that can be burned for recovery.

The measurement methods, including weighing, sampling, and analysis, must be of the highest quality. The accumulation of small but regular systematic errors in measurement can falsely indicate a loss of many dollars or, more seriously, obscure actual losses.

The general problem of the nuclear materials manager, therefore, is complex and must be solved largely by co-ordination. He must design a system that will not interfere with the process objectives, yet will ensure
safe handling and protection of the materials. In addition, his system must generate sufficient data to ensure product quality, statistically evaluate the process, and enable the necessary management reports.

**BASIC OBJECTIVES**

The basic objectives of a good materials control system are: (1) minimizing the number of material units to be accounted for; (2) establishing separate records for each major fabrication step and linking these in a manner that permits isolation of differences with a minimum of effort; (3) integrating the maximum number of controls into the minimum number of records to eliminate duplication; and (4) including a sufficient number of cross checks to ensure reliability.

**Material units**

All quantity controls of enriched uranium are in terms of grams. Accounting for the materials in process is simplified, however, if the control basis is changed to a piece count of components of known uranium mass as they are fabricated. For example, when 400 g $^{235}$U is fully contained in 20 identified fuel plates, the control should be by plate rather than gram. Furthermore, when these 20 plates are brazed into a single fuel element, the control should shift to the element. In general, then, the number of units should be progressively reduced as the materials flow through the process.

**Records**

Enriched fuel materials are controlled for two purposes by a complete record around each major fabrication step. First, the fabricator can balance the quantity of uranium fuel in the starting materials of each step against the quantity in its end products to confirm the intended fuel content, evaluate any process losses, and limit the size of the process area and the number of records that must be examined should any unusual differences occur. Second, use of the final balance of each step as the initial balance of the next step in the process links all data into a complete materials control history for the entire process and provides the basis for all evaluation of product fuel content and reporting for accountability.

As previously stated, the fuel manufacturer must control other general specifications of the fabricated pieces as well as the fuel content within close tolerances. Methods for meeting such specifications include mechanical and chemical measurements and statistical evaluation of variances.

**Integration of controls**

The records needed for each fabrication step should integrate both the general and fuel-control information into a dual-purpose form, thus reducing the number of forms needed and eliminating duplications. In addition one can detect important relationships between fuel-content data and process data that might otherwise go unobserved.
Cross checks

The danger of dependence on a single measurement of fuel content during any stage of the fabrication process becomes apparent when one considers the accidental variations that can occur. Balances can be off calibration, chemical analyses can be incorrect or improperly stated, and human error is always possible. To save time and expense, however, the fabricator must minimize the number of measurements and insure himself against intolerable fuel-content deviations with sufficient reliable cross checks. These fall into four categories.

Method and equipment checks

At the Oak Ridge National Laboratory, all balances for weighing fertile and fissile material are regularly calibrated against precise standards. Each calibration is recorded on a chart kept at the balance, and significant deviations dictate equipment removal for repair or replacement.

Random and back-up sampling

The fuel content of the fabricated component may also be confirmed by chemical analysis of random components. Because the component is destroyed, this type of sampling should be limited to the minimum number of samples from which a sound statistical evaluation can be made, and the components should be removed at the earliest possible stage of their processing.

During regular process sampling, an extra back-up sample should be taken and held for a repeat measurement should later data fail to correlate with the original results.

Statistical evaluation

Trends that are apparent in a regular review of all process data will often indicate control problems that are not obvious in the day-to-day data accumulations. In one evaluation of the difference between intended and chemically analysed content of uranium-aluminium alloy over a rather long period, we noted that new heats that included remelted aluminium-clad uranium-aluminium alloy plates usually had a higher uranium-to-aluminium ratio than intended. Investigation showed that the outer cladding material of the plates, which was of different composition than the high-purity fuel diluent material, contained small amounts of copper that were titrated with the uranium during the analytical procedure. Including an analysis for copper and adjusting the uranium analysis for the copper content corrected the situation.

The material balance

One of the most important methods of cross checking the routine process controls is the material balance. A complete and thorough materials in-
Inventory is correlated with the accounting book balance at the end of a selected period. We shall discuss this in detail later.

FABRICATED PRODUCTS

The types of enriched uranium fuel elements are almost as numerous and varied as the reactors now in existence. Indeed, one often has more than one specification for elements in the same reactor system. We shall, however, consider three general categories classified according to the physical form of fuel and illustrated in Fig. 1. To expand somewhat on these categories, Table I lists a representative selection of the materials combinations now in reactor service in the United States of America or manufactured by US firms.

CONTROL PROCEDURES

The development and fabrication of the many and varied types of fuel elements over the past 15 years at Oak Ridge National Laboratory have afforded an excellent opportunity for a parallel development and testing of materials control procedures. Reliable techniques for handling enriched fuel in alloy, dispersion, and bulk oxide form were developed and, in many cases, adopted directly as standards for subsequent commercial fabrication of these elements.

In every fabrication programme, successful control was achieved by establishing a unit procedure in the areas of (1) starting material in the as-received form, (2) fabrication of components, (3) component processing, and (4) scrap handling; these were tied together into a control summary so that the entire effort could be examined and analysed.

Control of starting material

Control must begin immediately upon receipt of the uranium materials. These materials fall into two general categories: solid metals and powdered materials. Solid metals include both pure uranium and alloys, since the fabricator will sometimes avoid the melting and casting step by procuring prealloyed uranium. Powdered materials include such compounds as UO₂, UN, and U₃O₈, which are not alloyed but mechanically mixed with diluent. For bulk oxide fabrication, the fabricator may receive premixed compounds, such as UO₂-ThO₂, UO₂-BeO, UO₂-C, and Si-SiC-UO₂.

Verification

Three quantities of the material should be listed in its accompanying documents: net weight of the uranium-bearing material, total uranium contained in the material, and the total ²³⁵U based on enrichment. Verification of all three figures is important to the fabricator, not only because the materials are expensive but also because the fuel may not meet specifications.
FIG. 1. Representative fuel element types: (a) alloy fuel plate element - MTR [1]; (b) dispersion fuel element plate - APPR [2]; (c) bulk oxide rod element - power reactor [3]
<table>
<thead>
<tr>
<th>Name and Location</th>
<th>Startup</th>
<th>Cladding Material</th>
<th>% $^{235}$U Enrichment</th>
<th>Fuel Material</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bulk Shielding Reactor, Oak Ridge, Tenn.</td>
<td>1950</td>
<td>Al</td>
<td>93</td>
<td>Uranium-Aluminum Alloy</td>
</tr>
<tr>
<td>Low-Intensity Training Reactor, Oak Ridge Tenn.</td>
<td>1950</td>
<td>Al</td>
<td>93</td>
<td>Uranium-Aluminum Alloy</td>
</tr>
<tr>
<td>Materials Testing Reactor, BNNS, Idaho</td>
<td>1952</td>
<td>Al</td>
<td>93</td>
<td>Uranium-Aluminum Alloy</td>
</tr>
<tr>
<td>Stationary Medium Power Reactor (APPR) Fort Belvoir, Va.</td>
<td>1955</td>
<td>SS</td>
<td>93</td>
<td>$U_3O_8$ Dispersed in SS</td>
</tr>
<tr>
<td>Engineering Test Reactor, BNNS, Idaho</td>
<td>1957</td>
<td>Al</td>
<td>93</td>
<td>Uranium-Aluminum Alloy</td>
</tr>
<tr>
<td>Vallecitos Boiling Water Reactor, Pleasanton, Calif.</td>
<td>1957</td>
<td>SS</td>
<td>93</td>
<td>$U_3O_8$ Dispersed in SS</td>
</tr>
<tr>
<td>Shippingport Atomic Power Station, Shippingport, Pa.</td>
<td>1957</td>
<td>Zircaloy</td>
<td>93</td>
<td>Uranium-Zirconium Alloy</td>
</tr>
<tr>
<td>Oak Ridge Research Reactor, Oak Ridge, Tenn.</td>
<td>1958</td>
<td>Al</td>
<td>93</td>
<td>Uranium-Aluminum Alloy</td>
</tr>
<tr>
<td>Dresden Nuclear Power Station, Morris, Ill.</td>
<td>1959</td>
<td>Zircaloy</td>
<td>1.5</td>
<td>Pelletized $U_3O_8$</td>
</tr>
<tr>
<td>Puerto Rico Nuclear Center, Mayaguez, Puerto Rico</td>
<td>1960</td>
<td>Al</td>
<td>20</td>
<td>Pelletized $U_3O_8$</td>
</tr>
<tr>
<td>Yankee Nuclear Power Station, Howe, Mass.</td>
<td>1960</td>
<td>SS</td>
<td>3.4</td>
<td>Pelletized $U_3O_8$</td>
</tr>
<tr>
<td>Consolidated Edison Thorium Reactor, Buchanan, N. Y.</td>
<td>1962</td>
<td>SS</td>
<td>93</td>
<td>Pelletized $ThO_2-U_3O_8$</td>
</tr>
<tr>
<td>Ballem Nuclear Power Facility, Ballem, Neb.</td>
<td>1962</td>
<td>SS</td>
<td>3.6</td>
<td>Uranium Molybdenum Alloy</td>
</tr>
<tr>
<td>Elk River Reactor, Elk River, Minn.</td>
<td>1962</td>
<td>SS</td>
<td>93</td>
<td>Pelletized $ThO_2-U_3O_8$</td>
</tr>
<tr>
<td>SEWN, Gargliano River, Italy</td>
<td>1963</td>
<td>Zircaloy</td>
<td>2.0</td>
<td>Pelletized $U_3O_8$</td>
</tr>
<tr>
<td>High Flux Isotope Reactor, Oak Ridge, Tenn.</td>
<td>1965</td>
<td>Al</td>
<td>93</td>
<td>$U_3O_8$ Dispersed in Aluminum</td>
</tr>
</tbody>
</table>
Net weight is verified with balances, accurate to ± 0.5 g for the higher enrichments. Total uranium can be chemically verified by potentiometric titration analysis. Isotopic enrichment is verified by mass spectrometry. If the fabricator receives alloyed or premixed material, he must develop and use reliable sampling techniques to ensure that the uranium is both in the proper proportion with the matrix material and homogeneously dispersed.

Handling and storage

The uranium materials, received in a critically safe container as shown in Fig. 2, are removed and check-weighed on suitable balances. Metals may be placed directly on the balance. Powdered compounds, however, must remain in their inner containers on the balances or be check-weighed in a contained area, such as a dry box, to protect the operator from toxicity and prevent contamination. It is often advantageous for the fabricator to

![Diagram of a shipping container with labeled parts: COVER SHEET, FUEL BEARING ALLOY OR POWDER PRESSED CORE, FRAME PIECE, COVER SHEET.](image)
supply his vendor with permanently tared inner containers so the filled container can be check-weighed.

Control records

Materials are usually processed by the fabricator in smaller batches than received. Control of the starting materials requires knowing how much material has been received, how much has been assigned to process, and how much is on hand. The preferred initial control is the simple in-out-balance perpetual inventory. To facilitate immediate materials tracing, however, it should include references for each quantity received and the process batches to which the smaller quantities have been assigned.

Fabrication of components

"Primary fabrication" includes all stages in which the fuel materials are being processed for containment in non-fuel metals. When uranium-aluminium alloy fuel plates are fabricated, for example, the uranium is first dispersed in aluminium by melting, and the alloy is cast in a graphite mould. The ingot is then reduced in thickness by rolling. Fuel cores are punched from the rolled plate, pressed into an aluminium frame of similar thickness, and covered with aluminium plates to form a composite "sandwich" (Fig. 3). Now the primary fabrication is complete. The fuel is contained within a composite fuel plate and cannot be altered in weight or content except by destruction of the component.

Often alloy fuels are impractical or impossible to fabricate, so that the fuel core is fabricated by dispersion of a powder. For example, aluminium alloys containing more than 30% U are very difficult to fabricate, and useful concentrations of uranium cannot be incorporated in stainless steel by alloying. In the powder dispersion method, the fuel compound and matrix are formed into a fuel compact by blending, cold pressing, sintering, and coining. This method permits excellent distribution of fuel and extremely accurate accounting for the critical ingredients. Also, the fuel components have two chief advantages over the alloy: (1) greater freedom of selection of fuel and matrix material for better performance characteristics; and (2) confinement of the fission product damage to the fuel compound, so that the matrix can better carry the in-service structural load.

Bulk oxide or other ceramic compound component fabrication involves ceramic and metallurgical techniques similar to those used in the dispersion method. Although the amount of fuel material in the component must be exact, the control is relatively simple since no fuel-matrix relations are involved and, in this regard, homogeneity is no problem.

Specification

In both alloy and dispersion fabrication, one must predetermine uranium content in the core materials to obtain the desired $^{235}$U content in the final component. Aronin and Klein [4] have shown that the density method can be utilized with a high degree of accuracy in determining proper composi-
tion of uranium-aluminium alloys. Their work has shown a correlation be-
tween calculated percentage by weight and analytical results within ± 0.19% 
in alloys containing up to 35% U. However, Martin and Leitten [5], at the
Oak Ridge National Laboratory, showed that suppression of UAl₃ formation in the alloy is difficult at the higher uranium percentages and that the correlation becomes inadequate above 24 wt. % U.

When the dispersion technique is used, the predetermination becomes even more difficult because of the additional variables involved. These variables, which have a direct effect on the density of the fuel core compact, include the uranium content of the fuel compound and the size, shape, and density of both the fuel and matrix particles.

Where homogeneous fuel cores are the same size and fabricated from a single melt, the average weight method for determining fuel content in each component is very reliable. The entire batch of components from a single heat is weighed and total weight divided by their number. If an extremely close tolerance is desired, the content is determined from a density measurement of each component.

Verification

A pilot fabrication effort with liberal destructive testing of the end products will be used by the fabricator before actual production begins. This facilitates prediction of the end result with enough certainty to ensure general control during actual production and, in many cases, is one of the best methods of product verification. Homogeneity of dispersion products, for instance, is often based on integrity of the process and depends almost wholly on intelligent operation and close control during fabrication. Even though many advances have been made in the field of non-destructive testing for confirming homogeneity, fuel quantity, and other specifications [6], any such control methods remain secondary to process integrity.

In alloy fabrication, however, content may be verified by wet chemical analysis of dip samples taken from each melt before casting. The samples are very representative, and measurement at this early stage permits some adjustment of the uranium content by adjusting the thickness of the fuel compact before primary fabrication is complete.

These analyses must be evaluated to determine if the uranium content of the material is acceptable. The establishment of criteria for acceptability depends on two uncertainties in alloy content determination: (1) the disagreement between the chemical analysis and the intended composition; and (2) the limits of error in the sampling and analysis. These must be examined collectively to determine if the fuel content specification has been met. The average analytical result rather than the intended composition is depended on to correct any mechanical or human errors that have occurred in the process.

Control records

The form records used in the control of enriched uranium during the primary fabrication process differ from plant to plant even more widely than do the fabricated shapes they control. In every case, however, they should answer three basic control questions: (1) Where is the material located? (2) What is its physical shape? (3) What is its quantity in relation to the matrix materials in which it is dispersed?
### ALLOY HEAT LOG

**Heat No.**
- E-514

**Type Material**
- U Alloy

**Intended wt % U**
- 15.95

**wt % U by Avg. Chem. Analysis**
- 15.97

**Fabrication Order No.**
- 1124 R78

#### BEGINNING MATERIALS

<table>
<thead>
<tr>
<th>Type Material</th>
<th>Identification</th>
<th>Net Weight</th>
<th>Total U</th>
<th>wt % U by Avg. Chem. Analysis</th>
</tr>
</thead>
<tbody>
<tr>
<td>Uranium Metal</td>
<td>IC - 842</td>
<td>710.14</td>
<td>710.14</td>
<td>648.13</td>
</tr>
<tr>
<td>Alloy Scrap</td>
<td>E - 497</td>
<td>1654.12</td>
<td>239.02</td>
<td>215.12</td>
</tr>
<tr>
<td>Scrap Components 3 Plates</td>
<td>E - 492</td>
<td>591.22</td>
<td>25.17</td>
<td>22.65</td>
</tr>
<tr>
<td>Scrap Components 2 Plates</td>
<td>E - 494</td>
<td>393.68</td>
<td>16.74</td>
<td>15.07</td>
</tr>
<tr>
<td>Scrap Components 2 Plates</td>
<td>E - 500</td>
<td>397.10</td>
<td>20.13</td>
<td>18.12</td>
</tr>
<tr>
<td>Other Matrix  High Purity Al Metal</td>
<td>2646.22</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Other Material</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Total Materials</td>
<td></td>
<td>6402.48</td>
<td>1021.20</td>
<td>919.09</td>
</tr>
</tbody>
</table>

#### ALLOY PRODUCTS

<table>
<thead>
<tr>
<th>Alloy Billets</th>
<th>(2) E - 514</th>
<th>E - 514</th>
<th>6096.28</th>
<th>975.58</th>
<th>876.22</th>
</tr>
</thead>
<tbody>
<tr>
<td>Alloy Samples</td>
<td>#1 AR 15.99</td>
<td>0.62</td>
<td>1.06</td>
<td>0.95</td>
<td></td>
</tr>
<tr>
<td></td>
<td>#2 AR 15.96</td>
<td>6.40</td>
<td>1.02</td>
<td>0.92</td>
<td></td>
</tr>
<tr>
<td></td>
<td>#3 AR 15.93</td>
<td>4.57</td>
<td>0.73</td>
<td>0.66</td>
<td></td>
</tr>
<tr>
<td>Waste Products</td>
<td>Heat Dress</td>
<td>315.80</td>
<td>50.43</td>
<td>45.39</td>
<td></td>
</tr>
<tr>
<td>Total Alloy Products</td>
<td></td>
<td></td>
<td>6429.67</td>
<td>1026.82</td>
<td>924.14</td>
</tr>
<tr>
<td>Beginning Materials – Alloy Product Difference</td>
<td></td>
<td>+27.19</td>
<td>+5.62</td>
<td>+5.06</td>
<td></td>
</tr>
</tbody>
</table>

#### COMPONENT PRODUCTS

<table>
<thead>
<tr>
<th>No. Acceptable Pieces</th>
<th>76 Cores</th>
<th>4482.91</th>
<th>715.92</th>
<th>644.33</th>
</tr>
</thead>
<tbody>
<tr>
<td>Average Weight Piece</td>
<td>(58.9%)</td>
<td>(9.42)</td>
<td>(9.42)</td>
<td></td>
</tr>
<tr>
<td>Scrap Materials</td>
<td>After Punching</td>
<td>1611.24</td>
<td>257.32</td>
<td>231.58</td>
</tr>
<tr>
<td>Waste Materials</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Total Products</td>
<td></td>
<td>6094.15</td>
<td>973.24</td>
<td>875.91</td>
</tr>
<tr>
<td>Alloy – Component Difference</td>
<td></td>
<td>-2.13</td>
<td>-0.34</td>
<td>-0.31</td>
</tr>
</tbody>
</table>

AR – Analytical Result

---

**FIG. 4a. Typical alloy heat log**

Figure 4a illustrates a practical log that records the desired information and supplies the necessary control facts. This form is readily adaptable to other alloy fabrication, such as the casting and extrusion of rods, since the same facts are required. Note that the uranium percentage applied to the net alloy weights is the average of the analyses of the dip samples. Note also that a great deal of information has been practically condensed into a single control record and reflects the relationship of the categories in a single presentation.
The type of control record used for dispersed fuel materials is very similar (Fig. 4b). Since there are fewer steps, the record is somewhat simpler. It does, however, adequately reflect the steps and provide the necessary control information. The same form is also useful for the primary step in bulk oxide fabrication.

### BLENDED MATERIALS BATCH LOG

<table>
<thead>
<tr>
<th>Blend No.</th>
<th>Type Material</th>
<th>wt % Uranium</th>
<th>Fabrication Order No.</th>
</tr>
</thead>
<tbody>
<tr>
<td>EB-284</td>
<td>VO₂-Aluminum</td>
<td>48.00</td>
<td>1127</td>
</tr>
</tbody>
</table>

#### BEGINNING MATERIALS

<table>
<thead>
<tr>
<th>Type Material</th>
<th>Identification</th>
<th>Net Weight</th>
<th>Total U</th>
<th>U²³⁵</th>
</tr>
</thead>
<tbody>
<tr>
<td>Uranium VO₂ Powder</td>
<td>IC-927</td>
<td>1011.63</td>
<td>890.23</td>
<td>178.05</td>
</tr>
<tr>
<td>Residue from Previous Blend</td>
<td>EB-283</td>
<td>12.21</td>
<td>5.86</td>
<td>1.17</td>
</tr>
<tr>
<td>Matrix Material Al Powder</td>
<td></td>
<td>843.01</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Other Material</td>
<td></td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Total Blend</td>
<td></td>
<td>1846.85</td>
<td>896.90</td>
<td>179.22</td>
</tr>
</tbody>
</table>

#### COMPONENT PRODUCTS

<table>
<thead>
<tr>
<th>No. Acceptable Pressed Comp.</th>
<th>19</th>
<th>1759.42</th>
<th>844.51</th>
<th>168.90</th>
</tr>
</thead>
<tbody>
<tr>
<td>Average weight Acceptable Comp.</td>
<td>-</td>
<td>(92.60)</td>
<td>(44.45)</td>
<td>(8.89)</td>
</tr>
<tr>
<td>Rejected Components</td>
<td>1</td>
<td>92.60</td>
<td>44.45</td>
<td>8.89</td>
</tr>
<tr>
<td>Unused Blend Residue</td>
<td>10.61</td>
<td>5.09</td>
<td>1.02</td>
<td></td>
</tr>
<tr>
<td>Total Component Products</td>
<td>1862.63</td>
<td>894.06</td>
<td>178.81</td>
<td></td>
</tr>
<tr>
<td>Blend-Component Difference</td>
<td>4.22</td>
<td>2.03</td>
<td>0.41</td>
<td></td>
</tr>
</tbody>
</table>

**FIG. 4b. Typical blended materials batch log**

**Component processing**

"Secondary fabrication" relates to work on the fuel components after they are completely contained in non-fuel metals. Since changes in the uranium content of the components are now impossible without their destruction, material control becomes relatively simplified; only a piece identification is necessary.

The most difficult task in controlling secondary materials is to maintain identification of small, individual components until they can be permanently marked. The use of batch containers is a good method for maintaining identification as well as transporting the pieces through the early stages of secondary processing when the components are in groups of identical pieces and can be interchanged. Such a container should be compartmented to accommodate any rejected pieces as they occur in the process. Once the components have been permanently marked, usually by etching or stamping, the batch container is no longer needed and can be diverted to new batches of components.
Control of the final assembly

If several of the small components are to be further processed into a fuel assembly, control shifts from individual component identification to assembly or fuel element identification. Components from different batches will often be mixed to attain fuel content within a close tolerance. The control shifts when the plates are selected and temporarily assembled by taping or tying and should be so recorded.

Control records

Secondary fabrication records fall into three categories: (1) component processing records; (2) component disposition records; and (3) assembled component records.

**BATCH CONTROL CARD**

<table>
<thead>
<tr>
<th>Stage</th>
<th>Processed By</th>
<th>Rejections and Reason</th>
<th>Inspected By</th>
</tr>
</thead>
<tbody>
<tr>
<td>1. Frame</td>
<td>GWC</td>
<td>OK</td>
<td>HJW</td>
</tr>
<tr>
<td>2. Cover</td>
<td>GWC</td>
<td>OK</td>
<td>HJW</td>
</tr>
<tr>
<td>3. Can</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>4. Weld</td>
<td>GJW</td>
<td>OK</td>
<td>AK</td>
</tr>
<tr>
<td>5. Hot Roll</td>
<td>CWH</td>
<td>OK</td>
<td>HJW</td>
</tr>
<tr>
<td>6. Cold Roll</td>
<td>CWH</td>
<td>1.057</td>
<td>HJW</td>
</tr>
<tr>
<td>7. Anneal</td>
<td>JWH</td>
<td>2,. Electrode</td>
<td>WWP</td>
</tr>
<tr>
<td>8. Fluoroscopy</td>
<td>JWB</td>
<td>3. Crease of Tab.</td>
<td>JWB</td>
</tr>
<tr>
<td>9. Shear</td>
<td>JWB</td>
<td>OK</td>
<td>JWB</td>
</tr>
<tr>
<td>10. Machine</td>
<td>CPR</td>
<td>OK</td>
<td>AK</td>
</tr>
<tr>
<td>11. Anneal</td>
<td>JWH</td>
<td>1. Electrode</td>
<td>WWP</td>
</tr>
<tr>
<td>12. Inspect</td>
<td>WWP</td>
<td>1. thin Edge</td>
<td>WWP</td>
</tr>
</tbody>
</table>

Total Accepted Components to Storage: 98
Total Scrapped Components: 8

**FIG. 4c. Typical batch control card**

Figure 4c illustrates a record for controlling the materials during component processing. This batch control record accompanies its group of components through all stages of processing, accounting for the components, and collecting information concerning rejections. When complete, the record is filed with the previous batch log (Fig. 4a or b) and the two serve as a complete materials and process control from receipt of the starting materials through component processing. If desired, the two cards may be merged into a single control record which is initiated with the starting materials.
At this point, the fuel components may be finished products or may require further assembly into multicomponent units. In either case, a record should be made to indicate the disposition of the finished accepted components. Figure 5a is a record that initially records the temporary storage of a finished component batch. Their shipment, or further processing into assembled units, is indicated as it occurs, and their disposition is permanently recorded.

Materials control during assembly is very similar to batch control. A record is made of those components selected for assembly into an element and follows them through the assembly process. The element assembly record, illustrated in Fig. 5b was originally designed for the collection of data during assembly of an MTR element containing curved plates. This record is also a good illustration of how one may integrate the maximum number of controls into the minimum number of records to eliminate duplication.

### COMPLETED COMPONENT DISPOSITION RECORD

<table>
<thead>
<tr>
<th>Disposition</th>
<th>Date</th>
<th>Number Components</th>
<th>Balance on Hand</th>
</tr>
</thead>
<tbody>
<tr>
<td>Received for Storage</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Element 1-109</td>
<td>6-12-58</td>
<td>68</td>
<td>576.4</td>
</tr>
<tr>
<td>Shipment 1-123</td>
<td>6-15-58</td>
<td>27</td>
<td>527.32</td>
</tr>
<tr>
<td>Element 1-121</td>
<td>6-24-58</td>
<td>16</td>
<td>142.46</td>
</tr>
<tr>
<td>Element 1-123</td>
<td>6-25-58</td>
<td>9</td>
<td>37.68</td>
</tr>
<tr>
<td>Element 1-127</td>
<td>6-27-58</td>
<td>4</td>
<td>0</td>
</tr>
</tbody>
</table>

FIG. 5a. Typical component disposition record

**Scrap handling**

Scrap materials, which can occur at any time during the fabrication process, must be disposed of by either recycling through the process or reclaiming through chemical recovery. When economically feasible, scrap may be buried or otherwise safely destroyed; this rarely occurs, however, with the use of highly enriched material.

Alloy residues of good quality can be remelted with additional uranium and aluminium to form a new ingot, so chemical recovery is not economically feasible unless the process is closed. Rejected components can often be remelted with the alloy residues if no contaminants that would damage the new alloy are present in the cladding materials.
Some residues, like heat dross from the top of the alloy melt, contain so many impurities that they must always be removed from the process and recovered. Also, rejected powder compacts can be recovered only by chemical means.

Since enriched scrap usually occurs in relatively small amounts, it is recovered most inexpensively by accumulating several small quantities into a large batch for a single recovery operation. Such a batch is, of course, limited by criticality considerations.

Control summary

The final control procedure in the fabrication of enriched uranium dictates a summary and general examination of the entire process. The total amount of uranium material that has entered the fabrication plant must be compared with the total amount on hand after processing, and process differences must be determined. In addition, all records should be examined to ensure that product specifications have been met and analysed for any trends that would affect the process or components.
The material balance

The materials before and after processing are best compared by the material balance. In this method, the process is either actually or theoretically cut off at a selected point. Usually, an actual cut-off occurs only if the product is changing and different specifications are involved (such as a different uranium enrichment), necessitating an equipment clean-up and resetting or retooling. Pure and unclad materials are weighed and counted and their uranium content is computed. The clad or contained materials are counted and identified, and fuel content is established from their fabrication records.

### ENRICHED URANIUM MATERIAL BALANCE

<table>
<thead>
<tr>
<th>Period: July 1 - 31</th>
<th>Total Uranium</th>
<th>U-235</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Grams</td>
<td>Grams</td>
</tr>
<tr>
<td>Balance on Hand, June 30</td>
<td>17,037</td>
<td>15,333</td>
</tr>
<tr>
<td>Total Receipts - July</td>
<td>25,274</td>
<td>22,747</td>
</tr>
<tr>
<td>Total Previous Balance and Receipts</td>
<td>42,311</td>
<td>38,080</td>
</tr>
<tr>
<td>Less Total Shipments - July</td>
<td>16,426</td>
<td>14,783</td>
</tr>
<tr>
<td>Balance on Hand, July 31</td>
<td>25,885</td>
<td>23,297</td>
</tr>
</tbody>
</table>

Materials Inventory

Beginning Materials:
- Metal 3,421 3,079

Materials in Process:
- Billets 2,652 2,387
- Rods 1,181 1,243
- Cures 1,829 1,546
- Remelt Alloy from Punchings 1,491 1,891

Product Materials:
- Plates 6,737 6,063
- Assembled Element 7,347 6,012

Scrap Materials for Recovery:
- Heat Dross
  - By Weight and % Uranium 998
  - By Difference 973
  - Recovered 924 924 832

**TOTAL INVENTORY**
- 25,836 23,253

**Process Difference (Loss)**
- 49 44

---

**FIG. 5c. Typical uranium material balance**

On completion, the inventory is checked against closing balances indicated on all previously discussed control records. The inventory listings are posted to a previously prepared worksheet which already bears the ending control record balances. In some cases, this worksheet is given to the personnel taking the inventory as a guide; but when this is done the advantage of a "blind" inventory requiring a thorough search is lost.

The uranium content values computed for the inventoried materials listed on the worksheet are now summed and drawn into an actual balance against all materials charged to the plant (Fig. 5c).
## Scrap, Waste, and Yield Record

**Uranium Aluminum Alloy**

For Period: July 1 to July 8

<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Net Weight</td>
<td>Total U</td>
<td>U235</td>
</tr>
<tr>
<td>E-514</td>
<td>76</td>
<td>3</td>
<td>1</td>
<td>3</td>
<td>8</td>
<td>8</td>
<td></td>
<td></td>
</tr>
<tr>
<td>E-514</td>
<td>76</td>
<td>1</td>
<td>3</td>
<td>2</td>
<td>3</td>
<td>3</td>
<td></td>
<td></td>
</tr>
<tr>
<td>E-515</td>
<td>76</td>
<td>2</td>
<td>1</td>
<td>4</td>
<td>7</td>
<td>7</td>
<td></td>
<td></td>
</tr>
<tr>
<td>E-516</td>
<td>74</td>
<td>1</td>
<td>3</td>
<td>1</td>
<td>9</td>
<td>9</td>
<td></td>
<td></td>
</tr>
<tr>
<td>E-516</td>
<td>74</td>
<td>1</td>
<td>1</td>
<td>3</td>
<td>1</td>
<td>0.0</td>
<td></td>
<td></td>
</tr>
<tr>
<td>E-517</td>
<td>76</td>
<td>2</td>
<td>1</td>
<td>1</td>
<td>9</td>
<td>9</td>
<td></td>
<td></td>
</tr>
<tr>
<td>E-517</td>
<td>76</td>
<td>2</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>0.0</td>
<td></td>
<td></td>
</tr>
<tr>
<td>E-518</td>
<td>76</td>
<td>1</td>
<td>2</td>
<td>3</td>
<td>1</td>
<td>1.0</td>
<td></td>
<td></td>
</tr>
<tr>
<td>E-518</td>
<td>76</td>
<td>1</td>
<td>2</td>
<td>3</td>
<td>2</td>
<td>2</td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>Totals</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td><strong>378</strong></td>
<td><strong>24</strong></td>
<td><strong>4</strong></td>
<td><strong>30</strong></td>
</tr>
</tbody>
</table>

**Fig. 5d. Typical scrap, waste, and yield record**

*Number denotes type of rejection.*
Process differences

Note that the uranium content of the heat dross in this particular balance is determined three ways: as the uranium percentage in the alloy, as the difference between inventory and book balance, and as the actual recovered content. Only the last is significant in determining the normal process differences that will occur, as the impurities in the heat dross will vary rather extensively from batch to batch. Indeed, on first drawing the balance using the percentage, one will more often experience a process gain rather than a process loss, a good first indication that the final balance will fall within acceptable limits of error if such a gain is small.

What, then, are acceptable limits of error for the process? The answer to this question is tied directly to two considerations: (1) the collective measurement limits of error for the process; and (2) the fuel specifications of the fabricated products. If the process difference falls within these limits, it is a reasonable one. If it does not, individual process circumstances must be considered and the process records must be re-examined to isolate the process stage or stages where it occurred. Sometimes, additional checks must be made, such as X-ray examination, further component sampling, or a chemical analysis of the related back-up dip samples. In any case, the fabricator must be satisfied as to the product fuel content, for there is no practical adjustment after the component has entered the reactor.

Process losses of raw material also affect the product cost, especially when expensive enriched uranium is the raw material. If a relatively large process loss occurs, even though the components meet specifications satisfactorily, the cost of enriched uranium requires that the process be re-examined and adjusted to make any possible reduction of this loss.

Process evaluation

In addition to a recheck of all measurements and values reflected in the process records, a thorough analysis and evaluation of the total data are important. Such factors as dimensions consistently on the high side set patterns of intended fuel content versus intended result, and constant recurrence of unusual effects or normal defects will often reflect areas in the process requiring further development work.

Another important aspect of any fabrication effort is the maintenance of a good ratio of product yield to materials processed. The scrap, waste and yield record (Fig. 5d) illustrates a good approach to determining this ratio. It also provides a scrap analysis for determining process trouble spots.

SUMMARY

Control of enriched uranium during its fabrication is important because of the scarcity, expense, and special hazards of the material, and because technical success in the manufacture of reactor fuel elements depends on how well its controls are designed and applied. The relation of these factors
to the particular fabrication process must be considered before one can properly select and establish controls for that process. The basic objectives in control system design include completely integrated records for a minimum number of material units in each major fabrication step confirmed by reliable cross checks.

Control procedures must be established to provide data needed for the ultimate material balance. From this summary, process gains or losses are determined which either confirm the product fuel content or indicate the need for further examination of the control data and fuel components.

Two other results are evident in developing proper control procedures. First, as production proceeds, the number of units to be controlled decreases; and the higher the rate of decrease, the more efficient and simplified the control procedure becomes. Second, the data generated during the control process can form significant patterns that point to possible trouble spots in the fabrication effort.

REFERENCES


BIBLIOGRAPHY

FOSTER, B.E., SNYDER, S.D., McCUNG, R.W., Continuous scanning X-ray attenuation technique for determining fuel inhomogeneities in dispersion core fuel plates, USAEC rep. ORNL-3737 (1965).

DISCUSSION

W. J. WRIGHT: You mentioned the use of density measurements to determine the uranium content of U-Al alloy plates. How good is this method?
R. G. CARDWELL: In addition to using the density method for pre-determining the desired wt. % of uranium in the alloy core of the final component, we can use it to confirm that this wt. % has been attained during fabrication. This is done by measuring the density of the alloy fuel compact
(plate core), before framing and cladding, and checking this result against the wt. %/density curve from which the original determination was made. However, whether or not this is a desirable control, and to what extent such a control is applied, depends on the fuel content accuracy required in the particular component fabricated.
A NUCLEAR MATERIALS MANAGEMENT CONTROL SYSTEM FOR URANIUM REACTOR FUEL ELEMENT FABRICATION (A HYPOTHETICAL CASE)

L.D. HAZELTON
THE DOW CHEMICAL COMPANY, GOLDEN, COLO., UNITED STATES OF AMERICA

Abstract — Résumé — Аннотация — Resumen

A NUCLEAR MATERIALS MANAGEMENT CONTROL SYSTEM FOR URANIUM REACTOR FUEL ELEMENT FABRICATION (A HYPOTHETICAL CASE). A control system for effective nuclear materials management requires the collection and dissemination of varying data. This paper describes a control system that will meet the requirements and at the same time eliminate duplication of effort by combining the data collection needs of accountability, production control and process control. The presentation is divided into three parts: the data required; collection, tabulation and distribution of the data; and how the data is used. Each topic will be covered in sufficient detail to provide the nucleus for establishing a control system.

SYSTEME DE CONTROLE DE LA GESTION DES MATIERES NUCLEAIRES APPLICABLE A LA FABRICATION D'ELEMENTS COMBUSTIBLES A BASE D'URANIUM POUR REACTEURS (CAS HYPOTHETIQUE). Un système de contrôle assurant la gestion efficace des matières nucléaires suppose que l'on rassemble et diffuse différentes données. L'auteur décrit un système de contrôle qui remplit ces conditions et évite toute répétition d'un même travail en groupant les besoins statistiques des services de comptabilité, de contrôle de la production et de contrôle des opérations. Le mémoire est divisé en trois parties: données requises; groupement, élaboration et diffusion des données; mode d'utilisation des données. Chacune de ces questions est analysée de façon suffisamment détaillée pour servir de base à la création d'un système de contrôle.

СИСТЕМА АДМИНИСТРАТИВНО-ХОЗЯЙСТВЕННОГО КОНТРОЛЯ ПРИ ПРОИЗВОДСТВЕ ТОПЛИВНЫХ ЕЛЕМЕНТОВ ИЗ УРАНА (ГИПОТЕТИЧЕСКИЙ СЛУЧАЙ). Система контроля для обеспечения эффективности административно-хозяйственного учета ядерных материалов требует собирания и распространения различных данных. Дается описание системы контроля, отвечающей этим требованиям и, вместе с тем, исключающей дублирование благодаря объединению усилий в собирании данных для отчетности, производственного и технологического контроля. Рассматривают три темы: требуемые данные; сбор, составление таблиц и распространение данных; способы использования данных. Каждая из этих тем разрабатывалась с достаточными подробностями, чтобы служить основой для создания системы контроля.

SISTEMA DE CONTROL DE MATERIALES NUCLEARES PARA LA ELABORACION DE ELEMENTOS COMBUSTIBLES DE URANIO (CASO HIPOTETICO). Todo sistema de control para una administración eficaz de los materiales nucleares requiere la compilación y distribución de diversos datos. En esta memoria se describe un sistema que satisface las exigencias fundamentales del control y al mismo tiempo evita la duplicación combinando las necesidades de los servicios de contabilidad, control de la producción y control de las operaciones. Se divide en tres partes que tratan, respectivamente, de los datos necesarios, de su compilación, tabulación, y distribución y de la manera de utilizarlos. Cada uno de estos temas es objeto de un estudio detenido cuya finalidad es facilitar las bases para el establecimiento de un sistema de control.

INTRODUCTION

In the early days of fabricating small quantities of uranium on a laboratory scale, control over the materials was easily accomplished. Usually
this consisted of physical control exercised by the individual performing the work. As demand for products made from uranium increased, larger production facilities were built and processing techniques for handling large quantities of material were developed. Today, in a modern plant, the fabrication of uranium reactor fuel elements is usually a large production operation utilizing complicated processes on materials that are relatively scarce and expensive. Many of these facilities are operated by private enterprise having full financial responsibility for the SS materials\(^1\). Inadequate control over SS materials in such a venture could easily make the difference between financial success and failure.

Since efficient operation requires strict adherence to schedules and specifications and a satisfactory accountability over the material at all times, SS material control systems have also evolved from a simple one-man visual operation to a complex system of data collection and dissemination. Originally, three systems of data collection - one for accountability, one for production control and one for process control - were maintained over the same processes and materials. This was done for several reasons, chief of which was the inability to co-ordinate a consolidated system that would feed data back to the user when he needed the information. The use of computer equipment for high speed data collection and dissemination has made possible an efficient system of control that will satisfy the needs of the three categories of control previously mentioned.

To understand better the problem of establishing a consolidated control system, the three major types of control are defined as follows:

(a) Accountability control

A system of control which reasonably ensures that all SS materials have been accounted for and are on hand; that materials have not been inadvertently lost; that materials have not been removed from the operation in an unauthorized manner, and that materials have been adequately measured.

(b) Production control

A system of control capable of providing detailed data about each phase of the process; the flow of parts through processing and the number of parts in each processing step; the quantity and type of feed materials on hand; the quantity and type of scrap awaiting recovery, and the quantity of finished product meeting specifications and awaiting shipment.

(c) Process control

A system of utilizing data from measurements, analyses and inspections to ensure that the product meets control specifications, and the use of these data and other information such as operation time, temperatures and pressures to develop new or better processing techniques.

The three systems of control are interrelated and dependent to some extent upon one another. Each requires a collection of large volumes of data, which, if accomplished separately, would result in significant duplication. Efficient use of manpower and machines requires that data used by

---

\(^1\) The term "SS materials" is an abbreviation for source and special nuclear materials which are defined as follows: Source materials are natural uranium and thorium as found in nature. Special nuclear materials are those nuclear materials that are man-made and include uranium enriched in the uranium-235 isotope beyond that found in nature, uranium depleted in the uranium-235 isotope below that found in nature and plutonium.
the three separate control systems be efficiently combined. The accomplishment of unified control eliminates the duplication of data-gathering and processing and provides management with better overall tools to effect control at a minimum cost.

This paper will describe a unified control system applicable to a large production facility fabricating uranium reactor fuel elements. My intent is to illustrate and explain some of the problems most likely to be encountered by those who are establishing control systems for the first time or those making improvements on an existing system.

The discussion is divided into three major parts as follows: (a) data required; (b) collection, tabulation and distribution of the data; (c) how the data is used.

Because of the word limitation, this discussion excludes the principles of computer programming and operation, and for the same reason process control programmes using the data will be mentioned, but not explained. The computer operation and process control programmes would each require a separate paper for adequate discussion.

DATA REQUIRED

One of the first things that must be known to establish a control system is what type of information about SS materials is needed. Making this decision usually involves a review of the end product of the proposed control programme or what the data will be used for. A brief statement of what will be done with the accumulated information prefaces the following outline of data required for each segment of control.

(a) To establish a control over materials as outlined in the definition for accountability, we must obtain data about the SS materials that will enable us to produce a book inventory (an inventory determined from the records), establish procedures for listing certain common data about physical inventories and provide procedures to document accurately the flow of material. The data needed are described as follows:

(i) The batch, lot or item identification, uranium weight, uranium-235 per cent, uranium-235 weight and impurity content of feed material received.

(ii) The batch, lot or item identification, net weight, uranium weight, uranium-235 weight, alloy weight of each part, lot or residue transferred from one major processing or control area to another within the plant.

(iii) The unit identification, net weight, uranium weight, uranium-235 weight, alloy per cent, alloy weight and impurity content of all finished product shipped from the plant.

(iv) The batch, lot or item identification of all parts or batches created.

(v) The batch, lot or item identification, net weight, uranium weight, uranium-235 per cent, uranium-235 weight, alloy per cent and alloy weight of all parts scrapped during fabrication.

(vi) Analytical results on samples taken from materials in process or on inventory.
(vii) The batch, lot or item identification, net weight, uranium weight, uranium-235 per cent, uranium-235 weight, alloy per cent, alloy weight and impurity content of all materials listed on physical inventories.

(b) Certain data are required to provide reasonable assurance that SS material measurements are valid. Usually this is accomplished by statistically comparing analyses on production samples to analyses on standard samples, evaluating check weighing programmes on scales and balances, validating physical inventory weights and accurately calibrating tanks and vessels for volume measurements. The data gathered from the control programme are limited to results of analytical, emission spectrographic and mass spectrographic analyses. Data on vessel calibration and check weighing programmes do not lend themselves to economical collection by means of a unified collection system. This information is obtained from special programmes and becomes a part of the procedures to assure adequately that measurements are reasonably valid.

(c) Production control needs information about SS materials to determine if the operation is meeting schedules and to plan and prepare future schedules. These data have to be furnished to production control on a timely basis, usually daily. The data are summarized as follows:

(i) The location of a batch or part at specific processing steps.
(ii) The number of parts or batches initiated.
(iii) The number of parts or batches scrapped and the reason for scrapping.
(iv) The quantity and type of scrap awaiting recovery process.
(v) The type and quantity of finished goods available for shipment.
(vi) The quantity and type of feed materials on hand.

(d) Process control (product control) needs data that can be used to determine if the product meets the buyer's specifications, if processing equipment is functioning correctly and as the basis for process and equipment improvement. The information is summarized as follows:

(i) Results of analytical, emission spectrographic and mass spectrographic analyses.
(ii) Results of radiographic analyses.
(iii) The quantity and type of units produced.
(iv) The number of units scrapped and the reason for scrapping.
(v) Dimensional inspection information.
(vi) Processing data such as time, temperature and pressure.

The data required for a control system over materials in a particular plant or operation will depend upon the product manufactured, type of processing and the desired control deemed necessary by management. The described data may not meet your specific requirements, but represent what we have found to be typical of a fuel element fabricating facility.

COLLECTION, TABULATION AND DISTRIBUTION OF THE DATA

Once the required data have been determined, the next step is to devise ways to collect these data. The first step in the collection of data for this purpose is to record the information on what we call source documents.
the years several types of documents have been used as source documents by uranium fabricators and processors. Some of these are described as follows:

(a) Batch cards

These forms may be of various designs. Usually they include an identification number, material weight, type of material and other processing information that may be necessary.

(b) Shipping documents

These forms are used to describe materials that are shipped from the facility. Material weights, part or item identification numbers and container numbers are usually shown.

(c) Receiving reports

These forms are used to show materials received from outside the facility. Shipment identification, item identification and material weights are listed.

(d) Inspection reports

Forms used to record inspection information about specific parts. They usually show part identification, material weight and specific dimensions.

(e) Analytical requisitions and reports

Forms used to request the analysis of samples, identify the batch or part represented by the sample, specify the type of analysis required and show the results of such analysis.

(f) Transfer receipts

Forms used to show movement of materials between major processing areas. Usually parts and batches are identified, material weights are given and material types are shown. Other pertinent information may be included.

(g) Batch or part initiation records

A production record showing creation of an identifiable entity. Identification number and location are shown. Material weight may be included.

(h) Scrap reports

A form showing parts scrapped in process. Minimum information is part identification, material weight and the reason for scrapping the part.

The next step in establishing a control system is to decide which source documents will be the best to use for collecting the required data. In some cases existing forms are satisfactory; existing forms can be modified; or it may be necessary to design new forms. Process flow charts are a help in making this decision. Figures 1 and 2 are examples of process flow charts for the uranium processing facility described in this paper.

(a) Figure 1 shows the flow of material through the various stages of processing, points of residue generation and processing steps where accountability data are generated. Material balance areas are shown by the broken lines. The large encircled letters A, B, etc. are keyed to the source documents used for recording the data.

(b) Figure 2 shows processing steps where production and process control data are generated.

The source documents used to record the data generated as outlined in the flow charts are illustrated in forms 1 to 10 in the Appendix.
FIG. 1. Process flow chart showing generation points for accountability data
FIG. 2. Process flow chart showing generation points for production and process control data
(a) Form 1, code symbol ®, illustrates the standard shipping document used to transfer materials between licensees\(^2\) and between licensees and AEC contractors.

(b) Form 2, code symbol (J), illustrates a feed storage control card that is used to record all feed received from suppliers. The shipper's data, codes and control numbers are preprinted before issuance to the feed storage area personnel. The balance of the data will be entered as each item is checked, weighed and sampled.

(c) Form 3, code symbol ©, illustrates a combined casting - rolling - shearing operation control card. Pertinent data such as composition and weight of casting charge, weight of ingot, weight of alloy material and other processing information needed to describe these operations are recorded on the card.

(d) Form 4, code symbol illustrates a general process control card used to document the movement of fuel elements from the shearing operation through the various stages of processing and inspection. Only data pertinent to each operation are shown each time the card is used. The initial process number and new process number will always be shown regardless of the other information recorded.

(e) Form 5, code symbol ©, illustrates a transfer receipt used to record the movement of finished fuel elements from the fabrication area to shipment storage. This form is used primarily as an information document and as a means of transferring responsibility from one area to another. Pertinent data describing the finished fuel element are already in the computer magnetic tape storage.

(f) Form 6, code symbol ©, illustrates a salvage preparation batch card. This form is used to record the batching of all residues generated from processing operations and to record the movement of the material from the point of origin to the recovery area.

(g) Form 7, code symbol ©, illustrates a receiving report used to record receipt of all scrap materials from off plant. Shipper's material identification and weights are recorded before issuance to the recovery receiving area. Pertinent receiver's data such as description, material weights, sample requisition number and process control number are recorded as they become available during the receiving operation.

(h) Form 8, code symbol ©, illustrates a form used to record all residues containing small amounts of uranium that are uneconomical to recover and are discarded.

(i) Form 9, code symbol ©, illustrates the form used to request specific laboratory analyses. The form also acts as the transfer document moving the material from production areas to the laboratory.

(j) Form 10 illustrates a universal form used for recording physical inventories.

Letter symbols (®, ©, etc.) shown on examples of source documents indicate the location of their use on the process flow charts.

---

\(^2\) A person or corporation licensed by the United States Atomic Energy Commission to possess specific SS materials.

\(^3\) Abbreviation for the United States Atomic Energy Commission.
Establishing codes for material types, processing information, inspection data, analytical methods, inventories, material location, processing steps and account identification are closely related to source document selection.

Codes should be numerical wherever possible and conform with a standard pattern designed for computer programming and operation. Codes should be devised in such a manner that they can be expanded without causing extensive revisions in the master computer programme; and finally, codes should be approved by the users of the data as well as the data processing centre.

The following numerical codes for classifying various data were assigned for use in our model control system:

(a) Material type codes

<table>
<thead>
<tr>
<th>Code</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>10 - 30 Metal</td>
<td></td>
</tr>
<tr>
<td>10</td>
<td>Feed material, buttons from recycle</td>
</tr>
<tr>
<td>11</td>
<td>Feed material, buttons from off-site</td>
</tr>
<tr>
<td>12</td>
<td>Feed material, scrap</td>
</tr>
<tr>
<td>13</td>
<td>Feed material, briquettes</td>
</tr>
<tr>
<td>14</td>
<td>Turnings, chips and fines</td>
</tr>
<tr>
<td>15</td>
<td>Skulls</td>
</tr>
<tr>
<td>16</td>
<td>Samples</td>
</tr>
<tr>
<td>17</td>
<td>Ingots</td>
</tr>
<tr>
<td>18</td>
<td>Sheet</td>
</tr>
<tr>
<td>19</td>
<td>Fuel elements in process</td>
</tr>
<tr>
<td>20</td>
<td>Finished elements</td>
</tr>
<tr>
<td>21</td>
<td>Scrapped elements</td>
</tr>
<tr>
<td>31 - 50 Recoverable liquids</td>
<td></td>
</tr>
<tr>
<td>31</td>
<td>Carbitol</td>
</tr>
<tr>
<td>32</td>
<td>Coolant and lubricating oils</td>
</tr>
<tr>
<td>33</td>
<td>Perclene and triclene</td>
</tr>
<tr>
<td>34</td>
<td>Miscellaneous organic solvents</td>
</tr>
<tr>
<td>35</td>
<td>Nitric acid solutions</td>
</tr>
<tr>
<td>36</td>
<td>Filtrates</td>
</tr>
<tr>
<td>37</td>
<td>Column product</td>
</tr>
<tr>
<td>38</td>
<td>Skull solutions</td>
</tr>
<tr>
<td>39</td>
<td>Miscellaneous solutions</td>
</tr>
<tr>
<td>51 - 70 Recoverable solids</td>
<td></td>
</tr>
<tr>
<td>51</td>
<td>Green salt UF₄</td>
</tr>
<tr>
<td>52</td>
<td>Orange oxide UO₃</td>
</tr>
<tr>
<td>53</td>
<td>Black oxide U₃O₈</td>
</tr>
<tr>
<td>54</td>
<td>Yellow cake UO₄</td>
</tr>
<tr>
<td>55</td>
<td>Sand, liner, slag and crucibles</td>
</tr>
<tr>
<td>56</td>
<td>Combustibles</td>
</tr>
<tr>
<td>57</td>
<td>Ash</td>
</tr>
<tr>
<td>58</td>
<td>Graphite and carbocel</td>
</tr>
<tr>
<td>59</td>
<td>Glass and metal</td>
</tr>
<tr>
<td>60</td>
<td>Plastics, tygon and rubber</td>
</tr>
<tr>
<td>61</td>
<td>Filter muds</td>
</tr>
</tbody>
</table>
HAZELTON

71 - 80 Discard materials
71 Discard solids
72 Discard solutions

(b) Standard fuel element identification codes

<table>
<thead>
<tr>
<th>Code</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>01</td>
<td>5% U-235 12&quot; X 4&quot; X 1/4&quot;</td>
</tr>
<tr>
<td>02</td>
<td>5% U-235 18&quot; X 4&quot; X 1/4&quot;</td>
</tr>
<tr>
<td>03</td>
<td>10% U-235 12&quot; X 4&quot; X 1/4&quot;</td>
</tr>
<tr>
<td>04</td>
<td>10% U-235 18&quot; X 4&quot; X 1/4&quot;</td>
</tr>
<tr>
<td>05</td>
<td>5% U-235 5% Molybdenum 12&quot; X 4&quot; X 1/4&quot;</td>
</tr>
<tr>
<td>06</td>
<td>5% U-235 5% Molybdenum 18&quot; X 4&quot; X 1/4&quot;</td>
</tr>
<tr>
<td>07</td>
<td>10% U-235 5% Molybdenum 12&quot; X 4&quot; X 1/4&quot;</td>
</tr>
</tbody>
</table>
| 08   | 10% U-235 5% Molybdenum 18" X 4" X 1/4"

These codes may be expanded to cover any number of standardized fuel elements for size, uranium-235 content and alloy content.

(c) Element codes

<table>
<thead>
<tr>
<th>Code</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>01</td>
<td>Normal uranium</td>
</tr>
<tr>
<td>02</td>
<td>Enriched uranium, more than 75% U-235</td>
</tr>
<tr>
<td>03</td>
<td>Enriched uranium, less than 75% U-235</td>
</tr>
<tr>
<td>04</td>
<td>Depleted uranium</td>
</tr>
<tr>
<td>05</td>
<td>U-235</td>
</tr>
<tr>
<td>06</td>
<td>Aluminium</td>
</tr>
<tr>
<td>07</td>
<td>Zirconium</td>
</tr>
<tr>
<td>08</td>
<td>Molybdenum</td>
</tr>
<tr>
<td>09</td>
<td>Tungsten</td>
</tr>
</tbody>
</table>

(d) Reject codes

<table>
<thead>
<tr>
<th>Code</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>01</td>
<td>Dimensional reject. Machining step 1</td>
</tr>
<tr>
<td>02</td>
<td>Dimensional reject. Machining step 2</td>
</tr>
<tr>
<td>03</td>
<td>Dimensional reject. Machining step 3</td>
</tr>
<tr>
<td>04</td>
<td>Radiography reject</td>
</tr>
<tr>
<td>05</td>
<td>Impurity reject</td>
</tr>
<tr>
<td>06</td>
<td>Alloy reject</td>
</tr>
<tr>
<td>07</td>
<td>Isotopic reject</td>
</tr>
<tr>
<td>08</td>
<td>Plating reject</td>
</tr>
</tbody>
</table>

(e) Analytical method codes

<table>
<thead>
<tr>
<th>Code</th>
<th>Method</th>
</tr>
</thead>
<tbody>
<tr>
<td>01</td>
<td>Colorimetric</td>
</tr>
<tr>
<td>02</td>
<td>Fluorimetric</td>
</tr>
<tr>
<td>03</td>
<td>Gravimetric</td>
</tr>
<tr>
<td>04</td>
<td>Gamma counting</td>
</tr>
<tr>
<td>05</td>
<td>Emission spectrographic</td>
</tr>
<tr>
<td>06</td>
<td>Mass spectrographic</td>
</tr>
<tr>
<td>07</td>
<td>Radioassay</td>
</tr>
<tr>
<td>08</td>
<td>Titration analysis, direct with dichromate</td>
</tr>
</tbody>
</table>
### (f) Measurement method codes

<table>
<thead>
<tr>
<th>Code</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>01</td>
<td>Weighed</td>
</tr>
<tr>
<td>02</td>
<td>Weighed, sampled and analysed</td>
</tr>
<tr>
<td>03</td>
<td>Volumetrically measured, sampled and analysed</td>
</tr>
<tr>
<td>04</td>
<td>Estimated by difference</td>
</tr>
<tr>
<td>05</td>
<td>Estimated from past experience</td>
</tr>
</tbody>
</table>

### (g) Operation and location codes

<table>
<thead>
<tr>
<th>Code</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>01</td>
<td>Feed storage</td>
</tr>
<tr>
<td>02</td>
<td>Casting</td>
</tr>
<tr>
<td>03</td>
<td>Rolling and shearing</td>
</tr>
<tr>
<td>04</td>
<td>Machining</td>
</tr>
<tr>
<td>05</td>
<td>Inspection</td>
</tr>
<tr>
<td>06</td>
<td>Plating</td>
</tr>
<tr>
<td>07</td>
<td>Final inspection</td>
</tr>
<tr>
<td>08</td>
<td>Shipment storage</td>
</tr>
</tbody>
</table>

### (h) Inspection identification codes

<table>
<thead>
<tr>
<th>Code</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>01</td>
<td>Length</td>
</tr>
<tr>
<td>02</td>
<td>Width</td>
</tr>
<tr>
<td>03</td>
<td>Thickness</td>
</tr>
</tbody>
</table>

### (i) Account numbers - Material balance areas

<table>
<thead>
<tr>
<th>Code</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>10-1</td>
<td>Feed storage</td>
</tr>
<tr>
<td>20-1</td>
<td>Foundry and rolling mill</td>
</tr>
<tr>
<td>30-1</td>
<td>Fabrication (includes machining, plating and inspection)</td>
</tr>
<tr>
<td>40-1</td>
<td>Shipment storage (includes shipment preparation and storage)</td>
</tr>
<tr>
<td>50-1</td>
<td>Residue processing</td>
</tr>
<tr>
<td>60-1</td>
<td>Laboratories and research and development</td>
</tr>
</tbody>
</table>

These codes may be further sub-divided if necessary, i.e., 10-1, 10-2, 10-3, etc.

### Control accounts

<table>
<thead>
<tr>
<th>Code</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>1000</td>
<td>Plant inventory control</td>
</tr>
<tr>
<td>2000</td>
<td>Plant receipts control</td>
</tr>
<tr>
<td>3000</td>
<td>Plant shipments control</td>
</tr>
</tbody>
</table>

Another important consideration is a standardized method of part or batch identification for materials as they travel from process to process. The process control numbering programmes used for the control programme described herein is outlined as follows:

#### (a) Processing numbering system

<table>
<thead>
<tr>
<th>Operation</th>
<th>Numbers</th>
</tr>
</thead>
<tbody>
<tr>
<td>Feed storage</td>
<td>1-00001 to 1-10000</td>
</tr>
<tr>
<td>Casting</td>
<td>00001 to 5000</td>
</tr>
<tr>
<td>Rolling</td>
<td>Casting number</td>
</tr>
</tbody>
</table>
Shearing 00001-XX to 5000-XX suffixes are added to casting processing number for each sheared piece
Machining Casting number with suffix
Inspection Casting number with suffix
Plating Casting
Final inspection Fuel element serial number as requested by customer
Shipment Fuel element serial number

Request for analyses
Samples taken from feed materials received from off-site 10-0001 to 10-9999
Samples taken from good metal 20-0001 to 20-9999
Samples taken from recoverable liquids 30-0001 to 30-9999
Samples taken from recoverable solids 50-0001 to 50-9999
Samples taken from low level residues ready for discarding 70-0001 to 70-9999
Samples taken from miscellaneous materials not previously described 90-0001 to 90-9999

The following steps briefly describe the use of the batch numbering system as the materials move from one process stage to another:
(a) Feed materials are separated into lots or batches. Specific numbers in numerical sequence are assigned to each lot or batch.
(b) When feed materials move into the foundry and become casting charges, they lose their batch identity. At this point, a casting process control number is assigned replacing the feed process control number. The casting process number identifies the ingot until it is rolled and cut into smaller rectangular shapes.
(c) Each rectangle is assigned a process control number after it is cut from the sheet. This number will identify the part throughout the production process.
(d) Part serial numbers are assigned after the part completes final inspection.
(e) Residues generated from processing are collected and batched according to material type. At this time a sequential process control number is assigned which identifies the batch until the material is inserted in a continuous process or combined with other batches.
(f) The product from the recovery operation is a metal button from the reduction process. This button is assigned a process control number which identifies it until used in a casting charge.
Transmitting data

The next step in our data accumulation process is to get the information to the data processing centre and into the computer magnetic tape storage system. At present, we have three methods available and none are satisfactory from an efficiency standpoint when compared with the high speed capabilities of the computer.

These three methods are described as follows:
(a) The source documents are sent to the data processing centre where keypunch operators punch the data into cards. The data in the punched cards are then transcribed onto magnetic tape.
(b) The data are keypunched into a transmitter located in the production area. This machine is electrically connected with a standard keypunch machine in the data processing centre. The information in the cards prepared by this equipment is then transcribed onto magnetic tape.
(c) The data are recorded by a series of pencil marks on special cards. This is known as the "mark sense method". The marked cards are sent to the data processing area where the data are transcribed onto magnetic tape. Since there is no way to satisfactorily verify the data inscribed on the marked cards, accuracy may be questionable.

In this control system, methods (a) and (b) are used.

(a) The following source documents are audited for propriety, coded where necessary and sent to the data processing centre for keypunching:

<table>
<thead>
<tr>
<th>Form</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>388</td>
<td>Shipping, receiving</td>
</tr>
<tr>
<td>1</td>
<td>Feed storage control card</td>
</tr>
<tr>
<td>2</td>
<td>Transfer receipts</td>
</tr>
<tr>
<td>4</td>
<td>Form 4</td>
</tr>
<tr>
<td>5</td>
<td>Form 5</td>
</tr>
<tr>
<td>6</td>
<td>Form 6</td>
</tr>
<tr>
<td>7</td>
<td>Form 7</td>
</tr>
<tr>
<td>8</td>
<td>Measured discard report</td>
</tr>
<tr>
<td>9</td>
<td>Analytical requisition</td>
</tr>
<tr>
<td>10</td>
<td>Physical inventory report</td>
</tr>
</tbody>
</table>

(b) The data on the following documents are keypunched into transmitting equipment described in method (b).

<table>
<thead>
<tr>
<th>Form</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>3</td>
<td>Casting, rolling, shearing control card</td>
</tr>
<tr>
<td>4</td>
<td>Part movement from one processing stage to another (Process control card)</td>
</tr>
</tbody>
</table>

The method used to transmit the data to the data processing centre is dependent upon the operation, data processing equipment and management requirements.

The next subject for our consideration is how to report physical inventories. There are two generally accepted ways to record a physical inventory. These are briefly described as follows:
(a) Hand list all desired information about every item, batch or part actually observed on inventory.
(b) Compare observed items with a list of such items prepared from book inventory data determined two or three days before the end of the ac-

\footnote{Forms 1 to 13 are to be found in the Appendix.}
counting period. This will require a reconciliation involving 10 to 15 per cent of the items and is just as effective as the 100 per cent hand listed method. Also, it is faster and easier to do.

Both methods of reporting physical inventories are used in this system of control. The format for hand listing the physical inventory is shown in form 10. Essentially, a prelisting of a book inventory contains the same data. Note that the inventory form is designed to provide space for listing a variety of items likely to be found on an inventory. Only the appropriate columns will be filled in with the necessary descriptive data for each item. Generally, the following data are common to all items listed on the physical inventory.

(a) Inventory control number
(b) Process control number
(c) Net weight or volume
(d) SS material weights

Note that inventory control numbers are used. This number corresponds to a like number printed on a strip of coloured adhesive tape which is affixed to each inventory item at the time it is recorded on the physical inventory listing. Inventory control numbers serve two purposes: (1) they facilitate a search for a particular item that may have to be re-checked and (2) they provide a device for a quick visual check that all items are inventoried.

Report formats

An important part in the control system is report format design and report submission schedules. Generally, each of the three major users of the data want the information presented in a different manner and at a different time.

The best way to describe a format for a machine listing of data is by illustration. However, to show a format for all listings and reports in the control system is not within the scope of this paper. Illustrated in forms 11, 12 and 13 are examples of typical listings for each of the three major uses of the data. The same general format is used for other listings.

HOW THE DATA ARE USED

Once the data have been collected and stored on magnetic tape, the next and most important phase of our programme is to use the data to effect the desired control over the materials. As mentioned in the Introduction, use of the data for accountability and production control will be described in some detail but word limitations prohibit any such discussion of process control uses.

Reports and listings for accountability, production control and process control purposes are obtained from the data stored on computer magnetic tape. A separate computer programme is needed for each type report and listing. When a report is required, the computer system is activated by the programme and in a matter of minutes the required report is prepared.

The following reports and listings are prepared for accountability uses:
(a) Accounting ledger

This ledger is a series of machine listings that duplicate a hand posted accounting ledger. All transactions affecting material balance areas are shown; book inventories are calculated and printed; physical inventory values for each material balance area are listed, and physical inventory values are compared with book inventory values and the difference, which we call "material unaccounted for", is printed. This ledger becomes part of the overall facility accounting record and is usually prepared on a monthly basis. The ledger also is documentary proof that approved control procedures were exercised over the SS materials. The ledger serves as a ready reference when historical information about materials is needed and provides the basis for an audit. The material unaccounted for, determined by comparing book inventory values with physical inventory values, is a measure of control over the materials. Statistical programmes have been devised that allow us to predict when the material unaccounted for is in or out of control. When this value falls outside the control limits, management is alerted to the fact that there probably has been a breakdown in the control system. Usually, this will initiate a detailed investigation to determine the cause and, if one is not readily discovered, a complete shutdown and re-inventory of the area or entire facility may result.

(b) Discrepancy report

This report is a listing of items that are unaccounted for regardless of the cause. This list is prepared by having the computer compare all items listed on the beginning inventory, all items originated during processing and all items received from off-site with all items listed on the ending inventory, all items shipped off-site and all items scrapped or started in a continuous recovery process. Comparison is made on process batch number or part number, whichever is appropriate. If there have been no mistakes, the discrepancy report will show a zero balance, but this rarely happens. For example, these types of errors often appear on the discrepancy report: (1) An item is not listed on inventory. (2) Numbers are transposed. (3) Material moves from one control area to another without documentation. Each discrepancy is investigated and corrected. When this has been accomplished, the nuclear materials manager is reasonably assured that all discreet items and unprocessed residue batches have been accounted for. However, the control does not guarantee that the correct SS material values have been recorded for each item. This control gives management a system to pin-point errors and assign responsibility to the individual. When employees become aware of this, they do a better job of handling the SS materials and more carefully follow procedures for initiating accountability documents.

(c) Physical inventory listing

This listing shows the location and status of all SS materials in each area at a given point in time. The accountability function uses these data as supplementary information to the ledger and for determining material unaccounted for previously mentioned. This listing becomes a part of the historical accountability records.

(d) Listings for each shipment

These listings consist of a machine-prepared tabulation of all pertinent data for each fuel element on each shipment. The following information is
typical: (1) Fuel element serial number. (2) Material type. (3) Net weight. (4) SS net weight. (5) Uranium-235 weight. (6) Dimensions. Keypunch operators punch serial number identification into blank data processing cards for each item on the shipping list. The cards are used to instruct the computer which parts are to be tabulated on the shipping lists. Copies of the shipment listing are forwarded to the customer at the time of shipment. The customer verifies receipt of the shipment by signing one copy. The signed copy is returned to the shipper and becomes a permanent record of the shipment.

(e) Shipment listings summary

These summaries are tabulations of fuel elements shipped for a given accounting period. The information is summarized as follows: (1) Total number of fuel elements shipped, categorized by type. (2) Net weight of SS material. (3) Uranium-235 weight.

These data are used for internal reporting purposes and serve as a quick reference when this type of information is needed by management.

(f) Analytical requisition summary

This summary is a listing of the analysis results on all sample requisitions sent from processing areas to the laboratory. The information on the listing is as follows: (1) Requisition number. (2) Number of batch sampled. (3) The results from the analyses. (4) Weight of sample transferred. (5) SS material in sample. (6) Analytical method used.

These data are used by accountability to support calculations of SS material and alloy values of material transferred internally, on physical inventories and shipped to customers.

(g) Measured waste discard report

This report lists all batches of material that are uneconomical to recover and are discarded. Information would be material type, volume or weight, analytical requisition number and results, uranium content and percentage of uranium-235.

It is a function of the statistical department to provide reasonable assurance that measurements made to determine quantities of SS materials are valid. Results from analytical, emission spectrographic and mass spectrographic analyses made on samples from production materials are listed and compared to like analyses made on standard control samples. Data from check weighing and calibration programmes along with the analysis results are statistically evaluated to determine the accuracy of the measurement programmes. This data is also used to calculate "confidence limits" determinations for any particular measurement sequence. This programme is particularly important to support the facility position when shipper-receiver differences exist regarding SS material content of feed or finished product.

Material unaccounted for determinations, assurance of control over parts and batches and statistical measurement evaluations are tools to aid management in determining if the control over the SS materials is adequate. Often a breakdown of control in an area will be pin-pointed in time for corrective action before substantial losses are incurred. These data are also used to estimate losses of SS materials in dilute residues that are uneconomical to recover. These values must be known within a reasonable degree
of accuracy since they are an important cost factor when making bids for fuel element fabrication contracts.

The listings and machine reports for production control purposes are outlined as follows:

(a) Material in process

The frequency of this report is daily and indicates the number of ingots and parts in each stage of processing at the close of the working day. These data are used by the production manager to evaluate past production performance and prepare future operating schedules. Sources of trouble causing production to lag behind schedule are identified allowing corrective action to be taken before the situation gets out of control.

(b) Material in feed storage

The frequency of this report is daily and itemizes each batch of feed material available for casting at the end of the working day. The listings will include batch, lot or item identification, uranium weight, per cent of uranium-235 enrichment, per cent alloy (if appropriate), and per cent impurities. These data are used by the production manager for preparation of schedules and future orders to feed material suppliers.

(c) Parts scrapped

The frequency of this report is daily and shows the type part scrapped, the reason for scrapping, the process operation, date and the identity of the individual working on the part when it was scrapped. This information is used to determine process and employee efficiency and is another factor for determining future schedules.

(d) Finished fuel elements available for shipment

The frequency of this report is weekly and describes each finished fuel element on hand that is ready for shipment as of the end of the day or week. Pertinent data such as fuel element serial number, net weight, uranium net weight, per cent of uranium-235 enrichment, uranium-235 weight, per cent alloy, alloy weight and type codes are listed. The production manager uses this information to plan succeeding shipments and as an aid in making future commitments.

(e) Fuel elements shipped

The frequency of this report is weekly and is on the same format as the listing of fuel elements available for shipment. In addition, the customer or recipient is identified. These data are used by the production manager to determine if the shipments to customers are on schedule and as another tool for future schedule preparation.

(f) Feed materials received from off-site

The frequency of this report is weekly and describes the feed received and is used to determine if suppliers are meeting their commitments.

(g) Feed materials from recovery process

The frequency of this report is weekly and describes the feed metal produced by recovery operations. Since casting feed is the end product of the recovery process, these data are used by the production manager to evaluate the efficiency of the recovery operation and to schedule future recovery production.
(h) Scrap materials available for processing

The frequency of this report is monthly and categorizes all scrap batches available for starting in the recovery process as of the month's end. These data are used to schedule the next month's recovery operation.

Process control

Process and product control procedures require a great amount of data that cannot be economically obtained through a data gathering system primarily designed for accountability and production control. However, some of the data collected for the primary purpose may be used by process control. The information available from the control system useful to process control engineers is defined as follows:

(a) The results of chemical, emission spectrographic and mass spectrographic analyses.
(b) Radiographic results.
(c) Number of good parts produced.
(d) Number of parts rejected and cause for rejection.
(e) Number of castings made.
(f) Number of castings rejected.
(g) Time and temperature from casting operation.
(h) Temperatures and pressures from rolling and shearing operation.

The above data are obtained for process control in two ways. If the information is used with other data in a computer orientated programme, the data will be extracted from the computer magnetic tape storage and used as required in the other programmes. The best example of this is the use of the chemical, emission spectrographic and mass spectrographic analysis data to statistically evaluate whether the product meets purity, uranium content and alloy content specifications.

The other method is to instruct the computer to print out the information in the form of a machine listing. The information on these lists is outlined as follows:

(a) The machine listing for the number of good parts produced describes all parts produced during a period of time, usually on a weekly or monthly basis. Usually the list describes the part by serial number, type, per cent alloy, net weight, uranium weight, uranium-235 weight and ingot reference.

(b) The listing of parts rejected describes the part rejected and the reason for the reject. The data listed are part number, part type and rejection code.

(c) The listing showing number of castings produced describes this information by casting number, type material in casting charge, weight of material in charge, temperature of metal at time of pouring, weight of alloy in charge, time required to melt casting charge, analytical requisition reference, per cent alloy, per cent uranium and per cent uranium-235.

(d) The listing of casting rejects is a recapitulation of all ingots failing because of improper amount of alloy or excessive impurity content. Information on this listing will be the same as under item (c) except that the reason for the rejects will also be shown.
(e) The listing for rolling mill data contains metal temperature and rolling mill pressures for each ingot passed through the rolling mill. Ingots are described by ingot number, type, per cent alloy, per cent uranium and per cent uranium-235.

Process control engineers use these listings along with other data to evaluate processing techniques, identify sources of trouble and as background information for developing new and better processing methods.

SUMMARY

A method of collecting and disseminating a variety of data for effective accountability, production control and process control over materials and process operation found in a typical uranium-fuel-element fabricating facility has been presented. The discussion and illustrations are focused on portions of the control system that are important in initiating a new system or when converting from an electrical-accounting machine system to automatic data processing methods. Since it is not possible to discuss all facets of the control system, the intent was to present enough information to provide guidelines to those interested in establishing a control system for the first time or in modernizing an already established system.
<table>
<thead>
<tr>
<th>Column</th>
<th>Details</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.</td>
<td>Transfer Series: A. B. C. (From) (To) (Number)</td>
</tr>
<tr>
<td>2.</td>
<td>Shipped By (Shipper): Name Address</td>
</tr>
<tr>
<td>3.</td>
<td>Shipped to (Receiver): Name Address</td>
</tr>
<tr>
<td>5.</td>
<td>Shipped To Account Of Lic. No. Name Address (For AEC Use Only): Lease No. Order No.</td>
</tr>
<tr>
<td>6.</td>
<td>(For AEC Use Only): Nuclear Material Draft (Form AEC-437) Number</td>
</tr>
<tr>
<td>7.</td>
<td>SS Material (Check One): (A) Enriched Uranium (B) Uranium-233 (C) Plutonium (D) Other</td>
</tr>
<tr>
<td>8.</td>
<td>Material Descriptions:</td>
</tr>
<tr>
<td>9.</td>
<td>This Transfer Involves: (A) Initiating Lease Responsibility (B) Transfer of Lease Responsibility (C) Return to AEC for Credit (D) No Change in Lease Responsibility (Transfer of Material Only)</td>
</tr>
<tr>
<td>10.</td>
<td>Material Quantities: A. Weight Units:</td>
</tr>
<tr>
<td>B.</td>
<td>Container No.</td>
</tr>
<tr>
<td>C.</td>
<td>Piece Count</td>
</tr>
<tr>
<td>D.</td>
<td>Gross Weight</td>
</tr>
<tr>
<td>E.</td>
<td>Tare Weight</td>
</tr>
<tr>
<td>F.</td>
<td>Net Weight</td>
</tr>
<tr>
<td>G.</td>
<td>Element Weight</td>
</tr>
<tr>
<td>H.</td>
<td>Weight % Isotope</td>
</tr>
<tr>
<td>I.</td>
<td>Isotope Weight</td>
</tr>
<tr>
<td>Totals</td>
<td></td>
</tr>
<tr>
<td>11.</td>
<td>The Above Items and Quantities Shipped On, 19 (Shipper's Signature)</td>
</tr>
<tr>
<td>12.</td>
<td>The Above Items and Quantities Received On, 19 (Receiver's Signature)</td>
</tr>
</tbody>
</table>
**FORM 2. Feed storage control card**

<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Shipper-Receiver Diff. - Net Wt.</td>
<td>U-235</td>
<td>Dr. Acct.</td>
<td>Acct. Title</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Sample Req. No.</th>
<th>% U-235</th>
<th>% Impurities</th>
</tr>
</thead>
</table>

<table>
<thead>
<tr>
<th>Weighed By</th>
<th>Date (Day of yr.)</th>
<th>Checked By</th>
<th>Date (Day of yr.)</th>
</tr>
</thead>
</table>

**Columns:**
- **Shipper's Control Number**
- **Receiver's Uranium Code**
- **Shipper's Net Uranium Weight**
- **Receiver's U-235 Code**
- **Shipper's U-235 Weight**
- **Shipper's Container Number**
- **Receiver's Control No.**
<table>
<thead>
<tr>
<th>Charge Preparation</th>
<th>Rolling and Shearing</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Operation No.</strong></td>
<td><strong>Operator No.</strong></td>
</tr>
<tr>
<td><strong>Process No.</strong></td>
<td><strong>MTC</strong></td>
</tr>
<tr>
<td><strong>Uranium Code</strong></td>
<td><strong>Furnace No.</strong></td>
</tr>
<tr>
<td><strong>U-235 Code</strong></td>
<td><strong>Crucible No.</strong></td>
</tr>
<tr>
<td><strong>Alloy Elem No-1 Code</strong></td>
<td><strong>Mold No.</strong></td>
</tr>
<tr>
<td><strong>Alloy Elem No-2 Code</strong></td>
<td><strong>Time of Heat</strong></td>
</tr>
<tr>
<td><strong>Total Charge Wt.</strong></td>
<td><strong>Date of Pour</strong> (Day of Year)</td>
</tr>
<tr>
<td><strong>Temperature of Melt</strong></td>
<td></td>
</tr>
<tr>
<td><strong>Date Rolled and Sheared</strong> (Day of Year)</td>
<td></td>
</tr>
<tr>
<td><strong>Net Wt. of Ingot</strong></td>
<td><strong>Sample Req. No.</strong></td>
</tr>
<tr>
<td>% <strong>Uranium</strong></td>
<td><strong>Density</strong></td>
</tr>
<tr>
<td>% <strong>U-235</strong></td>
<td>% <strong>Impurities</strong></td>
</tr>
<tr>
<td>% <strong>Alloy Elem No-1 Code</strong></td>
<td>% <strong>Alloy Elem No-2 Code</strong></td>
</tr>
</tbody>
</table>

**Remarks:**

**Supervisor’s Sig.:**
# PROCESS CONTROL CARD

<table>
<thead>
<tr>
<th>Field</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>Initial Process No.</td>
<td></td>
</tr>
<tr>
<td>New Process No.</td>
<td></td>
</tr>
<tr>
<td>Uranium Code No.</td>
<td></td>
</tr>
<tr>
<td>U-235 Code No.</td>
<td></td>
</tr>
<tr>
<td>Alloy Code No.</td>
<td></td>
</tr>
<tr>
<td>SS Net Wt. Uranium</td>
<td>Grams</td>
</tr>
<tr>
<td>Net Wt. U-235</td>
<td>% Uranium</td>
</tr>
<tr>
<td>Net Wt. Alloy</td>
<td>% U-235</td>
</tr>
<tr>
<td>Net Wt. Alloy</td>
<td>% Alloy</td>
</tr>
<tr>
<td>Total Net Wt.</td>
<td>% Impurities</td>
</tr>
<tr>
<td>Length Code No. 1</td>
<td></td>
</tr>
<tr>
<td>Width Code No. 2</td>
<td></td>
</tr>
<tr>
<td>Thickness Code No. 3</td>
<td></td>
</tr>
<tr>
<td>Radiography</td>
<td>Sample Req. No.</td>
</tr>
<tr>
<td>Accepted Code No. 1</td>
<td></td>
</tr>
<tr>
<td>Rejected Code No. 2</td>
<td></td>
</tr>
<tr>
<td>Credit Acct.</td>
<td>Acct. Title</td>
</tr>
<tr>
<td>Debit Acct.</td>
<td>Acct. Title</td>
</tr>
<tr>
<td>Transferor Sig.</td>
<td></td>
</tr>
<tr>
<td>Transferee Sig.</td>
<td></td>
</tr>
<tr>
<td>Date (Day of yr.)</td>
<td></td>
</tr>
</tbody>
</table>

FORM 4. Process control card
## FORM 5. Transfer receipt

<table>
<thead>
<tr>
<th>Line No.</th>
<th>Process Control Number</th>
<th>Element Identifications</th>
<th>Size Code (Grams)</th>
<th>Net Wt. of Plate (Grams)</th>
<th>% Uranium</th>
<th>% U-235</th>
<th>Net Wt. Alloy #1</th>
<th>Net Wt. Alloy #2</th>
<th>Net Wt. Alloy #3</th>
<th>Net Wt. Alloy #4</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>2</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>3</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>4</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>5</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>6</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>7</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>8</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>9</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>10</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>11</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>12</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>13</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>14</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>15</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>16</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>17</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>18</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>19</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>20</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

**Totals:**
- Net Wt.
- Total Size Grams
- Total U-235
- Total Alloy #1
- Total Alloy #2

**WTC:**

<table>
<thead>
<tr>
<th>Cr Accr</th>
<th>Dr Accr</th>
<th>Acct Title</th>
<th>Signature Transferor</th>
</tr>
</thead>
</table>

**Transaction Code:**
## SALVAGE PREPARATION BATCH CARD

**Prepared on Date (Day of Year):** 

<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>2</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>3</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>4</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>5</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>6</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>7</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>8</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>9</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>10</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>11</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>12</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

**TOTAL:** 

- Sample Req. No. 
- Salvage Operation No. 
- Salvage Batch Prepared by Operator No. 
- Remarks: 

- **Total Volume or Weight:** 
  - % Uranium 
  - % U-235 
  - % Alloy # 1 
  - % Alloy # 2 
  - % Impurities 
  - Credit Account 
  - Debit Account 

**Remarks:** 

**Salvage Batch Approved by Supervisor (Signature):** 

**Date (Day of Year):** 

**FORM 6. Salvage preparation batch card**
## Scrap Receiving Report

<table>
<thead>
<tr>
<th>Line No.</th>
<th>Receiver's Item Number</th>
<th>Shipper's Item Number</th>
<th>Receiver's Net Wt.</th>
<th>Shipper's Net Wt.</th>
<th>Shipper's U-235 Wt.</th>
<th>Shipper's Alloy No. 1</th>
<th>Shipper's Net Wt.</th>
<th>Receiver's % Uranium</th>
<th>Receiver's % U-235</th>
<th>Receiver's % Alloy No. 1</th>
<th>Receiver's Net Wt.</th>
<th>Receiver's % Alloy No. 2</th>
<th>Receiver's Net Wt.</th>
<th>Receive's % Alloy No. 2</th>
<th>Credit Acc #</th>
<th>Debit Acc #</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>2</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>3</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>4</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>5</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>6</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>7</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>8</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>9</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>10</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>11</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>12</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>13</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>14</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>15</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>16</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>17</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>18</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>19</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>20</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>21</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>22</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>TOTALS</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

**Total Received**

**Shipper-Receiver Difference**

Complied By: ____________________________

Approved By: ____________________________

---

**FORM 7. Scrap receiving report**
### FORM 8. Measured discard report

<table>
<thead>
<tr>
<th>Lane No.</th>
<th>Process Number</th>
<th>Volume</th>
<th>MTC</th>
<th>Sample No.</th>
<th>Viscosity Code</th>
<th>% Viscosity</th>
<th>Net Wt. Uranium</th>
<th>% U-235</th>
<th>Net Wt.</th>
<th>% U-235</th>
<th>Operation Number</th>
<th>Operator Number</th>
<th>Date</th>
<th>Day of Year</th>
<th>Operator Signature</th>
<th>Supervisor's Signature</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>2</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>3</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>4</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>5</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>6</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>7</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>8</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>9</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>10</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>11</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>12</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>13</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>14</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>15</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>16</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>17</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>18</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>19</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>20</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>21</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>22</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>23</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>24</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>25</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>26</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>27</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>28</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>29</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>30</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Total Uranium: 
Total U-235: 
Credit Acct: 
Debit Acct: 

Date of Report (Day of Year): [ ]
### REQUEST FOR ANALYSIS

<table>
<thead>
<tr>
<th>Dept.</th>
<th>Date</th>
<th>Batch or Process No.</th>
<th>Req. No.</th>
<th>Cont. No.</th>
<th>CHEMICAL LAB.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Chg. No.</td>
<td>Time</td>
<td>Sampled By</td>
<td></td>
<td>G</td>
<td>T</td>
</tr>
<tr>
<td>Matl. Type Code</td>
<td></td>
<td>Weight</td>
<td>G/G</td>
<td>Method</td>
<td></td>
</tr>
<tr>
<td>Matl. Description</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

**EMISSION SPEC. LAB.**

<table>
<thead>
<tr>
<th>Batch or Process No.</th>
<th>Sp. Gr.</th>
<th>VOLUME</th>
<th>G/L</th>
<th>PPM</th>
</tr>
</thead>
<tbody>
<tr>
<td>Plate No.</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Ca</td>
<td>Mg</td>
<td>*</td>
<td></td>
<td></td>
</tr>
<tr>
<td>B</td>
<td>Cu</td>
<td>*</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Fe</td>
<td>Al</td>
<td>*</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Li</td>
<td>Sl</td>
<td>*</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Mn</td>
<td>Cd</td>
<td>*</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Ni</td>
<td>Mo</td>
<td>*</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Cr</td>
<td>Sn</td>
<td>*</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Ca</td>
<td>Pb</td>
<td>*</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Na</td>
<td>K</td>
<td>*</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Wt %</th>
<th>C</th>
<th>H2O</th>
<th>F</th>
<th>NO3</th>
<th>O2</th>
<th>Pu</th>
<th>H2</th>
<th>AOI</th>
<th>Fe</th>
<th>Zr</th>
<th>Al</th>
<th>Mo</th>
<th>W</th>
</tr>
</thead>
<tbody>
<tr>
<td>235</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

**Remarks:**

---

**DATE**

**Approved**

---

**FORM 9. Request for analysis**
### INVENTORY REPORT

**Acct. No.**

**Acct. Title**

<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>2</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>3</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>4</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>5</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>6</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>7</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>8</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>9</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>10</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>11</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>12</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>13</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>14</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>15</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>16</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>17</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>18</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>19</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>20</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>21</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>22</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>23</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>24</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>25</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

**Taken By: ___________________________  Checked By: ___________________________  Approved By: ___________________________

**FORM 10. Physical inventory report**
<table>
<thead>
<tr>
<th>Column</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Inventory Control Number</td>
</tr>
<tr>
<td>2</td>
<td>Process Control Number</td>
</tr>
<tr>
<td>3</td>
<td>Fuel Element Type</td>
</tr>
<tr>
<td>4</td>
<td>Fuel Element Serial Number</td>
</tr>
<tr>
<td>5</td>
<td>Element Type Code</td>
</tr>
<tr>
<td>6</td>
<td>Material Type Code</td>
</tr>
<tr>
<td>7</td>
<td>Measurement Method</td>
</tr>
<tr>
<td>8</td>
<td>Analytical Requisition Number</td>
</tr>
<tr>
<td>9</td>
<td>Net Weight or Volume</td>
</tr>
<tr>
<td>10</td>
<td>Analytical Answer - g/g or g/l</td>
</tr>
<tr>
<td>11</td>
<td>Analytical Method Code</td>
</tr>
<tr>
<td>12</td>
<td>Net Uranium Weight</td>
</tr>
<tr>
<td>13</td>
<td>Percent U-235</td>
</tr>
<tr>
<td>14</td>
<td>U-235 Weight</td>
</tr>
<tr>
<td>15</td>
<td>Alloy No. Element Code</td>
</tr>
<tr>
<td>16</td>
<td>Percent Alloy No. 1</td>
</tr>
<tr>
<td>17</td>
<td>Alloy No. 1 Net Weight</td>
</tr>
<tr>
<td>18</td>
<td>Alloy No. 2 Element Code</td>
</tr>
<tr>
<td>19</td>
<td>Percent Alloy No. 2</td>
</tr>
<tr>
<td>20</td>
<td>Alloy No. 2 Net Weight</td>
</tr>
</tbody>
</table>

FORM 11. Physical inventory listing format
### DAILY PARTS IN PROCESS REPORT (PRODUCTION CONTROL)

<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

1. Process Control Number
2. Fuel Element Type
3. Fuel Element Serial Number
4. Number of ingots available for rolling and shearing
5. Number of fuel elements rolled and sheared
6. Number of fuel elements in Radiography
7. Number of fuel elements in Machining Operation No. 1
8. Number of fuel elements in Inspection No. 1
9. Number of fuel elements in Machining Operation No. 2
10. Number of fuel elements in Inspection No. 2
11. Number of fuel elements in Machining Operation No. 3
12. Number of fuel elements in Inspection No. 3
13. Number of fuel elements in Plating
14. Number of fuel elements in Final Inspection
15. Number of fuel elements finished available for shipment

**FORM 12.** Daily parts in process report format
<table>
<thead>
<tr>
<th>Control Spec.</th>
<th>Ag</th>
<th>Al</th>
<th>B</th>
<th>Ba</th>
<th>Be</th>
<th>Bi</th>
<th>Ca</th>
<th>Cd</th>
<th>Co</th>
<th>Cr</th>
<th>Cu</th>
<th>Fe</th>
<th>Ga</th>
<th>In</th>
</tr>
</thead>
<tbody>
<tr>
<td>Req. No.</td>
<td>Non-Control Sample Code Casting Plate No.</td>
<td>Sn</td>
<td>Sr</td>
<td>Ta</td>
<td>Th</td>
<td>Ti</td>
<td>U</td>
<td>V</td>
<td>W</td>
<td>Zn</td>
<td>Zr</td>
<td>C</td>
<td>Am</td>
<td>Ga</td>
</tr>
</tbody>
</table>

1. Analytical Requisition Numbers
2. Control Non-Control Sample Code
3. Casting Number Sample Taken From
4. Emission Specification Plate Number
5 - 18. Impurity Listings
19. Sum of the Impurities subtracted from 1.00

Listing of impurities is in same format as element identification symbols. For example:
If Column 5 shows the following: 5

Ag
K
Sn

10
5
20

the impurity values are Ag - 10 ppm, K - 5 ppm and Sn - 20 ppm.

FORM 13. Listing of impurities determined by emission spectrographic analysis format
DISCUSSION

W. J. WRIGHT: To what extent does your organization use quality control charts to predict and control long-term systematic process variations, as distinct from short-term random variations?

L. D. HAZELTON: Quality control charts are used for long-term predictions to the extent that control limits are established for specific product specifications for the life of the particular production run. The production run may be for six months, one year or even two years. By comparing monthly data — collected according to set procedures — with the quality control limits we can tell with 95% certainty that all items produced within the period will fall within the product specifications. This does not guarantee that 100% of the items are within the specifications. If the customer desires a 100% guarantee as to product specifications, he must be prepared to pay for the extensive sampling and analytical work required.

"Material unaccounted for" (MUF) — i.e. quantities arising from bias in sampling and analysis and from human error for a given quantity of product throughput — are predicted. When the MUF is within the control limits we feel that adequate weight control has been exercised. This does not apply to unit control, which requires accountability for 100% of the items.

F. TINGEY: Routine gamma scanning to determine the $^{235}$U content of individual fuel elements received for use in the Materials Testing Reactor and the Engineering Test Reactor has indicated frequently a shipper-receiver difference as large as 4%, when the fabricator determined the fuel element loading by density measurements on individual fuel plate cores, by gamma counting of the cores or by chemical analysis of dip samples from the melt. On one contract the fabricator's measurement on a lot in excess of 600 elements was approximately 2% low.
MANAGEMENT AND CONTROL OF EBR-II FUEL

G.K. WHITHAM, T.R. SPALDING AND M.J. FELDMAN
ARGONNE NATIONAL LABORATORY, IDAHO FALLS, IDAHO, UNITED STATES OF AMERICA

Abstract — Résumé — Аннотация — Resumen

MANAGEMENT AND CONTROL OF EBR-II FUEL. Unique problems relating to the management and inventory of fuel are being solved in the operation of Experimental Breeder Reactor II (EBR-II) at the National Reactor Testing Station in Idaho. The EBR-II plant complex incorporates an on-site facility, adjoining the reactor plant, for pyrometallurgically reprocessing spent fuel. Fuel removed from the reactor and also fuel which has been reprocessed by the Fuel Cycle Facility is highly radioactive, requiring remote handling at every step of the procedure used to accomplish removal, processing and return to the reactor. An additional complication is encountered in that fuel handling mechanisms, as well as the reactor, pumps and heat exchanger, operate submerged in a large tank containing sodium (86 000 gal) at a temperature of 700°F.

A digital computer which receives intelligence from punched cards controls these "under-sodium" transfers. Accurate information on each loading on the 636 reactor grid positions is maintained and controlled by computer tabulation of the punched cards. All loading changes are programmed by punched cards, processed and checked by computer methods. Fuel burn-up for all sub-assemblies at each loading change is automatically computed along with time available to maximum burn-up for any sub-assembly. Thus, a complete inventory of fissionable materials is maintained by computer control. Computer analysis and control of the number and frequency of reactor loading changes programmes operations to realize maximum plant utilization.

In the Fuel Cycle Facility, fuel burn-up information is converted to data which forms the basis for replenishment of uranium-235. The ingot weight of refined fuel plus dross is compared to the incoming fuel weight for process control. When the refined fuel then enters this stage of fabrication, the weight of fuel pins and scrap is carefully accounted for. A punched data card for each fuel pin is automatically produced when the cast pins are measured and weighed for acceptance. This card follows the fuel pin through the entire fabrication process, thus furnishing a complete history and data record for each of the 91 elements in a sub-assembly. One of the bits of information on these cards furnishes a precise account of an element's position in a particular sub-assembly. Data on fabrication are therefore useful as a comparison when making post-irradiation physical measurements and other evaluations on individual pins.

Accumulated data on the punched cards are programmed into the computer to provide additional control data for accountability, studies of process efficiency and statistical control of process variables.

GESTION ET CONTROLE DU COMBUSTIBLE UTILISE DANS LE REACTEUR EBR-II. L'exploitation du réacteur surgénérateur expérimental II (EBR-II) à la Station nationale d'essai des réacteurs (Idaho) permet actuellement de résoudre les problèmes très particuliers que posent la gestion et l'inventaire des combustibles. Sur le site même de la centrale nucléaire où fonctionne le réacteur EBR-II se trouve, à proximité du réacteur, une installation permettant d'effectuer le traitement du combustible par pyrométallurgie. Le combustible déchargé du réacteur, de même que le combustible qui a été traité dans cette installation, est fortement radioactif et doit être télémanipulé pour toutes les opérations, qu'il s'agisse de son déchargement du réacteur, de son traitement ou de son rechargement dans le réacteur. On se heurte à une autre complication due au fait que les télémanipulateurs, de même que le réacteur, les pompes et l'échangeur de chaleur, sont immergés dans une grande cuve contenant 325 900 litres de sodium porté à une température de 370°C.

Une calculatrice numérique qui reçoit les informations sur cartes perforées contrôle les transferts qui s'opèrent «sous sodium». Des informations sur chacun des chargements effectués à l'une des 636 positions de la "grille" du réacteur sont entretenues, et leur précision est contrôlée par traitement des cartes perforées dans la calculatrice. Toutes les modifications de la charge sont consignées sur cartes perforées qui sont ensuite traitées, et elles sont vérifiées dans la calculatrice. A chaque modification de la charge, le taux de combustion pour tous les sous-assemblages est calculé automatiquement ainsi que le délai dans lequel l'irradiation maximale est atteinte pour l'un quelconque des sous-assemblages. On dispose ainsi, par contrôle à la calculatrice, d'un inventaire complet des matières fissiles. L'analyse et le contrôle par la calculatrice du nombre et de la fréquence des modifications de la charge du réacteur permettent d'établir un programme d'opérations assurant une utilisation maximale de la centrale.
Dans l'installation de traitement du combustible, les informations sur l'irradiation de combustion sont transformées en données qui servent à déterminer les besoins en uranium-235. Pour contrôler les processus, on compare le poids du lingot de combustible raffiné et des déchets au poids de combustible reçu. Lorsque le combustible raffiné parvient au stade de fabrication des barreaux, on détermine soigneusement le poids des barreaux et des déchets. On établit automatiquement une carte perforée pour chaque barreau de combustible lorsqu'on procède aux mesures et aux pesées de contrôle des barreaux coulés. Ces cartes perforées suivent toutes les étapes de fabrication des barreaux de combustible et fournissent ainsi tous les renseignements nécessaires sur chacun des 91 éléments d'un sous-assemblage. L'une des informations portées sur ces cartes indique de façon précise la position d'un élément dans un sous-assemblage déterminé. Ces données de fabrication servent donc utilement de base de comparaison lorsqu'on procède, après irradiation, sur certains barreaux de combustible, à des mesures physiques et à diverses autres évaluations.

Les informations consignées sur les cartes perforées sont traitées à la calculatrice afin d'obtenir des renseignements supplémentaires utilisés pour le contrôle de la comptabilité des matières, l'étude du rendement des procédés et le contrôle statistique des variables de ces procédés.

ОБРАЩЕНИЕ С ТЕПЛОВЫДЕЛЯЮЩИМИ ЭЛЕМЕНТАМИ ЭКСПЕРИМЕНТАЛЬНОГО РЕАКТОРА-РАЗМНОЖИТЕЛЯ EBR-II И ИХ УЧЕТ. Исключительные по своему характеру проблемы, связанные с административно-хозяйственным учетом и инвентаризацией тепло- выделяющих элементов получают свое разрешение при эксплуатации экспериментального реактора-размножителя (EBR-II) на Национальной реакторной испытательной станции в Айдахо. Комплекс реактора EBR-II включает в себя смежную с реакторным помещением установку для пирометаллургической переработки отработанного ядерного топлива. Вынутое из реактора ядерное топливо, а также ядерное топливо, переработанное на установке по топливному циклу, обладают сильной радиоактивностью и требуют дистанционного обращения на каждой ступени операций по изъятию топлива из реактора, его переработке и возвращении в реактор. Дополнительное затруднение возникает из-за того, что механизмы по выгрузке, перемещению и загрузке топлива, такие как и сам реактор, насосы и теплообменник работают в погруженном состоянии в большом баке, наполненном натрием (86 000 галлонов) при температуре 700° Фаренгейта.

Цифровое электронное счетно-решающее устройство, получавшее информацию с перфорированных карточек, управляло этими перемещениями "под жидким натрием". Производимая счетно-решающим устройством табличная запись перфорированных карточек позволяет автоматически вести и контролировать точную информацию по каждой загрузке 636 позиций реакторной решетки. Все изменения загрузки программируются на основании перфорированных карточек, обрабатываются и проверяются автоматически при помощи счетно-решающего устройства. При каждом изменении загрузки выгорание топлива для каждой подборки подсчитывается автоматически одновременно со временем, необходимым каждой подборке для достижения максимального выгорания. Таким образом контроль при помощи счетно-решающего устройства позволяет вести непрерывный учет всех перемещений топлива и частоты изменений реакторной загрузки при помощи счетно-решающего устройства позволяет составлять программу эксплуатации для достижения наилучшего использования установки.

На установке по топливному циклу сведения по выгоранию ядерного топлива преобразуются в данные, служащие основой для пополнения урана-235. Вес переработанного топлива плюс отходы сравниваются с весом поступающего на переработку топлива, что позволяет регулировать технологию процесса. В процессе переработки на соответствующей стадии точно учитывают вес топливных стержней и лома. При измерении и взвешивании отлитых стержней на предмет их приемки для каждого стержня автоматически составляется перфорированная карточка со всеми данными. Эта карточка сопровождает соответствующий топливный стержень на протяжении всего технологического процесса изготовления, предоставляя таким образом полные сведения и зарегистрированные данные для каждого из 91 элементов подборки. Одни из заказов в двоичной системе информации на этих карточках дает точное указание о позиции каждого элемента в соответствующей сборке. Таким образом данные о ходе изготовления могут быть полезными для сопоставления при проведении физических промеров после облучения и составлении других оценок по каждому отдельному стержню.

Данные, накопленные на перфорированных карточках, служат для составления программы для счетно-решающего устройства с целью предоставления дополнительных контрольных данных для отчетности, изучения эффективности технологических процессов и для статистического контроля над участвующими в процессах переменными величинами.
ADMINISTRACION Y CONTROL DEL COMBUSTIBLE EMPLEADO EN EL EBR-II. La explotación del Reactor Reproductor Experimental II (EBR-II) de la National Reactor Testing Station de Idaho ha permitido resolver algunos de los problemas más singulares que plantea la administración y el inventario de combustibles. La planta del EBR-II comprende una instalación contigua al reactor destinada a la regeneración pirometalúrgica del combustible agotado. El combustible extraído del reactor y el que ha sido regenerado en la Fuel Cycle Facility son altamente radioactivos y exigen la manipulación a distancia en todas las etapas del procedimiento empleado para su extracción, tratamiento y retorno al reactor. Otra complicación consiste en que tanto los mecanismos para la manipulación del combustible como el reactor, las bombas y el intercambiador de calor están sumergidos en un tanque de grandes dimensiones que contiene sodio (325 900 litros) a 370°C.

Una calculadora numérica, con información suministrada por tarjetas perforadas, controla todas estas operaciones. La calculadora tabula las tarjetas y de esta manera se mantiene y controla una información exacta sobre cada una de las 636 posiciones correspondientes al retículo del reactor. Todos los cambios de carga se programan con ayuda de tarjetas perforadas, se sistematizan, y se verifican mediante la calculadora. En cada cambio de carga se calcula automáticamente el grado de combustión de todos los submontajes y el tiempo disponible en cualquiera de ellos hasta llegar al grado de combustión máximo. El control por calculadora permite así mantener un inventario completo de los materiales fisionables. Si se analizan y controlan mediante la calculadora el número y la frecuencia de los cambios en la carga del reactor, las operaciones pueden programarse de modo que se logre la máxima utilización de la planta.

En la Fuel Cycle Facility la información relativa al grado de combustión sirve para obtener los datos que se utilizan como base para el reabastecimiento en uranio-235. Para controlar el proceso, el peso del lingote de combustible refinado, más la escoria, se compara con el peso del combustible que entra. Cuando el combustible refinado llega luego a esta etapa de la fabricación se determina con sumo cuidado el peso de las varillas y de los recortes. Al pesar y medir las varillas coladas para ver si son aceptables se perfora automáticamente una tarjeta para cada varilla con los datos correspondientes. Estas tarjetas acompañan las varillas durante todo el proceso de fabricación y así se obtiene una historia completa y un registro de datos para cada uno de los 91 elementos que componen un submontaje. Uno de los datos que figuran en estas tarjetas indica con exactitud la posición del elemento en un submontaje determinado. Por consiguiente, los datos sobre la fabricación son útiles a título comparativo cuando después de la irradiación se efectúan mediciones físicas y otras evaluaciones en cada una de las varillas.

Con los datos acumulados en las tarjetas perforadas se programa la calculadora a fin de obtener datos adicionales de control para la contabilidad, para los estudios sobre el rendimiento de los procesos y para la verificación estadística de sus variables.

The Experimental Breeder Reactor II (EBR-II), located at the National Reactor Testing Station in Idaho, is the first fast power breeder in the United States Atomic Energy Commission programme to demonstrate operation on a closed fuel cycle [1-4]. Fuel is moved from the reactor to an adjacent Fuel Cycle Facility for pyrometallurgical reprocessing, re-enrichment, and refabrication. These combined facilities are designed for a high rate of fuel turnover and minimum fuel inventory, originally planned to be 1 1/2 core loadings (300 kg $^{235}$U). However, the programming for experiments may require an increase in inventory to as much as 2 1/2 core loadings.

FACILITY DESCRIPTION

The EBR-II complex (Fig. 1) consists of four separate buildings and a radiochemical laboratory for fuel and materials analysis. The reactor is located in a cylindrical steel containment building, along with its primary sodium coolant system and associated auxiliaries.

The Fuel Cycle Facility (FCF) adjoins the reactor building, with an interconnecting air lock for transfers of radioactive fuel.

The Sodium Boiler Plant contains sodium-to-water steam generators which produce high-pressure, superheated steam,
The Power Plant houses the main control room, the 20-MW(e) turbine generator, and the steam system auxiliaries.

PRIMARY TANK AND REACTOR

The reactor and its primary cooling system, including the main heat exchanger and primary pumps, are contained in a large vessel (primary tank) and operate submerged in 80 000 gal (US) of radioactive sodium at 700°F. This concept of a submerged system provides highly reliable containment for the primary coolant; however, it requires fuel handling equipment that operates in a sodium environment (Fig. 2).
The reactor consists of an enriched core, 19-in. diam. and 14 in. high, surrounded by 30 tons of fertile blanket material. Individual fuel subassemblies (Fig. 3) in the core section consist of 91 small-diameter, stainless steel clad, enriched elements. Depleted uranium forms the upper and lower breeding blanket sections, each 20 in. in length.

There are 636 subassembly positions in the reactor grid. The grid is subdivided into three radial zones: the central core, the inner blanket and the outer blanket. Grid holes of different sizes are used in the three zones. The subassembly lower adapters are sized for each zone so that they are not interchangeable, to avoid gross loading errors.
FUEL HANDLING

Fuel handling operations in the primary tank are centrally controlled from the reactor building fuel handling console (Fig. 4). The two general types of fuel handling permissible are: (1) restricted, which permits transfers to or from the submerged storage basket during reactor operation; and (2) unrestricted, which permits transfers to and from the reactor grid while the reactor is shut down. The console operator continually maintains surveillance of all the fuel handling operations which are programmed by punched cards. In addition, he must also initiate sequential steps by manually depressing permissive switches. He likewise controls manual operations, such as transfer arm movement. This provides positive control of each operation for maximum safety.

Prior to any grid transfer, the control rod drives are disengaged from the 12 fuel control rods. The reactor cover is then raised approximately 9 ft to permit fuel transfers. The rotating shield plugs, one rotating eccentrically within the other, permit orientation of the main fuel gripper over any grid location. The main fuel gripper is programmed to move downward, engage the subassembly, and then remove it from the grid. After the subassembly is raised, it is received by the transfer arm at a transfer position. The transfer arm moves the subassembly horizontally to a location above the storage basket. The basket is programmed to rotate to a desired orientation and then raises to a release elevation where the subassembly is left
FIG. 4. Fuel handling console

...in storage. The storage basket provides for sodium cooling of irradiated subassemblies and temporary storage for reprocessed subassemblies.

Subassemblies are removed from the storage basket via the transfer arm to the fuel unloading machine (FUM). This is a shielded coffin which moves on tracks to the primary tank transfer port. The subassembly is removed from the transfer arm by an extendable gripper located internally in the coffin. Forced circulation of argon is maintained to cool the subassembly while in the coffin. The subassembly is transferred to an interbuilding transfer coffin for removal to the FCF.

CRITICALITY HAZARDS CONTROL IN THE REACTOR BUILDING

When new or experimental subassemblies are manually loaded into the FUM, appropriate criticality hazards control procedures which control movement and limit storage of such fuel in the reactor building are in force.

PREPARING FUEL HANDLING INSTRUCTIONS

Most fuel handling equipment for reactor loading changes is automatically programmed by a digital computer from input information provided on
punched cards. The key-punched cards are prepared by an operator and
cross-checked by the computer. This is accomplished according to written
procedures under the supervision of the loading coordinator.

The preparation of the punched cards requires exact duplication of the
co-ordinate digital information to ensure proper orientation of the rotating
shield plugs. A set of "master" cards is used to machine-duplicate accurate
co-ordinate information on input cards. The subassembly identity, the date,
the sequential operation, the direction of transfer, and the assigned serial
numbers are also punched on the cards.

Cards of four different colours are used to simplify administrative con-
trol during fuel handling operations. A blue card is used for transfers from
the FUM to the storage basket. A red card is used for transfers removing
subassemblies from the grid. A green card is used for transfers to the
grid. A yellow card is used for transfers from the storage basket to the
FUM.

Upon completion of fuel handling operations, the digital computer pro-
duces a white output card, verifying the completion of the fuel handling trans-
fer and co-ordinate positions.

The prepared input cards are compared by computer checks, for pos-
sible errors, with a group of cards which include exact information on the
loading of the reactor grid and/or storage basket.

The input cards are further checked by two cognizant engineers. Written
loading instructions and loading charts are prepared, describing the loading
change in detail. Loading instructions (Fig. 5) are then approved by the
operations manager, physicist, loading co-ordinator, and technical support
group manager. The operations manager then authorizes the shift crews
to make the loading change.

HISTORICAL INFORMATION

The punched cards provide a complete historical record of the locations
and transfers for any subassembly which has been installed in either the
storage basket or reactor grid. These cards, including the white output
cards, are filed in two groups. One group represents all subassemblies
currently installed in the reactor grid or storage basket. The other group
provides a history of transfers for any subassembly and final disposition
of the subassembly.

IRRADIATION EXPOSURE CONTROL

The computer programme includes calculation of irradiation exposure
of each subassembly in megawatt-days. These tabulated data provide control
information for programming removal of fuelled subassemblies before reaching
maximum "burn-up" limit. Useful fuel life is limited by irradiation
damage to the fuel alloy.

A check on the irradiation history of each subassembly is incorporated
in the computer programme, eliminating the possibility of an error which
returns any fully irradiated subassembly to the grid. This is accomplished
by logging a number proportional to burn-up for each subassembly after each incremental irradiation. In this calculation, account is taken of the variation in burn-up as a function of reactor position by assigning a relative fission rate to each of the first six rows of the grid. The irradiation history of a given subassembly is kept as permanent record. If the irradiation of a subassembly is greater than some previously determined maximum, the computer will not continue the programme. The loading co-ordinator must then decide whether it is safe to install a subassembly as planned.

The computer scans the entire loading and calculates the maximum number of megawatt-days of reactor operation permissible before any subassembly attains the maximum permissible burn-up. This number is typed out as the final message.

The significance of monitoring the irradiation exposure of the fuel and blanket material is emphasized by the need to limit irradiation damage.
Accurate exposure data must be maintained on experimental fuels which are being irradiated.

The total exposure of any subassembly in the reactor varies according to position, power level, and exposure time. Moving a subassembly from one position to another, or returning a partially irradiated subassembly to the core, compounds the difficulty of computing the usage of $^{235}$U and $^{238}$U and production of $^{236}$U and $^{239}$Pu. Since both fission and capture in each of these isotopes differs throughout the core, the distribution of reaction rates, both axially and radially, for both $^{238}$U and $^{235}$U has been calculated and measured by various methods.

A second computer programme has been written to calculate the new isotopic weights after any given irradiation in megawatt-days (Fig. 6). This isotopic information is provided to the fuel cycle facility (FCF) upon transfer of spent subassemblies.

<table>
<thead>
<tr>
<th>SA No.</th>
<th>GRID POS.</th>
<th>U-235</th>
<th>U-238</th>
<th>U-236</th>
<th>U-239</th>
<th>FISSION</th>
<th>AV.-% FUEL BU</th>
<th>REACTOR MWD</th>
<th>DATE INSTAL</th>
<th># of TIMES INSTAL</th>
</tr>
</thead>
<tbody>
<tr>
<td>138</td>
<td>1 1 1</td>
<td>2769.3</td>
<td>3011.4</td>
<td>10.6</td>
<td>3.3</td>
<td>338.1</td>
<td>.5324 1631</td>
<td>415 4</td>
<td>1</td>
<td></td>
</tr>
<tr>
<td>138</td>
<td>1 1 1</td>
<td>2804.3</td>
<td>3017.3</td>
<td>4.9</td>
<td>.0</td>
<td>306.4</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>150</td>
<td>2 1 1</td>
<td>2785.0</td>
<td>3022.9</td>
<td>8.1</td>
<td>1.9</td>
<td>323.8</td>
<td>.2946 927</td>
<td>203 5</td>
<td>1</td>
<td></td>
</tr>
<tr>
<td>150</td>
<td>2 1 1</td>
<td>2804.0</td>
<td>3026.1</td>
<td>4.9</td>
<td>.0</td>
<td>306.5</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>128</td>
<td>2 2 1</td>
<td>2806.7</td>
<td>3044.1</td>
<td>9.3</td>
<td>2.6</td>
<td>334.3</td>
<td>.4064 1281</td>
<td>827 4</td>
<td>1</td>
<td></td>
</tr>
<tr>
<td>128</td>
<td>2 2 1</td>
<td>2833.8</td>
<td>3948.6</td>
<td>4.9</td>
<td>.0</td>
<td>309.9</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>152</td>
<td>2 3 1</td>
<td>2797.1</td>
<td>3015.8</td>
<td>6.1</td>
<td>.7</td>
<td>313.0</td>
<td>.1100 346</td>
<td>420 5</td>
<td>1</td>
<td></td>
</tr>
<tr>
<td>152</td>
<td>2 3 1</td>
<td>2804.4</td>
<td>3017.0</td>
<td>4.9</td>
<td>.0</td>
<td>306.7</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>103</td>
<td>2 4 1</td>
<td>2768.0</td>
<td>3008.8</td>
<td>10.5</td>
<td>3.3</td>
<td>336.9</td>
<td>.5162 1631</td>
<td>417 4</td>
<td>1</td>
<td></td>
</tr>
<tr>
<td>103</td>
<td>2 4 1</td>
<td>2802.0</td>
<td>3014.4</td>
<td>4.9</td>
<td>.0</td>
<td>306.4</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>145</td>
<td>2 5 1</td>
<td>2770.4</td>
<td>3011.5</td>
<td>10.5</td>
<td>3.3</td>
<td>337.1</td>
<td>.5162 1631</td>
<td>1031 3</td>
<td>1</td>
<td></td>
</tr>
<tr>
<td>145</td>
<td>2 5 1</td>
<td>2804.4</td>
<td>3017.1</td>
<td>4.9</td>
<td>.0</td>
<td>306.6</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>122</td>
<td>2 6 1</td>
<td>2799.3</td>
<td>3042.9</td>
<td>10.5</td>
<td>3.3</td>
<td>340.9</td>
<td>.5162 1631</td>
<td>1031 3</td>
<td>1</td>
<td></td>
</tr>
<tr>
<td>122</td>
<td>2 6 1</td>
<td>2833.8</td>
<td>3048.6</td>
<td>4.9</td>
<td>.0</td>
<td>309.8</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

FIG. 6. Fuel burn-up data

FUEL CYCLE FACILITY [5]

The FCF provides for pyrometallurgical processing of enriched uranium-fissium alloy (46 wt. % $^{235}$U, 49 wt. % $^{238}$U, 5 wt. % fission products) two weeks after its discharge from the reactor. The cycle includes complete remote fabrication of highly radioactive fuel elements and subassemblies from reprocessed fuel for return to the reactor. The FCF contains two cells (Fig. 7): one, with an atmosphere of air, is used for disassembly, reassembly, and test operations with clad fuel; and the other, with an argon atmosphere, is used for decanning, reprocessing, and fabrication operations with exposed fuel. Altogether, this is a flexible pilot plant in which all equipment can be installed, operated, maintained and repaired remotely.
The basic fuel cycle is as follows: disassembly, melt refining, fuel fabrication, and assembly. The fuel cycle is shown schematically in Fig. 8. The dismantling, decanning, chopping, melt refining [6], and slagging operations [7] involve spent reactor fuel. The end product of the pyrometallurgical refining operation is partially decontaminated, replenished fuel suitable for reactor use. In the fabrication operations, the ingot is cast into pins; the pins are demoulded, sheared, and inspected for acceptable dimension; the accepted pins are placed in sodium-filled cladding jackets, fitted with end closures, and welded; the welds are then leak checked and the elements bonded; the bonded elements are tested for bond integrity and for proper sodium level and are, at this point, accepted for reactor use.

The elements are assembled in groups of 91 for core subassemblies or 61 for control and safety subassemblies. The subassemblies are welded and subjected to tensile and straightness tests. The accepted subassembly is then transferred to the reactor for re-use. Figure 3 shows the fuel pin, the fuel element, and fuel section, and a core-type subassembly.

When a subassembly is transferred from the reactor to the FCF [8], the burn-up data provided by computer calculation is transmitted along with it. This information is first used to calculate the decay period necessary in the reactor storage basket to reduce radiation and self-heating to an acceptable level for handling in the FCF. After the subassembly is transferred from the reactor, fuel burn-up data are programmed in a computer.
to determine the enrichment (replenishment of $^{235}$U) and the required addition of $^{238}$U to dilute the noble fission products (Mo, Ru, Zr, Nb) that have built up, but are not removed by oxidative slagging of the spent fuel.

Following the injection casting operation [9], the castings are transferred to the pin processing station [10].

During this quality control operation an identity is given to each acceptable cast pin in the form of a punched card which includes such information as casting batch number and pin sequence. This identity is maintained all through fabrication, reactor operation, and disassembly. It is
possible to use these data for purposes of criticality control, accountability, process control, and post-irradiation fuel analysis.

At the pin processing station, the length, weight, diameter, and porosity of each element are measured and a volume and density computed. This information, obtained at the readout console of the remotely operated equipment, is punched on data cards.

Although the pins rejected at the processing station become recycle scrap, the data generated by these rejects (and the reason for rejection) are included on punched cards as part of the permanent record of that run. The card, as initially produced, is shown in Fig. 9.

Each data card is placed in an envelope. Information at each succeeding stage (including data on the process as well as on the container and position) is recorded by the operator on the envelope. Carriers for envelopes simulate the configuration of the fuel or element carriers in the cell and are used to transport the data packets between operations. In operations such as welding or bonding [11], the parameters of operation, such as time, temperature, etc., are recorded in a log book and referenced on a punched card.

When an element (clad fuel pin) is placed in a subassembly and that subassembly is completed and accepted, the data from the envelope are key-punched on the data card. The cards (as shown in B of Fig. 9) then represent a complete process history of the fuel elements in that subassembly.

The strength of this system is the ease with which data studies can be carried out. The weakness of the system is the reliance on the operators to ensure that the position identification of each unit is properly maintained. Since the system was inaugurated in early 1964, adequate experience enabled improvement in those areas that appeared sensitive to error. Our present system is capable of generating reliable data by which the refining and fabricating operations can be studied.

During movement of fuel, a verification of the number of pieces can be ascertained at any time by comparing the fuel in the container with the data packet. When a fuel transfer is made, the operator makes a simultaneous data packet transfer; therefore, information for accountability control and for criticality control is available.

Criticality control in the air and argon cells is accomplished by use of administrative control zones. Maximum fissium fuel content of the cells is 150 kg for the air cell and 250 kg for the argon cell. These maxima are further broken down in the operating zones to 25 kg per zone. Administrative control of each zone is the responsibility of a staff engineer, and no transfer in or out of a zone can be made without his permission.

Since the initiation of the data handling system, which was introduced mainly as a means of process control, additional uses have been found for it. These include data on sheared length as a measure of shear-blade wear, and average sodium height in the element as a measure of the quality of component dimensional control.

As experience is gained with the process, variations in present operating parameters are planned (such as casting pressure, casting temperature, and bonding temperature). The data presently being accumulated and the availability of the data handling system will allow for expeditious analysis of the effect on the succeeding processes of these planned variations.
| B | FIG. 9. A. Initial card at pin processor - B. Final form of data card. |
With the existing close control of identity of fuel going into a reactor subassembly, it is possible to make provisions to maintain the records on individual irradiated elements. As subassemblies are returned to the FCF from the reactor, a sample of a group of elements (10 to 20 out of 91) is selected, identified, and remeasured. This post-irradiation measurement, in part, parallels the procedures for the normal fuel cycle process.

The post-irradiation analysis of the fuel includes measurement of the fission gas pressure, the sodium level, and the dimensions of the fuel pin. This information is used to evaluate the condition of the core; i.e. those parameters which limit the useful life of the fuel.

In addition to individual comparison, the average diameter of the fuel and average sodium level of a sample group of elements are compared to the original batch data obtained during manufacture.

SPECIAL MATERIALS ACCOUNTABILITY

The accountability of fissionable materials in vaults and locked repositories is the direct responsibility of the Special Materials Section. It is also responsible for issuing all transfer documents which release fuel to the FCF or reactor supervisors who, in turn, are responsible for the material while in their custody. They likewise document all interbuilding transfers of special materials between the respective plant supervisors. The reactor operations manager maintains fuel accountability records with a punch card system.

The FCF uses a zone inventory control system which is displayed on the operator side of the cell zones. In addition, inter-cell transfers are recorded in log books. The sum of zone totals and the log book totals are compared daily by a representative of the fuel cycle. The total inventory is verified with special materials records.

The Special Materials Section maintains control and accountability of all fissionable materials by computer methods for all Argonne National Laboratory installations.

FUEL RECYCLE STUDIES

Recycling fuel in EBR-II introduces a variety of problems in fuel management. Considerations relating to fuel usage by the reactor include inventory, reactor power level, storage before and after processing, plant factors of both FCF and the reactor, and fuel utilization. These are not all completely compatible with respect to the optimizing of any of the various operating parameters or operating philosophies. The equilibrium relationship between the FCF production rate and the reactor burn-up rate can be expressed by a simple formula:

\[
\text{FCF production rate (SA/WK)} = \frac{(PF)(MW)(0.11)}{(BU)(FU)}
\]
where

\[ \begin{align*}
PF &= \text{plant factor} \\
SA &= \text{subassemblies} \\
WK &= \text{week} \\
MW &= \text{reactor power (MW)} \\
0.11 &= \text{a factor for neutron flux rates and neutron flux distribution} \\
BU &= (\text{in } % \text{ heavy element burn-up}) \text{ maximum burn-up allowed in any subassembly} \\
FU &= \text{fraction of fuel utilized for maximum burn-up in a given subassembly}
\end{align*} \]

The above equation indicates the FCF production rate if operating power plant factor and per-cent burn-up are kept constant (e.g. with the reactor operating at 45 MW to 1.5% burn-up with a plant factor of 0.83 and a FU of 0.91, the FCF must produce an average of 3.0 subassemblies a week). Ideal conditions are complicated by four major considerations:

1. **Fuel utilization.** The reactor core can be divided into nine regions which have approximately the same burn-up within a region. However, the burn-up rate for each of the nine regions is different, requiring nine shutdowns for a complete core change to achieve the maximum burn-up on each group. Furthermore, because of the flux gradient across a fuel subassembly, it might be economical for maximum fuel conservation to rotate the subassembly 180° halfway through its irradiation life for maximum fuel utilization. Such a system can be accomplished only at the cost of the plant factor.

2. **Plant factor.** Scheduled shut-down must be kept to a minimum if a high plant factor is to be maintained. Shut-down, fuel handling, and start-up require from two to three days regardless of the number of fuel transfers made. A small number of shut-downs is incompatible with maximum fuel utilization. Factored into this is also the necessary time for routine plant maintenance.

3. **Inventory.** The total fuel in process or in completed subassemblies in inventory must equal the sum of the following: present core loading, the next incremental loading, the last increment of subassemblies unloaded and cooling, the number of elements in the FCF under processing, some contingency subassemblies, the make-up material for repletion of fuel, and, finally, special experimental subassemblies. It is obvious that minimum inventory is not compatible with a high plant factor, and maximum inventory places high demand on storage space and adversely affects fuel economics.

4. **Storage.** Unless compatibility of fuel reprocessing and reactor burn-up is established, not only does inventory become a problem but storage of used subassemblies becomes a major consideration. The practical limit of storage and/or inventory forces eventual equilibrium between FCF production and the reactor consumption.

In order to assess the interrelation between these considerations, computer programmes have been written which include as variables in their inputs the following:

(a) Choice of various programmes for the number of changes per shut-down and grid position of subassembly.

(b) Reactor power level.

(c) Operating days between shut-downs (variable within the programme).
(d) Number of cooling days before transfer.
(e) Production rate of FCF.
(f) Inventory: fuelled subassemblies plus material in process.
(g) Shut-down time between operating cycles.
(h) Number of operating periods.

The programme has two modes of output. The first assumes the reactor operates on definite plant factor schedule, and gives an account of subassembly disposition, down time, and running time on a day-to-day or cycle-to-cycle basis. Any lack of fuel is indicated by a negative inventory at that time (Fig. 10). The second mode of output operates realistically and increases the reactor shut-down time whenever necessary for the fuel production to equal consumption. While these programmes are only as good as the input data assumptions, they are useful in planning reactor operation and FCF production.

ASSUMED CONDITIONS

<table>
<thead>
<tr>
<th>Condition</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Burn Up Not To Exceed</td>
<td>1.5% @ 45 MW</td>
</tr>
<tr>
<td>Recovery</td>
<td>1.00</td>
</tr>
<tr>
<td>Prior Run</td>
<td>0 Days</td>
</tr>
<tr>
<td>Cooling</td>
<td>10 Days</td>
</tr>
<tr>
<td>FCF Production Rate</td>
<td>3.0 SA/WK</td>
</tr>
<tr>
<td>Subassembly Inventory</td>
<td>35 Not In Reactor</td>
</tr>
</tbody>
</table>

COMPUTER TABULATION

<table>
<thead>
<tr>
<th>Plant Shutdown</th>
<th>Subassemblies Replaced</th>
<th>Subassembly Inventory</th>
<th>Subassemblies in Process &amp; Cooling</th>
<th>Days Shut Down</th>
<th>Days Total Operation</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>7</td>
<td>28</td>
<td>7</td>
<td>3</td>
<td>117</td>
</tr>
<tr>
<td>2</td>
<td>6</td>
<td>22</td>
<td>13</td>
<td>3</td>
<td>124</td>
</tr>
<tr>
<td>3</td>
<td>6</td>
<td>19</td>
<td>16</td>
<td>3</td>
<td>131</td>
</tr>
<tr>
<td>4</td>
<td>12</td>
<td>12</td>
<td>23</td>
<td>3</td>
<td>143</td>
</tr>
<tr>
<td>5</td>
<td>6</td>
<td>11</td>
<td>24</td>
<td>3</td>
<td>154</td>
</tr>
<tr>
<td>6</td>
<td>6</td>
<td>10</td>
<td>23</td>
<td>3</td>
<td>157</td>
</tr>
<tr>
<td>7</td>
<td>12</td>
<td>1</td>
<td>32</td>
<td>3</td>
<td>174</td>
</tr>
<tr>
<td>8</td>
<td>6</td>
<td>4</td>
<td>27</td>
<td>3</td>
<td>196</td>
</tr>
<tr>
<td>9</td>
<td>12</td>
<td>-3</td>
<td>34</td>
<td>3</td>
<td>205</td>
</tr>
<tr>
<td>10</td>
<td>7</td>
<td>12</td>
<td>19</td>
<td>3</td>
<td>258</td>
</tr>
<tr>
<td>11</td>
<td>6</td>
<td>11</td>
<td>20</td>
<td>3</td>
<td>270</td>
</tr>
<tr>
<td>12</td>
<td>6</td>
<td>10</td>
<td>21</td>
<td>3</td>
<td>281</td>
</tr>
</tbody>
</table>

FIG. 10. Typical fuel recycle programme computer study

SUMMARY

The punched card equipment for reactor fuel handling was part of the original facility design. The use of cards as control for inventory, calculat-
ing burn-up of each subassembly, historical records, and simplified administrative control has developed with experience. In all cases, the methods developed have simplified operations, saved time, reduced manpower, eliminated possible errors, and have resulted in a more reliable and safe system.

At the FCF, our experience indicates that the data handling system is an efficient tool for accountability and criticality control, a valuable tool for process analysis, and a technical tool for the individual and statistical analysis of the effect of the reactor environment on the fuel.

We conclude that, with the larger number of variables involved in the operation of the EBR-II complex, the data card-computer approach is a valid avenue for adequate control and analysis.

ACKNOWLEDGEMENTS

The authors are grateful for the valuable contributions from Mr. F. S. Kirn for developing computer programmes for fuel recycle studies. Also, the contributions of R. M. Fryer and V. G. Eschen, of the Fuel Cycle Facility staff, and Mr. L. H. Berkes, of the Special Materials Section, are appreciated. Special thanks are extended to F. W. Thalgott for many suggestions during preparation and editing of this paper.

REFERENCES

SPECIAL NUCLEAR MATERIAL CONTROL
BY THE POWER REACTOR OPERATOR

R. A. CORDIN
YANKEE ATOMIC ELECTRIC COMPANY, BOSTON, MASS.,
UNITED STATES OF AMERICA

Abstract — Résumé — Аннотация — Resumen

SPECIAL NUCLEAR MATERIAL CONTROL BY THE POWER REACTOR OPERATOR. A relatively new and extremely valuable fuel for electric power production, uranium, requires very careful inventory control from the time the reactor operator assumes financial responsibility for this material until, as partially expended fuel, it is transferred to another facility and the remaining part of its initial value is recovered.

Most power reactor operators were operating fossil-fuelled power plants before the advent of nuclear power and have long since established rather complete and adequate controls for these fossil fuels. The reactor operator must have no less adequate controls for the special nuclear material used in his nuclear plant.

Power reactor operation is not an ancient science and during its relatively short history our engineers and scientists have been constantly improving plant designs and methods of operation to reduce costs and make our nuclear plants competitive with fossil-fuelled conventional plants. Nuclear material management must be as modern and efficient as is humanly possible to ensure that technological advances leading to reduced costs are not lost by poor handling of nuclear fuel and the records pertaining to fuel inventory.

Nuclear material management requires the maintaining of complete and informative records by the power reactor operator. These records need not be complex to satisfy the criteria of completeness and adequacy. In fact, simplicity is extremely desirable.

Despite the fact that nuclear fuel is new and completely different to our conventional fuels no mystery should be attached thereto.

Nuclear material control as part of nuclear material management is not limited to simple inventory work but it is the basis for a great deal of other activity that is an inherent part of any power reactor operations such as irradiated fuel shipments, reprocessing of spent fuel, with its associated accounting for reclaimed fuel and material produced during reactor operation, and the establishing and maintaining of an adequate insurance programme.

CONTROLE DES MATIERES NUCLEAIRES SPECIALES PAR L'EXPLOITANT D'UNE CENTRALE NUCLEAIRE.
L'uranium, combustible relativement nouveau et extrêmement intéressant pour la production d'électricité, doit faire l'objet d'un contrôle très strict depuis le moment où l'exploitant de la centrale en devient financièrement responsable jusqu'à celui où, sous forme de combustible partiellement épuisé, la matière est transférée dans une autre installation et où l'on récupère ce qui reste de sa valeur initiale.

La plupart des exploitants de centrales nucléaires dirigeaient des centrales alimentées par des combustibles fossiles avant l'arrivée de l'énergie nucléaire et ils ont établi depuis longtemps un contrôle étendu et efficace de ces combustibles fossiles. L'exploitant d'une centrale nucléaire doit exercer un contrôle non moins efficace sur les matières nucléaires spéciales utilisées dans son installation.

La technique de l'exploitation des centrales nucléaires n'est pas ancienne et, au cours de son existence relativement brève, les ingénieurs et hommes de science des États-Unis ont constamment amélioré les plans des centrales et les méthodes d'exploitation afin de réduire les coûts et de permettre aux centrales nucléaires de concurrencer les centrales classiques. La gestion des matières nucléaires doit être aussi moderne et efficace que possible pour assurer que les progrès technologiques grâce auxquels les prix on pu être réduits ne soient pas mis en échec par des insuffisances dans la manipulation du combustible nucléaire et la tenue de la comptabilité des stocks.

Pour assurer la gestion des matières nucléaires, il faut que l'exploitant de la centrale établie et tienne à jour une comptabilité complète et détaillée, sans être nécessairement complexe pour autant, en fait, la simplicité est extrêmement souhaitable.

Bien que le combustible nucléaire soit nouveau et qu'il n'ait rien de commun avec les combustibles classiques, aucun secret ne doit s'y attacher.
Le contrôle des matières nucléaires dans le cadre de la gestion des matières nucléaires ne se limite pas à de simples travaux d'inventaire mais sert de base à beaucoup d'autres activités qui font partie intégrante du programme d'opérations de tout réacteur, par exemple les expéditions de combustible irradié, le traitement chimique du combustible épuisé et la comptabilité du combustible récupéré et des matières produites au cours du fonctionnement du réacteur, et l'institution et l'application d'un régime d'assurance satisfaisant.

**Специальный контроль наличия ядерных материалов оператором энергетического реактора.** Относительно новое и чрезвычайно ценное топливо для производства электроэнергии, а именно уран, требует очень тщательного учета с момента, когда оператор реактора принимает на себя материальную ответственность за этот материал, и до того момента, когда этот материал в виде частично отработанного топлива передается на другую установку, где регенерируется оставшаяся часть его первоначальной ценности.

Большая часть операторов энергетических реакторов, до появления в свете ядерной энергетики, работала на электростанциях с обычным ископаемым топливом и уже давно выработала достаточно полно и надежные формы контроля за его расходованием. Оператор реактора должен располагать не менее надежными способами учета ядерных материалов, используемых на его установке.

Эксплуатация энергетических реакторов не имеет большой истории, и на протяжении относительно короткого периода существования реакторов наши инженеры и ученые непрерывно совершенствовали конструкцию установок и улучшали методы эксплуатации с целью снизить себестоимость с тем, чтобы наши ядерные установки могли быть конкурентоспособными с работающими на ископаемом топливе электростанциями обычного типа. Административно-хозяйственный учет ядерных материалов должен быть по мере возможности современным и эффективным с тем, чтобы технологический процесс, направляемый на снижение себестоимости, не страдал бы из-за расточения ядерного топлива и небрежного его учета.

Административно-хозяйственный учет требует со стороны оператора энергетического реактора ведения полных и подробных учетных документов. Для удовлетворения требованиям полноты и надежности эти документы вовсе не должны быть сложными. Наоборот, простота их является чрезвычайно желательной.

Несмотря на то, что ядерное топливо является чем-то новым и совершенно отличным от топлива обычного типа, нельзя облекать его покровом таинственности.

Контроль наличия ядерных материалов как часть административно-хозяйственного учета ядерных материалов не ограничивается одной лишь работой по проверке наличия материалов, но он лежит в основе многих других видов деятельности, являющихся неотъемлемой частью эксплуатации любого энергетического реактора, как например, отправок облученного топлива, переобработки отработанного топлива со связанной с этим отчетностью при регенерации топлива и материала, произведенного при работе реактора. К этому кругу проблем относятся также выработка и осуществление надлежащей программы страхования.

**Control de materiales nucleares especiales por parte de quienes operan el reactor de potencia.** Combustible relativamente nuevo y sumamente valioso para la producción de energía eléctrica, el uranio requiere un control muy minucioso desde el momento en que la dirección de una central asume la administración y responsabilidad inherente a su posesión hasta que como combustible parcialmente agotado se transfiere a otra instalación en la que se recupera la parte que no se ha consumido.

Antes de que se descubriera la posibilidad de emplear la energía nuclear para producir electricidad, la mayor parte de las empresas que actualmente explotan centrales nucleares explotaban centrales alimentadas con combustibles fósiles y habían establecido sistemas de control relativamente completos y adecuados para los combustibles de ese tipo. Los responsables de las centrales nucleoelectricas deben disponer de sistemas no menos adecuados para controlar los materiales nucleares especiales que utilizan.

La explotación de los reactores de potencia no es una ciencia antigua, pero durante el tiempo relativamente corto que ha transcursado desde que se inició su empleo los ingenieros y hombres de ciencia han mejorado continuamente el diseño del equipo y los métodos de trabajo con objeto de disminuir los costos de producción y de lograr que las centrales nucleares puedan competir en el plano económico con las centrales clásicas. La administración de los materiales nucleares debe efectuarse con métodos modernos y eficientes a fin de que los adelantos tecnológicos que han permitido reducir los costos no resulten innecesarios debido a un control deficiente de los combustibles nucleares y a la falta de precisión en las operaciones contables.

La administración de materiales nucleares requiere que los explotadores de los reactores de potencia lleven una contabilidad completa y detallada. No es necesario que los documentos sean complejos si lo que se pretende es que sean completos y adecuados. En realidad, la sencillez es sobremaneira conveniente.
A pesar de que el combustible nuclear es un combustible nuevo y totalmente distinto de los de tipo tradicional, no hay que rodear su utilización de ninguna aureola de misterio.

El control de los materiales nucleares no es una simple labor de inventario, sino que constituye la base de muchas otras operaciones inherentes a la explotación de los reactores de potencia, por ejemplo: el transporte del combustible irradiado, su regeneración con la correspondiente contabilización del combustible recuperado y del material producido durante el funcionamiento del reactor, y, por último, el establecimiento de un sistema de seguros adecuado.

In establishing a programme of special nuclear material control the power reactor operator is taking a further step into an area that was unknown 20 years ago. At that time, in 1945, the nuclear technology was known to a rather small group of engineers and scientists and supplies of special nuclear material were also rather small. The question of peacetime economics was not pertinent during that period. Today, to the power reactor operator, such is not the case. He is using in his facility this special nuclear material, uranium, as a fuel, as a source of heat, for the production of electricity. This relatively new and extremely valuable fuel requires his careful control. Without such control, economic and intelligent operation of his plant would be difficult, if not impossible. Nuclear material control, or, if you will, nuclear material management, has become a vital necessity.

Since power reactor operation is relatively new and scientists and engineers are constantly improving plant designs and methods of operation to reduce costs, nuclear material management must be as modern and as efficient as is humanly possible to ensure that technological advances are not offset by poor handling of nuclear fuel and the records pertaining thereto.

Fuel control is not new to power reactor operators, for most of them were operating fossil fuelled power plants before the advent of nuclear power. The fossil fuels used – coal, oil or gas – were and are accurately metered and weighed as received and burned. Each month the power plant operator knows how much fossil fuel he has received, how much he has burned, and how much he has stockpiled. He is invoiced for his fuel purchased and his dollar costs are known. At each step in his fuel handling he knows these fuel costs for he has been able to set up the necessary controls to provide this information. It is this same type of control that the reactor operator should seek for his nuclear plant. At present, at least in most cases, he does not prepare the fuel he uses but, nevertheless, has the financial responsibility for every gram of special nuclear material that he has in his own facility or in the hands of his fuel fabricators or fuel reprocessors. In the United States of America this is the responsibility of the United States Atomic Energy Commission. Final and accurate fuel costs cannot be absolutely determined until spent fuel reprocessing is complete. Power reactor operators must estimate these fuel costs and while we feel these estimates are very close to actual costs, they are still estimates. Starting information, in so far as nuclear material in reactor cores is concerned, is therefore of the utmost importance to the reactor operator. This information should be accurate and timely.

In our approach for the same controls for nuclear fuels that we now have for fossil fuels, we might easily fall into a trap of oversimplification. We are dealing with a fuel cycle wherein the reactor is loaded with a quantity
of fuel based on information furnished by the processors. Estimated burn-ups are subtracted from this initial quantity during core life resulting in a final quantity removed from the reactor. With an accurate starting point our chances of having an accurate final answer are greatly enhanced.

Consider a facility whose burn-up in a given reactor core was in the order of 277 kg of the element. For the same power production, over 500,000 tons of coal would have been needed in a fossil fuel plant of the same efficiency. Surely it is easier to keep track of 277 kg, or 611 lb of uranium than it is to keep track of over one thousand million pounds of coal. Keeping these numbers in mind, the trap of oversimplification becomes evident. We can start nicely into this trap on the subject of losses. To be sure, certain losses are inevitable in the processing of material, but someone will have to pay for every gram lost. Contractual arrangements between processors and the reactor operator will determine who will make the payments — but someone will, and that someone is the reactor operator. If he does not pay them directly because his contract with the core fabricator calls for the fabricator to assume the responsibility for all losses, it is certain that the price the reactor operator pays for his core will contain the cost of the lost material. We must be sure that such losses as do occur are logical and factual and are not the result of any lack of care on our part. A lost kilogramme of uranium will have to be paid for, but it will not produce a single BTU in a reactor.

Very close to the loss situation is the problem of shipper-receiver differences. As has been pointed out many times this problem is one of long standing. In our industry this is, or can be, a very serious problem but is one which is almost non-existent for the operator of a fossil fuel plant. In the United States of America the Institute of Nuclear Material Management has stressed, since its very inception, the vital need for careful record-keeping and accurate measurements of all special nuclear material. They have repeatedly and strongly urged all persons charged with the responsibility for special nuclear material to use the utmost care in record-keeping, and pointed up the necessity for self-policing of our jobs in our industry.

Actually, when we speak of nuclear material management, management in the sense in which we use it connotes stewardship. In all our operations we have the stewardship of someone else's property and we should keep this trust inviolate. In this connection, no remarks are necessary in regard to integrity as this is tacitly understood, but our reporting should be accurate and on time and I believe that if we do use the inordinate amount of care that is a must in our material management work, we will see the problem of shipper-receiver differences become smaller and smaller with the passing of time. If we consider only the loss and the shipper-receiver difference problems, and these two are certainly not our only problems, I think we can safely say that we dare not try to oversimplify our controls for nuclear material management.

We should, of course, realize that a "factor of merit" is part of our record-keeping. As we get away from oversimplification, we are very likely to change into the directly opposite area and overcontrol our situation. Are we then overcontrolling to the extent that these very controls are costing us more than we save by their use? If this happens we have lost our "factor
of merit". Somewhere there lies a middle ground and here we must find our answer.

We are concerned with finding controls for a fuel which could easily be termed exotic, a fuel which we might even want to call romantic. This is quite all right — we are in an age where nuclear operations and space age projects are all blessed with a little romance and it is not going to hurt us very much if we keep this feeling, for I don't doubt that hundreds of years ago the people who first burned coal under boilers had this same feeling. However, let us not confuse any of the issues of nuclear material management or use by trying to make a mystery out of any of our operations. Again, firm and accurate records are needed, not mysticism generated because we are using a fuel that is relatively new. Let us not try to oversimplify, but let us be as factual as we possibly can in every step that we take in the handling of these materials.

Let us go back to our controls for a moment. In most cases in the United States of America when lease material is withdrawn from the USAEC for use in a reactor, the reactor operator immediately assumes financial responsibility. However, some time elapses between the withdrawal of this material from the USAEC and the installation of this material in the operator's reactor. During this time the material is moving through the hands of various processors. The reactor operator has a liability to the Commission of millions of dollars, so it is quite obvious that he wants to know where all these dollars are at all times. With the advent of private ownership of special nuclear material the power reactor operator will have to finance an inventory of material the value of which may approach the value of his plant.

We are not, as reactor operators, interested in only the dollars as dollars that we have scattered around in the hands of the processors or in our own facilities, although our interest in these dollars is and should be more than casual. We want to know where these dollars are, how long they have been there, and when they move. These material dollars are not static dollars. At each step along the processing line the dollar value per kilogram increases by the amount of work performed in the facility through which the materials are passing. Also, each day that goes by sees an increase in the use charges accrued. This information ties in with our needs for the economic control of our business but, as we said earlier, we are looking for these nuclear material controls for both the economic control and the intelligent operation of our own facility.

We must have proper, accurate information on which to base our burn-up calculations and our by-product material production. It is also necessary for us to have as much information as possible as promptly and as accurately as we can get it, for the purpose of establishing our insurance programme. Any reactor operator who has a spent core in storage, an operating core in the reactor, and a new core in his fuel vaults must, of necessity, have rather an elaborate and expensive insurance programme. He is protecting material which is not his but is, in the United States of America, the property of the USAEC. The reasons for this insurance programme are obvious; if the reactor operator does not have that fuel to return to the Commission, he has to pay for it. Of course, we do not anticipate that anybody
is going to walk into a power reactor operator's facility and walk out with a fuel assembly under his arm. However, common prudence demands that we protect our owners and for that reason we must insure this material. This is, of course, you understand, property insurance and not liability.

All the foregoing has concerned the handling of new material in the preparation of the reactor cores, but it also applies just as well to the reprocessing of spent fuel. The reactor operator's liability to the USAEC does not end until the material has been returned to this agency, and we are just as interested in the spent fuel discharged from our reactor and the material produced in our reactor as we are in the new fuel while it is being fabricated.

A power reactor operator's records should show the location of all the special nuclear material for which he has the responsibility, regardless of its physical location. To achieve the desired control, the following outline, I believe, represents the minimum requirements for the accountability records of a power reactor operator.

1. Ledger or sub-section of ledger for each converting, processing, or fabrication facility that handles the reactor operator's fuel. To provide continuous inventory of fuel movement until it arrives at the reactor operator's plant.

2. Ledger or sub-section of ledger for cold scrap materials returned to the Atomic Energy Commission to provide control for checking of credits by the Atomic Energy Commission to the reactor operator's lease account.

3. Ledger or sub-section of ledger for new fuel vault. To provide inventory control of new fuel received from fabricator until charged to reactor.

4. Ledger or sub-section of ledger for each core loaded to reactor. To provide continuing record of core as loaded, material burn-up, material production, and final composition of core.

5. Ledger or sub-section of ledger for spent fuel storage pit. To provide material inventory control of discharges from reactor.

6. Ledger or sub-section of ledger for shipments of irradiated fuel assemblies. To provide inventory control of spent fuel moving out of the reactor operator's facility.

7. Ledger or sub-section of ledger for total AEC lease account covering withdrawals, cold scrap returns, burn-up and payment therefore, and return of material which has seen irradiation. To provide control of total material responsibility under lease account and establish a dollar basis for use-charge accruals.

8. Ledger or sub-section of ledger for plutonium. To provide a record of plutonium production by cores and its disposition.

It is, of course, realized that at least every six months, when it becomes necessary to prepare a material status or inventory report for the Atomic Energy Commission, the reactor operator knows where his material is and when he receives transfer documents from his processors he knows that material has moved or is moving from one facility to another. Before the filing of a material status or inventory report there is, by use of the material activity schedule, an opportunity for the various processors of the operator's material to reach an agreement on balances. But such balance agreements executed only every six months do not really provide a com-
pletely satisfactory control of the multi-million dollar liability for which the reactor operator is responsible.

If we were conducting a small business in almost any field you might wish to choose and during the course of a year handled materials of any nature with the total valuation of the nuclear material going into a single power reactor core, you may be sure that we would produce monthly a balance sheet and a profit and loss statement showing just where we were at the end of each month. Probably the most important single factor in those statements would be the inventory report. If we were working with our own material and had such inventory valuations, we would use great care in our records, both in building them up and reproducing them for our operating statements. If we were handling someone else's materials, of any type or description, we would certainly be keeping very accurate records and on a monthly basis. I do not believe that we should do any less in the nuclear industry. Recognizing that these inventories are vital and must be correct, we therefore have to arrive at a point of agreement as to the timing for our inventory balance reports. How, then, shall we determine this point of agreement? Once again the trap of oversimplification faces us. We can immediately jump to a seemingly logical comparison and say that there are hundreds of banks which have more dollars in their vaults than many processors or reactor operators have grams of nuclear material on hand.

The banks balance their cash daily - why can't we do the same? The only trouble with this statement is that the comparison is not very valid. The banks do balance their cash everyday; however, for every dollar the banks have - they have one hundred cents. Now we have lost our comparison between nuclear facilities and banks.

Quite basically some facilities handling special nuclear material, especially processors and converters who may have contracts for fuel production for several reactor cores, do not have a certain number of grams of material, each one enriched to the same percentage. To ask them to balance their inventory of varying enrichments would be like asking the banks to balance all their dollars where some of these dollars contain twenty pennies, some dollars contain three pennies and some contain ninety pennies, an impossible situation as far as daily balances are concerned.

Inventory balances that are worth the paper they are written on are not easily obtained. If, for instance, the operator of a processing facility is going to give an accurate balance report to a reactor operator, he must inventory his entire holdings of material, not just that which he is holding for a single reactor operator.

The situation does not improve very much if we try to put it on a weekly basis, but would we not be approaching a pretty firm foundation if we established monthly balances? A very simple form could be used for this purpose. At least this type of material reporting would reduce our period of uncertainty to one-sixth of the present situation.

The practice of establishing monthly balances would, it seems, add greatly to the control that the reactor operator is seeking for his nuclear material. We must consider the fact that fuel processors are handling these special nuclear materials, and as the materials move between facilities transfer documents are prepared and distributed to the interested parties who record these transfers, but unless a shipment of material goes directly
to the reactor operator, he very possibly does not receive a copy of these transfer documents. It is true that information copies of transfer documents could be furnished to the reactor operator, even though he does not receive any of the fuel, but since it is not mandatory that information copies be furnished to anyone, this is a point where the system easily breaks down. When this happens the reactor operator might be completely unaware of material transfers between facilities and, of course, it is his money that is being transferred. I am sure that if we were transferring money instead of uranium everyone concerned would know about the transfer. A monthly balance report would obviate the possibility of the reactor operator being left without the complete knowledge of the whereabouts of his material. When the reactor operator is without this information, he has not only lost the control of the actual material movement, but he has lost also the control of his application costs represented by use charges.

Granted that the filing of monthly balance reports would add somewhat to the paper work the processors or fabricators now have, it would seem that such a procedure would definitely have the "factor of merit" we have mentioned before and for which we look in the application of cost, i.e. in this case the information would be of greater value than the cost of producing it. I believe that this particular factor is applicable to the processors and fabricators who would have to prepare the reports, as well as the reactor operator who very definitely benefits from such a control. When we consider the approaching date in the United States of America when special nuclear material passes into private ownership, then very definitely such a system of balancing would be sought, or, in fact, even demanded by the reactor operator. I believe the fact that we presently do not own the material we are using for fuel is not a very valid reason for not having such monthly balance reports at the present time.

As we are presently set up, is this monthly balance report a problem? It would seem not since we all operate or should operate accurate accountability ledgers, some of us with automated equipment and some with hand-posted books. I do not believe any of us are trying to operate without formal books of any kind. If ledger postings are timely and accurate, book balances should be available almost immediately at the close of business on the last day of the month. Perhaps we have no problem here except that of communications. By that I mean that in all probability we already have these monthly balances in report form, but we just have not bothered to tell anyone about them.

As we progress along this problem of accounting for materials, we must also be prepared to at least look into the matter of changes in enrichments as our material moves along. Here again is a matter of control. As material is withdrawn from the United States Atomic Energy Commission Production Department we have very well defined enrichments, and when the reactor operator asks for a predetermined and definite enrichment he gets material that is extremely close to his request. However, if a large amount is ordered out so that a core for a large power reactor may be fabricated, the enrichments coming from the Production Department will vary ever so slightly above and below the requested enrichment. How shall we handle transfers of this material as it passes through our processing and fabrication lines?
With a large amount of material involved, it is quite obvious that the first material withdrawn will probably reach the end of the processing and fabrication line before the last of the material is actually withdrawn. It is, therefore, impossible to apply a total average withdrawal enrichment to the first material moving through the various facilities. As this material moves along the line there should be no change in this enrichment. However, as we get down to smaller balances in various facilities, we reach the point where due to transfers of material at an assumed average for a certain shipment and the necessary roundings that take place between element and isotope, we are finally faced with a remaining balance in a facility at an enrichment which, at first glance, seems ridiculously high or low compared to the original withdrawal.

If we wait until all of the material has been withdrawn from the production facilities and then balance our figures with those submitted to us by the USAEC, we are in a position to establish the total average withdrawal enrichment. In most cases the attempt to make any adjustment during the fabrication of this material would be fraught with a great deal of danger. Here then is an area where our control is not lost, but has become tenuous until such time as the necessary adjustments can be made. Perhaps this is an area in which we can live without tight control as long as our records are timely and accurate and the necessary adjustment is made as soon as possible.

Quite obviously, if your reactor is designed to use 5% material and as you get to the end of a processing line you find that you have some material left over that your inventory shows as 10% material, you have something that you could not use in your reactor. It is also obvious that you could not pick up this enrichment unless somewhere along the line some blending took place, which I think you will agree is not in the realm of probability if we continue to operate with discipline. But, and this is most important, we must operate with complete and unending discipline.

So far we have been covering the aspects of control in regard to accountability of weights and we have lightly touched on the subject of varying enrichments which result from the compounding of rounding errors. If we are now at a point where you are asking yourselves, "Why are the reactor operators reaching so strongly for control?" Perhaps we can digress a moment and consider a possible situation with which the reactor operator might be faced.

If we did not have some kind of nuclear material control it is possible that when our reactor was put into operation after a fuelling or refuelling, we conceivably could have indications that all was not well within our nuclear system, resulting in a shut-down in the plant until a reason for the departure from normal operations was discovered. We would like to say that the fuel had nothing to do with it, and I think there would be pretty general agreement in a nuclear material management group that this statement could be made with complete safety since we do take a great deal of care to ensure proper handling in our facilities. When I say we, I mean reactor operators and processors.

But suppose the power reactor operator was faced with a shut-down of his plant and had to make a determination that the fuel was not at fault, just what does this mean to the reactor operator? Let us consider a large
plant; a plant that has an electrical output of 500 MW. This is the size plant
that is becoming rather common at present. A 500-MW plant would have
a saleable output of approximately 11 200 000 kWh in a 24-h day; if this power
is priced at 6.5 mills per kWh, which under present assumptions is rather
realistic, then every day that a turbo generator is out of service at the re-
actor operator's plant represents a loss of almost $73 000 income to the
reactor operator. This loss of $73 000 income per day would continue until
the reactor operator had determined that his fuel was as specified and that
he could with safety start his plant. This supposition may be one that would
never happen, but it is presented here as a possibility and a reason for de-
manding the utmost care in every phase of nuclear material management.

Not too long ago a workshop type of meeting was held by the Institute
of Nuclear Materials Management on shipper-receiver differences. In the
report issued following this meeting a possibility was postulated wherein
a shipment of enriched uranium from the production cascades was followed
to a processor, then to the fabricator, then to the reactor manufacturer who
finally prepared the fuel element, and the assumption was that each man in
the chain accepted the Production Department data and no shipper-receiver
differences occurred. If the spent core, after removal from the reactor
and shipment to the reprocessing plant, is found to have a discrepancy, such
a discrepancy (the workshop pointed out) would be obscured by so many oper-
ations that we would probably never know where the discrepancy occurred.
The workshop went on to state that this was not impossible but something
that could happen, and had, as a matter of fact, actually happened.

Since this unfortunate situation is known to have happened once, in all
probability it has happened several times. I cite this as one example
of operational problems that arise during a fuel preparation cycle. It is a
problem that must be solved to prevent recurrence. It is one of the reasons
the power reactor operator is seeking special nuclear material inventory
controls.

I can recommend no quick, easy and dull method of obtaining the con-
trols the reactor operator seeks. I believe the previously mentioned
monthly balance report is a long step in the right direction. A standardized
accountability ledger would, I believe, be another step forward. I have no
doubt but that most nuclear material managers use the same general type
of ledger but I also have no doubt that the accountable officer for Company X
feels that his ledger needs are perhaps slightly different from the needs of
the accountable officer in Company Y, and so he has devised a slightly dif-
f erent ledger or ledger page for his own use. If we establish an audit pro-
cedure for special nuclear material inventories a standardized ledger would
be invaluable.

The possibility of an audit procedure is intriguing. For many years
public accountants have audited the financial books of corporations and sub-
sequent to the audit have certified their findings to the corporation. Recently,
in the United States of America, the United States Atomic Energy Commission
has established an audit procedure for holders of licensed material. To the
best of my knowledge this has been very satisfactory and has provided the
Commission with the needed proof of inventories held by licensees. What
will become of the Commission' material audits after private ownership.
becomes mandatory, I do not know, but I believe some sort of material audit procedure is a most desirable part of nuclear material management.

Who would perform these material audits if the present governmental agencies discontinue this phase of their work, I am unable to say. There are at least two possibilities; an organization such as the Institute for Nuclear Material Management might be willing to set up an audit group available to members of the nuclear industry, or perhaps the public accounting firms would be willing to augment their staffs with men knowledgeable in nuclear fuel accountability work.

The special nuclear materials control that the power reactor operator needs should not be too difficult to set up and operate. Simplicity is a prime requisite and completeness and accuracy of records is a vital necessity. The attainment of these two goals should be the aim of every person engaged in nuclear materials management and no power reactor operator should try to operate his facility without exercising management control over the expensive fuel his plant requires.

**DISCUSSION**

D. JANISCH (Chairman): You spoke about the insurance of fissile material. I would like to ask two questions in this connection. Is this insurance operated by commercial insurance firms, and what rate is charged, i.e. as a percentage of the value insured?

R. A. CORDIN: Property insurance on nuclear facilities including fuel is available from "Nuclear Energy Property Insurance Associates" - a stock company pool - and a smaller mutual insurers' pool. The costs are approximately 5 cents per US $100.
PROFESSIONAL NUCLEAR MATERIALS MANAGEMENT

A. A. FORCELLA AND W. J. O'LEARY
ALLIS-CHALMERS MANUFACTURING COMPANY,
BETHESDA, MD., UNITED STATES OF AMERICA

Abstract — Résumé — Аннотация — Resumen

PROFESSIONAL NUCLEAR MATERIALS MANAGEMENT. This paper describes the scope of nuclear materials management for a typical power reactor in the United States of America. Since this power reactor is financed by private capital, one of the principal obligations of the reactor operator is to ensure that the investment is protected and will furnish an adequate financial return.

Because of the high intrinsic value of nuclear materials, appropriate security and accountability must be continually exercised to minimize losses beyond security and accountability for the nuclear materials. Intelligent forethought and planning must be employed to ensure that additional capital is not lost as avoidable additional costs or loss of revenue in a number of areas. The nuclear materials manager must therefore provide in advance against the following contingencies and maintain constant control or liaison against deviations from planning during (a) pre-reactor acquisition of fuel and fuel elements, (b) in-reactor utilization of the fuel elements, and (c) post-reactor recovery of fuel values.

During pre-reactor planning and operations, it is important that the fuel element be designed for economy in manufacture, handling, shipping, and replaceability. The time schedule for manufacturing operations must minimize losses of revenue from unproductive dead storage of high cost materials.

For in-reactor operations, the maximum achievable burn-up of the fissionable material must be obtained by means of appropriate fuel rearrangement schemes. Concurrently the unproductive down-time of the reactor for fuel rearrangement, inspections, and the like must be minimized.

In the post-reactor period, when the fuel has reached a predetermined depletion of fissionable material, the nuclear materials manager must provide for the most economical reprocessing and recovery of fissionable values and by-products.

Nuclear materials management is consequently an essential factor in achieving competitive fuel cycle and unit energy costs with power reactors.

GESTION INDUSTRIELLE DES MATIERES NUCLEAIRES. Le mémoire expose en quoi consiste la gestion des matières nucléaires dans une centrale nucléaire type, aux États-Unis. Comme le réacteur en question est partiellement financé à l'aide de capitaux privés, l'une des principales obligations de l'exploitant est d'assurer la protection et la rentabilité des investissements.

Étant donné que les matières nucléaires sont d'une valeur intrinsèque élevée, il faut constamment procéder à des contrôles appropriés allant au-delà des mesures de sécurité et de la comptabilité intéressant les matières nucléaires proprement dites afin de réduire les pertes au minimum. Il faut faire preuve de clairvoyance et planifier judicieusement pour prévenir toute perte supplémentaire de capital provenant de frais inutiles ou d'un manque à gagner dans divers secteurs de l'exploitation. C'est ainsi que le gestionnaire de matières nucléaires doit prendre des dispositions pour garantir la bonne marche des opérations et assurer le respect des plans d'exécution par une liaison et un contrôle constants, dans les domaines suivants : a) acquisition du combustible et des éléments combustibles, b) utilisation des éléments combustibles dans le réacteur et c) récupération, dans le combustible irradié, des produits et matières présentant de l'intérêt.

Pendant la période qui précède l'utilisation du réacteur, il faut faire une place importante dans la planification et les travaux préparatoires aux considérations d'économie dans la conception des éléments combustibles, en ce qui concerne leur fabrication, leur manutention, leur transport et leur remplacement. Les différentes étapes de la fabrication doivent être planifiées de façon à réduire au minimum le manque à gagner dû à des périodes improductives d'entreposage de matières très coûteuses.

Pendant la marche du réacteur, il faut assurer une combustion maximale de la matière fissile par des redistributions appropriées du combustible dans le cœur du réacteur. Parallèlement, les temps morts dus à ces redistributions, aux inspections et autres causes semblables doivent être réduits au minimum.
ОБРАЩЕНИЕ С ЯДЕРНЫМИ МАТЕРИАЛАМИ НА ПРОФЕССИОНАЛЬНОМ УРОВНЕ.

Дается описание объема работ на типичном энергетическом реакторе в США. По скольку этот реактор финансируется частным капиталом, одна из основных обязанностей оператора состоит в обеспечении сохранности капиталовложений и получении определенной прибыли.

Ввиду большой себестоимости ядерных материалов необходимо постоянно обеспечивать надлежащую безопасность и учет с целью сведения к минимуму потерь, не связанных с обеспечением безопасности и учета ядерных материалов. Необходимо все тщательно продумывать и планировать заранее, чтобы избегать ненужных затрат и расходования капитала или снижения размеров прибыли в ряде областей. Поэтому администратор, ведающий ядерными материалами, должен заранее учитывать все непредвиденные обстоятельства и осуществлять постоянный контроль над отклонениями от стандартов при планировании во время а) приобретения топлива и топливных элементов до их использования в реакторе, б) при использовании топливных элементов в реакторе, и в) при регенерации топлива после использования в реакторе.

Во время планирования и осуществления операций с топливом до его использования в реакторе важно, чтобы предусматривалась экономия топливных элементов при изготовлении, обработке, транспортировке, а также предусматривалась их заменяемость. График проведения производственных операций должен обеспечивать сведение к минимуму потерь прибыли в результате непродуктивного "мертвого" хранения дорогостоящих материалов.

При проведении операций внутри реактора необходимо добиваться максимально возможной степени выгорания расщепляющегося материала путем осуществления соответствующих схем перераспределения топливных элементов. Одновременно необходимо всячески уменьшать время, непродуктивно затрачиваемое на перегруппировку топливных элементов на остановленном реакторе, на инспекции и т.д.

Во время операций с ядерными материалами после их использования в реакторе, когда степень обогащения топлива достигла уровня предварительно установленного обеднения расщепляющегося материала, администратор в области ядерных материалов должен обеспечить наиболее экономическую переработку и регенерацию расщепляющихся материалов и побочных продуктов.

Обращение с ядерными материалами соответственно представляет собой важный фактор в деле осуществления конкурентоспособного топливного цикла и поддержания соответствующего уровня расходов на единицу энергии на энергетических реакторах.

ADMINISTRACION EFICIENTE DE MATERIALES NUCLEARES. En esta memoria se describen las modalidades de la administración de los materiales nucleares destinados a un reactor de potencia típico de los Estados Unidos. Este reactor está financiado con capital privado y, por tanto, una de las principales obligaciones de la dirección de la central es asegurar que las inversiones estén protegidas y produzcan un interés razonable.

Como los materiales nucleares tienen un valor intrínseco elevado es preciso ejercer sobre ellos una fiscalización constante a fin de evitar que las pérdidas rebasan los límites de seguridad. Es preciso prevenir mediante una planificación inteligente las pérdidas evitables de capital debidas al incremento de los costos o a la reducción del rendimiento en determinadas etapas de las operaciones. El administrador de materiales nucleares debe por consiguiente tomar de antemano las disposiciones necesarias para hacer frente a todas las eventualidades y mantener un control constante para evitar que se produzca alguna desviación de los planes durante a) la adquisición del combustible o de los elementos combustibles, b) el empleo de los elementos combustibles en el reactor y c) la regeneración del combustible una vez utilizado.

Por lo que respecta al primer punto, es muy importante que los elementos combustibles hayan sido diseñados de forma que su fabricación, su manipulación y su transporte resulten económicos y sea fácil sustituirlos. La programación de las operaciones de fabricación deberá reducir al mínimo las pérdidas de ingresos debidas a un almacenamiento improductivo excesivamente largo de materiales de costo elevado.

Para la etapa de su empleo en el reactor, será necesario un programa adecuado de redistribución de los elementos combustibles a fin de lograr el grado máximo de combustión del material fisionable. Además, habrá que reducir en la medida de lo posible los paros improductivos del reactor debidos a la redistribución del combustible, a las inspecciones, etc.
Cuando el combustible haya llegado a un grado predeterminado de agotamiento, el administrador tendrá que disponer la forma más económica de regeneración de los materiales fisionables y de recuperación de los subproductos.

La administración de materiales nucleares es por consiguiente un factor esencial para la reducción de los costos del ciclo del combustible y para lograr que el costo unitario de la energía producida resulte económicamente interesante.

1. INTRODUCTION

This paper describes the scope of nuclear materials management for a typical privately operated power reactor in the United States of America. This paper does not discuss nuclear materials management in connection with reactors operated by or for the United States Atomic Energy Commission.

Nuclear materials management is a relatively new profession. Many people consider this field to be almost synonymous with that of nuclear materials accountability. But with the increasing use of nuclear energy for peaceful purposes, particularly in the field of nuclear power reactors, the industry has been broadening its demands on accountability representatives. Consequently, nuclear materials management today encompasses far more than accountability.

In the broadest terms, nuclear materials management encompasses: (a) accountability for nuclear materials, (b) pre-reactor nuclear materials management, (c) in-reactor nuclear materials management, (d) post-reactor nuclear materials management, and (e) fuel-cycle cost analysis. This sequence is chronological and does not reflect the relative importance of the items. Accountability is a part of the three areas of materials management which form an endless nuclear fuel cycle. Fuel-cycle cost analysis is an independent activity that is based upon the other four areas. These areas will be discussed in the chronological order given.

2. ACCOUNTABILITY

Classical accountability originated as a security measure and was employed to ensure that uranium-235 did not fall into unauthorized hands and was not used for unauthorized purposes. Exclusive of the intrinsic worth of the 235U, a rigorous accounting for the physical presence or the irretrievable loss of every milligram of the material was an early requirement.

The distribution of 235U remained under Government control even after the production of this material had increased sufficiently to warrant its wide distribution for peaceful purposes, including the development of power reactors that are competitive with fossil-fuelled plants in areas of high-cost power. The emphasis on control, however, has changed gradually in the intervening years. The emphasis now is on both security measures and financial considerations. The mining, concentrating and use of this scarce and intrinsically valuable material has been subsidized by the Government. The Government, therefore, has had an obligation to maintain surveillance over it, and to assure the taxpayer that the agents of Government were not willfully or carelessly wasting 235U and valuable by-product fissionable materials.
The Government's surveillance programme, often called classical accountability, has been described piecemeal in many publications including various chapters of the USAEC Manual [1]. These and other pertinent publications have been adequately collated, unified and amplified as a unique area of the nuclear industry by Lumb in a publication [2] covering the acquisition, recording, evaluation, and presentation of quantitative data necessary for the control of source and special nuclear materials as defined by the US Atomic Energy Act of 1954. Recent publications on nuclear materials management have divided the accountability field into four distinct areas, each of which is treated separately because the requirements for one may not duplicate the requirements for the others. These areas are:

(a) Research and test reactors [3],
(b) Irradiated fuel reprocessing [4],
(c) Reactor fuel fabrication [5], and
(d) Power reactors [6].

The technology of accountability, or nuclear materials control, embraces a measurement system, an accounting system, a statistical analysis, and a quality control system.

A measurement system generates the quantitative data to be accumulated. These data must show: (a) the amount of source and special nuclear material, (b) where it is located, (c) what form it is in (e.g. spare fuel elements, virgin fuel, spent fuel, solid or liquid fuel material, isotopic species, enrichment scrap, etc.), (d) what blending or mixing, if any, has occurred, (e) how much material was originally received, (f) how much has been returned, (g) how much has been consumed or burned up, (h) how much has been reclaimed or recovered, and (i) how much has been lost.

An accounting system records and reports the above-generated data and attaches thereto the corresponding monetary value for the material.

A statistical analysis and quality control system evaluates the validity of the data generated by the measurement system. The usefulness of the measurements system is largely a function of the accuracy employed. If, for instance, a single gram of source and special nuclear material has a monetary value of $5, it is poor economics to employ an accounting system whose cost is $5 or more to recover that gram or to determine where it was lost (unless such an expenditure will preclude future losses; or unless safety, security, or irreplaceability is an overriding factor). It is also poor economics to install a system whose inadequacy leaves in doubt the source of any discrepancies encountered, or the probable reasons for losses that are experienced. Economics will indicate some effective middle ground as a basis for establishing system adequacy and cost.

Experience has shown that optimum operating and economic efficiency for a source and special nuclear materials system is achieved by subdividing a plant into a series of material balance areas (MBAs). In each of these areas, the input, output and inventory are treated in accordance with the items noted in the previous paragraphs. Thereafter, cross comparison of the audits for the various MBAs quickly shows discrepancies which may be the result of error, system inadequacy, losses, or other causes. This type of system [2, 3, 4, 5, 6] satisfactorily complied with Government requirements when the Government was the sole owner of source and special nuclear materials. There have naturally been many improvements over the years in
the mechanics of handling the complicated accounting procedures necessary to keep track of so precious a commodity. To minimize errors of omission and duplication, erroneous entries, and the like, some large operating plants have computerized their accounting systems. For instance, the General Electric Company, operator of the Hanford, Washington, plant, has published the format of the computer code it employs [7]. The change from a manual to a computerized accounting system has removed much of the book-keeping drudgery from nuclear materials management. It has eliminated many working papers and general ledgers while maintaining intact the fundamentals of the system described above. The General Electric Company's simplification is applicable not only to the Hanford plutonium production plant, but, after suitable modification, to university-owned research reactors or to power reactors owned by public utilities.

Now that private ownership of source and special nuclear materials is permissible, a public utility company must consider that a nuclear power reactor is no longer a subsidized research and development tool. Therefore, large losses of source and special nuclear material can no longer be tolerated in the construction and operation of a power reactor. With private ownership of nuclear materials, most nuclear power plants will be competing with fossil-fuelled power plants. The public utility company operating a nuclear power plant must account to its stockholders for profits and losses. Therefore, it can be expected to exercise a more thrifty nuclear materials control policy than did the Government, whose objective was to assess the feasibility of employing nuclear energy.

Recognizing this eventuality, George gives some cogent reasons for the necessity of source and special nuclear material control [8]. He and Lumb are among the first to visualize that with increasing participation of industry in the nuclear field and the consequent de-emphasis of Government regulation, industry will of necessity increase the responsibility of the specialists who have been handling classical accountability [2, 8]. The demands on these specialists have inevitably broadened to encompass nuclear materials management in pre-reactor technology, in-reactor technology, and post-reactor technology.

3. PRE-REACTOR NUCLEAR MATERIALS MANAGEMENT

In practice, the evolution from accountability control to nuclear materials management has been occurring for the last several years. A nuclear materials manager must be familiar with and experienced in a number of interrelated and interdependent technical areas of disciplines, some of which are unique requirements for competence in the nuclear field. It is not a prerequisite that the nuclear materials manager be expert in each discipline. However, he must possess a working knowledge which enables him to understand and use the information, and to direct his staff and associates in obtaining the necessary data in the fields of: (a) accounting, (b) nuclear physics, (c) health physics, (d) chemical engineering, (e) statistics, and (f) economics or business management.

This statement of required qualifications may, at first glance, seem to be preposterously exaggerated. However, their necessity is a function
of the complexity of the operation for which the nuclear materials manager is responsible together with the size of the investment that is being risked. In the case, for instance, of a nuclear materials manager responsible to a utilities group for the economical operation of a power reactor, one has simply to ask a few questions to visualize the extent to which the owners must depend upon the manager's specialized competence:

(a) Is it cheaper to make a single withdrawal of feed material (UF₆) from the cascades than to make several separate withdrawals? Will the lease charges on a single withdrawn lot (or cost of money plus loss of interest) more than offset a plurality of withdrawal charges spread over an appreciable time?

(b) Is the conversion plant (UF₆ to UO₂) critically limited, or can it handle the conversion at a rate consistent with a single withdrawal?

(c) At what rate can the manufacturer fabricate UO₂ powder to pellets and to finished fuel assemblies? Will UO₂ be a money-consuming inventory in the manufacturer's shop if a single withdrawal of UF₆ is made?

(d) Is it cheaper to lease fuel or to have private ownership?

Each question, and its answer, suggests a host of associated questions and ramifications; and each question may involve a considerable sum of money. The cost of money borrowed for one reactor fuel loading may be the margin between profit and loss in a highly competitive market. The physical loss of an appreciable portion of the ²³⁵U inventory could spell financial disaster, not only because of the inherent value of the source and special nuclear material, but also because of the consequent necessity to defray the costs of investigation and to defend against lawsuits for real or imaginary damages.

The duties and responsibilities of the nuclear materials manager begin before the fuel for a reactor is placed on order. Expensive delays may be incurred if adequate forethought and preparation are not devoted to the following subjects:

(a) The company must be qualified to receive or possess special nuclear materials. This means that the company or one of its plants must have a licence or some other accredited basis to receive or possess source and special nuclear material. As a corollary to this, any proposed converter, shipper, and fabricator must also be suitably accredited.

(b) To obtain accreditation for receiving or possessing source and special nuclear materials, it is essential that requisite personnel and an adequate mechanism be provided to conform with the health physics and safety requirements [9, 10] for all areas in which the source and special nuclear material will be located or through which it will pass.

(c) If the nuclear material is to be leased, a suitable accounting system must be provided to follow the transfers of the material and to report thereon periodically to the Government. If the source and special nuclear material is to be privately owned, elementary prudence dictates that an adequate accounting system be available for surveillance.

(d) A request for an allocation of starting material (UF₆) of the required enrichment in ²³⁵U must be made to the Government or cascade operator at a sufficiently early date to ensure its availability at the required time. To minimize lease charges or the cost of money, a schedule should be prepared
showing the information which the nuclear materials manager has obtained. He should have planned for: (a) the number of withdrawals, dates of withdrawals, and quantities of withdrawn UF₆, (b) dates of shipment of UF₆ to converter, and quantities to be shipped, (c) dates of shipment of UO₂ from converter to fabricator, and quantities to be shipped, and (d) dates of shipment of fuel elements from fabricator to reactor, and number of fuel elements to be shipped; with the amounts of contained source and special nuclear materials indicated.

These and other pertinent data, with special reference to shipping regulations and insurance, are discussed elsewhere in more detail [1, 11, 12, 13, 14, 15, 16, 17].

4. IN-REACTOR NUCLEAR MATERIALS MANAGEMENT

The nuclear materials manager must determine the optimum methods for realizing the full value of the intrinsically valuable nuclear fuel. This responsibility is a continuous one. It must be exercised when selecting and purchasing the fuel for a reactor, when the manager is establishing a system for accounting and surveillance of the fuel during the processing and manufacturing operations, when the fuel is being delivered to the reactor site, and when it is being loaded into the reactor. The nuclear materials manager must ensure that no step in the overall process results in diminishing fuel value.

In a power reactor that must be operated on a continuous basis, the amount of reactor downtime (periods when the reactor is inoperative because of the necessity to change fuel, perform maintenance, or for any other cause) represents not only loss of revenue but additional expense. The amount of reactor downtime must, therefore, be minimized by nuclear materials management planning (excluding, of course, unforeseeable events such as malfunctioning of equipment).

The most important single consideration affecting downtime of a reactor is the number of refuelling periods per reactor lifetime. If a reactor may be refuelled under power and without shut-down, there is little or no loss of revenue to affect economics. But if the reactor requires shut-down for refuelling, it is obviously preferable to plan for a shut-down every three years rather than every nine months. The interval between refuelling periods is a function of the nuclear lifetime of the fuel elements, that is, the amount of burn-up or depletion of fissionable isotope before subcriticality occurs [18]. This, in turn, is primarily dependent upon the initial enrichment of the fuel. Consequently, the nuclear materials manager must coordinate with the design engineers and the physicists to effect desirable compromises tending towards lower costs of power [19]. Three of the more important areas of compromise to which the nuclear materials manager must be a party include the areas of enrichment, fuel shuffling and the equilibrium operation of the reactor. These areas affect nuclear materials cost, inventory and utilization; and they correspondingly affect the cost of power.
4.1. Enrichment

The higher the enrichment in fissionable isotope, the greater is the initial cost of the fuel. The higher the enrichment, the longer is the nuclear lifetime and the greater the potential burn-up. But, the higher the enrichment, the more difficult and costly is the control to hold down excess reactivity. Consequently, in the fuel element design stage, a compromise must be reached about the enrichment that will be employed for longer burn-up and less downtime versus the cost of enrichment.

4.2. Fuel shuffling and the cost of control

Fuel shuffling also aids in realizing the full value of the nuclear fuel. During reactor operation, fuel burn-out is more rapid at the centre of the reactor core than at the outside. Therefore, when the reactor reaches subcriticality, the central portion of the fuel core will be most depleted in fissionable material. Some excess reactivity, however, will still remain in the outer fuel regions. Consequently, removing the entire fuel loading as a single batch is usually wasteful. Early in reactor design, the nuclear materials manager, in collaboration with the physicists and the design engineers, determines whether the fuel can be more economically rearranged or replaced in checkerboard fashion (between areas of high and low flux peaking) or in zone fashion (concentric zonewise from outside toward the centre of the core or vice versa). Zonewise rearrangement of fuel from the outside toward the centre has been found to be the most economical in most cases involving long burn-up periods. It is, therefore, necessary to determine whether two equal concentric zones of fuel elements, or three such zones, or some other number of zones, will afford the best compromise between highest burn-up and minimum downtime for a given reactor. An accurate inventory of rearranged or replaced fuel must, of course, be maintained.

4.3. Equilibrium operation of the reactor

The reactor is said to be on equilibrium operation when each downtime period for fuel replacement and rearrangement occurs at a constant time interval, and when a constant number of fuel elements of constant enrichment is employed to replace a corresponding number of spent fuel elements of constant average depletion or burn-up. In general, equilibrium operation of a reactor is the most economical method. Several reactor cycles of fuel replacement with associated periods of reactor operation (nuclear lifetime of the fuel zones) and of reactor downtime may be required to reach equilibrium. The decision will depend on the fuel management scheme employed. The total elapsed time from reactor start-up to the attainment of equilibrium may vary from 2 to 5 years. The difference in time between the minimum achievable by means of a particular fuel management scheme and the actual elapsed time for equilibrium operation by an alternate fuel management scheme represents money lost to lease charges, interest, or other liability. The nuclear materials manager must, therefore, balance out this disadvantage against any additional costs entailed by any shortening of the ap-
proach to equilibrium, such as by employing zone loadings of different enrichments with correspondingly complex schemes for reactivity control.

5. POST-REACTOR NUCLEAR MATERIALS MANAGEMENT

Before the removal of the first lot or zone of burned-up fuel, the nuclear materials manager must have ordered and stored a replacement lot. Upon attainment of subcriticality, the reactor is shut down; and the predetermined lot of burned-up fuel, usually the innermost zone of the reactor, is removed and stored in the cooling pool. It is inventoried out of the reactor and into the pool in the usual manner. The remainder of the in-reactor fuel is then rearranged in the reactor. A record is kept of each fuel element and its new location. This rearrangement, in its simplest form, involves moving to the empty places at the centre of the reactor a predetermined zone of less depleted fuel elements which had concentrically surrounded the removed lot. The next outer zone is then moved inward to fill the new vacancies, and this process is repeated until the outermost zone of the reactor is vacant. This vacant zone is then filled with fresh fuel elements, and reactor operation is restarted.

The spent fuel removed from the reactor is held in the cooling pool usually from 90 to 120 days, for decay of short-lived isotopes. At this point, additional responsibilities devolve upon the nuclear materials manager. The more important of these include the decisions for throw-away fuel, reprocessing batch size, shipping, refabrication and integral reprocessing of spent fuel.

5.1. Throw-away fuel

If the fuel was originally of low enrichment and, therefore, low in cost, it may not be worth the expense of chemically reprocessing it for removal of accumulated nuclear poisons and recovery of accumulated fissionable isotopes. This economic balance must be determined by the nuclear materials manager. It may be more economical to purchase new fuel, and even to have the fresh fuel toll-enriched instead of purifying the spent fuel for recycle into the re-enriching cascade.

If the decision is made not to reprocess, a cursory review of most of the references quoted up to this point will quickly establish that the spent fuel must not be literally thrown away. Since it costs money to dispose of spent fuel legitimately, the most economical method of disposition must be determined. Ordinarily, this is accomplished by burial at the reactor site or by arrangement with a commercial waste disposal agency.

5.2. Reprocessing batch size

If it has been determined that the spent fuel contains economically recoverable fissionable isotope values, this fuel will be held for chemical reprocessing. The next important consideration then becomes the difference in cost between shipping and reprocessing this zone of spent fuel immediately at the end of its cooling period, or accumulating two or more zones for simultaneous reprocessing. Cost calculations must be performed to determine the minimum penalty that will be incurred per batch of fuel shipped to
reprocessing. These cost calculations depend on the capacity of the reprocessing plant, the daily plant charges, the cost of minimum turn-round time to decontaminate the plant, and the amount of total spent fuel to be reprocessed. Against this cost must be charged the accumulation of use charges or inventory charges plus cost of storage space for the spent fuel after it has left the reactor. The minimum difference represents the most economical course to follow.

5.3. Shipping

From an administrative viewpoint, the shipment of spent, radioactive fuel is far more difficult than the shipment of virgin or fresh fuel. There are many Federal Government, state, municipal, local and carrier regulations with which the shipper must comply. The nuclear materials manager must, therefore, be fully conversant with these regulations because an unforeseen delay in the course of transportation could cause a serious financial loss. In addition to administrative features, there are such economic considerations as the manufacture, use and depreciation of shipping casks versus the cost of renting casks for specific shipments of radioactive fuel.

5.4. Refabrication

When reprocessing has been completed, the purified fuel will probably be too depleted in fissionable material to sustain a chain reaction in another reactor cycle. Therefore, it must be toll-enriched to a predetermined level, or some additional high-enriched fuel must be blended with it to provide the necessary excess reactivity. The nuclear materials manager will obviously take that approach which results in the lesser cost. The fuel is ready for return to the fabricator for repetition of the entire cycle after re-enrichment by either process.

5.5. Integral reprocessing of spent fuel

The nuclear materials manager must also be able to advise his management on the cost requirements for an integral reprocessing and refabrication plant. Some companies are offering such a plant as an adjunct to the nuclear power plant. With such an adjunct, the spent fuel is not shipped away from the power reactor but is reprocessed at the reactor site. The fuel is not completely decontaminated, as it would be in a conventional reprocessing plant. Instead, it is treated to remove the bulk of the fission-product activity, that is, to effect a fission-product decontamination factor of about $1 \times 10^3$. The resulting fuel will then have an activity level comparable to the steady-state level of the isotopic constituents. Under these circumstances, the partly reprocessed fuel may be handled remotely in "warm cells" and refabricated into fuel elements by remote means. This may be accomplished with or without the introduction of additional fissionable material to compensate for that which has been burned up. The reconstituted fuel is then returned to the reactor. A typical integral reprocessing and refabrication plant has been described elsewhere [20, 21].

Such a concept has certain obvious advantages. With an adjacent plant sized to handle the output of a reactor or reactor complex, there is no ne—
cessity to provide expensive shipping casks, to ship highly radioactive fuel long distances, or to pay for complete decontamination. The write-off of the capital cost and the operating cost of the adjunct plant are the disadvantages of this system. The concept is economically attractive, particularly when the operating reactor is far removed from an established reprocessing facility.

6. FUEL CYCLE COST ANALYSIS

The determination of all the above factors and associated costs, their unification and correlation for a specific reactor, and their summation as the total cost of the energy produced by a reactor constitute a nuclear power cost analysis. A fuel cycle cost is most meaningfully expressed in terms of unit energy cost for a plant, that is the cost of producing one electrical kilowatt of energy under plant equilibrium conditions. In the United States and Canada, unit energy cost is expressed in mills per kilowatt hour ($0.001/kWh).

Fixed charges associated with the retirement of capital costs and plant operating costs are the two largest components of nuclear power costs. These vary from country to country in accordance with the method of financing employed by various groups. These costs are the responsibility of the financiers rather than of the nuclear materials manager. Nevertheless, the nuclear materials manager must be aware of these assessed costs and of the basis on which they have been accumulated. Incorrect allocation of cost items can adversely affect the areas for which the nuclear materials manager is responsible.

Although the fuel cycle cost is the most complex, it is not the greatest component of nuclear power cost. Fuel cycle cost also exhibits the greatest variability not only between various reactor types (boiling water versus pressurized water, for example), but also within the same reactor type as a function of reactor design. There is ample experience and precedent for the financiers to establish fixed charges on a power plant irrespective of nuclear or fossil-fuel heating, but there are few data available on the over-all operating costs of nuclear plants. There is still less experience with actual nuclear fuel cycle costs. Consequently, there is still no common basis for allocating costs in these areas in the same arbitrary manner as is done in the assigning of fixed charges. A certain amount of educated guessing on in-reactor costs and post-reactor costs must be incorporated with the ascertainable pre-reactor costs to reach the probable truth on total cost.

The man best fitted by training and experience to make educated guesses in these areas is the nuclear materials manager. He is also best fitted to assemble and consolidate the many pieces of nuclear fuel cost data discussed earlier. He can be trusted not to overlook some essential items and to avoid the introduction of fictitious costs which would penalize nuclear plants.

A number of fuel-cycle cost analyses covering different types of reactors have been published [22, 23, 24, 25, 26]. In the United States, the most widely employed guide is popularly known as the AEC Cost Handbook [27]. At the Third United Nations International Conference on the Peaceful Uses of
Atomic Energy, no less than 35 papers dealt at least in part with fuel-cycle cost analysis. Difficulties encountered in attempting to correlate or compare most of the published papers may be traced to the lack of standardization in assembling the cost components employed. Such difficulties have been minimized in two publications [26, 27] which were prepared by personnel thoroughly familiar with nuclear materials management.

In summary, the nuclear materials manager is responsible for and must be adequately prepared to handle a broad spectrum of duties. In addition to keeping current account of the inventory of high cost source and special nuclear materials, the nuclear materials manager is responsible for literally managing these materials throughout their entire useful cycle from mine to burial. In managing these materials, he must see that they flow through the cycle with maximum efficiency and with the lowest attainable cost. Finally, he is responsible for recommending to management any changes consistent with physics data that will improve the competitive position of his plant.

REFERENCES

[16] UNITED STATES ATOMIC ENERGY COMMISSION, Indemnification of Atomic Energy Activities and Operations of Advisory Committee on Reactor Safeguards.
(26) VALLANCE, J.M., Proc. 3rd UN Int. Conf. PUAE I1 (1965) 56.
(27) UNITED STATES ATOMIC ENERGY COMMISSION, Guide to nuclear power cost evaluation, TID-7025 Vols 1-4, inclusive (1962).
CONTROL POSSIBILITIES ON THE BASIS OF THE MATERIALS BALANCE. The person responsible for handling a basic material needs to have, for each conversion plant:
- A breakdown between the different types of expenditures and the losses;
- Information on production yield, when failure to achieve a certain level is subject to penalties, as is generally the case for materials of such high value as basic nuclear materials.

For this purpose he has available the materials balance, which is simply a study of the "law of the conservation of matter" over a given accounting period. It can be written simply as

\[ q_e + q_v_i = q_s + q_v_f + q_p \]

where \( q_e \) denotes the quantity of incoming material, \( q_v_i \) the reserves at the beginning of the period, \( q_s \) the quantity of outgoing material, \( q_v_f \) the reserves at the end of the period, and \( q_p \) the losses.

Analysis of the different factors shows that these are variates defined by a mean value and a variance. There is thus an area of uncertainty for closing the balance sheet and, conversely, if the size of this area is fixed, the variance to be applied to each factor contained in the expression for the materials balance can be deduced from it.

If the balance does not come out within the area of uncertainty, there is an implied systematic error in the determination of at least one of the factors. Complete analysis of the terms by which the different items of the balance are assessed will reveal the factor responsible.

In the last part of the paper, the practical application of the method to a natural uranium refinery is described.
Возможные методы контроля на основе материального баланса: Лицу, отвечающему за основные материалы, необходимо иметь в отношении каждого перерабатывающего завода следующее:

- классификацию различных расходов и потерь;
- сведения об эффективности предприятия, которая не должна быть ниже определенного уровня, в противном случае это влечет за собой материальную ответственность, как это и случается с такими ценными материалами, как ядерные.

Материально ответственное лицо располагает материальным балансом, который является не чем иным, как выражением "закона сохранения материи" за данный отчетный период. Его можно выразить формулой:

\[ q_e + q_{vi} = q_i + q_{vf} + q_p, \]

где, \( q_e \) — поступающее количество материала, \( q_{vi} \) — все материалы, находящиеся на предприятии в начале отчетного периода, \( q_i \) — материалы, выходящие из предприятия, \( q_{vf} \) — все резервы предприятия на конец отчетного периода и \( q_p \) — потери.

Анализ различных факторов показывает, что эти факторы являются случайными величинами, определяемыми средним значением и отклонением. Из этого вытекает известная возможность погрешности при закрытии баланса. Наоборот, если предусмотреть размер этой области, то из этого можно определить отклонения, применяемые в отношении каждого фактора, содержащегося в выражении материального баланса.

Если баланс не сходится в упомянутой области, то это означает, что происходит систематическая ошибка при определении хотя бы одного из этих факторов. Полный анализ членов формулы, определяющих различные статьи баланса, должен привести к выявлению фактора, который вызывает ошибку.

В заключение дается описание практического применения метода на предприятии по очистке природного урана.

Controles basados en el balance de materiales. Toda persona encargada de administrar materiales básicos desea conocer los datos siguientes para cada planta de transformación:

- El balance pormenorizado de las diversas salidas y pérdidas.
- El rendimiento de fabricación en los casos en que de ser inferior a cierto valor se penaliza al fabricante como generalmente ocurre cuando se trata de materiales de valor tan elevado como los materiales básicos nucleares.

Para ello dispone del balance de materiales que es simplemente la ley de conservación de la materia estudiada en el curso de un ejercicio dado. Se puede escribir simplemente

\[ q_e + q_{vi} = q_i + q_{vf} + q_p, \]

donde \( q \) designa las cantidades correspondientes a las entradas, las reservas al comienzo del ejercicio, las salidas, las reservas al final del ejercicio y las pérdidas.

El análisis de los diversos factores muestra que se trata de variables aleatorias definidas por un valor medio y una variancia; de ellas resulta un área de incertidumbre para el cierre del balance. Inversamente, si se predetermina la amplitud de esa área se deducirán de ella las variancias correspondientes a cada uno de los factores que intervienen en la expresión del balance de materiales.

El análisis completo de las condiciones de evaluación de los diversos elementos del balance debe llevar al descubrimiento del factor erróneo.

La memoria termina refiriendo todas esas consideraciones a una refinería de uranio natural.

Le contrôle de l'utilisation des matières nucléaires dans une usine de transformation est basé sur le bilan matières. Nous nous proposons dans ce mémoire d'étudier les caractéristiques propres à un bilan et ses différents composants, et d'en tirer les conséquences concernant le contrôle.
1. CARACTERISTIQUES GENERALES DU BILAN MATIERES

Les lois de la conservation de la matière s'appliquent, bien entendu, aux usines de transformation des matières nucléaires: usines de concentration de minerais, raffineries, ateliers de fabrication des éléments combustibles de retraitement des combustibles irradiés, usines de séparation isotopique...

Pour une durée d'exercice donnée et chaque élément chimique intervenant dans les transformations, on peut donc écrire la relation

\[ q_e + q_{vi} = q_{vf} + q_s + q_p \]  

(1)

où les \( q \) désignent les quantités de l'élément considéré respectivement dans l'entrant, le volant de fabrication et les stocks des produits intermédiaires en début et fin d'exercice, les produits finis et les pertes de fabrication (\( e = \) entrant, \( vi = \) volant initial, \( s = \) sortant, \( vf = \) volant final, \( p = \) pertes).

Faire le bilan matières de l'exercice consiste à déterminer chaque terme des équations (1), ce qui permet au gestionnaire de localiser l'élément qu'il est chargé de comptabiliser.1

Dans tous les cas pratiques qui nous intéressent, le processus de détermination fera appel aux phases suivantes:
- mesure de la masse ou du volume des lots de matières
- échantillonnage des lots
- analyse de l'échantillon.

Au cours de chacune de ces opérations, on commet un groupe d'erreurs et il serait vain d'aborder l'une d'entre elles sans se soucier des conditions dans lesquelles les autres se produisent.

Ces erreurs ont les causes les plus diverses et se composent pour faire apparaître une erreur résultante, en sorte que les quantités déterminées sont des variables aléatoires soumises généralement à la loi normale et qu'on peut caractériser par les deux premiers moments de la loi de distribution:
- la moyenne (moment d'ordre 1)
- la variance (moment d'ordre 2).

La différence entre la moyenne et la valeur vraie traduit l'inexactitude de l'ensemble des opérations; on l'appelle habituellement l'erreur systématique. La variance est l'expression de la variabilité autour de la moyenne quant on répète les opérations; elle traduit l'imprécision de la mesure.

Il en résulte qu'il est impossible de connaître exactement les quantités d'éléments aux différents stades de la production. En l'absence de toute erreur systématique et en adoptant un intervalle de probabilité de 95% que l'on peut admettre comme une certitude pratique, la valeur mesurée ne différera pas de la valeur vraie inconnue de plus de deux écarts types (l'évaluation des différents écarts types s'obtient en recommençant un certain nombre de fois la même opération de détermination).

La relation (1) s'écrit alors:

\[ q_e \pm 2\sigma_{qe} + q_{vi} \pm 2\sigma_{qvi} = q_{vf} \pm 2\sigma_{qvf} + q_s \pm 2\sigma_{qs} + q_p \pm 2\sigma_{qp} \]  

1 CEA-CETAMA, Journée de l'Analyse 1963.
Les processus de détermination de chaque terme étant indépendants les uns des autres, les variables aléatoires correspondantes ne sont pas liées au sens statistique. On est donc en droit de composer leurs variances de façon additive et de remplacer l'égalité précédente par la relation:

\[
q_e + q_{vi} + q_s + q_p \leq 2(\sigma_{q_e}^2 + \sigma_{q_{vi}}^2 + \sigma_{q_s}^2 + \sigma_{q_p}^2) \text{ près (2)}
\]

Dans le cas où le bilan ne boucle pas, compte tenu de la «plage d'incertitude», on est sûr d'avoir mis en évidence une erreur systématique sur l'un au moins des postes. Ces erreurs sont évidemment sournoises. Leurs causes peuvent être multiples, le remède est unique: soumettre toutes les méthodes de détermination utilisées et leur mise en œuvre à l'examen critique le plus minutieux; la connaissance de l'ordre de grandeur de cette erreur grâce au bilan est un élément précieux pour la bonne réussite de l'enquête.

Le fait que le bilan soit satisfaisant ne permet pas, par contre, de conclure à l'absence d'erreur systématique, deux erreurs pouvant se compenser ou une erreur pouvant être dissimulée par l'étendue de la plage d'incertitude.

Du point de vue du contrôle, il est certes intéressant d'avoir une plage aussi petite que possible, mais augmenter la précision demande la multiplication ou le raffinement des déterminations, ce qui n'est pas sans incidence sur le prix de revient de l'opération. En fin de compte, c'est un compromis entre la valeur du produit et le coût du contrôle qui permettra de fixer l'importance de la plage.

Les matières nucléaires ayant une valeur élevée, le contrôle sera très poussé. En outre, dans certains cas, on sera amené à aller au-delà de «l'optimum économique» pour des raisons de sécurité, afin de parer à des dangers connus ou supposés, ou même quelquefois pour des raisons psychologiques.

2. ANALYSE DES DIFFÉRENTS COMPOSANTS DU BILAN MATERIERES

2.1. L'inventaire

La détermination des termes \( q_{vi} \) et \( q_{vf} \) se fait par les opérations d'inventaire en début et fin d'exercice. L'importance des volants par rapport au flux qui transite dans l'usine au cours de l'exercice varie considérablement suivant la nature des usines, mais elle est le plus souvent grande.

Le premier travail consistera à imposer à priori une précision globale à l'opération, compte tenu de cette importance, afin d'éviter de masquer des erreurs systématiques significatives sur les autres termes. Cette tolérance sera répartie entre les différents postes de l'inventaire. Un recensement rapide de la nature et de l'importance des produits à inventorier permettra de déterminer pour chaque poste le nombre de mesures à faire, d'échantillons à prélever, d'analyses à exécuter.

Le coût direct ou indirect (immobilisation de personnel ou d'installations) peut paraître alors trop élevé et on sera amené à adopter un compromis.
en admettant une précision globale inférieure, quitte à avoir ultérieurement une certitude plus grande en cumulant les résultats de plusieurs exercices.

À conditions égales, deux facteurs permettent d'atteindre plus facilement l'objectif requis:
- la durée de l'exercice: plus il sera long, moins l'importance du volant sera grande, par rapport à la production, ce qui autorisera une incertitude supérieure lors de l'inventaire;
- l'arrêt de l'usine: en général les circuits de fabrication sont alors vidés et les produits intermédiaires ou les déchets à recycler sont répartis dans un nombre limité de récipients faciles à jaugeer et à échantillonner. Sans préconiser un arrêt spécial pour l'inventaire, on aura donc intérêt à faire coïncider cette opération avec les arrêts prolongés correspondant soit à une fermeture annuelle pour congés, soit à une période de gros entretien, soit à un temps mort entre deux campagnes de traitement dû à un changement de fabrication ou à une surcapacité des installations.

Au point de vue de la comptabilité, la période d'exercice correspondant en général à l'année légale, il est intéressant de prévoir un arrêt suffisant en fin d'année, si un tel arrêt est compatible avec les programmes de production. Si cela n'est pas possible, il vaut mieux se contenter d'un inventaire sommaire permettant l'arrêt des comptes, les compensations se faisant par la suite lorsqu'on aura l'occasion d'opérer de façon précise.

2.2. Le contrôle des réceptions et des livraisons

La détermination de $q_e$ et $q_s$ est effectuée par le contrôle des réceptions et des livraisons. L'opération se répète sur des lots de produits représentant des quantités assez petites. Le nombre de lots mis en jeu au cours d'un exercice est donc important et la précision à obtenir sur chaque lot est $\frac{1}{n}$ fois celle que l'on désire obtenir en fin d'exercice si $n$ est le nombre de lots. On pourra donc accepter pour chaque lot une incertitude assez grande, c'est-à-dire utiliser des méthodes assez rapides et faire relativement peu d'analyses.

Par contre, ces opérations étant répétées un grand nombre de fois, il faut faire une guerre incessante aux erreurs systématiques qui s'ajoutent et peuvent entraîner en fin d'exercice un écart inacceptable. Le bilan matières permettra de mettre en évidence un tel écart si on a pris la précaution d'harmoniser entre elles les précisions sur les différents postes.

Bien entendu, l'analyse critique des méthodes de détermination utilisées et de leur mise en œuvre permettra de déceler les causes de ces erreurs, qui peuvent avoir une incidence commerciale (quantités d'espèces monétaires à échanger entre le client et le fournisseur) et technique (connaissance exacte des rendements).

Si, pour éviter les contestations commerciales, il suffit que la méthode de détermination définie contractuellement et détaillée dans un cahier de charges soit reproductible, il est plus nécessaire techniquement d'avoir des valeurs exactes, les usines de transformation de matières nucléaires ayant souvent un rendement minimal imposé très élevé pour éviter le gaspillage de produits rares et pour des raisons de sécurité.
2.3. Les pertes

Le contrôle des pertes a ceci de commun avec les opérations précédentes qu'il fait l'objet de déterminations fréquentes (il peut même se faire en continu).

La aussi, il n'y a pas lieu de rechercher des méthodes précises mais il faut éviter les erreurs systématiques. L'une d'entre elles peut être en particulier l'ignorance d'une source de pertes, même minime, si elle est continue, ou le fait de la négliger. A cet égard, les plus subtiles sont les pertes à l'atmosphère, que ce soit sous forme de fuite gazeuse, d'évaporation ou d'entrainement de poussières.

 Certes les industries nucléaires s'imposent les plus grandes précautions, non pour des considérations techniques, mais pour des raisons évidentes de sécurité. L'étanchéité des circuits est particulièrement soignée, les postes générateurs de poussières sont mis en dépression et le rejet à l'atmosphère se fait sous contrôle après récupération sur filtres très efficaces. Les appareils de détection de la radioactivité permettent de s'assurer de la bonne marche de l'installation; mais malgré tout les volumes rejetés sont souvent tels qu'il n'est pas interdit de penser qu'ils constituent tout de même une source de pertes que l'on ne prend pas en considération.

D'ailleurs les bilans usine indiquent «les pertes contrôlées», ce qui sous-entend bien qu'il en existe d'autres.

Une grande partie des pertes contrôlées intervient sous forme d'effluents liquides. Le contrôle des volumes est en général assez facile à faire, car le rejet n'est pas direct; les effluents sont analysés pour vérifier leur nonnocivité. Par contre, les teneurs sont très faibles et sont déterminées par des méthodes très sensibles mais peu précises, si bien que l'imprécision sur les pertes est très grande, même en fin d'exercice, malgré les opérations répétées de détermination. Heureusement, les transformations de matières nucléaires se font (c'est une conséquence de leur prix élevé) avec de hauts rendements, obtenus si nécessaire par de nombreux recyclages. La valeur absolue des pertes étant faible, leur imprécision n'influence finalement pas beaucoup sur l'incertitude du bilan.

3. EXEMPLE D'UNE USINE DE RAFFINAGE

3.1. Position du problème

Alimentée en concentrés liquides (nitrates d'uranyle) et solides (uranates de magnésie), une raffinerie produit des lignots d'uranium métal de pureté nucléaire. Le procédé est le suivant: après dissolution des uranates par l'acide nitrique et purification des nitrates d'uranyle par solvant, l'uranium est précipité par du NH₃ gazeux. Le diuranate formé est calciné en UO₃, lui-même transformé en UF₄ dans un four de réduction-fluoruration. L'élaboration du métal se fait par calciothermie.

Le Commissariat à l'énergie atomique (CEA) est propriétaire de l'uranium, la raffinerie travaillant à façon.

Les sorties de l'usine et les inventaires de fin d'année se font contradictoirement entre la raffinerie et le CEA. Par contre, la raffinerie ne
participe pas officiellement à la détermination de l'uranium dans les concentrés, qui s'opère contradictoirement entre le CEA et les usines de concentration. Mais, petit à petit, un système local de contrôle des réceptions a été mis en place.

Or, les inventaires accusent un déficit entre stock réel et stock théorique assez important (de l'ordre de 1% du flux ayant transité dans l'usine au cours de l'exercice) et les réceptions locales indiquent une différence encore plus sensible avec la valeur officielle.

Aucune étude d'ensemble n'avait été faite permettant de dire quelles seraient les limites admissibles pour le déficit compte tenu des processus de détermination utilisés.

3.2. L'enquête

Nous avons eu pour mission de trouver les raisons des différences constatées.

3.2.1. Écarts sur les réceptions

Nous avons commencé par étudier les méthodes utilisées pour déterminer l'uranium dans les concentrés. Celles qui aboutissaient aux valeurs officielles étaient détaillées dans un cahier des charges dont les clauses soigneusement élaborées étaient bien respectées.

Les opérations de contrôle dans la raffinerie avaient évolué dans le temps et, en nous fondant sur une étude statistique des écarts lot par lot, nous avons pu mettre en évidence des biais à plusieurs stades de sorte que les quantités avancées par les raffineries étaient dénuées de valeur.

A titre d'exemple, voici le relevé de quelques causes d'erreurs:
- fonctionnement défectueux d'une étuve de laboratoire ce qui entraînait une erreur importante sur la teneur des uranates,
- passage du poids net humide des uranates déterminé à la raffinerie au poids net sec par adoption de la teneur en H2O mesurée plusieurs mois auparavant à l'usine de concentration, car on n'était pas équipé localement en étuves industrielles pour mesurer cette teneur. Or, un certain nombre de containers, non étanches, voyaient leur humidité évoluer dans le temps,
- erreurs sur les poids nets de nitrates d'uranyle et leurs teneurs: le système local de vidange des containers étant défectueux, on les rinçait à l'eau.

3.2.2. Précision de l'inventaire

Nous avons assisté à un inventaire de fin d'année afin de calculer la précision de l'opération. Les conditions étaient idéales car l'usine était arrêtée, les circuits vidés, et il n'y avait aucun déchet recyclable. L'uranium à évaluer était essentiellement réparti dans deux grandes cuves de nitrate d'uranyle et de nombreux fûts d'UF4.

L'essentiel de l'imprécision est dû au poste nitrate d'uranyle, car on mesure des volumes, alors que l'on pèse les fûts d'UF4. Mais on aurait pu, à travail égal, arriver à mieux en multipliant les analyses sur les nitrates et en réduisant celles sur l'UF4.
Connaissant l'imprécision de la détermination sur les entrants officiels (1% pour un lot) et sur le métal sortant (très bonne puisqu'il s'agit de simples pesées), et ayant déterminé celle de l'inventaire, il restait à évaluer l'incertitude sur les pertes. Les pertes contrôlées sont faibles (~1%) de sorte que même en supposant une grande imprécision (100%) on arrive à la conclusion que la plage d'incertitude de fermeture du bilan est sans commune mesure avec le déficit trouvé à l'inventaire. Il fallait donc conclure à l'existence d'une erreur systématique sur la détermination de l'un au moins des postes du bilan matières.

3.2.3. Recherche des causes d'erreurs systématiques

L'erreur systématique ne pouvait provenir ni des inventaires, ni des sorties. Une étude succinte des pertes contrôlées et des points de pertes possibles autres que ceux pris en considération pour la raffinerie (essentiellement pertes à l'atmosphère par entraînement de poussières) nous a vite convaincus que, s'il y avait sous-évaluation, elle ne pouvait être de l'ordre de grandeur du déficit existant.

Notre effort s'est alors porté sur la détermination de l'entrant. Des recoupements d'après les résultats d'une autre raffinerie traitant exclusivement des nitrates d'uranyle nous ont permis de penser que la méthode d'évaluation de l'uranium contenu dans les uranates entraînait un biais voisin de 0,5%. Résumons rapidement le processus: après remplissage des containers on prélève 1 à 1,5% de la quantité d'uranates à la sonde, l'emplacement des coups de sonde étant donné par un gabarit. Le prélèvement est disposé sur un plateau en une couche d'épaisseur inférieure à 3 cm, pesé immédiatement et séché pendant 36 h au moins dans une étuve ventilée à une température de 140°C ± 10°C. Après refroidissement par air sec le plateau est pesé. On obtient ainsi le taux d'humidité.

L'ensemble des prélèvements correspondant à dix containers constituant un lot est ensuite broyé, mélangé et quarté pour constituer les échantillons servant à la mesure de teneurs en uranium. Les laboratoires qui reçoivent les échantillons sèchent à nouveau à la même température de 140°C les prises pour analyse, afin d'éliminer l'humidité acquise au cours des opérations de conditionnement de l'échantillon.

Nous avons pensé qu'il n'était pas impossible qu'en fin de compte l'état de siccité de l'échantillon servant à la détermination des teneurs soit différent de celui du prélèvement «sec» servant à calculer le taux d'humidité. Des expériences simples (complément de séchage dans une étuve de laboratoire immédiatement après la pesée à la sortie de l'étuve industrielle) ont confirmé ce point de vue, les écarts enregistrés étant de l'ordre de ceux recherchés. Des essais systématiques plus élaborés sont en cours et des solutions de rechange à l'étude (séchage concourent, calcination des prélèvements) pour éliminer cette cause d'erreur.

Nous avons ainsi abouti à expliquer une partie du déficit. Pour le reste les résultats sont moins nets. Un certain désaccord entre différents laboratoires peut laisser supposer que les résultats des analyses officielles de teneurs des nitrates d'uranyle seraient légèrement surestimés. La différence étant inférieure à la précision des méthodes utilisées, la vérifica-
tion sera assez longue. D'autre part, une étude détaillée des pertes dans l'usine de raffinage serait certainement fructueuse.

En conclusion, nous pouvons dire que, dès l'année prochaine, le déficit à l'inventaire constaté dans la raffinerie devrait être ramené dans la fourchette normale d'imprécision correspondant aux méthodes mises en œuvre, soit 0,2% environ.

DISCUSSION

D.E. GEORGE: In your oral presentation you referred to "unofficial" and "close" checks. What do you mean by this?

A. GILLES: By an official check I mean the type of control on which a commercial transaction is based, carried out when the material leaves the concentration plant. The refinery also checks the incoming material but, as this is not done in the presence of both parties, it is of no value for commercial transaction purposes from the point of view of the Commissariat à l'Energie atomique (CEA). Hence, it is an "unofficial" check.

J. SORNEIN: The fundamental problem here is the following. The CEA, which buys and owns the French uranium, is faced with the task of evaluating the quantities of uranium being handled at three successive stages, viz. (1) when the ore is delivered to the concentration plant, (2) when the concentrate is delivered to the refinery, and (3) when the metal, oxide or hexafluoride is delivered to the user. Obviously, the clearest picture of the actual movement of uranium is obtained at the third stage. The problem is to what extent should the accounting records for stages 1 and 2 reflect the discrepancies recorded at the end of the process and to what extent should these discrepancies be attributed to "non-controllable" refinery losses.
NUCLEAR MATERIALS MANAGEMENT IN THE URANOTHORIANITE PROCESSING UNIT OF THE BOUCHET PLANT. The uranothorianite processing unit of the Bouchet plant supplies thorium and uranium salts of nuclear purity, obtained from ore concentrates by a process consisting mainly of the following operations: grinding of the ore, dissolution in nitric acid, successive solvent purification with uranyl nitrate and thorium nitrate, concentration by evaporation and crystallization of the thorium nitrate, and decontamination of the effluents.

To maintain a continuous check on the uranium and thorium, material balances are drawn up daily for each chemical operation and monthly for the plant as a whole. This means that each management unit must determine as accurately as possible the amounts of incoming and outgoing material and the amounts involved in conversion.

While uranyl nitrate and thorium nitrate of nuclear purity can be determined with an accuracy of the order of 0.1%, numerous causes of uncertainty must be eliminated in determining other uranium- and thorium-bearing products. These originate mainly with inaccurately defined volumes or weights, or with heterogeneous and impure solutions or solids which are difficult to sample and analyse.

Thanks to progress in systematically reducing the incidence of these causes of uncertainty in the balances it has become possible during the last few years of operation to maintain quasi-continuous control of nuclear materials. This has proved particularly satisfactory in the case of uranium.

УБЩЕНИЕ С ЯДЕРНЫМИ МАТЕРИАЛАМИ В ЦЕХЕ ПО ОБРАБОТКЕ УРАНОТОРИАНИТА НА ЗАВОДЕ БУШЕ. Цех по обработке ураноторианита на заводе Буше поставляет ториевые и урановые ядерно-чистые соли, получаемые из рудных концентратов с помощью процесса, который состоит из следующих основных операций: измельчение руды, растворение руды в азотной кислоте, последовательная очистка растворителями нитрата урана и нитрата тория, концентрация выпариванием и кристаллизация нитрата тория, дезактивация эффлуентов.

В целях обеспечения постоянного контроля за наличием урана и тория ежедневно составляются материальные балансы по каждой химической операции и ежемесячно по всему заводу.
Составление этих балансов требует по возможности самого тщательного определения количества материалов на входе, на выходе и в процессе обработки в каждой рассматриваемой производственной единице обращения с материалами.

Если ядерно-чистые нитрат уранила и нитрат тория могут быть определены с точностью порядка 0,1%, то необходимо устранить причины, ведущие к ошибкам определения других продуктов, содержащих уран или торий. Причины заключаются, главным образом, в плохом определении объема или веса, либо в неоднородных и нечистых растворах и твердых телах, у которых трудно брать пробы или анализ.

Достигнутые успехи в целях систематического уменьшения воздействия этих ведущих к ошибке причин на материальные балансы позволили за последние годы эксплуатации разработать почти постоянный контроль за ядерными материалами. При этом особенно положительные результаты достигнуты в отношении урана.

ADMINISTRACION DE MATERIALES NUCLEARES EN LA PLANTA DE TRATAMIENTO DE URANOTORIANITA DEL CENTRO DE LE BOUCHET. La planta de tratamiento de uranotorianita del centro de Le Bouchet produce sales de torio y de uranio de pureza nuclear a partir de concentrados de mineral mediante un proceso que comprende las siguientes operaciones principales: moloturación del mineral, disolución en ácido nítrico, purificación por disolventes de los nitratos de uranilo y de torio, concentración por evaporación y cristalización del nitrato de torio, descontaminación de los efluentes.

A fin de asegurar el control del uranio y del torio, se hacen todos los días balances de materiales para cada operación química y, mensualmente, para el conjunto de la planta. Esos balances exigen una determinación lo más exacta posible en cada unidad administrativa de las cantidades de materiales a la entrada, a la salida y en curso de transformación.

El nitrato de uranilo y el nitrato de torio de pureza nuclear pueden determinarse con una exactitud del orden del 0,1%, pero para la determinación de los demás productos uraníferos o toríferos es preciso eliminar numerosas causas de incertidumbre debidas esencialmente a una definición imprecisa de volúmenes o pesos, o a la heterogeneidad e impureza de soluciones o sólidos difíciles de muestrear y de analizar.

Los progresos realizados para reducir sistemáticamente el efecto de estas causas de incertidumbre sobre los balances de materiales han permitido establecer durante los últimos años un control casi permanente de los materiales nucleares, particularmente satisfactorio en lo que respecta al uranio.

1. GENERALITES

L'atelier de traitement de l'uranotherianite, exploité depuis 1957 à l'usine du Bouchet, fournit des sels de thorium et d'uranium de très haute pureté, à partir des concentrés miniers extraits à Madagascar, par un procédé qui comprend les principales opérations suivantes: broyage et séchage du minerai, purification par solvant successivement du nitrate d'uranyle et du nitrate de thorium, concentration par évaporation et cristallisation du nitrate de thorium, décontamination des effluents liquides.

2. PRINCIPE DU TRAITEMENT DE L'URANOTHERIANITE

Le procédé auquel nous nous référions tout au long de ce mémoire a déjà été partiellement décrit dans des publications antérieures [1, 2]; la figure 1 en schématisé les traits essentiels.

2.1. Séchage et broyage du minerai

Le minerai titrant 5 à 30% en uranium et 40 à 60% en thorium est reçu en fûts d'un volume de 50 litres et d'un poids moyen de 180 kg, vidangé,
séché et broyé à une finesse de 150 µm par des opérations télécommandées, en enceinte ventilée et dépoussiérée.

2.2. Dissolution du minerai

Par solubilisation dans l'acide nitrique à 50% à l'ébullition sous reflux et après filtration des insolubles d'attaque, on obtient une solution impure de nitrates contenant l'uranium (40 à 90 g/litre), le thorium (100 à 250 g/litre) et de nombreuses impuretés (fer, plomb, radium, terres rares notamment) ainsi qu'un stérile à faible teneur en uranium (0,02 à 0,64%) et en thorium (0,3 à 0,5%).
2.3. **Purification par solvant de l'uranium**

L'indispensable séparation uranium-thorium que comporte le traitement de tout minerai contenant ces deux éléments est réalisée sur la solution impure de nitrates par extraction sélective et quantitative de l'uranium au tributylphosphate, dans une batterie de 16 mélangeurs décanteurs [3]. On obtient alors deux solutions: une solution de nitrate d'uranyle qui, du fait de sa très haute pureté, peut servir directement à la préparation du diuranate d'ammonium de pureté nucléaire; et une solution impure de thorium, débarrassée d'uranium.

2.4. **Purification par solvant du thorium**

Le nitrate de thorium produit au stade précédent subit par des procédés connus [4, 5], dans une série de cinq colonnes agitées, une purification au tributylphosphate qui donne naissance à deux nouvelles solutions: d'une part, un pied de colonne d'extraction, solution de nitrates métalliques, débarrassée d'uranium (moins de 20 mg/litre) et de thorium (moins de 0,5 g/litre) qui est rejetée après décontamination dans une station spécialisée de traitement des effluents; d'autre part, une solution pure de nitrate de thorium titrant 80 à 100 g/litre.

2.5. **Concentration et cristallisation du nitrate de thorium**

La solution pure de nitrate de thorium est concentrée dans un appareil à triple effet jusqu'à 600 g/litre, après déphosphoration préalable au diluant [6] destinée à éliminer les entraînements de solvant, puis cristallisée dans un finisseur; on produit par essorage des cristaux très purs de nitrate de thorium pentahydraté [7].

2.6. **Décontamination des effluents**

On élimine les radioéléments des pieds de colonne, avant rejet, en deux stades successifs: tout d'abord le radium, par entraînement au sulfate de plomb à pH 1,5; ensuite, les autres radioéléments dont l'uranium et le thorium non éliminés au stade précédent, par précipitation sodique à pH 7,5. Deux types de déchets sont recueillis: d'une part, un sulfate de plomb radioactif, de haute activité, titrant 0,1% en thorium et dépourvu d'uranium; d'autre part, un gâteau d'hydroxydes de faible activité, contenant 0,02 à 0,03% d'uranium et 0,3 à 0,4% de thorium.

3. **CONTROLE DE L'URANIUM ET DU THORIUM**

Si, comme dans toute industrie mettant en œuvre des produits rares et coûteux, une saine gestion économique requiert la mise en place d'un plan de contrôle suffisamment sensible des mouvements de matière, l'uranium et le thorium, par leur radioactivité propre ou celle de leurs descendants toujours associés, imposent des sujétions particulières. En particulier le respect de la sévère réglementation fixant les teneurs maximales de rejet.
dans les effluents exige un contrôle permanent de l'uranium et du thorium d'une rigueur inhabituelle dans les autres industries chimiques.

Pour assurer ce contrôle, des bilans sont dressés quotidiennement, à partir des données enregistrées sur les feuilles journalières de fabrication relatives à chaque opération chimique. Pratiquement, l'établissement de ces bilans nécessite la détermination des quantités d'uranium et de thorium contenues dans tous les produits entrant, sortant ou en cours de transformation dans chaque unité de gestion, dont le tableau I donne une liste récapitulative.

4. ELABORATION DES DONNEES QUANTITATIVES NECESSAIRES A L'ETABLISSEMENT DES BILANS MATIERE

4.1. Minerai d'uranotherianite

Le minerai est pesé fût par fût et des teneurs moyennes sont déterminées par lots d'arrivée.

a) Échantillonnage

Les fûts de minerai, groupés par lots de 30 à 40 t, sont repris par un basculateur placé dans une enceinte spéciale en raison de la production possible de poussières radioactives et du dégagement de thoron ou radon pendant les opérations de préparation physique de minerai. Après un rapide tamisage le minerai est déchargé dans une trémie de stockage en charge sur la sole doseuse qui alimente le séchoir vertical à plateaux fins et tournants. La cuillère actionnée par une minuterie électrique d'un échantillonneur automatique MINEMET, intercalé entre la sole doseuse et le séchoir, prélève toutes les 30 s un échantillon primaire représentant environ 1% du minerai débité.

Cet échantillon primaire, d'un poids de 300 à 400 kg par lot, est vidé dans une trémie mobile à trappe coulissante qu'un palan alimente au sommet d'une tour d'échantillonnage, entièrement automatique, supprimant tout contact du personnel avec les poussières ou le thoron (fig. 2). Le prélèvement effectué après concassage par un échantillonneur primaire MINEMET réduit le poids de l'échantillon à une dizaine de kilogrammes. Un second prélèvement, réalisé après broyage dans un broyeur à cylindre et deux broyeurs à disques, par un second échantillonneur MINEMET réduit finalement le poids de l'échantillon précédent à 600 g. Un répartiteur le divise en douze fractions recueillies dans des flacons disposés en couronne. On réunit les produits contenus dans les bouteilles respectivement et diamétralement opposées pour obtenir six échantillons définitifs pesant chacun environ 100 g.

b) Analyse chimique

- Détermination du thorium:

On dissout le minerai dans l'eau régale, puis on extrait le thorium par un mélange tributylphosphate - white spirit en milieu nitrate d'ammonium-acide nitrique. L'oxalate de thorium précipité dans la solution aqueuse de
<table>
<thead>
<tr>
<th>Unité de gestion</th>
<th>MATERIES CONTROLEES</th>
<th>A l'entrée</th>
<th>A la sortie</th>
<th>En cours de transformation</th>
</tr>
</thead>
</table>
| Préparation du minerai                 |                      | **Minerai tout venant**                                                   | **Minerai sec et broyé**                                                   | a) **Minerai tout venant**  
|                                        |                      |                                                                           |                                                                           | b) **Minerai sec**  
|                                        |                      |                                                                           |                                                                           | c) **Minerai broyé**  |
| Dissolution du minerai                  |                      | **Minerai sec et broyé**                                                   | a) **Stéries d'attaque**                                                   | a) **Pulpes d'attaque**  
|                                        |                      |                                                                           | b) **Solution impure de nitrates d'uranium et de thorium**                  | b) **Solutions de nitrates**  
|                                        |                      |                                                                           |                                                                           | c) **Gâteau de filtration**  |
| Purification par solvant de l'uranium   |                      | **Solution impure de nitrates de thorium et d'uranium**                   | a) **Solution pure de nitrates d'uranyle**                                 | a) **Solutions aqueuses de nitrates**  
|                                        |                      |                                                                           | b) **Solution impure de nitrates de thorium**                              | b) **Solutions organiques de nitrates**  |
| Purification par solvant du thorium     |                      | **Solution impure de nitrates de thorium**                               | a) **Solution pure de nitrates de thorium**                               | a) **Solutions aqueuses de nitrates**  
|                                        |                      |                                                                           | b) **Pieds de colonne d'extraction**                                        | b) **Solutions organiques de nitrates**  |
| Concentration et cristallisation du nitrates de thorium | | **Solution pure de nitrates de thorium** | **Nitrates de thorium cristallisé pentahydraté** | a) **Solutions de nitrates de thorium**  
|                                        |                      |                                                                           |                                                                           | b) **Suspensions de nitrates de thorium cristallisé**  |
| Décontamination des effluents           |                      | **Pieds de colonne d'extraction**                                         | a) **Effluents décontaminés**                                              | a) **Solutions de nitrates**  
|                                        |                      |                                                                           | b) **Sulfate de plomb**                                                    | b) **Pulpes de précipitation**  
|                                        |                      |                                                                           | c) **Gâteau d'hydroxydes**                                                 | c) **Déchets solides**  |
réextraction est transformé en oxyde par calcination à 1000°C durant une heure et demie.

- Détermination d'uranium:
Elle se fait sur les eaux mères de précipitation oxalique du thorium de la détermination précédente, par précipitation et calcination du diuranate d'ammonium après élimination des ions oxaliques. Une correction est effectuée par reprise nitrique diluée de l'oxyde calciné.

4.2. Nitrate de thorium cristallisé

Les cristaux de nitrate de thorium sont conditionnés en fûts préalablement tarés avec leur sac intérieur en polythène et ajustés au poids constant
de 330 kg. On préleve sur chaque lot de 100 fûts un échantillon primaire d'une vingtaine de kilogrammes, à l'aide d'une sonde de 20 mm de diamètre munie d'une lumière longitudinale de 9 mm de large et de 1 m de hauteur. Cet échantillon est réduit au diviseur à riffles en six échantillons de 400 g chacun, sur lesquels on détermine le thorium par calcination à 1000°C jusqu'à un poids constant.

4.3. *Nitrate d'uranyle pur*

La solution de nitrate d'uranyle est homogénéisée durant 24 h dans un réservoir de 50 m$^3$ par une pompe de recirculation. L'uranium est déterminé gravimétriquement par précipitation et calcination du diuranate d'ammonium.

4.4. *Déchets solides homogènes*

Les stériles d'attaque et de décontamination des pieds de colonne, conditionnés en fûts préalablement tarés avec leur sac intérieur en polythène, sont échantillonnés à la sonde à leur sortie sur des rouleaux, l'opérateur étant protégé par un mur de béton de la hauteur des fûts. La sonde cylindrique utilisée a un diamètre de 30 mm et une lumière de 11 mm de large sur une hauteur de 1 m. Les prélèvements, à raison d'un par fût, sont groupés pour analyse par lot de 3500 kg environ de stériles.

a) Détermination du thorium

Après dissolution nitrique et une extraction au tributylphosphate, on détermine le thorium par volumétrie à l'éthylènediaminetétracétique [8].

b) Détermination de l'uranium

Sur la solution nitrique d'attaque des stériles, on effectue une extraction par l'acétate d'éthyle et l'on détermine l'uranium par spectrophotométrie du complexe au thiocyannate d'ammonium [9].

4.5. *Autres produits*

Ce sont essentiellement diverses solutions aqueuses ou organiques de nitrates et des suspensions de minerai, de précipités chimiques (hydroxydes, sulfate de plomb) ou de cristaux de nitrate de thorium.

a) Echantillonnage

Les solutions clarifiées aqueuses ou organiques ne posent aucun problème particulier; par contre, toutes les suspensions exigent une parfaite homogénéisation qui n'est que très rarement réalisée. En effet, les solides se classent selon leur forte densité (cas des minerais d'uranotherianite) ou par suite des caractéristiques fonctionnelles de l'appareillage (auge des filtres rotatifs, lit fluidisé du cristallisoir continu). L'échantillon, malgré
un choix extrêmement soigné des conditions de prélèvement, n'est toujours qu'une approximation plus ou moins satisfaisante.

b) Détermination de l'uranium et du thorium

Présents en faibles quantités, ils sont dosés par les méthodes d'analyse utilisées pour la détermination de ces éléments dans les déchets solides \[8, 9]\ ; lorsqu'ils se trouvent en quantités plus importantes, on utilise les mêmes modes de détermination que pour les minéraux.

5. DISCUSSION DES BILANS MATIERE

Pour chaque unité de gestion, l'établissement d'un bilan matière nécessite des quantités qui sont le résultat du produit d'un poids ou d'un volume par une teneur déterminée par l'analyse chimique d'un échantillon du produit considéré.

En pratique, si les produits purs (nitrate d'uranyle et nitrate de thorium cristallisé) peuvent être comptabilisés avec une très bonne approximation puisqu'elle atteint couramment 0,1\%, tous les autres produits - matière première entrante ou produits en cours de transformation - sont beaucoup plus difficiles à comptabiliser pour des raisons très variables: volumes morts dans des appareils fermés (broyeur, sécheur, transporteurs du type redler), suspensions en mouvement durant une filtration ou un essorage, échantillons non représentatifs parce que les produits à déterminer sont hétérogènes ou ont des teneurs constamment variables en fonction du temps.

Par exemple, la figure 3 représente les variations des concentrations de thorium des phases aqueuses et organiques des 16 mélangeurs décanteurs de la batterie de purification au tributylphosphate du nitrate d'uranyle, alimentée cependant par des solutions de composition constante.

D'autre part, dans une installation industrielle, des considérations économiques limitent très vite le nombre possible d'analyses. En particulier, dans le cas cité précédemment d'une batterie de 16 mélangeurs décanteurs, il est absolument exclu d'effectuer journellement les 64 déterminations de volume, les 64 prélèvements et les 64 déterminations analytiques qu'exigerait l'établissement quotidien d'un bilan matière rigoureux. On tourne généralement cette difficulté par l'adoption d'une valeur supposée constante de la charge de ces appareils. Mais il est évident que cette façon d'opérer entache les bilans quotidiens d'une erreur non négligeable.

Par contre, si l'on établit les bilans comptables sur des périodes plus grandes, en fait le mois et le semestre, on minimise toutes ces incertitudes si l'on prend les précautions suivantes:
- établir le bilan comptable pour l'ensemble de l'atelier en comptabilisant l'uranium et le thorium en un nombre minimal de points qui sont ceux indiqués sur la figure 4;
- éliminer aussi souvent que possible les volumes ou les poids mal définis, par exemple en arrêtant et en rinçant avant l'instant choisi pour la comptabilisation les auges des filtres, les cuves agitées, les mélangeurs, le cristalliseur à lit fluide de cristaux;
6. CONCLUSION

En comparant les résultats d'un bilan comptable mensuel ou semestriel à l'inventaire physique réel, on obtient un écart qui permet de juger de la
validité du mode de gestion adopté. Dans le cas de l'uranium, les deux résultats s'écartent d'une valeur inférieure à 1% tandis que pour le thorium cette valeur peut atteindre quelques pour cent. La même comparaison entre bilan comptable et inventaire effectué par unité de gestion permet d'ailleurs parfaitement de localiser la principale cause d'incertitude à l'évaluation des matières nucléaires contenues dans le minerai entrant, minerai dont la proportion respective des teneurs en uranium et en thorium est effectivement répartie d'une manière très hétérogène. On a pu d'ailleurs minimiser cette incertitude sur l'élément le plus intéressant, l'uranium, en stratifiant les lots de 30 à 40 t en sous-lots relativement plus homogènes en uranium.
Toutefois, en ce qui concerne le thorium, les résultats ne sont pas aussi satisfaits, comme l'a déjà mis en évidence un groupe d'étude sur l'analyse chimique des matières nucléaires [10]. Néanmoins, les progrès réalisés pour réduire systématiquement l'incidence des incertitudes sur les bilans matière ont permis, au cours des dernières années, l'exploitation de l'atelier de traitement de l'uranorthionate de l'usine du Bouchet et la mise au point d'un contrôle quasi permanent des matières nucléaires particulièrement satisfaisant en ce qui concerne l'uranium.

REFERENCES

NUCLEAR MATERIALS MANAGEMENT IN A RECOVERY FACILITY FOR UNIRRADIATED ENRICHED URANIUM

G.R. JASNY
UNION CARBIDE CORPORATION, OAK RIDGE, TENN., UNITED STATES OF AMERICA

Abstract — Résumé — Аннотация — Resumen

NUCLEAR MATERIALS MANAGEMENT IN A RECOVERY FACILITY FOR UNIRRADIATED ENRICHED URANIUM. The United States Atomic Energy Commission's Y-12 Plant, in Oak Ridge, Tennessee, has been processing and recovering various forms of unirradiated enriched uranium for over twenty years. Today, the Y-12 recovery facilities consist of a semi-continuous train of unit operations including dissolution, combustion, evaporation, extraction, denitration, and hydrofluorination. The processing and storage equipment is of restricted geometry and has a combined capacity of several hundred kilograms of enriched uranium per month. Feed to the recovery operation comes both from the Plant and from other USAEC installations in the United States. This feed has included, at one time or another, practically every type of unirradiated enriched uranium scrap. Physical control is maintained successfully by the following techniques:

1. Careful design of equipment to permit cleaning and sampling and to minimize material trapping;
2. Continuous monitoring of all discards and waste streams including sewer and stack effluents and contaminated discards;
3. Periodic physical inventories;
4. Careful sampling and analysis of all external feed streams and product streams;
5. Stringent control of the quality of analytical measurements and of sampling;
6. Assignment of the responsibility for material control to operating personnel rather than to accounting personnel;
7. Careful indoctrination of operating personnel.

Information input to the nuclear material accounting system is achieved by means of a paper flow closely paralleling the physical flow of material, i.e. batch identity is maintained by means of individual batch cards and batch disposition is signalled by transfer of the batch card to the nuclear material accounting department. Most accounting operations are performed on electronic data-processing equipment. In addition to the records and calculations required for material balances, historical records of hold-up and concentration are kept to detect anomalies in input-output or inventory measurements.

Statistical analysis is used extensively in connection with the control of measurements and to minimize the number of chemical analyses. However, the application of statistics to the evaluation of inventory discrepancies has not been successful.

This paper will discuss the details of the material control techniques which have been listed and some of the problems associated with them.

GESTION DES MATIERES NUCLEAIRES DANS UNE INSTALLATION DE RECUPERATION D'URANIUM ENRICHI NON IRRADIE. L'usine Y-12 exploitee par la Commission de l'énergie atomique à Oak Ridge, Tennessee, procède depuis plus de 20 ans au traitement et à la récupération d'uranium enrichi non irradié se présentant sous diverses formes. À l'heure actuelle, les installations de récupération d’Y-12 exécutent une série d'opérations distinctes en chaîne semi-continue, par exemple, dissolution, combustion, évaporation, extraction, dénitrification et hydrofluoruration. Les installations de traitement et de stockage ont une géométrie restreinte et leur capacité globale est de plusieurs centaines de kilogrammes d'uranium enrichi par mois. L'uranium entrant provient de l'usine elle-même ainsi que d'autres installations de la Commission de l'énergie atomique, situées aux États-Unis. Depuis sa création, l’usine Y-12 a reçu pratiquement toutes les sortes de déchets d'uranium enrichi non irradié. Le contrôle des opérations est assuré à l'aide des méthodes suivantes:

1. Utilisation du matériel conçu de manière à faciliter le nettoyage et l'échantillonnage ainsi qu'à réduire au minimum le «piégeage» de matières;
2. Contrôle permanent de tous les rebuts et déchets liquides, en particulier des effluents évacués à l'égout ou dans l'atmosphère et des déchets contaminés;
3. Inventaires périodiques des stocks;
4. Echantillonnages et analyses de toutes les matières entrantes et sortantes;
5. Contrôle strict de la qualité des mesures analytiques et de l'échantillonnage;
6. Attribution de la responsabilité du contrôle des matières au personnel chargé des opérations plutôt qu'à des comptables;
7. Formation poussée du personnel chargé des opérations.

Le transfert des informations au service de comptabilité des matières nucléaires est assuré au moyen d'un système de fiches qui accompagnent les matières: chaque lot est identifié par une fiche individuelle et la livraison du lot est signalée par transfert de la fiche au service de comptabilité des matières nucléaires. La plupart des opérations comptables sont exécutées sur des appareils électroniques de traitement de l'information; outre les dossiers et calculs nécessaires pour l'établissement des bilans matières, on enregistre régulièrement des données sur la rétention et la concentration afin de faciliter la détection des anomalies dans les bilans entrée/sortie ou dans les inventaires.

L'analyse statistique est largement utilisée pour vérifier les mesures et réduire au minimum le nombre d'analyses chimiques. Toutefois, l'application des statistiques n'a pas donné de bons résultats pour l'évaluation des erreurs d'inventaire.

Le mémoire étudie de manière détaillée les méthodes de contrôle des matières indiquées plus haut et certains des problèmes qu'elles posent.
ADMINISTRACION DE MATERIALES NUCLEARES EN UNA PLANTA DE RECUPERACION PARA URANIO ENRIQUECIDO NO IRRADIADO. Desde hace más de veinte años, la planta Y-12 de la Comisión de Energía Atómica, situada en Oak Ridge, Tennessee, viene tratando y recuperando diversas formas de uranio enriquecido no irradiado. El proceso de recuperación consiste en un tren semicontinuo de operaciones unitarias: disolución, combustión, evaporación, extracción, desnitrificación e hidrofluoración. El equipo de tratamiento y almacenamiento es de dimensiones restringidas para evitar la criticidad y tiene una capacidad combinada de varios centenares de kg de uranio enriquecido al mes.

El material sometido a las operaciones de recuperación proviene tanto de la planta como de otras instalaciones que la CEA posee en los Estados Unidos. Este material incluye prácticamente todos los tipos de residuos de uranio enriquecido sin irradiar. El control físico se ejerce satisfactoriamente mediante las técnicas siguientes:

1. Diseño cuidadoso del equipo para facilitar la limpieza y el muestreo y reducir al mínimo el atascamiento del material;
2. Control continuo de todas las corrientes de desechos y residuos, incluidos los efluentes líquidos y gaseosos, y los desechos contaminados.
3. Inventarios físicos periódicos;
4. Muestreo y análisis minuciosos de todos los materiales que entran en la planta y de todos los productos que salen de ella;
5. Control estricto de la calidad de las mediciones analíticas y del muestreo;
6. Asignación de la responsabilidad del control de los materiales al personal técnico y no a los servicios de contabilidad;
7. Adecuado adiestramiento del personal técnico.

El suministro de datos a los servicios de contabilidad se logra estableciendo una corriente de información prácticamente paralela a la corriente física del material; la identidad de cada partida se mantiene mediante una serie de fichas individuales y la salida de una partida se indica pasando la ficha correspondiente al departamento de contabilidad de material nuclear. La mayor parte de las operaciones contables se llevan a cabo con un equipo electrónico de sistematización de datos. Además de los registros y de los cálculos necesarios para los balances de material, se llevan registros de carga y concentración que facilitan la detección de anomalías en la relación entrada-salida o en la evaluación de las existencias.

El análisis estadístico se emplea en gran medida para el control de las mediciones y para reducir al mínimo el número de los análisis químicos. De todas formas, la aplicación de la estadística a la evaluación de las discrepancias de inventario no ha dado buenos resultados.

En la memoria se discuten todos estos detalles de la técnica de control de materiales y algunos de los problemas que plantea la aplicación de esta técnica.

INTRODUCTION

In recent years, a new term has been incorporated into the language of quality control. This term is "total quality control". It implies a quality control programme that, instead of merely monitoring the quality of a finished product, reaches far back into plant design and the manufacturing process to control and optimize the conditions under which the product is made. By analogy, "total nuclear material management" should denote a programme concerned with as many as possible of the variables that can have an impact on the desired end result that is a satisfactory material balance. This paper will discuss several aspects of a "total" nuclear material control programme developed in a large US Atomic Energy Commission facility for the reprocessing of unirradiated, highly enriched uranium scrap.

The facility in question is located at the Oak Ridge Y-12 Plant, one of three Oak Ridge facilities operated by the Union Carbide Corporation for the US Atomic Energy Commission. The other two facilities are the Oak Ridge National Laboratory and the Oak Ridge Gaseous Diffusion Plant. The re-
covery plant is part of a vast, enriched uranium processing facility equipped to convert UF₆ to uranium metal and fabricate this metal into various components. For many years the Y-12 Plant has served as the recovery site for all highly enriched uranium salvage generated by other Atomic Energy Commission installations. This function has now, to a great extent, been absorbed by private processors, but over a period of years nearly every conceivable form of enriched uranium scrap has been processed at Y-12, including the major alloys and compounds, combustibles, solutions and miscellaneous solids [1].

The recovery plant has the dubious honour of being the oldest plant of its type in existence since Y-12 produced the first significant quantities of enriched uranium ever made. Lest this seniority should convey the impression of obsolescence, it must be pointed out that the plant is now in its third design generation. The first generation was essentially a large laboratory in which the most frequently encountered piece of processing equipment was a four-litre beaker. The second generation was a combination of laboratory equipment, large (200 gal (US)) dissolvers, and continuous liquid extraction systems. The present, third generation, plant is composed of inherently safe-geometry dissolvers, evaporators, extraction columns, storage tanks, denitrators and fluid beds all permanently interconnected. Within the plant, only the burning and dry crushing equipment has been spared the dictum of restricted geometry.

Since material control itself can have a number of different connotations, a further delineation of the area of concern is necessary. As used here, "material control" is understood to be limited to the steps taken to minimize the unintentional loss or diversion of enriched uranium and to provide, efficiently, the necessary ingredients of a good material balance. Other problems such as minimization of inventory, nuclear safety, contamination control, and prevention of theft, though they are important, will not be considered.

There are at least three sine qua non principles of nuclear materials control: (1) painstaking attention to the physical measurement of all inputs, outputs and inventories; (2) good control of the quality of all measurements; and (3) maintenance of a good record system. For years, these principles have been faithfully adhered to at Y-12. Nevertheless, a number of material balance crises have occurred over the years, each characterized by the nagging question, "Is this an indication of real loss?" Despite the best assurance of the statisticians that fluctuations were in control, operating management and Atomic Energy Commission officials have remained unimpressed, and the usual outcome of such crises has been a major clean-up of the plant, frequently followed by a significant gain of material. This experience has led to the conclusion that if one is to have total nuclear material control, three additional principles must be applied: (1) good facilities design; (2) thorough monitoring of discards and losses; and (3) continued training and motivation of personnel.
FACILITIES DESIGN

If the existing body of US published articles on nuclear materials management is used as a measure of interest, one is forced to the conclusion that recognition of the importance of good facilities design to successful material control has been slow in coming. Considerable emphasis has been placed on design as related to nuclear safety and process efficiency, but very little on design for material control. This situation is somewhat ironic because the very changes that have received emphasis as a result of these concerns, i.e. the use of long slender shapes and of continuous or semi-continuous processing equipment, have made the material control problem more difficult. Some results have been that volume measurements and samples are harder to obtain, and piping systems have grown in complexity with the attendant likelihood of material becoming trapped and unaccounted for. The more advanced the processing system, the stronger the temptation to cover breakdown eventualities and to provide flexibility by the addition of such extra components as valves, bypasses and overflow collectors. Nevertheless, good material control argues strongly for the elegance of simplicity.

Some general features found helpful at Y-12 are: the use of air lifts for material transfer and recirculation \[2\], full-flow ball valves, and, whenever possible, short, straight runs of pipe with as few flanges or screwed joints as possible.

The importance of good design to material control can, perhaps, be illustrated by a specific example — that of solution storage tanks.

We shall first consider the safe tank. A common feature of inherently safe nuclear design is the so-called safe tank — a long pipe 4-6 in. diam., 10-50 ft long, and frequently situated in horizontal arrays in which the tanks are on 2-ft centres. It is common practice to position such tanks with a slope of 0.1 in/ft. A recirculation system is generally provided to homogenize the contents of these tanks.

Some months ago, at the request of the Atomic Energy Commission Division of Nuclear Material Management, our plant initiated an experimental programme to study the problem of mixing the contents of safe tanks. Preliminary results have shown that mixing time, \( T \), is a function of the recirculating flow rate, \( Q \), according to an equation of the form \( \ln T = A - B \ln Q \), and that fairly high recirculation rates (of the order of at least 10 gal/min for a 6-in. tank, 20 ft long) are required to achieve good mixing in less than 1 h. If these results are confirmed, our own recirculation systems will probably be undersized and will have to be corrected or else much longer mixing times will have to be used. Another finding of this experimental programme is that mixing in banks of parallel tanks is enhanced by maintaining unequal flows to the individual tanks, a fact that had escaped us and for which no provision had been made.

There is another, potentially more serious, problem associated with slanted, horizontal safe tanks — that of volume measurement. The calibration curve for such tanks is in the shape of an elongated S on its side, thus \( \). This means that small increments of height in the sightglass can represent significant volumes unless the tank is almost empty or almost full. To this
uncertainty must be added the possibility of slight deviations from straight-
ness. The combination of all these factors means that the measurement of
significant quantities of enriched uranium solution in arrays of slanted
horizontal safe tanks is one of the most probable sources of inventory errors.
One obvious design change, which we have made when possible, has been
substitution of vertical arrays that eliminate volume uncertainties for practi-
cal purposes. But vertical tanks present problems of headroom that, ideally,
should be considered at the earliest plant design stage.

An alternative to the restricted-geometry tank, which is growing in
favour because it is cheaper, is the conventional large tank packed with
Pyrex Raschig rings containing boron. However, one of the drawbacks of
this storage method is its material-control limitations. Loosely packed
Pyrex rings tend to break and erode away, causing changes in the free vo-
lume of the tank; worse, the rings provide a large surface area for the
deposition of uranium compounds. The determination of the amount of
uranium left in the packed tank after the tank has been drained can be very
troublesome and so can the removal of the adsorbed material. It is clear
that the extent to which the engineering problems of ring breakage and ad-
sorption on the rings are solved will greatly affect the material-control pro-
erties of these tanks.

PHYSICAL MEASUREMENTS OF INPUTS, OUTPUTS, AND INVENTORIES

It can be argued that inventory errors, even though they cause much
trouble, have a way of correcting themselves in the long run, and that the
real essence of good material control is to correctly measure input and pro-
duct and to minimize and measure losses and discards, even if this means
reducing the emphasis on inventory. Three elements are required for such
a programme: (1) measurement of input and product, (2) measurement and
control of losses and discards, and (3) assurance of the quality of these
measurements.

Measurement of input and product

The measurement of product, involving almost pure and homogeneous
materials, presents relatively few sampling and analytical problems. On
the other hand, the measurement of the input to the reprocessing facility
is one of the fundamental difficulties of the scrap reprocessing business.

Since the Y-12 reprocessing plant receives materials from other parts of the
plant as well as from other installations, a double standard is maintained.
Locally generated scrap known to contain significant quantities of uranium
is carefully measured as it is transferred. For example, crucible oxide
generated during the casting of metal is dissolved and the resulting solution
is sampled and analysed to establish accountability. On the other hand,
materials known to contain little uranium on the average, i.e. mop water
and combustibles, are transferred to the recovery operation on the basis
of estimates that are historic values, periodically rechecked. This procedure
permits drawing approximate balances around major production areas. The
recovery area balance reflects the composite uncertainty of all the other balance areas and therefore tends to fluctuate more than they do.

When it comes to receipts from other installations, this relatively complacent attitude is totally absent. Generally, physical handling of outside shipments is in line with the procedures recommended in an excellent manual prepared by the US Atomic Industrial Forum [3]. Specific Y-12 procedures have been described by Barkman [4]. Briefly, the major precautions used to maintain good accountability on outside shipments include: segregation of individual shipments, weight verification, dissolution of alloys and impure solids before sampling, homogenization of solids and solutions, filtration of solutions, sampling of all parts of a shipment, and use of duplicate samples one of which is retained until agreement has been established with the shipper. Statistical analysis is routinely employed in evaluating the significance of apparent shipper-receiver discrepancies. The techniques used have been described by Smith [5].

Recently, over a period of several years, Y-12 recovered only 0.07% less than the shippers' estimates (after reconciliation of significant differences) in processing many thousand kilograms of enriched uranium contained in many hundreds of scrap shipments. Even allowing for the obvious advantages of the receiver over the shipper, and for the averaging effect of many shipments, this record is good evidence that close adherence to the basic plant precepts will insure a minimum of shipper-receiver discrepancies.

Measurement of inventory

Despite the relative shift in emphasis from the measurement of inventory to the measurement of inputs and outputs, the inventory process still commands a major part of the material-control effort. Because of the large amount of enriched uranium on hand, inventories are still taken on a monthly basis. However, it is possible that this frequency will be reduced because of our good experience in recent years.

In preparing for the inventory, an attempt is made to reduce to a minimum the number of individual batches of material that may range from 1000 to 2000. When possible, hold-up in mixing equipment such as safe tanks is adjusted to permit a precise volume measurement. Finally, processing during the month is controlled to achieve a composition of inventory that will permit good chemical and isotopic analysis of the bulk of the material on hand at the end of the month. For the past two years, an average of 95% of the enriched uranium on inventory in the recovery plant was measured at inventory time or was carried over on the basis of a previous measurement. Only 5% of the inventory was based on estimates.

Taking the inventory does not start until after all transfers into or out of the recovery area have been completed. Portable items are tagged and listed. Fixed-equipment readings are entered on preprinted sheets that must be filled out for every piece of equipment in the plant. Procedures for homogenizing materials and sampling are carefully detailed.

After completing the inventory, the area is thoroughly surveyed by a supervisory team that checks the tagging of individual batches, spot-checks sightglass readings and monitors empty equipment. This work is done with
the help of commercially available, portable, gamma-sensitive instruments\(^1\). Our experience has been that, in general, equipment that gives a reading of no more than 1 to 2 mR/h over background contains only insignificant amounts of uranium. Not only do these meter checks uncover trapped material that can usually be removed by flushing, but they also reduce the frequency of equipment and piping disassembly that previously was a routine practice in connection with inventory.

The "unaccounted-for" experience of the Y-12 recovery facility over 22 months, ending 3 months ago, included 10 "gains" and 12 "losses". These fluctuations ranged from a maximum gain of 1.6% of the ending inventory to a maximum loss of 1.7%. The absolute magnitude of the variations may be somewhat exaggerated because, as was pointed out previously, the recovery-area balance reflects the collective uncertainties of the transfers from the other enriched uranium processing areas at Y-12. Nevertheless, as would be expected in a condition of over-all control, the frequency of "gains" matches the frequency of "losses" very closely. The cumulative "unaccounted-for amount" from the beginning to the end of that period for the entire enriched uranium plant was only -0.037% of the inventory.

MEASUREMENT OF LOSSES AND DISCARDS

The possibility of accidental loss — or worse, undetected accidental loss — of enriched uranium hangs like a sword of Damocles over the heads of all who work with large quantities of this extremely valuable material. This possibility has been a matter of continued concern at Y-12, and a few years ago a comprehensive study of loss control and detection, which included a review of the handling of discards, was initiated. A special task force of engineers spent several months measuring track-out, testing stack samplers, sampling sewers and measuring contaminated metal. The improved monitoring network that resulted from their findings is, in our judgment, adequate to prevent large, accidental losses and minimize undetected losses. This network covers stacks, storm sewers, liquid waste streams and solid discards. Track-out losses, which proved to be relatively small, are not routinely measured but are included in our estimates of loss.

Liquid waste streams

Small, miscellaneous liquid batches are individually sampled before discard. In addition, two continuous liquid waste streams from the recovery plant must be checked: extraction raffinates and evaporator condensates. Discard limits for these streams are 2 ppm of enriched uranium. Condensates flow through a gamma monitor that automatically activates a bypass valve to retaining tanks whenever the gamma count corresponds to a uranium concentration greater than 2 ppm. The flow of the condensate that is discarded is metered and a composite sample of the stream is automatically taken. Raffinates, which do not readily lend themselves to gamma counting

\(^{1}\) Model FS-11A, Technical Associates, Burbank, Calif.
because they contain small quantities of fission products, are stored and analysed in intermediate safe tankage before discard. A rapid, fluorescent analysis method is used to minimize intermediate storage requirements. To detect leaks in coolers, the recirculating cooling tower stream is also continously sampled and gamma monitored.

**Solid wastes**

Every container of leached processing residue is, of course, sampled before discard. This, however, is not practical for contaminated metal items such as broken valves, old piping and worn-out pieces of equipment. To check this sort of material, a shielded gamma-counting facility has been developed. This facility consists of a shielded enclosure large enough to accommodate a 55-gal drum, a turntable, and a detection unit consisting of four 4 1/2-in.-diam. sodium iodide crystals, each with its own electronics and necessary sequence controls. Barrels containing the metal to be monitored are placed on a conveyor. Operation of the controls causes the barrel to enter into the shielded enclosure, rotate 360° in front of the gamma scintillation detectors, and emerge from the shield. Count-rate versus barrel rotation is displayed on a strip-chart recorder. An alarm sounds if the count-rate exceeds a present value corresponding to 10 g U/1000 lb. Active material may be approximately located in the barrel by noting the angular position of the peak count-rate on the chart trace, and its height above the bottom of the barrel may be found by selecting one of the four detectors that gives the highest output as indicated by the recorder trace.

**Storm sewers**

Storm sewers leaving the enriched uranium area are monitored to detect accidental spills. A typical storm-sewer sampling station consists of a 90° V-notch weir, flow sensing and recording instruments, a proportional sampling pump and a small evaporator. The flow sensing and recording instruments are equipped to provide a pneumatic signal to the pump controller so that a sample is taken that is always proportional to the flow. The evaporator is equipped with level controls for automatic and continuous operation. The product is removed when a concentration sufficiently high for analysis and subsequent sewer-loss evaluation has been attained, usually at the end of a week.

**Stacks**

All stacks removing air from the enriched uranium processing areas are equipped with continuous air samplers. In addition, vents from critical process equipment are provided with appropriate monitors and alarms. For example, the exhaust piping from the fluid beds in which UO₃ is converted to UF₄ is checked by a gamma monitor located immediately downstream from the filters and set to alarm at a count corresponding to 5 ppm ²³⁵U in the gas stream. Much of the sampling is accomplished through the use of traverse-type probes inserted through the stack wall. A sample of the exhaust air is pulled out of the stack stream through the probe and through
a sampling filter paper by vacuum. The filter paper is analysed periodically. The traverse-type probe itself consists of a tube mounted completely across the diameter of the stack with openings pointing upstream. The openings are spaced and sized to collect a sample across the stack cross-section. Parenthetically, in tests conducted a few years ago, the traverse-type probe was found to be capable of representative sampling of stack particulates, but only about 25% of the sample entering the probe reached the filter paper [6].

This loss and discard monitoring programme tells us that, over the past few years, total discards have averaged 0.05% of the plant throughput — throughput being defined as the sum of inventory and transfers out. Of this, only 0.02% was accounted for by stack losses, track-out and the like. More importantly, this programme gives us strong assurance that we are not suffering undetected losses. This assurance has resulted in minimizing emotional responses to the unavoidable fluctuations in the monthly material balances.

CONTROL OF QUALITY OF MEASUREMENTS

Measurements quality relies primarily on the use of chemical and isotopic control samples, the calibration and certification of weighing equipment, and the application of statistical techniques to the solution of sampling problems.

Control samples

Samples are submitted to the laboratory at regular intervals from each of a number of batches set aside for the control of laboratory precision and bias. These batches cover a wide range of material types and concentrations, every major source of routine samples being represented. The samples are submitted by production personnel, and, to laboratory personnel, are indistinguishable from the routine samples. Punched-card equipment is used in the identification of control-batch analyses and in statistical treatment of the reported results [7]. Personnel in the Quality Control Department prepare laboratory standards, maintain the control batches, plot control charts, and use standard statistical techniques to estimate bias and precision of laboratory measurement methods. If the results indicate an out-of-control condition, the laboratory is immediately notified. In addition, quarterly measurement quality reports are issued.

Certification of weighing equipment

About 30 sets of scales and balances are used for accountability weighings. These instruments range from 200-g analytical balances to 200-kg scales. This equipment is regularly checked and certified. Certification intervals vary between 4 and 13 weeks, depending upon the type of equipment and the amount of use. The equipment is found to be in satisfactory condition more than 90% of the time. Minor adjustments are made
about 10% of the time. No major equipment failures have been detected during the routine checks.

*Calibration of weight standards*

All weight standards are calibrated at 26-week intervals with reference to the plant primary standards. The primary standards are Class M weights calibrated by the US National Bureau of Standards.

*Sampling*

Control of the plant's sampling programme is exercised through statistically designed experiments rather than continuous monitoring. Sampling techniques are carefully tested before adoption, and thereafter whenever a process change indicates sampling re-evaluation to be desirable. These checks indicate that, as a rule, sampling of solutions is very reliable but that obtaining representative samples from the heterogeneous powder frequently encountered in uranium recovery can be a very challenging task.

**ACCOUNTING RECORDS SYSTEM**

Y-12 uranium accounting techniques have been described in detail by Bass [8]. They follow good, conventional accounting practices to achieve two basic functions: (1) recording the movement of material into and out of the material-balance area permitting a double-entry system of accounting, and (2) accounting for each discrete, identifiable batch of material in the material balance area to assure that each item has been correctly disposed to the system during the accounting period or recorded on the inventory.

Whenever material is inventoried or flows into or out of the recovery process, the action is paralleled by a batch card that can be one of two types: cards that identify materials to be recovered as to type and source prior to processing to a measurable state, and cards that identify and report information for measured materials. The batch cards are of standard format, consecutively numbered within categories, and, in several instances, are colour coded. The uranium accounting data system is centred on an IBM 1401 computer. The major activities of the system include the work necessary for the preparation of the following reports:

1. Plant inventory, transfer and balance reports.
2. Control reports that compare the input and output quantities of material on a batch or container basis with or without associated weights.
3. Production throughput and cost reports.

Data reported for the recovery process include: batch identification, net weight or volume, sample identification number, disposition data, cost category and units, cost accounting and a chemical form code. The chemical and isotopic enrichment analyses reported by the plant laboratory are associated with the batch and inventory data by means of the sample identification number, and the quantities of contained uranium are calculated. The data are summarized to provide listings of all transfers by shipper and/or receiver and the total inventory values.
By using the originating cost account, cost category, units of material transferred and standard cost of recovery values, production and cost-of-recovery reports are issued each month by the second working day.

The accuracy of the receipts, transfers and inventory listings is verified by preparing control reports that determine on a batch basis that all items on beginning inventory, received, or produced were either inventoried, transferred or consumed. This data system also checks the accuracy of inventory data by determining that inventory items are listed with the same weights and sample identification numbers as those used on beginning inventory, or on produced or receipt records.

In addition to this formal accounting programme, trend records of various types are maintained by the Nuclear Materials Accounting Department. These records include such items as: hold-up in various pieces of equipment, uranium concentrations at certain stages in the process as well as for certain streams, and inventory to throughput ratios. This information, used in conjunction with area balances, has been of considerable value in evaluating material balance discrepancies and in correcting errors.

**TRAINING AND MOTIVATION OF PERSONNEL**

Because it is not in the nature of man to treat a barrel of waste as though it were worth its weight in gold (which it often is when one deals with enriched uranium), training and motivation of the plant workers are particularly important to good material control in the recovery plant. Training operating personnel in sampling techniques, weighing and records keeping has proved particularly worthwhile as has the maintenance of explicit and up-to-date procedures for these activities. As a matter of basic policy, the responsibility for good material control in the recovery plant is unequivocally vested in the operating supervision. In retrospect, this policy has probably been our most effective motivating technique. The operating supervision's responsibility includes co-ordination of all material-control activities. A very useful tool in achieving good co-ordination is the inventory committee. This group has as its chairman the head of the recovery plant and is composed of representatives of the Material Accounting Department, the Laboratory Department, and other departments that process enriched uranium. The committee plans each inventory and reviews the subsequent balances, investigates significant discrepancies, and recommends procedural changes. Besides providing a good, routine communications channel between key groups the committee has resulted in a much better understanding of the problems faced by each of these groups and has done much to reduce friction between them.

**COST OF MATERIAL CONTROL PROGRAMME**

Good material control in enriched uranium recovery is not cheap, except when viewed against the consequences of bad material control. It is estimated that currently, material control costs at Y-12, exclusive of capital costs, amount to approximately 7% of the total recovery cost. This total
includes the cost of time lost during the inventory, sampling and analysis of samples used only for accountability, record keeping and data processing. A large number of samples are used for both process control and accountability. In fact, only 20% of all samples are taken solely for accountability.

CONCLUSION

In conclusion, I would like to make two points: The first is that nuclear materials control, which started out as the province of accountants, statisticians and analytical chemists, needs and deserves growing attention from engineers and operating management. The second is that no one element of this programme for total material control can be significantly downgraded without downgrading the entire programme and inviting continuing difficulty. Nevertheless, there can be varying degrees of emphasis. It is axiomatic that a prevented loss is far more desirable than the most erudite diagnosis of a loss that has occurred. Therefore, physical control of the material should be given a high priority in the allocation of nuclear material management efforts. This approach has met with very good success at the Y-12 recovery plant.

REFERENCES


DISCUSSION

J. SORNEIN: If I understand you correctly, you make the point that one only obtains a really clear picture of the materials balance in a plant of this sort when all the installations are cleaned out and all the recycling operations are completed at the end of a period of continuous operation. We fully agree. But how often do you think that these clean-outs should take place? Should one deliberately break down operational periods into smaller units so as to increase the effectiveness of the control?

G. R. JASNY: A double standard should be employed. All equipment used for processing an individual external shipment should be completely cleaned out before processing of another shipment begins. On the other hand, when it comes to the main processing train of the plant, the frequency of clean-outs should be a function of the degree of control achieved. If the control appears to be very effective, clean-outs should be kept to a minimum. They
are expensive, present contamination problems and increase the likelihood of a nuclear accident.

G. WELCH: Does the very small difference in the shipper-receiver discrepancies quoted in this paper represent the average of a very large number of transactions over a long period, or is it typical of individual differences?

G. R. JASNY: The figure quoted is the average result of many shipments and is not typical of individual shipper-receiver differences. Admittedly it is a ridiculously small number. It is significant inasmuch as it shows that measurement, quality control and records-keeping techniques do exist which, if used by both shipper and receiver, can minimize shipper-receiver discrepancies.

G. WELCH: Bearing in mind that all the enriched uranium used in the United States originally comes from Oak Ridge, do you think the smallness of this difference is possibly due to some "hidden" feature in American fuel cycles which might lead to positive discrepancies from one establishment being offset by negative discrepancies from another?

G. R. JASNY: I do not think so. I cannot quote exact numbers, but the recovery cycle constitutes only a small part of the total inventory and throughput. Furthermore, any analytical errors would cancel out since both shipments and receipts would be subject to them. Finally, there is no reason why Oak Ridge should try to hide shipper-receiver discrepancies.

F. TINGEY: I am intrigued by this figure of 0.07%. I cannot determine whether you are referring to your own internal material balance or whether this figure really relates to the shipper-receiver difference.

G. R. JASNY: This value, which represents a long-term average, is based on corrected transfer documents, the corrections being indicated and agreed upon by both parties. The significant point, I think, is that this agreement, even though it is based on occasionally corrected values, could not possibly be as good as this or even of the same order of magnitude unless our measurements and records were basically sound.

D. E. GEORGE: What were the upper and lower limits of these shipper-receiver discrepancies in the case of individual transfers?

G. R. JASNY: Individual differences have ranged from zero to several hundred per cent.

D. JANISCH (Chairman): This overall "loss" figure - 0.07% - relates to a recent period of several years. I am sure that countries which do not have processing plants at the moment would be interested in hearing about your experience in earlier years. Can you give some indication of this?

G. R. JASNY: The position with regard to shipper-receiver discrepancies and inventory was much worse in the past. As regards inventory, we had long periods of continued "loss" in the past but our current experience is that "losses" and "gains" balance each other out quite well in frequency. This, I think, is an indication of good control.

G. WELCH: Is all plant equipment in Y-12 completely washed out before each stock-taking or only for special enquiries?

G. R. JASNY: This is not done regularly and there has been no special investigation for the last two years. Tanks and equipment are cleaned periodically as individual items but at inventory time we rely on gamma
counters to survey equipment which is supposedly empty. Readings of 0.2 MR/h or less correspond to insignificant uranium holdups.

F. RONTEIX: You say tanks and equipment are cleaned out "periodically". Is this done (a) whenever a new shipper is involved, and (b) whenever there is a change in the rate of enrichment?

G. R. JASNY: The answer is "yes" in both cases. However, as soon as accountability has been established on a shipment, that shipment loses its individual identity and is added to the main processing stream.

F. RONTEIX: The USAEC only accepts scrap after it has been re-transformed into U₃O₈ or UF₆. When you speak of discrepancies in weight, are you referring to the scrap in its initial state, e.g. in the form of pieces of U-Al alloy plates, or to scrap that has been transformed into an analysable state?

G. R. JASNY: Accountability is established at the earliest reliable sampling point. In the case of alloys, this would be after the dissolution stage, when the solution and the residue are both sampled. With combustible fuels, it would be after burning.
Abstract — Résumé — Аннотация — Resumen

THE SYSTEM FOR CONTROLLING SOURCE AND SPECIAL NUCLEAR MATERIAL AT THE EUROCHEMIC REPROCESSING PLANT

W. FRENZEL AND W. SCHÜLLER
EUROCHEMIC, MOL, BELGIUM

The system for controlling source and special nuclear material at the Eurochemic reprocessing plant. Eurochemic is constructing a reprocessing plant near Mol (Belgium). The main characteristics of the plant and the different process steps are summarized. To maintain proper control of source and special nuclear material, the facilities of the company are divided into material balance areas. All transfers into and out of these areas are determined. Depending on their significance for the overall material balance, different precision requirements have been established according to a scale of significance which is briefly discussed.

To adjust the book balance, physical inventories are periodically performed. The corresponding inventory procedures are outlined. The books used for nuclear materials accounting and the principles of the accounting system are summarized.

SYSTEME DE CONTROLE DES MATIERES NUCLEAIRES BRUTES ET SPECIALES A L'USINE DE TRAITEMENT D'EUROCHEMIC. La société Eurochemic fait actuellement construire une usine de traitement chimique près de Mol (Belgique). L'auteur indique succinctement les principales caractéristiques de l'usine et les différentes étapes du traitement. Pour assurer un contrôle correct des matières nucléaires brutes et spéciales, les installations d'Eurochemic sont divisées en secteurs de mesures.

Tous les transferts entre ces secteurs de mesure sont déterminés quantitativement. Les mesures sont faites avec des degrés de précision différents qui ont été fixés en fonction de leur importance pour le bilan matières général; les auteurs examinent brièvement les critères appliqués à cette fin.

Pour vérifier le bilan, on procède périodiquement à des inventaires matériels. La façon dont sont menés ces inventaires est exposée dans ses grandes lignes. Les auteurs indiquent les livres utilisés pour la comptabilité des matières nucléaires et résument les principes du système comptable.

СИСТЕМА КОНТРОЛЯ НАД ИСХОДНЫМ И СПЕЦИАЛЬНЫМ ЯДЕРНЫМ МАТЕРИАЛОМ НА ЗАВОДЕ ЕВРОХИМИЧЕСКОЙ КОМПАНИИ ПО РЕГЕНЕРАЦИИ ЯДЕРНОГО ТОПЛИВА. Компания Еврохимик в настоящее время строит завод по переработке ядерного топлива около Моля (Бельгия). Суммируются главные характеристики завода и различных стадий обработки. Для проведения надлежащего контроля над исходным и специальным ядерным материалом установки компании разделены на зоны материального баланса.

Определяются все перемещения в эти зоны и из них. В зависимости от важности этих материалов для общего материального баланса установлены различные требования в отношении точности в соответствии со шкалой важности, которая кратко описывается.

Для правильного составления журнального баланса периодически проводятся физические инвентаризации. Иллюстрируются соответствующие методы инвентаризации. Дается краткое описание журналов учета ядерных материалов и принципов системы учета.

CONTROL DE MATERIALES NUCLEARES BASICOS Y DE MATERIALES FISIONABLES ESPECIALES EN LA PLANTA DE REGENERACION DE LA EUROCHEMIC. La compañía Eurochemic está construyendo una planta de regeneración en Mol (Bélgica). Los autores resumen las principales características de la instalación, y las distintas fases del proceso industrial. Para mantener un control adecuado de los materiales nucleares básicos y de los materiales fisionables especiales, las instalaciones de la compañía estarán divididas en diversas zonas de balance de materiales.

Toda entrada y salida de materiales de estas zonas quedará registrada. Teniendo en cuenta su importancia en relación con el balance global de materiales, se han fijado distintos requisitos en materia de precisión basados en una escala de valores que los autores describen brevemente.
Periódicamente se efectuarán inventarios de material a fin de proceder al balance de asientos. Se exponen los correspondientes procedimientos de inventario. Los autores describen los libros utilizados en la contabilidad de los materiales nucleares y enuncian los principios en que se basa el sistema de contabilidad.

The Eurochemic Company is a joint undertaking of the 13 member countries of the European Nuclear Energy Agency of the Organisation for Economic Co-operation and Development. It was founded in 1959 to construct and operate a nuclear fuel reprocessing plant at Mol, Belgium. The main plant is now at the stage of testing the process installation, and the Research Laboratory and the Fuel Storage Building are already in operation.

1. THE EUROCHEMIC PLANT

1.1. Characteristics of the plant

The plant is constructed for aqueous reprocessing of fuels used in the different European research and power reactors. Consequently the processing programme covers a large variety of fuel elements, which differ in size, shape, chemical composition, cladding material, fuel enrichment, burn-up and plutonium content. Pre-irradiation fuel enrichment covers the range between natural uranium and highly enriched uranium of 93\% \textsuperscript{235}U (MTR-type fuel).

From the point of view of the accounting of source and special nuclear material, it should be mentioned that many fuels are either sold or leased to the reactor operators under different bilateral or multilateral agreements.

The Eurochemic installations include the following facilities: fuel storage, chemical processing plant, analytical laboratory, research laboratory, waste treatment and storage, and product storage.

The reprocessing plant is designed for a nominal daily throughput of either 350 kg of uranium of less than 1.6\% enrichment, or 250 kg of uranium of enrichments between 1.6 and 5\%, or 5 kg of highly enriched uranium (as MTR fuel elements).

The size of some fuel batches in the schedule is quite small and their separate treatment does not appear technically and economically feasible. Therefore, it is foreseen that similar batches will be combined for a single solvent extraction run, after they have been dissolved separately and input analyses of source and special material have been carried out.

In addition, the depletion of fissile material during irradiation precludes in most cases the direct re-use of the purified product by the reactor operator who has shipped the fuel for reprocessing. Therefore, a certain amount of product blending and redistribution appear to be at least technically justified, if not necessary. These points stress the particular emphasis that must be placed on careful accounting of nuclear materials.
1.2. Characteristics of the process

1. Fuel elements are chemically declad and dissolved in one of three dissolvers, designed to treat the three classes of fuel enrichments previously mentioned.
2. The installations for first cycle feed preparation include a centrifuge for clarification of fuel and decladding solutions.
3. Separation of uranium, plutonium and fission products is carried out by a Purex-type flowsheet with subsequent final purification of the uranium on a silica-gel bed and of plutonium by oxalate precipitation.
4. Air-pulsed sieve plate columns are used for first cycle decontamination and partitioning, and for second cycle uranium purification. Mixer-settler batteries of the pump-mix type are used for second cycle plutonium purification.
5. The auxiliary systems include units for solvent recovery, nitric acid recovery and re-working of off-standard process solutions.
6. The standard chemical forms of the final products are a uranyl nitrate solution in nitric acid and plutonium dioxide powder. Plutonium can also be recovered as a nitrate solution if desired.
7. Uranium product solutions of up to 1% enrichment are stored in a conventional tank farm and shipped in tank trucks.
   The product of higher enrichment is stored in safe tanks for 5% and 93\% \textsuperscript{235}U, and is put into safe bottles for shipment.
   Plutonium oxide is packed into sealed cans and shipped in Class I containers (IAEA).
8. The first-cycle raffinate, containing most of the fission products, is concentrated and stored on site as high level waste.
   Low level wastes are either transferred to a waste treatment plant of the nearby Belgian nuclear centre, or are concentrated before storage in a medium level waste tank farm, which also receives the various de-cladding solutions.
9. Gaseous effluents are purified in an off-gas treatment system, mixed with the ventilation air from the building and discharged via the main stack.

2. CONTROL METHODS FOR SOURCE AND SPECIAL NUCLEAR MATERIALS

The primary purpose of the accounting system at Eurochemic is the control of shipments of source and special material to and from the site, as well as its distribution within the installations of the company.

These tasks are performed in the following manner:
1. The installations in which fissile material may be handled are divided into a number of material balance areas (MBA).
2. The quantities of fissile material transferred into or out of each MBA are determined and accounted.
3. All accounts are balanced monthly.
4. Physical inventories of fissile material are taken periodically to adjust the material balances.
2.1. The system of material balance areas (MBA)

According to operational and accounting criteria, the buildings concerned have been divided into six MBAs covering the following sections: fuel reception, storage and dissolution; chemical processing; analytical laboratory; research laboratory; effluent treatment and waste storage; and product storage.

The combination of the fuel storage area and the dissolvers into one MBA was necessary because accounting operations in this part of the plant are limited to counting fuel elements and recording the quantities of source and special material based on data calculated by the reactor operator.

The dissolvers, however, have been separated from the storage area by sub-division of the first MBA into two sub-areas. This facilitates a separate numerical inventory of the fuel storage and allows a comparison to be made between the calculated source and special material content of the fuel elements charged to the dissolver, and the determination of the corresponding in-put process.

A similar sub-division was considered practical for the chemical processing area, which covers a relatively complex installation. Three sub-areas have been established to cover feed preparation, solvent extraction and plutonium purification respectively.

2.2. Transfers of source and special material

Transfers of source and special material are considered either as external or internal. External transfers include reception of irradiated fuel in the fuel storage area, shipment of uranium or plutonium product from the product storage area, and in exceptional cases, delivery of inactive uranyl nitrate solution for plant start-up to the reagent make-up section or to the product storage, and movement of relatively small amounts of research material of various forms and compositions, to or from the research laboratory.

Internal transfers are those between two different material balance areas or sub-areas.

A comparison of the various internal transfers shows that they are of very different significance in establishing the material balance.

Therefore, all routine transfers have been classified according to a scale of significance, which expresses the daily quantity of source and special material crossing each transfer point in units of the daily plant throughput.

Three main groups can then be distinguished:

1. Transfer significance about 1: including feed input, bulk transfers between different plant sections or product output.
2. Transfer significance \(10^{-2}\) to \(10^{-3}\): e.g. rinse solutions.
3. Transfer significance of less than \(10^{-4}\): e.g. waste solutions.

Transfers of the third group have virtually no effect on the totals in the material balance, although they may be of interest for assessing process performance.
One of the most important applications of this classification is the corresponding adjustment of precision requirements for both the volume measurement and analytical determination of sample composition.

2.3. Physical inventories

The large number of process tanks, equipment and auxiliary vessels in the plant to be covered by a physical inventory requires many determinations of source and special material at one time. In addition, these determinations must be performed in a relatively short period for a physical inventory to be of any value, and therefore represent a substantial peak load on the analytical service.

A detailed examination of inventory points reveals that it is possible to perform a complete inventory with reasonable precision without making both the volume measurement and the sample analyses for each tank involved. The various tanks, therefore, have been grouped into the inventory categories summarized in Table I.

<table>
<thead>
<tr>
<th>TABLE I</th>
<th>INVENTORY CATEGORIES</th>
</tr>
</thead>
<tbody>
<tr>
<td>VC</td>
<td>Volume and Composition determined from level measurement and sample analysis</td>
</tr>
<tr>
<td>V</td>
<td>Volume determined by level measurement, composition derived from analysis of up- or down-stream sample</td>
</tr>
<tr>
<td>C</td>
<td>Composition determined by sample analysis; volume = standard value</td>
</tr>
<tr>
<td>D</td>
<td>Composition derived from analysis of up- or down-stream sample; volume = standard value</td>
</tr>
<tr>
<td>F</td>
<td>Flowsheet data used for both equipment volume and composition</td>
</tr>
<tr>
<td>B</td>
<td>Book balance used (applied for dissolvers only)</td>
</tr>
<tr>
<td>T</td>
<td>Transfer of solution to other vessel</td>
</tr>
<tr>
<td>O</td>
<td>No inventory made</td>
</tr>
</tbody>
</table>

3. BOOK-KEEPING

All transfer and inventory data are reported in writing to the Accountability Office, where they are used for calculation of the source and special material quantities.

The results are entered in a set of nuclear material accounting documents that include one storage basket list, two transfer journals and five summary ledgers.
3.1. Storage basket list

This list is kept in the Accountability Office to summarize all fuel movements to and from the storage baskets. It gives a survey of all loading and unloading records that report changes of the basket content. It also indicates the "legal situation" of the fuel. The "legal situation" depends on information from the shipper about the agreement ruling his reception of the fuel, e.g. a bilateral agreement. In addition to this storage basket list kept by the Accountability Section, a "basket record file" with more details of the basket contents is kept by the fuel storage operators.

3.2. Transfer journals

Two journals are used for booking transfers of source and special materials, the transfer journal for fuel storage area and the transfer journal for all other material balance areas. Separate book-keeping for the fuel storage is made for the following reasons:

1. Book-keeping of source and special material in this area is necessarily based on figures calculated by the reactor operator, whereas all quantities of material in the remaining areas are obtained from measurements of solution volume or product weight and sample analysis.
2. In the storage basin, a great variety of fuels may be present at the same time, i.e. fuels of different enrichment or ownership, which must be accounted separately before their true source and special material content can be determined by the plant input measurement. Separate treatment and accounting, however, is not technically practicable once the material has been fed into the continuous processing chain.

The first transfer journal is a catalogue of all fuel receptions in and removals from the irradiated fuel storage area, the removals normally being made for the preparation of dissolver charges. It has different accounts for each source and special material (uranium and plutonium); those for uranium are grouped into three classes of enrichment (<1.6% $^{235}$U, between 1.6 and 5% $^{235}$U, and > 5% $^{235}$U). At the end of each month, all entries in the journal are balanced for the different fuel suppliers.

The second transfer journal is used for daily entry of all transfers of process solutions or solid product. It has separate accounts for each material balance area, subdivided into total uranium, $^{235}$U and plutonium. At the end of each month, all transfers between every two MBAs are balanced and entered in the respective summary ledgers.

3.3. Summary ledgers

These ledgers are used for a monthly summary of all source and special material transfers between the various material balance areas. They are needed to prepare the monthly inventory and balance reports.

Summary ledgers are kept for the following different types of source and special material: depleted uranium, natural uranium, enriched uranium, uranium-233, and plutonium.

A double entry system is used; external transfers are posted to the "Eurochemic Plant Balance" account as well as to the account of the MBA
concerned, while internal transfers are entered in the accounts of both MBAs concerned.

All fuel receptions from each reactor station during the month under review are totalled in the transfer journal and entered in the summary ledger as "Off-site Receipts". The total from each reactor is posted separately. Off-site shipments to the various receivers are also separately totalled and posted correspondingly.

4. PRESENT STATE OF NUCLEAR MATERIALS CONTROL AT EUROCHEMIC

Since the Eurochemic reprocessing plant is not yet in operation, nuclear material control is still in a development phase. Nevertheless, accounting operations started some time ago, when the research laboratory was put into service. At the beginning of 1965 the fuel storage building began operating, and about 6000 kg of natural and slightly enriched fuels and some 150 MTR-type elements have been received until the date this report was submitted. Inactive plant testing started in June with tank calibrations. For this work, the accountability section prepared standard procedures and supervised the execution. Particular emphasis was placed on the calibration of tanks used for input and output determinations and internal bulk transfers of fissile material between different MBAs.

Some experience has been accumulated during accounting operations for the research laboratory and fuel element storage. Nevertheless, the efficiency of the system described for nuclear material accounting in the reprocessing plant remains to be demonstrated.
NUCLEAR MATERIALS MANAGEMENT AT THE JAPAN ATOMIC ENERGY RESEARCH INSTITUTE

C. MACHIDA AND I. HIRAMATSU
JAPAN ATOMIC ENERGY RESEARCH INSTITUTE,
TOKAI-MURA, IBARAKI-KEN, JAPAN

Abstract — Résumé — Аннотация — Resumen

NUCLEAR MATERIALS MANAGEMENT AT THE JAPAN ATOMIC ENERGY RESEARCH INSTITUTE. The procedures for nuclear material control are reviewed, based on the experiences at the Japan Atomic Energy Research Institute. Nuclear materials to be discussed here include:
- Both natural and enriched uranium for research;
- Imported enriched nuclear fuel elements for JRR-2 (10-MW CP-5), JRR-4 (1-MW swimming pool), JPDR (12.5-MW(e) BWR), and the critical assemblies for JMTR (50-MW light-water moderated) and for the propulsion reactor;
- Domestically-fabricated natural uranium fuel elements for JRR-3 (10-MW heavy-water moderated);
- Domestically-fabricated fuels for the critical assemblies manufactured from imported enriched uranium oxides;
- Domestically-fabricated enriched fuel elements for JPDR and for the propulsion reactor manufactured from imported enriched uranium hexafluoride.

Both thorium and plutonium are also under control, but excluded from the present paper. Entire administrative pattern for nuclear material control is first presented. The emphasis is placed on the domestic fabrication of enriched fuel elements from imported enriched uranium, and the details of the control procedures during and after the fabrication process are discussed. The control procedures include the chemical analysis for purity check, isotopic assay by mass spectrometry, physical and mechanical tests of fabricated products, and the careful prevention in the diversion of nuclear materials.

Administrative problems being attributed to Japanese domestic situation are presented; for example, the segregation, collection and efficient recovery and practical uses of residual uranium from the fabrication process. Methods for keeping records on the storage and uses of nuclear materials are also discussed. More satisfactory control procedures for other nuclear materials such as thorium and heavy water are under progress.

GESTION DES MATIERES NUCLEAIRES AU CENTRE JAPONAIS D'ETUDES NUCLEAIRES. Les auteurs exposent les méthodes de contrôle des matières nucléaires fondées sur l'expérience acquise au Centre japonais d'études nucléaires. Le mémoire traite de la gestion des matières suivantes:
- Uranium naturel et uranium enrichi utilisés pour la recherche;
- Eléments combustibles (uranium enrichi) importés pour les réacteurs JRR-2 (10 MW CP-5), JRR-4 (1 MW, piscine), JPDR (12.5 MW(e), eau bouillante), et les assemblages critiques pour JMTR (50 MW, ralenti à l'eau légère) et pour le réacteur de propulsion;
- Eléments combustibles à l'uranium naturel, fabriqués au Japon pour JRR-3 (10 MW, ralenti à l'eau lourde);
- Combustibles pour assemblages critiques, fabriqués au Japon à partir d'oxydes d'uranium enrichi importé;
- Eléments combustibles enrichis pour JPDR et pour le réacteur de propulsion, fabriqués au Japon à partir d'hexafluorure d'uranium enrichi importé.

Le thorium et le plutonium font aussi l'objet d'un contrôle, mais il n'en est pas question dans le mémoire. Les auteurs exposent en premier lieu toute la procédure administrative du contrôle des matières nucléaires. Ils mettent l'accent sur la fabrication, au Japon, d'éléments combustibles à partir d'uranium enrichi importé, et ils discutent les détails du contrôle exercé pendant et après la fabrication. Ce contrôle comporte l'analyse chimique pour déterminer la pureté du produit, le dosage isotopique par spectrométrie de masse, l'application d'épreuves physiques et mécaniques au produit fini et une surveillance très stricte visant à prévenir le détournement de matières nucléaires.
Les auteurs exposent des problèmes d’administration propres à la situation qui prévaut au Japon, par exemple la séparation, le ramassage, la récupération effective et les utilisations pratiques des résidus d’uranium provenant de la fabrication. Ils examinent également les méthodes de comptabilité concernant l’entreposage et les utilisations des matières nucléaires. Des procédés de contrôle plus satisfaisants sont à l’étude pour d’autres matières nucléaires telles que le thorium et l’eau lourde.

ОБРАЩЕНИЕ С ЯДЕРНЫМИ МАТЕРИАЛАМИ В НАУЧНО-ИССЛЕДОВАТЕЛЬСКОМ ИНСТИТУТЕ АТОМНОЙ ЭНЕРГИИ ЯПОНИИ. Методы контроля за ядерным материалом рассмотриваются в свете опыта, накопленного в Научно-исследовательском институте атомной энергии Японии. В состав ядерных материалов, о которых идет речь, входят:
- природный и обогащенный уран для исследовательских целей;
- импортированные ядерные топливные элементы из обогащенного материала для реакторов JRR-2 (10 МВт CP-5), JRR-4 (1 МВт бассейнового типа), JPDR (12,5 МВт(эн) типа BWR), критической сборки для реактора JMTR (50 МВт с замедлителем в виде обычной воды) и для реактора для двигателей;
- топливные элементы, местного изготовления из природного урана, для реактора JRR-3 (10 МВт с замедлителем в виде тяжелой воды);
- топливные элементы местного производства для критических сборок.
- топливные элементы, изготовленные из импортных обогащенных окисей урана;
- обогащенные топливные элементы местного производства для реактора JPDR и для реактора для двигателей, построенного из импортного обогащенного шестифтористого урана.

ОБРАЩЕНИЕ С ЯДЕРНЫМИ МАТЕРИАЛАМИ В НАУЧНО-ИССЛЕДОВАТЕЛЬСКОМ ИНСТИТУТЕ АТОМНОЙ ЭНЕРГИИ ЯПОНИИ. Методы контроля за ядерным материалом рассмотрываются в свете опыта, накопленного в Научно-исследовательском институте атомной энергии Японии. В состав ядерных материалов, о которых идет речь, входят:
- природный и обогащенный уран для исследовательских целей;
- импортированные ядерные топливные элементы из обогащенного материала для реакторов JRR-2 (10 МВт CP-5), JRR-4 (1 МВт бассейнового типа), JPDR (12,5 МВт(эн) типа BWR), критической сборки для реактора JMTR (50 МВт с замедлителем в виде обычной воды) и для реактора для двигателей;
- топливные элементы, местного изготовления из природного урана, для реактора JRR-3 (10 МВт с замедлителем в виде тяжелой воды);
- топливные элементы местного производства для критических сборок.
- топливные элементы, изготовленные из импортных обогащенных окисей урана;
- обогащенные топливные элементы местного производства для реактора JPDR и для реактора для двигателей, построенного из импортного обогащенного шестифтористого урана.

Как торий, так и плутоний также поставлены под контроль, но они исключаются из обсуждения в данном докладе. Все административная схема контроля над ядерными материалом представляется впервые. Упор делается на местное производство обогащенных топливных элементов из импортного обогащенного урана и подробно обсуждаются методы контроля в процессе изготовления и после него. В состав методов контроля включаются химический анализ на предмет проверки чистоты, изотопическая оценка методами масс-спектрометрии, физическая и механическая проверки готовой продукции и полное предотвращение использования ядерных материалов не по назначению.

Представляются проблемы административного характера, характерные для внутреннего положения в Японии, например разделение, сбор, эффективное восстановление и практическое использование отходов урана в результате процесса изготовления. Обсуждаются также методы ведения учетных документов при хранении и использовании ядерных материалов. В настоящее время разрабатываются более удовлетворительные методы контроля для других ядерных материалов, например тория и тяжелой воды.

ADMINISTRACION DE MATERIALES NUCLEARES EN EL ATOMIC ENERGY RESEARCH INSTITUTE DEL JAPON. En esta memoria se examinan los procedimientos de control de materiales nucleares establecidos sobre la base de la experiencia adquirida por el Atomic Energy Research Institute del Japón. Los materiales nucleares de que se trata son:
- El uranio natural y el uranio enriquecido, para trabajos de investigación;
- Los elementos combustibles enriquecidos importados para los reactores JRR-2 (10 MW, tipo CP-5), JRR-4 (1 MW, tipo piscina), JPDR (12,5 MW, eléctricos, tipo BWR), y los montajes críticos para el JMTR (60 MW, moderado por agua) y para el reactor de propulsión;
- Los elementos combustibles de uranio natural fabricados en el Japón para el JRR-3 (10 MW, moderado por agua pesada);
- Los combustibles fabricados en el Japón para montajes críticos elaborados a base de óxidos de uranio enriquecido importados;
- Los elementos combustibles enriquecidos fabricados en el Japón para el JRDR y para el reactor de propulsión, elaborados a base de hexafluoruro de uranio enriquecido importado.

El torio y el plutonio están también sujetos a control, pero en la memoria no se trata de ellos. Los autores describen en primer lugar la organización administrativa completa en que se basa el control de los materiales nucleares. Insisten particularmente en los elementos combustibles enriquecidos fabricados en el Japón a partir de uranio enriquecido importado y analizan detenidamente los procedimientos de control durante y después del proceso de fabricación. Estos procedimientos incluyen los análisis químicos para la comprobación de la pureza, las determinaciones de la composición isotópica por espectrometría de masas, los ensayos físicos y mecánicos de los productos fabricados y las medidas para prevenir la disolución de los materiales nucleares.

Se exponen algunos problemas administrativos atribuidos a la situación interna del Japón, por ejemplo: la separación, recolección y recuperación eficiente del uranio residual de los procesos de fabricación, y la
utilización práctica de ese uranio. Se analizan también métodos para mantener al día la información sobre el almacenamiento y el empleo de los materiales nucleares. En la actualidad se elaboran procedimientos más satisfactorios de control para otros materiales nucleares, tales como el torio y el agua pesada.

ADMINISTRATION ORGANIZATION

In the Japan Atomic Energy Research Institute (JAERI), four research reactors (JRR-1, -2, -3 and -4) and one power demonstration reactor (JPDR) are in operation, and one material testing reactor (JMTR) is now under construction.

In addition to the above, three experimental critical assemblies and one subcritical assembly are operated, and another critical assembly is also under construction (Table I).

Some research and development sections are associated with nuclear fuels. With the development of the Japanese nuclear power generation programme, the activities of the manufacturers of nuclear fuel have become larger and they have recently proposed the trial production of nuclear fuels for JAERI's reactors. JAERI accepted those proposals and agreed to cooperate with them in the development of the technology of nuclear fuel fabrication. However, at that time there was much to be improved in the administration of the nuclear fuels in the Institute: after discussions and preparatory works from April 1962 to September 1962, the following system of administration was put into practice.

Management of nuclear materials in the Institute is being undertaken by chiefs of the nuclear fuel section and other sections using such materials as well as group leaders in these sections. These group leaders are in charge of actual management under the supervision of section chiefs.

Group leaders keep the books for the accounting of their inventories, receipts and transfers, and also exercise their own discretion on how to use such materials. This organization is based on the following considerations:

1. The particular use of a nuclear material depends on the subject of research carried out under the group leader. Thus, it is proper for the leader to be responsible for the nuclear fuel used.

2. In the case of laboratories, where there are different research subjects and many research workers, the transfer of nuclear material occurs between the research groups. This internal transfer is one of the causes of discrepancy between the figures and the actual amounts.

3. Members of the laboratories are shifted sometimes according to a change in the research organization of the Institute; however, the groups, usually as a unit of research activities, remain unchanged. The nuclear fuel section belongs to the Division of Administrative Services and is responsible for the management of all the nuclear materials in the Institute. Therefore the chief of the nuclear fuel section is capable of direct supervision of all such section chiefs and group leaders. Also, any transfer of nuclear materials between the JAERI and other external organizations is done through the nuclear fuel section.

By this system nuclear materials management is under the general control of the nuclear fuel section, where, therefore, any information concerning uses, storage, transfer, etc., is available.
FEATURES AND PROBLEMS OF THE GROUPS

There are 54 material balance areas in the Institute where nuclear materials are handled. These are in reactor operation, reactor design, fuel research and development and other divisions, and also in the nuclear fuel section (Table II).

Reactor operation division

The reactor operation division includes four research reactors, one critical experimental assembly, one hot laboratory, in-pile loops, etc., and in addition there is the JPDR. The nuclear fuel problems raised include calculation of the fuels burnt, other nuclear fuel matters in the reactor operation schedules, storage of the spent fuels, etc. With regard to reprocessing of the spent fuels, highly enriched fuels will be reprocessed in the United States, and the slightly enriched or natural uranium fuels will be reprocessed in Japan; however, the US-Japan Chemical Reprocessing Contract has not yet been concluded and the reprocessing facilities within Japan are scheduled to start in 1971. Until then the storage of spent fuels is an important problem. Spent fuels of high enrichment (JRR-2, -4) are stored in cooling storage ponds, and those of natural uranium (JRR-3) and ruptured elements are stored in exposed drying pits. The cooling ponds are expensive, costing approximately $400,000 for each pond holding 100 MTR-type spent fuel elements. But this storage pond is advantageous in storing the spent fuels in good condition and in their easy handling. At present, the Institute stores 50 highly enriched spent fuel elements and these will be increased to about 270 by the end of 1969. We are greatly interested in the agreement between the United States and Japan. The dry pit with 100 storeholes (an 18-ton capacity track crane included) costs $90,000 and is much cheaper to construct. However, as it is uncertain whether or not the circulation of dry air is satisfactory enough to keep it in good condition, relatively cheap natural uranium spent fuel elements with a thick sheath are stored there. The other important aspect of nuclear fuel control in the reactor operation division relates to nuclear materials used for material irradiations and in the hot laboratory. The number of irradiations handled in 1964 was about 2000, 700 of which were done on requests from outside the Institute. Approximately 15% of the irradiations was for nuclear materials. It is difficult in the hot laboratory to determine the amount of nuclear materials. The amount of nuclear materials lost in the laboratory is determined by subtracting the amount weighed after the test from the initial amount. Actually, this determination is done by the proportionality method, in which the gross weight of the nuclear material taken into the laboratory is weighed, and later, after the test has been done, the remaining weight is weighed.

Reactor design division

In the reactor design division, two critical assemblies and instrumentation research facilities are in operation, the research groups being a total of nine. There are no particular problems in this division.
TABLE I
NUCLEAR REACTORS AND EXPERIMENTAL CRITICAL FACILITIES

<table>
<thead>
<tr>
<th>Name</th>
<th>Type</th>
<th>Output (t) (kW)</th>
<th>Forms of fuels</th>
<th>No. of assemblies</th>
<th>Amount of uranium (kg)</th>
<th>Enrichment (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>JRR-1</td>
<td>W.B.</td>
<td>50</td>
<td>Uranyl sulphate</td>
<td>-</td>
<td>6.6</td>
<td>20</td>
</tr>
<tr>
<td>JRR-2</td>
<td>CP-5</td>
<td>10 000</td>
<td>Improved MTR</td>
<td>24</td>
<td>4.6</td>
<td>90</td>
</tr>
<tr>
<td>JRR-3</td>
<td>NRX</td>
<td>10 000</td>
<td>NRX</td>
<td>246</td>
<td>6093.0</td>
<td>N.U.</td>
</tr>
<tr>
<td>JRR-4</td>
<td>Swimming pool</td>
<td>1 000</td>
<td>Improved ETR</td>
<td>20</td>
<td>3.3</td>
<td>90</td>
</tr>
<tr>
<td>JPDR</td>
<td>BWR</td>
<td>46 000 (12 500e)</td>
<td>UO₂</td>
<td>72</td>
<td>4575.0</td>
<td>2.6</td>
</tr>
<tr>
<td>TCA (LWCA)</td>
<td>-</td>
<td>-</td>
<td>UO₂</td>
<td>720</td>
<td>1163.9</td>
<td>2.6</td>
</tr>
<tr>
<td>AHCA</td>
<td>-</td>
<td>-</td>
<td>Uranyl sulphate</td>
<td>-</td>
<td>13.2</td>
<td>20</td>
</tr>
<tr>
<td>SHCA</td>
<td>-</td>
<td>-</td>
<td>UO₂ in graphite</td>
<td>-</td>
<td>76.0</td>
<td>20</td>
</tr>
<tr>
<td>JMTR a</td>
<td>Improved ETR</td>
<td>50 000</td>
<td>Improved ETR</td>
<td>27</td>
<td>6.5</td>
<td>90</td>
</tr>
<tr>
<td>JMTRCA a</td>
<td>-</td>
<td>-</td>
<td>Improved ETR</td>
<td>27</td>
<td>6.5</td>
<td>90</td>
</tr>
</tbody>
</table>

a Under construction.

Fuel research and development division

The administration of nuclear materials in the fuel research and development division is very complicated because of the varied types of nuclear material, their different uses, and transfers to and from other external establishments. The division includes a metals laboratory, a fuel irradiation laboratory, a ceramic fuels laboratory and a reprocessing research and testing plant, and is divided into 11 groups. The main feature of the division is the variation in the physical and chemical forms of such materials. The amount of nuclear materials must be shown in the amounts of uranium and uranium-235, not in the amount of the chemical compounds or other physical forms. One method is to trace the amount of nuclear materials, e.g. irrespective of the chemical forms, and the other is to separate one form from the other forms of nuclear materials, keeping separate books for the respective fuels. The former method was considered to be very difficult to carry out in the Institute, and the latter method was therefore employed.
<table>
<thead>
<tr>
<th>Divisions</th>
<th>Sections</th>
<th>Groups</th>
</tr>
</thead>
<tbody>
<tr>
<td>Administrative services</td>
<td>Nuclear fuel</td>
<td>Fuel management</td>
</tr>
<tr>
<td>Research reactor operation</td>
<td>Research reactor services</td>
<td>Utilization, business</td>
</tr>
<tr>
<td></td>
<td>Research reactor technology</td>
<td>Same as the section</td>
</tr>
<tr>
<td></td>
<td>JRR-1 operations</td>
<td>Same as the section</td>
</tr>
<tr>
<td></td>
<td>JRR-2 operation</td>
<td>Same as the section</td>
</tr>
<tr>
<td></td>
<td>JRR-3 operation</td>
<td>Same as the section</td>
</tr>
<tr>
<td></td>
<td>JRR-4 operation</td>
<td>Same as the section</td>
</tr>
<tr>
<td></td>
<td>Hot laboratory</td>
<td>Same as the section</td>
</tr>
<tr>
<td>JPDR operation</td>
<td>Section I (JPDR)</td>
<td>Group I</td>
</tr>
<tr>
<td></td>
<td>Section II (TCA)</td>
<td>Group IV</td>
</tr>
<tr>
<td>Reactor design and engineering</td>
<td>Fast reactor physics laboratory</td>
<td>Fast reactor physics (2)</td>
</tr>
<tr>
<td></td>
<td>Reactor physics experiment laboratory</td>
<td>Fast reactor experiment (2)</td>
</tr>
<tr>
<td></td>
<td>Heat design laboratory</td>
<td>Same as the section</td>
</tr>
<tr>
<td></td>
<td>Heat engineering laboratory</td>
<td>Same as the section</td>
</tr>
<tr>
<td></td>
<td>Reactor instrumentation laboratory</td>
<td>Same as the section</td>
</tr>
<tr>
<td></td>
<td>Material engineering laboratory</td>
<td>Same as the section</td>
</tr>
<tr>
<td></td>
<td>Shielding laboratory</td>
<td>Same as the section</td>
</tr>
<tr>
<td>Fuel research and development</td>
<td>Metals laboratory</td>
<td>Metal</td>
</tr>
<tr>
<td></td>
<td>Ceramic fuels laboratory</td>
<td>Ceramic fuels (2)</td>
</tr>
<tr>
<td></td>
<td>Plutonium fuels laboratory</td>
<td>Plutonium fuels</td>
</tr>
<tr>
<td></td>
<td>Fuel irradiation laboratory</td>
<td>Fuel irradiation (2)</td>
</tr>
<tr>
<td></td>
<td>Material irradiation laboratory</td>
<td>Material irradiation</td>
</tr>
<tr>
<td></td>
<td>Structure analyses laboratory</td>
<td>Structure analyses</td>
</tr>
<tr>
<td></td>
<td>Reprocessing laboratory</td>
<td>Reprocessing laboratory (2)</td>
</tr>
<tr>
<td></td>
<td>Reprocessing development laboratory</td>
<td>Reprocessing development laboratory</td>
</tr>
<tr>
<td>Divisions</td>
<td>Sections</td>
<td>Groups</td>
</tr>
<tr>
<td>--------------------------</td>
<td>-----------------------------------</td>
<td>---------------------------------------</td>
</tr>
<tr>
<td>Research</td>
<td>Nucleus physics I laboratory</td>
<td>Nucleus physics I</td>
</tr>
<tr>
<td></td>
<td>Nucleus physics II laboratory</td>
<td>Nucleus physics II</td>
</tr>
<tr>
<td></td>
<td>Fuel chemistry laboratory</td>
<td>Fuel chemistry (4)</td>
</tr>
<tr>
<td></td>
<td>Radiochemistry laboratory</td>
<td>Radiochemistry (4)</td>
</tr>
<tr>
<td></td>
<td>Reactor chemistry laboratory</td>
<td>Reactor chemistry</td>
</tr>
<tr>
<td></td>
<td>Analytical chemistry laboratory</td>
<td>Analytical chemistry laboratory</td>
</tr>
<tr>
<td>Health physics and safety</td>
<td>Environmental survey</td>
<td>Observation</td>
</tr>
<tr>
<td></td>
<td>Bioassay</td>
<td>Development</td>
</tr>
<tr>
<td></td>
<td>Radiation dosimetry</td>
<td>Development</td>
</tr>
<tr>
<td></td>
<td>Waste disposal and decontamination</td>
<td>Waste disposal decontamination</td>
</tr>
<tr>
<td>Radiisotope centre</td>
<td>Production</td>
<td>Business</td>
</tr>
<tr>
<td></td>
<td>Production development</td>
<td>Technology II</td>
</tr>
<tr>
<td>Radiisotope school</td>
<td></td>
<td>Radiisotope school</td>
</tr>
<tr>
<td>Nuclear engineering school</td>
<td></td>
<td>Business</td>
</tr>
<tr>
<td>JMTR project</td>
<td>JMTR administration</td>
<td>Business</td>
</tr>
<tr>
<td></td>
<td>Irradiation</td>
<td>Irradiation</td>
</tr>
<tr>
<td></td>
<td>Reactor</td>
<td>Reactor</td>
</tr>
<tr>
<td>Oarai field office</td>
<td>Administrative services</td>
<td>Administrative service</td>
</tr>
</tbody>
</table>

N.B. Figures in parentheses show numbers of groups using nuclear materials. The total number is 54 groups.

The second feature of the division is the frequent transfer of material from and to other outside establishments. There were about 110 such transfers to during the period January 1964 - April 1965, 50 in the fuel research and development division and another 50 concerning chemical analyses specimens related to nuclear fuel development.
Research division

The main feature of the research division is the small amounts of nuclear materials used. The kinds and forms of the nuclear materials are almost the same as those in the fuel research and development division; however, this division uses a few grams or several tens of grams of nuclear material compared with the kilograms used in the fuel research and development division. Reports to the government are in kilograms for natural uranium, depleted uranium and thorium, and in grams for enriched uranium, uranium-233 and plutonium. However, the actual administration is done by grams for natural uranium, depleted uranium and thorium, and 0.01 grams for enriched uranium, uranium-233 and plutonium. The use of these units is subject to the judgment of the chief of the nuclear fuel section.

These units are employed because of the importance of knowing the amounts that have been actually used. Accordingly, in the case of large units, i.e. kilograms, the addition of a few zeros is necessary. The aim is to assess not only the effective total amount but also to know the amounts in the respective groups.

Nuclear fuel section

The special feature of the nuclear fuel section is to act as a channel for nuclear materials between JAERI groups and the outside, and to carry out the administrative work concentrated in this section. The latter includes approval of the uses of nuclear materials, procedures to obtain or divert nuclear materials, reports to the government, ordering of fuel materials, weighing of the delivered nuclear materials, procedures to transfer materials to outside the JAERI, safeguards required according to the international treaty, etc. The section carries out the stock and storage of the returned materials, storage of scrapped materials and off-site control of nuclear material delivered to private fuel manufactures, etc. The nuclear material, to be used for fuel elements by the manufacturers, is inspected through reports submitted by these firms and actual inspection of fuel material in the factories. In this case, problems sometimes occur concerning differences in the weights of the fuel materials handed to such firms and those received from them. This is caused by inaccuracies in the weighing instruments used. The following methods are therefore used:

1. When the discrepancy is less than 1/10 000, the supplier's weight is final.
2. When the discrepancy is larger than 1/1000, the final weight is decided after consultations. Weighing in the presence of witnesses may be done, if necessary.
3. When the discrepancy is between 1/10 000 and 1/1000, the recipient (JAERI), selects either method (1) or (2).

In important deliveries of fuel material to the fuel manufacturers, and in deliveries of the finished fuel elements to the Institute, the weighing is done in the presence of the witnesses from the both sides.
THE METHOD OF ACCOUNTABILITY

The nuclear fuel management in the Institute consists of the following three phases:

(1) Report on the transfer of the nuclear materials

Transfer form of nuclear material inside JAERI (Table III)

In transfer from one group to another, three copies of the transfer form are completed by the shipper. One copy is retained and the remaining two are sent to the group receiving the materials. This latter group checks the materials against the completed transfer form, and then, when confirmed, one copy is sent to the nuclear fuel section. When transfer is made outside the JAERI, the group concerned submits three copies of the form together with the fuel to the nuclear fuel section. After this, the section is wholly responsible for transfer of the materials.

When nuclear materials are taken into the Institute from outside, they are transmitted to the nuclear fuel section, together with the attached document papers, where the materials are checked against the documents, and the section then issues the transfer forms, and the fuel materials are sent to the receiving group or groups together with the transfer forms.

Notification of shipping nuclear materials to outside establishment

This notification is made to the chief of the receiving establishment from the chief of the nuclear fuel section. This ensures that the necessary procedure is carried out on both sides, and provides the receiving party with information necessary to report to the government.

(2) Recording in the account book

Every material balance area completes the regular ledger with records of receiving, delivery, use and disposal (Table IV).

The nuclear fuel section also records the same items in the same ledger according to the transfer form submitted by the material balance areas.

The ledger is completed separately for groups, kinds and forms of nuclear materials. In the nuclear fuel section, other ledgers are also maintained for receiving from outside and delivery to outside respectively.

(3) Routine inspection of the fuel inventories

The nuclear fuel section carries out a routine inspection twice a year for all the material balance areas, as follows:

Checking the ledgers

The ledger held in the nuclear fuel section is checked against the ledgers in the material balance area. Any discrepancies are caused by neglecting
### TABLE III
NUCLEAR MATERIAL TRANSFER FORM INSIDE JAERI
(JAERI-NF Form 2-2)

<table>
<thead>
<tr>
<th>(Delivering section or lab.)</th>
<th>(Receiving section or lab.)</th>
<th>(Nuclear fuel section)</th>
</tr>
</thead>
<tbody>
<tr>
<td>To:</td>
<td></td>
<td>No.</td>
</tr>
<tr>
<td>Date:</td>
<td>Date:</td>
<td></td>
</tr>
<tr>
<td>Chief of delivering section or lab.</td>
<td></td>
<td>Signature</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Delivery</th>
<th>Receiving</th>
</tr>
</thead>
<tbody>
<tr>
<td>Delivery date</td>
<td>Receiving date</td>
</tr>
<tr>
<td>Delivery place</td>
<td>Receiving place</td>
</tr>
<tr>
<td>Building</td>
<td>Room</td>
</tr>
<tr>
<td>Person in charge</td>
<td>Person in charge</td>
</tr>
<tr>
<td>Signature</td>
<td>Phone</td>
</tr>
<tr>
<td>Group leader</td>
<td>Signature</td>
</tr>
<tr>
<td>Phone</td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Kind: Enriched U, Natural U, Depleted U, U-233, Thorium, Plutonium</th>
</tr>
</thead>
<tbody>
<tr>
<td>Chemical form:</td>
</tr>
<tr>
<td>Physical form:</td>
</tr>
<tr>
<td>Quantity:</td>
</tr>
<tr>
<td>Number, caps or bottles</td>
</tr>
<tr>
<td>Container No.</td>
</tr>
<tr>
<td>----------------</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Reason for delivery:</th>
</tr>
</thead>
<tbody>
<tr>
<td>Remarks:</td>
</tr>
</tbody>
</table>

This is to certify that the above nuclear material has been received.

Date: Chief of receiving section or lab. Signature

<table>
<thead>
<tr>
<th>Nuclear fuel section</th>
<th>Person in charge</th>
<th>Record of receiving</th>
<th>Record of delivery</th>
<th>Witness</th>
<th>Register</th>
<th>Keep this form for 10 years by law</th>
</tr>
</thead>
</table>
### TABLE IV

**NUCLEAR MATERIAL LEDGER FORM**  
*(For enriched uranium)*

<table>
<thead>
<tr>
<th>Date</th>
<th>Items</th>
<th>Slip No.</th>
<th>Received</th>
<th>Delivered</th>
<th>Disposed</th>
<th>Balance</th>
<th>Present</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Material</td>
<td>U</td>
</tr>
</tbody>
</table>

(Special information)  
Purchase, lease, other lease

Purchased price @ Amount Total price

<table>
<thead>
<tr>
<th>Reference No.</th>
<th>Material</th>
<th>Container No.</th>
<th>Status of material</th>
<th>Name of producer or origin user location</th>
<th>Fabricator</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Purchase, lease, other lease  
Building room lab.
to complete the ledger, escape of dissolved fuel materials in the drainage, etc.

Physical inventory of the nuclear fuels

If an error in weighing is feared, a standard scale and weights should be used. However, few problems occur with the usual scales. The discrepancy between the weight in the ledger and that of the actual material is caused by failure to issue the transfer form, by neglecting to complete the ledger with the depleted amount of fuel material, by an error in keeping the records, etc. These discrepancies are found and corrected at this stage.

CONTROL OF THE FUEL MATERIALS OUTSIDE THE JAERI

A special case of management is the control of nuclear materials handed to manufacturers for fabrication into fuel elements. This includes nuclear material for the second core of JRR-3, for the trial fuel for JPDR, and for the critical assembly for the JMTR.

These supplies are distributed under a contract indicating that the Institute delivers raw nuclear fuel obtained beforehand to a manufacturer and that a manufacturer returns not only the products but also scrapped nuclear materials to the Institute. For this transfer, there are the following three procedures:

Certificate on delivery and receiving of the materials and elements

When the Institute provides fuel to the manufacturer it issues a delivery form for the supply of nuclear material (Table V), to which a certificate on weight, quality, etc. is attached (see above). In the case of purchased raw materials, a certificate issued by the firm serves the same purpose. The report on the products and returned scraps by the manufacturer is done on the material transfer form prepared by the Institute. This report contains a summary report (Table VI), the product-weight certificate, returned material detail report and report of loss and error, etc.

Weighing of fuel materials

The weighing is done according to the procedure described above. However, when uranium metal turnings are sent, immersed in machine oil, there is a considerable discrepancy between those measurements. The shipment of the scraps in the form of the oxide or ingot is done as much as possible to avoid the weight discrepancy and to ensure safety.

Chemical analysis

Chemical analyses are usually performed on the supplied material, products and returned materials. These chemical analyses are carried out to determine the amount of uranium, and the impurities included. Mass spec-
**TABLE V**

**DELIVERY FORM FOR NUCLEAR FUEL MATERIAL SUPPLIED**
*(JAERI-NF Form 10-1)*

<table>
<thead>
<tr>
<th>No.</th>
<th>Addressee</th>
<th>Date:</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Japan Atomic Energy Research Institute</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Issuer</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Signature</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Receiver</th>
<th>Location</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Name of organization</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Person responsible for receiving</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Delivery point</th>
<th>Method of transportation</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Delivery date</th>
<th>Person in charge of transportation</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Person in charge at JAERI</th>
<th>Person in charge at the organization</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Name of commodity</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Name of chemical compound</th>
<th>Form</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Number of container(s)</th>
<th>Number of product(s)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Nuclear fuel material</th>
<th>Gross weight (g)</th>
<th>Weight of container (g)</th>
<th>Weight of chemical compound (g)</th>
<th>U weight (g)</th>
<th>Enrichment (%)</th>
<th>U-235 weight (g)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Reason for delivering of material supplied</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Radiation control</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Remarks</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
</tr>
</tbody>
</table>

**N.B.** Three copies are issued, one for the receiver, one for JAERI, and one to be returned to JAERI from the receiver as receipt.
TABLE VI
SUMMARY REPORT OF NUCLEAR FUEL MATERIAL SUPPLIED
(JAERI-NF Form 10-3)

<table>
<thead>
<tr>
<th>Item</th>
<th>Name of commodity</th>
<th>Weight of chemical compound (10 mg)</th>
<th>Weight of U (10 mg)</th>
<th>Enrichment (%)</th>
<th>U-235 (10 mg)</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Supplied material (to maker)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Product delivered</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Test sample</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1. For chemical analyses</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>2. For metallurgy</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>3. Others</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
| Total | | | | | | (1 ~ 3)
| Returned material | | | | | | |
| 1. Unused material supplied | | | | | | |
| 2. Product in excess | | | | | | |
| Total | | | | | | (1 ~ 2)
| Loss | | | | | | |
| 1. Fabrication loss | | | | | | |
| 2. Analyses loss | | | | | | |
| 3. Unknown loss | | | | | | |
| Total | | | | | | (1 ~ 2)
| Weighing error | | | | | | |
| Grand total | | | | | | |

Note: Product weight certificate (Form 10-4) and Returned material details (Form 10-5) are attached to this report.
trographic analyses are done on purchased UF₆, uranium metal, and on the finished fuels.

UTILIZATION OF RETURNED SCRAPS

The Institute asks the Atomic Fuel Corporation or other private companies for purification of natural uranium scraps. Scrap materials include highly enriched uranium (90%)-aluminium alloys, and slightly enriched uranium oxide. The former are produced in the fabrication of the fuel elements for the JMTR critical assembly; this scrap material is expected to total 1.5 kg. Using this scrap material, the Institute, with an additional 2.5 kg of uranium metal, intends to produce the fuel element for the JRR-2 (nine fuel elements). The ratio of scrap to the total is approximately 40%.

The slightly enriched uranium scrap materials include 2.6% enrichment uranium produced in the test production of the JPDR fuel assemblies and the scrap material from the fabrication for the nuclear ship critical experimental assembly (2.7 and 3.0% enrichments). The JPDR scrap material and part of the critical assembly are to be used for the second JPDR test fuel assemblies. In these cases it is necessary to obtain accurate information on the quality of the scrap materials.
THE MANAGEMENT OF NUCLEAR MATERIALS IN A RESEARCH ESTABLISHMENT

W.J. WRIGHT AND D.R. HOCKING
AUSTRALIAN ATOMIC ENERGY COMMISSION RESEARCH ESTABLISHMENT,
LUCAS HEIGHTS, N. S. W., AUSTRALIA

Abstract — Résumé — Аннотация — Resumen

THE MANAGEMENT OF NUCLEAR MATERIALS IN A RESEARCH ESTABLISHMENT. The functions of a nuclear materials management scheme are reviewed in relation to the activities of research establishments. Since these activities are normally non-repetitive, there is little opportunity to establish statistical quality and quantity control. The risks of an error in the material accounts must therefore be established from relatively few analytical measurements and the implications of this are discussed.

Similar arguments are applied to illustrate the difficulties of quality control on suppliers when a large variety of materials are being purchased in small quantities.

1. INTRODUCTION

Although the broad requirements and the principles of application are similar, in the matters of detail the management of nuclear materials in
a research establishment varies considerably from that in a production facility. Nuclear materials management in a research establishment is characterized by: (a) relatively small weights of material; (b) material is dispersed over many areas; (c) there is a large range of chemical-physical forms of material; (d) the residence time of material on each project is relatively long; (e) there is a wide variety of operations (experiments) in progress; and (f) there is less formal restraint on the handling of material by the higher qualified staff involved.

This report examines the functions required of a materials management scheme for a research establishment and attempts to define the basic approach required to achieve these functions.

2. THE FUNCTIONS OF A MANAGEMENT SCHEME

There are three basic reasons for the introduction of a materials management scheme

(a) Monetary control. Nuclear materials are expensive and may represent a large fraction of the investment in a research establishment; the management scheme must ensure that the material inventory is kept to a minimum and that the material is used wisely with minimum "dead stock". This requires a detailed knowledge of the programme objectives of the establishment in order to anticipate changes in demand, for example, when a change of programme may render some stock redundant.

A second object of monetary control is to ensure that the correct quantity and quality of material is received from suppliers. This aspect has received considerable attention in the literature, for example, in discussions on shipper-receiver discrepancies, although normally in relation to the control of production plants.

(b) Security and safety. The loss of nuclear material by diversion (deliberate or unintentional) may constitute a threat to the security and safety of a research establishment. The role which the materials management scheme plays in the safe operation of the establishment varies with the internal organization. At the research establishment of the Australian AEC, materials control is closely linked with control of criticality and environmental safety, all significant movements of materials being regulated through a central records office.

(c) Identification of material. The success of any research project hinges upon adequate knowledge of the properties of the starting materials and the knowledge that these properties can be reproduced as necessary. The materials control scheme thus serves an important purpose in ensuring that these data are readily available, that a consistent method of batch identification is used and that adequate stocks are reserved to ensure completion of the project.

The relative importance of these functions of material control may vary between establishments, depending upon the scope of the work undertaken and the amounts of material involved. These requirements determine the
complexity of the accounting system and, in particular, the accuracy necessary in the materials records.

3. THE ACCOUNTING SYSTEM

The focal point of any materials management scheme is the accounting system for recording the receipt and subsequent history of material. The literature contains a number of detailed examples of the actual mechanics of record-keeping, which may be adapted as required. The essential feature of these records in promoting the functions given above is their flexibility in reproducing the basic data according to various classifications.

Table I shows the classifications adopted by the AAEC to describe the material and its history. The classification of material in this way and the preparation of separate balance accounts has, in the past, involved the AAEC in considerable effort to maintain detailed ledgers and journals; even then the records were not readily available for use without considerable effort. These records have recently been programmed for the Commission's IBM 7040/1401 digital computer; the information on each batch of material is coded with the characteristics 1-12 and the computer produces a balance of material with each characteristic within about five minutes. The magnetic tape, on which the information is stored, is up-dated each week (or more often if justified) from the records of the various transactions of receipt, movement, disposal etc. during that period. The computer then produces a new series of materials balances in printed form; some of these balances, particularly those relating to dispersal of material and financial records are used directly by other groups (safety and financial accounts) within the AAEC, while the main accounting records with characteristics 1-5 are numbered and filed in an appropriate ledger for use by the materials control group.

4. THE RELIABILITY OF MATERIALS RECORDS

The reliability of materials records can be discussed in terms of the functions of the management scheme given in section 2.

Materials accounts are only estimates and, unlike a monetary system, there is often no absolutely correct value which can be recorded. The estimates can be improved by refinements in techniques and by repeated sampling but these improvements are expensive; the success of the accounting system is therefore most readily assessed by considering the cost of achieving a required level of accuracy in the records and balancing this against the risk of an error.

In inventory management alone relatively large errors (10% or so) on estimates of batch quantities are probably acceptable. Such errors are unlikely to affect the financial standing of the project or of the research establishment; nor are they likely to lead to embarrassment in continuity of supply of materials to the project. A similar situation exists in relation to control of safety; the margin for safe operation of a project is rarely so low that a 10% error in material inventory would make the project unsafe (although it may lead to errors in results of the experiments). Moreover
TABLE I
NUCLEAR MATERIALS CHARACTERISTICS

<table>
<thead>
<tr>
<th>Reason</th>
</tr>
</thead>
<tbody>
<tr>
<td>1. Element</td>
</tr>
<tr>
<td>2. Isotope</td>
</tr>
<tr>
<td>3. Chemical-physical form and analytical results</td>
</tr>
<tr>
<td>4. Quantity</td>
</tr>
<tr>
<td>5. Batch identification number</td>
</tr>
<tr>
<td>6. Holding officer (sub-account)</td>
</tr>
<tr>
<td>7. Laboratory or area involved</td>
</tr>
<tr>
<td>8. Supplier’s name</td>
</tr>
<tr>
<td>9. Country of origin</td>
</tr>
<tr>
<td>10. Internal project number</td>
</tr>
<tr>
<td>11. Inventory breakdown</td>
</tr>
<tr>
<td>(i) In stores</td>
</tr>
<tr>
<td>(ii) Not irradiated</td>
</tr>
<tr>
<td>(iii) Irradiated</td>
</tr>
<tr>
<td>12. Total project costs</td>
</tr>
<tr>
<td>(i) Value of inventory</td>
</tr>
<tr>
<td>(ii) Consumption</td>
</tr>
<tr>
<td>(iii) Other expenses</td>
</tr>
</tbody>
</table>

The actions in an emergency are not likely to be influenced by errors of this magnitude.

The role of the management scheme in collating material data (e.g. composition limits) for direct use in the research project is highly dependent upon the attitude of the research scientist and the complexity of his problem. It is generally unlikely that the materials control group could anticipate all the demands made upon it for such data without enormous expense on analytical facilities. If the project requirements for such data are well defined, specific instructions should be given for the suppliers to provide a certificate of analysis on each batch. In this case the data may be used in calculation of contained quantities for the materials records; the expense
will be entirely justified by the aims of the research project, instead of the requirements for materials accounting.

The main factors, therefore, which define the accuracy necessary in the materials records are: (1) the possibility of diversion of a quantity of material which could create a local hazard; and (2) the possibility of a financial loss due to the supply of a quantity or quality of material which is below specification.

Consideration of the first factor can be limited primarily to the diversion of fissile (special nuclear) material and to the quantities required to generate a self-sustaining reaction. The second factor is of general application to all nuclear materials.

(1) The possibility of diversion of special nuclear materials

The unknown diversion or loss of fissile materials presents a possible hazard particularly in the creation of a critical mass and consequent reaction. The circumstances often considered most likely for this accident are the build-up of fissile materials in waste drains or sumps.

This build-up of material can occur without detection by the materials accountants in three ways: (a) a single loss of an amount which will cause criticality; (b) a cumulative loss of this amount due to errors in measurement; or (c) a persistent bias in measurement.

The first two can be treated statistically if the mean and standard deviation of measurements are known accurately; the problem resolves into the calculation of the probability of obtaining a given value in a known population. This approach is normally adopted in production plants where operations are sufficiently repetitive to build up reliable statistical data. In a research establishment it is most unlikely that materials records can be brought under statistical control since the measurements are obtained from diverse operations and the true mean and standard deviations of the samples can never be estimated with adequate confidence. For example, it is unlikely that more than five samples would be taken for analysis from a delivery of fissile material because the quantities received would be small and the cost of taking more samples would be too high. The risk of a significant difference between the true value of the contained quantity and that estimated from the five samples can be relatively high and the resultant argument between shipper and receiver can be long and unsatisfactory.

There is no satisfactory solution to this problem. Inadequacy of data invariably implies a restriction on its use. The use of umpire samples, or agreement on measurement techniques, which are the normal solutions to shipper-receiver differences, are thus inadequate safeguards in these circumstances against an unknown loss of material.

The third problem, bias in measurements, is common to both research and production establishments and the accepted safeguard against a persistent bias is the inclusion of a "known" standard or calibrated sample in each measurement run. Since the standard value is determined from a large number of measurements (normally involving other parties), the confidence in this value is high; on the other hand, this confidence in the analytical measurements can be misplaced unless it has been demonstrated over a large number of measurements in the laboratory that the method is capable
of reproducing the standard value. Once again, the problem is more acute in a research establishment since a wider variety of methods are involved and the total opportunity for continuous verification of a method is less.

In short therefore, statistical control of materials accounts is difficult to achieve in a research establishment. The balance between the costs of achieving a given accuracy in the records and the risks of an error, must be assessed partly by personal judgement based upon detailed knowledge of the research programmes, and the operation of the establishment.

In principle, the AAEC has attempted to work with a small risk (say 1 in 20) that the recorded amount of material is in error by more than 5% of the true quantity received from a supplier. In subsequent actions, the records are maintained within an accuracy of 1-2% provided the material does not change chemical form (since these records are based upon repeated weighings with an accuracy of ±½% or better). If the material changes chemical form or is mixed with other materials, it is unlikely that the accuracy of materials accounts can be held within ±10% since the compositions tend to be complex and the errors in sampling are increased.

These targets have been chosen mainly as a guide to operations within the AAEC, considering the costs of analytical services and the complexity of experimental work, but it is recognized that the true accuracy and risk cannot be confirmed in all cases by adequate measurement. In assessing these points, the materials management group is required to check each operation for the possibility of undetected loss of material, to review the experimental procedures to achieve the maximum significance from the measurements of material quantities and to specify the frequency and type of measurements required for accounting.

Despite these precautions, the existence of materials accounting records alone is an inadequate safeguard against the diversion of material, unknowingly or otherwise. Thus it is necessary to monitor any area in which it is possible that material could accumulate and cause a hazardous situation. Moreover, the inadequacy of materials records admits the possibility of an undetected diversion of material. This situation highlights the need for extensive physical inventory checks associated with independent audit of the materials accounts; even then the inventory check is subject to the limitations imposed by taking a few samples, as discussed above.

(2) The check on suppliers

The verification of the quantity received from a supplier is subject to the limitations of measurements as discussed in the previous section. However, the supplier's liability also includes the quality of the product since this will affect the research programme and could involve a research establishment in a financial loss, even though the total contained quantity of material is as specified.

In practice, the solution to this problem depends mainly upon the mature judgement of the materials management group; it must assess the reliability of the suppliers in meeting the specification, and the risk to the research project that the material might be unsuitable. Against this it must weigh the costs of sampling and analysis and the probable limits of error in the results. To assist in this judgement, it is particularly useful to ob-
tain the processing records from the suppliers including the analysis of raw materials and any intermediate products which are involved. A research establishment must insist upon dealing with a reputable supplier but ensure that the specification is not so restrictive as to exclude unjustly a competitive bid.

The supply of relatively small quantities of material for research work is unfortunately subject to problems not encountered with large quantities. Suppliers have little incentive with these small orders to meet the specification in all respects, and the purchaser must, in many instances, carry the complete responsibility (and costs) of quality control. Moreover, the supplier is often reluctant to vary his standard specification limits to satisfy small orders. The suppliers' attitudes, of course, are completely understood and they emphasize the advantages of buying an established standard product. When it is considered that a research establishment may wish to buy some dozens of items in a year from numerous suppliers, it is clear that much additional effort in assessment of suppliers and their products is essential.

SUMMARY

This summary of the problems of management of nuclear materials in a research programme is not intended to imply that these problems are unique to research work, but rather to emphasize the areas where a "production" philosophy must be modified.

The conditions in a research organization persistently demand a personal assessment of the problems and decisions have to be made in many cases without the guidance of significant statistics on the risk involved. A conscientious manager is continually seeking to reduce the risk of error and to improve his service to the research scientist without unwarranted expenditure. To do this, he must use the principles of statistical sampling and quality control but recognize the limitations of statistics based on small samples. Above all, he must be conversant with the objectives and methods of the research projects, in order to appreciate fully the risks and implications of his decisions.

DISCUSSION

R.G. CARDWELL: I quite agree that it is difficult to gather sufficient data in a research organization to establish good deviation limits. I believe I understood you to say that you regard 10% error in inventory as a satisfactory measurement. Does this mean in effect that you stop searching when you have located 90% of your inventory?

W.J. WRIGHT: The guiding principle in inventories of this sort is to stop searching when it becomes too expensive to continue, or when the risk of error is reduced sufficiently in the light of the amount and importance of the material involved. In most of our work we can be confident that the errors will not exceed ±10%, although the extent of inventory, unfortunately,
often has to be a matter for individual judgement. One exception to this
statement is required for the burn-up of individual elements in the HIFAR
assembly: because the experimental reactor loading is so variable the in-
dividual elements are subject to relatively wide variations in burn-up, even
though the reactor's aggregate fuel consumption can probably be assessed
within the limits I mentioned.

R.G. CARDWELL: Are the internal project numbers you mentioned
in your oral presentation applicable only within one installation or do they
Correspond to similar project classifications throughout the A.A.E.C.?

W.J. WRIGHT: Until recently the project numbering system was limited
mainly to the research establishment. However, over the past two years
our responsibilities have been somewhat extended, to cover not only mining
and concentrates but certain other commercial activities. So far these jobs
have not been treated as separate projects, but the materials required for
them are batched and recorded under the same system.

J. SORNEIN (Chairman): Experimenters often give the special fissile
materials with which they work highly unusual physico-chemical forms and
properties; that in fact is their job, but one consequence is that the materials
are difficult to sample and analyse and sometimes very costly to recover.
I should be interested to know what practices your installations have adopted
with a view to accounting for lots of material that have been subjected to
such changes.

W.J. WRIGHT: It is true that the A.A.E.C. programme requires the
production of materials of complicated structure, which often involve mix-
tures of fissile and fertile material in the same sample. In many instances,
therefore, we have been obliged to adopt a dual system of accounting to take
care of both types of material. This does, of course, complicate accounting
and the problem of estimating the content of materials, particularly where
chemical recovery is contemplated; but doubtless the problem is not pe-
culiar to Australia's installations.

R.G. CARDWELL: We have found that, in cases where research quan-
tities are issued for a specific short-term job rather than for a continuing
project, the research workers are usually glad to avoid accountability
record-keeping by saving all materials, in whatever shape or form results
from the experiment, and returning them all together for reclamation or
disposal. Our experience has shown that this method considerably reduces
the loss factor on research quantities of nuclear material.

W.J. WRIGHT: Yes, I would certainly not encourage research workers
to make continuous transfers of material to the recovery stores during the
course of their projects. For one thing, they might want to repeat some
experiments or measurements. Moreover, continuous movements of small
quantities of material can lead to an accumulation of errors. The best way
of achieving an accurate balance is to return all the material when the pro-
ject is finished; the material returned can then be assessed against the known
input and residue balances.
ACCOUNTING SYSTEMS FOR HEAVY WATER AND FISSIONABLE MATERIALS

G. W. FLETCHER, H. B. REID AND W. G. JENKINSON
ATOMIC ENERGY OF CANADA LTD., CHALK RIVER, ONT., CANADA

Abstract — Résumé — Аннотация — Resumen

ACCOUNTING SYSTEMS FOR HEAVY WATER AND FISSIONABLE MATERIALS. Detailed accounting and reporting procedures used by Atomic Energy of Canada Limited (AECL) for maintaining adequate records and control of heavy water supplies and stocks of fissionable materials are described, along with the duties and responsibilities of those administering the system. An appraisal is made of these procedures with respect to their adaptability for use in rapidly expanding research and power programmes. In particular the use of electronic data processing equipment is evaluated.

A senior management committee is responsible for ensuring that there is a proper system for recording, reporting and controlling fissionable materials. The Production Planning and Control Branch (PP and CB) of the Operations Division at the Chalk River Nuclear Laboratories (CRNL) is responsible to the committee for keeping the over-all records and for the general administration of the system. The duties involved are detailed in the report.

The system for fissionable materials is segregated into several accountability units 15 of which are allocated to AECL departments and the others to Canadian industries and research organizations. A control ledger is kept by PP and CB for each of the units; however, the units are responsible for preparing detailed accounts of all material under their jurisdiction. The basic recording procedures covering the movement of materials between units, the changing of forms within units, the handling of gains and losses, and disposals, are outlined in the report. The transfer of this data to IBM cards, the ultimate processing through an IBM 1401 computer and the preparation of reports for management approval are described.

The heavy-water accounting system based on the same principles as used for the fissionable materials is explained. In this case the control ledger lists the pounds of D$_2$O allocated to each of the 15 accountability units. Again the basic recording methods and the use of a computer system are outlined.
1. INTRODUCTION

Atomic Energy of Canada Limited (AECL) has as its objectives the advancement of fundamental research in the field of nuclear science, the de-

registre de contrôle le poids de D₂O attribué à chacune des 15 sections comptables. La encore, les auteurs exposent les principales méthodes comptables et décrivent l'utilisation de la calculatrice.

SISTEMA DE CONTABILIDAD PARA EL AGUA PESADA Y LOS MATERIALES FISIONABLES. Los autores describen detalladamente los procedimientos de contabilidad e información que emplea la Atomic Energy of Canada Limited (AECL) para mantener registros adecuados y ejercer un control eficiente de los suministros de agua pesada y de las existencias de materiales fisionables, y exponen los deberes y las responsabilidades de las personas encargadas de la fiscalización. Evalúan estos procedimientos desde el punto de vista de su adaptabilidad a programas de investigación y de producción de energía en rápido desarrollo y tratan en particular del empleo de equipo electrónico para la sistematización de los datos.

Un comité administrativo superior vela para que se aplique un sistema adecuado de registro y fiscalización de los materiales nucleares. La Sección de Planeamiento y Control de la Producción de la División de Operaciones de los Laboratorios Nucleares de Chalk River (CRNL) responde ante dicho comité del mantenimiento de los registros y del buen funcionamiento del sistema. En la memoria se exponen detalladamente sus funciones.

Para los materiales fisionables, el sistema consta de varias unidades contables, quince de las cuales corresponden a departamentos de la AECL y las demás a industrias y centros de investigación del Canadá. La Sección de Planeamiento y Control de la Producción lleva un libro mayor para cada una de las unidades, pero éstas contabilizan detalladamente todos los materiales que se hallan bajo su jurisdicción. En la memoria se exponen a grandes rasgos los procedimientos básicos para el registro de las transferencias de materiales entre las unidades, el intercambio de formularios dentro de cada unidad, las pérdidas y ganancias y las evacuaciones. Se explica también cómo estos datos pasan a fichas IBM, su sistematización en una calculadora IBM 1401, y la preparación de los informes que se someterán a la aprobación de las autoridades administrativas.

El sistema de contabilidad para el agua pesada está basado en esos mismos principios. En el libro mayor se inscriben las libras de D₂O asignadas a cada una de las quince unidades contables. Los autores describen también sucintamente los métodos básicos de registro y la forma en que se emplea la computadora.
velopment of economic nuclear power and the production and sale of radio-isotopes. The various AECL establishments are shown in Appendix I. Chalk River Nuclear Laboratories (CRNL) is the oldest and largest establishment. Two high-flux reactors, NRX and NRU, provide the main irradiation facilities for research and isotope production; NRX being in operation since 1947. A second research and development centre is being established at Whiteshell, Manitoba, where an organic-cooled experimental reactor is scheduled for operation in the autumn of 1965. All of these reactors require heavy water as a moderator and enriched uranium for fuel or spiking.

In the nuclear power field, a 20-MW(e) power demonstration reactor (NPD) has been in operation at Rolphton, Ontario since 1962. A second station with CANDU-PHW 200-MW(e) reactor, designed by Power Projects and located at Douglas Point, Ontario, is scheduled for operation early in 1966. Another 1000-MW(e) nuclear plant will be built at Pickering, near Toronto, for operation in 1970-71. All of these reactors require natural uranium as fuel and heavy water for the moderator and the coolant.

Accounting systems for heavy water and fissionable materials have had to be adjusted to cope with the expanding programmes without increasing staff and to provide the necessary controls for safety and economy over broader areas.

2. REQUIREMENTS OF AECL'S ACCOUNTING SYSTEMS

2.1. General

At present there is in inventory 310,719 kg of heavy water valued at $17,300,000, along with 4100 kg of leased enriched uranium containing 225 kg of \(^{235}\text{U}\) valued at $2 million and 170,000 kg of natural uranium. The accounting systems must meet the requirements of organizations concerned with the criticality, health, operational and financial control of these special nuclear materials. The major groups are listed below.

2.2. Atomic Energy Control Board

This corporate regulatory body is directly responsible to the Canadian Government for controlling and supervising all aspects of development, application and use of atomic energy in Canada. Up-to-date records and procedures must meet all the requirements of this body to enable them to carry out their duties.

2.3. United States Atomic Energy Commission (USAEC)

The enriched uranium used in the research reactors is leased from the above organization. Regular reports on the use and distribution of this fissionable material must be submitted in a form compatible with the accounting system employed by the Division of International Affairs. Thus the AECL system must be capable of producing data quickly in a suitable form.
2.4. CRNL Divisions

Suitable data must be produced for planning, budget control, experimental records and operational control. The nuclear materials inventory has to be presented in accordance with the accounting principles laid down by the Finance Division in other precious materials.

3. ADMINISTRATION OF THE ACCOUNTING SYSTEMS

3.1. Fissionable materials

Due to the large number of groups involved in the use of these materials, a Fissionable Material Accountability Committee was appointed by Senior Management to see that adequate records were kept and that all requirements of the Atomic Energy Control Act were met. The members represent the Finance, Administration, Radiation and Industrial Safety, Operations, Engineering and Research Divisions.

The Production Planning and Control Branch of the Operations Division is responsible to the above Committee for keeping the necessary accounts. The Branch acts as secretary to the Committee, implements the decisions and operates the overall system.

There are 12 fissionable material accounts (Appendix III) and the entire inventory is segregated into material balance areas called, "accountability units". Twelve of these units are at CRNL while 27 others are located at other AECL establishments, contractor's plants, universities and other research centres.

The Production Planning and Control Branch maintains an overall ledger on all materials owned or leased by AECL and records the quantities in the 12 separate categories or accounts. In addition, a control ledger is maintained for each accountability unit in the same manner as for the overall record.

The designated head of each accountability unit is responsible for selecting and maintaining a detailed recording system for all fissionable material held by that unit. He is also responsible for the physical control of all material in his area.

In the handling of heavy water fewer groups are involved and so no committee is required for assisting in the administration of the accounting system. The Production Planning and Control Branch carries out the procedures as outlined above for the fissionable materials as directed by CRNL Management. There are only 15 accountability units involved.

4. RECORDING PROCEDURES

4.1. Preparation of vouchers

Formal vouchers are issued to cover all transactions involving special nuclear materials. Each accountability unit forwards the basic information to the Production Planning and Control Branch who prepares the document
for appropriate signatures. The voucher is the only authority by which changes can be made to ledgers and detailed records.

Transactions covered by the issuance of vouchers include the following: (a) transfers between accountability units, (b) gains and losses of materials, and (c) changes from one fissionable material account to another. The information recorded on the vouchers and subsequently on the ledgers is received as follows:

(a) Off Project: Shippers and receivers issue memoranda recording the weights of material shipped and received. The shippers weights are accepted by the receiver if the check is within 100 grams. When agreement has been reached the official voucher is issued to the parties concerned and the ledger adjusted.

(b) At CRNL: Requests for transfer are forwarded to the Production Planning and Control Branch giving weights, form, etc., of material to be transferred. After checking the requests and requesting more information if it is required the official voucher is issued and ledgers adjusted.

The accuracy of the information varies, of course, with the type of material involved. Where possible, weight and other checks are carried out. In the case of pure material the accuracy is estimated to be between $\frac{1}{2}$ and 1%. The error, however, may be in the order of several per cent or more in the case of scrap and other impure materials.

4.2. Introduction of electronic data processing

As AECL's programme expanded through the years the quantity of material to be accounted for, and the number of individual transactions, increased greatly. The Branch had only ten men engaged in the accounting work. They were having difficulty in keeping the records up to date and a great deal of overtime work was necessary. In the summer of 1963 a full-scale study of the system was initiated.

The study revealed the following facts:
(a) Considerable time was being spent on hand calculations for preparing routine reports. This caused delays of up to two weeks in performing normal recording procedures.
(b) Month-end reports could be issued on time only if overtime was worked.
(c) Special reports for management could only be prepared on the same basis as in (b).
(d) There was insufficient time to train personnel in new work. Consequently, errors occurred which were time-consuming.
(e) Reconciliations between ledgers and accountability unit monthly reports were not being performed on a regular basis due to the pressure of routine work. Discrepancies between the various ledgers were occurring, causing loss of control.

On the basis of this survey it was decided to introduce some form of electronic data processing into the fissionable-material and heavy-water accounting systems. Before the study, the only machine accounting being done was the detailed listing of fuel rods in the NRX and NRU reactors from
punched cards run through IBM payroll equipment which consisted of an IBM-407-10 for listing and sorting and an IBM 602-A for calculating. At this time, however, an IBM 1401 computer was installed at CRNL. The new machine is four times faster in its listing and tabulation function and ten times faster in its calculating function. This permitted much wider use of the machine and at the same time made the scheduling of reports easier. Thus procedures were developed for this machine according to the following plan: (a) The programmes for the fissionable material and heavy water would be worked out separately to a successful implementation into the system, (b) Both the new and the old systems would be carried out in parallel until all difficulties had been cleared and the staff had been trained to use the new system with confidence.

4.3. Maintaining control ledgers using the computer

Cards were set up to record the information from the formal vouchers. Programmes have been developed to, (a) print out detailed ledger sheets for each accountability unit showing the opening balance, the transactions for the period and the closing balance, (b) print out a summary sheet showing the total amounts held by the units.

Appendix II shows a detailed sheet for the fissionable materials held in five different accounts by one of the accountability units. The detailed information includes transfer number, date, quantity and general description of the material. Appendix III contains a summary sheet showing the quantity of material held in the 12 accounts by each unit. Appendix IV is an example of a detailed ledger sheet for heavy-water inventory records.

With the new procedure information from each voucher is transferred to an IBM card. The cards are assembled once a month and run through the computer. The detailed print-out is set in the official ledger books while the summary sheet is used in the preparation of monthly reports. Runs can be made more frequently if required.

4.4. Recording system for materials in storage

The Production Planning and Control Branch is responsible for the storage of special nuclear materials not in current use by the accountability units. A computer programme has been established for keeping storage records. The following operations are carried out for both the heavy-water and fissionable materials systems:

(a) The IBM cards are prepared from the formal vouchers covering the receipts and issues of materials.
(b) The print-out is used as the detailed inventory listing of the quantities of materials in storage assigned to the various accountability units.
(c) In the case of fissionable materials the cards are sorted according to their 12 accounts as well as their chemical and physical form.

Appendix V gives a typical detailed listing of fissionable materials in storage. The following information is outlined:

(a) Fissionable material account
(b) Chemical form
Appendix VI shows a typical heavy-water storage print-out sheet with the following details:

(a) Grade of heavy water
(b) Drum number
(c) Gross, tare and net weights
(d) Per cent D$_2$O
(e) Weight of D$_2$O
(f) Location in storage
(g) pH and conductivity
(h) Permanganate demand
(i) Beta-gamma activity
(j) Tritium content
(k) Transfer or voucher number
(l) Date.

4.5. Recording system for reactor fuel inventories

The programme, used to record the irradiation data for the fuel in the NRX and NRU reactors, calculates the MWd output of the rods and provides a detailed listing of the fissionable material according to its location within the reactor accountability unit. The following procedure is carried out:

(a) The reactor-physics office assigns a heat output factor to each fuel rod position based on the reactor loading.
(b) Each time the reactor is shut-down and the loading changed, a new set of heat output factors is issued.
(c) IBM cards are prepared for each fuel rod in the reactor listing the total power to date (TPD) and the heat output factor at the start of irradiation.
(d) When the heat output factors change, the TPD at that time is punched on each card. The cards are then run through the computer to determine the MWd output of each rod.
(e) A new card is prepared for each rod showing the previous MWd output, the TPD at start-up and the new heat output factor.
(f) Items (d) and (e) are repeated as required.
(g) A new card is prepared for any rod removed from the reactor showing the change in location from the reactor to the irradiated rod storage, and the final MWd output.

An IBM card for each fuel rod within the reactor accountability unit is also maintained. This card outlines the location, the fissionable material content and the irradiation history. Once a month the cards are sorted by location and rod number for the NRX and NRU accountability units. A detailed rod listing is made. The print-outs serve as the record of fissionable material within these units.
Appendix VII is a sample of these detailed listings. It contains the following information:

(a) Rod location
(b) Assembly type
(c) Rod series and number
(d) Quantity of fissionable material
(e) Insertion date in reactor
(f) Insertion TPD
(g) Reactor position
(h) MWd output of previous rod
(i) TPD at start, TPD at finish and f-factor
(j) Present rod MWd
(k) Pu content
(l) Removal date.

With the increased use of enriched uranium fuel in the research reactors it became necessary to adapt the computer system for the $^{235}\text{U}$ burn-up calculations in order to prepare regular reports for the Finance Division and the USAEC. A burn-up formula based on the MWd output of each rod was included in the programme. Appendix VIII illustrates the type of data recorded. The following details are printed out:

(a) Rod number
(b) Rod description
(c) Location
(d) Initial grams uranium
(e) Initial grams uranium-235
(f) Initial wt. % uranium-235
(g) Uranium content after previous run off
(h) Uranium-235
(i) MWd to date
(j) Uranium burn-up for period
(k) Uranium-235 burn-up for period
(m) Total uranium burn-up to date
(n) Total uranium-235 burn-up to date
(o) Present uranium content
(p) Present uranium-235 content
(q) Present wt. % U
(r) Present per cent burn-up

4.6. General comments on the new recording system

The complete transition from the manual system to the computer took place within a year and went remarkably smoothly. Difficulties with the fissionable-material and heavy-water accounting systems as listed above were eliminated and the results may be summarized as follows:

(a) With the manual system some 200 fissionable-material transfers per month were being made resulting in more than 500 manual postings to the control ledger. For the heavy-water system some 100 transfers per month were being made with 300 manual postings in the overall ledger. By having the computer print out the ledger sheets we have reduced the man-
hours per month from 130 to 25 despite a 30% increase in transfers, and eliminated a large number of errors in posting.

(b) Month-end reports can now be prepared quickly from the processed records. This has resulted in a more even work load over the month. Staff are more readily available for special work assignments, and for replacement of personnel on leave.

(c) Due to less routine work, the staff are now able to spend more time on the control aspects of fissionable-material and heavy-water accounting. Reports from contractors, and from AECL establishments (other than at CRNL), have been reconciled routinely against the control ledgers. Discrepancies have been readily resolved and the reports and systems used can be more closely scrutinized.

(d) An expanded accounting programme can now be carried on without any difficulty. The system provides a greater flexibility for preparing special reports. Information can now be obtained from the original transaction cards and analysed automatically on the computer.

5. REPORTING AND CONTROL PROCEDURES

5.1. Fissionable materials

Appendix IX lists the reports prepared each month. They are issued in accordance with the control requirements specified in section 2 above. The information contained in reports 1 to 4 is described in section 4. Report 5 is prepared by each accountability unit outside CRNL and gives a detailed listing of all material under their control. Report 6 is issued by all contractors following the completion of their work on any specific programme. It constitutes a complete balance sheet on all fissionable material assigned to them listing the quantities in the product and scrap and giving a complete statement of their losses.

All reports are checked against the overall summary report prepared on the computer. An additional check is made on the overall inventory by keeping a manual ledger which gives a running total of all material owned or leased by AECL.

The major accounts at CRNL are verified each month; the minor accounts are reported monthly but are only verified on a quarterly basis. For this purpose a copy of report 1 is circulated to the unit head asking certification for the amounts shown.

Losses must be fully explained by the accountability units before a gain/loss voucher is prepared. Requests for disposal are handled in a similar manner. Vouchers, when signed by the unit head, are submitted for approval to a sub-group of the Accountability Committee having representatives from the Operations, Finance and Health Physics Divisions. In addition, any major disposal must be approved by the General Manager, Works.

Auditing of records and stocks is carried out routinely by the Finance Division and the Production Planning and Control Branch. These audits are made at least once a year beginning in February for completion by the end of the fiscal year on March 31. In addition this may be also done at
any time by auditors from the Federal Government, the Atomic Energy Control Board or the USAEC (leased material).

5.2. Heavy water

Appendix X lists the heavy-water reports submitted on a monthly basis. Reports 1 and 2 are prepared on the computer and contain listings of the material held by the various accountability units and in storage examples of which are shown in Appendices 4 and 6. Report 3 is a summary prepared manually from reactor reports based on physical inventories wherever possible. Reports 4 and 5 are summaries prepared for inventory control and planning.

The summary reports for all units are checked against an overall ledger which shows the total quantity of heavy water owned or leased by AECL. Each month vouchers are issued to cover losses from each accountability unit as recorded in report 5 which is submitted to senior management.

Auditing of records and stocks is carried out routinely by the Finance Division and the Production Planning and Control Branch as is done for the fissionable materials.

6. CONCLUSIONS

In summary, the fissionable material and heavy water accounting systems meet the requirements of the Atomic Energy Control Board, the USAEC and AECL's senior management.

The recording system has been brought up-to-date within the last year and is now capable of handling increased data without difficulty. Reporting procedures have improved as a result of this more efficient system. Routine and special reports are now issued on schedule without overtime work despite an increase of 30% in the work load. Controls on the system have become more effective. It is hoped that these will become even tighter, especially in the policing of fissionable material losses at contractor's plants.

It should be noted that while the basic requirements of a system are constant, namely, to provide a system of accounting and control to satisfy safety, regulatory and economic requirements, continual evaluation of the detailed system is necessary to take care of changing programmes. This study is also necessary to ensure that errors or inadequacies are detected and corrected, bearing in mind that over-control can be almost as serious as under-control. This is particularly true when the materials are being used in a research and development programme which is constantly changing.
### APPENDIX II

**PP & CB CONTROL LEDGER ON WE & P ACCOUNTABILITY UNIT FOR THE MONTH OF JANUARY, 1965**

#### FISSIONABLE MATERIAL LEDGER

<table>
<thead>
<tr>
<th>MATERIAL</th>
<th>REFERENCE NUMBER</th>
<th>DATE</th>
<th>PARTICULARS</th>
<th>RECEIPTS</th>
<th>ISSUES</th>
<th>BALANCE</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>NAT U -KG-</strong></td>
<td></td>
<td>31/12/4</td>
<td>BROUGHT FORWARD</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>TRSF. 9370</td>
<td>06/01/5</td>
<td></td>
<td>3 NRU NAT UO2 F/M FUEL RODS IN 1 ASSEMBLY</td>
<td>0.9</td>
<td>0.9</td>
<td>0.9</td>
</tr>
<tr>
<td>TRSF. 9307</td>
<td>01/01/5</td>
<td></td>
<td>TWO SURPLUS FLATS TO PMS AS SCRAP FOR RECOVERY</td>
<td>0.8</td>
<td>0.6</td>
<td>0.2</td>
</tr>
<tr>
<td>TRSF. 9400</td>
<td>21/01/5</td>
<td></td>
<td>1 PK 111 FAST NEUTRON TRANSFORMER ROD</td>
<td>27.2</td>
<td>27.2</td>
<td>0.0</td>
</tr>
<tr>
<td><strong>ENR U -KG-</strong></td>
<td></td>
<td>31/12/4</td>
<td>BROUGHT FORWARD</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>TRSF. 9373</td>
<td>08/01/5</td>
<td></td>
<td>THREE ONLY CONCENTRIC U/AL ALLOY TUBES</td>
<td>1.0</td>
<td>1.1</td>
<td>0.1</td>
</tr>
<tr>
<td>TRSF. 9366</td>
<td>04/01/5</td>
<td></td>
<td>FOUR CONCENTRIC U/AL ALLOY TUBES</td>
<td>1.2</td>
<td>1.0</td>
<td>0.2</td>
</tr>
<tr>
<td>TRSF. 9377</td>
<td>11/01/5</td>
<td></td>
<td>TWO CONCENTRIC U/AL ALLOY TUBES</td>
<td>2.7</td>
<td>0.8</td>
<td>1.9</td>
</tr>
<tr>
<td>TRSF. 9365</td>
<td>01/01/5</td>
<td></td>
<td>ENR U/AL ELEMENTS FOR FAB OF NRU 12-EL ENR RODS</td>
<td>1.0</td>
<td>1.0</td>
<td>0.0</td>
</tr>
<tr>
<td>TRSF. 9381</td>
<td>12/01/5</td>
<td></td>
<td>FIVE 12-EL NRU ENR FUEL RODS</td>
<td>1.1</td>
<td>1.1</td>
<td>0.0</td>
</tr>
<tr>
<td>TRSF. 9380</td>
<td>12/01/5</td>
<td></td>
<td>ENR U/AL ELEMENTS FOR FAB OF NRU 12-EL ENR RODS</td>
<td>6.7</td>
<td>6.3*</td>
<td>0.4*</td>
</tr>
<tr>
<td>TRSF. 9369</td>
<td>05/01/5</td>
<td></td>
<td>THREE 12-EL NRU ENR FUEL RODS</td>
<td>6.2*</td>
<td>5.8*</td>
<td>0.4*</td>
</tr>
<tr>
<td>TRSF. 9413</td>
<td>21/01/5</td>
<td></td>
<td>FOUR CONCENTRIC TUBES FAB INTO RODS FOR NRU</td>
<td>2.4</td>
<td>2.4*</td>
<td>0.0</td>
</tr>
<tr>
<td>TRSF. 9399</td>
<td>18/01/5</td>
<td></td>
<td>FIVE CONCENTRIC U/AL ALLOY TUBES</td>
<td>2.4*</td>
<td>2.4*</td>
<td>0.0</td>
</tr>
<tr>
<td>TRSF. 9399</td>
<td>19/01/5</td>
<td></td>
<td>FIVE ELEMENT NRU ENRICHED FUEL RODS</td>
<td>2.4*</td>
<td>2.4*</td>
<td>0.0</td>
</tr>
<tr>
<td><strong>CSP U -KG-</strong></td>
<td></td>
<td>31/12/4</td>
<td>BROUGHT FORWARD</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>TRSF. 9425</td>
<td>29/01/5</td>
<td></td>
<td>DEPLETED UO2 PELLETS SWEATED IN ZIRCALOY</td>
<td>2.4</td>
<td>2.4*</td>
<td>0.0</td>
</tr>
<tr>
<td><strong>PLUTONIUM -G-</strong></td>
<td></td>
<td>31/12/4</td>
<td>BROUGHT FORWARD</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>TRSF. 9373</td>
<td>08/01/5</td>
<td></td>
<td>THREE ONLY CONCENTRIC U/AL ALLOY TUBES</td>
<td>614.0</td>
<td>614.0</td>
<td>0.0</td>
</tr>
<tr>
<td>TRSF. 9366</td>
<td>04/01/5</td>
<td></td>
<td>FOUR CONCENTRIC U/AL ALLOY TUBES</td>
<td>628.4</td>
<td>628.4</td>
<td>0.0</td>
</tr>
<tr>
<td>TRSF. 9367</td>
<td>11/01/5</td>
<td></td>
<td>TWO CONCENTRIC U/AL ALLOY TUBES</td>
<td>311.6</td>
<td>311.6</td>
<td>0.0</td>
</tr>
<tr>
<td>TRSF. 9365</td>
<td>01/01/5</td>
<td></td>
<td>ENR U/AL ELEMENTS FOR FAB OF NRU 12-EL ENR RODS</td>
<td>824.2</td>
<td>824.2</td>
<td>0.0</td>
</tr>
<tr>
<td>TRSF. 9381</td>
<td>12/01/5</td>
<td></td>
<td>FIVE 12-EL NRU ENR FUEL RODS</td>
<td>1,030.0</td>
<td>1,030.0</td>
<td>0.0</td>
</tr>
<tr>
<td>TRSF. 9376</td>
<td>08/01/5</td>
<td></td>
<td>ONE ENRICHED U/AL ELEMENT</td>
<td>1,080.0</td>
<td>1,080.0</td>
<td>0.0</td>
</tr>
<tr>
<td>TRSF. 9380</td>
<td>12/01/5</td>
<td></td>
<td>ENR U/AL ELEMENTS FOR FAB OF NRU 12-EL ENR RODS</td>
<td>619.0</td>
<td>619.0</td>
<td>0.0</td>
</tr>
<tr>
<td>TRSF. 9369</td>
<td>05/01/5</td>
<td></td>
<td>THREE 12-EL NRU ENR FUEL RODS</td>
<td>2,671.5</td>
<td>2,671.5</td>
<td>0.0</td>
</tr>
<tr>
<td>TRSF. 9399</td>
<td>18/01/5</td>
<td></td>
<td>FIVE CONCENTRIC TUBES FAB INTO RODS FOR NRU</td>
<td>620.4</td>
<td>620.4</td>
<td>0.0</td>
</tr>
<tr>
<td>TRSF. 9401</td>
<td>18/01/5</td>
<td></td>
<td>THREE 12-EL NRU ENR FUEL RODS TO NRU REACTOR</td>
<td>619.0</td>
<td>619.0</td>
<td>0.0</td>
</tr>
<tr>
<td>TRSF. 9399</td>
<td>19/01/5</td>
<td></td>
<td>FIVE CONCENTRIC U/AL ALLOY TUBES</td>
<td>31.6</td>
<td>31.6</td>
<td>0.0</td>
</tr>
<tr>
<td>TRSF. 9476</td>
<td>29/01/5</td>
<td></td>
<td>THREE 12-EL NRU ENR RODS FOR REWORK</td>
<td>1,030.0</td>
<td>1,030.0</td>
<td>0.0</td>
</tr>
<tr>
<td>TRSF. 9399</td>
<td>19/01/5</td>
<td></td>
<td>FIVE 12 ELEMENT NRU ENRICHED FUEL RODS</td>
<td>5,704.5*</td>
<td>5,704.5*</td>
<td>0.0</td>
</tr>
</tbody>
</table>

*Note: BALANCE figures are rounded to the nearest whole number.*
## APPENDIX VIII

### SUMMARY OF FISSIONABLE MATERIAL AT 31 JANUARY 1965
FOR ACCOUNTABILITY UNITS AT CRNL

#### ATOMIC ENERGY OF CANADA LIMITED

#### SUMMARY OF FISSIONABLE MATERIAL INVENTORY
AS AT JAN. 31, 1965

<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>BIOLOGY</td>
<td>6.6</td>
<td>103.0</td>
<td>.4</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>CHEMICAL OPERATIONS</td>
<td>84.6</td>
<td>254.8</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>CHEMISTRY</td>
<td>82.0</td>
<td>2.6</td>
<td>4.1</td>
<td>1.3</td>
<td>56.9</td>
<td>14.5</td>
<td>127.8</td>
<td>83.2</td>
<td>3.3</td>
<td>5.2</td>
<td>24.8</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>F. N. C. S.</td>
<td>3450.6</td>
<td>-</td>
<td>631.6</td>
<td>.2</td>
<td>6277.9</td>
<td>4192.4</td>
<td>32964.5</td>
<td>205.7</td>
<td>650.6</td>
<td>-</td>
<td>265.2</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>N. R. X. REACTOR</td>
<td>583.2</td>
<td>6663.6</td>
<td>2.5</td>
<td>529.8</td>
<td>169.1</td>
<td>2198.5</td>
<td>2065.3</td>
<td>34756.9</td>
<td>197.1</td>
<td>399.1</td>
<td>213.9</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>N. R. U. REACTOR</td>
<td>217.6</td>
<td>925.2</td>
<td>5.6</td>
<td>41.3</td>
<td>220.7</td>
<td>5055.6</td>
<td>22803.9</td>
<td>235.0</td>
<td>571.5</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>PHYSICS</td>
<td>55.4</td>
<td></td>
<td>52.9</td>
<td>20.4</td>
<td>3.4</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>REACTOR RESEARCH</td>
<td>39034.4</td>
<td>2.7</td>
<td>10.7</td>
<td>1779.6</td>
<td>16.9</td>
<td>6390.5</td>
<td>3.1</td>
<td>3.7</td>
<td>2.2</td>
<td>59.6</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>W. E. &amp; P.</td>
<td>247.2</td>
<td>6.2</td>
<td>45.9</td>
<td>1.1</td>
<td>2949.9</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>ENGINEERING 1</td>
<td>912.8</td>
<td>231.6</td>
<td>8.9</td>
<td>330.3</td>
<td>124.8</td>
<td>40.3</td>
<td>2830.9</td>
<td>3701.2</td>
<td>8.3</td>
<td>9.7</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>ENGINEERING 2</td>
<td>.2</td>
<td></td>
<td>3.9</td>
<td>.3</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>METALLURGY</td>
<td>146.9</td>
<td>35.8</td>
<td>44.1</td>
<td>30.0</td>
<td>1.6</td>
<td>1.9</td>
<td>570.3</td>
<td>1069.6</td>
<td>707.1</td>
<td>16.1</td>
<td>2.2</td>
<td>.4</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>TOTALS</td>
<td>44736.7</td>
<td>7946.1</td>
<td>713.7</td>
<td>740.9</td>
<td>8286.9</td>
<td>211.3</td>
<td>7566.0</td>
<td>53474.8</td>
<td>62261.1</td>
<td>1117.9</td>
<td>977.8</td>
<td>573.6</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

### Notes
- The table above provides a summary of fissionable material inventory as at January 31, 1965, for accountability units at CRNL.
- The units are provided in kilograms (kg) and grams (g).
## APPENDIX IV

**PP & CB HEAVY-WATER CONTROL LEDGER**  
**ON CHEMICAL OPERATIONS ACCOUNTABILITY UNIT**  
**FOR THE MONTH OF JANUARY, 1965**

### HEAVY WATER LEDGER

**CHEM. OPS. - CLEANING**

**AS AT JAN. 31, 1965**

<table>
<thead>
<tr>
<th>Reference Number</th>
<th>Date D M Y</th>
<th>PARTICULARS</th>
<th>Receipts</th>
<th>Issues</th>
<th>Balance</th>
</tr>
</thead>
<tbody>
<tr>
<td>31/12/4</td>
<td></td>
<td>Brought Forward</td>
<td></td>
<td></td>
<td>2,988.4</td>
</tr>
<tr>
<td>6499 04/01/5</td>
<td>2 DRUMS</td>
<td>548.7</td>
<td></td>
<td>1,904.9</td>
<td></td>
</tr>
<tr>
<td>6501 04/01/5</td>
<td>3 DRUMS</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>6504 05/01/5</td>
<td>1 DRUM</td>
<td>30.5</td>
<td></td>
<td>990.7</td>
<td></td>
</tr>
<tr>
<td>6505 05/01/5</td>
<td>2 DRUMS</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>6509 07/01/5</td>
<td>3 DRUMS</td>
<td>480.1</td>
<td></td>
<td>1,338.2</td>
<td></td>
</tr>
<tr>
<td>6512 07/01/5</td>
<td>5 DRUMS</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>6518 12/01/5</td>
<td>1 DRUM</td>
<td>504.0</td>
<td></td>
<td>180.1</td>
<td></td>
</tr>
<tr>
<td>6523 15/01/5</td>
<td>1 DRUM &amp; CARBOY</td>
<td>40.1</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>6524 15/01/5</td>
<td>1 DRUM</td>
<td></td>
<td></td>
<td>180.1</td>
<td></td>
</tr>
<tr>
<td>6528 18/01/5</td>
<td>1 DRUM &amp; CARBOY</td>
<td>542.1</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>6529 18/01/5</td>
<td>3 DRUMS</td>
<td>458.9</td>
<td></td>
<td>454.5</td>
<td></td>
</tr>
<tr>
<td>6531 20/01/5</td>
<td>1 DRUM</td>
<td></td>
<td></td>
<td>413.4</td>
<td></td>
</tr>
<tr>
<td>6532 20/01/5</td>
<td>1 DRUM</td>
<td></td>
<td></td>
<td>413.4</td>
<td></td>
</tr>
<tr>
<td>6538 21/01/5</td>
<td>2 DRUMS</td>
<td>633.4</td>
<td></td>
<td>823.6</td>
<td></td>
</tr>
<tr>
<td>6547 25/01/5</td>
<td>3 DRUMS</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>6548 26/01/5</td>
<td>3 DRUMS</td>
<td>552.5</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>6552 28/01/5</td>
<td>1 DRUM</td>
<td>453.5</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>6554 28/01/5</td>
<td>2 DRUMS AND CARBOY</td>
<td>877.0</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>6559 29/01/5</td>
<td>ADJUSTMENT FOR UP-GRADING</td>
<td>44.0</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

**ADJUSTMENT FOR UP-GRADING**

<table>
<thead>
<tr>
<th>Reference Number</th>
<th>Date D M Y</th>
<th>PARTICULARS</th>
<th>Receipts</th>
<th>Issues</th>
<th>Balance</th>
</tr>
</thead>
<tbody>
<tr>
<td>6559 29/01/5</td>
<td>ADJUSTMENT FOR UP-GRADING</td>
<td>44.0</td>
<td>5,850.3</td>
<td>1,844.0</td>
<td></td>
</tr>
</tbody>
</table>

**APPENDIX VIII**

**PP & CB HEAVY-WATER CONTROL LEDGER**  
**ON CHEMICAL OPERATIONS ACCOUNTABILITY UNIT**  
**FOR THE MONTH OF JANUARY, 1965**
## APPENDIX V

### PAGE FROM PP & CB FISSIONABLE MATERIAL STORAGE

#### ACCOUNTABILITY UNIT INVENTORY

<table>
<thead>
<tr>
<th>CHEN</th>
<th>PHYS</th>
<th>FISSILE MATERIAL</th>
<th>WEIG WT</th>
<th>STORED</th>
<th>TYPE</th>
<th>ISO-OP</th>
<th>ENRICH</th>
<th>REASON</th>
<th>TRSF.</th>
<th>REF.</th>
<th>REC.</th>
<th>NUMBER</th>
<th>NO.</th>
<th>POSN</th>
<th>REMARKS</th>
</tr>
</thead>
<tbody>
<tr>
<td>03</td>
<td>03</td>
<td>148.6</td>
<td>93.19</td>
<td>T</td>
<td>FFR 416</td>
<td>13</td>
<td>14/06/3</td>
<td>MAU OTER TUBE 845/3</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>03</td>
<td>01</td>
<td>202.5</td>
<td>93.19</td>
<td>T</td>
<td>FFR 416</td>
<td>11/07/2</td>
<td>MAU OTER TUBE 844/3</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>03</td>
<td>01</td>
<td>200.9</td>
<td>93.19</td>
<td>T</td>
<td>FFR 416</td>
<td>17/12/3</td>
<td>MAU OTER TUBE 844/1</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>03</td>
<td>01</td>
<td>201.3</td>
<td>93.19</td>
<td>T</td>
<td>FFR 416</td>
<td>17/12/8</td>
<td>MAU OTER TUBE 844/3</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>03</td>
<td>01</td>
<td>202.8</td>
<td>93.19</td>
<td>T</td>
<td>FFR 416</td>
<td>12/08/8</td>
<td>MAU OTER TUBE 844/4</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>03</td>
<td>01</td>
<td>3.2</td>
<td></td>
<td>T</td>
<td>754</td>
<td>03</td>
<td>05/01/7</td>
<td></td>
<td>MAU OTER TUBE 844/4</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>03</td>
<td>01</td>
<td>140.1</td>
<td></td>
<td>T</td>
<td>7908</td>
<td>14/11/2</td>
<td></td>
<td></td>
<td>MAU OTER TUBE 844/2</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>03</td>
<td>01</td>
<td>205.9</td>
<td>93.16</td>
<td>T</td>
<td>FFR 828</td>
<td>03</td>
<td>29/01/5</td>
<td>MAU OTER TUBE 844/3</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>03</td>
<td>01</td>
<td>205.0</td>
<td>93.16</td>
<td>T</td>
<td>FFR 828</td>
<td>03</td>
<td>29/01/5</td>
<td>MAU OTER TUBE 844/3</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>03</td>
<td>01</td>
<td>205.9</td>
<td>93.16</td>
<td>T</td>
<td>FFR 828</td>
<td>03</td>
<td>29/01/5</td>
<td>MAU OTER TUBE 844/3</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>03</td>
<td>01</td>
<td>206.0</td>
<td>93.16</td>
<td>T</td>
<td>FFR 828</td>
<td>03</td>
<td>29/01/5</td>
<td>MAU OTER TUBE 844/3</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>03</td>
<td>01</td>
<td>206.0</td>
<td>93.16</td>
<td>T</td>
<td>FFR 828</td>
<td>03</td>
<td>29/01/5</td>
<td>MAU OTER TUBE 844/3</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>03</td>
<td>01</td>
<td>153.0</td>
<td>93.19</td>
<td>T</td>
<td>TRSF 939</td>
<td>5/01/5</td>
<td></td>
<td>MAU OTR TUBE 854/2</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>03</td>
<td>01</td>
<td>154.8</td>
<td>93.19</td>
<td>T</td>
<td>TRSF 939</td>
<td>5/01/5</td>
<td></td>
<td>MAU OTR TUBE 854/2</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>03</td>
<td>01</td>
<td>154.4</td>
<td>93.19</td>
<td>T</td>
<td>TRSF 939</td>
<td>5/01/5</td>
<td></td>
<td>MAU OTR TUBE 854/2</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>03</td>
<td>01</td>
<td>157.1</td>
<td>93.19</td>
<td>T</td>
<td>TRSF 939</td>
<td>5/01/5</td>
<td></td>
<td>MAU OTR TUBE 854/2</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>03</td>
<td>04</td>
<td>140.0</td>
<td>93.15</td>
<td>T</td>
<td>FFR 740</td>
<td>03</td>
<td>28/01/4</td>
<td>MAU OTER TUBE 854/3</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>03</td>
<td>03</td>
<td>206.0</td>
<td>93.16</td>
<td>T</td>
<td>FFR 826</td>
<td>03</td>
<td>29/01/5</td>
<td>MAU OTER TUBE 854/3</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>03</td>
<td>03</td>
<td>6.3</td>
<td>90.00</td>
<td>T</td>
<td>TRSF 867</td>
<td>7/09/3</td>
<td>75470</td>
<td>MAU OTER TUBE 854/3</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>03</td>
<td>06</td>
<td>67.8</td>
<td>93.15</td>
<td>T</td>
<td>FFR 757</td>
<td>07</td>
<td>17/09/4</td>
<td>MAU OTER TUBE 854/3</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>03</td>
<td>06</td>
<td>36.6</td>
<td></td>
<td>T</td>
<td>TRSF 937</td>
<td>08/01/5</td>
<td>754819</td>
<td>MAU OTER TUBE 854/3</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>03</td>
<td>06</td>
<td>11.5</td>
<td>92.20</td>
<td>T</td>
<td>TRSF 937</td>
<td>08/01/5</td>
<td>754819</td>
<td>MAU OTER TUBE 854/3</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>03</td>
<td>06</td>
<td>124.2</td>
<td></td>
<td>T</td>
<td>MAU OTR TUBE 815/1</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>03</td>
<td>09</td>
<td>252.4</td>
<td>92.86</td>
<td>T</td>
<td>C-6024</td>
<td>20/10/9</td>
<td>75353</td>
<td>MAU OTER TUBE 854/3</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>03</td>
<td>09</td>
<td>312.5</td>
<td>92.86</td>
<td>T</td>
<td>TRSF 939</td>
<td>01/01/4</td>
<td>75353</td>
<td>MAU OTER TUBE 854/3</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>03</td>
<td>09</td>
<td>1.6</td>
<td>99.88</td>
<td>T</td>
<td>TRSF 5112</td>
<td>01/01/4</td>
<td>75353</td>
<td>MAU OTER TUBE 854/3</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>03</td>
<td>09</td>
<td>4.9</td>
<td>92.00</td>
<td>T</td>
<td>TRSF 5122</td>
<td>01/01/4</td>
<td>75353</td>
<td>MAU OTER TUBE 854/3</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>03</td>
<td>09</td>
<td>7.5</td>
<td>92.00</td>
<td>T</td>
<td>MAU OTER TUBE 854/3</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>03</td>
<td>10</td>
<td>6.0</td>
<td>92.00</td>
<td>T</td>
<td>TRSF 8253A</td>
<td>21/11/2</td>
<td>7518</td>
<td>MAU OTER TUBE 854/3</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>03</td>
<td>10</td>
<td>5.0</td>
<td>91.17</td>
<td>T</td>
<td>TRSF 8729</td>
<td>14/11/3</td>
<td>75584</td>
<td>MAU OTER TUBE 854/3</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>03</td>
<td>10</td>
<td>49.0</td>
<td>91.17</td>
<td>T</td>
<td>TRSF 8729</td>
<td>14/11/3</td>
<td>75584</td>
<td>MAU OTER TUBE 854/3</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>03</td>
<td>01</td>
<td>49.0</td>
<td></td>
<td>T</td>
<td>TRSF 9249</td>
<td>07/08/4</td>
<td>75584</td>
<td>MAU OTER TUBE 854/3</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>03</td>
<td>01</td>
<td>49.0</td>
<td></td>
<td>T</td>
<td>TRSF 9249</td>
<td>07/08/4</td>
<td>75584</td>
<td>MAU OTER TUBE 854/3</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

---

**INVENTORY OF FISSILE MATERIAL IN MFCS STORAGE**

**DATE:** 31 JANUARY 1965

**CHEMICAL FORM:**

- PHYS: PHYSICALLY FISSILE MATERIAL
- MATERIAL: MATERIAL STORED
- WEIGHT: WEIGHT STORED
- WT: WT STORED

**TYPE:**

- FORM: FORM OF MATERIAL
- PHYS: PHYSICAL FORM
- FISSILE: FISSILE MATERIAL
- MATERIAL: MATERIAL STORED
- HEIGHTS: HEIGHTS STORED

**REASON:**

- TRSF.: TRANSFER
- REASON: REASON FOR TRANSFER
- TRSF.: TRANSFER

**DATE:**

- STORAGE: STORAGE DATE
- BLDG.: BLDG. NUMBER

**REMARKS:**

- REMARKS: REMARKS
- NO.: NO.
- POSN.: POSN.
<table>
<thead>
<tr>
<th>TYPE</th>
<th>HW DRUM NO.</th>
<th>WEIGHT LOSS</th>
<th>GROSS</th>
<th>TARE</th>
<th>NET PERCENT</th>
<th>DATE</th>
<th>DDG</th>
<th>WEIGHT</th>
<th>CONDUIT - PERMANENT BETA</th>
<th>CONDUIT - PEAK BETA</th>
<th>CONDUIT - PERMANENT BETA</th>
<th>CONDUIT - PEAK BETA</th>
<th>CONDUIT - PERMANENT BETA</th>
<th>CONDUIT - PEAK BETA</th>
</tr>
</thead>
<tbody>
<tr>
<td>PRODUCTION PLANNING &amp; CONTROL BRANCH - FHC HEAVY WATER INVENTORY</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>DATE</td>
<td>DDG</td>
<td>WEIGHT</td>
<td>CONDUIT - PERMANENT BETA</td>
<td>CONDUIT - PEAK BETA</td>
<td>CONDUIT - PERMANENT BETA</td>
<td>CONDUIT - PEAK BETA</td>
<td>CONDUIT - PERMANENT BETA</td>
<td>CONDUIT - PEAK BETA</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>DATE</td>
<td>DDG</td>
<td>WEIGHT</td>
<td>CONDUIT - PERMANENT BETA</td>
<td>CONDUIT - PEAK BETA</td>
<td>CONDUIT - PERMANENT BETA</td>
<td>CONDUIT - PEAK BETA</td>
<td>CONDUIT - PERMANENT BETA</td>
<td>CONDUIT - PEAK BETA</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>DATE</td>
<td>DDG</td>
<td>WEIGHT</td>
<td>CONDUIT - PERMANENT BETA</td>
<td>CONDUIT - PEAK BETA</td>
<td>CONDUIT - PERMANENT BETA</td>
<td>CONDUIT - PEAK BETA</td>
<td>CONDUIT - PERMANENT BETA</td>
<td>CONDUIT - PEAK BETA</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>DATE</td>
<td>DDG</td>
<td>WEIGHT</td>
<td>CONDUIT - PERMANENT BETA</td>
<td>CONDUIT - PEAK BETA</td>
<td>CONDUIT - PERMANENT BETA</td>
<td>CONDUIT - PEAK BETA</td>
<td>CONDUIT - PERMANENT BETA</td>
<td>CONDUIT - PEAK BETA</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>DATE</td>
<td>DDG</td>
<td>WEIGHT</td>
<td>CONDUIT - PERMANENT BETA</td>
<td>CONDUIT - PEAK BETA</td>
<td>CONDUIT - PERMANENT BETA</td>
<td>CONDUIT - PEAK BETA</td>
<td>CONDUIT - PERMANENT BETA</td>
<td>CONDUIT - PEAK BETA</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>DATE</td>
<td>DDG</td>
<td>WEIGHT</td>
<td>CONDUIT - PERMANENT BETA</td>
<td>CONDUIT - PEAK BETA</td>
<td>CONDUIT - PERMANENT BETA</td>
<td>CONDUIT - PEAK BETA</td>
<td>CONDUIT - PERMANENT BETA</td>
<td>CONDUIT - PEAK BETA</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>DATE</td>
<td>DDG</td>
<td>WEIGHT</td>
<td>CONDUIT - PERMANENT BETA</td>
<td>CONDUIT - PEAK BETA</td>
<td>CONDUIT - PERMANENT BETA</td>
<td>CONDUIT - PEAK BETA</td>
<td>CONDUIT - PERMANENT BETA</td>
<td>CONDUIT - PEAK BETA</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>DATE</td>
<td>DDG</td>
<td>WEIGHT</td>
<td>CONDUIT - PERMANENT BETA</td>
<td>CONDUIT - PEAK BETA</td>
<td>CONDUIT - PERMANENT BETA</td>
<td>CONDUIT - PEAK BETA</td>
<td>CONDUIT - PERMANENT BETA</td>
<td>CONDUIT - PEAK BETA</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>DATE</td>
<td>DDG</td>
<td>WEIGHT</td>
<td>CONDUIT - PERMANENT BETA</td>
<td>CONDUIT - PEAK BETA</td>
<td>CONDUIT - PERMANENT BETA</td>
<td>CONDUIT - PEAK BETA</td>
<td>CONDUIT - PERMANENT BETA</td>
<td>CONDUIT - PEAK BETA</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>DATE</td>
<td>DDG</td>
<td>WEIGHT</td>
<td>CONDUIT - PERMANENT BETA</td>
<td>CONDUIT - PEAK BETA</td>
<td>CONDUIT - PERMANENT BETA</td>
<td>CONDUIT - PEAK BETA</td>
<td>CONDUIT - PERMANENT BETA</td>
<td>CONDUIT - PEAK BETA</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>DATE</td>
<td>DDG</td>
<td>WEIGHT</td>
<td>CONDUIT - PERMANENT BETA</td>
<td>CONDUIT - PEAK BETA</td>
<td>CONDUIT - PERMANENT BETA</td>
<td>CONDUIT - PEAK BETA</td>
<td>CONDUIT - PERMANENT BETA</td>
<td>CONDUIT - PEAK BETA</td>
</tr>
<tr>
<td>Location</td>
<td>Fuel Identification</td>
<td>Weight</td>
<td>Material</td>
<td>Type</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>----------</td>
<td>--------------------</td>
<td>--------</td>
<td>----------</td>
<td>------</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

**APPENDIX VII**

**PAGE FROM NRU REACTOR ACCOUNTABILITY UNIT**

**FISSIONABLE MATERIAL INVENTORY**

---

**NOTE:** The table contains detailed information about fuel identification, weight, material, and type for various locations within the reactor. Each row represents a specific location with corresponding details.
## APPENDIX VIII

### PAGE FROM PROGRAMME CALCULATING NRX AND NRU ENRICHED URANIUM AND $^{235}$U BURN-UP IN ENRICHED URANIUM FUEL RODS

**BURNUP IN ENRICHED URANIUM RODS LEASED FROM THE USAEC**  
31 DEC. 1964

<table>
<thead>
<tr>
<th>ROD NUMBER</th>
<th>DESCRIPTION</th>
<th>LOCAL. INITIAL</th>
<th>CONTENT AT 1 JUL. 1964</th>
<th>WMD</th>
<th>BURNUP FOR PERIOD</th>
<th>BURNUP TO DATE</th>
<th>PRESENT CONTENT</th>
<th>PRES- END PER- CENTU</th>
</tr>
</thead>
<tbody>
<tr>
<td>DP0016</td>
<td>NRX 7-EL</td>
<td>5 525.0</td>
<td>489.0 93.14</td>
<td>326.2</td>
<td>251.3 201.7</td>
<td>12.6 15.0</td>
<td>211.4 252.7</td>
<td>313.6 236.3</td>
</tr>
<tr>
<td>DP0018</td>
<td>NRX 7-EL</td>
<td>5 525.0</td>
<td>489.0 93.14</td>
<td>320.0</td>
<td>243.4 204.3</td>
<td>9.1 10.9</td>
<td>214.1 256.0</td>
<td>310.9 233.0</td>
</tr>
<tr>
<td>DP0022</td>
<td>NRX 7-EL</td>
<td>5 525.0</td>
<td>489.0 93.14</td>
<td>322.8</td>
<td>247.3 201.2</td>
<td>8.7 10.4</td>
<td>210.9 252.1</td>
<td>314.1 236.9</td>
</tr>
<tr>
<td>DP0023</td>
<td>NRX 7-EL</td>
<td>5 525.0</td>
<td>489.0 93.14</td>
<td>320.7</td>
<td>244.8 202.8</td>
<td>8.2 9.9</td>
<td>212.5 254.1</td>
<td>312.5 234.9</td>
</tr>
<tr>
<td>DP0025</td>
<td>NRX 7-EL</td>
<td>5 525.0</td>
<td>489.0 93.14</td>
<td>326.8</td>
<td>252.1 190.7</td>
<td>1.7 2.0</td>
<td>199.9 238.0</td>
<td>325.1 250.1</td>
</tr>
<tr>
<td>DP0027</td>
<td>NRX 7-EL</td>
<td>5 525.0</td>
<td>489.0 93.14</td>
<td>372.5</td>
<td>306.7 187.0</td>
<td>43.5 52.0</td>
<td>196.0 234.3</td>
<td>329.0 254.7</td>
</tr>
<tr>
<td>DP0028</td>
<td>NRX 7-EL</td>
<td>5 525.0</td>
<td>489.0 93.14</td>
<td>349.5</td>
<td>279.1 209.4</td>
<td>44.0 52.5</td>
<td>219.5 262.6</td>
<td>305.5 226.6</td>
</tr>
<tr>
<td>DP0029</td>
<td>NRX 7-EL</td>
<td>5 525.0</td>
<td>489.0 93.14</td>
<td>338.5</td>
<td>266.0 209.2</td>
<td>32.7 39.1</td>
<td>219.2 262.1</td>
<td>305.8 226.9</td>
</tr>
<tr>
<td>DP0030</td>
<td>NRX 7-EL</td>
<td>5 525.0</td>
<td>489.0 93.14</td>
<td>370.2</td>
<td>303.9 203.1</td>
<td>58.0 69.4</td>
<td>212.8 254.4</td>
<td>312.2 234.5</td>
</tr>
<tr>
<td>DP0031</td>
<td>NRX 7-EL</td>
<td>5 525.0</td>
<td>489.0 93.14</td>
<td>375.7</td>
<td>310.4 186.5</td>
<td>46.2 55.1</td>
<td>195.5 233.7</td>
<td>329.5 255.3</td>
</tr>
<tr>
<td>DP0034</td>
<td>NRX 7-EL</td>
<td>5 525.0</td>
<td>489.0 93.14</td>
<td>359.4</td>
<td>267.1 203.4</td>
<td>27.6 33.0</td>
<td>213.2 254.9</td>
<td>311.8 234.1</td>
</tr>
<tr>
<td>DP0035</td>
<td>NRX 7-EL</td>
<td>6 525.0</td>
<td>489.0 93.14</td>
<td>370.1</td>
<td>303.8 201.1</td>
<td>55.9 66.8</td>
<td>210.8 252.0</td>
<td>314.2 237.0</td>
</tr>
</tbody>
</table>

**3091.1 28792.8 25137.3 21888.5 2682.7 3206.9 8457.2 10111.7 22454.6 18519.6**

| FB 136 NRU 12-EL | 3 223.4  | 138.5 62.01 | 223.4  | 138.5 57.8 | 60.6 72.4 | 60.6 72.4 | 162.8 66.1 | 40.65 52.27 |
| FB 158 NRU 12-EL | 3 223.4  | 138.5 62.01 | 223.4  | 138.5 55.7 | 58.4 69.8 | 58.4 69.8 | 165.0 68.7 | 41.64 50.40 |
| FB 159 NRU 12-EL | 3 223.4  | 138.5 62.01 | 223.4  | 138.5 51.6 | 54.1 64.7 | 54.1 64.7 | 169.3 73.8 | 41.59 46.71 |
| FB 161 NRU 12-EL | 3 223.4  | 138.5 62.01 | 223.4  | 138.5 52.8 | 59.3 66.2 | 59.3 66.2 | 168.1 72.3 | 41.01 47.80 |
| FB 162 NRU 12-EL | 3 223.4  | 138.5 62.01 | 223.4  | 138.5 48.0 | 50.3 60.1 | 50.3 60.1 | 173.1 78.4 | 45.29 43.39 |
| FB 163 NRU 12-EL | 3 223.4  | 138.5 62.01 | 223.4  | 138.5 53.9 | 56.5 67.5 | 56.5 67.5 | 166.9 71.0 | 42.46 46.74 |
| FB 164 NRU 12-EL | 3 223.4  | 138.5 62.01 | 223.4  | 138.5 48.7 | 51.0 61.0 | 51.0 61.0 | 172.4 77.9 | 44.95 44.04 |
| FB 165 NRU 12-EL | 3 223.4  | 138.5 62.01 | 223.4  | 138.5 41.0 | 49.6 52.1 | 49.6 52.1 | 179.8 84.8 | 40.06 37.62 |
| FB 166 NRU 12-EL | 3 223.4  | 138.5 62.01 | 223.4  | 138.5 46.6 | 48.0 50.4 | 48.0 50.4 | 174.6 80.4 | 45.88 42.17 |
| FB 167 NRU 12-EL | 3 223.4  | 138.5 62.01 | 223.4  | 138.5 45.0 | 47.2 56.4 | 47.2 56.4 | 176.2 82.1 | 46.59 40.72 |
| FB 168 NRU 12-EL | 3 223.4  | 138.5 62.01 | 223.4  | 138.5 49.3 | 51.7 61.8 | 51.7 61.8 | 171.7 76.7 | 44.67 44.62 |
**APPENDIX IX**

**SUMMARY OF FISSIONABLE MATERIAL REPORTS**

<table>
<thead>
<tr>
<th>Item</th>
<th>Report Description</th>
<th>Prepared by</th>
<th>Reference</th>
<th>Issued</th>
<th>For</th>
<th>Use</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.</td>
<td>Summary of fissionable material held by each accountability unit.</td>
<td>PF&amp;CB on computer</td>
<td>Appendix No. 3</td>
<td>Monthly</td>
<td>1. Atomic Energy Control Board</td>
<td>Control Purposes</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>2. Finance Division</td>
<td>Control Purposes</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>3. Fissionable Material Accountability Committee</td>
<td>Control Purposes</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>4. CRNL Accountability Units</td>
<td>Control Purposes</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>5. PF&amp;CB</td>
<td>Record Purposes</td>
</tr>
<tr>
<td>2.</td>
<td>Summary of detailed fissionable material inventory held in PF&amp;CB fissionable material storage accountability unit.</td>
<td>PF&amp;CB on computer</td>
<td>Appendix No. 5</td>
<td>Monthly</td>
<td>1. Finance Division</td>
<td>Control Purposes</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>2. PF&amp;CB</td>
<td>Record Purposes</td>
</tr>
<tr>
<td>3.</td>
<td>Summary of detailed fissionable material inventory held in NRX and NRU accountability units.</td>
<td>PF&amp;CB on computer</td>
<td>Appendix No. 7</td>
<td>Monthly</td>
<td>1. Finance Division</td>
<td>Control Purposes</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>2. NRX &amp; NRU reactors</td>
<td>Control Purposes</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>3. PF&amp;CB</td>
<td>Record Purposes</td>
</tr>
<tr>
<td>4.</td>
<td>Enriched uranium and uranium-235 burn-up reports for NRX and NRU reactors.</td>
<td>PF&amp;CB on computer</td>
<td>Appendix No. 8</td>
<td>Monthly</td>
<td>1. USAEC</td>
<td>Control Purposes &amp; billing for uranium burn-up.</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>2. Finance Division</td>
<td>Control Purposes</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>3. Fissionable Material Accountability Committee</td>
<td>Control Purposes</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>4. PF&amp;CB</td>
<td>Record Purposes</td>
</tr>
<tr>
<td>5.</td>
<td>Monthly reports from AECL establishments other than CRNL and AECL contractors.</td>
<td>Accountability unit involved prepared manually</td>
<td>-</td>
<td>Monthly</td>
<td>1. Atomic Energy Control Board</td>
<td>Control Purposes</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>2. Finance Division</td>
<td>Control Purposes</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>3. PF&amp;CB</td>
<td>Reconciliation against PF&amp;CB ledgers</td>
</tr>
<tr>
<td>6.</td>
<td>Fissionable material balance reports at completion of an AECL contract.</td>
<td>AECL contractors involved prepared manually</td>
<td>-</td>
<td>-</td>
<td>1. PF&amp;CB</td>
<td>Control Purposes</td>
</tr>
</tbody>
</table>
## APPENDIX X

### SUMMARY OF HEAVY-WATER REPORTS

<table>
<thead>
<tr>
<th>Item</th>
<th>Report</th>
<th>Prepared by</th>
<th>Reference</th>
<th>Issued</th>
<th>For</th>
<th>Use</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.</td>
<td>Summary of heavy water held by each accountability unit.</td>
<td>PP&amp;CB on computer</td>
<td>Appendix No. 4</td>
<td>Monthly</td>
<td>1. Senior management</td>
<td>Control Purposes</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>2. Finance Division</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>3. PP&amp;CB</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Control Purposes</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Record Purposes</td>
<td></td>
</tr>
<tr>
<td>2.</td>
<td>Detailed heavy water inventory of PP&amp;CB heavy water storage Accountability unit.</td>
<td>PP&amp;CB on computer</td>
<td>Appendix No. 4</td>
<td>Monthly</td>
<td>1. Finance Division</td>
<td>Control Purposes</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>2. PP&amp;CB</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Control Purposes</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Record Purposes</td>
<td></td>
</tr>
<tr>
<td>3.</td>
<td>Heavy water inventory report from NRX, NRU and NPD heavy water accountability units</td>
<td>NRX, NRU &amp; NPD heavy water accountability units prepared manually</td>
<td>Monthly</td>
<td>1. Senior management</td>
<td>Control Purposes</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>2. Finance Division</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>3. PP&amp;CB</td>
<td>Record Purposes</td>
</tr>
<tr>
<td>4.</td>
<td>Summary of heavy water stocks held in PP&amp;CB storage</td>
<td>PP&amp;CB prepared manually</td>
<td>Monthly</td>
<td>1. Senior management</td>
<td>Control Purposes</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>2. Finance Division</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>3. PP&amp;CB</td>
<td>Record Purposes</td>
</tr>
<tr>
<td>5.</td>
<td>Summary of D2O losses</td>
<td>PP&amp;CB prepared manually</td>
<td>Monthly</td>
<td>1. Senior management</td>
<td>Control Purposes</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>2. Finance Division</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>3. PP&amp;CB</td>
<td>Record Purposes</td>
</tr>
</tbody>
</table>
DISCUSSION

J. CHENOUARD: Are your burn-up data calculated values?

G.W. FLETCHER: The burn-up data are calculated from the MWd output of the rods on the assumption that 1.253 g of $^{235}$U are destroyed for every MWd of energy produced (1.048 g by fission and 0.205 g by non-fission capture yielding $^{236}$U).

J. CHENOUARD: The last column in the table entitled "Burn-up in Enriched Uranium Rods Leased from the USAEC" (Appendix 8) shows "per cent burn-up". Could you tell us whether the burn-up ratios above 50% were calculated in regard to the theoretical burn-up or the limiting experimental burn-up for a given type of fuel?

G.W. FLETCHER: These are average values related to the original $^{235}$U content and not to the limiting experimental burn-up.

W.J. WRIGHT: In Appendix 2 you have a full description rather than a coded description of the materials included in the computer balance. Bearing in mind the difficulties of programming for all forms of material, do you think that this is necessary or desirable?

G.W. FLETCHER: Yes, we have found it necessary to identify the material in this way. However, the number of such miscellaneous pieces of material is kept to a minimum, and codes have been devised to sort them on a broad basis.

R.G. CARDWELL: There is no doubt a good deal of truth in this: It is often difficult to condense all desired input data for computer calculation and print-out of inventory storage reports. However, we have found that although many people request storage, their reasons for doing so tend to fall into a few basic categories. These can be reduced to numerical designations and the numbers fed to a computer in such a way that the reasons for requests are printed out either by their classification number or in their complete word description, which is keyed by the classification number.

W.J. WRIGHT: Are suitable business-type machines available for keeping material balance records?

G.W. FLETCHER: Yes, they are; but having bought the IBM-1401 computer for other purposes we have been able to use it profitably for this task as well.

E. LOCKETT: You state that three separate audits are carried out: one by the Production Planning and Control Branch (PP&CB), one by the Atomic Energy Control Board and one by your accountants. Do the accountants attempt to make a physical check on special materials?

G.W. FLETCHER: Our contractors are subject to audit by these three groups, although the check of the PP&BC is more in the nature of an operational control, carried out whenever there is a discrepancy between the control ledger and actual stocks which cannot be cleared up by examination and discussion of the records. The audit performed by the accountants of the Finance Division does include physical inspection of materials as well as an examination of the ledgers.

F. RONTEIX: As our task here is to compare methods of managing nuclear materials in different countries, I might say that I am particularly struck by the similarity between the Canadian system, which has just been
so ably described by Mr. Fletcher, and the system which we have in France. This similarity will, of course, encourage us to pursue our efforts.

I should like to make one specific comment on Mr. Fletcher's paper. From the appendices we see that enriched uranium is accounted for as total uranium and as $^{235}$U, but in a single inventory. It strikes me that this is not enough if one wishes to consider the uranium from the economic point of view as well as from the standpoint of criticality and control of utilization; for this purpose each different enrichment should appear separately in the ledger. For example, 100 kg total uranium and 20 kg $^{235}$U could represent either a single lot of 20%-enriched uranium or the average of two lots, one enriched to 98%, the other to 2%. It has seemed to us in France that the only solution is to keep a separate account for each isotopic mixture. Then it is no longer necessary to keep a special account for $^{235}$U as it can be obtained simply by multiplying total uranium by the isotopic fraction for each entry. This is a fairly vexed question, however, and as our time is limited it would perhaps be best for those who are interested to discuss it privately.
DETERMINATION OF THE URANIUM CONTENT IN SCRAP

K. NAKAJIMA AND N. SAITO
ATOMIC FUEL CORPORATION, TOKAI-MURA, IBARAKI-KEN, JAPAN

Abstract — Résumé — Аннотация — Resumen

DETERMINATION OF THE URANIUM CONTENT IN SCRAP. Uranium accountability problems related to recovered precipitate cakes and metal scrap are described in this paper.

Waste solution from the uranium refining pilot plant utilizing the Excer process is neutralized and sent to aging tanks. Condensed slurry-type precipitates are filtered with a filter press. To determine the uranium content of the cakes, sampling experiments were continued over a two-year operating period, and the following values were obtained for 171 lots:

- Average uranium content $\bar{X}$: 10.29
- Variance between lots $\sigma^2_1$: 4.36
- Variance between filter plates $\sigma^2_2$: 0.014
- Variance within filter plates $\sigma^2_3$: 2.02
- Variance of reduction and analysis $\sigma^2 + \sigma^2_4$: 0.349.

Based on these results, a proper sampling plan was established. In this plan variance of mean value $V(\bar{X})$ is 4.65, variation without $\sigma^2_1$ is 0.29, and coefficient of variation is about 5%. Derived values from this sampling method using precipitate cakes as feed materials for a recovery process, agree satisfactorily with the analysis of the resulting product.

Since uranium metal scrap in the form of turnings or sawdust ignites easily in air, it is difficult to use the direct weighing method and consequently the weight-volume-density method was adopted.

As a result of theoretical analysis of errors associated with this method it was found that both a lower specific gravity of liquid and a higher density scrap sample result in a higher degree of precision, and also that measurement errors of liquid density and total volume seriously affect the derived scrap weight.

During actual weighings, the following observations were noted: (a) Uranium metal scraps are oxidized even under machine oil, and fine oxidized particles cause a measurement error; and (b) The measurement error of oil density becomes negligible using a proper method.

Using these results, detailed techniques were adopted. In this way, the accuracy of this method increased and determinations of uranium scrap weights have been carried out satisfactorily.
Etant donné que les déchets d’uranium métallique sous forme de copeaux ou de limaille s’enflamment facilement à l’air, il est difficile de pratiquer la pesée directe. C’est pourquoi la méthode poids/volume/densité a été adoptée.

A la suite d’une étude théorique des erreurs inhérentes à cette méthode, les auteurs ont constaté que l’on obtenait un plus haut degré de précision en abaissant la densité du liquide et en augmentant celle de l’échantillon de déchets, et également que les erreurs dans les mesures de la densité du liquide et du volume total avaient une forte incidence sur le poids calculé des déchets.

Lors des mesures en cours d’opérations, les observations suivantes ont été faites: a) les déchets d’uranium métallique s’oxydent même dans l’huile de graissage et la présence de fines particules oxydées fausse les mesures; et b) les erreurs dans la mesure de la densité de l’huile peuvent être rammées à une valeur négligeable si l’on utilise la méthode la plus appropriée.

Sur la base des résultats obtenus, des procédés ont été mis au point dans le détail; la méthode ainsi améliorée en précision a permis de procéder à la pesée des déchets d’uranium d’une manière satisfaisante.

OPРЕДЕЛЕНИЕ СОДЕРЖАНИЯ УРАНА В ОТХОДАХ. Дается описание проблем учета урана, связанных с восстановлением прессованного осадка и металлических отходов.

Раствор отходов опытной установки по очистке урана, в которой применяется процесс Эксера, нейтрализуется и направляется в баки выдержки. Конденсированные осадки типа шлама фильтруются с помощью фильтропресса. Для определения содержания урана в прессованном осадке эксперименты по отбору проб продолжались в течение двух лет в ходе эксплуатации, и для 171 партии были получены следующие значения (%):

Среднее содержание урана X: 10,29.
Изменение между партиями σ²: 4,36.
Изменение между пластинами фильтра σ²: 0,014.
Изменение внутри пластин фильтра σ²: 2,02.
Изменение восстановления и анализа σ²: 2,049.

На основании этих результатов был разработан соответствующий план отбора проб, в этом плане изменение среднего значения V(X) составляет 4,65, изменение без о² составляет 0,29%, коэффициент вариации — около 5%. В случае использования прессованных осадков в качестве сырьевых материалов в процессе восстановления выявленные на основании применения этого метода отбора проб значения удовлетворительно согласуются с результатами анализа полученного продукта.

По сколько отходы металлического урана в виде стружки или опилок легко воспламеняются на воздухе, трудно использовать метод непосредственного взвешивания. Поэтому принят метод веса — объема-плотности.

В результате теоретического анализа ошибок, связанных с этим методом, было установлено, что при использовании жидкости с пониженной плотностью и образца отходов с повышенной плотностью достигалась более высокая степень точности, а также что ошибки при измерении плотности жидкости и полного объема оказывают большое влияние на выводимое значение веса отходов.

При фактических взвешиваниях отмечалось следующее: a) отходы металлического урана окисляются даже под действием машинного масла, и окисленные небольшие частицы являются причиной ошибок при измерении; b) ошибка измерения плотности масла становится пренебрежимо малой при использовании надлежащего метода.

На основании этих результатов были разработаны подробные методики. Таким путем была увеличена точность этого метода и удовлетворительно определялся вес отходов урана.

DETERMINACION DEL CONTENIDO DE URANO EN RESIDUOS. Se exponen en esta memoria algunos problemas de contabilidad del uranio relacionados con la recuperación de tortas de precipitados y de recortes metálicos.

Las soluciones de desechos, procedentes de la planta piloto de refincación de uranio a base del método Excer, se neutralizan y se envían a tanques de envejecimiento. Los precipitados fangosos se separan con un filtro-prensa. Para determinar el contenido de uranio de las tortas se realizaron durante dos años experimentos de muestreo con la planta en marcha. Se obtuvieron los siguientes valores para 171 tandas:

Contenido promedio de uranio, X: 10,29
Variancia entre las tandas σ²: 4,36
Variancia entre las placas del filtro σ²: 0,014
Variancia dentro de las placas σ²: 2,02
Variancia de la reducción y del análisis σ²: 0,349.
Sobre la base de estos resultados se estableció un plan de muestreo con las siguientes características: variación del valor medio $V(X)$: 4,65; variación sin $\sigma^2$: 0,29; coeficiente de variación: un 5%. Cuando un proceso de recuperación se efectúa a base de tortas de precipitado, los valores obtenidos mediante este método de muestreo concuerdan satisfactoriamente con los resultados del análisis del producto.

Como los residuos de uranio metálico en forma de virutas o de partículas son pirofóricos, es difícil emplear el método de pesada directa. Por consiguiente, se ha adoptado el método peso-volumen-densidad.

El análisis teórico de los errores asociados a este método han puesto de manifiesto que un líquido de poca densidad y una muestra muy densa permiten obtener un mayor grado de precisión y que los errores cometidos al medir la densidad del líquido y el volumen total influyen seriamente sobre el peso de los residuos.

Al hacer las pesadas directas se pudo observar lo siguiente: a) Los residuos de uranio metálico se oxidan aunque estén recubiertos de aceite de máquina, y las pequeñas partículas oxidadas originan un error de medición, b) El error en la medición de la densidad del aceite tiende a desaparecer cuando se sigue un método adecuado.

Partiendo de estos resultados se han adoptado técnicas que han aumentado la precisión de ese método y han permitido determinar satisfactoriamente el peso de los residuos de uranio.

The Tokai Refinery of the Atomic Fuel Corporation (AFC) has a uranium refining pilot plant utilizing the Excer process, the flow sheet of which is shown in Fig. 1. Measurement of the uranium content of the main processes is carried out rather easily by normal measuring methods, on the other hand, measurement of the uranium content of secondary processes, for instance, that of slags, metal scrap, recovered precipitate cakes etc. contains many difficult problems. In this paper, problems related to recovered precipitate cakes and metal scrap are described.

1. SAMPLING METHOD FOR RECOVERED PRECIPITATE CAKES

Waste solutions containing 0.1 to 2 g/litre uranium from the plant are neutralized 3 to 10 times a day in a 1.5-m³ volume neutralizer tank, and are sent alternately to two 10-m³ volume aging tanks. After settling, upper liquids are drained and condensed slurry-type precipitates are filtered with a filter press. The batch weight of recovered precipitate cakes is about 200 kg and their uranium content is about 10% in wet base.

Uranium distribution within cakes is heterogeneous as determined by analytical samples, so sampling experiments were continued over a two-year operating period for 171 lots.

1.1. Experimental method

The following sampling methods, A and B, were applied alternately and analytical results were recorded on control charts. To estimate the variance of reduction and analysis, some samples were divided into halves and both of them were analysed. In this case one of the analytical values was chosen at random for recording on the control charts.

1.1.1. Sampling method A: see Fig. 2a

Two samples are taken at random from the 1st, 4th, 7th, 10th, 13th, 16th, 19th, 22nd, 25th, and 28th filter plates, and one sample from each plate is collected together and mixed in sample bottle 1, the other samples are treated in the same way in sample bottle 2.
1.1.2. Sampling method B: see Fig. 2b

Two samples are taken at random from the 1st, 7th, 13th, 19th, and 25th filter plates, collected together and mixed in sample bottle 1, and two samples from 4th, 10th, 16th, 22nd, and 28th filter-plates are treated in sample bottle 2 in the same manner.

Samples 1 and 2 are both analysed for each lot.

1.2. Results

The $\overline{X}$-control chart of 171 lots is shown in Fig. 3, the R-control charts of sampling method A and B are shown in Figs. 4 and 5, and the $\overline{X}$-R control chart for estimation of variance of reduction and analysis is shown in Fig. 6. From the above figures the following values were obtained:

- Total average of uranium content $\overline{X}$: 10.29%
- Average of uranium content of sampling method A $\overline{X}_A$: 10.34%
- Average range of sampling method A $R_A$: 0.842%
- Average of uranium content of sampling method B $\overline{X}_B$: 10.24%
- Average range of sampling method B $R_B$: 0.844%
1. SAMPLING METHOD A

2. SAMPLING METHOD B

FIG. 2. Schematic presentation of sampling method A and B

Average of uranium content of samples for estimating the variance of reduction and analysis $\overline{X_M}$: 11.15%
Average range of samples for estimating the variance of reduction and analysis $R_M$: 0.667%

1.3. Discussion

The following relations exist for sampling method A:

$$\text{Expectation } E\left(\frac{R_A}{d_2}\right)^2 = \frac{\sigma_w^2}{mn/2} + \sigma_R^2 + \sigma_M^2$$ \hspace{1cm} (1)

$$\text{Variance } V\left(X_A\right) = \sigma_L^2 + \frac{\sigma_b^2}{m} + \frac{\sigma_w^2}{nh} + \frac{\sigma_R^2}{2} + \frac{\sigma_M^2}{2}$$ \hspace{1cm} (2)

where

$X_{A1}, X_{A2}$: one set of analytical data
$\overline{X_A}$: average of $X_{A1}$ and $X_{A2}$
$R_A$: range of $X_{A1}$ and $X_{A2}$
$\sigma_L^2$: variance between filtered lots
$\sigma_b^2$: variance between filter plates
$\sigma_w^2$: variance within filter plates
$\sigma_R^2$: variance of reduction
$\sigma_M^2$: variance of analysis
$m$: number of plates sampled (10 in this case)
n$: number of samples for each plate (2 in this case)
$n$: average of $n$
d_2: a factor of control line of $\overline{X}$-$R$ control chart (1.128 in this case)
Similarly for sampling method B:

\[
\text{Expectation } E\left(\frac{R_B}{d_2}\right)^2 = \frac{\sigma_B^2}{m} + \frac{\sigma_w^2}{mn} + \sigma_R^2 + \sigma_M^2 
\]

(3)

\[
\text{Variance } V\left(\bar{X}_B\right) = \frac{\sigma_L^2}{m} + \frac{\sigma_R^2}{m} + \frac{\sigma_W^2}{mn} + \frac{\sigma_R^2}{2} + \frac{\sigma_M^2}{2} = V\left(\bar{X}_A\right) 
\]

(4)

where

- \(X_{B1}, X_{B2}\): one set of analytical data
- \(\bar{X}_B\): average of \(X_{B1}\) and \(X_{B2}\)
- \(R_B\): range of \(X_{B1}\) and \(X_{B2}\)
For the data for estimating variance of reduction and analysis

\[ \text{Expectation } E \left( \frac{R_M}{d_2} \right)^2 = \sigma_R^2 + \sigma_M^2 \]  \hspace{1cm} (5)

where

- \( X_M, X_2 \) : one set of analytical data
- \( \bar{X}_M \) : average of \( X_M \) and \( X_2 \)
- \( R_M \) : range of \( X_M \) and \( X_2 \)

The following relations are derived from the above formulas:

Estimate of variance of reduction and analysis

\[ \hat{\sigma}_R^2 + \hat{\sigma}_M^2 = \left( \frac{R_M}{d_2} \right)^2 \]  \hspace{1cm} (6)

Estimate of variance within filter plates

\[ \hat{\sigma}_w^2 = \frac{m \bar{R}}{2} \left\{ \left( \frac{R_A}{d_2} \right)^2 - \left( \frac{R_M}{d_2} \right)^2 \right\} \]  \hspace{1cm} (7)

Estimate of variance between filter plates

\[ \hat{\sigma}_b^2 = \frac{m}{2} \left\{ \left( \frac{R_B}{d_2} \right)^2 - \left( \frac{R_A}{d_2} \right)^2 \right\} \]  \hspace{1cm} (8)

Observed values in these formulas were \( \hat{\sigma}_R^2 + \hat{\sigma}_M^2 = 0.349 \), \( \hat{\sigma}_w^2 = 2.02 \), and \( \hat{\sigma}_b^2 = 0.014 \).
The variance between lots $\sigma_L^2$ can be obtained by using the following formula:

$$\hat{\sigma}_L^2 = \frac{S^2}{2} - \frac{1}{2} \left( \frac{R}{d_2} \right)^2$$  

(9)

where $S$ is the unbiased variance of total lots in this case $S^2 = 4.64$ and the estimate of variance between lots is $\hat{\sigma}_L^2 = 4.36$.

The fact that variation within filter plates is rather large while variation between filter plates is negligible shows that precipitate slurry flows into each plate uniformly.

It is clear from control charts that these sampling methods are satisfactory to detect variation between lots. The alternative application of sampling methods A and B for production operations is unsuitable because of their complicated procedures, therefore a simplified sampling method was established.

1.4. New sampling method

The 10th and 20th filter plates of each filtered lot are chosen and nine samples are taken from each filter plate, as shown in Fig. 7. In this sampling method,

$$\text{Variance } \text{V} (\bar{X}) = \sigma_L^2 + \frac{\sigma_R^2}{m} + \frac{\sigma_M^2}{n} \cdot \frac{m + n}{2}$$  

(10)

where

- $X_1$, $X_2$: one set of analytical data
- $\bar{X}$: average of $X_1$ and $X_2$
- $m$: number of plates sampled (2 in this case)
- $n$: number of samples (9 in this case)
FIG. 7. Position of nine samples for each plate

Other symbols are same as in Eqs. (1) and (2). Using the numerical values already obtained, $V(\bar{X}) = 4.65$.

Excluding $\sigma_t^2$ from $V(\bar{X})$, $\sigma_E^2 = 0.29$ and is almost the same as that of sampling methods A and B (0.28). The coefficient of variation becomes $\sigma_E/\bar{X} = \sqrt{0.29/10.29} = 0.052$, and this value indicates that this method is effective in practice.

1.5. **Comparison with values of dissolution**

Recovered precipitate cakes are used as materials for the recovery process. In this case, comparison of derived values from this sampling method with the analysis of resulting solutions was carried out for 40 successive lots.

\[
\begin{align*}
\text{Total of derived values} & : 1072.3 \text{ kg of U} \\
\text{Total of analytical result} & : 1080.5 \text{ kg of U}
\end{align*}
\]

Taking $d$ as the difference between both values in each lot, the average of the differences $\bar{d} = 0.205 \text{ kg}$ and the sum of the square of the residual $\Sigma (d-d)^2 = 110.86$. Application of Student’s $t$ test, showed no significant difference between both values.

2. **DETERMINATION OF URANIUM CONTENT IN METAL SCRAP**

For the fabrication of the secondary charge fuel elements for JRR-3 during 1963 AFC supplied refined ingot metal. After the fuel elements had been manufactured, scrap such as turnings, saw-dust etc. was sent back to AFC for recovery. There was a large discrepancy between the shipper’s and receiver’s estimates of uranium content as shown in TABLE I. The reason for this discrepancy was examined both theoretically and experimentally, and detailed procedures were established. As a result, the discrepancy has been greatly decreased.

Uranium metal scrap easily ignites in air, so instead of direct weighing an indirect calculation method is used; after immersing the scrap in inert liquid, the weight of scrap is calculated from Eq (11) using values of total weight, total volume, and specific gravities of uranium and liquid. In the case of scrap from JRR-3 fuel element fabrication, machine oil was used as the immersing liquid.
2.1. Theoretical analysis of errors

The formula for calculating uranium content is

\[ W = \frac{\rho (W_T - W_t - \rho_o V)}{(\rho - \rho_o)} \] (11)

where

- \( W \): uranium net weight (kg)
- \( W_T \): total weight (kg)
- \( W_t \): tare weight (kg)
- \( V \): total volume (1)
- \( \rho \): specific gravity of uranium
- \( \rho_o \): specific gravity of liquid

Differentiating the logarithms of both sides of Eq. (11), the formula of relative error is obtained.

\[
\frac{dW}{W} = -\frac{\rho_o}{\rho (\rho - \rho_o)} \, d\rho + \frac{(W_T - W_t - \rho V)}{(W_T - W_t - \rho_o V)(\rho - \rho_o)} \, d\rho_o + \frac{1}{(W_T - W_t - \rho_o V)} \, dW_T \]

\[
\frac{1}{(W_T - W_t - \rho_o V)} \, dW_t + \frac{1}{(W_T - W_t - \rho_o V)} \, dV
\]

In this formula, \( \rho \approx 18.9 >> \rho_o \) and \( \rho V >> W_T, W_t \). It was found that the greater the specific gravity of the liquid, the greater the resulting relative error, and the higher the bulk density of the scrap in the measuring drum, the smaller the relative error.

In practice a 200-litre volume drum with level gauge is used and the approximate values of the terms in Eq. (12) and their fluctuations are as follows:

- \( W_T = 240 \text{ kg} \) \( \Delta W_T = 0.2 \text{ kg} \) (minimum scale of balance)
- \( W_t = 20 \text{ kg} \) \( \Delta W_t = 0.2 \text{ kg} \) (minimum scale of balance)
- \( V = 180 \text{ l} \) \( \Delta V = 1.0 \text{ l} \) (about 4 mm depth)
- \( \rho = 18.9 \) \( \Delta \rho = 0.1 \) (in original ingot \( \rho : 19.0 \sim 18.8 \))
- \( \rho_o = 0.93 \) \( \Delta \rho_o = 0.004 \) (measurement error)

in this case \( W = 55.3 \text{ kg} \).

Values of the terms in Eq. (12) are as follows,

\[
\frac{\rho_o}{\rho (\rho - \rho_o)} \Delta \rho = 0.0003
\]

\[
\frac{(W_T - W_t - \rho V)}{(W_T - W_t - \rho_o V)(\rho - \rho_o)} \Delta \rho_o = 0.013
\]

\[
\frac{1}{W_T - W_t - \rho_o V} \Delta W_T = 0.0038
\]

\[
\frac{1}{W_T - W_t - \rho_o V} \Delta W_t = 0.0038
\]

\[
\frac{\rho}{W_T - W_t - \rho_o V} \Delta V = 0.018
\]

and the possible maximum relative error is \( \Delta W/W = 0.039 = 3.9\% \).
## TABLE I

**DISCREPANCY OF SHIPPER AND RECEIVER**

*(Before the establishment of detailed procedure)*

<table>
<thead>
<tr>
<th>No.</th>
<th>Shipper's weight A (kg U)</th>
<th>Receiver's weight B (kg U)</th>
<th>Difference A - B (kg U)</th>
<th>Error rate (%)</th>
<th>Specific gravity of oil Shipper</th>
<th>Specific gravity of oil Receiver</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1</td>
<td>46.7</td>
<td>43.6</td>
<td>3.1</td>
<td>6.6</td>
<td>0.930</td>
<td>0.941</td>
</tr>
<tr>
<td>2</td>
<td>22.8</td>
<td>21.9</td>
<td>0.9</td>
<td>4.0</td>
<td>0.930</td>
<td>0.938</td>
</tr>
<tr>
<td>3</td>
<td>49.2</td>
<td>38.1</td>
<td>11.1</td>
<td>29.1</td>
<td>0.934</td>
<td>0.997</td>
</tr>
<tr>
<td>4</td>
<td>29.9</td>
<td>27.8</td>
<td>2.1</td>
<td>7.0</td>
<td>0.930</td>
<td>0.935</td>
</tr>
<tr>
<td>5</td>
<td>40.1</td>
<td>37.4</td>
<td>2.7</td>
<td>6.7</td>
<td>0.930</td>
<td>0.949</td>
</tr>
<tr>
<td>6</td>
<td>40.7</td>
<td>37.6</td>
<td>3.1</td>
<td>7.6</td>
<td>0.930</td>
<td>0.940</td>
</tr>
<tr>
<td>7</td>
<td>34.9</td>
<td>27.6</td>
<td>7.3</td>
<td>29.9</td>
<td>0.930</td>
<td>0.944</td>
</tr>
<tr>
<td>8</td>
<td>45.3</td>
<td>42.9</td>
<td>2.4</td>
<td>5.3</td>
<td>0.930</td>
<td>0.941</td>
</tr>
<tr>
<td>9</td>
<td>42.2</td>
<td>40.2</td>
<td>2.0</td>
<td>4.7</td>
<td>0.930</td>
<td>0.930</td>
</tr>
<tr>
<td>10</td>
<td>39.9</td>
<td>39.3</td>
<td>0.6</td>
<td>1.5</td>
<td>0.932</td>
<td>0.933</td>
</tr>
<tr>
<td>11</td>
<td>57.0</td>
<td>53.5</td>
<td>3.5</td>
<td>6.1</td>
<td>0.929</td>
<td>0.946</td>
</tr>
<tr>
<td>12</td>
<td>21.3</td>
<td>20.4</td>
<td>0.9</td>
<td>4.2</td>
<td>0.931</td>
<td>0.938</td>
</tr>
<tr>
<td>13</td>
<td>23.8</td>
<td>20.9</td>
<td>2.9</td>
<td>12.2</td>
<td>0.930</td>
<td>0.940</td>
</tr>
<tr>
<td>14</td>
<td>27.5</td>
<td>25.7</td>
<td>1.8</td>
<td>6.5</td>
<td>0.939</td>
<td>0.942</td>
</tr>
<tr>
<td>15</td>
<td>10.0</td>
<td>7.2</td>
<td>2.8</td>
<td>28.0</td>
<td>0.935</td>
<td>0.940</td>
</tr>
<tr>
<td>16</td>
<td>42.7</td>
<td>38.3</td>
<td>4.4</td>
<td>10.3</td>
<td>0.934</td>
<td>0.942</td>
</tr>
<tr>
<td>17</td>
<td>62.9</td>
<td>60.1</td>
<td>2.8</td>
<td>4.5</td>
<td>0.936</td>
<td>0.940</td>
</tr>
<tr>
<td>18</td>
<td>61.9</td>
<td>58.0</td>
<td>3.9</td>
<td>6.3</td>
<td>0.936</td>
<td>0.944</td>
</tr>
<tr>
<td>19</td>
<td>43.6</td>
<td>42.7</td>
<td>0.9</td>
<td>2.1</td>
<td>0.934</td>
<td>0.943</td>
</tr>
<tr>
<td>20</td>
<td>49.8</td>
<td>47.3</td>
<td>2.5</td>
<td>5.0</td>
<td>0.934</td>
<td>0.943</td>
</tr>
<tr>
<td>21</td>
<td>12.9</td>
<td>11.6</td>
<td>1.3</td>
<td>10.1</td>
<td>0.934</td>
<td>0.940</td>
</tr>
<tr>
<td>22</td>
<td>61.9</td>
<td>52.9</td>
<td>9.0</td>
<td>17.0</td>
<td>0.934</td>
<td>0.973</td>
</tr>
<tr>
<td>23</td>
<td>68.8</td>
<td>45.5</td>
<td>23.3</td>
<td>48.6</td>
<td>0.934</td>
<td>1.045</td>
</tr>
<tr>
<td>24</td>
<td>28.3</td>
<td>30.6</td>
<td>-2.3</td>
<td>7.5</td>
<td>0.934</td>
<td>0.932</td>
</tr>
<tr>
<td>25</td>
<td>45.1</td>
<td>41.6</td>
<td>3.5</td>
<td>8.4</td>
<td>0.934</td>
<td>0.944</td>
</tr>
<tr>
<td>Total</td>
<td>1007.2</td>
<td>912.7</td>
<td>94.5</td>
<td>10.4</td>
<td>0.933</td>
<td>0.948</td>
</tr>
</tbody>
</table>
It is clear from these values that measurement errors of liquid density and total volume seriously affect derived scrap weights. The main cause of the large error is the factor in term $\rho_0$ together with the measurement error in term $V$.

2.2. Practice

In paragraph 2.1, $\rho$ and $\rho_0$ are treated as constant values, but in fact, the oxidation rate of uranium metal is rather high even in machine oil and it causes an overestimate of about 8% in uranium content for the oxidized portion. Specific gravities of oil also change with time, fine particles of uranium oxide suspend in oil and uranium content sometimes amounts to more than 5%, in this case the apparent specific gravity of oil increases to more than 1.0.

It seems that the foregoing was the main reason for the discrepancy between the shipper's and receiver's values. It is necessary to measure $\rho_0$ in each drum every time uranium weighing is carried out, and attention must be paid to proper sampling of the oil. More correct values are obtained using a 500-ml volumetric flask than with a pycnometer at the same room temperature as when weighing scrap. Using the results given in paragraphs 2.1 and 2.2, detailed techniques of this weighing method were adopted, as shown in paragraph 2.3.

### TABLE II

<table>
<thead>
<tr>
<th>No.</th>
<th>Weight (kg U)</th>
<th>Difference A-B (kg U)</th>
<th>Error rate A-B x 100</th>
<th>Error rate A-B x 100</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Direct (A)</td>
<td>Indirect (B)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>1</td>
<td>5.6</td>
<td>5.7</td>
<td>0.1</td>
<td>1.8</td>
</tr>
<tr>
<td>2</td>
<td>10.9</td>
<td>10.8</td>
<td>-0.1</td>
<td>-0.9</td>
</tr>
<tr>
<td>3</td>
<td>21.2</td>
<td>21.1</td>
<td>-0.1</td>
<td>-0.5</td>
</tr>
<tr>
<td>4</td>
<td>31.8</td>
<td>31.9</td>
<td>0.1</td>
<td>0.3</td>
</tr>
<tr>
<td>5</td>
<td>42.6</td>
<td>42.7</td>
<td>0.1</td>
<td>0.2</td>
</tr>
<tr>
<td>6</td>
<td>52.5</td>
<td>52.7</td>
<td>0.2</td>
<td>0.4</td>
</tr>
<tr>
<td>7</td>
<td>62.6</td>
<td>62.7</td>
<td>0.1</td>
<td>0.2</td>
</tr>
<tr>
<td>8</td>
<td>73.0</td>
<td>73.2</td>
<td>0.2</td>
<td>0.3</td>
</tr>
<tr>
<td>9</td>
<td>83.3</td>
<td>83.5</td>
<td>0.2</td>
<td>0.2</td>
</tr>
<tr>
<td>10</td>
<td>93.5</td>
<td>93.8</td>
<td>0.3</td>
<td>0.3</td>
</tr>
<tr>
<td>11</td>
<td>97.3</td>
<td>97.6</td>
<td>0.3</td>
<td>0.3</td>
</tr>
<tr>
<td>MEASUREMENT DATA</td>
<td>DRUM No</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>------------------</td>
<td>---------</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>SHIPPER</td>
<td>GROSS WEIGHT (W_G)</td>
<td>TARE WEIGHT (W_T)</td>
<td>kg</td>
<td></td>
</tr>
<tr>
<td>DATE</td>
<td>ROOM TEMPERATURE</td>
<td>°C</td>
<td></td>
<td></td>
</tr>
<tr>
<td>WEIGHT OF MEASURING VESSEL (W_M)</td>
<td>kg</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>WEIGHT OF VESSEL AND WATER (W_W)</td>
<td>kg</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>MEASURING VOLUME (V = W_W - W_T)</td>
<td>l</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>TOTAL WEIGHT (W_T)</td>
<td>kg</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>LIQUID TEMPERATURE</td>
<td>°C</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>SPECIFIC GRAVITY OF LIQUID (\rho_0)</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>WEIGHT OF URANIUM SOLID (W) = \frac{18.90 (W_T - W_T - \rho_0 V)}{18.90 - \rho_0}</td>
<td>kg</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>WEIGHT OF LIQUID (W_L = W_G - W_T)</td>
<td>kg</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>URANIUM CONTENT OF LIQUID (R)</td>
<td>kgU / kg LIQUID</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>SUSPENDED URANIUM IN LIQUID (W')</td>
<td>kg</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>SHIPPER'S VALUE (S)</td>
<td>kg</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>DIFFERENCE (S - W)</td>
<td>kg</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>(S - W) / W \times 100</td>
<td>%</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>TOTAL DELIVERY</td>
<td>kg</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

**FIG. 8.** Measurement record

### 2.3. Detailed procedure of weighing method

(a) A 200-litre drum with level gauge is used as measuring vessel, and 180 litre is used as the standard oil level.

(b) Uranium content of one weighing must be more than 40 kg.

(c) The volume of the container must be measured by weighing the same volume of water after every measurement.

(d) The shipper must use new machine oil for each measurement and send the scrap with the oil to AFC. AFC must use this same oil for measurements.

(e) An oil sample is taken in the centre of every drum, and specific gravity is measured by a 500-ml volumetric flask in the same room.
# Table III
**Discrepancy of Shipper and Receiver**
*(After the establishment of detailed procedure)*

<table>
<thead>
<tr>
<th>No.</th>
<th>Shipper's weight A (kg U)</th>
<th>Receiver's weight B (kg U)</th>
<th>Difference A - B (kg U)</th>
<th>Error rate (A-B)/B x 100 (%)</th>
<th>Specific gravity of oil</th>
<th>Shipper</th>
<th>Receiver</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>53.5</td>
<td>53.7</td>
<td>-0.2</td>
<td>0.4</td>
<td>0.926</td>
<td>0.924</td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>72.1</td>
<td>71.2</td>
<td>0.9</td>
<td>1.3</td>
<td>0.926</td>
<td>0.926</td>
<td></td>
</tr>
<tr>
<td>3</td>
<td>68.3</td>
<td>67.9</td>
<td>0.4</td>
<td>0.6</td>
<td>0.926</td>
<td>0.926</td>
<td></td>
</tr>
<tr>
<td>4</td>
<td>47.4</td>
<td>48.0</td>
<td>-0.6</td>
<td>1.2</td>
<td>0.926</td>
<td>0.924</td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>51.4</td>
<td>52.4</td>
<td>-1.0</td>
<td>1.9</td>
<td>0.926</td>
<td>0.923</td>
<td></td>
</tr>
<tr>
<td>6</td>
<td>57.3</td>
<td>57.6</td>
<td>-0.3</td>
<td>0.5</td>
<td>0.926</td>
<td>0.919</td>
<td></td>
</tr>
<tr>
<td>7</td>
<td>53.1</td>
<td>53.2</td>
<td>-0.1</td>
<td>0.2</td>
<td>0.926</td>
<td>0.918</td>
<td></td>
</tr>
<tr>
<td>8</td>
<td>46.0</td>
<td>46.9</td>
<td>-0.9</td>
<td>-1.9</td>
<td>0.926</td>
<td>0.917</td>
<td></td>
</tr>
<tr>
<td>9</td>
<td>70.2</td>
<td>70.2</td>
<td>0</td>
<td>0</td>
<td>0.926</td>
<td>0.927</td>
<td></td>
</tr>
<tr>
<td>10</td>
<td>82.7</td>
<td>82.7</td>
<td>0</td>
<td>0</td>
<td>0.970</td>
<td>0.890</td>
<td></td>
</tr>
<tr>
<td>11</td>
<td>39.7</td>
<td>38.8</td>
<td>0.9</td>
<td>2.3</td>
<td>0.925</td>
<td>0.923</td>
<td></td>
</tr>
<tr>
<td>12</td>
<td>55.6</td>
<td>54.1</td>
<td>1.5</td>
<td>2.8</td>
<td>0.923</td>
<td>0.922</td>
<td></td>
</tr>
<tr>
<td>13</td>
<td>82.3</td>
<td>82.5</td>
<td>-0.2</td>
<td>-0.2</td>
<td>0.939</td>
<td>0.927</td>
<td></td>
</tr>
<tr>
<td>14</td>
<td>50.9</td>
<td>49.3</td>
<td>1.6</td>
<td>3.2</td>
<td>0.928</td>
<td>0.927</td>
<td></td>
</tr>
<tr>
<td>15</td>
<td>59.7</td>
<td>57.7</td>
<td>2.0</td>
<td>3.5</td>
<td>0.931</td>
<td>0.926</td>
<td></td>
</tr>
<tr>
<td>16</td>
<td>52.7</td>
<td>51.7</td>
<td>1.0</td>
<td>1.9</td>
<td>0.923</td>
<td>0.923</td>
<td></td>
</tr>
<tr>
<td>17</td>
<td>48.3</td>
<td>46.8</td>
<td>1.5</td>
<td>3.2</td>
<td>0.927</td>
<td>0.925</td>
<td></td>
</tr>
<tr>
<td>18</td>
<td>62.0</td>
<td>60.1</td>
<td>1.9</td>
<td>3.2</td>
<td>0.937</td>
<td>0.925</td>
<td></td>
</tr>
<tr>
<td>19</td>
<td>56.5</td>
<td>54.8</td>
<td>1.7</td>
<td>3.1</td>
<td>0.920</td>
<td>0.916</td>
<td></td>
</tr>
<tr>
<td>20</td>
<td>60.1</td>
<td>58.9</td>
<td>1.2</td>
<td>2.0</td>
<td>0.920</td>
<td>0.916</td>
<td></td>
</tr>
<tr>
<td>21</td>
<td>99.7</td>
<td>99.4</td>
<td>0.3</td>
<td>0.3</td>
<td>0.902</td>
<td>0.895</td>
<td></td>
</tr>
<tr>
<td>22</td>
<td>88.5</td>
<td>87.6</td>
<td>0.9</td>
<td>1.0</td>
<td>0.904</td>
<td>0.900</td>
<td></td>
</tr>
<tr>
<td>23</td>
<td>68.6</td>
<td>68.4</td>
<td>0.2</td>
<td>0.3</td>
<td>0.906</td>
<td>0.902</td>
<td></td>
</tr>
<tr>
<td>24</td>
<td>54.2</td>
<td>53.7</td>
<td>0.5</td>
<td>0.9</td>
<td>0.923</td>
<td>0.918</td>
<td></td>
</tr>
<tr>
<td>25</td>
<td>95.6</td>
<td>97.2</td>
<td>-1.6</td>
<td>-1.6</td>
<td>0.926</td>
<td>0.926</td>
<td></td>
</tr>
<tr>
<td>Total</td>
<td>1576.4</td>
<td>1564.8</td>
<td>11.6</td>
<td>.07</td>
<td>0.922</td>
<td>0.919</td>
<td></td>
</tr>
</tbody>
</table>

(f) The measurement record form is shown in Fig. 8.

(g) Delivery weight of uranium is the sum of solid weight and suspended uranium weight in oil. While solid weights are those of the receiver, de-
termination of the suspended uranium weight in oil is a little complicated. When a shipper's value is (1) less than 2% larger than a receiver's, the difference is regarded as the suspended uranium weight, (2) more than 2% larger than a receiver's, the suspended weight is determined by analysis, or (3) smaller than a receiver's, the suspended weight in oil is regarded as 0.

Following this procedure, measuring experiments using solid uranium metal were carried out. The results (TABLE II) show good agreement with direct and indirect weighing. In practice both values show good agreement as shown in TABLE III.
A METHOD OF IDENTIFICATION AND INSPECTION FOR INVENTORY CONTROL OF IRRADIATED FUEL ELEMENTS*

E. M. KINDERMAN AND J. S. MILLS
STANFORD RESEARCH INSTITUTE, MENLO PARK, CALIF., UNITED STATES OF AMERICA

Abstract — Résumé — Аннотация — Resumen

A METHOD OF IDENTIFICATION AND INSPECTION FOR INVENTORY CONTROL OF IRRADIATED FUEL ELEMENTS. The reactor discharge and irradiated fuel storage locations should be examined in any complete special nuclear materials inventory. They present, however, special physical difficulties to the auditor or inventory control manager. In these locations the distance between object and observer and the poor illumination and optical distortion caused by the shielding media (water, lead glass) contribute to the difficulties of materials inventory.

In an attempt to overcome these difficulties and to provide a system for positive identification of reactor discharged fuel, we have developed a compact and convenient optical examination system utilizing a fixed examination station, a periscope, a telescope, and a camera. This system has been tested in a cobalt irradiation pool by examination of test coupons which were marked in various ways, and then treated in a hot, circulating water loop. In the test system, a water-filled periscope, 11-ft long, a catadioptric telescope placed 16 ft from the periscope, and a 35-mm camera for recording the observations were used. The telescope, camera and mountings are all readily portable since they weigh in total less than 22 lb and have maximum dimensions of 29 in.

Photographic observations of test resolution charts in air 20 ft away from the object lens of the telescope have demonstrated that the system can resolve markings 22 μm in width. This corresponds to a resolution of 0.8 second of arc. Tests performed on the water system have shown that resolution in this system is better than 50 μm at 20 ft. This corresponds to a resolution of about 1 second of arc. It is clear that the resolution can never be better than the air path system. The present system used in an actual fuel inventory should adequately reproduce any inventory markings.

METHODE D'IDENTIFICATION ET D'INSPECTION PERMETTANT DE PROCEDER A L'INVENTAIRE DES ELEMENTS COMBUSTIBLES IRRADIES. Tout inventaire complet des matières nucléaires spéciales exige un contrôle au point de déchargement du réacteur et au point de stockage du combustible irradié. Or, ce contrôle pose des difficultés matérielles particulières au vérificateur ou à l'agent chargé de l'inventaire. En effet, ces deux emplacements, la distance entre l'objet et l'observateur, le mauvais éclairage et la distorsion optique provoquée par les protections (eau, verre au plomb) gênent les opérations d'inventaire.

Pour surmonter ces difficultés et pouvoir identifier positivement le combustible déchargé, les auteurs ont mis au point un système optique peu encombrant et commode comprenant un poste fixe d'observation, un périscope, un télescope et un appareil photographique. Ce système a été soumis à des essais consistant à examiner, dans une piscine d'irradiation au cobalt, des échantillons portant différentes marques d'identification, qui ont été traités dans une boucle de haute activité à circulation d'eau. Ces essais ont été faits avec un périscope rempli d'eau, de 3,35 m de long, un télescope catadioptrique placé à 4,9 m du périscope et un appareil photographique (format 35 mm) pour enregistrer les observations. Le télescope, la caméra et les supports sont facilement transportables car ils pèsent moins de 10 kg en tout et leurs dimensions maximales n'excèdent pas 75 cm.

Les photographies dans l'air de mires graduées servant à déterminer le pouvoir de résolution et situées à 6 m de l'objectif du télescope ont montré que ce système permet de discriminer des graduations de 22 μm, ce qui correspond à un pouvoir de résolution de 0,8 secondes d'arc. Les essais effectués dans l'eau ont montré

* This work was supported under Contract 224/RB with the International Atomic Energy Agency.
qu'à une distance de 6 m, ce système peut résoudre des marques de moins de 50 µm, ce qui correspond à un pouvoir de résolution de 1 seconde d'arc environ. Il est évident que le pouvoir de résolution ne peut jamais être supérieur à celui du système utilisé dans l'air. Le système expérimenté doit permettre de distinguer toutes marques faites en vue de l'inventaire du combustible.

METODO IDENTIFICACIÓN Y PROVERKИ ПРИ ИНВЕНТАРНОМ КОНТРОЛЕ ОБЛУЧЕ-НЬХ ТОПЛИВНЫХ ЭЛЕМЕНТОВ. Места разгрузки реактора и хранения облученного топлива необходимо проверять при любой полной инвентарной проверке специальных ядерных материалов. Это, однако, связано с особыми трудностями с точки зрения физики, с которыми сталкивается контролер или руководитель операции по проведению инвентарного контроля. В этих местах расстояние между объектом и наблюдателем и плохое освещение и искажение света, вызываемое средой защиты (вода, свинцовое стекло), делают проведение инвентарной проверки еще более трудной.

Чтобы преодолеть эти трудности и создать систему, которая обеспечит опознавание выгруженного топлива из реактора, разработана компактная и удобная система оптической проверки с применением неподвижной станции проверки, перископа, телескопа и камеры. Эта система проверена в бассейне для облучения кобальта путем изучения пробных образцов с различной маркировкой, которые обрабатывали в петле с горячей циркулирующей водой. В системе проверки применяли наполненный водой перископ длиною 11 футов, катадиоптрический телескоп, расположенный в 16 футах от перископа, и 35 мм камеру для регистрации наблюдений. Телескоп, камера и сборки всегда можно передвинуть, поскольку они все вместе весят менее 22 футов и имеют максимальный размер 29 дюймов.

Фотографические наблюдения за проверочными таблицами разрешения в воздухе на расстоянии 20 футов от линзы объектива телескопа показали, что система может давать разрешение 22 микрона в ширину. Это соответствует разрешению 0,8 сек дуги. Предварительные испытания, проведенные с водой системой, показали, что разрешающая способность этой системы на расстоянии 20 футов лучше 50 микрон. Это соответствует разрешению приближенно 1 сек дуги. Ясно, однако, что разрешающая способность водной системы никогда не может быть лучше воздушной. В любом случае нынешняя система, применяемая при инвентарной проверке топлива, должна надлежащим образом воспроизводить любые инвентарные отметки.

METODOS DE IDENTIFICACION E INSPECCION PARA EL CONTROL DE LAS EXISTENCIAS DE ELEMENTOS COMBUSTIBLES IRRADIADOS. Cuando se hace un inventario completo de materiales nucleares especiales hay que inspeccionar el punto de descarga del reactor y el de almacenamiento del combustible. Pero esos dos lugares presentan dificultades materiales de carácter peculiar para los inspectores o los encargados de controlar el inventario. La distancia entre el objeto y el observador, la escasa iluminación y la distorsión óptica debida a los medios de blindaje (agua, cristal) son factores que contribuyen a dificultar las operaciones inherentes al inventario de materiales.

Para resolver estas dificultades y poder identificar positivamente el combustible descargado de un reactor, los autores han ideado un dispositivo de inspección óptica compuesto de una estación fija, un periscopio, un telescopio y una cámara fotográfica. Con este dispositivo se han hecho pruebas en una pileta de irradiación por cobalto en las que se han examinado varias muestras marcadas diversamente y tratadas en un circuito de agua caliente. Para estas pruebas se emplearon un periscopio lleno de agua, de 3,35 m de longitud, un telescopio catadióptrico colocado a 4,9 m del periscopio, y una cámara de 35 mm para registrar las observaciones. El telescopio, la cámara y los soportes son portátiles, pues pesan en total menos de 10 kg y su dimensión máxima es de 75 cm.

Las observaciones fotográficas de diagramas de resolución tomadas como pruebas en aire, a 6 m del objetivo del telescopio, pusieron de manifiesto que ese sistema puede resolver señales de comprobación de 22 µm de ancho, lo que corresponde a una resolución de 0,8 segundos de arco. Ensayos llevados a cabo con el dispositivo sumergido en agua mostraron que en este caso el poder de resolución es superior a 50 µm a 6 m, lo que corresponde a una resolución de 1 segundo de arco. Es evidente que el poder de resolución nunca puede ser superior al del sistema en aire. El sistema descrito empleado en un inventario real de combustible, reproducirá adecuadamente cualquier señal de comprobación.

1. INTRODUCTION

Accurate inventory of special nuclear materials is required because of the high specific value of all of these materials. Special nuclear ma-
terials in the form of fuel elements and fuel-element assemblies are among the easiest to identify and control. However, even these are difficult to identify positively when the fuel element or assembly is stored following irradiation in and discharge from a reactor. In addition to providing material accounting control, such identification may enhance operating efficiency and safety. For example, proper identification may prevent "green" or unaged fuel elements from being transported to the fuel reprocessing plant and processed before complete decay of iodine-131.

Visual observation and identification is the simplest of all techniques. Photographic inspection is nearly as simple and in addition has the value of permanent recording. It permits, for example, comparison of markings recorded at two stages in the fuel-element history: at final assembly in the fuel manufacturers' plants and in the irradiated fuel storage pool at the utility power plant.

The first of these stages (the fuel element manufacturers' plants) offers no difficulty or hazard to the person seeking to photograph some particular and characteristic marking. (This could be a lot or piece number, an inspector's stamp, or a weld with characteristic appearance.) The second stage requires some radiation shielding between the material and inspector. Very commonly this shielding is the water in a fuel-element storage pool. Photographic recording under water shielding or in high-radiation fields seems more difficult than in the first instance.

Under Contract 244/RB with the International Atomic Energy Agency, we have investigated the techniques and procedures required for photographing objects under water while retaining the high resolution necessary for unequivocal identification. We have also investigated techniques for preserving any marking placed on the element or assembly during its manufacture and our ability to record identification markings photographically at other stages of the fuel cycle.

2. APPARATUS

The basic apparatus used to investigate the underwater recording consisted of a "periscope", telescope, and camera. The camera used was a Nikkon "F" single-lens reflex camera. This camera uses 35-mm film. Six telescope systems were investigated, and in each case the same Nikkon "F" camera was used. All telescopes were of the catadioptric, folded-optical-system kind. This type of telescope was chosen so that a readily portable recording system could be developed for the IAEA. One each of the following telescopes was tested:

Nikkon Tele-Nikkor
Zeiss Mirator
Wollensak Mirrotel
Questar
Vega Maksutov
Tinsley
Of these particular samples the Vega Maksutov with a 6-in. objective lens had the best resolution – about 18 μm at 20 ft (air path). The Tinsley with a 5-in. objective lens had the next best resolution – 22 μm at 20 ft. The Tinsley telescope was finally chosen for the remaining tests because of its smaller size and ease of transport.

The resolution was measured in each case using the arrangement of Fig. 1. With this arrangement photographs were taken from 20 ft of a composite resolution chart made from a 1/16-scale National Bureau of Standards and an ITEK Corporation resolution chart. Measurement of film resolution was made with a Unitron measuring microscope which was also used to develop the resolution chart.

The Tinsley-Nikkon system resolution of 22 μm at 20 ft is equivalent to 0.8 second of arc. The depth of field is about 0.07 in. for a 20-ft air path at this resolution. Such resolution is better illustrated by example. Figure 2 shows two US one dollar bills, one a forgery which was kindly lent to us by officials of the US Treasury Department. Details in Fig. 2 are shown in the enlargement of Fig. 3. The photographs in Figs. 2 and 3 were taken through the telemicroscope system 20 ft from the bills.

Because of our self-imposed requirement for high resolution, we designed a simple periscope to use with the camera-telescope. The variable character of ordinary water surfaces caused by vibration or agitation of the water, thermal gradients and particulate matter in the water were all expected to degrade the resolution to a variable, but probably undesirable, degree.

The periscope was intended to minimize such difficulties. However, because of its size, a periscope would be needed at each fuel storage site (even though the rest of the system is portable). We emphasized simplicity of design in the prototype model which we tested. The principal features of the periscope are shown in Fig. 4. The tube is a 6-in. aluminum pipe, black anodized to minimize reflections. The lower window is an optical flat made from non-browning glass (Corning 7940). This window showed no evidence of discoloring, but the ordinary quartz optical flat (shown in Fig. 5) that supported the water column discolored severely because of radiation exposure during the test period. Its transparency was restored by simple annealing. The mirrors first used were high quality, "flat", silver-surfaced mirrors. Testing of the system revealed that these were not true optical flats. Substitution of a 4-in. optically flat mirror (furnished by Tinsley Optical Company) for the upper mirror improved the resolution to a marked, and satisfactory, degree. The periscope was filled with water from the lower horizontal window to the top of the upper mirror.

Lighting was one of our more troublesome problems. The periscope, as finally used, had an airtight chamber at the bottom that housed two electronic flash units (see Figs. 4 and 5). These flash units (Mecablitz 109) help to stop motion by serving effectively as a shutter. A quartzline lamp was installed outside the airtight chamber to assist the operator in focusing the telescope and making visual inspections. The electronic flash units did not prove as dependable as we had hoped. Failure of these flash units occurred, partly because the airtight chamber leaked and admitted water, and partly because the components were used extensively during the system develop-
ment. Our time schedule prohibited immediate replacement of the necessary components, therefore, the pictures on which the data for complete system resolution (discussed below) are based were taken using the quartzline lamp alone. We expect the ultimate resolution of the entire system will improve with good electronic flash lamps.

FIG. 1. Tinsley telescope (Telemicroscope)
FIG. 2. Enlargement of 35-mm photograph of bills
FIG. 3. Enlargement of FIG. 2 using telemicroscope
2.1. Procedures

Testing of the periscope-telescope camera system was carried out in a pool normally used for $^{60}$Co-irradiation experiments. For most of the tests the periscope rested on the pool bottom approximately $2\frac{1}{2}$ ft from a 1000-Ci source. The periscope rose approximately 3 ft above the water surface of the pool. The telescope and camera were mounted on ways which were placed on a laboratory table (see Appendix A, Figs. 5 and 11). The table was weighted with lead to reduce vibration. Testing consisted of photographing various objects under different lighting and water conditions in the pool and periscope. Successful resolution tests were made when the experiment pool looked as it does in Fig. 6. The detailed procedures for aligning and focusing the system are given in Appendix A.

Preferred physical manipulation techniques for filling the periscope were evolved during the test period. We found that distilled water was re-
required. The periscope must be filled in a way that prevents the formation of bubbles against the top mirror, otherwise no special techniques are required. Gentle stirring of the 11-ft distilled water column by aeration was tried but did not seem to improve resolution. Many of the initial photographs were taken using the flash lamp system. However, in the later stages of the project after the flashlamp units failed, steady illumination with the quartzline lamps was used. Exposures of 3 to $\frac{1}{2}$ second were used then.

The character of the film used and its development was of potential importance to the resolution. In our investigations we successfully used Panatomic X (Kodak) and Linograph Shellburst film. The latter is a high-contrast, high-speed film. Accufine, Dectol, and Diafine developer solutions were tried. Diafine, a two-solution developer, was used for most of the final high-resolution water photographs which were taken on Shellburst film. Two minutes were allowed for development in each of the Diafine solutions.
Final photographs of a National Bureau of Standards high contrast resolution chart, reduced by one-half and one-quarter, were used to measure the resolution of the entire system. The best resolution obtained was equivalent to at least 1 second of arc for a 25-ft air path. The actual path was 1 ft of pool water, 1 ft of air, 11 ft of periscope water, and 16 ft of air to the objective lens of the telescope. This resolution is demonstrated in Fig. 6 when the one-quarter-scale chart is definitely resolved for 56 and perhaps for 80 lines. These lines are respectively 0.0575- and 0.039-mm wide. In these charts the space between the lines is equal to the line width. The uneveness of the appearance in various positions of the figure is caused by the relative motion of the target during the relatively long time of photographic exposure. The dark portion of the upper left corner (Fig. 7) is caused by the ocular prism obstructing the optical path. The low intensity continuous light source required this long exposure.

For a more convincing demonstration see Fig. 8 where the same two one dollar bills are shown again. This time, instead of 20 ft of air, the periscope system path of 12 ft of water and 16 ft of air was used. Here, too, the imperfect illumination has caused some trouble, but the contrast between the workmanship in the two bills is evident. Another demonstration
of this method of identification is offered by Fig. 9, where the two bills are photographed in air but through the telemicroscope. A comparison of Figs. 8 and 9 demonstrates the degradation due to the water path.

In addition to the proof of the general method, which was the primary aim of our research, we investigated briefly techniques for marking stainless steel and zirconium which are the dominant cladding materials in fuels designed for water reactors. Resistance of the markings to water of high velocity, pressure, and temperature was tested briefly (up to 500 h) on test specimens immersed in a water loop operated at 425 to 450°F and 325 to 450 lb/in². The linear water intensity across the test specimens was 10 to 12 ft/sec.

Primary attention was given to hand or stamp markings on the test pieces. Various coating solutions were applied to both untreated and autoclaved material. The coatings tried are listed in Table I, along with results of loop test. All trials recorded here were with Type 304 (reactor grade) stainless steel coupons. These samples were prepared by first degreasing, washing in acetone, and then in a 20% HCl solution. All of the samples were marked with a signature. They were plated with copper, platinum (both plated and flame sputtered), zinc, gold, and ceramic paint. A control, or unplated, sample was inserted in each test series for later comparison.

For each of the coating materials and methods used, the procedures were varied until apparently adherent coatings were obtained, or until the
experimentor chose to reject the technique completely. Some of the unsuccessful attempts included any kind of plating on zirconium. The deposits formed on zirconium had, in some cases, good appearance but were not adherent.

In all cases the exposed stainless steel surfaces acquired an adherent, thin, reddish corrosion layer that did not obscure the markings. Therefore,
the results of this test seem to indicate that the reactor grade stainless, where used alone, is as good for identification purposes as any protective coating which we applied.

The coatings and markings were studied by marking micrographs of each coupon before and after submission to the corrosion loop. In addition,
**Table I**

Corrosion Test Results

<table>
<thead>
<tr>
<th>Plating Material</th>
<th>Coating Technique</th>
<th>Condition of Plating after Corrosion Tests</th>
</tr>
</thead>
<tbody>
<tr>
<td>Gold</td>
<td>Electrodeposit</td>
<td>Remained intact</td>
</tr>
<tr>
<td>Platinum</td>
<td>Electrodeposit</td>
<td>Obliterated</td>
</tr>
<tr>
<td>Platinum</td>
<td>Fired paste</td>
<td>Obliterated</td>
</tr>
<tr>
<td>Platinum</td>
<td>Flame sputtered</td>
<td>Remained intact</td>
</tr>
<tr>
<td>Ceramic</td>
<td>Fired paint</td>
<td>Obliterated</td>
</tr>
<tr>
<td>Copper</td>
<td>Electrodeposit</td>
<td>Partially intact</td>
</tr>
<tr>
<td>Zinc</td>
<td>Electrodeposit</td>
<td>Partially intact</td>
</tr>
<tr>
<td>None</td>
<td>(these were control samples)</td>
<td></td>
</tr>
</tbody>
</table>

**FIG. 10.** Gold-plated coupon with I.D. markings through periscope
selected samples were photographed through the telemicroscope-periscope system. The pictures of Figs. 10 and 11 are of the same electroplated gold sample in air and under water (through the periscope) respectively. Some of the markings of Fig. 10 were pencilled on for identification (i.e., the D, upper lefthand corner of the unplated region). The picture using the air path was taken first.

2.2. Suggestions for improvement of the optical systems

The basic design concepts of both the periscope and telescope systems have proven to be good. Because the periscope offered the most difficult problems, its possible improvement is worthy of discussion.

The flash lamp system proved the least reliable. The Mecablitz lamps used were designed for more common use in photography, but a water-tight flash lamp unit is more desirable. This can be obtained either commercially or by design and construction in the case of custom requirements.

If the flash lamp is water-tight, there is no reason for the water-tight box at the bottom of the periscope. This would eliminate one of the optical flats (Fig. 5) used to support the water column in the aluminium pipe. Thus, the number of non-browning windows needed can be reduced to one.
Rotation of the bottom chamber may be desirable where the vertical water column is held fixed. This feature could be incorporated in a final model. However, particular care must be exercised in such a design because of possible radiation damage to O-rings.

Because we found the mirrors to be the most critical optical components of the periscope system, it is apparent that high quality mirrors must be used. In the present model a low quality mirror is used in the lower position because it has the least effect on resolution. In future designs these mirrors should be the best available.

The quartzline lamp on the outside of the lower window was installed primarily for viewing and focusing, but was later used for photography. It stands away from the air-tight box and thus precludes the possibility of moving a fuel element or bundle near the window. The lamp is also subject to damage while the fuel element or assembly is being moved into position. In the final model it will be necessary to recess this lamp.

The small depth of field will make it difficult to observe large areas of curved surfaces in one photograph. Flat surfaces are preferred.

3. CONCLUSIONS

The model constructed and tested has satisfied the basic design requirements. Such a system will permit identification and detection of irregularities in reactor fuel inventories. Of course, the periscope is only necessary for those reactors using water as a loading and storage media. With moderate care one will be able to standardize the photograph size by adjustment of the target-objective lens distance to have the same effective path length for successive photographs. This would be desirable in the case where stereo projection of the negatives is used.

The telescope system has a resolution of about 0.8 second of arc. It can be reproduced at a cost of about $2000, and is easily transported in normal air travel.

ACKNOWLEDGEMENTS

It is a pleasure to acknowledge the excellent help of Mr. Zev Pressman with the photographic and optical problems and the general support of Mr. Frank Galimba.

APPENDIX A

DESCRIPTION OF TELESCOPE OPERATION

This description assumes that the telescope-ocular-camera system is assembled on the ways (base). With the aid of Figs. 12 and 13, and the following list, one can obtain sharp photographs of a target and also view the target before and after the photograph is taken.

(1) Remove the lens cap.
(2) Align the system so that the telescope is pointed at the target by
FIG. 12. Right-side photo of telescope with camera

FIG. 13. Left-side photo of telescope with camera
sighting along the edges of the telescope. Horizontal positions are obtained by simply moving the front or rear of the base in the desired direction. Vertical alignment is obtained by adjustment (the feet or the large screws) at the four corners of the base.

3. Place the ocular view prism in the optical path by pulling the prism knob (left side) all the way out and locking it by turning clockwise. If no light can be seen through the ocular eye piece, release the ocular cable release. Focus the ocular eye piece on the cross hair.

4. Estimate the distance from telescope objective lens to target, correcting for any water path by multiplying the water path distance by two-thirds and adding to the air path.

5. Move camera-ocular system along the ways until the estimated distance is read off on the index indicator (see Fig. 12). (The two large Allen screws on the lower right camera-ocular mount must be loosened for this operation.)

6. Set the fine focus micrometer (to the rear and below the camera) to 0.500 in.

7. Look through the ocular eye piece and adjust the coarse focus (micrometer on telescope) until the image is sharp.

8. If the image cannot be made sharp, move the camera-ocular mount again with the sharpest focus available on the coarse focus micrometer.

9. Lock the camera-ocular mount in place by tightening the two large Allen screws (loosened in step 5).

10. Final sharpness can be obtained by adjustment of the fine focus micrometer at the rear and below the camera.

11. The telescope may be focused on an undesirable part of the target because step 2 is not an accurate alignment procedure. Each operator can work out for himself the best procedure for final alignment. Repetition of the preceding steps may be necessary in order to obtain good results.

The next procedure makes the telescope-ocular-camera system a photographic system.

1. Set the ocular shutter speed at time.

2. Cock the ocular shutter.

3. Press the ocular cable release (a locking cable release) until the full travel is realized and lock into this position.

4. Looking through the camera eye piece, recheck the focus for sharpness.

Obtaining good photographs depends on the lighting, shutter speed, film, developer, etc. The operator should familiarize himself with the Nikkon F camera.

DISCUSSION

M.J. HIGATSBERGER: Was any blurring observed on the optical window close to the fuel element when your system was operated in a high radiation field?

E.M. KINDERMAN: The exposure times and radiation levels were of course limited, but within those limits no blurring was detected.
MATERIALS CONTROL SYSTEM FOR ENRICHED URANIUM CHEMICAL PROCESSING FACILITY FOR IRRADIATED NUCLEAR MATERIALS

F.H. TINGEY AND B.S. LEWIS
PHILLIPS PETROLEUM COMPANY,
IDAHO FALLS, IDAHO, UNITED STATES OF AMERICA

Abstract — Résumé — Аннотация — Resumen

MATERIALS CONTROL SYSTEM FOR ENRICHED URANIUM CHEMICAL PROCESSING FACILITY FOR IRRADIATED NUCLEAR MATERIALS. Systems for management and control of nuclear material in an enriched uranium chemical processing facility are not too different from those historically used in the chemical industry. The difference is one of degree rather than substance. The monetary and strategic value of the material being processed as well as the potential health and safety hazards inherent in handling nuclear material often dictate a level of effort and system detail above that which might seem reasonable to the casual observer. Among those areas of consideration which are of particular importance in the development of an effective nuclear materials management (NMM) system as based on experience gained at the Idaho Chemical Processing Plant are the following: organizational structure, accounting system, measurement system, implementation and integration of system, records and reports, internal audit, and inventory management.

SYSTEME DE CONTROLE DES MATIERES APPLICABLE A L'URANIUM ENRICHI DANS UNE INSTALLATION DE TRAITEMENT CHIMIQUE DES MATIERES NUCLEAIRES IRRADIEES. Les systèmes de gestion et de contrôle des matières nucléaires dans une installation de traitement chimique de l'uranium enrichi ne sont pas très différents de ceux qui sont classiquement utilisés dans l'industrie chimique. Cette différence est plus de degré que de nature. La valeur monétaire et stratégique des matières traitées ainsi que les risques sanitaires et les dangers inhérents à la manipulation des matières nucléaires exigent souvent des précautions et une minutie d'organisation que l'observateur non averti peut trouver exagérées. D'après l'expérience acquise à l'usine de traitement chimique de l'Idaho, les questions qui présentent une importance particulière pour la mise au point d'un système efficace de gestion des matières nucléaires sont les suivantes; organisation du système, système de comptabilité, système de mesure, application et intégration du système, registres et rapports, vérifications comptables internes, et gestion des matières à l'inventaire.

СИСТЕМА КОНТРОЛЯ МАТЕРИАЛОВ НА УСТАНОВКЕ ПО ХИМИЧЕСКОЙ ПЕРЕРАБОТКЕ ОБОГАЩЕННОГО УРАНА ДЛЯ ОБЛУЧЕНИХ ЯДЕРНЫХ МАТЕРИАЛОВ. Система административно-хозяйственного учета и контроля ядерного материала на установке по переработке обогащенного урана не многим отличается от методов, широко используемых в химической промышленности. Разница заключается скорее в степени, чем по существу. Денежная и стратегическая ценность обрабатываемого материала, а также потенциальная опасность для здоровья и безопасности, присущие обращению с ядерными материалами, часто требуют больших усилий и более подробной разработки по сравнению с тем, что может показаться разумным случайному наблюдателю. Среди такого круга рассматриваемых вопросов, которые представляют особенное значение при выработке эффективной системы административно-хозяйственного учета ядерных материалов, на основании опыта, приобретенного на установке по химической переработке в Айдахо, можно указать на следующие: организационная структура, система подотчетности и система измерения.

CONTROL DE MATERIALES EN UNA PLANTA DE TRATAMIENTO DE URANIO ENRIQUECIDO IRRADIADO. Los sistemas de administración y control de materiales nucleares adoptados en las plantas de tratamiento químico de uranio enriquecido no difieren mucho de los corrientemente empleados en la industria química. La diferencia
es de grado y no de fondo. El valor monetario y estratégico de los materiales que se tratan en la planta y los riesgos para la salud y seguridad de las personas, inherentes a la manipulación de materiales nucleares, imponen un esfuerzo y una minuciosidad muy superiores a lo que podría parecer razonable al observador ocasional. Según la experiencia adquirida en la planta de tratamiento químico de Idaho, entre las cuestiones que mayor importancia revisten para el desarrollo de un sistema eficaz de administración de materiales nucleares figuran las siguientes: estructura orgánica, sistema de contabilidad, sistema de medición, aplicación e integración del sistema, registros e informes, intervención interna y administración de las existencias.

1. INTRODUCTION

The purpose of this paper is to present an accounting system with associated techniques and procedures which has been successfully used for controlling SS materials in an irradiated fuel processing complex. Basically, the systems used for management and control in most chemical industries are similar in design and application. The primary difference is one of degree. The monetary and strategic value of SS material being processed as well as the health and safety hazards inherent to handling nuclear material often dictates a level of effort and system detail that might seem unreasonable to the casual observer.

A good accounting system is a basic requirement of effective SS materials management, but is of questionable value when the numbers that it reflects cannot be properly evaluated. It is therefore necessary to integrate the functions of accounting, chemistry, statistics, and engineering into an effective programme. A solution to integrating these functions is to implement written procedures designed to guide each department toward a common goal, materials management. The chemical operator must be oriented in material control procedures and impressed with the operational necessity of generating accurate data for material balance summation and evaluation. He should also be advised as to his responsibility for the generation and distribution of material transfer data. Analytical data must be evaluated in regard to precision and accuracy. It is therefore desirable to have a quality control laboratory supplemented with a technician certification programme to aid the statistician in evaluating the accuracy of the analytical methods and individual performances. The statistician in turn must assist or direct the accountant in making a proper evaluation regarding the numbers he uses for material balance calculations.

2. OPERATIONAL STRUCTURE

It is recommended that the nuclear materials control staff be responsible and report to the general project manager. To effect process control, data is generated by the process operator. This information can be made in turn to flow to the nuclear materials control office for inventory summation and evaluation. The accountability function under this procedure, therefore, is an adjunct of the process control mechanics. However, to evaluate properly the data, the nuclear material accountant must have at his disposal statistical counsel and direction. He must also maintain proper communications with operational personnel concerning process flow information re-
FIG. 1. Organizational chart of the Idaho Nuclear Materials Management Group

requirements. It is, therefore, proper that he should occupy a position in the organization structure to help him implement procedural changes concerning the control of fissile materials. An organization chart showing the internal structure of the Idaho Nuclear Materials Management Group and the position it occupies in the total plant operation is given in Fig. 1. Note that the Chief of the Operations Analysis Branch reports directly to the Project Manager. He also is the direct supervisor of Statistics, Safeguards (Nuclear Safety), and Nuclear Materials Control. The statistician is responsible for evaluating the data from the quality control programme and directs the accountant concerning the interpretation of the measurement data.

The safeguard function of the Operations Analysis Branch is to evaluate the nuclear safety hazards associated with the handling and transfer of SS material and devise rules and procedures to prevent an inadvertent criticality. The group is administered by the Chief of the Operations Analysis Branch, who acts as chairman of a committee whose members are specially trained in nuclear criticality problems.

Nuclear materials control is also a major responsibility of the Operations Analysis Chief. This group is functionally divided into three subsections: (1) audit and inspection (including material utilization), (2) material accounting, and (3) summation and reporting. This latter function is usually handled by a person (Nuclear Materials Control Representative) who is familiar with the overall operation and would therefore offer some supervisory aid to other members of the staff.
3. ACCOUNTING SYSTEM

3.1 Material balance area accounting

The control system, from a records point of view, must be designed to reflect the inter and intra area movement of all fissile material and provide an efficient means of obtaining perpetual inventories for the recovery process and all associated auxiliary functions. If we segregate specific plant geographical areas according to their relative purpose, we can control the flow of SS material and likewise compute perpetual inventories by the "material balance area" technique.

Each material balance area should be assigned a numerical or alphabet-numerical code designation which will descriptively identify the transfer of material from one material balance area to another. A typical code designation for a chemical processing facility is shown below:

<table>
<thead>
<tr>
<th>Material balance area</th>
<th>Area code</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fissile storage and process dissolvers</td>
<td>100</td>
</tr>
<tr>
<td>Engineering design (plant mockup)</td>
<td>200</td>
</tr>
<tr>
<td>Process recovery</td>
<td>300</td>
</tr>
<tr>
<td>Analytical laboratories</td>
<td>500</td>
</tr>
<tr>
<td>Residue and waste storage</td>
<td>700</td>
</tr>
<tr>
<td>Product storage</td>
<td>800</td>
</tr>
<tr>
<td>Chemical development study</td>
<td>900</td>
</tr>
</tbody>
</table>

The code number assigned to each material balance area can be further subdivided into more specific categories for the purpose of identifying particular locations within each material balance area. For example, the process recovery area (Code 300) can be subdivided into the following categories:
- 302 - Feed input measurement tanks for aluminium uranium processing
- 304 - Feed input measurement tanks for stainless steel uranium processing
- 330 - First cycle raffinate measurement tanks
- 335 - Process equipment waste measurement vessels
- 340 - Second cycle raffinate measurement tanks
- 380 - Product storage vessels

Assignment of material within a material balance area is to custodians.

The book inventories for each material balance area then can be compiled from transfer information supplied by the custodians of the various material balance areas. This transfer information is the key to the accounting system. Therefore, emphasis will be placed on techniques of obtaining the transfer data and compiling it into material balance area inventory values. The compilation of the transfer information will be discussed in a later section entitled "Records and reports".
3.2 Transfer data requirements

3.2.1 Reactor operation transfer form

Reactor fuel transferred to the processing facility storage basin is accompanied by transfer information concerning fuel-type piece count, identity, and fissile quantities. This information should be made available to the storage basin operator and the nuclear materials control accountant. A typical example of a transfer form required for this purpose is shown in Fig. 2. Note that provision is made to show both reactor values and chemical processing measurements. Usually, this transfer form is held by the processing operator until the chemical measurements have been determined and can be entered. At this time, a copy of the form is returned to the reactor operator. The values applied can be those determined either from plant input or output. If the output value is used, some provision must be made to identify and prorate processing losses and product recovered to the particular reactor batch.

3.2.2 Fissile storage receipt form

A typical SS storage-unloading record is shown in Fig. 3. This form has been designed to permit teleprint scale weight entries for each fuel storage bucket. Scale weights are entered by teleprint machine. This device eliminates operator error in reading and entering weight values. Fuel element identification for each storage bucket is entered to the right. Space is provided at the top of the form for recording material type, reactor operator identification symbol, shipment number, and date. The form can also be used to show intra-bucket transfer information should it become necessary to transfer fuel arrays within the basin proper. The bottom portion of the form is reserved for the signatures of the storage basin operator and supervisor.

3.2.3 SS material requisition

The material requisition form is shown as Fig. 4 and is required to show fuel element transfers from the storage buckets to the process dissolvers. Section I of the requisition contains batch information obtained from predetermined batching schedules. Section II contains similar information required in the unloading record previously discussed. Section III shows dissolver information concerning number of fuel charged, batch number, dissolver number, and date. Section IV is reserved for the nuclear materials control accountant for recording gross weights and fissile quantities.

3.2.4 Bucket record perpetual inventory file

A perpetual inventory card file can be maintained for each storage bucket located in the fuel storage basin. This card should be designed to accommodate both fuel gross weights and fissile quantities. The bucket transfer information concerning receipts, removals, and residual balance
### SS SHIPPING FORM

#### 1. FROM - (SHIPPER)
- SS STATION:
- PERSON: __________ (SS REPRESENTATIVE)
- ADDRESS: __________

#### 2. TO - (RECEIVER)
- SS STATION:
- ATTENTION: __________ (SS REPRESENTATIVE)
- DELIVER TO ADDRESS: __________

#### 4. BILL OF LADING No.:
- CARRIER: __________
- ROUTING: __________
- CAR No. & INITIALS: __________
- SEALS NOS: __________

#### 10. MATERIAL DESCRIPTION
- SHIPPER'S DATA
  - A. Transfer Authority: __________
  - B. From-Project No: __________
  - C. Date Material Shipped: __________
  - D. Date Doc. Dispatched: __________
  - E. Signature of Shipper (SS Representative): __________
  - F. Measurement Method: __________
  - G. Net Wt. - L.E.: __________
  - H. Chem. - L.E.: __________
  - I. Isotopic - L.E.: __________
  - J. Total - L.E.: __________
  - K. Lot No: __________
  - L. Con. No: __________
  - M. Gross: __________
  - N. Tare: __________
  - O. Net: __________
  - P. Element: __________
  - Q. Isotope: __________

- RECEIVER'S DATA
  - A. To-Project No: __________
  - B. Date Material Received: __________
  - C. Date Document Dispatched: __________
  - D. Signature of Receiver (SS Representative): __________
  - K. Measurement Method: __________
  - F. Net Wt. - L.E.: __________
  - G. Chem. - L.E.: __________
  - H. Isotopic - L.E.: __________
  - I. Total - L.E.: __________

#### 11. DOCUMENTATION
- a. Page: __________ of __________ Pages
- b. Copy: __________ of __________ Copies
- c. Series: __________

#### 12. SHIPPER'S DATA

#### 13. RECEIVER'S DATA

#### 9. No: __________

#### 16. TRANSFER SERIES
- A. FROM: __________
- B. TO: __________
- C. SHIPPING FORM No: __________
- D. DISTRIBUTION OF COPIES
  - 1
  - 2
  - 3
  - 4
  - 5

#### FIG. 2.
Typical SS shipping form
#### FIG. 3. Typical SS storage-unloading record

Table:

<table>
<thead>
<tr>
<th>Material Type</th>
<th>From</th>
<th>To</th>
<th>Number</th>
<th>Cask Number</th>
<th>Weight (Kg.)</th>
<th>Individual Element Identification or Count</th>
</tr>
</thead>
<tbody>
<tr>
<td>Batch No.</td>
<td>Bucket No.</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1</td>
<td>G</td>
<td>T</td>
<td>N</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>G</td>
<td>T</td>
<td>N</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>3</td>
<td>G</td>
<td>T</td>
<td>N</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>4</td>
<td>G</td>
<td>T</td>
<td>N</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>G</td>
<td>T</td>
<td>N</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>6</td>
<td>G</td>
<td>T</td>
<td>N</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>7</td>
<td>G</td>
<td>T</td>
<td>N</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>8</td>
<td>G</td>
<td>T</td>
<td>N</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>9</td>
<td>G</td>
<td>T</td>
<td>N</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>10</td>
<td>G</td>
<td>T</td>
<td>N</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>11</td>
<td>G</td>
<td>T</td>
<td>N</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>12</td>
<td>G</td>
<td>T</td>
<td>N</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>13</td>
<td>G</td>
<td>T</td>
<td>N</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>14</td>
<td>G</td>
<td>T</td>
<td>N</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>15</td>
<td>G</td>
<td>T</td>
<td>N</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>16</td>
<td>G</td>
<td>T</td>
<td>N</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>17</td>
<td>G</td>
<td>T</td>
<td>N</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>18</td>
<td>G</td>
<td>T</td>
<td>N</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>19</td>
<td>G</td>
<td>T</td>
<td>N</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>20</td>
<td>G</td>
<td>T</td>
<td>N</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>21</td>
<td>G</td>
<td>T</td>
<td>N</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>22</td>
<td>G</td>
<td>T</td>
<td>N</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>23</td>
<td>G</td>
<td>T</td>
<td>N</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>24</td>
<td>G</td>
<td>T</td>
<td>N</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

1. This constitutes an accurate record of materials transferred to the CPI BB Storage Area.

II. The above listed material was transferred within the CPI BB Storage Area from bucket(s) "A" to bucket(s) "B".

### Operator

I. This certifies acceptance of responsibility for the above listed materials placed in the CPI BB Storage Area.

Supervisor

Operator

Posted to Container Stock Ledger:

Clerk ____________ Date ____________

Audited by:

Clerk ____________ Date ____________

---

The gross weights are given in the "Unloading Record" and "SS Material Requisition Forms". The fissile quantities are available on the "Reactor Operator Transfer Form". A copy of these forms, as well as all subsequent forms discussed, is transferred to the nuclear materials control office.
3.2.5. Process recovery transfer forms

Basically, the control of uranium contained in the various streams of a separations process is contingent on the application of strategically located measurement transfer points throughout the recovery process. The decision as to the number and location of each measurement point is made on the basis of process control, nuclear safety and accountability considerations. Actually, the overriding consideration is process control once the...
plant is designed and built with nuclear safety in mind. Usually measurement points and frequency conducive to rigorous process control will satisfy the nuclear safety and accountability requirements.

After the necessary measurement points have been determined, an internal transfer form accounting system can be installed. More specifically, each transfer point measurement will be represented by an internal transfer form designed to accommodate batch transfer information between the various measurement points. Typical examples of these transfers are included below.

3.2.5.1. Feed input measurement transfer forms. The feed input measurement vessels must be equipped with precise, volumetric instrumentation as well as means for mixing and sampling of contents. The batch uranium values determined at this point are not only of prime concern to the reactor operator, but also must be used for plant input calculations in determining process imbalances. The internal transfer form used for this purpose is shown in Fig. 5. This form is initiated by the chemical operator. Note that the upper portion of the form contains information relative to volumetric calculations, while the lower portion contains wet chemistry data applicable to specific gravity, acidity, concentration, mass distribution, etc. The chemical operator enters the volumetric transfer data, checks the wet chemistry requirements and transfers the form and batch sample solution to the analytical laboratory. The analytical results are entered on the transfer form before it is forwarded to the nuclear materials control office. The upper portion of the form contains a computation square used by the control accountant to compute total batch uranium and isotope values. The code symbol in the upper right-hand corner (110-302) indicates to the chemical operator and nuclear material control accountant that this transfer represents a batch transfer from 110 (dissolving area) to 302 (feed input measurements for aluminium processing).

3.2.5.2. Waste residue measurement transfer form. Waste residue streams collected from the extraction process are transferred to instrumented vessels for waste batch measurements. The transfer form required at this transfer station is identical to the feed input measurement transfer. The only change required is the code symbol in the upper right-hand corner. The symbol in this case could be 330-710 or 340-710; 330 would designate 1st cycle extraction waste collection while 340 would represent 2nd and 3rd cycle extraction waste collection. The number 710 represents a particular waste tank storage vessel in the process waste material balance area (700).

3.2.5.3. Laboratory sample transfer form. This transfer form has been designed to transfer samples from the process streams to the analytical laboratory. Normally, these samples are taken from extraction or evaporator streams and contain sufficient concentration to warrant accountability or process control consideration. The transfer form used in this case is shown in Fig. 6. The small box near the centre of the form is used by the nuclear material control accountant to calculate sample uranium weights. Again the code symbol (360-510) is used to identify the transfer series.
3.2.5.4. **Product transfer form.** The product sample transfer form shown in Fig. 7 was designed to accommodate analytical data and product bottle weights. The upper half of the transfer form shows the product bottle, reference sample, and the lab sample net weights. The transfer is similar to those previously mentioned because it provides a calculation box for total uranium and isotopic computations. The form actually serves two purposes: (1) to transfer uranium product from the process area to the product storage vault (transfer series 380-810), and (2) to transfer product sample solution from process to the analytical laboratory (transfer series 380-510).
PHILLIPS PETROLEUM COMPANY
ATOMIC ENERGY DIVISION
REPORT OF SAMPLE TRANSFER AND ANALYSIS

<table>
<thead>
<tr>
<th>Sample Code</th>
<th>Charge Account</th>
<th>Vessel No.</th>
<th>Total Sample Vol.</th>
<th>Remarks</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>LL</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>DR</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>T</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>oC</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Sample Data (to be filled in by Analytical Laboratory):

<table>
<thead>
<tr>
<th>Sample No.</th>
<th>Bottle No.</th>
<th>Volume (ml)</th>
<th>Gross (g)</th>
<th>Tare (g)</th>
<th>Net (g)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Sample Equity (SF Department Only):

<table>
<thead>
<tr>
<th>Totals (ml)</th>
<th>(g)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Sample Determinations Desired

<table>
<thead>
<tr>
<th>Sample Results</th>
<th>Average</th>
<th>Units</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>mg/g</td>
</tr>
<tr>
<td></td>
<td></td>
<td>mg/ml</td>
</tr>
</tbody>
</table>

Sample Results Determinations Desired

<table>
<thead>
<tr>
<th>Sample Results</th>
<th>Average</th>
<th>Units</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>N</td>
</tr>
</tbody>
</table>

FIG. 6. Internal transfer form: 360-510

Transfer codes can be seen at the heading of each calculation box in the upper portion of the form.

4. MEASUREMENT SYSTEM

4.1. Chemist tester programme

One primary concern in reviewing process control data is assurance of the validity of the analytical methods and competency of the chemist or
FIG. 7. Product sample transfer form

technician involved. Too often the quality of analytical data is associated solely with the analytical method with no consideration being allowed for performance of the chemist applying the method. Although this procedure has some merit, it overlooks the importance of the individual in connection with data quality. Therefore, what is needed is some method of testing the chemist’s efficiency before he can obtain a qualification rating for analysing process control samples.

We have mentioned that the process operator must be aware of the importance of the volumetric data he supplies. The operations supervisor will
verify the transfer data to ensure himself that the operator has interpreted and recorded the correct data. However, in the case of the chemist tester this procedure is not feasible. He must assure his supervisor that he is qualified to perform certain analytical method determinations which are to be used for material control calculations.

This might be done as follows. The quality control laboratory director periodically submits synthetic process standards for routine analysis. The analyst determines the uranium concentration by different approved analytical methods. The differences between the results obtained by the analyst and the true synthetic value for a particular method are used to calculate the bias and precision of that method and enable the supervisor to make an objective appraisal of the chemist's performance. In the event of sub-standard performance, the chemist tester undergoes a training programme whereby he is instructed and practises on methods for which qualification is desired. When the individual is satisfied that he is qualified, synthetic control samples are again submitted to him for analysis. If his performance is such that he falls within the bias and/or precision established for the method or methods, his qualification will be accepted. However, if the precision of the data established by the chemist tester is outside the limits established in the quality control laboratory, it indicates that more education or intensive training is in order.

A significant bias on the part of the individual consistently obtained on a particular method may require a critical evaluation of the procedure as it is interpreted and applied by him.

4.2. Stoichiometric considerations

Although it is generally agreed that the chemical analysis of dissolved fuel batches with associated volume measurements provides the most precise measurement of residual content of spent fuels, certain problems associated with the control of this measurement and resolution of discrepancies which might arise therefrom suggest an alternate measurement technique be available for checking purposes.

One problem that may be encountered in the determination of volume is a bias in the calibration curve, thereby causing a systematic error in each batch measurement. This additive error sometimes results in a larger limit of uncertainty about the total input measurement than the sum total error of all random components. Another problem arises in those cases where input samples held for umpiring shipper-receiver differences change significantly due to evaporation and plating. In such cases they become of little value in settling disputes.

An effective alternative in certain cases is to determine the burn-up values stoichiometrically from pre and post mass distributions of the processed fuel batches. We define for a given segment of fuel: \( U \) = Uranium content, \( E = ^{235}U \) fraction, \( Z = ^{236}U \) fraction, with subscripts 0 and 1 denoting pre- and post-irradiated values, respectively. In the first method it is assumed only that the quantities of isotopes \(^{235}U\) and \(^{236}U\) originally present change significantly through fission or capture during irradiation. If other isotopes are involved the method can be easily extended to cover those
cases provided at least one isotope quantity remains essentially unchanged during irradiation.

Using the above notation, one can write:

\[ U_0 - U_1 = U_0 E_0 - U_1 E_1 + U_0 Z_0 - U_1 Z_1 = U_0 (E_0 + Z_0) - U_1 (E_1 + Z_1) \]

or

\[ \text{Burn-up} = U_0 E_0 - U_1 E_1 = U_0 \left[ E_0 - E_1 \left( \frac{1 - E_0 - Z_0}{1 - E_1 - Z_1} \right) \right] \]

Certain observations are worth noting:

(a) In any given case burn-up involves multiplying \( U_0 \) by a factor that is considerably less than 1, and hence the calculation is somewhat self-compensating for errors in \( U_0 \).

(b) The measurements \( E_1, Z_1 \) are highly reproducible and amenable to rigid bias control.

The second method is based upon the fission "stoichiometry" of \( ^{235}\text{U} \). Using the above notation it can be shown that:

\[ U_1 = U_0 \left( \frac{\alpha + Z_0}{\alpha + Z_1} \right) \]

where \( \alpha = \) capture-to-fission cross-section ratio for \( ^{235}\text{U} \).

Thus the burn-up can be calculated from

\[ \text{Burn-up} = U_0 \left[ E_0 - E_1 \left( \frac{\alpha + Z_0}{\alpha + Z_1} \right) \right] \]

The observations for the first method apply here also. The primary difficulty is the determination of an acceptable value for \( \alpha \) since \( \alpha \) is energy dependent and could vary widely over a given reactor. On an entire core basis, however, a theoretical spectrum average value could be derived a priori and thus serve as a basis for calculation.

5. RECORDS AND REPORTS

The accounting records should be designed to reflect internal measurement data transaction and inventory summation. The process data entered into the records must also be oriented such that the information can be easily retrieved for production reports, criticality control, operational efficiency, cost, fiscal accounting, and nuclear parameter estimations.

The record system at the Idaho Chemical Processing Plant is designed to fulfill these requirements. It consists primarily of one "Book of Original Entry" (Within Area Transfer Journal) and one "Book of Final Entry" (Material Balance Summary Ledger). The Within Area Transfer Journal is used to record daily within area material movement transactions. The summations of this journal are periodically transferred to the Material Balance
Summary Ledger for material balance inventory purposes. Perhaps the best way to describe the purpose of the transfer journal is to discuss the source from which it is compiled. The processing flow diagram depicting the various measurement points for materials control is given in Fig. 8. Note that the material balance areas discussed earlier have well defined boundaries. The movement of SS material affecting more than one material balance area is accompanied by an internal transfer form. The transfer code also mentioned earlier defines the locations involved in the transaction. Typical examples of the various transfer codes are shown; to mention a few, 110-302 represents dissolver fuel batches transferred to the processing parameter for input measurement determination (aluminium only); 330-710 represents 1st cycle extraction raffinate transferred to permanent waste storage; 360-510 shows transfer of concentrated stream samples to the analytical laboratory; and 380-810 represents transfer of product to the product storage vault.

The accumulation of each series transfer by batch sequence represents a Within Area Transfer Journal. All series may be posted to a formalized journal or bound individually for each reporting period. In either case, the total for each series is transferred to the Material Balance Summary Ledger. The ledger shown in Fig. 9 is comprised of a plant balance summary column (left) and seven material balance areas. The memorandum column at the extreme right is used to accumulate waste loss transactions for the purpose of maintaining perpetual accounting of the waste storage tanks. The description column at the left shows beginning inventory, receipts, shipments, intra area transfers, losses, and ending inventory. Off site receipts and/or shipments are posted directly to the ledger from the transfer form shown in Fig. 2. The intra area transfers are posted from the Within Area Transfer Journal. Losses are normally indicated by the transfer series 330-710, 335-710, and 340-710. Laboratory and research and development losses are also entered with a corresponding entry required in the Plant Balance column and the material balance area affected. All removals and receipts are indicated by red and black entries respectively. The ending inventories are computed at the bottom of the ledger. The total plant inventory must be equal to the summation of the respective material balance area inventory values.

The perpetual accounting of inventory items (by piece count and identification) can be maintained on index or computer cards. The raw data are obtained from the internal transfer form. Inventory listings represented by the computer or file cards are prepared completely independent of the record function. The total uranium values perpetually maintained for a particular material balance area must be in agreement with the total inventory value shown in the Summary Ledger. This reconciliation is made at intervals not to exceed one month.

If computer facilities are available, it would be highly advantageous to use computer cards rather than a file index. This arrangement would facilitate the easy retrieval of various inventory composition reports regarding material type, programmatic assignment, enrichment, etc.
FIG. 8. Enriched process stream SS material accountability flow diagram
6. INTERNAL AUDIT

We have discussed the procedures by which material balance area inventories are compiled. The nuclear materials manager must use the information the procedures are designed to produce to make decisions. One important criterion for making these decisions is the periodic physical inventory and records audit.

Most industries welcome annual audits by certified public accountants. The auditor provides management with an independent appraisal of the accounting system and performance and issues a formal statement concerning the fairness of the inventory values. The nuclear materials control group should have a member on its staff who is capable of rendering the same service as the certified public accountant. It is unreasonable to expect that the records and reports issued to management will contain useful information when the measurement data from which they are generated cannot be properly evaluated. In addition to determining the accuracy of the measurement data, the nuclear materials auditor must be trained to observe weakness in an established accounting system and issue recommendations and suggestions to management for administrative action. If management shows an interest in materials control, it will stimulate the people transmitting measurement data to be more interested in producing good numbers. Too often, if nothing is done to change the quality of the numbers, one is inclined to lose interest and the system disintegrates. It is, therefore, mandatory that the auditor survey all standard practice manuals regarding the control of SS material and assure management that deviations from these procedures are not conducive to good SS material management. If changes of existing procedures are in order, it should be his responsibility to detect the inadequacies and obtain a decision from management to implement corrective action.

At the Idaho Chemical Processing Plant, the Nuclear Materials Control Auditor has been assigned an additional responsibility. He must review the Standard Practice Manuals concerning criticality safeguards regulations established and approved by the Nuclear Safety Committee. These procedures define fissile quantity limitations for each storage location within the chemical processing area. These limitations must be adhered to for health and safety reasons. If deviations from established procedures are observed, it is the auditor’s responsibility to report same to the Nuclear Safety Committee Chairman.

7. INVENTORY MANAGEMENT

Too often, we find that certain SS material quantities accumulate at a chemical processing plant and remain a burden to management in terms of storage costs and health and safety hazards. It is sound business practice to have a high inventory turnover. The nuclear materials control manager can play an important role in inventory management by establishing procedures to excess, recover, or transfer those items which are of no further use to process or technical personnel. For instance, the NMM accountant has at his disposal all the information concerning the inventory holdings of each custodian. He is therefore the most logical man to co-ordinate the
reduction of inventory items. Periodically, he should prepare a list of those items he finds (during the physical inventory audit) to be excess to each custodian's needs. This list is submitted (through management channels) to all custodians for their review. In those cases where a particular individual wishes to retain custody of a particular item, he must justify (in writing) the programmatical need for such item. This justification is then submitted to management. The list not only serves to inform management of unnecessary inventory items, but can represent valuable information to custodians who are contemplating the purchase of a specific item which has been declared excess by another person. We have found such to be the case in several instances at the Idaho Chemical Processing Plant.

DISCUSSION

Y. FRUCHARD: You are in charge of a group dealing with criticality risks, and I take it that normally this group simply applies the regulations prescribed by an external authority. I should like to know whether the regulations in question provide for every eventuality or whether your group is supposed to supplement them with certain procedural rules of its own.

F.H. TINGEY: The Nuclear Safety Committee does not normally establish procedure, mainly because it cannot always be familiar with the engineering problems associated with particular transfers of material. The operational or technical group that wishes to move material generally lays down the procedure in great detail, and these instructions, together with associated drawings, are submitted to the Nuclear Safety Committee. Independent calculations, if required, are assigned to individual Committee members (and in fact we usually have two independent calculations made). The results are reviewed, after which the Committee indicates whether it believes any hazard is involved in the proposed operation. If it does, its next step is to make recommendations as to how the hazard can be eliminated.

Y. FRUCHARD: During the first stage of operations in a fuel processing plant it is rather difficult to keep an accurate materials balance. Does one, during this stage, rely entirely on the materials balance for nuclear safety or are there other safeguards as well?

F.H. TINGEY: Several different systems are used in chemical processing plants. The main one ensures safety by imposing restrictions on geometry and concentration. A secondary safeguard consists in a batch system which places limitations on $^{235}U$ content. The basis of this operation is what we call a "dissolver operating line" - a limit on the quantity of material allowed in the dissolver. To put it as briefly as possible, the material entering the dissolver must not exceed a specified quantity which takes into account all uncertainties in (i) the amount of uranium charged, (ii) the reactor burn-up calculations, and (iii) the cumulative measurements made in the dissolver. In practice this system forces us to make periodic heel reductions in the dissolver, which can then be regarded as clean again so that the cycle can be repeated.

C.R. JASNY: I should like to comment briefly on one aspect of the problem dealt with in Mr. Tingey's paper. Even organizational arrangements which give the SS materials control officer ready access to top management
cannot prevent every disagreement between operators, analysts and accountants. However, the establishment at Oak Ridge of an "accountability committee" chaired by the head of the recovery plant, and composed of representatives of production, laboratory and SS control, has helped to keep such disagreements to a minimum. The committee plans inventories, reviews their results, investigates discrepancies and recommends changes in procedure or equipment.
RECORDING, REPORTING AND GENERATION OF QUANTITATIVE DATA

(Session IV)
METHODES D'INVENTAIRE
DANS UN ETABLISSEMENT DE TRANSFORMATION

G. BILLY
COMMISSARIAT A L'ENERGIE ATOMIQUE, PARIS, FRANCE

Abstract — Résumé — Аннотация — Resumen

INVENTORY METHODS IN A CONVERSION PLANT. The main purposes of the inventories are: to safeguard the interests of the Commissariat, to evaluate losses, and to check the measures taken for waste storage. On stocktaking the investigator can either carry out a physical inventory, participate in the inventory, or check the inventory. The latter method gives the best results. The stocktaking operations, preceded by a preparatory meeting to determine the methods to be applied, are carried out in two stages: the weight inventory check and the physical auditing check.

Some questions arising in connection with the inventory are disputes between consignors and consignees regarding weight and content; choice of weighing equipment, waste, and evaluation of losses.

Inventory methods should not differ greatly from one country to another. It would appear desirable to stress the difficulties which arise during operation and to consider jointly ways of surmounting them.

METHODES D'INVENTAIRE DANS UN ETABLISSEMENT DE TRANSFORMATION. Les inventaires ont notamment pour but de sauvegarder les intérêts du Commissariat, évaluer le montant des pertes, et contrôler les mesures prises pour le stockage des déchets. Pour le recensement, l'enquêteur peut effectuer matériellement l'inventaire, y participer, ou le vérifier. Cette dernière méthode donne les meilleurs résultats. Les opérations de recensement, précédées d'une réunion préparatoire pour définir les modalités à appliquer, ont lieu en deux temps: la vérification de l'inventaire pondéral, et la vérification de la comptabilité physique.

Les questions qui peuvent être soulevées en cours d'inventaire sont des contestations sur le poids et les teneurs entre expéditeurs et destinataires, le choix du matériel de pesée, les déchets et l'évaluation des pertes.

Les méthodes d'inventaire ne doivent guère différer d'un pays à l'autre. Il semble préférable de souligner les difficultés qui se présentent en cours d'opération et d'étudier, en commun, les moyens qui permettent de les surmonter.

МЕТОДЫ ИНВЕНТАРИЗАЦИИ НА ПРЕДПРИЯТИИ ПО ПЕРЕРАБОТКЕ ЯДЕРНЫХ МАТЕРИАЛОВ. Основными задачами инвентаризации являются: соблюдение интересов Комиссариата; определение размеров потерь; контроль мер, принимаемых для хранения отходов. Инспектирующее лицо может проводить фактическую инвентаризацию, участвовать в ней или проверять ее. Этот последний метод дает наилучшие результаты. Операции по учету, которым предшествует подготовительное совещание для определения способов учета, проводятся в два этапа: сначала проверяется наличие по весу, а затем документация.

Вопросы, возникающие в связи с инвентаризацией и касающиеся весов, содержания поковок, выбора весов для взвешивания, норм отходов и оценки потерь, разрешаются отправителем и получателем.

Методы инвентаризации в одном государстве не должны сильно отличаться от таковых в другом государстве. Необходимо подчеркнуть трудности, возникающие в процессе инвентаризации, и желательность отыскания совместных путей их преодоления.

METODOS DE INVENTARIO EN UNA PLANTA DE TRANSFORMACION. La finalidad principal de los inventarios es la de salvaguardar los intereses de la Comisión, evaluar la cuantía de las pérdidas y controlar las medidas adoptadas para el almacenaje de los desechos. Para el recuento de las existencias el inspector puede efectuar materialmente el inventario, participar en él o comprobarlo. Este último procedimiento es el que da mejores resultados. Las operaciones de recuento, precedidas de una reunión preparatoria para fijar las modalidades, se efectúan en dos tiempos: la comprobación del inventario ponderal y la comprobación de la contabilidad física.

Las cuestiones que pueden plantearse durante el inventario se refieren a las discrepancias en el peso y el contenido, la elección del equipo de pesada, los residuos y la evaluación de las pérdidas.
1. INTRODUCTION

Des produits comme l'uranium, le plutonium, le thorium et leurs composés sont soumis, par le Commissariat à l'énergie atomique, à une réglementation particulière définie par un Code des matières de base. Ces produits peuvent être détenus soit par le CEA lui-même soit, pour son compte, par des entreprises ou organismes extérieurs.

Pour des raisons de sécurité, mais aussi pour des raisons fiscales, le CEA s'est toujours refusé à vendre des matières de base sur le territoire français, à l'exception de très faibles quantités destinées principalement à des laboratoires de produits chimiques. Les entreprises de transformation, notamment, agissent donc à titre de façonniers et si elles sont détentrices desdits produits, elles n'en sont pas propriétaires.

Toute quantité de matières de base appartenant au CEA ou louée par lui et entreposée ailleurs que dans ses établissements entre dans la catégorie des «stocks à l'extérieur». Un des départements du CEA, dit alors Département maître d'œuvre, est toujours responsable de l'un de ces derniers.

L'ensemble formé par les établissements du CEA et les entreprises ou organismes extérieurs est divisé en «unités de gestion des matières de base».

Certaines entreprises, dont l'activité principale, pour ne pas dire essentielle, est de transformer les matières en question, constituent des unités de gestion indépendantes placées sous la responsabilité des Départements maître d'œuvre. Dans d'autres cas, l'ensemble des stocks à l'extérieur d'un même département constitue une seule unité de gestion.

Tout mouvement entre deux unités de gestion fait l'objet d'un bon de livraison; toute variation quantitative du stock en dépôt dans l'une d'elles fait l'objet, soit d'un bon d'entrée s'il s'agit d'une acquisition ou d'une location, soit d'un bon de variation de stock dans les autres cas.

Chaque unité de gestion dispose d'un ou plusieurs registres de mouvements sur lesquels sont ouverts chaque année autant de comptes que cette unité détient de matières de nature et de composition isotopique différentes et, pour certaines matières, d'origines différentes. Les bons sont enregistrés au jour le jour, dans l'ordre de leur arrivée, et les comptes sont arrêtés à la fin de chaque mois. Cette comptabilité physique est récapitulée à l'échelon du département et par certains services centraux.

Les établissements de production ou de transformation diffusent mensuellement un bilan matières de leur exploitation. Ce bilan doit se raccorder exactement avec le registre des mouvements arrêté à la fin du mois. Afin de pouvoir comparer le stock comptable au stock physique l'entreprise doit procéder à des inventaires annuels.

Ces inventaires sont vérifiés sur place et sur pièces par des représentants du CEA, périodiquement pour les entreprises importantes, occasionnellement pour les autres. Ces contrôles ont pour but de sauvegarder les intérêts du CEA et notamment de permettre de...
- connaître dans toute la mesure du possible le montant exact des pertes,
- vérifier la façon dont les déchets sont stockés afin d'en rendre le recyclage économique,
- étudier la mise en place par l'entreprise d'une procédure qui lui permette de tenir une comptabilité matières correcte,
- s'assurer, le cas échéant, de l'application des règles de sécurité.

En outre, les mouvements et l'état des stocks de certains produits font l'objet de déclarations à des organismes extérieurs - EURATOM, USAEC - qui procèdent à des recensements sur place. Il convient donc également de s'assurer que les déclarations faites à ces organismes sont bien conformes à la réalité.

2. MODALITÉS DU RECENSEMENT

Plusieurs méthodes s'offrent aux représentants du CEA pour accomplir leur tâche dans le domaine en question:
- l'enquêteur effectue matériellement l'inventaire,
- il y participe concurremment avec l'entreprise,
- il contrôle l'inventaire préparé par le transformateur.

Cette dernière méthode a jusqu'à présent donné de très bons résultats; elle permet, dans les meilleurs délais, d'une part, de vérifier rapidement l'état des existants d'après les feuilles d'inventaire; de l'autre; de rapprocher les chiffres ainsi obtenus des différents postes du bilan matières d'exploitation.

2.1. Préparation de l'inventaire

La date et les modalités de l'inventaire sont précisées au cours d'une réunion préparatoire à laquelle participent, outre l'enquêteur, les représentants de l'usine et les agents qualifiés du Département maître d'œuvre.

La date, le plus souvent, est laissée à la libre appréciation de l'entreprise. Elle correspond dans toute la mesure du possible à la fermeture des ateliers: fin de campagne, période d'entretien ou départ en vacances. À la limite, le recensement des ateliers aura lieu un samedi et un dimanche. Le cas s'est déjà produit, les jours précédents ou suivants étant affectés au magasin. Les ateliers auront été approvisionnés en conséquence afin de limiter, voire de supprimer tout mouvement entre ces deux grandes zones.

Quant aux modalités, elles ont pour but, le recensement étant effectué, de faciliter le rapprochement des résultats avec les postes du bilan et de permettre, pour chaque rubrique, la conversion en élément contenu des poids nets tels qu'ils figurent dans les feuilles d'inventaire.

Les bilans matières d'exploitation sont établis par nature de produits, par composition isotopique pour une nature donnée, et éventuellement par origine lorsqu'il s'agit de matières fournies par un pays étranger. Ils distinguent entre autres

- le stock en magasin: matières premières,
  produits finis,
- le volant de fabrication,
- les déchets: recyclables par l'entreprise,
  recyclables par une usine de production,
  jugés sans valeur.

Un premier travail consiste donc pour l'entreprise à
- définir les secteurs qui détiennent des matières de base,
- désigner pour chacun d'eux, le responsable et le personnel d'exécution,
- attribuer à ces secteurs des tranches de numéros dont l'importance est fonction de l'ampleur du stock.

Chaque responsable reçoit, en quantité suffisante, un lot d'étiquettes où figurent:
- la nature du produit et sa teneur isotopique (il convient éventuellement de préciser l'origine),
- le type de l'alliage,
- la teneur du produit en alliage,
- la désignation du produit,
- le nombre de pièces,
- le numéro de la caisse ou du fût,
- le numéro de la palette,
- le poids brut, la tare et le poids net.

Enfin, il est opportun de rappeler, lorsqu'il s'agit par exemple d'une usine de fabrication d'éléments combustibles, que
- les numéros des billettes et lingots doivent être apparents sur les caisses d'origine avec le poids qui figure dans les bordereaux d'expédition du CEA,
- les barreaux usinés et les cartouches font l'objet d'un dénombrement affecté d'un poids moyen unitaire défini par le CEA pour chaque type de fabrication.

2.2. Déroulement des opérations

Chaque qualité de produit et chaque type d'alliage seront inventoriés séparément selon les rubriques du bilan mensuel, chaque caisse ou fût ayant été taré préalablement. Les étiquettes sont remplies au fur et à mesure des pesées ou des dénombrements, numérotées selon les indications reçues et accrochées aux emballages. Les caisses et fûts sont ensuite rangés sur des palettes numérotées au moment de leur usage. Une même palette ne supportera en règle générale que des emballages contenant des produits identiques parvenus à un même stade d'élaboration. Une fiche récapitulative est établie par palette; elle comprend:
- le numéro des caisses,
- la désignation des pièces par caisse et leur nombre,
- la nature du produit et, éventuellement, le type de l'alliage,
- le poids brut, la tare et le poids net.

2.3. Vérification de l'inventaire

L'enquêteur procède en premier lieu à la reconnaissance des lots. Il s'assure, par secteur, qu'ils sont bien répertoriés et qu'il y a concordance
entre les mentions portées par les étiquettes et celles qui figurent sur la fiche récapitulative. L'expérience a montré que cette première étape est indispensable. Il suffit que l'entreprise ait omis de peser ou d'enregistrer quelques caisses, les poids variant entre 100 et 200 kg, pour que le recensement ne puisse être utilement exploité.

Les vérifications de lots ont lieu après cette reconnaissance.

Les pesées de contrôle sont effectuées par sondages dont l'importance est fonction de la présentation et des écarts constatés en cours d'opération. En fait, les poids annoncés correspondent généralement à la réalité, compte tenu d'ailleurs de la sensibilité de la bascule utilisée.

Les produits, qui font l'objet d'un dénombrement, sont recensés en totalité. Il suffira alors pour les billettes et lingots, après avoir vérifié les numéros, de considérer le poids donné par l'expéditeur, et pour les barreaux usinés ou les éléments combustibles, de multiplier les quantités de chaque type différent par le poids moyen défini statistiquement par le CEA. Ce sont là encore de simples vérifications, car l'entreprise a déjà effectué les mêmes opérations pour remplir ses étiquettes.

2.4. Vérification de la comptabilité matières

Tant la préparation du recensement que l'inventaire physique lui-même ont eu pour objectif de reconstituer les différents postes du bilan matières d'exploitation à partir des fiches récapitulatives qui reflètent l'état exact des existants.

Les agents de l'entreprise et les représentants du Département maître d'œuvre établissent ainsi un bilan qui correspond à la réalité, tandis que l'enquêteur, à partir des mêmes fiches, répartit les produits stockés par catégorie d'uranium et par type d'alliage. Les résultats de ces dépouillements, effectués de deux manières différentes, sont ensuite comparés entre eux. Les divergences, lorsqu'elles existent, donnent lieu à de nouveaux pointages. Il paraît indispensable d'effectuer ce travail sur place sitôt le recensement terminé.

En principe, il conviendrait que l'entreprise ait également mis à jour le bilan de fabrication à la date de l'inventaire en tenant compte des mouvements intervenus depuis la fin du mois précédent. Cette opération demande cependant un certain délai car bien souvent la même cellule de l'usine prépare l'inventaire, participe à sa réalisation, seconde l'enquêteur et ventile les résultats.

Il est rare qu'on doive attendre plus de trois semaines la répartition de ce « stock comptable ». Il est alors aisé de déterminer, soit en poids net soit après conversion en élément contenu, outre la quantité de matières entreposées dans l'usine et déjà connue:
- la différence d'inventaire,
- le ou les postes du bilan qui devront être régularisés.

Un procès-verbal d'inventaire est établi contradictoirement, qui rappelle le montant du stock physique. Il est signé par le responsable de l'entreprise, le représentant du Département maître d'œuvre et l'enquêteur. De son côté, le Département en question fait émettre un bon de variation de stock qui permet aux gestionnaires centraux des matières de base de mettre à jour leur stock comptable.
S'il paraît normal qu'un recensement permette de déceler des pertes non déclarées, il est en revanche bien inquiétant de constater un «boni» d'inventaire. Bien souvent, le boni provient d'entrées non enregistrées, l'expéditeur ayant omis de rédiger un bon de livraison. En conséquence l'émission d'un bon de variation de stock a pour effet d'accroître le stock global du CEA aussi longtemps qu'il n'a pas été possible de déterminer l'origine du ou des mouvements en litige.

3. QUESTIONS SOULÈVÉES EN COURS D'INVENTAIRE

On examine ici certaines des plus importantes et nombreuses questions susceptibles d'être soulevées en cours d'inventaire.

3.1. Contestation sur les poids et teneurs

Les destinataires constatent parfois en leur défaveur des écarts dont l'importance ne peut être attribuée ni à l'imperfection du matériel de pesée, ni aux méthodes d'analyse. Quoi qu'il en soit, les poids indiqués par l'expéditeur doivent toujours être reportés, sans modification, dans la comptabilité matières. Si la différence se situe en dehors de la fourchette communément admise pour un produit donné, le destinataire, avec l'accord du Département, peut alors établir un bon de variation de stock en y portant, par exemple, la mention «différence sur pesée». Si le destinataire estime ne pas recevoir, à chaque livraison, la quantité annoncée, le Département maître d'œuvre envoie un contrôleur pour vérifier le bien-fondé de la réclamation, et, le cas échéant, attire l'attention de l'expéditeur sur le libellé de ses bordereaux de livraison.

Il n'en demeure pas moins que les cas sont rares où le poids annoncé correspond exactement au poids reçu; mais les différences sont assez minimes pour ne pas provoquer de réaction. Toutefois, en fin d'exercice, la différence entre la somme des entrées «comptables» et la somme des matières premières utilisées et enregistrées dans les cahiers des ateliers de fabrication peut se traduire par une perte non négligeable. Il semble alors préférable de distinguer celle-ci des pertes en fabrication, en la qualifiant arbitrairement d'occulte.

3.2. Choix du matériel de pesée

Le matériel de pesée est vérifié périodiquement par un service officiel du gouvernement français; aussi n'entre-t-il pas dans les intentions de l'enquêteur de traiter des bascules défectueuses qu'il lui sera toujours loisible de ne pas utiliser. En revanche il semble utile d'insister sur les avantages des bascules automatiques dites à tickets. D'une part, seule l'erreur systématique subsiste, l'erreur subjective étant ainsi supprimée; d'autre part, au fur et à mesure de leur impression, les tickets sont fixés sur les emballages correspondants. Ainsi les erreurs de lecture et les erreurs de transcription sont supprimées, tandis que la numérotation automatique des tickets permet de s'assurer de la continuité des opérations. S'il n'est pas possible de disposer d'une telle bascule, il convient de prendre cer-
taines précautions afin d'éviter les erreurs de lecture dues à la fatigue visuelle, erreurs effectuées, en général, toujours dans le même sens.

La meilleure solution consiste à affecter deux agents à la lecture de l'aiguille; l'un d'eux relèvera, par exemple, du service détenteur; l'autre des services généraux.

Lorsque les effectifs ne permettent pas d'immobiliser deux personnes, il ne faut pas craindre de procéder à des relèves fréquentes.

Après avoir adopté l'une de ces deux méthodes, on a pu constater que les poids indiqués par l'usine correspondaient bien aux résultats des pesées de contrôle.

3.3. Présentation des déchets

C'est là, semble-t-il, un des postes qui pèsent lourdement sur l'exactitude du stock physique. La vérification en est difficile, sinon impossible, car les déchets sont fréquemment mélangés à de l'huile pour des raisons de sécurité. En outre, les poids indiqués n'ont pas toujours une grande signification car la teneur en élément contenu résulte parfois de l'estimation des techniciens de l'entreprise. L'enquêteur se trouve donc, selon la nature des déchets, en présence de teneurs imprécises ou en présence de teneurs forfaitaires et provisoires fixées par les usines de retraitement.

En fait, la perte réelle n'est connue qu'après le recyclage des déchets. Comme ce recyclage est effectué par campagnes, il peut s'écouler de longs mois entre la date de l'inventaire et la réception d'informations précises concernant ces déchets.

L'enquêteur profitera de la vérification de ce poste pour s'assurer du bon état de conservation des produits à récupérer. Le façonnier doit, en effet, éviter de recueillir dans un même fût des déchets d'origines et d'alliages divers. Le principe est le même, que la récupération ait lieu dans un des ateliers de l'entreprise ou, après transfert, dans une usine de retraitement. Les déchets doivent être individualisés en vue d'un recyclage économique.

Il se présente cependant des cas où ce principe n'est pas applicable: les pièces à usiner, notamment, peuvent être constituées par des alliages d'un type différent, voire par des métaux divers. Là encore, il convient de faire estimer par les ingénieurs la part qui revient à chacun des constituants.

3.4. Evaluation des pertes

Pour une campagne de production ou une série de fabrication donnée, il est important de connaître le montant des pertes globales. Ces pertes comprennent:
- les pertes déclarées par l'entreprise dans chacun de ses bilans mensuels,
- les pertes occultes dont il a déjà été question,
- les pertes sur déchets,
- les pertes qui résultent de la différence d'inventaire.

Dans certaines entreprises, il convient de tenir compte de dépôts dans les appareils et les conduites. Ces dépôts font fréquemment l'objet d'une rubrique spéciale dans les bilans matières sous le nom de «hold up».
Lorsque cette rubrique existe elle sera déduite de la différence d'inventaire et figurera dans un poste « matières non inventoriables ». Lorsqu'elle n'existe pas, un renvoi indiquera le montant estimé du hold up. Il est aisé de concevoir le caractère aléatoire de cette notion. L'enquêteur avant de donner un chiffre aura également recours au technicien chargé de suivre pour le compte du CEA les travaux de l'usine.

3.5. Cas de sous-traitants

C'est là un problème annexe qui se pose lorsque le façonnier, avec l'accord du CEA d'ailleurs, sous-traite une partie ou un stade quelconque de la fabrication. La prise en compte des bons de livraison - entrées et sorties - au titre de l'entreprise principale ne permet pas de saisir le montant exact des pertes. On peut en donner l'exemple suivant (les chiffres sont donnés en élément contenu):

Soit A l'entreprise principale, et B le sous-traitant. Le CEA livre, par exemple, 500 kg de matières premières à A. A en expédie 300 kg à B, qui lui renvoie 200 kg de produits ouvrés; par hypothèse, les pertes non déclarées de B sont de 100 kg. A achève la fabrication, adresse au CEA les produits finis à raison de 150 kg et conserve une certaine quantité de déchets, soit 50 kg, qui proviennent de cette ultime transformation.

Le bilan matières de A est établi comme suit:

<table>
<thead>
<tr>
<th>Entrées du CEA</th>
<th>500 kg</th>
<th>Sorties vers B</th>
<th>300 kg</th>
</tr>
</thead>
<tbody>
<tr>
<td>de B</td>
<td>200 kg</td>
<td>vers le CEA</td>
<td>150 kg</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>450 kg</td>
</tr>
</tbody>
</table>

Le stock comptable est donc de 250 kg et se décompose en:

- matières premières 200 kg
- déchets 50 kg

Ces quantités sont vérifiées au cours d'un recensement et la différence d'inventaire est nulle. Faut-il en conclure que la transformation a été effectuée sans pertes?

C'est pourquoi, il semble opportun, dans ce cas, d'assimiler les deux façonniers A et B à une seule entreprise. Le bilan matières est présenté ainsi:

<table>
<thead>
<tr>
<th>Entrées du CEA</th>
<th>500 kg</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sorties vers le CEA</td>
<td>150 kg</td>
</tr>
<tr>
<td>Stock comptable</td>
<td>350 kg</td>
</tr>
</tbody>
</table>

Le stock physique est toujours de 250 kg et la différence d'inventaire fait ressortir une perte non déclarée de 100 kg, conforme à la réalité.

Si les sous-traitants éventuels détiennent également des matières de base à la date de l'inventaire, il conviendra de procéder à un recensement dans l'ensemble des usines intéressées par une fabrication donnée.
4. CONCLUSION

Il semble bien difficile d'apporter des idées neuves sur les méthodes proprement dites d'inventaire dans un établissement de transformation. Les phases essentielles ont été traitées dans la seconde partie de ce mémoire; elles résultent d'une mise au point effectuée en liaison avec les industriels au fur et à mesure que des difficultés matérielles surgissent en cours d'opération. Il n'en demeure pas moins une certaine imprécision dans l'exploitation des résultats. Quelques exemples en ont été donnés dans la troisième partie.

Il serait particulièrement intéressant de connaître les moyens utilisés par les représentants des autres nations qui participent à ce Colloque, en vue de limiter ou de supprimer la part des valeurs estimées, encore trop importante, dans la détermination d'un stock physique.

DISCUSSION

D.E. GEORGE: Various speakers, including yourself, have taken it for granted that materials of differing isotopic content may not be mixed. Does this apply even when there may be an economic advantage in mixing?

G. BILLY: Much enriched uranium comes from the United States. It is subjected to USAEC control and I do not think different isotopic contents can be mixed without prior consent. In the event of mixing, however, the product obtained will call for a materials accounting procedure corresponding to the new isotopic content.

F. RONTEIX: I should like to add to what Mr. Billy has said in response to Mr. George's question. In the central enriched-uranium store in France we have a number of waste lots containing small quantities of uranium generally of low enrichment. We propose mixing them for reprocessing. The quantities involved are so small (several tens of kilograms per lot) that the cost of reprocessing them individually would exceed the value of the recoverable uranium. We shall try to obtain levels suitable for re-utilization.

We have not done this yet because our reprocessing plant has been too much in demand.

Since these lots contain United States uranium, we shall certainly ask the USAEC for permission to carry this out.

J. DOWNING: The United States and France in fact agreed some years ago that French-held material of US origin can be mixed for certain technical and economic reasons.
PLANT MEASUREMENT, SAMPLING
AND ANALYSIS FOR ACCOUNTANCY PURPOSES
WITH PARTICULAR REFERENCE TO
SEPARATION PLANTS AT WINDSCALE

A. S. DAVIDSON, F. ELLIOTT, R. POWELL AND K. A. SWINBURN
UNITED KINGDOM ATOMIC ENERGY AUTHORITY,
WINDSCALE AND CALDER WORKS, CUMBERLAND,
UNITED KINGDOM

Abstract — Résumé — Аннотация — Resumen

PLANT MEASUREMENT, SAMPLING AND ANALYSIS FOR ACCOUNTANCY PURPOSES WITH PARTICULAR REFERENCE TO SEPARATION PLANTS AT WINDSCALE. All figures for special materials accountancy purposes contain one or more of three parameters, namely plant measurement, sampling and analysis.

For plant measurements it is best to use weighings wherever possible, e.g. the plutonium in the feed to the New Separation Plant is based on the weight of uranium rods fed. Methods used for measuring volumes are discussed, e.g. (a) pneumercator, and (b) tracer method using radioactive caesium. Judged accuracies and precisions of each method are given.

The sampling of solutions in a batch process is done by using evacuated tubes after thorough homogenization of the solution. For "flowing" streams, where high accuracy is required, a continuous sampler has been devised and is used on the feed to the New Separation Plant. Drilling of metal ingots is the normal sampling method at Windscale, whereas for other solids the method is to homogenize as far as possible (e.g. a Y cone blender is used for plutonium oxide) and then sample.

For the chemical analysis, the precision required of the method depends on the number of determinations in each accounting period. Thus a large number of analyses are required for an accurate but imprecise method. It may be more economical to use fewer determinations with a more precise method. Methods for determining plutonium are discussed in detail, e.g. (a) radiochemistry, (b) colorimetrically using thoronol, (c) plutonium separation, complexing with EDTA and back titration of the excess EDTA, (d) titrimetry (e) isotope dilution followed by mass spectrometry and (f) differential spectrometry. Methods for determining uranium are outlined, e.g. (a) gravimetry as U₂O₅, (b) ether extraction followed by gravimetry or colorimetry, (c) titrimetry, (d) isotope dilution followed by mass spectrometry and (e) enrichment determination by mass spectrometry or line shift emission spectrography. With all accounting methods standards are used to give quality control and to show any inaccuracy which requires to be corrected for. Precisions of all the methods are given and the methods actually used for the New Separation Plant are indicated.

MESURES, ECHANTILLONNAGES ET ANALYSES EN USINE A DES FINS COMPTABLES, NOTAMMENT DANS LES INSTALLATIONS DE SEPARATION DE WINDSCALE. Tous les chiffres intéressant la comptabilité des matières spéciales comportent un ou plusieurs des paramètres suivants: mesures, échantillonnages et analyses.

Pour ce qui est de la mesure en usine, il est préférable de procéder à des pesages toutes les fois que cela est possible; ainsi, le calcul du plutonium entrant dans la nouvelle usine de séparation est fondé sur le poids des barres d'uranium entrantes. Les auteurs examinent les diverses méthodes de mesure des volumes, à savoir: a) «pneumercator», et b) méthode des radioindicateurs utilisant le radiocésium. Le mémoire indique l'exactitude et la précision reconnues à chaque méthode.

On procède à l'échantillonnage des solutions par lots, en utilisant des pipettes à vide, après complète homogénéisation. Pour les opérations en continu, qui exigent une grande exactitude, on a conçu une échantillonneuse à fonctionnement continu qui est utilisée sur le circuit d'entrée de la nouvelle usine de séparation. La méthode habituelle d'échantillonnage pratiquée à Windscale consiste à forer des lingots métalliques; pour d'autres solides, on homogénéise dans toute la mesure du possible (pour l'oxyde de plutonium, par exemple, on utilise un mélangeur conique en Y), après quoi on procède à l'échantillonnage.

Quant à l'analyse chimique, la précision requise pour une méthode donnée dépend du nombre de dosages effectués au cours de chaque période comptable. Ainsi, une méthode exacte mais peu précise exige un grand
El número de análisis puede ser más económico si se reducen el número de dosajes y se aumenta su precisión. Los autores examinan en detalle los métodos de dosaje de plutonio, entre los que se encuentran: a) chimie des corps radioactifs, b) colorimétrie utilisant du thoronol, c) séparation du plutonium complexé avec EDTA et rettage de l'EDTA excédentaire, d) titrimétrie, e) dilution isotopique suivie de spectrométrie de masse, et f) spectrométrie différentielle. Se respetan igualmente los métodos de dosaje de l'uranium, notamment les suivantes: a) gravimétrie de U₃O₈, b) extraction à l'éther suivie de gravimétrie ou de colorimétrie, c) titrimétrie, d) dilution isotopique suivie de spectrométrie de masse, et e) détermination de l'enrichissement par spectrométrie de masse ou spectrophotographie des émissions accompagnées d'un déplacement des raies. Para todas las méhodos contables, se utiliza una variedad para asegurar la calidad y hacer resaltar toda inexactitud que es necesario corregir. Los autores indican la precisión de cada método y señalan las medidas efectivamente utilizadas en la nueva instalación de separación.
La precisión requerida del método de análisis químico depende del número de determinaciones que se efectúen en cada período contable. Así, para un método minucioso pero impreciso es necesario proceder a un gran número de análisis. Puede resultar más económico efectuar menos determinaciones con un método más preciso. En la memoria se exponen detalladamente los métodos para la determinación del plutonio: a) radioquímicos, b) colorimétricos, con toronol, c) separación del plutonio, formación del complejo con exceso de EDTA y retrovaloración del reactivo, d) volumetría, e) dilución isotópica seguida de espectrometría de masas, y f) espectrometría diferencial. También se reseñan los métodos de determinación del uranio: a) gravimetría en forma de $^{235}$UO$_2$, b) extracción con éter seguida de gravimetría o calorimetría, c) volumetría, d) dilución isotópica seguida de espectrometría de masas y e) determinación del enriquecimiento por espectrometría de masas o por espectrografía del desplazamiento de las líneas de un espectro de emisión. En todos los procedimientos contables se emplean patrones para controlar los análisis y descubrir las inexactitudes que deben corregirse. En la memoria se dan precisiones sobre todos los métodos y se indica cuáles de ellos se emplean en la nueva planta de separación.

1. INTRODUCTION

The Windscale Nuclear Fuel processing complex is now centred on the New Separation Plant [1, 2] which was commissioned in 1964. This plant consists essentially of four units: (a) the separation plant, whose main function is to provide separation of plutonium and uranium from each other and from fission products, (b) a thermal denitration plant, to convert the uranium nitrate concentrate to uranium trioxide before reprocessing in the Springfields fuel element plant, and to recover nitric acid, (c) a plutonium finishing plant from which pure plutonium is normally withdrawn as either oxide or metal, and (d) a plutonium recovery facility in which plutonium from plutonium rich wastes and experimental programme returns can be recovered.

The accountancy system used at Windscale may be summarized as follows. The feed of plutonium to the separation plant dissolver is obtained by multiplying the measured plutonium/uranium ratio in the dissolver solution by the weight of fuel fed. Corrections are applied to the nominal feed for burn-up losses and for the weight of impurities in the fuel. These corrections are normally insignificant for fuel of 3000 MWd/t irradiation, and even at 20 000 MWd/t the burn-up correction is only about 2.8%. The plutonium in the product from the separation plant is calculated from the plutonium concentration of a sample and the measured volume of the product. To this is added the plutonium which has been lost in raffinates, to obtain the total output of the plant. The separation plant output is the finishing plant input. The total output of the finishing plant is obtained by weighing the product and measuring the plutonium in the waste and raffinates. An overall process balance may be struck by comparing the plutonium input to the separation plant with the output represented as product from the finishing plant plus raffinates and wastes from both separation and finishing plants. A uranium balance is struck by comparing the weight of uranium fed to the primary separation plant with that observed in the uranium trioxide product, corrections being applied for process losses. Thus it will be seen that each figure obtained comprises one or more parameters of plant measurement, sampling, and analysis.

In the near future there will be added a head-end treatment plant capable of handling a wide range of fuels of varying enrichment. It is anticipated
that fuels fed through this plant will be dealt with batchwise. However, the accountancy system will remain the same in principle with the added refinements such as isotopic measurements which may be called for to control the plant operation. This paper reviews the system of plant measurements, sampling and analysis which currently form the basis for nuclear materials management within the complex.

2. PLANT MEASUREMENTS

The accountancy system outlined above demands that accurate and precise measurements of plant parameters are required only on the following points:

(a) the weight of fuel fed to the dissolver
(b) the volume of plutonium-rich concentrates which are transferred from either the separation or recovery facilities to the finishing plant
(c) the weight of plutonium metal or oxide
(d) the weight of uranium trioxide.

Normally liquors from these sources represent more than 99% of the accountable material so that other plant streams may be metered less stringently. For example should a particular raffinate be known to contain only 0.05% of accountable material, then a relatively inaccurate or imprecise assessment of volume will not vitiate the account balance.

2.1. Weighing

2.1.1. Irradiated fuel

In the former butex process [3] fuel was weighed in small batches before being fed to the dissolver. Many years of experience with this technique have shown that the weight of the rods fed agrees with the measured weight of the rods before irradiation to better than 0.2% [4]. This pre-irradiation weight, corrected for impurities and burn-up is currently used to define the weight of fuel fed to the new plant.

2.1.2. Product weighing

(a) Plutonium ingots are first of all cleaned mechanically to remove surface slag and then weighed before being sealed in their transit container.
(b) Plutonium dioxide is weighed after the homogenization and sampling process described in section 3.4.2.
(c) Uranium trioxide is packed into tared drums and weighed immediately.

2.2. Volume measurements

For raffinates any of a number of conventional volume measurement methods may be used since the accuracy required on these liquors for accountancy purposes is not very high. In practice raffinates are commonly
discharged into a series of volumetrically calibrated batch tanks interconnected by overflow weirs. These tanks are discharged in turn when full. Where the product plutonium (or uranium) level in the raffinate is negligible, batch tanks may not be interposed in the discharge cycle, and in such cases the net daily outflow is either metered with a conventional integrating flowmeter or calculated from flowsheet data.

Only two methods are normally used for high precision measurements of process plant liquor tanks at Windscale, differential pressure level gauges (pneumercators) and tracer methods.

2.2.1. Pneumercator

This form of liquid level indicator is well known outside the nuclear fuel processing field but has special advantages when dealing with radioactive materials since it has no moving parts in contact with the liquor being measured. These gauges, which are used to measure the volume of batch tanks, must be calibrated by the addition of known quantities of water to the tank, but once the calibration has been established the precision of measurement is largely a function of tank design. This is apparent from the facts that the gauge measures liquid levels and that the volume of liquid for any given height is a function of the shape of the tank.

Volumes measured by this type of gauge are dependent for their accuracy on the calibration, a typical procedure for which is given in Ref. [41]. Precisions of a single determination of vessel volume vary from ±0.2% for a vessel of cylindrical design to ±0.2% for vessels with the measuring legs located in a restricted neck at the top of the vessel [5].

2.2.2. Tracer method

This method is of limited applicability and can only be employed where subsequent processing of the liquor whose volume is being measured will eliminate the tracer. Its advantage is that no plant instrumentation is required and it can be applied to a tank of any geometry which can be homogenized and sampled. The method is simple in principle: a known quantity of tracer material contained in a known volume of solution is added to the tank containing liquor whose volume is to be determined. After the whole system has been homogenized a sample of liquor is taken and the tracer concentration redetermined. The volume of liquor in the tank can then be calculated. The volume of tracer solution added usually needs to be small compared with the volume being determined, but the analytical method for the determination of the tracer concentration need not be absolute so long as no bias exists between the measurements made. In practice $^{137}\text{Cs}$ [6] has been successfully used as a tracer for the determination of volumes of plutonium recovery liquor, the concentration of $^{137}\text{Cs}$ in the final solution being adjusted so as to permit its determination in the presence of plutonium or uranium without chemical separation.

The method using $^{137}\text{Cs}$ has a precision of 2-3% due mainly to the statistics of counting; however, when aggregated over a lengthy campaign an unbiased estimate of liquor volumes is obtained.
3. SAMPLING

Because of the radioactive environs from which plant process samples have to be taken, sampling facilities must be carefully designed into processing plants so that meaningful results can be derived from the subsequent analyses. Thus sample points must allow access to process points which are, or can be made to be, representative of the condition which is being studied. Large tanks of highly active liquor must be capable of being homogenized so that useful information can be gained from samples drawn from a single point.

Two fixed-point sampling devices are commonly employed at Windscale; these are the pulse sampler, described below, and the vacuum lift sampler. The latter consists in essence of a sample feed line connected to an intermediate holding vessel which can be filled with plant solution by reducing the pressure within the vessel. A further line from this vessel is connected to a hypodermic needle, so that when a rubber-capped evacuated sample phial is pierced by this needle, sample flows into it from the holding vessel. An essential requirement of the design and operation of this system is that facilities must be provided to ensure that stagnant liquor does not accumulate in the sample withdrawal pipe, and that the system can be repeatedly flushed through with the solution to be sampled.

The actual sampling techniques employed at mainstream accounting points are given below.

3.1. The highly active feed solution

The solution from the dissolver is metered to the first extractor of the primary separation stage by a constant flow device. The pulsed sampler \[?] is placed after this device and is designed to deliver a small pulse of solution at intervals, which can be varied between one and three minutes. Provided the period of feed variations is large compared with this sampling period, then, by collecting these pulses over a 24-h period a sample may be withdrawn which is representative of the irradiated fuel fed to the dissolver in that time.

Basically the sampler consists of a small hollow cylinder submerged in the liquid to be sampled. Liquid is allowed to flow into the cylinder through a small hole in its base and is then expelled by an air pulse of predetermined amplitude and frequency, so that a pulse of solution of fixed volume is discharged to the collecting vessel. At the same time surplus liquid is ejected from the cylinder and the cycle can then restart. To obtain a completely representative sample, experience has indicated that great attention to design and manufacturing detail is essential. The sizing of the inlet/outlet ports on the cylinder, the dimensions of the cylinder and of the sample discharge pipe are critical and were determined on the basis of an extensive experimental programme, supported by detailed computational study of the various design parameters. Considerable attention is paid to the finishing and profiling of the discharge line to avoid liquor crystallization. As the unit was destined to operate under highly active conditions it is designed in the event of failure to be easily removed and replaced.
3.2. **Plutonium nitrate concentrate**

Tanks containing plutonium product solutions are first homogenized by thorough mixing using an installed recirculatory pumping system. In this way eversafe tanks of up to 500 litres capacity can be homogenized in four hours [8]. Liquor samples are then withdrawn using a conventional vacuum lift technique, the sample finally being transferred into an evacuated rubber-capped glass phial.

3.3. **Uranium trioxide**

Samples are taken from top, middle and bottom of drums using a sampling thief. These samples are ground and homogenized on a rotary mixer before samples are taken for analysis. Since the material is hygroscopic, these operations must be carried out within 30 minutes of the drum being opened. Analytical results are referred to the weight of product dried at 110°C.

3.4. **Plutonium metal and oxide**

3.4.1. Metal

The ingot is mechanically cleaned to remove surface slag inclusions and then sampled by drilling in a nitrogen-filled glove box. The actual drilling operation is carried out in an argon-purged inner container. Approximately 10 g of drillings are taken from the base of the ingot using a clean, sharp, 0.25-in. drill. Drillings are collected in a Polythene-lined aluminium can which is then sealed before removal from the glove box.

3.4.2. Oxide

Plutonium dioxide is prepared in batches by the ignition of plutonium oxalate at temperatures of 250°C or greater. Whole batches are homogenized by blending in a Y cone blender contained in a glove box which is purged with dry nitrogen. After a 10-min blending period, random grab portions totalling 10-30 g, as required, are taken and weighed immediately into tared containers. The bulk of the material is then weighed at the same time. The tared containers are re-weighed immediately before the sample is withdrawn in the laboratory for analysis so that allowance can be made for moisture picked up after the oxide was removed from the production line.

3.5. **Raffinates**

Raffinates which are sent to holding tanks before treatment or discharge are sampled using a vacuum lift sampler. Flowing streams are sampled by pulse samplers of the same basic design as those used on the dissolver solution.

Facilities for these operations are an integral part of the separation plant equipment, slight variations in design being made where necessary.
to deal with the differing activity levels and nature of the radioactive liquors being handled. All samplers are designed to minimize liquor hold-up and to permit thorough flushing of sample lines before a sample is taken.

4. THE ANALYSIS OF PROCESS SAMPLES

The percentage of the total product throughput which is represented by the sample to be analysed decides the quality of the analytical method needed for a particular determination. Thus, in the case of the Windscale process the following high-precision determinations are required: (a) The ratio of the plutonium concentration to the uranium concentration in the feed solution. (b) The plutonium concentration in the main plutonium product liquor from the primary separation plant (this is also the feed to the plutonium finishing plant), and of liquors derived from the plutonium recovery facility. (c) The assay of the plutonium metal or oxide product (if required). (d) The uranium content of the uranium trioxide product from the thermal denitration plant.

For all other determinations of plutonium or uranium in other plant streams, e.g. raffinates and solid wastes, less precise methods suffice.

TABLE I

COMPOSITION OF TYPICAL FEED SOLUTION
(3000 MWd/t, 125-d cooling)

<table>
<thead>
<tr>
<th>Isotopic composition</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{240}$Pu 16%, $^{241}$Pu 4%</td>
</tr>
<tr>
<td>$^{235}$U 0.44%, $^{236}$U 0.05%</td>
</tr>
<tr>
<td>$^{241}$Am</td>
</tr>
<tr>
<td>$^{242}$Cm</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Concentration</th>
<th>Isotopic composition</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{240}$Pu 620 mg/litre</td>
<td></td>
</tr>
<tr>
<td>$^{235}$U 500 g/litre</td>
<td></td>
</tr>
<tr>
<td>$^{241}$Am 5 x $10^{-3}$ Ci/litre</td>
<td></td>
</tr>
<tr>
<td>$^{242}$Cm 0.15 Ci/litre</td>
<td></td>
</tr>
<tr>
<td>$^{242}$Cm 110 Ci/litre</td>
<td></td>
</tr>
<tr>
<td>$^{242}$Cm 3 M</td>
<td></td>
</tr>
</tbody>
</table>

4.1. The analysis of samples of feed solution

From the analytical view point the methods employed are largely governed by the sample matrix on which the determination is to be made. The product and concentrates are rarely difficult from this point of view, but the feed solution represents a real problem to the analyst. The composition of a typical feed solution derived from the processing of natural uranium fuel irradiated to 3000 MWd/t in a Magnox reactor and cooled for 125 days is given in Table I.
The problems to be faced in the analysis of this sample arise essentially from the need to determine the plutonium concentration at low levels in a sample of very high \( \beta-\gamma \) activity. (The determination of uranium which is necessary to complete the Pu/U ratio determination poses few problems.) Not all of the methods currently used at Windscale were available when the first processing plant was commissioned, in fact no proven chemical methods existed for the determination of plutonium in the dissolver solutions nor was a prime plutonium standard available, thus a non-chemical method was necessary.

4.1.1. Radiometric determination of plutonium

This has two immediate advantages: (a) methods of alpha counting are well established. (b) the method is applicable to the whole range of plant samples.

Co-precipitation techniques, e.g. lanthanum trifluoride, were not successful in this application because of a variable bias, and in consequence the emphasis has been on the alpha counting of sources prepared by direct evaporations of sample aliquots [9]. Corrections were then applied to these counts for uranium, americium-241 and curium-242 in the feed solution. Interference from fission products was eliminated electronically by appropriate discriminator settings in the alpha counters.

Early process samples were derived from fuel of short irradiation so that the magnitude of the corrections applied to the alpha counts was normally small. In this way a satisfactory 'alpha' balance was obtained through the process. When mass spectrometry and alpha spectrometry equipment became available it was possible from isotopic measurements to calculate the specific activity of plutonium being processed and thus convert the alpha balance into a true plutonium mass balance. As pure plutonium metal became available from the process its specific activity was determined by absolute alpha counting using a low geometry, and very satisfactory agreement (better than 1%) was observed when compared to the value calculated from isotopic measurements.

However, although in principle rapid and economical in use the method proved to be relatively imprecise, ±4% (3\sigma) being observed in practice for a single determination. Furthermore, as fuel irradiations increased, corrections to alpha counts began adversely to affect the accuracy of the determination and thus to necessitate, for example, the precise determination of americium-241 and curium-242 in the feed sample. This determination was normally carried out by extracting the plutonium from the feed sample with trioctylphosphine oxide and attributing the residual alpha count to \(^{241}\text{Am}\) and \(^{242}\text{Cm}\) [10]. Later the processing programme of the U.K.A.E.A. established fuel irradiations of 3000 MWD/t as the norm, and in this fuel alpha emission from plutonium isotopes constitute only about 30% of the total alpha emissions in the feed samples. A change of technique was obviously needed.

4.1.2. Chemical methods

4.1.2.1. Plutonium. The work of Drummond and Welch [11], which made possible the assay of a pure plutonium solution, was an essential feature
of the development of accurate chemical methods. A colorimetric method was developed which was based on the extraction of plutonium (IV) into Hyamine/benzene followed by a colorimetric finish using thoronol [12].

Further work by Boase, Foreman and Drummond [13], following that of Milner and Woodhead [14] on the plutonium (IV) EDTA system resulted in the development of what is now the preferred chemical method for the determination of plutonium in the feed solution. In this approach the Hyamine extraction stage is retained but the plutonium (IV) is then complexed with EDTA and the excess reagent is back titrated with zinc using dithizone as an indicator, and a photometric endpoint.

4.1.2.2. Uranium. As indicated previously the determination of uranium in the feed sample has never been a real problem. The only difficulties which have arisen have resulted from the attempts made to apply standard chemical techniques to highly active working conditions. Obviously gravimetric methods are inappropriate under these conditions but any one of a number of titrimetric methods may be used. At Windscale the preferred method [15] is based on the reduction of uranium (VI) to uranium (IV) with aluminium in hydrochloric acid followed by titration of uranium (IV) to uranium (VI) with standard ceric sulphate.

4.1.3. The determination of the plutonium/uranium ratio by isotope dilution and mass spectrometry

This method which was developed at AERE by Webster [16] is now the only technique routinely [17] used at Windscale for the determination of the plutonium/uranium ratio. It is inherently precise and its accuracy is defined by the accuracy of the prime plutonium and uranium standards against which it is calibrated and the accuracy of the addition of tracer. The principle of the method is as follows: $^{242}\text{Pu}$ and $^{233}\text{U}$ in a known ratio, close to that anticipated in the sample, are mixed with the sample, and, following a chemical equilibration stage, the uranium and plutonium fractions are separated by anion exchange prior to a mass spectrometric determination of the $^{242}\text{Pu}/^{239}\text{Pu}$ and $^{233}\text{U}/^{238}\text{U}$ ratio. Simultaneously the isotopic composition of the Pu and U in the feed are determined and by combining these with the $^{242}\text{Pu}/^{239}\text{Pu}$ and $^{233}\text{U}/^{238}\text{U}$ ratios the Pu/U ratio in the feed is calculated.

4.2. The analysis of plutonium rich samples

These samples of plutonium product and recovery liquors normally contain between 2 mg and 300 mg/ml of plutonium in approximately 3 M nitric acid. Other cations, e.g. uranium, iron, nickel, chromium, do not together exceed 1% of the plutonium content and the total fission product content is less than 1 $\mu$C/ml. These essentially pure solutions of plutonium may be analysed by a variety of methods, but currently titrimetry or differential spectrophotometry are favoured for routine analyses.
4.2.1. Titrimetric methods

Several titrimetric methods are available which are suitable in this concentration range. All of these methods are based on either the plutonium (III)/plutonium (IV) or plutonium (IV)/plutonium (VI) systems. In the former case a variety of reducing agents, e.g. zinc amalgam [18], chromous sulphate [19-20], titanous chloride [21], are used to effect total reduction of the plutonium to plutonium (III) which is then oxidized by standard ceric sulphate or potassium dichromate to plutonium (IV). In a typical example [22] of the second system plutonium ions are oxidized to plutonium (VI) by perchloric acid, following which the plutonium (VI) is reduced to plutonium (IV) with an excess of ferrous sulphate which is then back titrated with cerium (IV). The technique described by Corpel and Regnaud [23] is currently favoured at Windscale because of its ability to function satisfactorily in nitric acid solutions. Corrections are applied for the presence of uranium, iron and molybdenum where appropriate.

4.2.2. Differential spectrophotometry

The use of differential spectrophotometry for the estimation of plutonium was first described by Phillips [24] and has been in use for a number of years at Windscale [25]. Recent work by Bell and Boase [26] has shown that for optimum results nitrate ion should be eliminated or, if this is not possible, both standard and sample should contain the same molarity of nitrate ion. Other variations from the Phillips procedure suggested include a tighter control of chloride ion concentration and the addition of a greater amount of aluminium ion to control fluoride.

4.3. Plutonium product assay

4.3.1. Plutonium metal

Metal ingots are normally weighed and analysed for impurities, the true metal weight being obtained by difference; however, it is sometimes necessary for contractual purposes to assay samples from ingots. For this purpose the metal is cleaned, either electrolytically or mechanically, dried, weighed and dissolved in hydrochloric acid. From this point a number of methods have been employed, the exact technique being normally discussed during contract negotiations. Amongst the methods used have been:

(a) direct titration of the plutonium (III) produced on dissolution of the metal in hydrochloric acid, with ceric sulphate [27]
(b) the titanous chloride reduction, ceric sulphate oxidation method [23]
(c) ignition to stoichiometric plutonium dioxide with corrections applied for impurities
(d) differential spectrophotometry [28].

4.3.2. Plutonium dioxide

Any one of the methods used for metal assay can be used for this material once the material has been quantitatively dissolved [29]. Oxide
prepared by low temperature ignition (about 300°C) is not normally troublesome in this respect being completely soluble in either nitric or sulphuric acid. However as the temperature of ignition increases the oxide becomes progressively more insoluble until prolonged refluxing with nitric/hydrofluoric acid mixtures is needed [30].

4.4. Uranium product assay

Uranium trioxide from the thermal denitration plant is essentially non-active so that a precise gravimetric assay can be carried out. The purity of the product with respect to other metallic impurities is normally so satisfactory that a result of sufficient accuracy is obtained by direct ignition of the UO₃ to U₃O₈ at 850°C [31]. If, however, a chemical separation is required for uranium, either a direct ether extraction [32] or cellulose column [33] separation is employed.

4.5. Raffinate and process waste sample analyses

4.5.1. Plutonium determination

All raffinate samples are analysed by radiochemical methods [34]. Where possible sample aliquots are evaporated directly on to the counting tray and the alpha count obtained converted to plutonium by reference to the specific activity of the plutonium being processed. Modified techniques are needed to deal with, for example, the raffinate from the first mixer settler, a sample which contains little plutonium but essentially all of the ²⁴¹Am, ²⁴²Cm and fission product activity from the fuel. For this sample, plutonium is extracted with Hyamine in benzene in a shielded facility, following which the extract is removed from the facility and used to prepare trays for alpha counting [35].

A modified lanthanum trifluoride co-precipitation method is used to measure plutonium in the uranium product stream [36]. Plutonium in solid waste from the finishing line is estimated to an accuracy of ±25% by measuring the intensity of the 380-keV γ-photon emissions from ²³⁹Pu in a specially designed apparatus [37]. The prime function of this apparatus is to segregate the waste before recovery of plutonium from it.

4.5.2. Uranium determinations

Conventional colorimetric [39] or fluorimetric methods are applied throughout for raffinate sample analysis. Once again modifications to handling technique are necessary to deal with samples which are highly radioactive.

4.6. Measurements of isotopic abundances

Measurements of isotopic abundances of plutonium and uranium are often called for as part of the analytical inspection needed to fulfil contractual obligations. Although the full range of techniques is available in the Windscale laboratories for the determination of both stable and radioisotopes,
naturally demand centres on the determination of $^{235}\text{U}$, $^{236}\text{U}$, and $^{238}\text{U}$, and $^{239}\text{Pu}$, $^{240}\text{Pu}$, and $^{241}\text{Pu}$. Solid source mass spectrometry using an A.E.I. M.S.5 instrument is normally used for all this work, but we have in addition a uranium hexafluoride mass spectrometer for $^{235}\text{U}$ measurements. Emission spectrographic methods are also available and commonly used for samples containing 5 to 90% of $^{235}\text{U}$ [40].

4.7. The standardization, control, and precision of analytical methods

4.7.1. Prime standards

All analytical methods used for accountancy purposes are standardized by reference to prime uranium and plutonium standards obtained from within U.K.A.E.A. Plutonium standard is prepared from plutonium nitrate solution, and involves: (a) precipitation of plutonium oxalate in oxalate deficient conditions (b) successive recrystallizations of $\text{Pu(NO}_3\text{)}_4$; approximately 80% of the $\text{Pu(NO}_3\text{)}_4$ being recrystallized at each stage, (c) the pure $\text{Pu(NO}_3\text{)}_4$ dissolved to a calibrated volume and then this solution is standardized by igniting a known aliquot to $\text{PuO}_2$ at 1200°C. All dilutions, aliquots etc. are made by weighing.

For uranium standards uranyl nitrate solution is purified by successive ether extractions, following which it is standardized by ignition to $\text{U}_3\text{O}_8$ at 800°C.

4.7.2. Control

Control of the accuracy and precision of all analytical methods is essential for successful accountancy, particularly during long campaigns. This control is maintained by the regular analysis of simulated plant samples prepared from primary standard material. Statistically based quality control charts are drawn up and used throughout a campaign to provide accuracy and precision information on the various analytical methods used. Routine analytical results are always corrected for any method bias which these controls reveal.

4.7.3. Typical accuracy and precision data

The analysis of the same type of sample by the same method in a situation which demands the best possible performance of both method and operator offers a unique opportunity to collect valuable data over a very long period.

Usually the precision obtained is somewhat worse than that claimed by the method originators, which is not surprising when a large number of different operators of a varying level of competence are employed. Typical precision data obtained in routine accountancy analyses in the Windscale laboratories are given in Table II.
TABLE II
PERFORMANCE DATA OF ROUTINE ANALYTICAL METHODS

<table>
<thead>
<tr>
<th>Determination</th>
<th>Analytical method</th>
<th>Precision (3σ) of single determination (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>b. Isotope dilution [17]</td>
<td>±2</td>
</tr>
<tr>
<td></td>
<td>b. Chromous sulphate-ceric sulphate [20]</td>
<td>±1.2</td>
</tr>
<tr>
<td>3. Pu metal assay</td>
<td>a. Differential spectrophotometry [28]</td>
<td>±0.2</td>
</tr>
<tr>
<td></td>
<td>b. Titanous chloride-ceric sulphate [23]</td>
<td>±0.2</td>
</tr>
<tr>
<td>4. Pu oxide assay</td>
<td>a. Differential spectrophotometry [29]</td>
<td>±0.4</td>
</tr>
<tr>
<td></td>
<td>b. Titanous chloride-ceric sulphate [23]</td>
<td>±0.4</td>
</tr>
</tbody>
</table>

5. THE ACCURACY AND PRECISION OF THE OVERALL ACCOUNT

The bias (accuracy) of the accounts is the summation of the systematic errors in plant measurement, sampling and analysis. As shown previously the analytical information obtained from standards provides a method of correction which is automatically applied. It is therefore necessary, in order to prevent an account from biasing, to ensure that there are no systematic errors in plant measurements by frequent recalibrations of instruments and by constant attention to sampling techniques.

The precision (repeatability) of accountancy is expressed in terms of the coefficient of variation of the process, and is a measure of the degree of confidence with which an accountancy figure may be quoted. A full statistical summation of the variances of all accountancy processes is given by Tingey [41].

Given that the degree of confidence in the quantity of fissile material can be previously defined the amount of effort necessary in all stages can be calculated in relation to the campaign length. Safety may dictate that from plant design parameters losses of say 2 kg or greater should be positively indicated, whereas this may be an uneconomic quantity in relation to the weight of fissile material being processed.

In general provided the economic factor is met then the precision may be improved almost to any reasonable limits by replicating the necessary stages.
of the process. For example, in the feed point the weight of uranium fed is known very precisely, sampling is representative, so that errors arising from analysis are the most important. It is known that the precision of a single Pu/U ratio measurement is ±2%. Thus, if economy throughout a campaign dictates that throughput shall be known to ±0.2% then approximately 100 determinations of Pu/U ratio should be made during the period of the campaign.

REFERENCES

SAMPLING URANIUM HEXAFLUORIDE*

W.D. McCLUEN
UNION CARBIDE CORPORATION, OAK RIDGE, TENN., UNITED STATES OF AMERICA

Abstract — Résumé — Аннотация — Resumen

SAMPLING URANIUM HEXAFLUORIDE. Uranium hexafluoride is the commonly accepted chemical form in which transfers from and returns to production channels are effected, including financial settlement. Adequate measurement of the material becomes an inherent necessity. This implies that there is accurate weight or volume determination, representative sampling with respect to chemical and isotopic content, and definitive analytical effort. Uranium hexafluoride is frequently considered only in the gas phase. However, measurement for transfer of responsibility is almost always made in the liquid phase with actual shipment being in the solid phase. The discussion herein is confined to the sampling operation in the liquid phase of material enriched in the uranium-235 isotope packaged in 5-in. I.D., double-valve shipping cylinders. This cylinder is chosen because of its universal applicability and ease of shipment.

Receipts into the sampling station will be as cylinders of solid uranium hexafluoride. Representative sampling involves attaining a state of homogeneity for chemical and isotopic content. This is achieved by liquefying the contents through heating, shaking the container to ensure mixing, and withdrawing a liquid sample through a calibrated pipette into a sample container of the proper size. Experimental data are available to show the adequacy of techniques described. The problem of attaining chemical homogeneity before sampling is more difficult than that for isotopic homogeneity.

A discussion of auxiliary equipment associated with the sampling operation is included since this is an important part of a successful sampling programme. In addition, the variability of limits of uncertainty upon the cylinder contents is briefly indicated for multiple samples and multiple analyses on single samples.

PRELEVEMENT D'ECHANTILLONS D'HEXAFLUORURE D'URANIUM. L'hexafluorure d'uranium est la forme chimique sous laquelle l'uranium est le plus communément prélevé ou introduit dans le circuit de production et fait l'objet de transactions financières. Dans ces conditions, il est indispensable de pouvoir le doser avec la précision nécessaire. Il faut donc déterminer exactement les poids ou les volumes, établir des échantillons qui soient représentatifs du point de vue de la composition chimique et isotopique, et procéder aux analyses avec le plus grand soin. Bien souvent, l'hexafluorure d'uranium intervient en phase gazeuse. Cependant, lorsqu'il les expéditions proprement dites se font en phase solide. Le mémoire se borne à étudier le prélèvement d'échantillons sur la phase liquide d'une matière enrichie en uranium-235, conditionnée dans des bouteilles d'expédition à deux soupapes, d'un diamètre intérieur de 127 mm. Cette bouteille a été choisie parce qu'elle est d'un emploi universel et facile à expédier.

Le service d'échantillonnage reçoit l'uranium dans des bouteilles contenant de l'hexafluorure d'uranium solide. Pour que ces échantillons soient vraiment représentatifs, il faut que la matière soit homogène en ce qui concerne sa composition chimique et isotopique. A cet effet, on liquéfie le contenu en le chauffant, on agite le conteneur pour bien mélanger et on préleve un échantillon liquide, avec une pipette étalonnée, que l'on verse dans un récipient spécial pour échantillons. L'auteur dispose de données expérimentales qui montrent que les méthodes décrites sont efficaces. Il est plus difficile d'obtenir l'homogénéité chimique avant le prélèvement de l'échantillon que d'obtenir l'homogénéité isotopique.

Le mémoire contient également une étude du matériel auxiliaire utilisé dans les opérations de prélèvement; en effet, le succès des opérations d'échantillonnage dépend dans une large mesure de ce matériel. L'auteur indique en outre les variations des limites d'incertitude concernant le contenu des bouteilles dans les cas de prélèvement d'échantillons multiples et d'analyses multiples portant sur des échantillons simples.

ОТБОР ПРОБ ШЕСТИФТОРИСТОГО УРАНА. Шестифтористый уран является обычно принятой химической формой урана, в которой он передается в производство и возвращается...

* This paper is based on work performed at the Oak Ridge Gaseous Diffusion Plant operated by Union Carbide Corporation for the USAEC.
обратно, включая решение финансовых вопросов. Проведение необходимых измерений становится существенной необходимостью. Это означает, что точно определяется вес или объем, производится отбор характерной пробы в отношении химического и изотопного состава и проводятся окончательные анализы. Шестифтористый уран часто учитывается только в газовой фазе. Однако измерения с целью передачи ответственности почти всегда производятся в жидкой фазе, причем на практике его транспортировка осуществляется в твердой фазе. Данный документ ограничивается описанием отбора проб в жидкой фазе обогащенного изотопом урана-235 материала, содержащегося в двух вентильных транспортных цилиндрах с внутренним диаметром 12,5 см. Этот цилиндр выбран ввиду его универсального применения и удобства транспортировки.

На станции отбора проб материал поступает в виде цилиндров с твердым шестифтористым ураном. Отбор характерной пробы связан с обеспечением однородности химического и изотопного состава. Это достигается в результате сжигания содержимого путем нагрева, встряхивания контейнера с целью смешения его содержимого и помещения жидкого образца с помощью калиброванной пипетки в соответствующий контейнер для образцов. Имеются экспериментальные данные, которые говорят о достаточности описанных методов. Проблема достижения химической однородности до отбора проб является более трудной, чем проблема достижения изотопной однородности.

Дается описание вспомогательного оборудования для операции по отбору проб, поскольку это важно для ее успешного выполнения. Кроме того, кратко рассматривается изменчивость пределов неопределенности в отношении содержимого цилиндров для многократных проб и многократных анализов единичных проб.

1. INTRODUCTION

In recent years, the basic production of uranium enriched in the 235 isotope has been in the chemical form uranium hexafluoride. This establishes the starting chemical and measurement form insofar as a production facility is concerned. This also applies to the initial processor.

The big problem that faces the producer or processor of uranium hexafluoride, as with most other materials of production, is the development of accurate, reliable measurements of those components which are subject to variation. Basically, there are but two variable components: first, the total uranium content, i.e. how pure is the material; and second, the isotopic content, or what part is represented as uranium-235. These factors
determine the price. They are also the prime requisites of accountability. But even if accountability was not involved, the information would be needed for judgments of nuclear safety and health physics.

In the broad perspective, adequate measurement involves weighing, sampling, and laboratory analysis. There are well developed standards to judge the adequacy of weights and laboratory analyses, but such is not the case with respect to sampling. However, the acceptability of the measurement cycle depends significantly on the representativeness of the sample.

In the early consideration of developing a paper on this subject, it was decided to confine all remarks to the 5-in. ID (inside diameter) cylinder because of its broad usage. However, since that time, events have occurred which now suggest the advisability of including the 12-in. ID cylinder in the discussion.

2. MINIMIZING THE PROBLEM

The ideal situation is to sample a completely homogeneous quantity of uranium hexafluoride. Attaining isotopic homogeneity is not particularly difficult, but attaining chemical homogeneity is quite another matter. The direction to take, however, is clear; eliminate the impurity. From an operational point of view, the potential impurities are likely to be more volatile and of a lower molecular weight than uranium hexafluoride. What this means is that in the condenser system there is a built-in step of purification. Some impurity, however, such as hydrogen fluoride, is soluble to some degree in liquid uranium hexafluoride and will drain into a cylinder with the product material. Minimization of the hydrogen fluoride and other volatile impurities which become entrained in the liquid may be accomplished by evacuating the void volume over the solidified uranium hexafluoride. A further step can be taken after liquefaction through the same technique. Experience has shown that a series of short evacuations from a hot cylinder is more effective than a single long evacuation. The reason for this is that the vapour over the liquid uranium hexafluoride will become preferentially enriched in the volatile impurity if given sufficient time. It is true that somewhat more uranium hexafluoride will be evacuated through this procedure. This is modest, however, with respect to the total.

The discussion thus far has been confined to cylinders which have been filled by liquid drain. Quite frequently, 5-in. ID cylinders are also filled by direct condensation from a flowing gas stream, particularly when the target withdrawal quantity is relatively small. Those withdrawals which have isotopic enrichments of uranium-235 sufficiently low to permit the use of 12-in. ID cylinders are normally in amounts warranting the use of liquid condenser systems. It may be seen that the design of the 5-in. ID cylinder (Fig. 1) permits filling by both methods. Two valves are provided, one of which has a dip pipe or leg. When condensing from a flowing gas stream, the flow is directed into the cylinder through the dip-pipe valve. Venting is effected through the remaining valve. The base of the cylinder is refrigerated in this technique. Directing the gas flow in through the dip pipe causes a preferential filling near the bottom and results in optimum weight condensation. Heat, of course, is maintained on the valves. If the cylinder is
filled by liquid drain, only one of the valves is used. It may be observed that a single valve is provided on the 12-in. ID cylinder (Fig. 2). Those cylinders which are filled by cold condensation are held at condensing temperatures which tend to minimize the trapping of impurity. Additional precautions taken to eliminate impurity involve venting at ambient temperatures and again when liquefied, as discussed earlier.

Little attention need be given to the isotopic homogeneity. Even where deliberate changes are made in the uranium-235 content, the act of heating the cylinder to liquefy the contents will result in a thorough isotopic mixing through the heat-induced convection currents. However, an added assurance is introduced through agitating the liquid uranium hexafluoride.

3. THE SAMPLING OPERATION

The first step in the sampling operation is liquefying the uranium hexafluoride. Any controlled heating medium is considered satisfactory for this step. A minimum of at least 200°F should be attainable. This suggests saturated steam as a source since there is a built-in upper temperature control limit. If other sources of heat, such as electricity, are used, it is especially important that a maximum temperature of 300°F be not exceeded for the two types of cylinders discussed herein. The reason is that fill limits...
SM-67/23

FIG. 2. Twelve-inch uranium hexafluoride cylinder

have been based on a temperature of 300°F. The heating cycle for 5-in. ID cylinders is approximately two hours and for 12-in. ID cylinders is about four hours in the 200-300°F range to attain complete liquefaction. These are suggested minimum times.

After heating, the cylinder is agitated to aid homogenization. In the case of the 5-in. ID cylinder, which has a gross weight of the order of 50 kg when filled, this is accomplished through the use of a special device shown in Fig. 3. It can be seen that the cylinder fits into a sleeve and is locked in place by the shoulder near the top of the cylinder. The weight is reasonably balanced about a central shaft which permits manual rotation of the cylinder. This can be seen in Fig. 4.

For the 12-in. ID cylinder, having a full gross weight of about 280 kg, no special fixture for rotation is used. Homogenization is approached by rocking the cylinder over a ledge which is slightly off-centre, as shown in Fig. 5. The idea in both cylinder sizes is to attain a rapid axial movement of the liquid uranium hexafluoride. Experience has indicated that about five agitating cycles will produce a state of homogeneity beyond which no change can be seen.

After the homogenizing step, the cylinder is placed in the sampling position, as is indicated by Fig. 6 for the 5-in. ID cylinder and Fig. 7 for the 12-in. ID cylinder. Referring to Fig. 6, for example, it will be noticed that
there is a sample metering device, or pipette, immediately below the cylinder. The pipette is connected to the cylinder by tubing through one of the two cylinder valves. This valve does not have a dip pipe attached. Attached to the bottom of the pipette is a sample container, while extending from the side is the purge and evacuation line.

The sample pipette is a calibrated sight gauge in which the level of liquid uranium hexafluoride may be observed. Its internal free volume may be sized to suit the particular sampling requirements. Ordinarily, a sample is drained from the cylinder into the pipette. The sample is isolated in the pipette and is then drained to the sample tube and isolated. The sample system is purged and evacuated. After the sampling is complete, the cylinder is removed to a storage location.

Multiple cylinder lots of material having a common uranium-235 content are frequently composited, i.e. one sample will represent up to six cylinders. This is done in the interest of analytical economy. Each cylinder in the composite group is represented by a liquid aliquot. All aliquots are drained to a common sample tube which must have sufficient capacity to hold the total sample. It is important that the individual aliquots be standardized to the amount of parent material which each represents. In addition, care should be taken that the sample container is not overfilled to
guard against hydrostatic rupture in the laboratory. No downward adjustment of sample weight should be attempted. If the sample is over the target, it should be salvaged, and a second, properly sized sample taken.

4. VERIFYING THE SAMPLING OPERATION

Verification of the cylinder sampling technique is divided into two parts: that for uranium content, and that for isotopic enrichment. In the case of uranium content, the contents of a cylinder are deliberately diluted with an impurity such that a predetermined uranium concentration will be produced. The standard sampling procedure is then followed. For example, a 5-in. ID cylinder was filled with a quantity of uranium hexafluoride having a well-defined uranium content. A diluent charge was added which would produce a calculated value of 66.718% uranium. Fifteen samples were withdrawn by the standard technique. The mean result of these samples was 66.71%, which is easily within the limit of measurement uncertainty. In a similar test on a larger cylinder, the calculated and measured values were identical at 67.54% uranium.
Verifying the uranium-235 measurement is somewhat similar to that for uranium except that the diluent is uranium hexafluoride of a different isotopic content. In a particular test on a 5-in. ID cylinder, a quantity of material enriched in the uranium-235 isotope was diluted to a calculated target of 3.50%. Fifteen individual samples were then removed by the standard sampling technique. The mean of these samples was found to coincide exactly with the calculated value. Similar agreement was found in a separate test on the larger cylinder. Of added interest is the fact that sampling tests which omit the agitation step will produce representative isotopic results after dilution if the heating times required for total liquefaction are essentially doubled. It is theorized that the convection currents effect homogenization. This is not true, however, for total uranium content.

Control over the composite sample representativeness is effected by periodically comparing the weighted mean sample results of the individual cylinders comprising the composite with the routine composite sample results. This means that each cylinder is sampled twice—once individually, and once as a part of the composite group. In our experience, there has never been an observed instance of a non-representative composite sample for either uranium or uranium-235 content.
FIG. 6. Mixing a 12-in. cylinder of uranium hexafluoride

TABLE I

GUIDE TO RELATIVE REPORTED URANIUM ANALYSES OF UF₆

<table>
<thead>
<tr>
<th>No. of Determinations</th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
<th>5</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.052</td>
<td>0.036</td>
<td>0.030</td>
<td>0.026</td>
<td>0.023</td>
</tr>
<tr>
<td>2</td>
<td>0.035</td>
<td>0.025</td>
<td>0.020</td>
<td>0.018</td>
<td>0.016</td>
</tr>
<tr>
<td>3</td>
<td>0.031</td>
<td>0.022</td>
<td>0.018</td>
<td>0.016</td>
<td>0.014</td>
</tr>
<tr>
<td>4</td>
<td>0.029</td>
<td>0.021</td>
<td>0.017</td>
<td>0.014</td>
<td>0.013</td>
</tr>
<tr>
<td>5</td>
<td>0.027</td>
<td>0.019</td>
<td>0.016</td>
<td>0.013</td>
<td>0.012</td>
</tr>
</tbody>
</table>

5. PRECISION OF REPORTED ANALYSES

Analytical precisions and relative uncertainties of reported analyses with respect to the container being described are dependent upon the labora-
FIG. 7. Sampling position for 12-in. uranium hexafluoride cylinder

tory capability and the representativeness of the field sample. A very minor part of the over-all uncertainty is attributable to field sampling if the proper procedure has been followed. The major source of uncertainty is associated with laboratory analytical effort. Although outside the basic scope of this paper, it is worthwhile to look at the total relative uncertainty in statements of uranium and uranium-235. These are generally applicable to all standard containers utilized for the transport of enriched uranium hexafluoride. In Table I can be seen the relative precision in per cent of the reported analysis for uranium in individual cylinders for multiple samples and multiple determinations per sample. Similarly, Table II shows an array of relative values for uranium-235.

Axiomatically, it may be said that when the contribution of analytical uncertainty is high with respect to total uncertainty, the greatest improvement in measurement precision can be made by increasing the number of analyses per sample rather than by increasing the number of samples. This is specifically applicable to uranium hexafluoride.
<table>
<thead>
<tr>
<th>No. of Determinations</th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
<th>5</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.16</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>0.15</td>
<td>0.11</td>
<td>0.09</td>
<td>0.08</td>
<td>0.07</td>
</tr>
<tr>
<td>3</td>
<td>0.15</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>4</td>
<td>0.15</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>0.15</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>6</td>
<td>0.15</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>7</td>
<td>0.15</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>8</td>
<td>0.15</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>9</td>
<td>0.15</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>10</td>
<td>0.14</td>
<td>0.10</td>
<td>0.08</td>
<td>0.07</td>
<td>0.06</td>
</tr>
</tbody>
</table>

Blank regions show no improved precision.
Abstract — Résumé — Аннотация — Resumen

SAMPLING OPERATIONS INVOLVING SUPPLIERS AND CUSTOMERS. Sampling operations play an important part in all commercial transactions involving raw materials or finished products. Their importance generally varies with the value of the products in question.

The relatively high listed prices involved call for special efforts as regards sampling, to provide more satisfactory information concerning the material being processed. Depending on the circumstances, sampling operations may be designed to supply either quantitative or qualitative information about the materials. These two aspects may be regarded as separate or complementary.

Where quantitative information is concerned, the task is one of determining the amount of valuable material present, with the precision dictated by economic considerations (listed prices, exchange rate fluctuations, etc.) and technical considerations (possibility of processing an ore, effect of its composition on a process, the balance sheet, etc.).

As regards qualitative information, the main aim is conformity with technical specifications. Because of the rapid development of nuclear industry, these specifications are too often based on insufficiently realistic considerations. Reliance is more often placed on standard analysis reports than on specifications which have actually been studied, since the experience necessary to draw up rational specifications is not always available. Accordingly, a great effort must be made to reassess the problem of specifications in transactions involving nuclear materials.

By careful study of the problem, it should be possible to classify the materials subject to specification on the basis of various requirements (considerations of neutron physics, corrosion, process, accessibility after irradiation, etc.). These studies should permit simplification of acceptance analyses and even of fabrication controls, thereby reducing the cost of analysis and no doubt also favourably affecting the price of the materials.

In the assessment of materials the idea of sampling is closely linked with methods of analysis, and the paper emphasizes the interdependence of these two factors and the difficulty of treating them separately. Each sampling study is thus a special case depending on the nature of the material and the problems of analysis involved.

A subsidiary aspect of the controls connected with sampling, and one which is sometimes of considerable importance, is the problem of the containment of the products. The choice of packaging materials is a basic factor in avoiding contamination or deterioration of the product and in ensuring that samples taken at any time will be as representative as possible.

In the light of these considerations, numerous examples are cited as illustrations of the solutions adopted for a number of materials in the French nuclear industry. These examples relate to the sampling of uranium ores, uraniferous concentrates, the calcium used in reduction, uranium metal and various salts of nuclear elements.

OPERATIONS D’ÉCHANTILLONNAGE ENTRE FOURNISSEURS ET CLIENTS. Les opérations d’échantillonnage jouent un rôle important dans toutes transactions commerciales de matières premières ou de produits finis, l’importance de ce rôle étant généralement fonction de la valeur des produits intéressés par les transactions.

Le prix d’ordre relativement élevé impose donc un effort tout particulier dans le domaine de l’échantillonnage, pour une meilleure connaissance de la matière en œuvre. Selon les cas, les opérations d’échantillonnage viseront la connaissance quantitative ou qualitative des matériaux, ces notions pouvant être considérées isolément ou se compléter.

Pour la connaissance quantitative, on s’attache à déterminer la quantité de matière noble. Avec l’exactitude nécessaire selon les considérations économiques (prix d’ordre, fluctuations des cours, etc.) ou techniques (possibilité de traitement d’un minerai, influence de sa composition sur ce procédé, bilan, etc.).
ОТБОР ОБРАЗЦОВ ПРИ ТОРГОВЫХ СДЕЛКАХ МЕЖДУ ПОСТАВЩИКАМИ И ПОКУПАТЕЛЕМИ. Отбор образцов играет важную роль во всех торговых сделках по сырьевым материалам и готовой продукции, причем важность этой роли в общем зависит от значения продукта, предусматриваемых сделками.

Вот почему относительно высокая цена требует особых усилий для отбора образцов с тем, чтобы лучше знать соответствующие материалы. В различных случаях образцы выбираются с целью получения сведений о количестве или качестве материалов, и эти сведения можно рассматривать отдельно или они могут дополнять друг друга.

Что касается сведений о качестве продукта, то стремятся определить количество благородного материала с необходимой точностью в зависимости от экономических (цены, колебания цен) или технических соображений (возможность обработки руды, влияние ее состава на процесс обработки, выход и пр.).

Что касается получения сведений о качестве, то следует в особенности следить за соответствием техническим условиям. С учетом быстрого развития ядерной промышленности эти условия довольно часто увязываются с соображениями, которые не всегда являются весьма реальными. Часто пользуются аналитическим балансом, чем действительно изученными техническими условиями, поскольку не всегда имеется опыт, необходимый для определения разумных условий. Поэтому нужно предпринимать большие усилия для решения проблемы спецификаций в сделках по ядерным материалам.

Глубокое изучение проблемы должно позволить классифицировать элементы, поддающиеся под спецификацию по различным показателям (нейтронная характеристика, коррозия, способ обработки, доступность после облучения и пр.). Это должно облегчить приемные анализы и даже методы контроля за изготовлением, а поэтому снизить себестоимость анализа и, без сомнения, оказать также положительное влияние на цену этих материалов.

Что касается сведений о материалах, то понятие отбора образцов тесно связано с методами анализа, и при этом подчеркивается взаимозависимость двух факторов, которые трудно рассматривать отдельно. Поэтому любое изучение, связанное с отбором образцов, представляет собой особый случай веду характера материала и вытекающий из этого проблем анализа.

Дополнительным аспектом, который иногда нельзя обрабатывать со счета и который имеет отношение к контролю при отборе образцов, является кондегенерация продуктов. Выбор упаковки остается основным фактором, позволяющим избежать загрязнения или потери продукта с тем, чтобы отбор образцов был в любое время по возможности гидравлическим.

С учетом этих замечаний и на различных примерах приводятся решения, применимые к определенному количеству материалов во французской ядерной промышленности. Эти примеры относятся к отбору образцов урановых руд, к урансодержащим концентратам, к восстановленному кальцию, металлическому урану и различным солям ядерных элементов.
OPERACIONES DE MUESTREO ENTRE SUMINISTRADORES Y CLIENTES. Las operaciones de muestreo desempeñan un papel muy importante en todas las transacciones comerciales de materias primas o de productos acabados; por lo general, esa importancia está en función del valor de las materias o de los productos que son objeto de la transacción.

El precio relativamente elevado impone, pues, un esfuerzo particular en lo que respecta al muestreo, a fin de lograr un mejor conocimiento de los materiales de que se trate. Las operaciones de muestreo tienen por objeto, según los casos, el conocimiento cuantitativo o cualitativo de los materiales; estos dos conceptos pueden considerarse por separado o completarse.

Para conocer los materiales cuantitativamente se determina la cantidad de materia noble con la exactitud que exijan las consideraciones económicas (precio, fluctuaciones del mercado, etc.) o técnicas (posibilidad de tratamiento de un mineral, influencia de su composición sobre un proceso, balance, etc.).

La finalidad principal del análisis cualitativo es comprobar si el material cumple las especificaciones técnicas. Debido a la rápida evolución de la industria nuclear, esas especificaciones están con demasiada frecuencia vinculadas a consideraciones que no siempre se ajustan suficientemente a la realidad. Lo más frecuente es utilizar como base un boletín de análisis tipo y no una especificación realmente estudiada, porque no siempre se posee la experiencia práctica necesaria para establecer especificaciones racionales. Conviene, pues, realizar un gran esfuerzo para estudiar de nuevo el problema de las especificaciones en las transferencias de materiales nucleares.

Un estudio detenido del problema debe permitir clasificar los elementos sometidos a especificación con arreglo a diversos imperativos (neutrónica, corrosión, proceso, accesibilidad después de la irradiación, etc.). Es probable que dicho estudio permita además simplificar los análisis de entrada e incluso los controles durante la fabricación, con lo que se reduciría el precio de costo de los análisis y seguramente también el precio de los materiales.

Para el conocimiento de los materiales la noción de muestreo está estrechamente relacionada con los métodos de análisis; en la memoria se insiste en la interdependencia de esos dos factores, que difícilmente pueden tratarse por separado. Cada estudio de muestreo es, pues, un caso especial que depende de la naturaleza de los materiales y de los problemas analíticos que plantean.

Un aspecto secundario de los controles relacionados con el muestreo, pero que a veces no puede desprenderse, es el acondicionamiento de los productos. La elección del embalaje sigue siendo fundamental para evitar que el producto se contamine o evolucione y conseguir que el muestreo sea en todo momento lo más representativo posible.

Partiendo de estas consideraciones los autores exponen las soluciones adoptadas en la industria nuclear francesa para varios materiales; los ejemplos citados se refieren al muestreo de minerales de uranio, concentrados uraníferos, calcio de reducción, uranio metálico y diversas sales de elementos nucleares.

Les opérations d'échantillonnage jouent un rôle important dans toutes les transactions commerciales de matières premières ou de produits finis. Elles sont également primordiales pour les bilans d'usine. Toutefois, l'importance de ces opérations est liée à la valeur du produit considéré et aux quantités en œuvre.

Le prix d'ordre généralement élevé des matériaux nucléaires impose donc, dès qu'il s'agit de quantités quelque peu importantes, un effort tout particulier dans le domaine de l'échantillonnage. Celui-ci doit permettre, selon les besoins, la connaissance quantitative des produits considérés, leur connaissance qualitative, ou les deux simultanément.

La connaissance quantitative nécessite le dosage de la matière noble (par exemple, uranium dans un concentré ou un composé) avec une exactitude déterminée par des considérations économiques (prix d'ordre, fluctuation des cours, etc.), techniques (intérêt ou possibilité de traiter un minerais, influence de la composition d'un produit sur un procédé, bilan, etc.), ou de sécurité (criticité).
La connaissance qualitative vise surtout la conformité à des spécifications techniques et, tout particulièrement, au respect de limites imposées pour certains composants chimiques et fixées par des considérations techniques (influence sur un procédé, corrosion, etc.).

Chaque problème d'échantillonnage reste un cas d'espèce. Il importe en effet de définir chaque fois
- la fréquence et la quantité des prélèvements; celles-ci sont fonction des quantités en œuvre, de la nature du produit (granulométrie, homogénéité, répartition de l'humidité, possibilité d'altération, etc.);
- le traitement du prélèvement (homogénéisation, quantité nécessaire pour une analyse représentative, conditionnement du prélèvement, etc.).

Il faut insister ici sur les liens étroits à maintenir entre l'échantillonnage et l'analyse, car la précision désirée conditionne directement l'étude des méthodes d'analyse. D'autre part, il sera toujours nécessaire de disposer d'une méthode d'analyse très précise pour faire l'étude d'échantillonnage dans les meilleures conditions possibles. Dans quelques cas, on peut espérer qu'une méthode moins précise pourra ensuite suffire pour des transactions courantes de produits issus d'une fabrication stable.

1. ÉCHANTILLONNAGE FOURNISSEURS-CLIENTS

Le client reçoit un produit généralement livré, s'il ne s'agit pas d'un métal, en fût ou tout autre récipient. Si l'importance des tonnages en cause ne justifie pas une station particulière d'échantillonnage à la réception, on a souvent recours à l'échantillonnage à la sonde. Une autre méthode, préférable, est le passage de la totalité du contenu du récipient, après homogénéisation si possible, sur un échantillonneur constitué par un ensemble de diviseurs en cascade. Un tel appareil peut être facilement construit pour des quantités correspondant à la contenance des récipients commerciaux courants et être réalisé en géométrie sûre (fig. 1 à 3).

Toutefois les fournisseurs (usines productrices) sont parfois mieux placés pour étudier un système d'échantillonnage sûr et en continu. Il est alors recommandé d'associer le client à cette étude; celui-ci pourra accepter l'échantillon prélevé dans l'usine productrice, pour autant que le produit ne soit pas susceptible d'évolution ultérieure et que l'échantillonnage soit fait en présence d'un de ses représentants, occasionnel ou permanent.

Il est alors nécessaire d'apporter également un grand soin à l'emballage des produits pour éviter toute pollution lors des manipulations ultérieures, par exemple à l'ouverture des récipients.

Nous examinerons maintenant un certain nombre d'exemples de ce qui a été fait, dans le domaine de l'échantillonnage, dans l'industrie nucléaire française. Un point à souligner est le fait que beaucoup de matériaux sont traités sous contrat dans des firmes privées qui interviennent comme facteurs. De ce fait, les matériaux transitent dans un certain nombre d'usines et les problèmes d'échantillonnage aux divers stades prennent une importance accrue, tant pour des questions de composition que pour des questions de bilan.
2. ECHANTILLONNAGE DES METAUX

2.1. Echantillonnage du calcium

Nous commencerons, à titre d'exemple, par le cas du calcium métallique utilisé parallèlement au magnésium pour la réduction du fluorure d'uranium. Un cahier des charges fixe la pureté du calcium en tenant compte du cahier des charges fixant la pureté de l'uranium.

Le calcium est livré aux raffineries, en copeaux ou en grenaille, par lots de 600 à 800 kg. L'unité de fabrication est un lingot de quelques dizaines de kilogrammes, obtenu par distillation. Ces lingots sont hétérogènes entre eux et même au sein du lingot (fig. 4).
Le contrôle de qualité du calcium pouvait se faire soit sur chaque lingot constituant le lot, soit sur l'ensemble du lot. Pour écarter la première solution, trop lourde, on a été conduit à effectuer une étude très complète.

Pour effectuer le contrôle sur le lot, il fallait d'abord assurer son homogénéisation (la masse unitaire d'un copeau est de l'ordre de 100 mg). Il fallait ensuite déterminer le nombre de prélèvements, donc d'analyses à effectuer, et l'importance de chacun des prélèvements. Les conditions d'homogénéisation, le nombre et l'importance des prélèvements ont été établis par les mathématiciens du CEA; nous décrirons plus en détail l'étude expérimentale qui a été faite pour déterminer les valeurs des paramètres figurant dans les équations.
Une première expérience a été faite avec un lot de calcium dont une partie a été artificiellement polluée par du fer. On a introduit dans l'appareil à homogénéiser 40 kg d'un alliage de calcium contenant 135 ppm de fer et 400 kg de calcium contenant moins de 5 ppm de fer. Plusieurs prélèvements effectués au cours de chacun des passages successifs du lot dans l'appareil ont donné un ensemble de résultats dont l'exploitation a montré qu'il fallait trois passages successifs dans l'homogénéiseur pour assurer une homogénéité correcte. Mais cette expérience n'a pas permis de déterminer le nombre et l'importance des prélèvements à faire pour le contrôle des lots puisque, dans ce cas, les prélèvements avaient été fixés à priori.
Une deuxième expérience a été envisagée, expérience pour laquelle les mathématiciens souhaitaient avoir quatre populations de copeaux différant entre elles dans les rapports 15, 9, 5 et 1.

Les chimistes devaient former ces quatre populations et être capables ensuite de les reconnaître. Un marquage radioactif n'était pas possible. Un marquage par introduction d'impuretés aurait exigé un gros travail analytique, car une centaine de prélèvements était envisagée. Finalement les populations choisies ont été les suivantes:

- a) copeaux normaux, brillants;
- b) copeaux peu oxydés, noirs;
- c) copeaux oxydés, blancs;
- d) copeaux fluorescents.

Les populations 2 et 3 ont été obtenues par oxydation limitée des copeaux par une exposition plus ou moins longue à l'air sec. La population 4 a été obtenue par trempage des copeaux dans une solution benzénique de naphtaline, essorage et séchage.

De nombreux échantillons ayant été prélevés au cours de quatre passages successifs du lot de copeaux dans l'appareil à homogénéiser, l'analyse de chaque échantillon a pu être faite par comptage visuel des copeaux de chaque catégorie. De cette manière une journée a suffi pour effectuer l'ensemble de l'expérience et des «analyses» et, après exploitation mathématique des résultats, il est apparu que le lot homogène était convenablement représenté par trois prélèvements de 80 à 100 g chacun.

L'homogénéiseur rotatif (fig. 5) a permis au cours du dernier passage de prélever la quantité d'échantillon désirée, grâce à l'ouverture prérégulée d'une trappe sur une goulotte spécialement prévue.

Les conclusions de cette étude ont été vérifiées à posteriori, d'une part, par le fait que l'uranium produit avec le calcium homogénéisé et contrôlé de cette façon répond aux exigences du cahier des charges, et d'autre part,
par le fait qu'une pollution accidentelle dans la fabrication de calcium a été décelée immédiatement.

Cet exemple montre l'intérêt d'une étude statistique de l'échantillonnage aussi bien pour le producteur que pour le client, puisqu'elle évite toute contestation sur les caractéristiques du produit. Cette étude est rentable également pour l'analyste, puisque le nombre d'analyses nécessaires pour caractériser un lot a été dix fois moins grand que celui exigé par la méthode qui paraissait normale. Il est bien évident d'ailleurs que le prix de revient du calcium profite heureusement de cette diminution du nombre des analyses.

2.2. Echantillonnage de l'uranium

Avant de prendre une décision sur le mode de prélèvement d'un échantillon de métal, il importe de connaître les variations de composition au sein même de ce métal. On peut rencontrer:
- des hétérogénéités grossières;
- des hétérogénéités locales (inclusions).

Des études de ce genre ont été faites sur des lingots d'uranium brut de raffinage, à différents niveaux de pollution. On a fait des prélèvements de copeaux répartis dans plusieurs plans (fig. 6).

La figure 7 représente à titre d'exemple les résultats de dosage du fer. Pour l'une des billettes où la concentration en fer est en moyenne égale à 85 ppm, la répartition est très homogène. Toutes les valeurs trouvées
FIG. 6. Etude de prélèvement sur lingot d'uranium FIG. 7. Répartition du fer dans des lingots d'uranium

... sont en effet comprises entre 75 et 95 ppm, alors que la précision de l'analyse est de l'ordre de 10%. Mais pour l'autre billette, dont la teneur en fer est de l'ordre de 1000 ppm, la répartition paraît fortement hétérogène. Si l'on considère seulement les prélèvements au centre du lingot, on constate une variation progressive d'une extrémité à l'autre. Mais cette corrélation ne se retrouve pas sur les génératrices extérieures.

L'hétérogénéité des lingots peut varier suivant les éléments considérés. Par exemple, on a examiné 1000 barreaux d'uranium refondus sur lesquels on a fait des prélèvements en tête et en pied. Parmi tous les éléments analysés, seul le silicium est réparti de façon nettement hétérogène. En effet, on a trouvé que dans 3% des barreaux la teneur en silicium dépassait 100 ppm pour les deux échantillons, dans 18% des barreaux elle dépassait 100 ppm à la tête, et dans 1,5% des barreaux elle dépassait 100 ppm uniquement au pied.

On a pu déterminer ainsi, pour les productions courantes d'uranium, des conditions de prélèvement d'échantillons qui offrent des garanties suffisantes et peuvent être considérées comme représentatives.

Notons que, pour autant que la production reste à un niveau inférieur aux spécifications des cahiers des charges, on a pu diminuer notablement le nombre d'analyses effectuées en groupant certains échantillons pour effectuer des analyses moyennes à côté d'analyses individuelles plus espacées et conserver de bons renseignements sur la production. Si le niveau des impuretés remonte, le système des analyses de contrôle est immédiatement resserré.

2.3. Échantillonnage du zirconium

Pour le zirconium, élaboré en lingots de l'ordre de 500 kg, un système analogue a été étudié et un échantillon pondéré moyen préparé à partir de
prélevements de copeaux en différents points du lingot, compte tenu de la répartition moyenne des impuretés dans le lingot.

3. ECHANTILLONNAGE DES MINERAIS

3.1. Généralités

Le minerai d'uranium extrait en France, soit par les exploitations du CEA, soit par des sociétés privées qui le rendent au CEA, est traité dans trois usines (l'Écarpière, Bessines et le Forez). Ces usines produisent des concentrés commerciaux expédiés aux usines de raffinage du Bouchet et de Malvesi.

Les usines de l'Écarpière et de Bessines sont la propriété de la Société industrielle des minerais de l'ouest (SIMO), filiales des établissements Kuhlmann et du CEA. L'usine du Forez appartient au CEA et est gérée par la SIMO.

Ces usines traitent le minerai à façon pour le CEA; elles reçoivent une rémunération qui est fonction du tonnage traité et du rendement obtenu. Il est donc important, à la fois pour le CEA et pour la SIMO, de connaître le tonnage et la teneur du minerai traité. Nous décrirons donc le système de contrôle utilisé à l'usine de Bessines.

3.2. Principe du traitement (fig. 8)

Le minerai d'une teneur moyenne en uranium voisine de 0,15% est concassé et débourbé dans un atelier appartenant au CEA. Il est livré à la SIMO sous deux formes: des grains de granulométrie 0,5 à 50 mm et des pulpes qui contiennent le minerai de dimension inférieure à 0,5 mm.

L'usine SIMO comprend trois lignes de traitement: chacune est capable de traiter 700 tonnes de minerai par jour. Les grains et les pulpes sont mélangés par un procédé de broyage et de classification qui sépare des «sables» (0,15 à 0,5 mm) et des «fines» (moins de 0,15 mm). Les sables d'une teneur moyenne en uranium voisine de 0,06% sont lixivités par l'acide sulfurique dans des tambours tournants; les fines d'une teneur moyenne en uranium voisine de 0,25% le sont dans des cuves munies d'agitateurs mécaniques.

3.3. Comptabilisation du minerai

La comptabilisation se fait sur chaque ligne de traitement, en tête de chaque section d'attaque, séparément sur les sables et sur les fines, à la fois par détermination du tonnage traité et analyse de la teneur en uranium du minerai.

a) Détermination sur les sables

La détermination du tonnage de sables traité journallement se fait par pesée des sables égouttés, sur des bascules Weiller pesant en continu sur bandes transporteuses.
Deux échantillons sont prélevés dans la goulotte alimentant ces bascules :
- l'un pour la détermination de l'humidité,
- l'autre pour l'analyse de la teneur en uranium.

Le premier échantillon est obtenu en récupérant une fois par poste la totalité du produit débité pendant quelques secondes, pour obtenir un échantillon pesé puis séché à l'étuve qui permet de déterminer l'humidité à attribuer au poids de sables donné par les bascules. L'étude des humidités successives a montré une faible dispersion se répartissant pratiquement au hasard et justifiant cette façon de procéder.

Les échantillons pour teneur sont fournis par des échantillonneurs rectilignes automatiques classiques du type Minerais et Métaux.

b) Détermination sur les fines inférieures à 0,15 mm

Le tonnage de produits traité journellement sur chaque ligne dans les pulpes fines se fait par mesure du débit et détermination du poids de solide dans l'unité de volume.

A cet effet, un échantillonneur automatique fournit, à l'entrée de chaque circuit de cuves d'attaque, un échantillon recueilli chaque jour qui sert à la fois à la détermination du poids de solide contenu dans un litre de pulpe et à l'analyse de la teneur en uranium de ce solide.

Le volume de pulpe entrant en tête de chacune des trois lignes d'attaque des fines est donné par un débitmètre Altoflux. Cet appareil est fondé sur la mesure de la force électromotrice induite par le déplacement du liquide,
que l'on peut considérer comme conducteur, dans un champ électromagnétique créé par une bobine inductrice entourant une portion de la canalisation dans laquelle passe la pulpe. Un amplificateur électronique actionne un totalisateur de débit dont la lecture journalière fournit le volume de pulpe traité.

Les débitmètres sont vérifiés chaque jour dans les conditions mêmes du traitement, en détournant une certaine quantité de pulpe, pendant environ dix minutes, dans une cuve étalonnée. Les petits écarts qui apparaissent au cours de ces vérifications, de caractères aléatoires ou dus à de faibles dérivés de l'amplificateur, sont notés et pris en considération pour corriger le tonnage journalier traité. Si cet écart atteint 3%, une deuxième vérification est effectuée avant de procéder à la retouche éventuelle du réglage des appareils. On évite ainsi des interventions trop fréquentes qui rendraient difficile l'interprétation des données.

L'étude des fluctuations statistiques montre que la précision de cet échantillonnage doit être excellente par suite de la composition des écarts aléatoires entre les lignes de traitement et entre les sables et les fines. Les opérations d'échantillonnage sont faites par le personnel de l'usine chimique en présence d'un représentant du CEA et les analyses journalières sont effectuées par les laboratoires locaux du CEA et de la SIMO.

3.4. Précision

La précision statistique de l'ensemble des opérations contribuant à déterminer la quantité d'uranium traitée a été calculée à l'échelle de l'année; elle est de l'ordre de 0.2% au niveau de deux écarts types (95%). Les erreurs accidentelles ou systématiques sont évidemment plus difficiles à chiffrer; elles ne peuvent être évitées que par des études objectives et détaillées. De telles études ont été entreprises conjointement par la SIMO et le CEA, qui ont servi à établir un cahier des charges de l'échantillonnage où sont détaillées les opérations, ainsi que la fréquence et la nature des contrôles à effectuer périodiquement pour l'entretien des appareils de mesure.

4. ECHANTILLONNAGE DE CONCENTRES D'URANIUM

4.1. Généralités

Les concentrés d'uranium produits en France sont des uranates de magnésie à l'Ecarpière et à Bessines, des nitrates d'uranyle liquides au Forez et à Gueugnon (cette dernière usine recevant non pas des minerais, mais des préconcentrés obtenus en République du Gabon). Deux systèmes d'échantillonnage sont donc utilisés que nous décrirons rapidement.

4.2. Uranates de magnésie (fig. 9)

a) Les uranates titrent entre 65 et 70% d'uranium sur sec et renferment de 5% (l'Ecarpière) à 15% (Bessines) d'humidité. Ils sont chargés dans
des conteneurs métalliques de 1 m³ de capacité. L'échantillonnage se fait par lots de dix conteneurs représentant 7 à 9 t d'uranium.

b) Le prélèvement des échantillons se fait à l'aide d'une sonde de section triangulaire maniée à la main. On effectue neuf prélèvements par conteneur représentant au total de 10 à 15 kg, soit 1 à 1,5% du poids de l'uranate humide.

c) La totalité de l'échantillon prélevé sur un conteneur est recueillie immédiatement sur un plateau en acier inoxydable, pesé, puis placé dans une étuve réglée à 140°±10°C. Le séchage dure au moins 36 h (en pratique près de 48 h). L'échantillon séché est repesé et on calcule ainsi l'humidité de l'uranate de chaque conteneur.

d) Le poids de l'uranate humide est déterminé par pesée de chaque conteneur avant et après remplissage. Cette pesée se fait sur une bascule à cadran qui donne le poids à 500 g près.

e) Après séchage, chaque échantillon de conteneur est broyé à moins d'un millimètre puis divisé. On recueille 1500 g environ de poudre à partir de laquelle on prélève à la spatule un échantillon représentant un millième du poids d'uranate sec du conteneur.

Les échantillons correspondant aux dix conteneurs du lot sont mélangés dans un broyeur à boulets, puis broyés à environ 100 μm. On prélève enfin à la spatule le contenu de dix flacons de 125 cm³ qui sont cachetés à la cire et envoyés aux laboratoires d'analyse.

4.3 Nitrates d'uranyle (fig. 10)

a) Le nitrate d'uranyle titre 400 à 425 g/litres d'uranium; il a une densité voisine de 1,55 et un pH d'environ 1,5. Il est chargé dans des con-
teneurs de 2,5 m³ de capacité, à parois d'acier recouvertes d'un revêtement anti-corrosion.

L'échantillonnage se fait par lots de quatre conteneurs représentant environ 9 m³ de nitrate, soit 3,6 à 3,8 t d'uranium.

b) A partir d'une cuve de stockage de 50 m³, des pompes alimentent une cuve d'homogénéisation qui est remplie jusqu'au niveau d'un trop-plein réglé à la capacité d'un conteneur.

Après homogénéisation (15 min), la cuve est vidée complètement dans le conteneur placé sur une bascule et préalablement taré. Un ajutage permet de prélever 1 à 2 litres d'échantillon dans un récipient couvert (pour éviter les pertes par évaporation).

c) Pour chaque conteneur, on prélie à partir de l'échantillon précédent une quantité de nitrate représentant un millième de son poids net. Les quatre prélèvements du lot sont mélangés, puis on remplit des flacons envoyés aux laboratoires d'analyse.

5. ÉCHANTILLONNAGE DE PRODUITS INTERMEDIAIRES SORTANT DES RAFFINERIES ET BILAN URANIUM DANS UNE UNITÉ DE RAFFINAGE

5.1. Généralités

L'uranium est élaboré dans les usines françaises à partir des concentrés solides et liquides mentionnés à la section 4. Le processus comprend
- la purification, qui consiste (après dissolution nitrique dans le cas de concentré solide et directement si le concentré est liquide) à extraire au phosphatede tributyle l'uranium sous forme de nitrate d'uranyle, après réextraction à l'eau déminéralisée. Le diuranate d'ammonium est ensuite précipité à l'ammoniaque, puis séché et calciné sous forme de trioxyde d'uranium $UO_3$;

- la réduction - fluoration, qui consiste dans un seul réacteur à réduire par l'ammoniac le trioxyde en bioxide qui est ensuite transformé par du gaz fluorhydrique en tétrafluorure d'uranium $UF_4$;

- l'élaboration, au cours de laquelle le tétrafluorure d'uranium est transformé en uranium métal brut par du magnésium (ou du calcium), le sous-produit de cette transformation étant pour l'usine considérée des scories de fluorure de magnésium.

5.2. Echantillonnage et bilan uranium

Nous décrirons les opérations d'échantillonnage qui intéressent les produits intermédiaires pouvant faire l'objet de transactions et qui interviennent également pour le bilan avec d'autres produits.

La figure 11 schématiser l'établissement du bilan uranium d'une usine de raffinage. Elle fait apparaître que, pour établir le bilan uranium à un instant donné, l'installation étant arrêtée, on doit déterminer les teneurs en uranium, donc prélever des échantillons sur les produits suivants:
a) Produits entrant en fabrication

- Concentrés uranifères solides ou liquides (uranate ou nitrate)
- Déchets uranifères métalliques

b) Produits en cours de traitement

A la veille d'un bilan uranium les «en cours de traitement» sont réduits qualitativement au strict minimum afin d'annuler l'incidence des liquides ou des solides hétérogènes de nature mal définie et de poids difficile à déterminer. C'est ainsi qu'on se trouve uniquement en présence de (voir fig. 11):

Au stade purification

- Nitrate d'uranyle impur titrant environ 400 g/litres d'uranium
- Nitrate d'uranyle de pureté nucléaire titrant environ 120 g/litres d'uranium
- Trioxyde d'uranium se présentant sous forme de petites écailles orange d'une densité apparente de 1,0 contenant approximativement 81,5% d'uranium, conditionné en fûts de 115 litres pesant en moyenne 115 kg

Au stade réduction - fluoration

- Tétrafluorure d'uranium se présentant sous forme de petits grains vert d'une densité apparente de 1,8 contenant approximativement 76% d'uranium, conditionné en fûts de 200 litres pesant en moyenne 360 kg

Au stade élaboration - décapage - refusion

- Scories de fluorure de magnésium
- Uranium métal sous forme de lingots de différents types:
  
  210 kg, diamètre 290 mm
  450 kg, diamètre 290 mm
  450 kg, diamètre 400 mm

c) Produits sortant de fabrication

- Trioxyde d'uranium UO₃ décrit ci-dessus
- Tétrafluorure d'uranium UF₄ décrit ci-dessus
- Uranium métal sous forme de lingots des différents types
- Sesquioxyde d'uranium U₃O₈ d'une densité apparente de 2,8 contenant environ 84,0% d'uranium conditionné en fûts de 115 litres pesant en moyenne 320 kg
ARTAUD et al.

- Effluents liquides constitués par les eaux mères ammoniacales de précipitation du diuranate d'ammonium et titrant en moyenne 0,010 g/litres d'uranium et par les pieds de colonnes d'extraction au phosphate de tributyle du nitrate d'uranyle et titrant en moyenne 0,025 g/litres d'uranium.

- Fluorure de magnésium récupéré se présentant sous forme de poudre sèche stockée en fûts de 115 litres pesant en moyenne 230 kg et répondant aux caractéristiques principales suivantes:
  
  - densité apparente: 2,0
  - teneur moyenne en uranium: 0,3%

**Produits entrant en fabrication**

a) Concentrés uranifères solides ou liquides (uranates ou nitrates)

Les concentrés sont échantillonnés selon des règles analogues à celles décrites à la section 4.

b) Déchets uranifères métalliques

Ces déchets peuvent provenir de diverses usines (usinage). La teneur en uranium de ces déchets est estimée et une vérification est effectuée par détermination de la teneur en uranium de l'U₃O₈ auquel ils ont donné naissance. Ceci sous-entend évidemment que le jour du bilan tous les déchets uranifères métalliques ont été transformés en U₃O₈.

![Diagram of a tube with dimensions and labels](image)

**FIG. 12.** Sondes de prélèvement pour produits intermédiaires

L'U₃O₈ conditionné en fûts pesés à 100 g près est échantillonné fût par fût à l'aide d'une sonde (voir fig. 12). Cette sonde est constituée par un tube de 1200 mm de long et de 19 mm de diamètre, muni d'une lumière longitudinale de 1000 mm de long et de 9 mm de large.

<table>
<thead>
<tr>
<th>Phi int</th>
<th>UC₃</th>
<th>UF₃</th>
<th>U₃O₈</th>
<th>FeMn</th>
</tr>
</thead>
<tbody>
<tr>
<td>L₁</td>
<td>1200</td>
<td>25</td>
<td>19</td>
<td>14</td>
</tr>
<tr>
<td>L₂</td>
<td>1000</td>
<td>1000</td>
<td>1000</td>
<td>700</td>
</tr>
<tr>
<td>I</td>
<td>11</td>
<td>15</td>
<td>9</td>
<td>5</td>
</tr>
</tbody>
</table>
Il est donné un seul coup de sonde par fût, ce qui représente un prélèvement de 200 g, soit 0,06% de la quantité totale de $\text{U}_3\text{O}_8$ échantillonné.

Produits en cours de traitement

a) Nitrate d'uranyle (impur ou de pureté nucléaire)

Au moment de la vidange complète et du rinçage de l'installation de purification, les nitrates d'uranyle sont rassemblés dans des cuves de stockage après avoir transité par une cuve homogénéisée et pesée. À chaque pesée un échantillon représentant 0,05% en volume du volume total échantillonné est prélevé par un échantillonneur continu à orifice calibré.

b) Trioxyde d'uranium

Le stock intermédiaire de trioxyde d'uranium, conditionné en fûts pesés à 100 g près, est échantillonné à la sonde (voir fig. 12) au rythme d'un coup de sonde par fût représentant un prélèvement de 200 g, soit 0,17% de la quantité totale de $\text{U}_3\text{O}_8$ échantillonné.

La teneur en uranium est déterminée par calcination directe au four à moufle et pesée de 1$\text{U}_3\text{O}_8$ obtenu. Cette méthode conduit à une approximation de ± 0,1%.

c) Tétrafluorure d'uranium

Le stock intermédiaire de tétrafluorure d'uranium homogénéisé, conditionné en fûts pesés à 500 g près, est échantillonné à la sonde (voir fig. 5) au rythme d'un coup de sonde par fût représentant un prélèvement de 500 g, soit 0,14% par rapport à la quantité totale de $\text{UF}_4$ échantillonné.

d) Scories de fluorure de magnésium

L'uranium contenu dans les scories de fluorure de magnésium est, par définition, égal à la chute de rendement d'élaboration.

e) Uranium métal

La quantité d'uranium métal est déterminée par pesée sur une bascule.

Produits sortant de fabrication

a) Trioxyde d'uranium (voir ci-dessus)
b) Tétrafluorure d'uranium (voir ci-dessus)
c) Uranium métal (voir ci-dessus)
d) Sesquioxyde d'uranium (voir ci-dessus)
e) Effluents

Avant rejet les effluents sont parfaitement homogénéisés dans leur cuve étalonnée. Leur volume est calculé, et après prélèvement de l'échantillon,
la teneur en uranium est déterminée à 0,5% près, par colorimétrie au dibenzoylméthane.

f) Fluorure de magnésium récupéré

Le fluorure de magnésium récupéré, conditionné dans des fûts pesés à 500 g près et groupés par lots de fabrication, est échantillonné à la sonde (voir fig. 5) au rythme d'un coup de sonde par fût représentant un prélèvement de 150 g, soit 0,07% par rapport à la quantité totale de fluorure échantillonné.

5.3. Conclusions

Dans une unité de raffinage de l'uranium, le rendement étant voisin de 100%, l'importance des effluents est minime vis-à-vis de l'uranium entrant en fabrication sous forme de concentrés uranifères solides ou liquides.

C'est ainsi que depuis quelques années l'effort, en matière d'échantillonnage et d'analyse, a surtout porté sur la détermination des teneurs en uranium dans les concentrés uranifères de toute nature. Les progrès réalisés nous conduisent aujourd'hui à une incertitude n'excédant pas 0,10% sur l'uranium total mis en jeu. Notons toutefois que les concentrés liquides permettent d'arriver à une bien meilleure précision que les concentrés solides.

6. PROBLEMES DES SPECIFICATIONS LIEES A L'ECHANTILLONNAGE

Le contrôle qualitatif des produits est, comme nous l'avons dit précédemment, étroitement lié au problème des spécifications techniques. En raison de l'évolution rapide de l'industrie nucléaire, ces spécifications restent trop assujetties à des considérations qui ne sont pas assez réalistes. On s'appuie plus souvent sur des bulletins d'analyse type portant sur de nombreux éléments que sur des spécifications réellement étudiées, car on n'a pas toujours l'expérience nécessaire pour établir des spécifications rationnelles.

Il est nécessaire de faire un gros effort pour repenser le problème des spécifications dans les transactions qui intéressent les matériaux nucléaires.

Une étude sérieuse du problème doit permettre de classer les éléments soumis à des spécifications en fonction de divers impératifs (neutronique, corrosion, procédé, accessibilité après irradiation, etc.). Cet effort devrait permettre d'alléger les analyses de recette et même les contrôles de fabrication, donc de diminuer le prix de revient des analyses; sans doute aurait-il également une incidence bénéfique sur le prix de ces matériaux.

7. CONCLUSIONS

Nous avons examiné un certain nombre d'exemples d'échantillonnage et d'études d'échantillonnage dans le cadre des transactions de matériaux
nucléaires en France. Les solutions adoptées en ont en général montré la validité.

Pour conclure, nous insisterons une fois de plus sur le fait qu'il n'existe aucune solution générale à l'échantillonnage, qui est toujours un cas d'espèce; il faut continuellement avoir à l'esprit que l'échantillonnage, aussi bien étudié soit-il, peut être remis en cause par des considérations secondaires comme l'étalonnage de cuve, des variations de température de liquide, des pollutions dues à l'emballage, des modifications ultérieures de l'échantillon (variation d'humidité par exemple), qui annihilent les efforts faits au cours des études et éliminent la représentativité de l'échantillon prélevé.

BIBLIOGRAPHIE


DISCUSSION

J. JENNEKENS: Your paper describes a mechanical proportional-sampling system. Can you give an indication of the accuracy of mechanical sampling of uranium concentrates as compared with manual sampling?

J. ARTAUD: I have no valid data to give regarding the use of this type of sampler in the French uranium concentrates industry. The only case in which figures have been compared was for liquid concentrates, but this is a very special case.

J. JENNEKENS: Table I of Mr. McCluen's paper* suggests that the greatest improvement in precision in uranium analysis of UF₆ can be made by increasing the number of analytical determinations per sample rather than by increasing the number of samples. This is evidently because of the cancelling out of random errors in the analytical determination. I would like to know whether French experience corroborates this conclusion.

I should also like to point out that while the precision of the results is increased by multiple determinations the absolute accuracy remains unchanged. This is because systematic errors (e.g. non-representative sampling) are unchanged. It would appear, therefore, that a compromise between multiple sampling and multiple determinations is required in order to minimize both random and systematic errors.

J. ARTAUD: Personally, I have no experience with UF₆. This, too, is a special case.

K. NAKAJIMA (Chairman): Perhaps Mr. Bennett would like to comment on this.

C. BENNETT: There is no question of accuracy here. We are dealing only with estimates of the precision attainable using various schemes of sampling and analysis. Mr. McCluen has been able to estimate the components of variance from each of these sources, and has inferred that that

* These proceedings (SM-67/31).
due to sampling is reasonably small. For economic reasons he therefore prefers to obtain the necessary precision by reducing the analytical component through repeated determinations on a single sample. Any procedure not involving repeated sampling will lead to a fixed contribution to the variance due to sampling error, and the precision attainable by repeated determinations is limited by this fixed (albeit small) contribution due to the sampling error. Stated algebraically, n replicate deformations on a single sample will lead to an over-all precision $\sigma$ given by

$$\sigma^2 = \sigma_5^2 + \sigma_A^2/n$$

where $\sigma_5^2$ is the component due to sampling and $\sigma_A^2$ the component due to analysis. In this case the attainable precision, regardless of the size of n, is greater than $\sigma_5^2$. Operationally, we can state that once a sample is taken, the error due to sampling becomes fixed, and the most we can do by repeated analysis is determine more precisely the content of the sample. If we are willing to assume that the sample is sufficiently representative (which is the case when $\sigma_5^2$ is small), this is a perfectly satisfactory procedure.
URANIUM ORE AND CONCENTRATE SAMPLING

F.E. McGINLEY, D.L. BROWN AND R.W. LANGRIDGE
UNITED STATES ATOMIC ENERGY COMMISSION, GRAND JUNCTION, COLO., UNITED STATES OF AMERICA

Abstract — Résumé — Аннотация — Resumen

URANIUM ORE AND CONCENTRATE SAMPLING. The Grand Junction Office of the United States Atomic Energy Commission has been responsible for procuring large quantities of natural uranium in both ores and concentrates. The techniques used for sampling ores are necessarily different from those used for concentrates. Each step in the overall measurement and sampling systems for both ores and concentrates is discussed, giving particular attention to the accuracy and precision of that step. During the years 1948-1964, a total of 58 million tons of ore was sampled in about 40 different mechanical sampling plants in the western United States. All plants have been required to weigh, sample and analyse ore in accordance with practices satisfactory to the USAEC. The ordinary principles of ore sampling, as used for years in the mining industry, have been followed. However, sufficient check sampling and other tests were performed to ensure that the uranium content of the variety of ores sampled was as accurately determined as economically feasible.

Concentrates containing about 129,000 t of $\text{U}_3\text{O}_8$ were purchased from domestic producers during the last 17 years. This uranium was contained in approximately 10,000 lots, each of which was weighed, sampled, and analysed in accordance with carefully controlled procedures. These lots were received at USAEC-owned sampling facilities at Grand Junction or Weldon Spring, Missouri, both of which are contractor operated. The average lot consists of about 50 drums (55-gallon size) and weighs approximately 35,000 lb. Because concentrate varies so much in both physical and chemical characteristics, it is necessary to sample each drum. Through the years, various sampling systems were used, such as pipes, open auger, enclosed augers, and falling stream sampling. Falling stream sampling is the most accurate, provided precautions are taken to prevent changes in weight due to exposure to the atmosphere. Because of the tendency of concentrates to sorb or desorb moisture, depending upon the atmosphere to which they are exposed, special sub-sampling, drying, and sample preparation procedures were continuously developed to improve the accuracy and precision of the overall purchase sampling. Extensive use was made of statistics to analyse and interpret measurement and sampling evaluation data.

ECHANTILLONNAGE DES MINERAIS ET DES CONCENTRES D'URANIUM. Les services de la Commission de l'énergie atomique situés à Grand Junction (Colorado) sont chargés de fournir de grandes quantités d'uranium naturel sous forme de minerais ou de concentrés. Les méthodes utilisées pour échantillonner les minerais sont nécessairement différentes de celles utilisées pour les concentrés. Les auteurs étudient les différentes opérations de mesure et d'échantillonnage pour les minerais et les concentrés, en tenant plus particulièrement compte de l'exactitude et de la précision de ces opérations. Entre 1948 et 1964, 58 millions de tonnes de minerais au total ont été échantillonnées dans 40 installations différentes d'échantillonnage mécanique situées dans l'ouest des États-Unis. Toutes ces installations ont dû peser, échantillonner et analyser le minerai selon des méthodes agréées par la Commission de l'énergie atomique. Les principes courants d'échantillonnage de minerai utilisés depuis des années dans l'industrie minière sont appliqués. Il est toutefois procédé à des échantillonnages de contrôle et à d'autres essais pour s'assurer que la teneur en uranium des différents minerais échantillonnés est déterminée avec toute la précision économiquement possible.

Au cours des 17 dernières années, des concentrés contenant environ 129,000 tonnes d'U3O8 ont été achetés à des producteurs du pays. Cet uranium se trouvait dans 10,000 lots environ, dont chacun a été pesé, échantillonné et analysé selon des méthodes soigneusement contrôlées. Ces lots ont été expédiés aux installations d'échantillonnage de Grand Junction ou de Weldon Spring (Missouri), toutes deux propriétés de la Commission de l'énergie atomique et dirigées par des exploitants sous contrat. Un lot moyen comprend environ 50 fûts (de 210 litres) et pèse environ 16 tonnes. Comme les caractéristiques physiques et chimiques de ces concentrés varient considérablement d'un fût à l'autre, il est nécessaire d'échantillonner chaque fût. Au cours de ces années, on a eu recours à différents procédés d'échantillonnage - tubes, sondes ouvertes, sondes fermées ou écoulement du minerai. Ce dernier procédé est le plus précis à condition que l'on prenne les précautions re-
quises para prevenir los cambios de peso resultantes de la exposición a la atmósfera. Como las concentraciones tienden a absorber la humedad ambiente o a perder su humedad según las condiciones atmosféricas, hemos recurrido mucho a la estadística para analizar y interpretar los datos y medidas. Para prevenir los cambios de peso a consecuencia de la exposición a la atmósfera, hemos utilizado una técnica de reducción de volumen y preparación de muestras para calcular la cantidad de muestras que se deben tomar, especialmente cuando las concentraciones tienden a absorber la humedad ambiente o a perder su humedad por efecto de la atmósfera a la que están expuestas. Como métodos de reducción de volumen y preparación de muestras, utilizamos una técnica de reducción de volumen y preparación de muestras para calcular la cantidad de muestras que se deben tomar, especialmente cuando las concentraciones tienden a absorber la humedad ambiente o a perder su humedad por efecto de la atmósfera a la que están expuestas.
de cada barril. Durante estos años se han empleado diversos sistemas de muestreo: por tubos, barrenas cerradas, barrenas abiertas, muestreo en corriente libre. Este último es el más exacto, a condición de que se tomen las precauciones necesarias para impedir los cambios de peso debidos a la exposición a los factores atmosféricos. Teniendo en cuenta la tendencia de los concentrados a perder o absorber humedad según la atmósfera a que se hallan expuestos, se han ideado procedimientos especiales de submuestreo, secado y preparación de muestras con objeto de aumentar la precisión de las operaciones. Para analizar e interpretar los datos proporcionados por las mediciones y el muestreo se recurre en gran medida a la estadística.

INTRODUCTION

The first of many quantitative measurements of uranium, as it enters the production chain, are those performed at the ore buying station, or mill, where the ore is processed. The United States Atomic Energy Commission (AEC) becomes accountable for uranium when it is received for purchase by the AEC in the form of ore or concentrate. However, since the expiration of the AEC or purchase programme on 31 March, 1962, only uranium in concentrate has been purchased.

Historically, raw materials transactions have been in terms of contained pounds of U₃O₈, but the AEC also maintains source materials accountability records in kilograms of uranium (U). In this presentation, we are concerned only with natural uranium, since it has been established that the isotopic composition of both ores and concentrates is essentially constant.

The purpose of this paper is to describe the measurement methods used in the United States for determining the uranium content of domestic ores and concentrates. Ore sampling deals with large quantities of low value (about $20 per ton) material, whereas concentrate sampling involves smaller quantities of high value ($12 000-$16 000/t) material. The differing characteristics of the two materials require different sampling methods, and, for this reason, ore sampling will be discussed separately from concentrate sampling.

ORE SAMPLING

General Information

The AEC began purchasing uranium ores in July 1948 at the Monticello, Utah, buying station. Ores were purchased in accordance with a guaranteed price schedule to stimulate domestic production. As the need arose, additional ore buying stations were constructed in other areas by both the AEC and private industry. In all, some 40 mechanical uranium ore sampling plants have operated in the western United States, and a total of approximately 58 million tons of ore were sampled during the years 1948-1964. Of this total, approximately 3.5 million tons of ore were purchased by the AEC at 16 locations during its ore purchase programme. Currently, there are 15 uranium ore sampling plants in operation in the United States and all are privately owned.

The AEC has always taken an active interest in the weighing, sampling and analysing of uranium ores at both AEC and privately owned facilities. The principles of sampling uranium ore are the same as those applied by the mining industry in sampling other ores of comparable value and heterogeneity.
Ore is delivered, either by truck or railway wagon, to the ore sampling plant where it is accumulated in lots. Lot sizes vary among plants, depending, of course, upon the size of the operation; but, generally, they are a minimum of 10 t and may be as large as 1500 t. The mine-run ore usually contains 1-10% moisture and is less than 12 in. (30 cm.) in size. The majority of ores are relatively easy to crush and sample, although each sampling plant must be capable of accurately sampling the most heterogeneous ore it receives. The average ore contains about 0.25% U₃O₈, but individual ore lots may range from less than 0.10 to several percent U₃O₈. The most common ores are sandstones containing uranium minerals disseminated as grain coatings and cement between sand grains. Individual ore particles may vary several fold in uranium content. For this reason, it has been general practice to require a minimum sample weight at different particle sizes, regardless of the size of the lot, unless it is demonstrated that smaller sample weights produce representative final samples of the type of ore handled at a particular location.

At all AEC ore sampling plants, periodic precision checks were made by re-sampling a specified number of lots each month. In addition, between-plant comparisons were made by sampling the same lot at two or more buying stations, both AEC and private. For the average grade ore, sampling plants ordinarily obtained over-all sampling and assaying results that agreed within ±0.02% U₃O₈.

A typical flowsheet for uranium ore sampling is shown in Table I. Unit equipment has not been standardized and consequently many different types are used. Generally, three or four stages of crushing and mechanical sampling are employed and the most favoured kinds of samplers are chain-bucket, straight-line and continuously rotating arc-path cutters [1].

In buying custom ore, the seller or shipper is given a sample representing each lot delivered. The lot is paid for on the basis of the buyer's U₃O₈ analysis, unless the seller submits his assay and requests an analysis by an independent laboratory (umpire). The umpire is chosen from a list of qualified laboratories, and the settlement assay is the middle one.

Each step in the over-all measurement of the uranium content of ores will be described now in greater detail.

Weighing

Trucks and railway wagons of ore are weighed on platform-type scales, dumped, and then re-weighed empty in order to obtain the tare and the net wet weight of ore. The scales are calibrated periodically with standard weights and the AEC requires all scales to be certified for accuracy by State inspectors twice each year. The scales are a registering beam-type, with print-out mechanism, and the precision generally is within ±0.1% at 95% confidence.

Moisture determination

Immediately after weighing, a grab moisture sample of 10-20 lb is taken from each load to determine the dry weight of ore. Before drying can occur
# TABLE I

## TYPICAL FLOWSHEET FOR URANIUM ORE SAMPLING

- **Truck Shipment**
  - (-12 in)
  - **Platform Scales**

- **Moisture Sample**
  - **Lot Receiving Area**
  - **Truck Empty**

- **Crushing & Mixing**
  - **Primary Crusher**
    - (2 in-6 in)
  - **Secondary Crusher**
    - (1/2 in-2 in)

- **Oven - 110°C**
  - **Moisture Scale**
  - **Percent Moisture**

- **Rejects**
  - **No. 1 Mechanical Sampler**
    - (10-20%)
    - **Crusher (Jaw, Rolls, etc.)**
      - (1/4 in-1 in)
    - **Rejects**
  - **No. 2 Mechanical Sampler**
    - (5-15%)
    - **Crusher (Jaw, Rolls, etc.)**
      - (1/8 in-1/2 in)
    - **Rejects**
  - **No. 3 Mechanical Sampler**
    - (5-20%)
  - **Sample Container**
  - **Ore Dryer** (if required)

- **Mechanical Blender (10 min)**
  - **Stationary Sampler (Riffles)**
    - (100 lb)
  - **Grinder (Cone and Ring Type)**
    - (10 mesh)

- **Mechanical Blender (10 min)**
  - **Stationary Sampler (Riffles)**
    - (10 lb)
  - **Oven** (at 110°C 24 hr)

- **Pulverizer (Disk Type)**
  - (40 mesh)

- **Mechanical Blender (10 min)**
  - **Stationary Sampler (Riffles)**
    - (2½ lb)

- **Sieve (120 mesh)**

- **Mechanical Blender (10 min)**

- **Sample Pulps** (4 to 6 100 g each)
  - **Rejects Bin**
  - **Stockpiles**
  - **Process**
these samples are crushed to about 1/2 in. maximum particle size, mixed, and then approximately a 3-lb portion is removed for drying at 110°C to constant weight [2]. Screen tests on sandstone ores have shown that normally there is no marked concentration of moisture in the fines. Also, repetitive moisture sampling of sandstone ore containing 3-5% moisture showed that the moisture determination has a precision of ±0.6% at 95% confidence. This moisture determination precision, in terms of percent contained U₃O₈, is less than the equivalent of ±0.002% U₃O₈, so, therefore, is acceptable [3].

**Mechanical sampling**

Although hand sampling of uranium ores was used in a few instances, mechanical sampling is preferred because it is inherently more accurate. However, mechanical sampling in itself does not guarantee accuracy, and it is necessary to examine sampling plants individually. It is common practice to determine the adequacy of mechanical sampling by increment sampling tests. Such tests involve the collection, or cutting, of increments of ore at specific particle sizes at pre-determined time intervals while ore lots are actually being sampled. Each increment is analysed and the standard deviation of the analyses is calculated. From these data, it is determined if a sufficient number of cuts is being taken at each particle size tested, using the following formula:

\[
N = \frac{S^2 t^2}{E^2}
\]

where S is the pooled standard deviation of the incremental samples in percent U₃O₈, t is the percentile of the S distribution at 95% confidence, and E, the allowable error, is the analytical error at 95% confidence expressed in percent U₃O₈.

Representative test data collected at one of the AEC sampling plants were as follows:

\[
S = 0.079\% \text{ U}_3\text{O}_8 \text{ (approximately } 1\frac{1}{2}\text{-in. particle size)}
\]

\[
t = 2.201, \text{ based on 12 increments}
\]

\[
E = 0.003\% \text{ U}_3\text{O}_8 \times 2.00 = 0.006
\]

\[
N = \frac{(0.079)^2 (2.201)^2}{(0.006)^2} = 840 \text{ sample cuts required}
\]

The actual number of cuts taken by the sampler during the period equalled 1275, so the frequency of sampling was more than adequate [3].

A typical screen analysis of sandstone ore crushed to pass a 28-mesh screen is shown in Table II. Because uranium is concentrated in the finer size fractions, care must be exercised in all steps of the sampling operation to avoid excessive dust losses that could downgrade the sample.
TABLE II

TYPICAL SCREEN ANALYSIS OF SANDSTONE ORE

<table>
<thead>
<tr>
<th>Tyler sieve opening (mesh)</th>
<th>Per cent by weight</th>
<th>Per cent ( \text{U}_3\text{O}_8 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>- 28+115</td>
<td>80.2</td>
<td>0.178</td>
</tr>
<tr>
<td>-115+200</td>
<td>13.8</td>
<td>0.319</td>
</tr>
<tr>
<td>-200+325</td>
<td>4.4</td>
<td>0.568</td>
</tr>
<tr>
<td>-325</td>
<td>1.6</td>
<td>0.992</td>
</tr>
<tr>
<td>Calculated grade</td>
<td>-</td>
<td>0.227</td>
</tr>
</tbody>
</table>

Sample preparation

The preparation of pulps suitable for chemical analysis is the final step in the sampling operation. The procedure shown in Table I has been tested and proved satisfactory for the most heterogeneous ores. It is important, in sample preparation, to reduce the maximum particle size and blend prior to each reduction is sample quantity. All equipment must be clean before beginning sample preparation and sample identity must be carefully maintained.

Analysis of ore samples

Uranium ore samples are commonly analysed by volumetric and X-ray spectrometric methods. In a laboratory where accuracy is carefully controlled through the use of standard samples, one may expect a precision of ±0.005% \( \text{U}_3\text{O}_8 \) at 95% confidence. Similarly, the precision of the average of three determination would be ±0.003% \( \text{U}_3\text{O}_8 \) [4]. In ore buying, it has been found that two competent laboratories can agree quite easily within ±0.005% \( \text{U}_3\text{O}_8 \) in analysing duplicates of properly prepared ore samples.

To maintain a high degree of accuracy and precision among government, mill and umpire laboratories, the AEC has for several years conducted an analytical comparison programme on ore samples. Two unknown samples are analysed each month by all participating laboratories and the analyses are submitted to the AEC (Grand Junction Office) for comparison. Assuming the average analysis is the true assay, the standard deviation and the 95% confidence limits are calculated, plotted in graph form, and sent to each participating laboratory. With between 20 and 30 laboratories participating, typical analytical confidence limits are shown in Table III.

CONCENTRATE SAMPLING

General information

The term "concentrate" refers to products that are essentially impure uranium oxides, and sodium and ammonium uranates. Concentrates vary
TABLE III
TYPICAL ANALYTICAL CONFIDENCE LIMITS

<table>
<thead>
<tr>
<th>Sample No.</th>
<th>Average per cent U₃O₈ with 95% confidence limits</th>
</tr>
</thead>
<tbody>
<tr>
<td>ACP 71</td>
<td>0.120 ± 0.003</td>
</tr>
<tr>
<td>ACP 67</td>
<td>0.208 ± 0.004</td>
</tr>
<tr>
<td>ACP 58</td>
<td>0.325 ± 0.005</td>
</tr>
<tr>
<td>ACP 48</td>
<td>0.452 ± 0.005</td>
</tr>
<tr>
<td>ACP 53</td>
<td>1.203 ± 0.018</td>
</tr>
</tbody>
</table>

considerably in both chemical and physical characteristics, and the characteristics of each concentrate are determined by the chemical process and method of drying employed in its production. The uranium content of concentrates measured in terms of per cent U₃O₈ varies from 75 to 98% and averages about 85% on a dry weight basis. It is necessary that concentrates be less than 1/4 in. in particle size and contain less than 10% moisture to permit accurate sampling.

Concentrates are delivered in 55-gal, full removable-head, steel drums with ring closures. A lot consists of from 20 to 75 drums, but averages about 50 drums totalling about 35 000 lb and contain about 30 000 lb of U₃O₈. Deliveries are made to the AEC at either Grand Junction, Colo., or Weldon Spring, Mo., where AEC contractors receive, weigh, sample and analyse each lot for payment purposes. Approximately 10 000 lots, containing about 129 000 lb of U₃O₈, were received from domestic producers during the last 17 yr.

Characteristics of concentrates

The concentrates range in particle size from 1/4 in. (6.35 mm) to less than 325 mesh (44 μm) and the U₃O₈ content may vary between particle sizes by as much as 10%. The concentrates may lose or gain weight in a drying oven at 110°C, and generally are quite sensitive to changes in the water vapour content of the air. The sensitivity of a typical concentrate to changes in the dew point of the drying oven air is shown in Fig. 1. Because of this sensitivity to dew point changes, the "drying" of concentrates at 110°C is not what is normally considered drying. Most concentrates lose weight until reaching equilibrium with the oven atmosphere. Thereafter, the weight is not stable, but varies measurably with changes in atmospheric relative humidity or dew point. Therefore, it is necessary, in accurate concentrate sampling, that the packaged analytical sample should be in the same "state of dryness", or, in other words, have the same absolute water content and per cent U₃O₈ as the "moisture" sample at the conclusion of the loss-on-drying measurement (LOD), whatever the equilibrium level reached in the oven atmosphere.
Sampling problems

The problems encountered in sampling uranium concentrates can be attributed to the following:

(a) Segregation of particles during drum filling,
(b) Selective removal by auger samplers of coarse particles in preference to fine particles, or vice versa,
(c) Poor flow characteristics,
(d) Hygroscopic nature, and
(e) Loss of moisture through excessive heating during pulverization.

Each of these sampling problems has been solved through improved sampling procedures and improved equipment.

Weighing

The drums are tared before filling and, upon receipt at the sampling site, are individually weighed to the nearest 1/2 lb. The drum weights range from 500 to 1000 lb and average about 800 lb.

The accuracy of the scales is maintained by frequently checking with standard weights. The precision maintained in routine weighing is approximately ±14 lb per 35,000 lb lot at 95% confidence.

Primary sampling

The official primary samplers have evolved from a simple pipe, through an open auger with template, to the now standardized semi-automatic enclosed augers and falling stream samplers.

Early in 1959, a compact, dust tight, two-stage falling stream sampler was constructed at Grand Junction. Later, a very similar unit was built at Weldon Spring. Drums are elevated to a dumping station above the unit where they are emptied into an upper surge hopper equipped with a stirring mechanism. The hopper discharges to a vibratory feeder that empties into the first-stage cutter box. Here, a reciprocating straight-line sample cutter removes a portion of the stream, directing it to a second vibratory feeder that discharges into the second-stage cutter box. The second-stage straight-line sample cutter discharges into the sample container. Both sample cutters have interchangeable heads for regulating the quantity of sample to be taken. The rejects from both cutter boxes drop into a lower surge hopper from which empty drums are filled.

The falling stream sampler at Grand Junction was evaluated for both accuracy and precision. The number of cuts required per lot for acceptably precise sampling was determined by increment sampling tests using the most heterogeneous concentrate available. The minimum number of sample cuts per lot required for assurance of acceptable precision was calculated.

---

1 See SHELEY, W.J. and ZIEGLER, W.A., "Generation of Quantitative Data while Sampling Ores and Concentrates", these Proceedings.
CONCURRENT DRYING TESTS

- --- PULVERIZED CONCENTRATE
- --- NOT PULVERIZED CONCENTRATE
- --- DEW POINT

FIG. 1. Dependence of weight loss on dew point of oven air when drying uranium concentrates at 110°C.
by the previously described formula except, in this case, \( t \) was at 99% confidence. Substituting actual data:

\[
N = \frac{S^2_t}{E^2} = \frac{(0.478)^2(2.552)^2}{(0.049)^2} = 620
\]

The first-stage falling stream cutter samples the feed stream 17.5 times/min and the rate of feed to the cutter is approximately 4 min/drum. Thus, the 3500 cuts taken from an average 50-drum lot is quite sufficient, but, should the number of drums in a lot fall below 10, the rate of feed can be decreased to obtain sufficient cuts.

The accuracy of the falling stream sampler was determined indirectly because there are no standard concentrate lots available. Using again the most heterogeneous concentrate available, the accuracy of the sampler was determined by comparing the results obtained by sampling the lots "as received" with the results of sampling the lots following pulverization. In each case, the sampling of the pulverized lot was designated as the standard.

Typical screen analyses of the concentrate before and after pulverizing are shown in Table IV.

**TABLE IV**

<table>
<thead>
<tr>
<th>Tyler sieve opening (mesh)</th>
<th>Per cent by weight Before</th>
<th>After</th>
</tr>
</thead>
<tbody>
<tr>
<td>+9</td>
<td>66</td>
<td>0</td>
</tr>
<tr>
<td>-9+42</td>
<td>27</td>
<td>23</td>
</tr>
<tr>
<td>-42</td>
<td>7</td>
<td>77</td>
</tr>
</tbody>
</table>

The average falling stream sampling difference between the "as received" lots and the lots after pulverizing was 0.04% \( \text{U}_3\text{O}_8 \) and this difference was not significant. Therefore, the falling stream sampler at Grand Junction was considered to be an accurate means of obtaining the primary sample of a lot of concentrate. Subsequently, many lots were sampled and then resampled in this unit, and the precision of falling stream sampling was found to be equal to, or better than, \( \pm 0.12\% \text{U}_3\text{O}_8\) at 95% confidence.

Once the accuracy of the falling stream sampler was established, it served as a standard for evaluating the accuracy of the alternate primary sampler, the enclosed auger. Lots were first auger sampled and then sampled by the falling stream unit. These comparisons revealed that some concentrates, particularly granular ones, were difficult to sample accurately by auger methods. Such concentrates are routinely sampled by the falling stream samplers. Table V illustrates the accuracy of enclosed auger sampling as related to particle size. The data in Table V show that concentrates A and B require falling stream sampling for acceptable accuracy.
TABLE V
ACCURACY OF ENCLOSED AUGER SAMPLING AS RELATED TO PARTICLE SIZE

<table>
<thead>
<tr>
<th>Concentrate</th>
<th>Per cent by weight of particles greater than 42 mesh in size</th>
<th>Average difference of auger less falling stream (% U₃O₈)</th>
<th>Significance level (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>93</td>
<td>±0.17</td>
<td>99</td>
</tr>
<tr>
<td>B</td>
<td>80</td>
<td>±0.10</td>
<td>98</td>
</tr>
<tr>
<td>C</td>
<td>53</td>
<td>±0.02</td>
<td>42</td>
</tr>
<tr>
<td>D</td>
<td>27</td>
<td>±0.02</td>
<td>67</td>
</tr>
</tbody>
</table>

The enclosed auger sampling precision was found to be in the same range as the falling stream sampling (i.e., equal to, or better than ±0.12\% U₃O₈ at 95\% confidence).

Generally, the weight of primary samples obtained by enclosed auger samplers, when drilling two holes per drum, is either 1.0 or 1.5\% of the lot, depending upon the size of auger. The falling stream samplers are adjusted to cut primary samples equal to 1.0\% of the weight of the lot.

**Secondary sampling**

Secondary sampling reduces the primary sample of 200-600 lb to six or seven 1500-g portions before any particle size reduction or blending. This is accomplished in an airtight sampler, as shown in Fig. 2. The primary sample container is positioned above the sampler, the valve is opened and the concentrate passes down a short chute to the vibratory feeder, and then flows off the feeder trough into the rejects drum below. The sample cutter consists of six or seven slots, or drawers, which pass back and forth through the falling stream of concentrate. The feed rate and the number of cuts are adjusted so that 1500 grams are collected in each drawer. The uniformity of the samples in each set is indicated by the precision of a single sample in terms of per cent U₃O₈ and per cent loss-on-drying (LOD) at 110°C. These precisions are ±0.12\% U₃O₈ and ±0.08\% LOD at 95\% confidence.

The accuracy of the secondary sampler was determined by comparing the results of sampling "as received" primary samples with the results of sampling the primary samples after thorough mechanical blending. The average difference between the two samplings was found to be less than 0.01\% U₃O₈ and not significant. The precision of a single routine secondary sampling was found to be ±0.09\% U₃O₈ at 95\% confidence.

**Sample preparation**

The sample preparation of concentrates includes the moisture determination or loss-on-drying at 110°C, pulverizing, blending, redrying of the pulverized sample, if necessary, and the packaging of the analytical samples.
The weight loss during drying is not necessarily all due to water, since ammonia and organic compounds sometimes evolve during drying.

The sample preparation method at present in use at Grand Junction is called the "Concurrent Drying Method". In this method, two of a set of replicate secondary samples are combined, pulverized to minus 80 mesh, and blended. The pulverized sample then is dried concurrently in the same oven at 110°C with another two of the unpulverized replicate secondary samples. These latter two samples are the moisture samples and they are dried until there is no additional weight loss. This loss-on-drying is the reported lot moisture. Immediately upon the conclusion of the moisture determination, the pulverized analytical samples are packaged and sealed in jars while hot. The advantage of this method is that both the analytical and moisture samples are exposed simultaneously to the same atmospheric conditions and, at the completion of the moisture determination, both samples are in the same state of dryness, as shown in Fig. 1.

During the development of the Concurrent Drying Method, the use of controlled atmosphere ovens was evaluated. It was found that equipment
to supply air of constant humidity was costly and conditions were somewhat difficult to maintain. Beyond that, such atmospheres were not needed for the Concurrent Drying Method.

As mentioned earlier, some concentrates are very hygroscopic and will gain appreciable weight, even in a drying oven at 110°C. Such concentrates are routinely prepared by the "Natural Moisture Method". In this method, no drying is performed and the pulverizing, blending and packaging of the analytical samples is done as rapidly as possible with minimum atmospheric exposure. At Weldon Spring, atmospheric exposure is minimized by routinely preparing samples of such concentrates within a glove bag, or a white bench, under controlled humidity conditions.

**Tertiary sampling**

Although tertiary sampling is not used in official purchase sampling, the sampler and its use is described for a better understanding of the important role it has played in the development of improved sample preparation methods.

The tertiary sampler, Fig. 3, is a small falling stream sampler of special design, and consists of a feeder to deliver a stream of concentrate to cutters located on the periphery of a rotating disk. The total periphery is composed of cutters through which the concentrate falls into sample containers, thereby generating no rejects. The sampler is totally enclosed to minimize the effect of atmospheric exposure.

Since the sampler generates no rejects, accuracy is assured within the precision of the sampler that was found to be, for the average of two samples, ±0.07% U₃O₈ at 95% confidence.

To ascertain the accuracy of the various sample preparation methods, a 1500-g secondary sample is fed to the tertiary sampler and samples of approximately 125, 62.5, and 42 g are obtained. Two samples from opposite sides of the sampler are totally dissolved and each is analysed for U₃O₈. The average of these analyses is the standard value to which the lot moisture (LOD) and U₃O₈ analysis of analytical sample are compared. If the analytical sample has been properly prepared, its U₃O₈ analysis, adjusted to the so-called "wet basis", or secondary sample basis, using the lot moisture, will agree with the average tertiary sample U₃O₈ analysis.

**Sample pulverization studies**

In developing the sample preparation methods currently in use, various pulverizers were investigated. Of the three general types studied — ball mills, impact pulverizers and disk pulverizers — the disk pulverizer was found to be the most suitable for the variety of concentrates received.

Nevertheless, using the tertiary sampler, it was found that during pulverization certain concentrates were upgraded in percent U₃O₈ through loss of volatiles. This occurred when the space or gap between the rotating and stationary plates was too small, as illustrated by the data in Table VI.

---

2 See SHELLEY, W.J. and ZIEGLER, W.A., "Generation of Quantitative Data while Sampling Ores and Concentrates", these Proceedings.
FIG. 3. Tertiary sampler for concentrates

TABLE VI

THE UPGRADING EFFECT OF PULVERIZATION ON CERTAIN CONCENTRATES

<table>
<thead>
<tr>
<th>Disk or plate gap (mm)</th>
<th>Equivalent Tyler sieve (mesh)</th>
<th>Per cent LOD</th>
<th>Per cent U₃O₈ dry weight basis</th>
</tr>
</thead>
<tbody>
<tr>
<td>Not pulverized</td>
<td>-</td>
<td>5.37</td>
<td>5.94</td>
</tr>
<tr>
<td>0.13</td>
<td>115</td>
<td>-</td>
<td>-</td>
</tr>
</tbody>
</table>

Further study showed that plate gaps, if carefully adjusted with a thickness gauge, are reproducible, and that pulverization to 80 mesh, instead of minus 115 mesh, virtually eliminated the possibility of upgrading. These
studies prompted a recent revision of the sample preparation method at Grand Junction. The practice now is to set the pulverizer plate gap with a gauge and eliminate the sieving of the entire sample. To ensure that the desired particle size is attained, a quick sieve test is run immediately after blending the pulverized sample. This revision reduces both the time required for pulverization and the atmospheric exposure of the concentrate during sample preparation. The small increase in particle size of the analytical sample had no adverse effect on the precision of the U$_3$O$_8$ determination in the laboratory.

**Concentrate chemical analysis**

Since sample preparation procedures are designed to ensure that the analytical sample is in the same "state of dryness" as the moisture sample at the termination of drying, analytical samples are analysed as received in the laboratory with no further drying. The total weight of the bottle and sample is recorded on the sample bottle label and, before analysis, the analyst confirms that there has been no significant change in weight because of seal failure. The U$_3$O$_8$ and impurity determinations are then performed.

The U$_3$O$_8$ analytical method is a macro wet chemical method [5]. Laboratory accuracy and precision are evaluated by the use of concentrate standards, high purity U$_3$O$_8$-black oxide standards, and inter-laboratory comparisons. The precision routinely maintained at Grand Junction is approximately ±0.05% U$_3$O$_8$ at 95% confidence for the reporting of the average of two analyses. Inter-laboratory analyses commonly agree within ±0.20% U$_3$O$_8$.

**CONCLUSION**

The AEC has been responsible for procuring large quantities of natural uranium in both ores and concentrates. For this reason, the uranium measurement methods (specifically, the weighing, sampling, and analysing) have been subject to continuing AEC scrutiny. Ore sampling presents no unusual problems if adequate and representative quantities of samples are taken, dust losses are kept at a minimum, and the samples are properly prepared and analysed.

Because of the varying physical and chemical characteristics of concentrates, many more sampling problems have been encountered. These problems have been solved through the development and use of specialized equipment and by improvements in sampling procedures. Falling stream sampling has proved the most accurate, provided concentrates are adequately protected to avoid significant gain or loss in weight. By ensuring that the analytical and moisture samples are in the same state of dryness, it has been possible to eliminate laboratory drying of concentrate samples and thereby improve the accuracy and precision of the U$_3$O$_8$ analyses.

It is important, in any sampling, to establish and maintain a programme to evaluate all measurement methods. Such programmes have been responsible for continuing improvement in the accuracy attained in sampling both ores and concentrates. Significant contributions to the improvement of
measurement methods have been made by Lucius Pitkin, Inc., and Mallinckrodt Chemical Works, the AEC contractors at Grand Junction and Weldon Spring respectively.

ACKNOWLEDGEMENTS

The authors wish to express their appreciation to Lucius Pitkin, Inc., for providing all unreferenced data used in this paper, and to members of the Grand Junction Office of the AEC for their many helpful suggestions.

REFERENCES


DISCUSSION

J. SORNEIN: The results given for the methods of sampling and analysing concentrates at Grand Junction are very impressive. These results, however, although they seem to indicate an excellent and very accurate sampling and analysis system (fidelity and reproducibility), may in fact be inaccurate because of a systematic error. If a customer of USAEC had another method giving results with an equally low degree of scatter but centred on average values which were systematically lower, what would be the attitude of USAEC?

F.E. McGINLEY: First, we would collect sufficient data to be sure that a bias does in fact exist, and then we would seek to determine the cause of the bias and take whatever steps were necessary to eliminate the bias. By careful investigation of each step in the measurement methods, and through co-operation between the parties involved, it should always be possible to eliminate biases between facilities.
GENERATION OF QUANTITATIVE DATA WHILE SAMPLING ORES AND CONCENTRATES

W.J. SHELLEY AND W.A. ZIEGLER
MALLINCKRODT CHEMICAL WORKS, ST. CHARLES, MISS., UNITED STATES OF AMERICA

Abstract — Résumé — Аннотация — Resumen

GENERATION OF QUANTITATIVE DATA WHILE SAMPLING ORES AND CONCENTRATES. The ultimate purpose of the sampling process is to establish quantitative data as to the content of a valuable or noxious material occurring in a gross quantity of material. In addition, subsidiary data used for additional purposes such as process control, specification adherence, or similar auxiliary and subsidiary purposes may be collected during the process.

The sampling process involves the weighing of the gross amounts of material; subsequent subdivision of such gross amounts by any one of a number of sampling methods such as augering, removing a sample from a moving stream, coning and quartering, or blending and thiefing. Ultimate dissolution of a quantity of material and the subsequent removal of a portion for further processing has been examined but found to be impractical in application.

With the removal of this representative sample portion of the gross material, this portion is then subjected to several steps including weighing, homogenization by grinding and blending, possibly drying, and is further subdivided to provide several representative aliquots for analysis. During each of these operations, the influence of the climatic conditions, equipment and operating personnel must be minimized so as to retain a portion for final analysis that is truly representative of the gross quantity. Analytical methods have generally been well established for materials containing known ranges of constituents important in uranium processing.

Practices in the United States of America during the twenty years of uranium processing have been partially described elsewhere. The methods employed have varied primarily with equipment, and economics of the situation, the urgency of installation, the ultimate precision desired. Problems which have existed with installations have been recognized in many ways and subsequently corrected.

Difficulties which have existed in the evolving practices, their methods of recognition, and current work to further improve the sampling process and secure appropriate accuracy and precision are discussed. Data relative to the current work is presented in a brief tabular fashion.

During the evolution of the sampling, handling, and analytical procedure in the United States, continuous attention has been fixed upon the reliability of the process. Since a wide variety of materials require sampling, constant evaluation through development of reference sampling techniques, duplicate sampling, assay sample exchange and statistical examination of accuracy and precision is maintained.

OBTENTION DE DONNEES QUANTITATIVES LORS DE L'ECHANTILLONNAGE DE MINERAIS ET DE CONCENTRES. Le but ultime de l'échantillonnage consiste à obtenir des données quantitatives en ce qui concerne la teneur d'une matière précieuse ou nuisible présente dans une substance brute. En outre, on peut réunir, au cours de l'opération, des données subsidiaires servant à des fins complémentaires, telles que le contrôle de l'opération, la stricte observation des spécifications ou d'autres fins analogues.

L'opération d'échantillonnage nécessite le pesage de la matière brute, ainsi que la subdivision ultérieure de cette matière selon l'une quelconque des méthodes d'échantillonnage suivantes: carottage, prélèvement d'échantillons d'un courant de matières en mouvement, concassage ou mélange et prélèvement à la pipette. On a examiné la possibilité de procéder à la dissolution d'une quantité de matière, suivie du prélèvement d'une fraction aux fins de traitement ultérieur, mais cette méthode s'est révélée peu pratique.

Après avoir prélevé l'échantillon de matière brute, on le soumet à plusieurs opérations successives - pesage, homogénéisation par broyage et mélange et éventuellement séchage - après quoi on le subdivise en plusieurs parties aliquotes aux fins d'analyse. Au cours de chacune de ces opérations, il convient de réduire au minimum les effets attribuables aux conditions climatiques, au matériel et aux opérateurs de manière à disposer pour l'analyse finale d'un échantillon strictement représentatif de la matière brute. D'une manière géné-
ПОЛУЧЕНИЕ КОЛИЧЕСТВЕННЫХ ДАННЫХ ПРИ ВЫБОРКЕ ПРОБ ИЗ РУД И КОНЦЕНТРАТОВ. Конечной целью выборки проб является получение количественных данных относительно содержания ценного или бесполезного материала в его общем количестве. По- мимо этого, в ходе выборки проб можно собирать также вспомогательные данные, нужные для дополнительных целей, как то: регулирование технологических процессов, соблюдение спецификаций или для аналогичных вспомогательных или подсобных надобностей.

Выборка проб сопряжена со взвешиванием общих количеств материала с последующим разделением этих общих количеств на основании одного из многих методов выборки проб, как, например, выборка при помощи спирального бура, взятия проб из движущегося потока, квартования, конусования, смешивания или, наконец, взятия жидких проб. Была рассмотрена возможность растворения в конце процесса некоторого количества материала с последующим отбором части раствора для дальнейшей переработки, но такой метод оказался практически непригодным.

После отбора представительной пробной части общего количества материала эта часть подвергается ряду операций, включая взвешивание, гомогенизацию посредством измельчения или смешивания, иногда сушку, а затем дальнейшее дробление на части для получения нескольких представительных аналитических проб для анализа. На протяжении всех этих операций следует сводить до минимума влияние условий погоды, оборудования и обслуживающего персонала, с тем чтобы оставленная для конечного анализа часть действительно хорошо отражала настоящий состав общего количества. В отношении материалов, для которых известны пределы составных частей, представляющих большое значение для обработки урана, аналитические методы обычно уже хорошо отработаны.

Проводимая в Соединенных Штатах на протяжении двадцати лет практика обработки урана была частично описана в других публикациях. Использованные методы менялись, главным образом, в зависимости от рода оборудования, экономических соображений, срочности создания установки и желательной в конечном результате точности. Возникающие на установках затруднения вызывались теми или иными способами и затем устраивались.

В докладе обсуждаются затруднения, возникшие в выработке установившейся практики, методы их изложения и текущая работа по дальнейшему улучшению метода выборки проб и обеспечения нужной достоверности и точности. Данные, касающиеся текущей работы, представлены в сжатой форме в виде таблиц.

При усовершенствовании методов выборки проб, практики обращения с материалами и методов проведения анализов непрерывное внимание уделялось в Соединенных Штатах надежности технологических процессов. Ввиду того, что выборка проб необходима для самых разнообразных материалов, непрерывно проводится оценка посредством выработки методов приготовления калиброванных проб, двойной выборки проб, обмена пробами для анализа и статистического изучения достоверности и точности.

ОБТОЧЕНИЕ ДАТOS QUANTITATIVES DURANTE EL MUESTREO DE MINERALES Y CONCENTRADOS. El principal objetivo de muestreo consiste en obtener datos cuantitativos sobre las sustancias valiosas o nocivas contenidas en una cantidad bruta de material. Permite también reunir datos auxiliares para el control de los procesos, la observancia de las especificaciones y otros fines similares de índole subsidiaria.

Las operaciones comprenden la determinación del peso de las cantidades brutas del material y la subdivisión de esas cantidades por un método cualquiera de muestreo: taladrado, extracción de muestras en corriente
continua, división en conos y cuarteo, mezcla y muestreo en tanques. Se ha estudiado la posibilidad de recurrir a la disolución de una cantidad determinada de material y a la separación de una parte del material disuelto a fin de someterlo a un tratamiento ulterior, pero este procedimiento no ha dado buenos resultados.

Una vez obtenidas, las muestras representativas del material bruto son objeto de varias operaciones –pesada, homogeneización por trituración y mezcla, posiblemente secado– y luego se las subdivide a fin de obtener varias partes alícuotas para el análisis. Durante cada una de esas operaciones es preciso reducir al mínimo la influencia de las condiciones climáticas, del equipo y del coeficiente individual de las personas que lo emplean, de manera que para el análisis final se disponga de una porción verdaderamente representativa del material bruto. En general, se han establecido ya excelentes métodos de análisis para los materiales que contienen, en un orden de magnitud conocido, constituyentes importantes para el tratamiento del uranio.

Los métodos empleados durante los veinte últimos años en los Estados Unidos para el tratamiento de esta sustancia han sido ya parcialmente descritos. Esos métodos han variado según el equipo, las condiciones económicas, la urgencia de los trabajos y las exigencias en cuanto a la precisión de los resultados. Los problemas surgidos en las instalaciones han sido debidamente examinados y resueltos.

En la memoria se analizan las dificultades con que se tropezó al establecer las prácticas actuales, los métodos empleados para detectar esas dificultades y los trabajos realizados para perfeccionar las operaciones de muestreo y dotarlas de exactitud y precisión adecuadas. Se presentan sucintamente algunos datos relativos a esos trabajos.

Durante la progresiva elaboración de los procedimientos de muestreo, manipulación y análisis se ha prestado continua atención en los Estados Unidos a la seguridad de las operaciones. Dada la gran variedad de los materiales que requieren muestreo, se procede a una evaluación constante de los resultados mediante el empleo de técnicas de muestreo de referencia, muestreo duplicado, intercambio de muestras de ensayo y análisis estadístico de la precisión.

1. INTRODUCTION

Sampling of ores and particularly of the more valuable concentrates is performed to secure from a gross quantity of material a small, uniform representative portion that can be accurately and precisely assayed to provide a basis for payment and determine adherence to specifications. Other needs for a sample may also exist: determination of amenability to further physical beneficiation or chemical processing, establishment of material accountancy, and process control.

Since the accuracy of all subsequent determinations is dependent upon the removal of a representative sample, a sampling procedure must be devised for each material. The characteristics of the material must be known. Although similarities may exist, individual ores and concentrates will be different. All of the approximately 40 uranium concentrates processed by the Uranium Division have exhibited variation from lot to lot, from drum to drum, and often along any random straight path drawn through a drum. The particle size distribution is an important variable. Ores normally contain much larger particles than do concentrates. The moisture sensitivity of the material is an extremely important characteristic which is often overlooked.

2. SUMMARY OF SAMPLING PROCESS

Weighing of the gross lot of material or component units is the first processing operation. Ores, which are normally handled in bulk, are weighed in trucks or rail cars. Because of their smaller volume, greater value, and attendant dusting hazard, concentrates are transported and
weighed in drums. The next operation is the removal of a gross sample, which is then reduced in volume and particle size by grinding, blending, and splitting. Frequently, but not necessarily, samples will be dried or even fired at a high temperature. The final product is a group of representative aliquots, contained in sealed bottles or other containers, suitable for processing by analytical laboratories for the determinations of uranium and other constituents of interest.

3. WEIGHING

A typical lot of uranium concentrate has been described at this meeting by McGinley et al. [4]. The first major step at Weldon Spring is the weighing of the cleaned and dried drums on a print-weigh scale. This scale provides a visual reading and prints both drum numbers and weights to the nearest half-pound on a card and a paper tape.

Since weights are of primary importance, each drum weight is determined twice. At the Weldon Spring sampling plant, the second weighing is performed by an electronic strain gauge mounted on the platform of the print-weigh scale. The gauge measurement is read on a dial to the nearest pound. During routine operation, the expected drum weight from the supplier's tally sheet is set on the strain gauge dial. If the permissible difference between this setting and the measured weight is exceeded, an alarm is sounded and a red light flashes; otherwise, a green light indicates satisfactory agreement.

Additional support of the weight data is obtained by scale calibration. Scales are calibrated at zero and at the average drum weight of the lot before, after, and often during the weighing of the lot to ensure continued scale accuracy. Control charts of these check weighings provide a history of scale performance. Additional zero weight checks are made after every tenth drum throughout the weighing of a lot.

Ideally, in a sampling operation three events should occur simultaneously: weighing of the lot, withdrawal of the sample, and weighing of the sample. A time lapse between these operations may allow changes, such as water sorption, to occur between the two weighings, causing the sample to be non-representative. Since the ideal case is impossible, it is essential to work rapidly and to protect the material from the influence of the atmosphere as much as possible.

4. PRIMARY SAMPLING

There are several hand methods [1] of sampling which are used for bulk materials, such as ores. Grab sampling consists of taking small, equal portions by scoop or shovel at random or regular intervals from the mass of material to be sampled. Coning and quartering consists of piling the ore into a conical heap, dividing it radially into four quarters, taking opposite quarters as the sample and rejecting the other two. Shovel sampling is performed, usually during loading or unloading bulk materials, by selecting every nth shovelful to make up the sample. All hand sampling is highly sub-
FIG. 1. Hand-held open ship auger with two types of catch pans for collecting the sample

ject to technique and tends to oversample the fine material while rejecting the coarse material.

Dissolution of the entire lot with subsequent dip sampling of the presumably homogeneous solution is often suggested as a most accurate means of sampling. While the principal is good, practical difficulties usually prevent success. If not practical for routine use, dissolution of a large batch of material is often advocated for the occasional testing or checking of other sampling methods or techniques. Unfortunately, in our experience it has never succeeded. Tanks can seldom be calibrated as accurately as volumetric glassware. Plant conditions are not as rigorous as laboratory conditions; consequently, complete dissolution is difficult to achieve. Temperature control and resultant thermal expansion corrections are less reliable; spillages are more common on a plant scale; and finally plant operators do not normally have the quantitative skills of the laboratory analyst.

Augering is probably the most economical method of sampling if the material, such as concentrates, is packaged in drums which are to be transshipped without dumping. Although there are many designs, augers fall into
the two general categories of open and enclosed augers. In the United States of America open augers, as pictured in Fig. 1, have typically been ship augers of about 1 3/16 in. diameter powered by a hand-operated 3/4-in. drill. The augering is performed through a hole in a catch pan which collects the sample brought to the top. Contents of the pan are then dumped into a sample container. Open augering may not give good vertical representation of the drum, but will preferentially remove the material at the top of the drum at the expense of the lower layers [7]. Since drums of material are frequently segregated vertically, a biased sample may result.

Enclosed augers may either be the ship-auger type or have a central shaft with one or more flights as shown in Fig. 2. In either case it will be surrounded by a cylindrical sheath which does not rotate. Material removed in drilling may be discharged through a side hose at the top or it may be stored in the sheath for discharge by reversing the auger after withdrawal from the drum.
FIG. 3. Vezin and Geary-Jennings samplers [3]

FIG. 4. Falling-stream sampling system installed at Weldon Spring using two stages of Geary-Jennings samplers
In our experience, the enclosed auger gives a better vertical representation than the open auger, but neither give a good sample of the bottom two inches where the heaviest concentration of fines or foreign matter may occur. All augering, even though multiple holes are drilled, is subject to error if radial segregation is present in a drum. Radial segregation can readily occur since drums are normally filled from the middle, causing the coarser and denser particles to flow to the outside. Concentration differences of as much as 2% $\text{U}_3\text{O}_8$ have been found from centre to exterior circumference. Therefore, when an augering method is used, the material sampled must constantly be re-evaluated to ensure its continued amenability to auger sampling.

The most accurate method of primary sampling employs a falling-stream system where a moving cutter removes all of the falling stream part of the time. Such cutters fall into the two general categories of arc-path or straight-path samplers. Slot widths of the cutter should be at least three times the diameter of the largest particles to be sampled, four times or more is preferable. Obviously, the speed of travel through the stream is one control of the sample size collected, but the speed of the cutter should not be so great as to knock particles away.

The most popular and probably the best performer of the arc-path samplers for concentrates is the Vezin type sampler shown in the left half of Fig. 3. Ore or concentrate falls from a belt or vibrating feeder or is fed through a chute as a vertically falling stream which is cut by the rotating radially oriented slots of the cutter. Such a device will have one to usually not more than four slots. Material collected by the slots falls into the sample chute while the bulk of the material falls by into the reject stream. Mechanically, the Vezin sampler has the advantage of simple rotary motion, but it will not cut equal percentages from all parts of a stream if the slot sides are not perfect radii.

Of the straight-path samplers most variations exist only in the drive mechanisms; one of the most frequently encountered is the Geary-Jennings type shown in the right half of Fig. 3. Again the ore or concentrate falls from a moving belt or other feeder in a vertical stream through which passes a rectangular slot. Sample collected is usually diverted through an angled chute into a sample receptacle, and the gross reject material falls directly downward. The amount of sample collected is controlled by feed rate, slot width, and cutter speed and frequency. This sampler cuts every part of any shaped stream proportionately and is potentially the most accurate type of any we know.

A falling-stream system using two stages of Geary-Jennings sampling in use at the Weldon Spring plant is shown in Fig. 4. Drums are elevated to the top level where the lid is removed and replaced by a dumping cone, which is an integral part of the dumping mechanism. After the "apex" of the inverted cone is sealed to the dumping port in the top hopper, an iris valve in the cone is opened to allow the contents to flow into the stirred hopper. Material discharges from the hopper through a rotary valve (not shown) and vibratory feeder to the housing of the first Geary-Jennings sampler. The first-stage sample is fed through a vibratory feeder to the second sampler. The second-stage sample is collected in a drum, and the reject from both
stages is collected in the lower hopper from which drums are filled. This system is built as tightly as possible, and air does not flow in or out during normal operation.

Falling-stream sampling will probably be more economical operationally than augering if it is incorporated into the packaging facility at a production installation or the dumping operation at a consuming site. For materials handled by bulk methods, it is the only practical method for acceptable accuracy and precision for all but the cheapest of materials.

5. SAMPLE PREPARATION

The goal of the sampling process is to produce a small amount, as little as 0.5 g, of material having the same chemical composition as the whole or original quantity. For the average uranium concentrate lot this is a volume reduction of 25 million to 1!

In order that proportionality and accurate representation both be maintained, certain criteria are necessary for estimating the size of sample which may be able to replicate the whole quantity within specified limits [2]. An estimate which can satisfy practical requirements may be obtained by starting with an idealized example and then finding the conditions under which its solution will apply to actual conditions.

For any single sample withdrawal step from a larger body of material assumed to contain two basic kinds of particles A and B, it can be shown by application of statistics that the minimum sample weight w which will have the desired precision is

\[
w = v p (1 - p) \left( \frac{d_1 d_2}{d^3} \right) \left( \frac{P_1 - P_2}{P} \right) \left( \frac{1}{\sigma_s^2} \right)^2
\]

where
- \( v \) = average volume of the unit particle
- \( p \) = fraction of particles A relatively rich in uranium
- \( 1-p \) = fraction of particles B relatively poor in uranium
- \( d \) = average particle density
- \( d_1 \) = density of particles A
- \( d_2 \) = density of particles B
- \( P \) = average uranium assay
- \( P_1 \) = uranium assay of particles A
- \( P_2 \) = uranium assay of particles B
- \( \sigma_s \) = relative standard deviation of P due to sampling.

Of these variables the supplier of the ore or concentrate sets all except the relative standard deviation which is determined by the precision the sampler requires.

Since an actual sampling case is much more complex than the two-component case, the practical application of this formula involves some mathematical idealization of an actual concentrate. A sieve analysis is performed on a fairly large sample. The average particle volume can then be estimated from the weighted average of the sieve sizes. The density and
the assay of every constituent of interest, normally uranium and loss on
drying, are determined on each sieve fraction. Similar sieve fractions are
then grouped together to give two general types of particles with high and
low assays respectively. Various combinations can be tried to ensure that
the most rigorous case is included. The standard deviation may be chosen
strictly by judgment, or it may be related to the precision of the uranium
assay, since the latter is the most precise determination performed on the
material. It may be desirable to decide that the combined precision of the
sampling plus assaying should not be significantly greater (at the 95% con-
fidence level) than the precision of the assay alone. Since the total variance
is equal to the sum of the individual variances, the allowable sampling stan-
dard deviation is readily calculable. A more rigorous treatment of this
calculation is given by Nelson et al. [6].

From the pragmatic viewpoint it is evident that the minimum sample
size depends on:

1. Particle size distribution
2. Maximum particle size
3. Density differences among particles
4. Assay differences among particles
5. Distribution of assay differences

In sampling work only items (1) and (2) can be changed intentionally
as desired to fulfil the requirements of (6). Any change caused by the sam-
pling process to items (3), (4), and (5) would probably introduce error.

With the minimum sample size considerations in mind, the subsequent
steps to primary sampling can be determined. By either augering or falling-
stream sampling, a 0.75-1.00% sample, approximately 130 kg, is removed.
Previous minimum sample size determinations have shown some of the most
heterogeneous concentrates to have minimum sizes of as little as 400 g.
Therefore, further subdivision is appropriate, and the primary sample is
passed through a secondary sample splitter, such as that described by
McGinley [4], where each of six cutter slots and drawers collects 1.5 kg,
well above the minimum sample size.

Further sample preparation is outlined in Fig. 5, which illustrates dried-
sample preparation. Each of the first four drawerfuls from the secondary
splitter is weighed into a stainless steel pan and then dried in a forced-draught
oven at 110°C to constant weight.

The concentrate as purchased must meet a maximum particle size speci-
fication of 1/4 in., but it is too coarse for the analyst. Therefore, it is
ground in a pulverizer to about 60 mesh, thus reducing the maximum par-
ticle size as well as compressing the particle size distribution toward the
finer material. A further volume reduction can be made, but first blending
is necessary to homogenize the material as much as possible to enable an
accurate subdivision. Successive splits are made, on a Jones riffle at Wel-
don Spring, to reduce the 6 kg to about 2.2 kg. It has been determined that
concentrate fine enough to pass US standard No. 100 sieve has a sufficiently
small minimum sample size for the analyst. Further grinding is necessary
for most materials. Extra effort and possible excessive overheating of the sample are avoided by first screening the material and then grinding the relatively small amount of oversize by hand, further compressing the particle size distribution and reducing the maximum particle size. Since further subdivision will be necessary, the sample is next blended and then reheated in a pan in an oven for 48 h at 110°C to return it to the previous condition. Samples are then scooped from the hot pan as rapidly as possible and are placed in the final sample bottles, approximately 150 g per bottle.

Most concentrates readily absorb water from or yield it to the atmosphere. Many, including the dried samples, are good desiccants. Therefore, the nature of the bottles and the sealing operation are critical for proper preservation of the sample. Considerable work has been done to perfect and prove the present practice [5]. Best results at Weldon Spring have been obtained with Kerr home-canning bottles having two-piece caps. The circular lid is lined around the perimeter with a mastic material which forms the seal with the top of the glass jar. All samples to be stored for as long as overnight are vacuum-sealed. The mastic sealant is softened by heating of the lid on a small modified hot plate. Then, in a vacuum desiccator, the bottle is evacuated to about 5-10 cm of mercury. When full atmospheric pressure is suddenly admitted to the desiccator, the mastic is driven strongly
against the glass top making a tight seal, the integrity of which can always be easily checked by tapping on the lid. Various treatments and prolonged storage have shown that a 150-g sample will not pick up more than 0.014% of its weight in a year’s time. These bottles are then placed in a polyethylene bag which is sealed tightly at the top and imprinted with a Mallinckrodt seal. The final package is illustrated in Fig. 6. Any break in the plastic bag or absence of the seal imprint would then indicate tampering.

One of the greatest and most difficult problems in sample handling and preparation is the exchange of moisture with the atmosphere. In the dried-sample preparation described above, it is assumed that the wet weight of concentrate before drying is the same as that of the very same material when it was in the drums. In the essential preparative steps of grinding, screening, and splitting, however, the moisture content would unquestionably be changed. Therefore, an effort is made to dry the concentrate by a measured amount to a point to which it can be returned by redrying just before bottling.

A number of concentrates, however, will normally gain weight in an oven and demonstrate the fact that drying is dependent not only on temperature but also on humidity of the ambient atmosphere. These concentrates must be prepared by the natural moisture method previously described [4]. Exposure during sample processing may cause some moisture pick-up, of course, but it is minimized by shortcutting some of the steps, and the resultant error is not nearly as great as that from the prolonged exposure in the oven.

It is common practice in the laboratory drying of most chemicals to heat them above the boiling point of water for a period of time or to constant
weight. Since the boiling point is exceeded at the commonly used temperature of 110°C, it is assumed that all the water will evaporate. Such assumption is applicable only to adsorbed water and hydrates decomposable at that temperature. There is nothing significant about the boiling point temperature for the chemically bound or absorbed water in ores and concentrates. This is illustrated by the data of Table 1. Up to the temperature where all chemically bound water is removed, almost any loss-on-drying value may be secured at a given humidity by selecting the heating temperature.

As illustrated in another paper to this Symposium [4], there is a definite humidity effect upon the amount of drying. At a given temperature, every water-containing solid has a specific water-vapour pressure which, of course, tends to rise with increasing temperature. If the atmosphere above the solid possesses a lower vapour pressure, the solid will lose water. If the atmosphere has a higher vapour pressure, water will be absorbed by the solid. The atmospheric humidity which represents a vapour pressure equal to that above the solid, thus resulting in no net water exchange, is defined as the equilibrium humidity of that material. Adjustment of the atmospheric humidity will result in a different loss-on-drying within a reasonably limited range.

It is convenient to measure atmospheric humidity as the dew point in degrees Fahrenheit. If the equilibrium dew point of a concentrate sample at room temperature is lower than the dew point of the ambient atmosphere, the sample will gain weight by sorption of moisture. However, if the sample is placed in an oven, its temperature will rise, possibly causing it to emit water vapour and lower its equilibrium dew point value. But if the oven is supplied with ambient air, the atmospheric dew point inside the oven is the same as that outside! Heating the air does not withdraw water; it merely increases the capacity of the air to contain water. Therefore, samples dry only because they are heated, and they can dry only so far as to obtain an equilibrium dew point equal to the dew point of the ambient atmosphere.

One of the last steps in the production of concentrates at the mill is the drying of a polyuranate precipitate at temperatures of 140°C or more. Those heated above 400°C may become dry enough that their equilibrium dew points, even at 110°C, are so low that they pick up water vapour from the atmosphere, and samples of these materials cannot be oven-dried but must be processed by the natural moisture method. Many of these concentrates are better desiccants than anhydrous calcium chloride, which has an equilibrium dew point of -43°F, and will absorb atmospheric water faster than magnesium perchlorate.

The rate of sorption is also an important variable since the equilibrium dew point does not necessarily indicate how fast weight gains may occur. The most important influence upon the rate of sorption is the degree of exposure. Fortunately, thick static beds absorb relatively slowly. For example, 100 g of one of the more sensitive concentrates was left open in the room on a balance pan in a bed averaging about one centimetre thick. It gained 8% of its weight in eight hours. Had it been poured through the air so that each particle were thoroughly exposed, an equivalent gain could have been accomplished in much less than an hour. Other experiments have shown
TABLE I

COMPARISON OF DRYING CONDITIONS

<table>
<thead>
<tr>
<th>Concentrate Lot</th>
<th>Ambient Air 110°C., 5 days</th>
<th>Dried Air at 110°C. 8 days</th>
<th>Dried Air at 110°C. 15 days</th>
<th>Ambient Air 160°C., 3 days</th>
</tr>
</thead>
<tbody>
<tr>
<td>AA-1</td>
<td>0.44</td>
<td>1.44</td>
<td>1.59^a</td>
<td>2.31</td>
</tr>
<tr>
<td>AC-1</td>
<td>2.35</td>
<td>3.16^b</td>
<td>3.24^b</td>
<td>3.25</td>
</tr>
<tr>
<td>U-1</td>
<td>0.00</td>
<td>0.14</td>
<td>0.14</td>
<td>0.16</td>
</tr>
<tr>
<td>AA-2</td>
<td>6.92</td>
<td>7.22^c</td>
<td>--</td>
<td>8.16</td>
</tr>
</tbody>
</table>

^aLost about 0.02% per day after 8 days.

^bLost about 0.01% per day after 8 days. (Accepted as constant weight at 8 days.)

^cConstant weight obtained in 4 days.

Weight changes of as much as 1.60% in five minutes to occur in 20-g samples at 70°F with air flows through them of 6 litres/min at humidity levels commonly encountered.

An obvious way to prevent samples from changing weight is to expose them to atmosphere containing their equilibrium humidities. Even humidities close to the equilibrium value are an improvement since the extent of water exchange is limited, and the rate of exchange is lowered by the reduced humidity differential. This approach is currently being made with the White Bench shown in Fig. 7. This device is a laboratory work table completely enclosed between the end panels by a polyvinyl chloride sheet twice as long as the table. The flexibility of the sheet allows the operator, using the one set of armholes and viewing panel, to have complete access to the entire length and breadth of the table, thus making it far more versatile than a conventional dry box. The White Bench has been equipped to receive air from a 100-lb pressure airline, the dew point of which is lowered to -100°F by a Gilbarco dryer, or the off-gas from a liquid nitrogen storage tank with a dew point of about -75°F. Room air can also be introduced. The White Bench itself has a recirculation system in which containers of magnesium perchlorate are placed to aid in drying the atmosphere. This extra pre-
caution is necessary because diffusion of atmospheric water vapour through the 100 ft$^2$ of plastic prevents a dew point differential from the room air of greater than 80°F. A search is being made for a less permeable but equally flexible and tough plastic film.

Recent developments have enabled us to determine the equilibrium dew point of a concentrate by measuring the dew point of the equilibrated air above it in a partially filled bottle. The range of dew points extends from room temperature to -100°F. With a less permeable enclosure on the White Bench we will then be able to determine the dew point of a fresh sample from the secondary splitter, adjust the White Bench atmosphere to that value, and expose the sample to air of its equilibrium humidity during the critical period of sample preparation.

6. ANALYTICAL TESTING

The uranium assay of concentrates demands great attention when it is realized that at the current United States domestic price for uranium an error of 0.01% $\text{U}_3\text{O}_8$ on a typical 37,000-lb lot is equivalent to $\$29$. The assay method developed and used in our laboratory is unlike that used in most other locations. A 0.5-0.6-g sample is dissolved in mixed acids with tin and chromium being volatilized as the chlorides and silicon as the fluoride. In the presence of EDTA to complex impurities, uranyl ammonium phosphate is precipitated by addition of ammonia with careful pH control. The precipitate is filtered and dissolved in sulphuric and hydrochloric acids. The solution is then passed through a lead reductor and titrated with cerium
(IV) in the presence of iron(III) to an iron(II)-1, 10-phenanthroline end point. Each sample has one determination performed by each of two analysts. If there is agreement within the expected tolerance, the average of the two results is reported. Currently, this analysis has a limit of error at the 95% confidence level of ± 0.084% U₃O₈.

Careful control must be maintained on the assay work. In addition to the usual titrant standardizations against standard U₃O₈, it is helpful to have a standard concentrate sample which is assayed periodically to detect bias which may develop. A General Analytical Evaluation programme is currently participated in by four co-operating laboratories. Every three weeks a set of samples from three concentrate suppliers is distributed for assay. For each of the three types of concentrate the sample submitted may be a current production sample or a reference sample. The samples are disguised so that it is never known whether the current period's samples are production or reference. From the determinations on the reference samples, movements in assay level, if any, of the participants can be followed, and their precisions over a long time period can be calculated. From all the data the relative levels of the participants is determined, and the relative problems with the different types of concentrate can be determined.

In addition, for both uranium assays and the various impurity determinations the Weldon Spring laboratory maintains a concentrate recycle programme whereby certain previously tested lot samples are selected periodically in a random manner and re-analysed under a disguised label. The pairs of results are then used for precision determination which includes the variables of time, different analysts, different standard solutions, glassware, etc.

The evaluation programme just described is performed to provide the kind of assurance necessary to validate the quality of measurements made on materials of high value.

Although not as critical from a monetary standpoint as the assay, impurity determinations can achieve economic importance when specification limits are exceeded, because of the financial penalties attached. Typical specification impurities and methods of testing are given in Table II.

7. EVALUATION

Again, because of the great value of uranium materials constant evaluation of sampling methods is required. Primarily, one is concerned about bias. Repeated commission of only a small error on each lot can result in a total error of several thousands of dollars. Errors leading only to lower precision are not quite so serious since they are not cumulative, and over a long period of time the net error should remain near zero. However, an error of either kind could deal a serious economic blow to a small supplier making infrequent shipments.

As previously mentioned, auger sampling requires constant vigilance to see that a material's sampling characteristics do not change. The augering accuracy is determined at Weldon Spring by comparison with results from sampling the same lots on the falling-stream system, which is used as a reference. Comparisons are made of the loss on drying, both wet- and
**TABLE II**

**TYPICAL CONCENTRATE SPECIFICATION IMPURITIES**

<table>
<thead>
<tr>
<th>Impurity</th>
<th>Typical Concentration Range&lt;sup&gt;a&lt;/sup&gt;</th>
<th>Method of Determination</th>
</tr>
</thead>
<tbody>
<tr>
<td>Arsenic</td>
<td>&lt; 0.10 -- 0.80%</td>
<td>Spectrographic</td>
</tr>
<tr>
<td>Boron</td>
<td>&lt; 0.005 -- 0.011%</td>
<td>Spectrographic</td>
</tr>
<tr>
<td>Calcium</td>
<td>0.01 -- 0.71%</td>
<td>X-ray fluorescence</td>
</tr>
<tr>
<td>Carbonate</td>
<td>&lt; 0.01 -- 2.05%</td>
<td>CO₂ evolution-gravimetric</td>
</tr>
<tr>
<td>Copper</td>
<td>&lt; 0.01 -- 0.08%</td>
<td>Spectrographic</td>
</tr>
<tr>
<td>Dysprosium</td>
<td>&lt; 0.001 -- 0.005%</td>
<td>Spectrographic</td>
</tr>
<tr>
<td>Europium</td>
<td>&lt; 0.001 -- 0.001%</td>
<td>Spectrographic</td>
</tr>
<tr>
<td>Fluoride</td>
<td>&lt; 0.01 -- 0.16%</td>
<td>Spectrophotometric</td>
</tr>
<tr>
<td>Total other halides (as Cl⁻)</td>
<td>0.000 -- 0.35%</td>
<td>Titrimetric-Volhard</td>
</tr>
<tr>
<td>Gadolinium</td>
<td>&lt; 0.001 -- 0.005%</td>
<td>Spectrographic</td>
</tr>
<tr>
<td>Insoluble U₃O₈</td>
<td>&lt; 0.01 -- 0.49%</td>
<td>X-ray fluorescence</td>
</tr>
<tr>
<td>Molybdenum</td>
<td>&lt; 0.01 -- 0.45%</td>
<td>Spectrographic</td>
</tr>
<tr>
<td>Phosphate</td>
<td>&lt; 0.05 -- 4.40%</td>
<td>X-ray fluorescence</td>
</tr>
<tr>
<td>Samarium</td>
<td>&lt; 0.001 -- 0.005%</td>
<td>Spectrographic</td>
</tr>
<tr>
<td>Sulfate</td>
<td>0.09 -- 12.07%</td>
<td>X-ray fluorescence</td>
</tr>
<tr>
<td>Thorium</td>
<td>&lt; 0.01 -- 3.12%</td>
<td>X-ray fluorescence</td>
</tr>
<tr>
<td>Vanadium</td>
<td>&lt; 0.01 -- 1.15%</td>
<td>X-ray fluorescence,</td>
</tr>
<tr>
<td>Zirconium</td>
<td>&lt; 0.30 --</td>
<td>Spectrographic</td>
</tr>
<tr>
<td>Total extractable organic</td>
<td>&lt; 0.01 -- 0.26%</td>
<td>Gravimetric</td>
</tr>
</tbody>
</table>

<sup>a</sup>Dried sample basis

Dry-basis U₃O₈ assays, and total pounds of U₃O₈ found by each sampling. Weighing variation is included in only the latter comparison and contributes very little to the total error. Whenever a significant difference develops between methods in the wet-basis U₃O₈ comparison for no known correctable reason, consideration is given to switching the routine sampling method to falling stream.

Duplicate auger sampling is also performed to obtain precision data. Current relative precision values for the pounds of U₃O₈ found in the lot range for different types of concentrate from ± 0.073 to ± 0.158%, where the assay method itself is the biggest error contributor.
The checking of falling-stream sampling is more of a problem since there is no superior ultimate reference method. However, such a system must be thoroughly tested and proved after construction and before use. Test programmes on two falling-stream systems installed at Weldon Spring have been reported [8, 9, 10]. Continuing evaluation is made by duplicate sampling, comparing one falling-stream system against the other, and once a year comparing a lot with a sample from the secondary splitter. The precision of the Weldon Spring falling-stream sampling is currently ± 0.115% U₃O₈, wet basis. The most recent comparison on the sampling of one whole lot by both the primary and secondary samplers gave exact agreement on the loss-on-drying results and an absolute difference of only 0.03% U₃O₈, wet basis.

It is also desirable to check the validity of the sample preparation. This may be done by use of the tertiary sampler as described by McGinley et al. [4]. Alternatively, this check may be made by a procedure that has been named the absolute moisture method. A sample large enough, or aliquots which may later be summed, to exceed the minimum sample size is cut from the contents of one drawer of the secondary splitter and heated to as high a temperature as necessary, even to 1000°C, to evolve all volatile components and attain a steady weight. Such a plateau of steady weight can be followed with a thermobalance or by monitoring the off-gases with a mass spectrometer. Then the loss in weight is calculated. Upon completion of the regular sample preparation a portion of the prepared sample is similarly heated to determine a second loss in weight. Any net difference remaining between the two losses in weight, after correction for the loss on drying of the regular preparation, is a measure of the error introduced by sample preparation.

Inter-site sampling comparisons have also been made. Because of the large number of details that must be watched and the extensive planning required, such a programme becomes quite expensive but is valuable for exhibiting unsuspected differences due to equipment, standard operating procedures, climate, etc.

In any phase of the sampling activity, maintenance of consistently acceptable accuracy and precision requires constant and repetitive confirmation subjected to the rigours of statistical analysis.

REFERENCES

DISCUSSION

W.R. SHIELDS: I should like to mention that in the past few years it has been established that natural uranium has isotopic variation (±0.1% for $^{235}$U and ± an order of magnitude for $^{234}$U). The variation of $^{235}$U is too small for economic considerations but the variation of the $^{234}$U is large enough to affect the operation of a gaseous diffusion plant. Is this not reflected in the price and specifications of natural uranium?

W.J. SHELLEY: I do not know of any case where the $^{234}$U content is specified or taken into account in fixing the price.

D.E. GEORGE: That is so, at least as far as the United States is concerned - though I understand from Mr. McCluen that the isotopic concentration of UF$_6$ was verified prior to introduction into the diffusion cascade. Buyers should nevertheless be cautious, because there is now very much depleted uranium available, and unless proper precautions are taken a buyer may pay for natural uranium, but actually receive depleted uranium.

P. VERTÈS: Determining the moisture content of a concentrate is a delicate and important operation. I should like to ask Mr. Shelley whether this gives rise to special difficulties when the moisture content is relatively high - well above the USA standard? For example, with concentrates containing 10–25% H$_2$O?

W.J. SHELLEY: Difficulties have been experienced when concentrates have contained sufficient water to interfere with the mechanical operations of equipment through excessive adherence or changes in flow characteristics. Concentrates containing 0–30% water have been sampled successfully.

P. VERTÈS: Has any segregation of moisture been noted inside the container on concentrates which are initially homogeneous? If so, at what contents does this happen and what are the resulting difficulties in respect of sampling?

W.J. SHELLEY: We have found free water on the inside of the container and on top of the material. In such an event, special arrangements are made with the vendor for acceptable sampling methods.

P. VERTÈS: Of the various sampling methods used in the Mallinckrodt Works, one involving the dropping of concentrates onto a continuously rotating sampler seems to enjoy your preference. Why is this method of sampling not used exclusively?

W.J. SHELLEY: Selection of the sampling method is dependent on production needs, availability of labour, etc. We would prefer to use the falling stream method entirely. However, alternative methods should be available for use on scraps and residues.
METODO DE MUESTREO DOBLE PARA EL CONTROL DE CALIDAD DE UN PROCESO DE FABRICACION

J.A. CERROLAZA, A. LAGO Y ROSA M. MONTOJO
JUNTA DE ENERGIA NUCLEAR, MADRID
ESPAÑA

Abstract — Résumé — Аннотация — Resumen

DOUBLE-SAMPLING METHOD FOR CARRYING OUT QUALITY CONTROL OF A FABRICATION PROCESS.

In fabricating components for use in reactors, two basic factors are of decisive importance, namely the need for very high quality, which allows of only a very small percentage of defective parts, and the very high cost of each of the components. Because of these two considerations, control during fabrication has to follow an operating curve with a very high power and, provided that this condition is fulfilled, the number of parts destroyed in each inspection has to be kept to a minimum. The usual methods, which are based on the determination of the error of first kind, cannot be used because they are generally not effective enough.

Cavé has developed a method in which the size of the samples and the limits of control are established as a function of the errors of first and second kind. This method, although applicable, is not really practicable for the inspection of reactor components since the samples required are too large. The present paper presents a method which is similar to that of Cavé but which relies on double sampling and thus considerably reduces the average size of the sample.

METHODE DU DOUBLE ECHANTILLONNAGE POUR LE CONTROLE DE LA QUALITE D'UN PROCEDE DE FABRICATION. La fabrication des éléments constitutifs des réacteurs nucléaires présente deux caractéristiques essentielles, qui la conditionnent: la qualité supérieure exigée réduit fortement le pourcentage des pièces defectueuses que l'on peut admettre et le prix de revient de chaque élément est très élevé. Étant donné ces deux conditions, les contrôles en cours de fabrication doivent se multiplier à mesure que l'on avance dans la chaîne des opérations; d'autre part, il faut que le nombre des pièces détruites lors de chaque inspection soit aussi réduit que possible, tout en respectant la première condition. Les méthodes habituelles de contrôle qui visent essentiellement à limiter le risque inhérent à la première condition ne sauraient être appliquées car, d'une manière générale, elles ne sont pas suffisamment efficaces.

Cavé a mis au point une méthode dans laquelle la taille des échantillons et le degré de contrôle sont déterminés en fonction des risques inhérents à la première et à la deuxième condition. Bien que cette méthode soit applicable, elle est fort peu pratique quand il s'agit de contrôler des pièces de réacteurs, car les échantillons qu'elle exige sont de dimensions excessives. Le mémoire expose une méthode analogue à celle de Cavé mais qui consiste à établir un double échantillonnage dans lequel la taille moyenne de l'échantillon est beaucoup plus réduite.

МЕТОД ДВУХ ОБРАЗЦОВ ДЛЯ ОСУЩЕСТВЛЕНИЯ КОНТРОЛЯ ЗА КАЧЕСТВОМ В ПРОЦЕССЕ ИЗГОТОВЛЕНИЯ. При изготовлении компонентов для использования в реакторах решающее значение имеют два основных фактора, а именно: необходимость обеспечить очень высокое качество, допускающее лишь очень малый процент дефектных деталей, и очень высокая стоимость каждого из компонентов. По этим соображениям в ходе изготовления необходимо тщательно придерживаться кривой производственных операций, и при соблюдении этого условия число деталей, разрушающихся при каждой проверке, должно быть минимальным. Обычные методы, основанные на определении ошибки первого рода, не могут быть использованы, так как они вообще недостаточно эффективны.

Каве разработал метод, при котором размер образцов и пределы контроля являются функцией ошибок первого и второго рода. Этот метод, хотя он и применим, невозможно использовать для проверки компонентов реактора, поскольку при этом нужны слишком большие образцы. Дается описание метода, аналогичного методу Каве, но основанного на использовании двух образцов, благодаря чему значительно уменьшается средний размер образца.
METODO DE MUESTREO DOBLE PARA EL CONTROL DE CALIDAD DE UN PROCESO DE FABRICACION.

La fabricación de componentes que han de ser empleados en reactores nucleares presenta dos características fundamentales que condicionan el proceso: la exigencia de una calidad muy elevada que limita a valores muy bajos el porcentaje de piezas defectuosas admisibles, y el coste muy alto de cada uno de los componentes. Estas dos condiciones obligan a que el control durante la fabricación posea una curva de operación con potencia muy elevada, y, por otra parte, a que el número de piezas destruidas en cada inspección sea tan pequeño como se pueda, siempre que se cumpla la condición anterior. Los métodos usuales de control, basados en fijar el riesgo de primera especie, no son aplicables, ya que en general su eficacia no es suficiente.

Cavé ha desarrollado un método en que, tanto el tamaño de las muestras como los límites de control, se fijan en función de los riesgos de primera y segunda especie. Este método, si bien es aplicable, resulta poco práctico en el control de componentes para reactores, ya que se necesitan muestras de un tamaño excesivamente grande. En el presente trabajo se desarrolla un método semejante al de Cavé, pero en el que se establece un muestreo doble, con lo que el tamaño medio de la muestra es mucho más reducido.

1. INTRODUCCION

1.1. Los métodos de control por variables de la estabilidad de una fabricación, normalmente empleados, constituyen un contraste de hipótesis. Si se acepta que la producción sigue una distribución normal, por observación de la media y de la varianza de una muestra, o de la media y del recorrido, se contrastan las hipótesis de que la esperanza y la varianza de la producción se mantienen estables.

Las salidas de control de la varianza son poco frecuentes, por lo que el interés principal se centra en el estudio de la estabilidad de la esperanza.

En caso de que el intervalo natural de tolerancia sea menor que el intervalo de tolerancia impuesto por la especificación, se considera que la producción está dentro de control cuando la media aritmética de una muestra de n piezas se mantiene dentro del intervalo, $T_i + Kw$, $T_i - Kw$, donde $T_i$ y $T_s$ son los límites de tolerancia inferior y superior, respectivamente, y $W$ el recorrido medio de una serie de muestras.

Los valores de $K$ que se emplean corrientemente en Europa, corresponden a un riesgo de primera especie del 5% para una producción que posea un 2% de piezas defectuosas [1, 2].

Se puede calcular el riesgo de segunda especie que se comete con estos contrastes, pero no se halla tabulado. En general ello no es importante, ya que en las fabricaciones ordinarias la experiencia anterior puede dictar cuál es el tamaño de muestra más adecuado para conseguir un compromiso óptimo entre el gasto de la inspección y el riesgo de segunda especie.

1.2. Sin embargo, en una fabricación delicada, esta apreciación puede resultar poco afortunada, por lo que es más conveniente elegir en forma objetiva el método de inspección más adecuado.

R. Cavé [3] ha desarrollado un método de control que, basado en las mismas hipótesis que el anterior, posee la ventaja de que el plan de inspección se establece de tal modo que la curva de eficacia de la inspección pase por dos puntos previamente elegidos.

Es decir que, dados un valor del riesgo $\alpha$ y dos producciones - una (considerada de calidad aceptable) con un porcentaje de defectuosas $p_1$ y otra (considerada inaceptable) con un porcentaje de defectuosas $p_2$ - se puede elegir un valor de $n$ y otro de $\gamma$ de tal forma que la inspección tenga una probabilidad $\alpha$ de rechazar la primera producción y la misma probabilidad de aceptar la segunda.
Si consideramos que existe una sola tolerancia $T_1$, el criterio que se sigue en la inspección es aceptar la producción si $x < T_1 - \tau \sigma$ y rechazarla si $x > T_1 - \tau \sigma$.

1.3. La tecnología de componentes y materiales que han de ser empleados en reactores nucleares presenta dos características fundamentales que condicionan todo el proceso y el criterio con el que se deben seleccionar los métodos de control.

En primer lugar, estos componentes han de ser de una calidad muy alta. Los perjuicios que pueden derivarse de un fallo en el reactor son muy graves. Para evitarlos, los componentes empleados han de cumplir una especificación extraordinariamente severa y el riesgo de segunda especie, es decir la probabilidad de que se acepte por bueno un componente que no cumple la especificación, ha de ser extraordinariamente bajo.

La segunda característica es el elevado coste de estos componentes, lo cual, en parte, es consecuencia de la calidad que se les exige.

Este coste aconseja disminuir el riesgo de primera especie para evitar desechar como malo un producto caro y de buena calidad.

1.4. Estas dos características hacen imprescindible una inspección muy cuidadosa del producto terminado. Por ello, normalmente se realiza al final de la fabricación una inspección cien por cien.

Pero esto no es suficiente. La inspección cien por cien está limitada a aquellas propiedades que pueden inspeccionarse mediante métodos no destructivos.

En el curso de la fabricación debe realizarse también una inspección por muestreo. Así se mantiene la fabricación dentro de control, lo que disminuye el número final de rechazos. Además ello constituye una garantía suplementaria de la buena calidad del producto.

Como consecuencia de lo dicho en 1.3 los métodos de control durante la fabricación se han de elegir cuidadosamente y deben basarse en la curva de eficacia. Tanto los riesgos de primera especie, como los de segunda, deben ser muy pequeños, y los porcentajes de piezas defectuosas, $p_1$ y $p_2$, correspondientes a una fabricación que se desea aceptar y a una que se quiere rechazar, también deben ser pequeños.

 Esto implica que el tamaño de las muestras ha de ser muy grande. Así por ejemplo, si admitimos un riesgo del 2% de rechazar una producción con un 0,01% de piezas defectuosas y de aceptar una producción con un 0,2% de piezas defectuosas, el método de Cavé da una muestra de 48 piezas. Unas muestras tan grandes suponen un coste de inspección muy elevado, más aún si se considera el valor unitario de cada pieza, así como el coste de muchos de los métodos de inspección que deben emplearse.

En resumen, podríamos decir que los métodos de control usuales, aplicados a la industria nuclear, resultan o demasiado caros o demasiado poco eficaces, a consecuencia de las altas calidades exigidas y el elevado coste de los componentes de reactores.

1.5. En la Junta de Energía Nuclear se pensó que sería deseable establecer un método adecuado para la inspección de elementos nucleares; este método tendría que poseer una curva de eficacia muy potente con un tamaño de la muestra lo más pequeño posible.
En el presente trabajo describimos el método de inspección que se ha desarrollado y se está empleando con resultados satisfactorios.

2. DESCRIPCIÓN DEL METODO

2.1. Nuestro método constituye una prolongación del método de Cavé. En su forma de aplicarse se asemeja a los métodos de muestreo doble que se emplean en el control de recepción de un lote. Es decir, que si consideramos que solamente existe un límite de tolerancia $T_1$, se toma una muestra de tamaño $n$ y se determina la media aritmética de la muestra, $\bar{x}$. 

a) Si $\bar{x} < T_1 - K_1\sigma$, se admite que la fabricación está dentro de control.

b) Si $\bar{x} > T_1 - K_2\sigma$, se decide que la fabricación está fuera de control.

c) Si $T_1 - K_1\sigma < \bar{x} < T_1 - K_2\sigma$, se toma una segunda muestra del mismo tamaño $n$. En la muestra total, de $2n$ piezas, se determina la media aritmética $\bar{x}'$.

i) Si $\bar{x}' < T_1 - K_3\sigma$, se admite que la fabricación permanece dentro de control.

ii) Si $\bar{x}' > T_1 - K_3\sigma$, se decide que la fabricación está fuera de control. Naturalmente, en el caso de que existan dos límites de tolerancia, la media muestral debería compararse también con

$$T_1 + K_1\sigma; \quad T_1 + K_2\sigma$$

y, si procediera, con

$$T_1 + K_3\sigma$$

2.2. Al desarrollar el procedimiento consideramos que el poder elegir dos valores distintos para $n_1$ y $n_2$ daba más flexibilidad al método, pero desde el punto de vista de realización práctica, ello presentaba algunos inconvenientes. Si se decidía que ambas muestras debían tener un mismo tamaño $n$ se evitaban errores y complicaciones por parte de los inspectores, por lo que se adoptó este criterio.

3. CALCULO DE $n$, $K_1$, $K_2$ Y $K_3$

3.1. El método se basa en las siguientes premisas:

a) La producción sigue una distribución normal de desviación típica $\sigma$, invariable.

b) Se desea operar con un riesgo $R$ de rechazar una producción con un porcentaje de defectuosas $p_1$.

c) Se desea operar con un riesgo $R$ de aceptar una producción con un porcentaje de defectuosas $p_2$.

d) El tamaño esperado de la muestra, $N$, debe ser mínimo.

3.2. Este tamaño esperado no es más que la esperanza matemática del tamaño de la muestra y, fijado el método de inspección, depende del porcentaje de piezas defectuosas en la producción actual. Por lo tanto, no se puede conseguir que $N$ sea mínimo para cualquier producción.
Considerando que en una producción bajo inspección $p$ coincidirá con $p_1$ o será muy próxima a ella, se decidió concretar 3.1. d) en la siguiente forma:

El tamaño esperado de la muestra, para una producción con $p_1$ defectuosas, debe ser mínimo.

3.3. Sean:

- $\lambda_1$, la diferencia, expresada en unidades típicas, entre $T_1$ y el valor medio de la fabricación con $p_1$ defectuosas.
- $\lambda_2$, la diferencia, expresada en unidades típicas, entre $T_1$ y el valor medio de la fabricación con $p_2$ defectuosas.
- $x$, la probabilidad de que la media de una muestra de tamaño $n$, de una población con $p_1$ defectuosas, sea $> T_1 - K_1 \sigma$.
- $y$, la probabilidad de que la media de una muestra de tamaño $n$, de una población con $p_1$ defectuosas, sea $> T_1 - K_2 \sigma$.
- $z$, la probabilidad de que la media de una muestra de tamaño $2n$, de una población con $p_1$ defectuosas, sea $> T_1 - K_3 \sigma$.
- $X$, $Y$, $Z$, los valores correspondientes a $x$, $y$, $z$, en una población con $p_2$ defectuosas.

Denominemos:

$$F(a) = \int_a^{\infty} \frac{1}{\sqrt{2\pi}} e^{-\frac{u^2}{2}} \, du$$

Si

$F(a) = b$, diremos que $b = g(a)$

3.4. De acuerdo con la nomenclatura establecida en 3.3, las hipótesis de 3.1 se formularán:

$$y + (x-y)z = R \quad (3.4.1)$$
$$Y + (X-Y)Z = 1 - R \quad (3.4.2)$$

$$N = E(n) = n(1-x+y) + 2n(x-y) = n(1+x-y) = \min \quad (3.4.3)$$

La resolución analítica del sistema (3.4.1), (3.4.2) y (3.4.3) conduce a un complicado conjunto de ecuaciones no algebraicas. Pero fácilmente se puede obtener la solución por métodos numéricos.

3.5. En efecto, de acuerdo con (3.3) tendremos:

$$\lambda_1 = g(p_1) \quad (3.5.1)$$
$$\lambda_2 = g(p_2) \quad (3.5.2)$$
$$\sqrt{n}(\lambda_1-K_1) = g(x) \quad (3.5.3)$$
$$\sqrt{n}(\lambda_1-K_2) = g(y) \quad (3.5.4)$$
$$\sqrt{2n}(\lambda_1-K_3) = g(z) \quad (3.5.5)$$
$$\sqrt{n}(\lambda_2-K_1) = g(X) \quad (3.5.6)$$
\[ \sqrt{n} \left( \lambda_2 K_2 \right) = g \left( Y \right) \]  
(3.5.7)

\[ \sqrt{n} \left( \lambda_3 K_3 \right) = g \left( Z \right) \]  
(3.5.8)

Si llamamos

\[ t_1 = \frac{K_1}{\lambda_1} \]  
(3.5.9)

\[ t_2 = \frac{K_2}{\lambda_1} \]  
(3.5.10)

\[ t_3 = \frac{K_3}{\lambda_1} \]  
(3.5.11)

\[ L = \frac{\lambda_2}{\lambda_1} \]  
(3.5.12)

\[ a = g \left( x \right) \]  
(3.5.13)

\[ b = g \left( y \right) \]  
(3.5.14)

\[ c = \frac{1}{\sqrt{2}} g \left( z \right) \]  
(3.5.15)

\[ A = g \left( X \right) \]  
(3.5.16)

\[ B = g \left( Y \right) \]  
(3.5.17)

\[ C = \frac{1}{\sqrt{2}} g \left( Z \right) \]  
(3.5.18)

y elegimos una variable auxiliar

\[ t = c \frac{\left( 1-L \right)}{1-t_3} \]  
(3.5.19)

se llega fácilmente a que

\[ A = a-t \]  
(3.5.20)

\[ B = b-t \]  
(3.5.21)

\[ C = c-t \]  
(3.5.22)

\[ N = t^2 \left( \frac{1+x-y}{\lambda_1-\lambda_2} \right) \]  
(3.5.23)

En consecuencia, (3.4.2) y (3.4.3) equivalen a:

\[ F \left( Y \right) + \left[ F \left( X \right) - F \left( Y \right) \right] F \left( \sqrt{2} C \right) = \text{I-R} \]  
(3.5.24)
Estas ecuaciones unidas a la (3.4.1) y a las (3.5.13) - (3.5.22), permiten determinar fácilmente con el auxilio de una máquina de calcular, los valores de \(x, y, t\) que satisfacen el sistema.

3.6. Para ello, una vez fijado el valor de \(R\), basta elegir un par de valores \(x, y\). Por medio de (3.4.1) se halla \(z\). Mediante (3.5.13), (3.5.14) y (3.5.15) se obtienen \(a, b, y c\).

Se toma un valor de \(t\) con el que se hallan \(A, B y C\) mediante (3.5.20), (3.5.21) y (3.5.22). Se calculan \(X, Y, Z\) y se halla el valor de

\[
\psi = \frac{F(X) - F(Y)}{F(Z)} + F(Y)\tag{3.6.1}
\]

Se varía \(t\) hasta que:

\[
\psi = 1 - R
\]

Si \(t\) es el valor que cumple esta ecuación se calcula

\[
M = t^2 (1 + x - y)\tag{3.6.2}
\]

Se varían ahora \(x\) e \(y\), repitiendo todo el proceso hasta alcanzar el mínimo de \(M\).

La solución hallada depende exclusivamente de \(R\), es decir que se ha calculado una terna \(x^*, y^*\) y \(t^*\) que cumple con las condiciones (3.4.1) (3.5.24) y (3.5.25), y en cuyos valores no interviene \(p_1\) ni \(p_2\).

3.7. Volviendo a las ecuaciones (3.5.3) a (3.5.12) se deduce de ellas que si \(x^*, y^*, t^*\) son la solución hallada y

\[
a^* = g(x^*)\tag{3.7.1}
\]

\[
b^* = g(y^*)\tag{3.7.2}
\]

\[
c^* = \frac{1}{\sqrt{2}} - g(z^*)\tag{3.7.3}
\]

\[
M^* = t^2 (1 + x^* - y^*)\tag{3.7.4}
\]

se realiza:

\[
K_1 = \lambda_1(1 - \frac{a^*}{t^*}) + \lambda_2 \frac{a^*}{t^*}\tag{3.7.5}
\]

\[
K_2 = \lambda_1(1 - \frac{b^*}{t^*}) + \lambda_2 \frac{b^*}{t^*}\tag{3.7.6}
\]

\[
K_3 = \lambda_1(1 - \frac{c^*}{t^*}) + \lambda_2 \frac{c^*}{t^*}\tag{3.7.7}
\]
que permiten determinar con facilidad la solución del problema para cualesquiera valores de $p_1$, $p_2$ y el $R$ propuesto.

3.8. En las figuras 1 a 12 se han construido unos ábacos para resolver gráficamente las ecuaciones (3.7.5) a (3.7.9) con riesgos del 0,2%, 0,3%, 0,4%, 0,5%, 0,75% y 1,0%.

![Abaco para la determinación de $K_1$, $K_2$ y $K_3$ en función de $p_1$ y $p_2$ para un riesgo del 0,2%](image-url)
4. CONCLUSIONES

4.1. Se ha establecido un método de inspección por muestreo doble que consiste en tomar una muestra de \( n \) objetos y determinar la media. Según el valor de ésta, se acepta la producción, se rechaza, o se decide tomar una segunda muestra del mismo tamaño. En este último caso la media de la muestra total decide de la aceptación o del rechazo de la fabricación.

4.2. Se establecen las fórmulas que permiten determinar, mediante análisis numéricos, un grupo de parámetros que dependen exclusivamente del riesgo elegido. La determinación del tamaño de la muestra, el tamaño esperado y los límites de control, son funciones sencillas de estos parámetros y de \( p_1 \) y \( p_2 \).
4.3. Asimismo, se dan unos abacos en los que se pueden hallar el tamaño de la muestra, su esperanza y los límites de control, en función de $p_1$ y $p_2$, para seis valores seleccionados del riesgo.

4.4. Este método ha dado resultados satisfactorios en su aplicación en la JEN. Con él se consigue un gran ahorro en el coste de la inspección. Por ejemplo, para el caso descrito en 1.4 el tamaño de la muestra sería de 26 piezas, con un tamaño esperado de 31.5, lo que supone un ahorro del 33% respecto a un muestreo simple con la misma eficacia.
FIG. 4. Abaco para la determinación de n y N en función de p₁ y p₂ para un riesgo del 0.3%
<table>
<thead>
<tr>
<th>P %</th>
<th>K1</th>
<th>K2</th>
<th>K3</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.00</td>
<td>0.9</td>
<td>0.8</td>
<td>0.7</td>
</tr>
<tr>
<td>2.3</td>
<td>2.2</td>
<td>2.1</td>
<td>2.0</td>
</tr>
</tbody>
</table>

**FIG. 5.** Abaco para la determinación de K₁, K₂ y K₃ en función de P₁ y P₂ para un riesgo del 0.4%
FIG. 6. Abaco para la determinación de $n$ y $N$ en función de $p_1$ y $p_2$ para un riesgo del 0.4%
FIG. 7. Abaco para la determinación de $K_1$, $K_2$ y $K_3$ en función de $p_1$ y $p_2$ para un riesgo del 0.5%
FIG. 8. Abaco para la determinación de $n$ y $N$ en función de $p_1$ y $p_2$ para un riesgo del 0.5%
FIG. 9. Abaco para la determinación de \( K_1 \), \( K_2 \) y \( K_3 \) en función de \( p_1 \) y \( p_2 \) para un riesgo del 0,75%
FIG. 10. Abaco para la determinación de $n$ y $N$ en función de $p_1$ y $p_2$ para un riesgo del 0,75%
<table>
<thead>
<tr>
<th>$p_i$</th>
<th>$K_1$</th>
<th>$K_2$</th>
<th>$K_3$</th>
<th>$P_x$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.00</td>
<td>2.3</td>
<td>2.1</td>
<td>3.00</td>
<td>3.50</td>
</tr>
<tr>
<td>0.9</td>
<td>2.4</td>
<td>2.2</td>
<td>3.10</td>
<td>3.25</td>
</tr>
<tr>
<td>0.8</td>
<td>2.5</td>
<td>2.1</td>
<td>3.00</td>
<td>3.00</td>
</tr>
<tr>
<td>0.7</td>
<td>2.6</td>
<td>2.00</td>
<td>2.75</td>
<td>2.75</td>
</tr>
<tr>
<td>0.6</td>
<td>2.7</td>
<td>2.00</td>
<td>2.50</td>
<td>2.50</td>
</tr>
<tr>
<td>0.5</td>
<td>2.8</td>
<td>1.50</td>
<td>2.25</td>
<td>2.25</td>
</tr>
<tr>
<td>0.4</td>
<td>2.9</td>
<td>1.25</td>
<td>2.00</td>
<td>2.00</td>
</tr>
<tr>
<td>0.3</td>
<td>3.0</td>
<td>1.00</td>
<td>1.75</td>
<td>1.75</td>
</tr>
<tr>
<td>0.2</td>
<td>3.1</td>
<td>0.9</td>
<td>1.50</td>
<td>1.50</td>
</tr>
<tr>
<td>0.1</td>
<td>3.2</td>
<td>0.8</td>
<td>1.25</td>
<td>1.25</td>
</tr>
<tr>
<td>0.09</td>
<td>3.3</td>
<td>0.7</td>
<td>1.00</td>
<td>1.00</td>
</tr>
<tr>
<td>0.08</td>
<td>3.4</td>
<td>0.6</td>
<td>0.9</td>
<td>0.9</td>
</tr>
<tr>
<td>0.07</td>
<td>3.5</td>
<td>0.8</td>
<td>0.8</td>
<td>0.8</td>
</tr>
<tr>
<td>0.06</td>
<td>3.6</td>
<td>0.7</td>
<td>0.9</td>
<td>0.9</td>
</tr>
<tr>
<td>0.05</td>
<td>3.7</td>
<td>0.6</td>
<td>0.8</td>
<td>0.8</td>
</tr>
<tr>
<td>0.04</td>
<td>3.8</td>
<td>0.5</td>
<td>0.7</td>
<td>0.7</td>
</tr>
<tr>
<td>0.03</td>
<td>3.9</td>
<td>0.4</td>
<td>0.6</td>
<td>0.6</td>
</tr>
<tr>
<td>0.02</td>
<td>4.0</td>
<td>0.3</td>
<td>0.5</td>
<td>0.5</td>
</tr>
<tr>
<td>0.01</td>
<td>4.1</td>
<td>0.2</td>
<td>0.4</td>
<td>0.4</td>
</tr>
</tbody>
</table>

**FIG. 11.** Abaco para la determinación de $K_1$, $K_2$ y $K_3$ en función de $p_1$ y $p_2$ para un riesgo del 1.0%
FIG. 12. Abaco para la determinación de n y N en función de $p_1$ y $p_2$ para un riesgo del 1,0%

BIBLIOGRAFÍA

A MULTI-SCALER RECORDING SYSTEM AND ITS APPLICATION TO RADIOMETRIC "OFF-LINE" ANALYSIS

H. BISBY
ATOMIC ENERGY RESEARCH ESTABLISHMENT, HARWELL, BERKS., UNITED KINGDOM

Abstract — Résumé — Аннотация — Resumen

A MULTI-SCALER RECORDING SYSTEM AND ITS APPLICATION TO RADIOMETRIC "OFF-LINE" ANALYSIS. In large complex reprocessing plants a great deal has been done over the past few years to provide in-line instrumentation for the contemporary analysis of process stream content and characteristics. However, these instruments have a qualitative rather than a quantitative part to play in the overall control of the plant. Quantitative information, which must be obtained for control and accounting purposes, demands and relies upon the efficient use of laboratory techniques and instrumentation for the precise analysis of representative samples taken from the process streams.

These techniques, in particular those involving pulse counting systems, can be made automatic with modern instrumentation, such as will be described, in which the data is obtained in digital form in electronic stores (escanners). To support a large plant there will be many separate counting systems of this kind, independently controlled and therefore having no time correlation between them.

The automatic recording system described in the paper provides a common data read-out facility for more than 50 independently operating counting systems, recording scaler data, together with associated sample and system identification and the absolute time occurrence of each read-out. The data can be recorded, in forms suitable for subsequent processing by a computer, on a variety of tape and card punches, serial and parallel printers or magnetic tape. In addition, the whole recording system, including the scalers in any one system, can be checked for correct operation on an automatic routine basis which does not interfere with the operation of other counting systems.

It is concluded that the effective quantitative control of a plant rests on a rapid efficient sample analysis under laboratory conditions. It is probable that future developments of "off-line" facilities rather than on-line instrumentation will be possible and more worthwhile. The desirable characteristics of instrumentation for such a laboratory have been reviewed and an example demonstrates the value of unitized instrumentation and a comprehensive, flexible data recording system. Possible future developments have been mentioned in data processing which could lead to a completely automatic service becoming available.
par des machines à imprimer - en série ou en parallèle - ou sur bande magnétique. En outre, il est possible de vérifier le bon fonctionnement de tout le système d’enregistrement, y compris celui de chaque échelle de comptage, grâce à un dispositif entièrement automatique qui ne gêne en aucune manière le fonctionnement des autres appareils.

Les auteurs concluent que la qualité du contrôle quantitatif dans une installation dépend de la rapidité et de l’efficacité des analyses d’échantillons en laboratoire. Selon toute probabilité, la mise au point dans l’avenir de dispositifs non incorporés dans le circuit sera possible et permettra d’obtenir de meilleurs résultats qu’un appareillage incorporé. Les auteurs passent en revue les caractéristiques souhaitables de l’appareillage destiné à un laboratoire comme celui dont il est question ici et montrent par un exemple la valeur d’un appareillage par secteurs et d’un système souple et complet d’enregistrement des données. Les progrès possibles des techniques de traitement des données dont ils ont parlé permettront peut-être d’organiser un service complètement automatique.

МУЛЬТИСКАЛЯРНАЯ РЕГИСТРИРУЮЩАЯ СИСТЕМА И ЕЕ ПРИМЕНЕНИЕ К РАДИО-МЕТРИЧЕСКИМ И "ВНЕЛИНЕЙНЫМ" АНАЛИЗАМ. За последние несколько лет на крупных комплексных заводах по переработке ядерного топлива проделана большая работа с целью их обеспечения контрольными приборами для современных анализов технологических потоков и характеристик. Однако эти приборы играли большую качественную, чем количественную роль в общем контроле завода. Количественная информация, требующаяся для контроля и учета, основывается на эффективном использовании лабораторных методов и приборов для точного анализа характерных образцов, взятых из технологического потока.

Эти методы, и в частности те, которые связаны с импульсными счетными системами, могут быть автоматизированы с помощью современных приборов, таких, каких будут описаны в докладе, и от которых данные получаются в виде цифр в электронных накопителях (персчетные устройства). Для управления большим заводом нужно иметь много независимо управляемых отдельных счетных систем такого рода и, таким образом, не имеющих между собой временной корреляции.

Автоматическая система записи данных, описанная в настоящем докладе, представляет собой устройство отсчета показаний общих данных более, чем 50 независимо действующих счетных систем, данных записывающего счетного устройства, а также образца и системы идентификации и абсолютно точного времени каждого отсчета показаний. Данные могут записываться в таком виде, который пригоден для последующей обработки на вычислительной машине, на различного рода лентах и перфокартах, последовательно или параллельно печатающих устройствах или магнитных. Кроме того, можно автоматически проверять равномерность работы, записывающей системы, включая персчетные устройства в любой системе, что не ограждается на работе других счетных систем.

Делается вывод, что эффективный количественный контроль на заводе основывается на быстром проверяемом аналогичном образцом в лабораторных условиях. Вероятно в будущем возможно и более целесообразно разработать устройство "вне потока", а не приборов на потоке. Дается обзор желательных характеристик приборов в такой лаборатории, и на примере демонстрируется ценность использованных приборов и точной гибкой системы регистрации данных. Упоминаются возможные разработки в будущем в области обработки данных, которые могут привести к созданию полностью автоматической системы.

UN SISTEMA DE REGISTRO DE MULTIPLES ESCALIMETROS Y SU APLICACION AL ANALISIS RADIOMETRICO DISCONTINUO. Mucho se ha hecho durante los últimos años en las grandes plantas de recuperación con objeto de disponer de un sistema continuo de instrumentos para el análisis del contenido y de las características de la corriente de materiales de un proceso. Ahora bien, esos instrumentos desempeñan un papel más bien cualitativo en el control general de la planta. La información cuantitativa que requieren los servicios de control y contabilidad exige el uso eficiente de técnicas e instrumentos de laboratorio para el análisis exacto de muestras representativas tomadas de la corriente de materiales.

Esas técnicas, sobre todo las que implican sistemas de recuento de impulsos, pueden automatizarse utilizando los instrumentos modernos descritos en la memoria, con los cuales se obtienen datos numéricos de depósitos electrónicos (escalimetros). En una planta de gran capacidad se necesitan muchos instrumentos de recuento de este tipo controlados independientemente y, por lo tanto, sin correlación alguna de tiempo entre ellos.

El sistema de registro automático descrito en la memoria está dotado de un dispositivo común de lectura para más de 50 instrumentos independientes de recuento; registra los datos de los escalimetros, identifica la
muestra y el instrumento e indica el tiempo absoluto en que se produce cada lectura. Los datos pueden registrarse con una gran variedad de cintas y fichas perforadas, con impresiones seriadas o paralelas, o con cinta magnética, para su ulterior sistematización mediante una calculadora electrónica. Además, el funcionamiento de toda la instalación de registro, incluidos los escalímetros de cualquiera de los sistemas, puede verificarse automáticamente sin interferir en el funcionamiento de otros instrumentos de recuento.

Los autores llegan a la conclusión de que la eficacia del control cuantitativo de una planta reside en el análisis rápido y eficaz de las muestras en un laboratorio. Es probable que el desarrollo de instalaciones para el análisis discontinuo resulte más factible y conveniente que el de un sistema de análisis continuo. En la memoria se examinan las características que deberían tener los instrumentos de un laboratorio para ese tipo de análisis y se demuestra con un ejemplo la utilidad de la unificación de los instrumentos y de un sistema amplio y flexible de registro de datos. Se menciona la posibilidad de llegar a un servicio totalmente automático de elaboración de los datos.

1. INTRODUCTION

In large reprocessing plants, a good deal has been achieved in the past few years to increase the sensitivity and reliability of "on-line" instrumentation for the contemporary analysis of process stream for fission products, uranium and transuranic elements [1, 2]. However, where the environmental conditions militate against the known transducers or measurement techniques, for example where highly acidic streams are present, where unwanted radiations arise from interfering elements or from neighbouring streams, or where contamination build-up on the sample vessel occurs, these "on-line" instruments have only a qualitative, rather than a quantitative, part to play in the overall control and accountancy of the processes.

Quantitative information for these purposes must therefore be obtained by a precise analysis of representative process samples of which only refined laboratory techniques are capable. To be most effective, these techniques must be applied in such a way that the measured data is analysed, processed and made available for operators and management within the shortest possible time period after sample procurement. It is in these fields where revolutionary changes and improvements are most likely to occur in the near future, rather than in the further improvement of "on-line" instruments.

Already, instrumentation of great sophistication is available in the laboratory for producing data, analysing and recording it. In addition, increasing use will be made of data processors for correlating the measured data from many different samples taken at any one time to give values for process efficiency, flow-sheet product balances and the stock balances of process reagents. To achieve this, the speed of analysing and processing data will have to be increased and ultimately must become automatic to the extent that the processor will not only compute but will also be programmed to, or directly, exercise control over the variable parameters of the sampling and analysing equipment.

2. INSTRUMENTATION FOR AN ANALYTICAL LABORATORY

Before considering the data-processing aspect, it is worthwhile to consider the characteristics which the electronic equipment should have in order to provide these analyses in a modern "off-line" laboratory.
In the first place, the instruments should be contemporary with the rapid developments taking place in electronic techniques and solid state devices so that the maximum advantage can be obtained from these in terms of increased speed and precision of analysis. The equipment should be as flexible as possible in its operational role, thus allowing a change in plant operation or process characteristic to be accommodated by the analytical laboratory either by using existing equipment or by minor changes, but in both cases without further major expenditure on new equipment and without the delays which the procurement of new equipment often implies.

The greater the extent to which the equipment can be made automatic in operation, the smaller the team of technicians required to operate it. Trained scientific effort can be thus relieved of routine work and used more effectively or, where fully automatic systems are economically prohibitive, lower grade effort can be employed.

As the number or complexity of instruments increases with increased automation, their reliability should be increased. This is an important factor in relation to the cost of providing an analytical service since "down-time" caused by instrument failure, the size of the maintenance team and the stockholdings of spare units and components all make their contributions to it.

A second factor is the size of the laboratory, related to the physical size of the instrumentation, and the cost of heat extraction and cooling systems of the laboratory which are related to the heat generated by the instrumentation, so that both these characteristics must be considered and their contribution kept small.

In these requirements there are obvious contradictions since contemporary instrumentation implies frequent replacement of obsolete units and therefore recurrent expenditure on new units. Increased complexity and flexibility implies a greater capital cost of instrumentation. Increased reliability is often only possible by reducing complexity and using well-tried components and techniques, thereby precluding the concept of contemporary instrumentation.

Contradictions such as these are apparent in other fields, particularly in research work which relies extensively on instrumentation and for this latter case a unit system of electronics has been designed and made available at AERE, Harwell, and other national research centres in the United Kingdom, for the purpose of detecting, measuring and analysing data which appear in pulse form, as an example from radiation directors. This system is therefore directly applicable to the analytical services of a process plant.

3. THE HARWELL\(^1\) 2000 SERIES SYSTEM

The design philosophy of this system [3] requires that each function in a counting channel, i.e. amplification and discrimination (DC and pulse), pulse amplitude selection, the measurement of count and time, analogue to digital conversion, AC-DC power conversion etc., should be separately pro-

\(^1\) HARWELL is the registered trade mark of the United Kingdom Atomic Energy Authority.
vided by separate units, these units being modular in physical dimensions and otherwise standardized in terms of electrical supplies, connectors and information transmission between units. The 2000 Series contains a large selection from a wide variety of "basic" transistorized units for these functions.

Over three years' experience with the system has shown the following features to be substantiated:

(a) Flexibility in use, in that an operation role of one assembly can be changed merely by substituting or adding other basic units.

(b) An expansion of the facilities available from an assembly of basic units can be rapidly obtained by the redevelopment, or the additional development, of a small number (usually one) of new units.

(c) Advantage can be taken of advances in electronic components and techniques as these occur, by redevelopment of basic units.

The above features make it possible to maintain laboratory equipment in a contemporary state, at a reasonable investment cost.

(d) Standardization of the electrical aspects of the system enables expert circuit design effort to concentrate on those units which demand specialist techniques and in this way contributes to more thorough design.

(e) The features of compatibility which must exist between units require that high standards should be maintained throughout all design work.

(f) The mechanical standardization of units (including built-in ventilation) allows an assembly, rendered non-operational by a faulty unit, to be rapidly brought back into operation by substitution of the faulty unit by one of a similar type.

(g) The specification of a single-function unit, being simpler than that for a multi-functional unit, offers better control over manufacture, inspection and testing.

All these make their contribution towards increased reliability and an overall decrease in 'down time'.

(h) Where many counting channels are involved, each employing similar basic units, the numbers of spare units required on stand-by against failure is small.

There are other advantages available from such a comprehensive unit system of electronics, however, the above points demonstrate its applicability to the analytical laboratory.

4. AUTOMATIC DATA RECORDING

The measured data in most counting channels is in digital form and is stored in scalers (registers). In the Harwell 2000 Series, the scalers have visual indication in BCD code of their store content and in addition some have electrical access to the stores via a 50-way connector (Fig. 1). If the scaler is suitably addressed, the content of all decades is available at this connector together with signals to show whether the scaler has overflowed, or is connected, addressed and switched on. These scalers are of three varieties which have resolving times and decade stores respectively of $1.0 \mu s$, 6; $0.1 \mu s$, 6; $0.01 \mu s$, 4.
Three read-out systems are available, however the one of interest for this paper is known by the Type Number 3016 [4]. The situation to be considered is one of an analytical laboratory having several counting rooms, each allocated to a specific task and each having several counting channels into which samples from process streams are regularly supplied for measurement of analysis. Any one or more channels could be associated with an automatic sample-changer and in any case the sequence of "count, readout, reset, sample-change or hold" will be uncorrelated in time with similar
sequences in other channels. Furthermore the time at which each sample measurement is made will be referenced to some zero time. This is just one of the situations with which the Automatic Read-out System 3016 will deal and provide recorded data on a common, central recording facility which may be any two of such devices as parallel or serial printers, card or tape punch or magnetic tape recorder.

The data from any one channel may be accumulated in \( n \) scalers depending on the type of analysis being performed. A common value for \( n \) is 2, one scaler accumulating count pulses, the other clock pulses so that time or count-control of the measurement can be available. In the 3016 system the value of \( n \) can be, however, any number from 1 to 396 subject to the limitation set out in the next paragraph. This group of \( n \) scalers is referred to as a 'module' in what follows and each scaler is connected to a common 50-line highway via its 50-way connector and a junction unit (Fig. 2) whose function, additionally, is to allocate to each scaler a unique address. In this way \( n \) scalers are connected to the module controller which in its fully automatic mode of operation, generates for all these scalers a repetitive sequence of "count, print, reset, sample-change/hold". The count period can be started by a clock pulse and ended by limit signals from any scaler. The sample-change programme step demands action from associated equipment and awaits acknowledgment before proceeding further on the sequence. The programme steps can be individually selected by manual control or sequenced for single or repetitive cycles. Four addresses are allocated to each module controller, these being occupied by three groups of six digits on which preset data can be set up manually (i.e. for labelling the data from the \( n \) scalers), the fourth group allowing data to be introduced from an external source or sample changer. Thus \( (n + 4) \) addresses are occupied by each module.

There can be \( N \) modules of \( n \) scalers, each module connected by the 50-line highway to a common recording controller which serves 400 addresses i.e. \( N (4 + n) = 400 \) defines the maximum number of separate modules or counting channels. When the recording controller selects an address, the scaler (or preset data group) having that address is connected to the highway so that the contents are displayed in decimal code at the recording controller. Normally, the display shows the contents of the selected scaler and its address, but when the recording controller receives a print command from one of the \( N \) modules, it searches at the rate of 1000 addresses per second for the first address in the requesting module. The contents of this address are gated onto the highway, displayed and offered to the recording devices. When these have accepted the data, the recording controller searches for the next occupied address and so on until the data has been accepted from the last address of that module. The recording controller then returns to the preset address or continues to search sequentially if a second print command has appeared in the meantime. Thus queues of modules requiring read-out are treated systematically.

The recording controller distributes the data for recording as 10 digits, three address digits, six content digits and one digit for identifying the source of data (scaler, prescaler, preset data) so that the read-out onto a printer could be as shown in Table I when the channels 029 and 030 contain two scalers as described earlier.
FIG. 2. Automatic read-out from multiple independent modules onto common recording facilities
### TABLE I

**TYPICAL PRINTED RECORD FROM 3016 SYSTEM**

<table>
<thead>
<tr>
<th>Record</th>
<th>Abs. Time 1</th>
<th>Channel</th>
<th>Detector</th>
<th>Gain</th>
<th>E.H.T.</th>
</tr>
</thead>
<tbody>
<tr>
<td>091</td>
<td>263479</td>
<td>029</td>
<td>06</td>
<td>4405</td>
<td>1470</td>
</tr>
<tr>
<td>092</td>
<td>263479</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>093</td>
<td>001000</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>094</td>
<td>024617</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>101</td>
<td>029064</td>
<td></td>
<td></td>
<td>B/gd. 3.245</td>
<td></td>
</tr>
<tr>
<td>102</td>
<td>405147</td>
<td></td>
<td></td>
<td>Sample No. 007</td>
<td></td>
</tr>
<tr>
<td>103</td>
<td>032450</td>
<td></td>
<td></td>
<td>Plant Loc. 111</td>
<td></td>
</tr>
<tr>
<td>104</td>
<td>007111</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>221</td>
<td>263482</td>
<td>030</td>
<td>07</td>
<td>3900</td>
<td>1400</td>
</tr>
<tr>
<td>222</td>
<td>263482</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>223</td>
<td>002531</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>224</td>
<td>010004</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>231</td>
<td>030073</td>
<td></td>
<td></td>
<td>B/gd 3.243</td>
<td></td>
</tr>
<tr>
<td>232</td>
<td>000140</td>
<td></td>
<td></td>
<td>Sample No. 011</td>
<td></td>
</tr>
<tr>
<td>233</td>
<td>032430</td>
<td></td>
<td></td>
<td>Plant Loc. 295</td>
<td></td>
</tr>
<tr>
<td>234</td>
<td>011295</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Absolute time, referenced to some zero time is provided again by a common facility by which as many as four scalers common to several or all N modules can be recorded as the first batch of addresses in each module. These scalers are supplied with clock pulses but are not reset after printing. In the example given in Table I, only two scalers are demonstrated and the clock pulses have been supplied to them in parallel to provide a redundancy check on the scalers, but of course the scalers could be connected in cascade to give up to 12 decades \(10^{12}\) of clock pulse intervals. The three groups of preset data would typically be coded, as shown, to give channel number (three digits), type of detector (two digits), amplifier gain setting (four digits), E.H.T. value (four digits), background count rate (five digits). The fourth group would be coded as sample number (three digits) and plant location appropriate to that sample (three digits).

The whole system of scalers, junction units, controllers and recording machines can be automatically and systematically checked for correct operation at any time or according to a programme (for example, after X printouts, X being a presetable value). The test [5] is a means of generating pulse trains, containing exactly 111111 pulses (pulse recurrence frequency = \(10^6\) pulses/sec) which are applied to all scalers in a module. The scalers
are not reset between trains so that their contents will be therefore 111111; 222222; 333333 etc. after successive pulse trains and the ability to read out and record every digit from all decades of all scalers can be checked. Any print demand arising from any other module during the check period of a particular module is ignored until that check is completed, and, to avoid the risk that misuse of the check facility might occupy the recording controller indefinitely, there is an automatic release from checking after a few seconds.

5. A TYPICAL ASSEMBLY

An assembly, which would be typical of one associated with an analytical service laboratory, has now been in full operation for more than 12 months. This is in the Rutherford Laboratory of the Science Research Council at Chilton, Berks (previously NIRNS). Target materials are exposed to the heavy-ion beam of Nimrod and then required to be analysed for induced radioisotopes. The scale of the operation requires 12 counting modules, to deal with the number of irradiated targets, and these are disposed four to each of three counting rooms which occupy an area approximately 45 m x 15 m. The counting modules comprise Harwell 2000 Series analysing equipment and scalers whose number (n) has varied over the year from 2 to 8 per module. Two modules are fitted with automatic sample changers. Each module has its local module controller so that a total of 12 measurements can proceed independently of each other, but the recorder controller, parallel printer (15 scalers per second speed) and serializer plus punch-card machine (1 scaler per second) are disposed in a separate room towards one end of the suite, i.e. outside the "contamination zone". The principal measurements made are concerned with the identification of isotopes, by means of their half-life, and establishment of individual isotope yield at the termination of sample irradiation in Nimrod, which times are staggered successively on each sample with respect to an absolute time zero. Recorded measured data together with preset data is fed to an off-line computer which provides an analysis in terms of what isotopes can be present and their yield with probable error values.

6. FUTURE DEVELOPMENTS

An analytical laboratory of a process plant equipped with a similar, but probably larger, instrumentation scheme to that described above with off-line access to a computer would be about as up-to-date as present equipment availability will allow. Nevertheless, such a laboratory would be able to provide a large through-put of samples since it would be amenable to 24-hour operation with a small complement of staff. The capital cost of instrumentation would be high (of the order of £1500 per counting channel on average) but this would be more than compensated for, if in transistorized unit form throughout, by reduction of 'down time' and increased efficiency without loss of quantitative status. Furthermore, the whole laboratory could be physically quite small.
At present, the variety of preset and variable parameters associated with the measuring equipment are manually operated and local to the particular units (i.e. knobs, switches, connectors, etc. on front panels). This introduces no great disadvantage if samples must be loaded by hand into the counting assemblies. It is conceivable, however, that spot samples could be supplied automatically from a plant by means similar to those used for the irradiation of samples in reactors, whereby samples are transported rapidly over large distances between analytical laboratories and reactor channels using pneumatic tubes. If such a facility were made available between the processes and analytical laboratory, it would be most desirable if remote control of instrument parameters were to be possible from the common data-recording station.

Such a scheme would overcome the two major difficulties which beset on-line techniques and which have so far failed to be accommodated, i.e. on-line environment and contamination build-up on sample vessels, since in this scheme the sample would be divorced from its original environment and the spot sample vessel could be decontaminated before re-use or, if inexpensive, discarded after use.

It might then be a subsequent development to connect the data recorders on-line to a computer which not only exercises control over the parameters of the various counting modules to be appropriate to the sampling programme, but could in fact modify the sampling programme to comply with the current state of the process. For example, if a deviation from flow-sheet values became apparent to the computer, it could increase the sample rate in the appropriate section of the plant. With normal counting techniques an increase of sample taking would either create a demand for more counting channels or reduce the time of and therefore the accuracy of, a measurement affected by the divergency. There would be a clear case in such circumstances for applying the sequential analysis technique [6] to the counting channels for those samples arising from the unaffected area of the plant, thus conserving counting time, and divert the extra samples to the counting channels which would then become available.

7. SUMMARY

It is concluded that the effective quantitative control of a plant rests on a rapid efficient sample analysis under laboratory conditions. It is probable that future developments of "off-line" facilities rather than on-line instrumentation will be possible and more worthwhile.

The desirable characteristics of instrumentation for such a laboratory have been reviewed and an example demonstrates the value of unitized instrumentation and a comprehensive, flexible data-recording system. Possible future developments have been mentioned in data processing which could lead to a completely automatic service becoming available.

ACKNOWLEDGEMENTS

The author would like to express his appreciation of useful discussions on this topic with members of Electronics Division, Harwell, and in par-
ticular Mr. J.G. Cuninghame of Chemistry Division AERE for information concerning his work described in section 5.

REFERENCES


EVALUATION OF MEASUREMENT METHODS;
NUCLEAR SAFETY AND CRITICALITY CONTROL

(Session V)
HANDLING AND MEASUREMENT OF PLUTONIUM SHIPMENTS*

J. T. BYRNE
THE DOW CHEMICAL COMPANY, GOLDEN, COLO., UNITED STATES OF AMERICA

Abstract — Résumé — Аннотация — Resumen

HANDLING AND MEASUREMENT OF PLUTONIUM SHIPMENTS. The major forms in which power reactor plutonium can be shipped are metal, nitrate solution, and oxide. In this paper, recent studies on the measurement problems associated with handling and shipping plutonium are summarized. Factors affecting the stability of plutonium in these forms have been identified and current procedures evaluated.

Gas evolution, hydrogen ion consumption, and precipitation may occur in plutonium nitrate solutions. Conditions which lead to these effects have been defined. With proper control of these effects, nitrate shipments can be made with average shipper-receiver differences under 0.2%. The standard deviation of the difference can be kept under + 0.3%. Compositions in the range of 50-250 g Pu/litre, 2-10 N nitric acid, and up to 5000 ppm metallic impurities have been tested. Procedures for bulk measurements (both weight and volume), complete transfer, sampling, and analysis of plutonium nitrate in batches up to 10 litres are available.

The major contributor to shipper-receiver differences in shipments studied was the analytical determination of plutonium in solution. Many methods have been reported. However, interplant comparisons have shown biases between widely used methods. Smallest differences are observed when the same analytical method is used for both shipper and receiver measurements.

Plutonium oxide cans may pressurize during shipment if the oxide is low-fired, causing loss of material when the can is opened. However, properly prepared oxide can be shipped in batches of 2-12 kg with a measured loss of less than 0.2%. Procedures for blending, sampling and transferring have been tested. Thermal and radiation effects have been measured. Moisture adsorption and desorption on oxide must be controlled if shipper-receiver differences are to be minimized. Calorimetric assay of the plutonium can eliminate sampling errors and reduce analytical cost. Accuracy and precision of calorimetric and “wet” methods are comparable.

Metal shipments have presented little problem in measurements. Oxide formation due to self-heating and atmospheric corrosion is controlled by proper packaging. Analytical measurements can be based on direct assay of plutonium or on 100% minus impurities. Results on interplant sample exchanges show good agreement. The cost of shipping plutonium depends upon the form and size of the shipment. Factors affecting the shipping cost are evaluated.

MANIPULATION ET MESURE DU PLUTONIUM DESTINE A ETRE EXPEDIE. Le plutonium utilise dans un reacteur de puissance peut etre expedi essentiellement sous trois formes: metal, solution de nitrate et oxyde. L'auteur analyse brievement des etudes recentes consacrees aux problemes de mesure que posent la manipulation et l'expedition du plutonium. II definit les facteurs qui affectent la stabilité du plutonium sous ces formes et procede a une evaluation des methodes couramment utilisees.

Dans les solutions de nitrate de plutonium, il peut se produire une evolution du gaz, une consommation d'ions hydrogene et des precipitations. Les conditions qui determinent ces effets sont etudies dans le memoire. En combatant ces effets de facon appropriee, on peut expeditor du nitrate de telle sorte que l'ecart moyen entre la quantite expediee et la quantite reque soit inferieure a 0,2%. L'ecart type pour cette difference peut etre maintenu a ± 0,3%. Il a ete procede a des essais sur des solutions de 50 a 250 g de plutonium par litre, avec acide nitrique 2 a 10N et jusqu'a 5000 ppm d'impuretés metalliques. Il existe des methodes de mesure de la masse (poids et volume), les methodes de transfert, d'echantillonnage et d'analyse du nitrate de plutonium en lots pouvant atteindre jusqu'a 10 litres.

Les erreurs entre les quantites expediees et reques sont essentiellement dues au dosage analytique du plutonium en solution. De nombreuses methodes permettent de l'etablir mais des comparaisons effectuees entre plusieurs centrales font apparaître des differences systematiques entre les resultats obtenus a l'aide de methodes

* Work performed under USAEC Contract AT (29-1)-1106.
largely utilized. The errors are smaller when the same analytical method is used for measurements at the place of dispatch and receipt.

In the sealed containers containing plutonium oxide, the pressure may increase during transportation if the oxide has not been properly treated and this results in a loss of material when the container is opened. However, properly prepared oxide can be shipped in lots of 2 to 12 kg with a loss of less than 0.2%. Various methods of mixing, sampling, and transfer have been subjected to tests. The thermal and radiation effects have been measured. If you want to decrease the differences between the quantities dispatched and received, the adsorption and desorption of moisture on the oxide should be controlled. The calorimetric determination of plutonium allows the elimination of sampling errors and reduces the cost of analyses. The calorimetric and "wet" methods are comparable in terms of their accuracy and precision.

The deliveries of plutonium in the form of metal present few problems of measurement. An appropriate packaging prevents the formation of oxide due to self-heating and atmospheric corrosion. Analytical measurements can be based on either direct determination of plutonium or on subtracting impurities from the 100% content. The results of inter-plant exchange of samples are in good agreement. The costs of transportation of plutonium depend on the type and dimensions of the consignments. The author analyzes the factors that affect the cost of transportation.

ОБРАЩЕНИЕ С ГРУЗАМИ ПЛУТОНИЯ И ИХ ИЗМЕРЕНИЕ. Плутоний энергетических реакторов можно перевозить преимущественно в виде металла нитратного раствора, или оксид. Рассмотрены недавние исследования, связанные с обращением с плутонием при его перевозке: Установлены факторы, влияющие на устойчивость плутония в этих видах, и проведена критическая оценка существующей практики.

Нитратные растворы плутония могут выделять газы, поглощать ионы водорода и давать осадки. Выяснены условия, ведущие к таким результатам. При надлежащих мерах против такого рода эффектов при отправках нитрата можно свести до 0,2% разницу в количествах между отправителем и получателем. Можно добиться того, чтобы стандартные отклонения в этой разнице не выходили за пределы ±0,3%. Произведены испытания составов в диапазоне 50—250 г плутония на 1 л раствора, от 2 до 10N азотной кислоты и с металлическими примесями в количествах до 5000 частей на миллион. Сравниваются способы обмера (по весу и по объему), полного перевода из одного состояния в другое, выборки проб и анализа нитрата плутония в партиях до 10 л каждая.

В исследованных отправках главной причиной разницы между отправителем и получателем было аналитическое определение плутония в растворе. В этой связи сообщалось о многих методах. Однако межзаводские сравнения показали различия между широко используемыми методами. Наименьшая разница получается, когда отправитель и получатель используют при измерениях тем же самым аналитическим методом.

В случае изготовления окиси при низкотемпературном обжиге в контейнерах с оксидом плутония может появиться давление, что ведет к потере части материала при открытии контейнера. Тем не менее надлежащим образом изготовленную окись можно отправлять партиями 2—12 кг с измеренной потерей менее 0,2%. Описываются испытанные методы образования смесей, выборки проб и перевода из одного состояния в другое. Измерены тепловые и радиационные воздействия. Поглощение и выделение тепла в окиси должен быть под контролем, чтобы свести к минимуму разницу в показаниях прибора. Калиметрический способ определения количества плутония может помочь избежать ошибок при выборке проб и сократить стоимость производства анализов. Сравниваются надежность и точность калиметрического и "мокрого" методов.

Отправки в виде металла не вызывали значительных затруднений при измерении. С образованием окиси от самонагрева и атмосферной коррозии можно бороться, создав соответствующую конструкцию контейнера. Калиметрический анализ можно основывать или на непосредственном определении плутония, или на вычитании примесей из 100—процентного количества. Сообщаются результаты обмена пробами между отдельными установками. Стоимость перевозки плутония зависит от вида, в котором он отправляется, а также от величины груза. Приводится оценка факторов, влияющих на стоимость перевозки.

MANIPULACION Y MEDICION DE LOS ENVIOS DE PLUTONIO. El plutonio destinado a los reactores de potencia suele enviarse en forma de metal, de solución de nitrato o de óxido. En la memoria se resumen algunos estudios recientes sobre los problemas de medición asociados al transporte y a la manipulación del
plutonio, se determinan los factores que influyen sobre la estabilidad de ese elemento en las formas mencionadas y se evalúan los procedimientos corrientemente empleados para su medición.

En las soluciones de nitrato de plutonio puede producirse desprendimiento de gas, consumo de iones hidrógeno y precipitación. El autor expone las condiciones en que tales efectos se producen; si se les controla adecuadamente, se puede enviar nitrato sin que la diferencia entre las mediciones efectuadas por el remitente y el destinatario exceda de 0,2%. La desviación tipo respecto de esa diferencia es de ± 0,3% como máximo. Se han ensayado composiciones dentro de los siguientes límites: de 50 a 250 g de plutonio por litro de ácido nítrico 2-10 N y hasta 5000 partes por millón de impurezas metálicas. Existen diversos procedimientos de medición global (tanto en peso como en volumen) transvase completo, muestreo y análisis del nitrato de uranio en tandas de 10 l como máximo.

En los envíos estudiados, el factor que más ha influido en la diferencia entre los resultados de las mediciones efectuadas por el remitente y el destinatario ha sido la determinación analítica del plutonio en solución. Para esta determinación se conocen muchos métodos, pero comparaciones entre diversas plantas han puesto de manifiesto ciertas desviaciones sistemáticas entre algunos de los más empleados. Las diferencias mínimas se observan cuando el remitente y el destinatario emplean el mismo método analítico para efectuar sus mediciones.

El óxido de plutonio puede provocar presiones en los envases durante el transporte y esas presiones pueden ocasionar pérdidas de material al desenvasarlos. Pero, si se prepara adecuadamente, el óxido puede enviarse en partidas de 2 a 12 kg con una pérdida inferior a 0,2%. Se han ensayado procedimientos de mezcla, muestreo y transvase. Se han medido los efectos térmicos y radiactivos. Es necesario controlar la adsorción y la desorción de la humedad sobre el óxido si se desea reducir al mínimo la diferencia entre las cantidades expedidas y las recibidas. El ensayo calorimétrico del plutonio puede eliminar los errores de muestreo y reducir los costos de análisis. La exactitud de los métodos calorimétricos con la de los métodos «húmedos» son comparables.

El envío de plutonio plantea pocos problemas de medición. Un embalaje adecuado permite evitar la formación de óxido por autoencalentamiento y la corrosión debida a los factores atmosféricos. Las mediciones analíticas pueden basarse en el ensayo directo del plutonio o en el 100% menos las impurezas. Los resultados obtenidos con el intercambio de muestras entre diversas plantas están en buen acuerdo. El costo de los envíos de plutonio depende de su forma y de su importancia. En la memoria se evalúan los factores que influyen en dicho costo.

1. INTRODUCTION

1.1. Measurement requirements

Measurements of the material in plutonium shipments are necessary to meet accountability, safety, and process control requirements. The principal accountability requirement is to determine the amount of plutonium in the shipment. This could require a bulk measurement using either weight or volume methods. A plutonium assay is made on a sample of the shipment and the total weight of plutonium calculated. If the measurement of the sample is on a different basis (weight or volume) than the bulk measurement of the shipment, a density determination is required to convert to a common basis. For non-specific plutonium assay methods, it may be necessary to determine quantitatively those elements interfering in the assay.

An isotopic assay is sometimes required for accountability purposes. Since power reactor plutonium consists of a mixture of fissile, fertile, and parasitic nuclides, the value of the shipment is determined by the isotopic composition. The usual method for determining the isotopic composition of plutonium is by mass spectrometry.

The two major unsafe conditions that must be avoided in any shipment of plutonium are: (1) loss of containment of the plutonium and (2) rearrange-

1 If the calorimetric method of assay is applicable, no bulk measurement is required.
ment of the plutonium into a form that could be super-critical. Measurements important to containment are volatile impurity content of solids and concentrations of plutonyl (VI) and corrosive acid in liquids. The amount of heat generated by radioactive decay in a shipment may also be important in ensuring containment of the plutonium in a safe geometrical form. The heat generation can be predicted [1] through knowledge of the amounts and concentrations of radioactive isotopes (principally plutonium) in the shipping container.

Nuclear safety is ordinarily ensured by imposing a maximum limit on the amount of plutonium (80% of critical mass) or on the size (92% of critical dimension) of the shipping container. Measurements of the beta, gamma, and neutron radiation from the shipment may also be necessary to ensure that the radiation does not exceed the shielding capability of the shipping container.

The receiver of a plutonium shipment may impose additional specifications on the shipment to enable him to maintain good process control in his use of the material. These specifications may require determination of anionic and/or cationic impurities in the shipment, specific surface area, particle size, density, and acidity.

1.2. Purpose of this report

In this report, the problem of handling and measuring plutonium after chemical separation from fission products and uranium will be considered. A critical comparison will be made of different forms and methods of shipment. Past problems in handling and measuring plutonium shipments will be identified and areas for possible improvements in measurement accuracy, safety, and cost of shipping plutonium will be suggested.

2. PLUTONIUM SHIPPING EXPERIENCE

2.1. Form of plutonium in shipments

In the United States of America (and other countries) metal shipments have been used extensively. Metal provides a convenient shipping form if the plutonium exists in that form during some stage of its processing. A typical specification would require 99.5% plutonium purity.

For nuclear fuel applications, it is expected that shipments will consist principally of nitrate and oxide. Initially, the Nuclear Fuel Services Plant (West Valley, N.Y.) will ship plutonium as nitrate solution.

The USAEC has published specifications [2] for plutonium nitrate to be purchased by the US Government. These include a limit of 5000 parts of total impurity per million parts of plutonium. The nitrate solution acidity is limited to the range 2-10 M and the plutonium concentration to 50-250 g/litre.

The lower limit in nitric acid molarity is intended to minimize plutonium polymer formation. This limit necessitates concentration of the 0.3-0.6 molar acid anion exchange column product. As a result, the expected composition range for shipping solutions is 200-250 g Pu/litre and 3-6 molar nitric acid. If evaporation is used to concentrate the solution, a significant portion
of the plutonium is converted to the hexavalent state. Major metallic impurities expected are iron, chromium, nickel, uranium and titanium. Entrained organic material could include ion exchange resin, tributyl phosphate, hydrocarbon diluent, and degradation products of these materials.

Recent international shipments (USA to EURATOM; UK to France) have been in the form of plutonium oxide. It is anticipated that USAEC specifications for plutonium oxide purchase will be based on the assumption that the above nitrate solution may be converted to oxide without further purification.

### 2.2. Analytical methods

Most assay measurements on plutonium shipments have been made using a titration with ceric solution. The titration can be carried out using either volume or weight burettes for the addition of titrant. The endpoint can be detected either potentiometrically or photometrically. Interfering elements, such as iron, uranium, molybdenum, and vanadium, must be determined separately.

Plutonium nitrate solution samples must first be treated to remove the nitrate by fuming with sulphuric acid or by reaction with formic acid. The plutonium is then reduced to the trivalent state using liquid zinc amalgam, a lead reductor, a Jones reductor, or a titanous solution. The relative standard deviation of the ceric titration for plutonium nitrate samples is usually ±0.1-0.2%.

Oxide samples are dissolved in a nitric-hydrofluoric acid solution [3] or in a Wichers' bomb [4]. If necessary, refractory oxides first may be fused with ammonium bifluoride [3] or another flux.

The analytical method after dissolution is the same as for the nitrate solution. The relative standard deviation of plutonium determination in oxide dissolved in a nitric-hydrofluoric acid solution is usually less than ±0.1%.

One of the most convenient methods [5] for the determination of plutonium in metal consists of dissolving metal in hydrochloric acid and titrating with a ceric sulphate solution within a few minutes of dissolution. For some alloys, reduction with a lead or Jones reductor may be necessary. An indirect assay based upon subtraction of impurities from 100% is useful for oxidized samples if facilities are available for complete spectrographic and chemical analysis.

In a recent exchange of two 99.9% plutonium metal samples among four USAEC laboratories, the standard deviation was ±0.05%. The average results for multiple determinations by these laboratories are shown in Table I.

Standardization of ceric sulphate solution for plutonium titration can be done with arsenious oxide, sodium oxalate, pure plutonium metal, or with some plutonium compounds.

Plutonium compounds which have been evaluated for this purpose are dicaesium plutonium hexachloride [6] and plutonium sulphate tetrahydrate [7]. Pure metal standards are currently available [8] from the US National Bureau of Standards.
TABLE I
PLUTONIUM METAL SAMPLE EXCHANGE

<table>
<thead>
<tr>
<th>Laboratory</th>
<th>Sample No. 1</th>
<th>Sample No. 2</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>n&lt;sup&gt;a&lt;/sup&gt;</td>
<td>Aver. % Pu</td>
</tr>
<tr>
<td>A</td>
<td>16</td>
<td>99.94</td>
</tr>
<tr>
<td>B</td>
<td>16</td>
<td>99.94</td>
</tr>
<tr>
<td>C</td>
<td>24</td>
<td>99.89</td>
</tr>
<tr>
<td>D</td>
<td>16</td>
<td>99.95</td>
</tr>
<tr>
<td>Average</td>
<td>72</td>
<td>99.92</td>
</tr>
</tbody>
</table>

<sup>a</sup> Number of determinations run over a period of 3 months.

<sup>b</sup> Standard deviation.

Descriptions of other plutonium assay methods appear in a recent USAEC publication [9]. Comparisons of some of these methods have been made at Los Alamos [10] and at Aldermaston [11].

A recent method [12] that has unique advantages is based on calorimetric measurement. This method provides a non-destructive means of determining total plutonium content in sealed shipping containers of oxide or metal. Bulk measurement and sampling are not required since the plutonium content is directly related to the heat output from the container. Accurate isotopic analysis is required, however. The precision and accuracy of the method compare favourably with the ceric titration method. This method is particularly suited for insoluble and non-homogeneous oxide shipments.

2.3. Physical methods

2.3.1. Bulk measurements [13, 14]

The total weight of material shipped is determined by subtracting the tare weight of the container from the gross weight of the filled container. The net weight of the shipment can ordinarily be determined with an uncertainty much less than ±0.1%.

Volume measurements may be used for large amounts (greater than 10 l) of solution by transfer of the solution to a calibrated tank. Errors due to transfer line losses and evaporation may occur.
2.3.2. Sampling

When nitrate solution shipments are thoroughly agitated before withdrawing samples, it has been demonstrated [13] that the sampling uncertainty (relative standard deviation) is less than ±0.05%. On the other hand, sampling uncertainties of ±0.4% have been reported using in-line methods in a system in which samples are withdrawn during vacuum transfer of 10 litres of nitrate solution. The larger error may have been due to changes in the concentration due to evaporation caused by the vacuum transfer.

The use of a template for sampling of plutonium oxide has been recommended [14]. With a template, relative standard deviations due to sampling of ±0.1-0.2% have been reported.

The method used for metal sampling depends upon the form of the metal. For a shipment containing a large number (greater than 12) of metal "buttons", each button can be sampled in a different place. The sampling place is selected using a template. Both top and bottom of the button should be sampled. The sample is taken by drilling. A \( \frac{1}{4} \) in. diam. high-speed tool steel drill is recommended. A chlorinated hydrocarbon coolant (1,1,1-trichloroethane) can be used while drilling the sample. If carbon contamination is to be avoided, drilling can be done without coolant in an inert atmosphere. An iron determination should be made on an aliquot of the same solution used for the plutonium assay so that an accurate correction can be made to account for contamination from the drill bit.

2.3.3. Transfer

Solutions of plutonium nitrate can be transferred in and out of shipping containers using either gravity or vacuum [13]. The gravity method is preferred since errors may be introduced through evaporation of the solution during vacuum transfer. When 3 litres of plutonium nitrate solution are poured from a polyethylene bottle (5-in. diam.), a heel of less than 0.2% remains. This heel can be removed quantitatively with five 100-ml washes of 7 M nitric acid.

Plutonium oxide powder (particle size less than 1 \( \mu \)m) can be transferred from a metal can containing 2 kg of oxide with a loss of less than 0.02%. Serious loss of oxide powder could result if the can is pressurized [14]. Losses may occur from metal shipments if oxide dust is not carefully brushed from the shipping container into the receiving vessel. Oxidation is minimized if the metal is canned in a dry atmosphere.

2.4. Shipping packages

The present regulation applying to shipment of fissile material for the USAEC and its contractors is given in USAEC Manual Chapter 0529. This manual chapter, issued on 11 May 1964, is entitled "Safety Standards for the Preparation of Radioactive and Fissile Material for Transportation". The regulation requires a criticality evaluation of the package before and
after structural integrity tests. The chapter defines three modes of shipments as follows:

Class I: An infinite number of undamaged packages in any arrangement will remain sub-critical and 250 such packages after accident test conditions will be sub-critical.

Class II: The number of packages is limited to the lesser of two calculated values; one based on the undamaged package and the other based on the package after accident test conditions.

Class III: Administrative control by the shipper ensures the safety of the shipment.

A proposed United States Federal Regulation, 10 CFR 71, which will govern shipments by licensees was issued for comment on 16 November 1964. Some differences exist between the contractor regulation in Manual Chapter 0529 and the proposed licensee regulation. However, the classes of the shipments are essentially the same.

The IAEA regulations governing shipment of fissile materials include provisions similar to those in the US proposals.

2.4.1. Metal

A "birdcage" container has been extensively tested [15] for use in Class II metal shipments. The metal (2 kg) is hermetically sealed in a tin-plated can. If this operation is done in a glovebox with a dry atmosphere (less than 5°F dew point), oxidation of the metal during shipment is minimized. The can is placed in a pressure vessel. Up to 6 kg of plutonium metal can be shipped in a package.

2.4.2. Nitrate solutions

Nitrate solutions can be shipped in polyethylene bottles inside stainless steel pressure vessels. If serious corrosion of the pressure vessel by the nitrate solution is not expected, the polyethylene bottles can be omitted. The pressure vessel is mounted in a shipping package so as to provide a safe spacing from other containers. Some packages that have been approved by the USAEC for Class I shipment utilize wood (white oak) between the pressure vessel and the outer container to provide structural strength, thermal insulation, and neutron moderation [16]. A cadmium liner absorbs the moderated neutrons originating from external sources.

The outer container in these packages consists of one or two 55-gal drums. The pressure vessel should contain a vent valve so that the receiver can release pressure safely. A similar shipping package proposed [16] for Class II shipments utilizes a "birdcage" structure to position the pressure vessel inside the 55-gal drum. Vermiculite is used to provide thermal insulation. A 5-litre UK shipping container [17] uses Greenheart timber instead of white oak.

2.4.3. Oxide

Packages for shipping up to 12 kg of plutonium as oxide have been designed and tested [18]. The plutonium oxide is contained in 4-in. diam. by
4-in. high tin-plated cans which are hermetically sealed. This can, in a plastic bag, is then sealed in a standard No.3 tin-plated can. Three such No.3 cans can be shipped in one pressure vessel. The pressure vessel is shipped inside a 30-gal steel drum. For a Class I shipment, the pressure vessel is mounted in white oak. For Class II shipment, it is mounted in a tubular metal "birdcage" and vermiculite is added to fill the voids.

2.5. Metal shipments

At Rocky Flats, plutonium metal has been received in the form of reduction "buttons". In most cases, shipper-receiver differences of less than 0.1% are achieved.

Data tabulated from recent shipments involving over 100 sets of measurements in a period of one year showed average shipper-receiver differences of +0.007 and -0.013% between Rocky Flats (receiver) and two separate shippers. In both cases, the standard deviation of the differences was ±0.11%. For these shipments, the average proportion of the plutonium collected as oxide by the receiver was 0.06% (standard deviation ±0.07%).

2.6. Nitrate solution shipments

In 1957 the USAEC Rocky Flats Plant (Colorado) undertook to measure the uncertainties and to check for biases in its own measurements. This was done in an effort to resolve continuing shipper-receiver discrepancies on shipments of plutonium nitrate from Hanford to Rocky Flats. During the year in which these controls were maintained, the average shipper-receiver difference was 1.35% of the amount of plutonium per batch. The shipper's (Hanford) value averaged higher than the receiver's (Rocky Flats). The average shipper-receiver difference was statistically significant. It could not be accounted for by the analytical uncertainties measured at Rocky Flats.

Recently, systematic studies of the plutonium nitrate [13] and plutonium oxide [14] measurement problems have been carried out at Rocky Flats. Measurement procedures and shipping containers that could minimize shipper-receiver differences were developed. After the containers and procedures were developed, actual interplant shipments were carried out between Hanford and Rocky Flats using the recommended procedures and containers. In this way it was demonstrated that measurements could be made with accuracy and precision of better than ±0.3%, provided correct procedures were used. In this study, several factors causing instability in the shipping solutions were identified.

Gas (mostly oxygen) evolution was observed in plutonium nitrate solutions that contain plutonyl (VI) due to reduction of plutonyl (VI) by products of its own alpha radiation. The rate of the reduction is dependent on solution composition. Approximately one hydrogen ion is consumed for each ion of plutonyl (VI) reduced, thereby decreasing the acid concentration.

Gas evolution from solutions containing plutonyl (VI) ion causes pressure build-up in sealed containers. This pressure build-up can be avoided by letting the solution age for a pre-determined time before sealing the container. The exact time required would have to be determined experimentally since it depends on the amount of plutonyl (VI) in the solution as well as on
the concentrations of other components. The rate of plutonyl (VI) reduction could probably be increased measurably by adding a chemical reductant to the solution.

Even with gas formation, and in some cases slight precipitate formation, shipper-receiver differences of less than 0.3% were achieved with solutions whose plutonium composition meets the USAEC specifications [2]. A major contributor to shipper-receiver differences was the analytical determination of plutonium in solution. A suitable method has been demonstrated, but interplant comparisons have shown a significant analytical bias between two widely-used and recommended methods. When the same analytical laboratory did all analyses for an interplant shipment, shipper-receiver differences less than 0.2% were obtained.

2.7. Oxide shipments

In 1964 39 lots of plutonium oxide, totaling 45 kg Pu, were shipped from England to France [19]. Careful measurements of samples of the shipment were made at Windscale and samples were sent with the shipment so that comparative measurements could be made in France. It was concluded that the shipper-receiver difference of -0.21% was acceptably small. A correction of +0.11% was made to the French results for moisture adsorption by the plutonium oxide samples.

Also in 1964 a shipment of several hundred kilograms of plutonium oxide from Hanford to EURATOM was begun. Data are not yet available to compare the shipper-receiver agreement on measurements for this shipment.

In a recent study [14] an average shipper-receiver difference of +0.14% with a standard deviation of ±0.23% was reported for 12 interplant shipments between Rocky Flats and Hanford of oxide prepared from plutonium oxalate or plutonium peroxide. Using analytical laboratory results from the same laboratory only, the average shipper-receiver difference for these same interplant shipments was +0.10% with a standard deviation of ±0.18%. The positive shipper-receiver difference (+0.14%) for the interplant shipments was explained by the fact that all receiving samples were analysed in an atmosphere of lower humidity than that in which the oxide was initially prepared. Consequently, the analyses showed a progressive increase of plutonium concentration because of loss of adsorbed moisture. The interplant shipments also included two batches of oxide prepared by denitrification of plutonium nitrate at 400°C. Results from this material were erratic, indicating heterogeneity and instability because of incomplete calcination. The following specifications have been recommended to give accurate measurement and safe shipment of plutonium oxide:

1. The oxide shall not contain plutonium metal or pyrophoric impurities.
2. The weight per cent of volatile components measured by loss of weight in 2 h at 200°C in a dry (less than 5°F dew point) dynamic atmosphere shall not exceed 1%.
3. The weight per cent of volatile components measured by additional loss of weight in 2 h at 800°C in a dry (less than 5°F dew point) dynamic atmosphere shall not exceed the quotient of 0.5 ÷ (density in grams plutonium per cubic centimetre container) (calorie output in calories per second-gram plutonium) (days of unvented storage intended).
(4) The thermal output of the total plutonium in the shipping package shall not exceed 12 calories per second.
(5) The plutonium content of the primary container volume shall not exceed 4.0 g Pu/cm³.
A study of the properties of plutonium oxide which could influence shipment specifications has been reported [20].

3. CRITICAL COMPARISON OF RESULTS

3.1. Changes during shipment

The major material changes occurring in a plutonium shipment are: (1) decrease of metal weight through oxidation, (2) decrease of solution weight and increase of plutonium concentration through evaporation, (3) decrease of oxide weight and increase of plutonium concentration due to moisture desorption from oxide (moisture adsorption could cause a weight increase and a plutonium concentration decrease), and (4) "heels" left in container after transfer of bulk material. Typical measured magnitudes of these changes are shown in Table II.

The largest changes in Table II are those associated with concentration changes due to solution evaporation and desorption of moisture from oxide. These changes also show the greatest variability. It is clear that accurate measurement of plutonium concentration in solution and in oxide is a critical factor in minimizing shipper-receiver differences.

3.2. Measurement precision

The measurement precisions reported for plutonium metal, nitrate, and oxide are compared in Table III. The bulk measurement (weight or volume) uncertainty for these three forms of shipment is not significant. The nitrate measurement seems to be limited by the analytical determination of plutonium in solution. The oxide measurement is limited by sampling. Control of moisture in the atmosphere in which the oxide is handled may decrease the sampling error. If either of these variables were improved, the uncertainties of nitrate or oxide shipment could be close to that of the metal shipment. These uncertainties can be decreased by multiple measurement. To achieve the precisions for shipper-receiver difference shown in Table II, duplicate analyses were run on four shipper and four receiver nitrate samples. For the oxides, duplicate analyses of three samples were used.

3.3. Measurement accuracy

The major factors that have been known to cause measurement bias in recent studies are:

(1) Bias in plutonium determination in nitrate solution: average differences of nearly 1% in plutonium concentration were obtained for 12 inter-plant shipments between Hanford and Rocky Flats. The difference was virtually eliminated when all samples were analysed by the same laboratory. The reason for this difference and for other reported discrepancies between
<table>
<thead>
<tr>
<th>Form of shipment</th>
<th>Metal</th>
<th>Nitrate [13]</th>
<th>Oxide [14, 19]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Shipper</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Receiver</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Number of containers</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>% weight change</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>% Pu concentration change</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>% heel</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>% shipper-receiver difference</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

**Table II**

**TYPICAL MATERIAL CHANGES DURING PLUTONIUM SHIPMENTS**

<table>
<thead>
<tr>
<th>Form of shipment</th>
<th>Metal</th>
<th>Nitrate [13]</th>
<th>Oxide [14, 19]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Shipper</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Receiver</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Number of containers</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>% weight change</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>% Pu concentration change</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>% heel</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>% shipper-receiver difference</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

- **Variability of changes (% Pu)**
  - Standard deviation of weight change: ±0.11
  - Standard deviation of concentration change: ±0.07
  - Standard deviation of heel change: ±0.01
  - Standard deviation of shipper-receiver difference: ±0.11

- **Change measured in 15-g PuO₂ samples shipped in separate containers.**
- **Rocky Flats analyses used for all calculations.**
- **"Heel" is material remaining in container after transfer. +0.06% means 0.06% of plutonium shipped remains in container after bulk transfer.**
- **Difference reported for 44.9-kg shipment (39 batches).**
### TABLE III
TYPICAL MEASUREMENT PRECISION FOR PLUTONIUM SHIPPING FORMS

<table>
<thead>
<tr>
<th>Measurement operation</th>
<th>Standard deviation (% Pu)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Weight of volume</td>
<td>&lt;0.01</td>
</tr>
<tr>
<td>Sampling</td>
<td>a</td>
</tr>
<tr>
<td>Pu concentration</td>
<td>0.05</td>
</tr>
</tbody>
</table>

a = not measured.

Laboratories in plutonium nitrate analysis is not known. Nitrate solution can contain plutonium in four valences and in hydrolyzed and polymerized species. Changes can occur from chemical and radiolytic reaction mechanisms. An interlaboratory comparison programme is currently being sponsored by the USAEC to evaluate methods for determination of plutonium in nitrate solution.

(2) Moisture exchange between plutonium oxide and the atmosphere: in the recent Rocky Flats study and in the UK-French oxide shipment, the major cause of measurement bias was the adsorption or desorption of water from oxide samples. The magnitude of this bias was 0.14% in the Rocky Flats-Hanford interplant studies. The bias can be eliminated by using the calorimetric measurement method which does not require either sampling or exposure of oxide to the atmosphere between shipper and receiver measurements. Presumably, the bias could also be eliminated by standardization of shipper and receiver glovebox humidity. The moisture problem is greater with plutonium oxide that contains more than 1% moisture or 4% other volatile impurities. Preparation of oxide to meet the specifications given in section 2.7 yields a sufficiently stable material.

For maximum accuracy, other factors that can cause bias must also be carefully controlled. These include proper bulk measurement calibration, representative sampling, complete transfer of material, and analytical standardization.

### 3.4. Economics of shipment

Table IV summarizes the major cost components for shipping different forms and different amounts of plutonium. Plutonium metal is the most economical shipping form now known. However, there would be a serious economic penalty in converting nitrate to metal solely for shipping convenience. The USAEC has published a cost of $1.50 per gram ($1500 per kilogram) for converting nitrate solution to metal. The same argument ap-
TABLE IV
ESTIMATED PLUTONIUM SHIPPING COSTS
(Class 1, 2000 miles, rail)

<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>kg Pu/package</td>
<td>2</td>
<td>6</td>
<td>6</td>
</tr>
<tr>
<td>Container cost * 25^b</td>
<td>$30/kg Pu</td>
<td>$5/kg Pu</td>
<td>$1/kg Pu $1/kg Pu</td>
</tr>
<tr>
<td>Filling</td>
<td>44</td>
<td>20</td>
<td>20</td>
</tr>
<tr>
<td>Analysis</td>
<td>23</td>
<td>40</td>
<td>15</td>
</tr>
<tr>
<td>Transport</td>
<td>66</td>
<td>6</td>
<td>6</td>
</tr>
<tr>
<td>Receiving</td>
<td>37</td>
<td>19</td>
<td>23</td>
</tr>
<tr>
<td>Total $/kg Pu</td>
<td>$200/kg Pu</td>
<td>$40/kg Pu^a</td>
<td>$86/kg Pu $65/kg Pu</td>
</tr>
</tbody>
</table>

^a Estimated cost for a 200 to 300-litre shipping container (not yet designed).
^b Container life of 25 shipments assumed.

plies to the conversion of nitrate to oxide for shipping convenience. This conversion would cost over $100 per kilogram. If the shipper can produce an oxide that is directly usable by the receiver, the oxide form of shipment becomes most economical.

The development of large shipping containers for plutonium nitrate could give the nitrate form an economic advantage. The nitrate would also have an advantage in situations where the processor and/or the receiver do not desire to convert a nitrate solution to oxide, or where the oxide needed for processing purposes was not compatible with that required for safe shipment and accurate measurement.

The unit cost of shipping plutonium is within a factor of two of $100 per kilogram. This is the same range as the anticipated cost of fabricating plutonium into fuel elements for fast reactors (~20% Pu - 80% U). At the USAEC price for plutonium of $10 000 per kilogram (for $^{239}$Pu and $^{241}$Pu), the shipping cost is about 1% of the plutonium value. Shipping can be a significant factor in the economics of using plutonium, and even small reductions in unit shipping cost can be important in commerce.

4. CONCLUSIONS

4.1. Present status of plutonium shipping

Plutonium can be shipped as metal, nitrate, or oxide with measurement uncertainties less than 0.1-0.2% if proper procedures are used. The most
critical procedures are those related to analytical determination of plutonium (in nitrate solution) and exposure of plutonium oxide and metal to atmospheric moisture. Instability can be caused by hexavalent plutonium in nitrate solutions and by incomplete calcination of plutonium oxide.

Unit shipping costs are approximately $100 per kilogram (within a factor of two). The receiver’s process requirement usually determines the shipping form. Shipping costs can be a major factor in the recycle of plutonium in power reactors.

4.2. Recommendations for improved plutonium shipping

Three main areas of investigation appear to be fruitful if the safety, measurement precision, and cost of plutonium shipping are to be improved.

(1) The safe shipping of large quantities of plutonium nitrate solution requires a better understanding of the radiation chemistry in the solution. The mechanism of radiolysis in plutonium nitrate solutions is not well understood. The radiolysis mechanism is closely related to the gas production phenomena which presents a possible means of loss of containment.

(2) The determination of plutonium in nitrate solutions is not as precise as that of plutonium in solid materials. If the advantages of shipping plutonium as a nitrate solution are to be fully realized, a better understanding, and an improvement in the precision, of the methods for determining plutonium in nitrate must be achieved.

(3) Because of the strong dependence of unit shipping costs on the amount of plutonium in the container from which a sample must be taken, one of the most obvious directions for reduction of unit costs is to develop larger solution containers. The larger shipping container for plutonium nitrate solution presents serious engineering problems in structural integrity and containment. To surmount the nuclear safety hazards involved, it is necessary to utilize criticality poisons or to go to geometrically-safe containers.

ACKNOWLEDGEMENTS

Much of the work summarized in this report was carried out at Rocky Flats by R.L. Delnay, W.E. Domning, F.J. Milner, J.D. Moseley, and F.L. Oetting, under the sponsorship of S.C.T. McDowell, USAEC Division of Nuclear Materials Management. R.J. Sloat (General Electric Company) co-ordinated the interplant shipments with Hanford.

REFERENCES

Y. MOTODA: In Table IV you refer to rail shipment costs. Have you any idea what the cost of ocean transport would be, and whether it might compare favourably with railway rates?

J.T. BYRNE: Certain studies are in fact being carried out in the United States on water transport of irradiated fuel elements. I understand that in some conditions water transport costs can be lower than rail costs, but I have no exact information on the subject.

J.J. DOWNING: Can you give us the dimensions of the 50-kg plutonium container?

J.T. BYRNE: One large plutonium nitrate shipping container that has been proposed¹ has over-all dimensions of approximately 48-in. diam. X 48-in. height. The Pu(NO₃)₄ solution is contained in an annulus 2 in. wide. Other large containers have been designed with dimensions of about 30 in. diam. X 60-70 in. length. A nuclear poison is included in one container of this size.

G.A. WELCH: You refer to special measures which have been used to reduce shipper-receiver differences of plutonium nitrate from about 1.4% to 0.3%. Could you say what those special measures are, and what the two widely-used analytical methods are which yielded a significant discrepancy?

¹ Reference [16].
J.T. BYRNE: The procedures which save shipper-receiver differences of 0.3% are described in USAEC Report RFP-436\textsuperscript{2}. The most important source of bias seems to lie in the use of different analytical methods for determining plutonium in shipper and receiver laboratories. The two methods used for analysis in the study cited above were X-ray absorption and a volumetric method using cerium sulphate titration and photometric endpoint detection\textsuperscript{3}. Both are described in USAEC Report TID-7029\textsuperscript{4}. It was not determined which of these methods was in error.

J. SCHMETS: What are the specifications for fission-product activity in plutonium? Are they imposed by the reprocessing plant, by the user or by official regulations?

J.T. BYRNE: The USAEC specifications for purchase of plutonium nitrate solution are given in the US Federal Register for 25 March 1965 (Vol. 30, p. 3886). They include a limit on beta and gamma activity but none on fission products as such.

S.N. CEJA: What are the heat-removal problems associated with the shipping containers described in your paper?

J.T. BYRNE: The containers for plutonium nitrate solutions present no such problems. The heat rise in plutonium oxide containers can be predicted from the experimental data given in USAEC Report RFP-418\textsuperscript{5}. A temperature increase of about 100°C is expected in the plutonium oxide containers proposed in USAEC Report RFP-501\textsuperscript{6}.

W.J. WRIGHT: You mentioned that there are certain difficulties in concentrating \(\text{Pu(NO}_3\text{)}_2\) for shipment. Does this mean that \(\text{PuO}_2\) is more likely to be the form adopted for large-scale shipments?

J.T. BYRNE: \(\text{PuO}_2\), prepared by calcination at a temperature of 400°C or higher, appears to be a safe and economical form for shipment. There will undoubtedly be situations where the cost of converting nitrate solutions to oxide cannot be justified, and the nitrate solution would then be preferred for simple economic reasons.

\textsuperscript{2} Reference [13].
\textsuperscript{3} See Analyt. Chem. 34 (1962) 346.
\textsuperscript{4} Reference [3].
\textsuperscript{5} Reference [1].
\textsuperscript{6} Reference [18].
STATISTICAL CONTROL OF MEASUREMENT QUALITY

C. A. BENNETT
BATTLEF MEMORIAL INSTITUTE, RICHLAND, WASH., UNITED STATES OF AMERICA

Abstract — Résumé — Аннотация — Resumen

STATISTICAL CONTROL OF MEASUREMENT QUALITY. Effective nuclear materials management, and hence design and operation of associated material control systems, depend heavily on the quality of the quantitative data on which they are based. Information concerning the reliability of the measurement methods employed is essential to both the determination of data requirements and the evaluation of results obtained.

Any method of analysis should be (1) relatively free from bias and (2) reproducible, or, in more usual terminology, precise.

Many statistical techniques are available to evaluate and control the reproducibility of analytical results. Economical and effective experimental designs have been developed for the segregation of different sources of measurement error. Procedures have been developed or adapted for use in maintaining and controlling the precision of routine measurements. All of these techniques require that at least some measurements must be duplicated, but duplication of all measurements can be justified only when the detection of every gross error, or mistake, is extremely important.

Three types of measurement bias can be considered: (1) bias relative to a standard, (2) bias relative to prior experience, and (3) bias relative to a group. The first refers to the degree to which the measurements obtained deviate systematically from some "standard" which is unbiased either (1) by definition, or (2) because all known sources of bias have been removed. The second is concerned with the presence of systematic differences over a period of time. The third type of bias concerns the relationship between different physical entities or individuals at a given time. Recent developments in statistical methodology applicable to the evaluation of all three types of bias are discussed. Examples of the use of the statistical techniques discussed on Hanford data are presented.

CONTROLE STATISTIQUE DE LA QUALITE DE LA MESURE. La gestion efficace des matières nucléaires et, par conséquent, la conception et le fonctionnement des systèmes de contrôle de ces matières dépendent beaucoup de la valeur des données quantitatives sur lesquelles ils sont fondés. Il importe donc de connaître le degré de précision des méthodes de mesure employées, afin de déterminer les exigences en matière de données et d'évaluer les résultats obtenus.

Toute méthode d'analyse doit être 1. relativement exempte d'erreur systématique, 2. reproductible, autrement dit précise.

On dispose de nombreuses méthodes statistiques pour évaluer et contrôler la reproductibilité des résultats des analyses. On a mis au point des moyens économiques et expérimentaux pour séparer les diverses sources d'erreurs de mesure. On a créé ou adapté des méthodes permettant de maintenir et de contrôler la précision des mesures courantes. Toutes ces techniques exigent qu'au moins un certain nombre de mesures soient faites deux fois; mais la répétition de toutes les mesures n'est justifiée que lorsqu'il est extrêmement important de détecter toute erreur grossière.

On peut considérer trois sortes d'erreurs systématiques dans les mesures: 1. l'erreur par rapport à un étalon, 2. l'erreur par rapport aux résultats de mesures précédentes, 3. l'erreur propre à un groupe. La première est la déviation systématique des mesures obtenues par rapport à un «étalon» exempt de déviation, soit par définition, soit parce que l'on a supprimé toutes les causes connues de déviation. La deuxième concerne des différences systématiques observées sur un certain laps de temps. La troisième concerne le rapport entre différentes entités ou individualités physiques à un moment donné. L'auteur expose les moyens les plus récents de la méthodologie statistique applicables à l'évaluation de chacune de ces trois sortes d'erreurs systématiques. Il donne des exemples d'utilisation des méthodes statistiques appliquées aux données concernant le réacteur de Hanford.
СТАТИСТИЧЕСКИЙ КОНТРОЛЬ КАЧЕСТВА ИЗМЕРЕНИЙ. Эффективный контроль и учет ядерных материалов и, следовательно, организация и действие связанных с этим систем контроля за материалами, в значительной степени зависит от качества количественных данных, на которых они основаны. Информация относительно надежности применяемых методов измерений необходима как для определения потребностей в данных, так и для оценки полученных результатов. Почти любой метод анализа можно было бы считать хорошим, если бы он 1) был относительно свободен от систематических ошибок и 2) был воспроизводимым, или, употребляя более принятую терминологию, точным.

Для оценки и контроля за воспроизводимостью аналитических результатов имеется много статистических методов. Были разработаны экономичные и эффективные экспериментальные конструкции для выделения различных источников ошибок измерений. Были разработаны или приспособлены процедуры для поддержания и контроля за точностью повседневных измерений. Все эти методы требуют дублировать по крайней мере некоторые измерения, однако дублирование всех измерений может быть оправдано только в том случае, когда обнаружение каждой грубой ошибки или потенциальной ошибки имеет исключительную важность.

Можно учитывать следующие три вида ошибок в измерениях: 1) систематическая ошибка, относящаяся к эталону, 2) систематическая ошибка, относящаяся к предыдущему опыту и 3) систематическая ошибка, относящаяся к группе. Первая ошибка касается степени, с которой полученные измерения систематически отклоняются от предельного "эталона", который не имеет ошибок, либо 1) согласно определению, либо 2) потому что устранены все известные источники ошибок. Вторая ошибка связана с наличием систематических расхождений того или иного периода времени. Третий вид ошибок касается связи между различными свойствами или индивидуумами в данное время. Обсуждаются недавние достижения в области статистической методологии, применяемой к оценке всех трех видов ошибок. Приводятся примеры использования статистических методов на основе данных Хэнфорда.

CONTROL ESTADISTICO DE LA CALIDAD DE LAS MEDICIONES. La eficacia de la administración de los materiales nucleares y, por tanto, la organización y el funcionamiento de los sistemas de control dependen, sobremanera de la exactitud de los datos cuantitativos en que están basados. La información sobre la seguridad de los métodos de medición empleados es esencial tanto para determinar las necesidades en lo que respecta a los datos como para evaluar los resultados obtenidos.

Todos los métodos de análisis deben estar relativamente exentos de desviación y ser reproducibles o, por decirlo de una manera más corriente, precisos.

Para evaluar y controlar la reproducibilidad de los resultados de los análisis se dispone de varias técnicas estadísticas. Se han ideado procedimientos experimentales eficaces y económicos para descubrir el origen de los errores. También se han ideado o adaptado procedimientos para mantener y controlar la exactitud de las mediciones corrientes. Todas esas técnicas requieren la repetición de algunas mediciones, pero la repetición de todas ellas sólo está justificada cuando es de suma importancia detectar todos los errores serios.

En las mediciones cabe considerar tres tipos de desviación o inclinación viciosa: 1) desviación respecto de un patrón aceptado, 2) desviación respecto de experiencias anteriores y 3) desviación respecto de un grupo. El primer tipo se refiere el grado en que los valores obtenidos se separan sistemáticamente de un patrón que está exento de desviaciones ya sea por definición ya sea por haberse eliminado todas las causas de desviación conocidas. El segundo tipo se refiere a la presencia de diferencias sistemáticas después de un determinado período de tiempo. El tercer tipo concierne a las relaciones entre distintas entidades físicas en un momento determinado. En la memoria se analizan algunos procedimientos recientes de metodología estadística que pueden servir para evaluar esos tres tipos de desviación. Se dan, además, algunos ejemplos del empleo de esas técnicas estadísticas con datos obtenidos en Hanford.

1. INTRODUCTION

The design and operation of material control systems, which are in turn essential to effective nuclear materials management, depend heavily on the quality of the quantitative data on which they are based. Information concerning the reliability of the measurement methods employed is necessary both to determine data requirements and to assess the validity of derived
indices such as process shifts and material unaccounted for. This paper is concerned with the use of statistical analysis to evaluate and/or control the accuracy and reliability of measurement methods.

2. STATISTICAL APPROACH TO MEASUREMENT QUALITY

The simplest statistical model of a measurement process can be written in the form

\[ x_i = \mu + \epsilon_i \]

where \( x_i \) is the actual measurement obtained; \( \mu \) is a fixed constant identifying some "true" characteristic of the item being measured (such as temperature, presence, concentration, weight); and \( \epsilon_i \) is the "error" or "noise" of measurement associated with the \( i \)th measurement. Generally \( \mu \) is the quantity desired, and the \( \epsilon_i \) are deviations from the desired result; i.e. \( \mu \) represents the signal and the \( \epsilon_i \) the noise. In a more general model, both \( \mu \) and the \( \epsilon_i \) may be complex. For example, \( \mu \) may depend on some associated characteristic of the item being measured, in which case we have a regression model; or \( \mu \) itself may vary randomly from item to item, in which case we can speak of product variability as opposed to measurement error; or \( \mu \) may vary with time, in which case we might speak of process trends or process shifts. Similarly, the "error" \( \epsilon_i \) may be a combination of several errors arising from different sources. An example of this situation is considered in section 3.

The \( \epsilon_i \) are generally considered to be random variables or combinations of random variables. A random variable is a quantity whose behaviour is predictable only en masse in the sense that any individual value is chosen at random from a range of possible values in accordance with a given distribution of the probability of any particular value. Two important characteristics of any random variable are its expected value, or long-term average, and its variance, which is the expected value of the squared deviation from the expected value and measures the expected variability in individual values of the random variable. If we designate the expected value of \( \epsilon_i \) by \( E(\epsilon_i) = \delta \), then the variance of \( \epsilon_i \) is \( \sigma^2_\epsilon = E(X_i - \delta)^2 \). \( \delta \) is the bias, or systematic error, in the measurement. If the errors of measurement \( \epsilon_i \) are such that \( \delta = 0 \), the method is unbiased, i.e. on the average it will give the correct answer. If \( \sigma^2_\epsilon \) is small, the method is precise, i.e. on the average the error will not deviate very far from its expected value, or, in the case of an unbiased measurement, from zero. Obviously in the case of precise measurements, the sum of two errors will also be small, so that two different measurements will differ only by a small amount.

With respect to both precision and bias, it is the job of the statistician to define and measure the extent to which they exist in the measurements under consideration and to identify if possible the nature of the sources from which they arise. The improvement of precision (except to the extent to which it can be improved by replication) and the elimination of bias are primarily technical matters in the field of application being considered. However, in practice the greatest success will be accomplished if the use of
statistical techniques is intertwined with the technical development in such a fashion that each reinforces the other. Neither the statistician nor the scientist can achieve optimum results by himself and each must recognize the contribution of the other.

Note that the use of the term "accuracy" has been deliberately avoided in the above discussion. One of the reasons for this is that it is difficult to give a quantitative or even semi-quantitative definition of what is meant by accuracy. In a qualitative sense it refers to the degree to which a measurement gives an answer which is different from the "true" value. One difficulty with the quantitative definition arises because these deviations may be either random or systematic, i.e. they may be due either to the presence of bias or to a lack of precision. The other and more fundamental difficulty is the definition of what is meant by a "true" value. A little philosophical thought indicates that in many instances this value is defined only by the process itself, i.e. the measurement under consideration may be "accurate" by definition. We are then concerned only with whether or not the deviations are systematic or random. In other cases, such as the measurement of fundamental constants, a true value may in fact exist and in this case the definition of accuracy as some appropriate combination of precision and bias may have meaning. It is only in this situation that it makes sense to talk of an absolute bias. In most situations considered in section 4 we will be concerned with detecting biases relative to a defined quantity.

3. EVALUATION AND CONTROL OF PRECISION

3.1. Estimation of errors from repeated measurements: variance components

The simplest method of estimating measurement error is from repeated measurements. Every chemist is familiar with the use of duplicate results as a measure of precision and a check against gross errors or mistakes. The mean deviation or mean square deviation of replicate results can be used to estimate the variance, which, as indicated above, is a measure of the precision. Simple extensions of the duplication of measurement results can be used to evaluate individual sources of measurement error as well as product variability.

The data shown in Table I (from Ref. [1]) are the results of a special study of the uranium content of two types of U₃O₈ "produced" by essentially the same process from two types of waste material. The lots of each type represent consecutive processing, and the sampling and analytical techniques used were identical. Two distinct samples were taken from each lot by a thief technique, and after additional preparation submitted to the laboratory, where two different chemists did independent duplicate determinations of the metal content in per cent by weight. The pairs of chemists assigned were not necessarily the same, nor necessarily distinct, depending on chance assignment in the laboratory. Estimates of the variance components obtained from these data by standard analysis of variance techniques for nested replication are given in Table II.
### TABLE I

Results of a Special Study on Consecutive Lots of Metallic Oxide

<table>
<thead>
<tr>
<th>Lot</th>
<th>Sample A</th>
<th></th>
<th></th>
<th></th>
<th>Sample B</th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Chemist 1</td>
<td>Chemist 2</td>
<td>Chemist 1</td>
<td>Chemist 2</td>
<td></td>
<td>Chemist 1</td>
<td>Chemist 2</td>
<td></td>
</tr>
<tr>
<td></td>
<td>1</td>
<td>2</td>
<td>3</td>
<td>4</td>
<td>1</td>
<td>2</td>
<td>3</td>
<td>4</td>
</tr>
<tr>
<td>Type 1</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1</td>
<td>84.1</td>
<td>84.0</td>
<td>84.3</td>
<td>84.3</td>
<td>84.1</td>
<td>84.0</td>
<td>84.1</td>
<td>84.1</td>
</tr>
<tr>
<td>2</td>
<td>84.1</td>
<td>84.0</td>
<td>84.0</td>
<td>83.9</td>
<td>84.2</td>
<td>84.2</td>
<td>83.7</td>
<td>84.6</td>
</tr>
<tr>
<td>3</td>
<td>83.5</td>
<td>83.5</td>
<td>83.4</td>
<td>83.6</td>
<td>83.4</td>
<td>83.3</td>
<td>84.0</td>
<td>83.5</td>
</tr>
<tr>
<td>4</td>
<td>84.2</td>
<td>84.2</td>
<td>84.2</td>
<td>84.3</td>
<td>84.1</td>
<td>83.7</td>
<td>84.1</td>
<td>84.6</td>
</tr>
<tr>
<td>5</td>
<td>83.7</td>
<td>83.8</td>
<td>83.3</td>
<td>83.3</td>
<td>83.2</td>
<td>83.1</td>
<td>83.1</td>
<td>83.2</td>
</tr>
<tr>
<td>6</td>
<td>84.0</td>
<td>84.2</td>
<td>83.8</td>
<td>84.2</td>
<td>84.1</td>
<td>84.3</td>
<td>84.2</td>
<td>84.1</td>
</tr>
<tr>
<td>7</td>
<td>84.0</td>
<td>83.8</td>
<td>83.8</td>
<td>84.0</td>
<td>83.6</td>
<td>83.8</td>
<td>83.9</td>
<td>83.8</td>
</tr>
<tr>
<td>8</td>
<td>83.8</td>
<td>83.9</td>
<td>84.0</td>
<td>83.9</td>
<td>84.0</td>
<td>84.0</td>
<td>84.0</td>
<td>84.0</td>
</tr>
<tr>
<td>9</td>
<td>84.2</td>
<td>84.5</td>
<td>84.3</td>
<td>84.1</td>
<td>83.8</td>
<td>83.7</td>
<td>83.8</td>
<td>83.8</td>
</tr>
<tr>
<td>10</td>
<td>83.6</td>
<td>84.0</td>
<td>84.0</td>
<td>83.7</td>
<td>83.9</td>
<td>84.1</td>
<td>84.2</td>
<td>83.7</td>
</tr>
<tr>
<td>11</td>
<td>84.6</td>
<td>84.6</td>
<td>84.0</td>
<td>83.4</td>
<td>84.4</td>
<td>84.5</td>
<td>83.9</td>
<td>84.1</td>
</tr>
<tr>
<td>12</td>
<td>83.3</td>
<td>82.9</td>
<td>83.2</td>
<td>83.9</td>
<td>82.9</td>
<td>83.7</td>
<td>83.3</td>
<td>83.4</td>
</tr>
<tr>
<td>13</td>
<td>84.5</td>
<td>84.5</td>
<td>84.0</td>
<td>84.2</td>
<td>83.7</td>
<td>84.0</td>
<td>84.0</td>
<td>83.9</td>
</tr>
<tr>
<td>14</td>
<td>83.8</td>
<td>83.8</td>
<td>83.5</td>
<td>83.6</td>
<td>84.3</td>
<td>84.1</td>
<td>83.8</td>
<td>83.8</td>
</tr>
<tr>
<td>15</td>
<td>84.2</td>
<td>84.1</td>
<td>83.8</td>
<td>83.8</td>
<td>83.8</td>
<td>83.8</td>
<td>83.9</td>
<td>83.9</td>
</tr>
<tr>
<td>16</td>
<td>84.2</td>
<td>83.4</td>
<td>83.7</td>
<td>84.1</td>
<td>84.4</td>
<td>84.5</td>
<td>84.0</td>
<td>84.0</td>
</tr>
<tr>
<td>17</td>
<td>83.3</td>
<td>83.4</td>
<td>83.9</td>
<td>84.0</td>
<td>82.2</td>
<td>82.3</td>
<td>82.4</td>
<td>82.7</td>
</tr>
<tr>
<td>18</td>
<td>83.6</td>
<td>83.7</td>
<td>83.6</td>
<td>83.5</td>
<td>84.1</td>
<td>84.0</td>
<td>84.4</td>
<td>84.2</td>
</tr>
<tr>
<td>Type 2</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1</td>
<td>83.4</td>
<td>83.4</td>
<td>83.6</td>
<td>83.5</td>
<td>83.7</td>
<td>83.5</td>
<td>83.1</td>
<td>83.4</td>
</tr>
<tr>
<td>2</td>
<td>84.2</td>
<td>84.1</td>
<td>84.3</td>
<td>84.2</td>
<td>84.2</td>
<td>84.2</td>
<td>84.3</td>
<td>84.2</td>
</tr>
<tr>
<td>3</td>
<td>83.5</td>
<td>83.5</td>
<td>84.2</td>
<td>84.5</td>
<td>83.4</td>
<td>83.7</td>
<td>83.9</td>
<td>84.0</td>
</tr>
<tr>
<td>4</td>
<td>83.4</td>
<td>83.3</td>
<td>83.5</td>
<td>83.1</td>
<td>84.2</td>
<td>84.2</td>
<td>83.3</td>
<td>83.1</td>
</tr>
<tr>
<td>5</td>
<td>83.2</td>
<td>82.8</td>
<td>83.1</td>
<td>82.7</td>
<td>83.0</td>
<td>83.0</td>
<td>83.2</td>
<td>82.7</td>
</tr>
<tr>
<td>6</td>
<td>80.2</td>
<td>80.7</td>
<td>80.8</td>
<td>80.7</td>
<td>80.3</td>
<td>80.4</td>
<td>80.2</td>
<td>79.0</td>
</tr>
<tr>
<td>7</td>
<td>80.9</td>
<td>80.6</td>
<td>80.3</td>
<td>80.6</td>
<td>81.0</td>
<td>81.1</td>
<td>80.7</td>
<td>81.0</td>
</tr>
<tr>
<td>8</td>
<td>83.3</td>
<td>83.5</td>
<td>83.5</td>
<td>83.4</td>
<td>83.9</td>
<td>83.7</td>
<td>83.7</td>
<td>83.7</td>
</tr>
<tr>
<td>9</td>
<td>82.9</td>
<td>82.6</td>
<td>82.8</td>
<td>82.9</td>
<td>83.1</td>
<td>83.1</td>
<td>82.9</td>
<td>82.7</td>
</tr>
<tr>
<td>10</td>
<td>83.8</td>
<td>83.8</td>
<td>83.9</td>
<td>83.8</td>
<td>83.4</td>
<td>83.6</td>
<td>84.0</td>
<td>83.8</td>
</tr>
<tr>
<td>11</td>
<td>83.8</td>
<td>83.4</td>
<td>83.6</td>
<td>83.8</td>
<td>83.8</td>
<td>83.6</td>
<td>83.9</td>
<td>84.0</td>
</tr>
<tr>
<td>12</td>
<td>83.2</td>
<td>82.5</td>
<td>83.0</td>
<td>83.5</td>
<td>84.3</td>
<td>83.5</td>
<td>83.8</td>
<td>83.8</td>
</tr>
<tr>
<td>13</td>
<td>83.4</td>
<td>83.4</td>
<td>83.3</td>
<td>83.3</td>
<td>83.5</td>
<td>83.5</td>
<td>83.2</td>
<td>83.3</td>
</tr>
</tbody>
</table>
TABLE II

Estimated Variance Components

<table>
<thead>
<tr>
<th>Source of Variation</th>
<th>Type 1</th>
<th>Type 2</th>
</tr>
</thead>
<tbody>
<tr>
<td>Lots</td>
<td>0.0620</td>
<td>1.3673</td>
</tr>
<tr>
<td>Samples</td>
<td>0.0683</td>
<td>0.0252</td>
</tr>
<tr>
<td>Chemists</td>
<td>0.0201</td>
<td>0.0407</td>
</tr>
<tr>
<td>Determinations</td>
<td>0.0423</td>
<td>0.0492</td>
</tr>
</tbody>
</table>

All of the last three sources of variation, including sampling, can be considered as measurement error, as opposed to process variability. Note that for Type 1 lots the estimated measurement error is actually greater than the variation between lots. However, for Type 2 material the variation between lots is appreciably greater than the variation in measurement, although this is primarily due to the large discrepancies in lots 6 and 7. The overall measurement error is comparable for the two types of material, as might be expected. Although the difference in sampling variation approaches significance, no apparent reason for this could be found, and the difference is in the opposite direction from that to be expected from the nature of the material processed.

As indicated in this example, the error in a chemical measurement arises generally from two sources, laboratory error and sampling error. Which of these two is the more important will depend on the circumstances involved. There are some cases, such as in the sampling of true solutions or comparatively homogeneous mixtures, where the error of sampling is minor and the analytical error is the controlling factor. On the other hand, there are situations, such as in the sampling of coal, where elaborate schemes must be devised to obtain a sufficiently reproducible and representative sample, whereas the analytical determinations on the sample thus obtained can be carried out with relatively little error. In any event, it must be remembered that effective control can be maintained only if all sources of error are controlled. To obtain a truly independent measurement one must repeat the entire measurement process, including the sampling and sample preparation as well as the laboratory determination.

Many methods have been advanced to control the reproducibility of analytical results. All of these methods have the common feature that more than one independent result must be obtained. More than one analysis on the same material can very seldom be justified on the basis of the increased accuracy obtained from the averaging of the replicate results. Therefore it must be recognized that in the general situation these replicate analyses represent unproductive work on the part of the laboratory done purely for purposes of control. They merely allow an "inspection" of the quality of the product produced. The common practice of running duplicate analyses is somewhat analogous to 100% inspection, and suffers from many of the disadvantages of this practice. In the first place, it is time-consuming. Because of this, the inspection tends to be somewhat cursory in the sense that
the duplicate analyses are frequently not truly independent results, but mere repetitions by the same chemist at the same time on the same sample and on the same apparatus. In the second place, there is a tendency to try to inspect quality into the product. That is, we come to depend on the "repeatability" of an analysis as assurance of its quality. Figure 1 shows graphically 38 pairs of duplicate results on a standard iron solution containing 29.27% iron, submitted by the quantitative analysis class in a large university. On the type of control chart shown, each point represents a pair of duplicate results. Note that although the checks between duplicate results are well within control, as indicated by the 45-degree control lines, the points are not in control with respect to the standard value. The variation of the average results about this value is several times the magnitude of the variation between duplicates.

FIG. 1. Plot of duplicate determinations on a standard iron solution containing 29.27% iron. Scales are % - 25

There is a place for the use of duplicate analyses just as there is a place for 100% inspection. If the detection of every gross error is sufficiently important to justify duplicate analyses on every sample, it is certainly sufficiently important to justify complete repetition of the entire measurement scheme rather than some ineffectual control involving the repetition of only a minor portion of the analytical process. In cases where such duplication (or replication) is not justified, the available effort should be spent on the effective replication of a portion of the measurements, i.e. a sampling inspection, rather than a cursory inspection of all measurements. For example, every tenth sample might be subjected to a referee analysis, with more intensive checking when gross errors are detected. Such a
scheme is, of course, completely analogous to the sampling inspection of a continuous production process. The referee analysis need not necessarily be a replication of the same analytical procedure.

The technique involved in evaluating measurement error in the above situation falls in the field of variance component analysis, since we are trying to estimate the two components, measurement error and product variability, which go to make up the total variation in the observed measurements. The simplest case, of course, is that in which one or more determinations have been made on each production batch in such a fashion that the total variation in the measurements can be divided into two independent parts, one of which furnishes an estimate of the variation within batches, and the other an estimate of the variation between batches. Since the first of these is affected only by measurement errors, while the second is affected by both measurement errors and product variability, it is possible by equating both of these estimates to their average values to obtain unbiased estimates of both the measurement error and the product variability. A similar procedure can, of course, be used where we have two or more nested classifications. The necessary methodology is given in most texts in applied statistics. A particularly good treatment is given in a recent text by Mandel[2].

3.2. Estimation of error from associated measurements: covariance components

The methods of the preceding section apply primarily to singling out the relative contribution of several sources of error in a single measurement. A complementary situation exists when associated measurements with distinct error variances are made on the same series of items. For example, shipments may be analysed by both shipper and receiver; both a direct and an indirect measurement of content may be possible; or a series of fuel elements may be weighed on several available scales. The estimation of the individual errors in this type of situation is considered quite generally by Jaech in a recent paper [3] based on earlier work by Grubbs [4].

To illustrate the principles involved, consider the simple case of two different unbiased measurements of different precision on a single series of items. As in section 2, the model for the two measurements \( x_i \) and \( y_i \) can be written

\[
  x_i = \theta_i + \epsilon_i \quad y_i = \theta_i + \eta_i
\]

Assuming the errors to be independent, and independent of the variation between items, the sample variances (mean square deviations) \( s_x^2 \) and \( s_y^2 \) and the sample covariance (mean cross-product of deviations) \( s_{xy} \) are estimates of \( \sigma^2 + \sigma^2 \epsilon, \sigma^2 + \sigma^2 \eta, \) and \( \sigma^2 \), respectively, where \( \sigma^2 \) is the true item to item variability, and \( \sigma^2 \epsilon \) and \( \sigma^2 \eta \) are the respective measurement errors.\(^1\)

---

1 As indicated in section 2, it is common statistical practice to indicate the expected value (mean) and variance of a random variable by the Greek letters \( \mu \) and \( \sigma^2 \) respectively. The standard deviation \( \sigma \) is the square root of the variance. When more than one random variable (or error) is involved, subscripts differentiate between the respective means and variances. Similarly, it is common practice to designate parameters associated with random variables or the associated probability distributions by Greek letters, and the corresponding quantities (statistics) obtained from a sample of observations by Roman letters. "Hats" are commonly used to indicate values of population parameters estimated from a sample by same function of appropriate sample statistics.
TABLE III

Product Content of Each of 24 Production Lots Obtained by Each of Two Analytical Methods

<table>
<thead>
<tr>
<th>Lot</th>
<th>Method I</th>
<th>Method II</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.79</td>
<td>0.64</td>
</tr>
<tr>
<td>2</td>
<td>0.81</td>
<td>0.58</td>
</tr>
<tr>
<td>3</td>
<td>0.86</td>
<td>1.09</td>
</tr>
<tr>
<td>4</td>
<td>0.82</td>
<td>0.74</td>
</tr>
<tr>
<td>5</td>
<td>0.82</td>
<td>0.62</td>
</tr>
<tr>
<td>6</td>
<td>0.87</td>
<td>0.86</td>
</tr>
<tr>
<td>7</td>
<td>1.00</td>
<td>1.01</td>
</tr>
<tr>
<td>8</td>
<td>0.88</td>
<td>0.99</td>
</tr>
<tr>
<td>9</td>
<td>1.03</td>
<td>0.87</td>
</tr>
<tr>
<td>10</td>
<td>0.98</td>
<td>0.86</td>
</tr>
<tr>
<td>11</td>
<td>0.90</td>
<td>1.28</td>
</tr>
<tr>
<td>12</td>
<td>0.87</td>
<td>0.64</td>
</tr>
<tr>
<td>13</td>
<td>0.90</td>
<td>0.80</td>
</tr>
<tr>
<td>14</td>
<td>1.04</td>
<td>1.20</td>
</tr>
<tr>
<td>15</td>
<td>0.97</td>
<td>1.17</td>
</tr>
<tr>
<td>16</td>
<td>1.03</td>
<td>0.51</td>
</tr>
<tr>
<td>17</td>
<td>1.02</td>
<td>0.45</td>
</tr>
<tr>
<td>18</td>
<td>0.79</td>
<td>0.98</td>
</tr>
<tr>
<td>19</td>
<td>0.89</td>
<td>0.72</td>
</tr>
<tr>
<td>20</td>
<td>0.81</td>
<td>0.80</td>
</tr>
<tr>
<td>21</td>
<td>0.79</td>
<td>0.87</td>
</tr>
<tr>
<td>22</td>
<td>0.85</td>
<td>0.64</td>
</tr>
<tr>
<td>23</td>
<td>0.77</td>
<td>0.85</td>
</tr>
<tr>
<td>24</td>
<td>1.00</td>
<td>0.52</td>
</tr>
</tbody>
</table>

EQUATING the estimates to their expected values enables us to solve for both the product variability and the measurement error in both methods.

As an example, consider the data given in Table III, which shows the product content (coded by subtracting a constant) of each of twenty-four production lots obtained by each of two analytical methods. These data are plotted in Fig. 2. Note the apparent lack of correlation between these two measurements, which is immediately indicative of the fact that most of the variability in these measurements is due to measurement error rather than to actual variability in the lots themselves. For these data we obtain

\[ s_x^2 = 0.0516 \quad s_{xy} = 0.00132 \quad s_y^2 = 0.0081 \]

and the estimated variances are

\[ \hat{\sigma}_0^2 = 0.00132 \quad \hat{\sigma}_e^2 = 0.0503 \quad \sigma_\eta^2 = 0.0068 \]

\[ \sigma_\theta^2 = 0.0363 \quad \sigma_\varepsilon^2 = 0.224 \quad \sigma_\eta = 0.082 \]

As anticipated \( \hat{\sigma}_0^2 \) is small compared to \( \hat{\sigma}_e^2 \) and \( \sigma_\eta^2 \).
4. DETECTION OF BIAS

4.1. Types of bias

Within the general definition of bias given in section 2, three distinguishable types can be considered: (1) bias relative to a standard, (2) bias relative to prior experience, and (3) bias relative to a group. Bias relative to a standard refers to the degree to which the method of measurement under consideration deviates systematically from some "standard" value which by implication is much more precisely and exactly determined than the method under consideration. By the same token, it is to be expected that this standard will be unbiased either (1) by definition, such as the definition of the standard metre, or (2) because it is determined in such a fashion that we have technical reasons to believe that all possible biases have been removed, such as is the case with the isotopic standards now available through the Bureau of Standards. Bias relative to prior experience refers to the existence of a systematic change in the method of measurement or process under consideration, i.e. whether or not present measurements are consistent with those previously obtained, or have deviated in some systematic fashion. It is this type of bias that concerns itself with process shifts. It is also in this context that the statistical use of the term bias agrees most closely with the dictionary definition which refers to bias as prejudice or a leaning of the mind. Both sudden shifts from previous experience or gradual drifts in the measurement or process must be considered. Finally, bias relative to a group refers to the degree to which several different and parallel methods differ systematically from each other. This type of bias is typified by the lack of consistency between laboratories, measurement situations, or individuals performing a given analysis. Only the first of these types has any sense of absolute bias contained in it. The last two are purely relative, the second being concerned with the relationship between measurements made over a period of time, the last with the relationship between different physical entities or individuals at a given time.
Before proceeding to examine the fundamentals of the techniques used to detect and measure these various types of bias, the reasons why a consideration of the bias in measurements is so important should be briefly discussed. The first and most obvious reason, especially when the bias concerned is a systematic deviation from a standard, is that in the presence of bias, or systematic errors, the total absolute error in the sum of a series of measurements will accumulate in direct proportion to the number of measurements involved, rather than proportional to the square root of the number of measurements, as is the case with random errors. Similarly, a sum of measurements will have precisely the same percentage bias as the individual measurement, whereas the percentage error due to random fluctuations will tend to decrease as $1/\sqrt{n}$. Putting this in the simplest possible terms, random deviations or fluctuations tend to disappear when a large number of measurements are accumulated but biases do not. The second and more compelling reason for studying bias is that independent of any consideration of deviation from a standard value, we are almost always interested in the consistency of our comparisons or processes. It is essential to effective control that we establish the stability of our measurement methods and processes over both time and space. Only when this stability has been established are we in a position to concentrate on the absolute nature of our measurements.

4.2. Bias relative to a standard

Consider first the problem of detecting bias relative to a standard. Perhaps the minimum assumption which allows the use of statistical procedures is that in the absence of bias the measurements would fall above and below the standard value with about equal frequency. Thus in a series of n measurements, about n/2 would be expected to fall above and n/2 below the standard value. However, in any given situation this expectation will not always be realized, i.e. when a coin is flipped ten times, the result is not always precisely five heads and five tails, but occasionally four heads and six tails or three heads and seven tails. The precise probability of these combinations, assuming equal probability of each, can be determined. Thus, if eight measurements above the standard value and two measurements below are observed, the conclusion is that in the absence of bias, where this is defined to mean approximately equal frequency above and below the standard, the probability of such an occurrence would be 0.0344. The analyst must now decide whether or not this is something that would happen by chance or whether this probability is sufficiently small to support the conclusion that bias is actually present.

Notice that the presence of bias, i.e. the violation of the assumption of equal frequency, tends to make such one-sided occurrences more probable, i.e. tends to increase the chances of concluding that bias is present. A large positive bias compared to the variation in the measurements will make it almost certain that a measurement will fall above the standard. In this case some such split as nine-one or ten-zero will be highly probable, and a conclusion that bias is present relatively certain. However, if the bias is small relative to the variation in the measurements, it is likely to
cause only a minor deviation from equal frequency, and in this case, a split such as six-four, which actually reflects the true situation, will be very difficult to distinguish from a chance deviation based on equal frequency. The degree to which the probability of an unusual occurrence is affected by the presence of bias is called the "power" of the test with respect to the detection of bias. Tests based on relatively small number of measurements, such as that considered in the above example, may have a high power with respect to large biases but very little power with respect to small biases. As can be seen, very small biases will be almost impossible to detect even with a very large number of measurements.

The simple test described above is called the "sign test" since it depends only on the direction of the deviation and not its magnitude. Because it involves only the assumption of about equal frequency above and below the standard value, it is a non-parametric test. This test is described in many standard elementary texts on statistics, for example Refs. [5] or [6].

A slight variation from the sign test which is still non-parametric is given by a type of test known as the "signed rank test". In this case not only the sign of the differences is considered, but they are also ranked in order of magnitude. The sum of the ranks of those differences having positive signs is then computed, and a similar sum for the differences with negative signs. The smaller of these, known as the Wilcoxon sum, can then be examined to determine whether it is sufficiently small to warrant a rejection of the hypothesis of a symmetric distribution about the standard value. This test is slightly more powerful than the sign test in that it at least considers the order of magnitude of the deviation as well as its direction. Tables of critical values for this test can be found in Ref. [4], which also contains additional references to original work on this test. The use of this type of statistic in estimation is somewhat more complicated.

Virtually all the parametric methods for detecting bias relative to a standard are based on the assumption that in the absence of bias the measurements are normally distributed about the standard value \( \mu \), with some fixed but known variance \( \sigma^2 \), and that the presence of bias means that the mean of observations is displaced from the true mean by some unknown amount \( \delta \). Under these assumptions the exact Student "t" test for significance of the deviation of an observed from a true mean is available, and the power of this test at any given significance level can be plotted as a function of \( \delta / \sigma \), i.e. as a function of the amount of bias present measured in terms of the precision of measurements [6]. Such curves are again referred to as the power curve or operating characteristic curve of the test. If \( \sigma \) is known or can be approximated, the number of measurements required to detect a given amount of bias with a given probability can be determined. In addition to the exact procedure, many approximate procedures based on functions of order statistics, such as the range, mid-range, and median are also available. These are rather completely described in Ref.[5]. They have the advantage of being almost as powerful as the exact procedure and at the same time involving much less computation.

Parametric tests of this type have the advantage of being more sensitive to a given level of bias than the non-parametric procedures, and of giving estimates of the bias present directly in terms of deviations of the
average of the measurements from their standard value. They have the
disadvantage that the significance level of a given test can depend not only
on the amount of bias present but on the degree to which the assumptions
concerning the nature of the distribution are violated. In this sense a good
test (referred to in statistical terms as one which is robust) is not only one
which has high power with respect to the alternative being studied, but which
is only moderately or slightly affected by violations of the other assumptions
concerning the nature of the statistical distribution. Many studies have
shown that the Student "t" test is quite robust, i.e. the significance levels or
probabilities based on the assumption of a normal distribution are not greatly
changed by non-normality. However, the test is moderately affected by the
contamination of the measurements with occasional measurements with a
much larger variation, a situation frequently encountered in chemical
measurements. Under these conditions the sensitivity of the sign test may
become greater than that of the "t" test.

4.3. Bias relative to prior experience

The problem of detecting process trends and shifts has by and large
been treated independently of the above considerations, although the sta-
tistical methods used are closely related. The question here is one of de-
termining whether and when a shift in the process or measurement has taken
place, i.e. is there a bias in recent results compared to previous ones. The
simplest situation involves the assumption that successive measurements
are equally variable but that the mean value has changed with time. In order
to fix ideas in mind, consider a series of n measurements in time order.
The problem then is to determine whether or not the expected value of these
measurements has changed during the time period represented. Two si-
tuations can occur: (1) a fixed series of measurements is available, and
the problem is to determine whether or not a shift has occurred; (2) a con-
tinuing series of measurements is available, and the problem is to deter-
mine at the earliest possible time whether and when a shift has occurred.

The first case can be subdivided into: (a) the situation where there is
some information as to when the shift may have occurred, and the question
is whether it occurred, and (b) the situation where there is no information
as to when the change, if any, occurred. (a) is virtually equivalent to the
problem of determining whether two samples have different means, while (b)
can be identified with the question of whether or not a given sample is a
random sample from a fixed distribution.

In case (1a) the Student "t" test for the significance of difference be-
tween two means is the most powerful when the assumption of a normal distri-
bution of random errors and a fixed and equal variance before and after the
shift is valid. This test and its power function are again described in most
standard statistical texts, and as before, many highly efficient and more
easily computed variations based on order statistics are available.

Case (1b) can be approached by testing for a violation of the assumption
that the given measurements are a random sample from a fixed distribution.
A non-parametric method for this purpose based on the distribution of runs
was developed by Swed and Eisenhart and the later work of Mood and
Mosteller on the theory of runs is also applicable. In particular, the examination of a series of measurements in terms of the runs above or below the median (middle value) of the measurements gives a relatively sensitive indication of the presence of long-term trends or drifts. Tables are available (see, for example, Ref. [6]) showing the probability that this number will be above or below a given value in the case of a true random series, and hence the significance of a given observed number of runs can be assessed. These critical values do not depend on any assumptions concerning the nature of the distribution. If it can be assumed that the variations in the measurements, aside from any long-term trend or drift, are normally distributed, then the mean square successive difference developed by Hart and von Neumann can be used as a test of long-term trends in the means of the observations. References to original work in this problem and a table of percentage points are again given in Ref. [3]. The principle here is quite simple; if the estimate of the variance obtained from successive differences is much smaller than the overall variance of the observations, this can be accounted for only by the presence of a long-term trend of some sort in the mean values. A complete theoretical examination of this problem, known to statisticians as the slippage problem, is given in Ref. [7].

Consider now the second of the two situations outlined above, that in which a continued series of measurements is available, and the problem is to determine at the earliest possible time whether or not a process shift has taken place. The selection of shifts of this type was inherent in Shewhart’s early work on the control chart, in that this type of variation was one of the many types of “assignable” causes which he wished to detect through the use of a control chart. It has been shown since that time, however, that the Shewhart control chart, while extremely sensitive to sudden large shifts, is relatively insensitive to gradual changes or process drift. Ad hoc modifications in the control chart principle which attempted to take this into account were based on the plan of establishing warning limits as well as action limits, so that several successive points outside the warning limits were considered equivalent to one point outside the action limits. Olmstead has also considered the length of runs on a control chart above or below the central line required to establish the significance of a possible change in process level.

In recent years there has been a renewal of interest in the use of some type of control chart specifically designed to look for changes in process level rather than sudden erratic values. This has led to the consideration of various charts based on weighted means of the past observations, and to the development by Page and others of the cumulative sum chart. The latter examines continuously not the observations themselves but the nature of the changes in their cumulative sum. A fine expository summary of the use of cumulative sum charts and their relationship to other types of control charts is given by Page [8], and a more extensive discussion of when and how to use cumulative sum charts is given by Ewan [9]. Another discussion of all these methods, and a comparison based on the average run length required to detect a given change in process level, is given by Freund in a recent issue of Industrial Quality Control [10]. Fundamentally, all of these discussions indicate that the newer types of charts are more sensitive than the
Shewhart chart to small or gradual changes in process as opposed to sudden large shifts.

4.4. Bias relative to a group

Finally, let us consider the statistical methods which are applicable to the study of relative bias among a number of machines or methods. Here two types of questions can be asked: (1) Are the methods or machines as a whole consistent, and (2) what are the relative biases of these machines with respect to each other, or with respect to some one machine chosen as a standard. The first situation occurs when each of a group of analysts is asked to repeatedly analyse a given standard and was discussed in section 3.1. The second situation occurs when successive measurements on a series of objects or solutions are obtained by two or more methods and was discussed in section 3.2. In either case, the basic question concerning the presence of bias is whether or not the variability between measurements or between methods exceeds the variability within methods, i.e. does the bias between measurements contribute to the total variability. Whether these biases are considered as fixed, or as representative samples from a distribution of biases which might occur between similar machines or individuals, is the basic problem of fixed versus random models, which has received much attention from statisticians since about 1950. The details of the methods involved can be found in Refs. [2, 6, 11, 12]. Another recent discussion applicable to this type of problem, with particular reference to the estimation of physical constants, has been given by Youden [12].

5. CONCLUDING REMARKS

It is obvious that the above exposition has been intended to elucidate principles rather than present details. In practice, the statistical approach is capable of almost infinite variation, depending on the particular problem being attacked. Other papers in these proceedings have used the methods discussed, or adaptations of them, to solve specific problems. This paper has been concerned with the statistical approach rather than specific accomplishments through the use of statistical techniques. It is essential that this approach be used (either formally or informally) if we are to obtain the maximum information concerning the quality of source data and the effectiveness of control indices.

REFERENCES


DISCUSSION

W. R. SHIELDS: It strikes me that there is a difference between bias (systematic shift) and the limit of error of bias, which reflects inability to control a systematic shift. Bias as such is no problem, but the limit of error of bias represents the limiting parameter of measurement.

C. A. BENNETT: I quite agree. Every source of error generally includes a random component and a fixed component, and certainly a source of bias can also be a source of random variability. However, I am concerned primarily with non-random changes in bias with time, between instruments, and between different analysts.

M. J. HIGATSBERGER: Bias can usually be reduced if one uses different methods to measure the data, e.g. chemical means, mass spectrometry, or non-destructive methods. In some cases deviations can be adjusted by a "least square" approach, and inconsistent values might even be rejected.

C. A. BENNETT: Let me say only that it is dangerous to "reject" data, for not infrequently a rejected value contains more information than any other.
NUCLEAR MATERIALS MANAGEMENT IN RELATION TO SAFETY AND CRITICALITY CONTROL: A STUDY OF NON-INHERENTLY SAFE SYSTEMS. The production, utilization and handling of certain fissionable materials are associated with a particular type of danger, the risk of criticality. The engineer must try to avoid this risk, at the least possible expense, without hindering too much the work of production or conversion. Roughly speaking, there are three ways of eliminating this risk: limitation of solution concentrations, limitation of mass, and judicious choice of geometries. The tendency is to employ the third method as much as possible, i.e. to operate with inherently safe systems. Nevertheless, non-inherently safe systems are still common in nuclear installations and recourse sometimes has to be had to the first or second procedure. These methods are bound up with nuclear materials management.

In installations where small quantities of fissionable materials are handled it is often possible to ensure nuclear safety if the mass of fissionable material permitted in each room is less than the minimum critical mass. In larger installations this type of control, based entirely on materials management, becomes impossible.

The paper considers the relation between management and nuclear safety in the special case of a plant for the aqueous processing of irradiated fuels (the fuels are based on natural uranium). From the point of view of criticality control such a plant comprises, broadly speaking, three sections:

In the first section, where fairly dilute solutions are treated, the type of control required is that over concentration. This control (based on analysis and the use of neutron counters) is easy to carry out but it is difficult to ensure that no deposits are formed. Although experience has shown that there is no particular reason for fearing such deposits, the existence of a deficit in the material balance gives rise to apprehension in respect of criticality. In this section, materials management, still imperfectly carried out, is of prime importance for safety.

The second section is where concentrated solutions and wet precipitates are treated. This is the realm of safe geometry. Materials management is of less direct interest here but is still an important factor as far as safety is concerned (where special action, cleaning of equipment, etc. is necessary).

In the third section, for the treatment of dry products, control by mass is required and materials management presents no particular difficulty.

An additional field which, although less obvious, is nevertheless of paramount importance from the point of view of safety is that of recycling and waste processing. A look at the list of criticality accidents in the United States of America is sufficient to show that five out of six accidents were connected more or less directly with waste processing. If we examine these accidents in greater detail, we can observe that in most cases one of the contributory causes to a criticality accident was faulty management of fissionable materials.

The "over-all" importance of the relation between safety and nuclear materials management should also be stressed. Although this management may not always form the basis of "primary" criticality control, it is still indispensable throughout any process since what is done in this respect in one section of a plant is of significance with regard to the safety of neighbouring sections, especially that concerned with waste processing.
manutention de certaines matières fissiles comportent un type de danger particulier qui est le risque de criticité. L'art de l'ingénieur est d'éviter ce risque, au moindre prix, sans trop gêner le travail de production ou de transformation. Pour écarter ce risque, il existe, en première approximation, trois méthodes préventives: une limitation de la concentration des solutions, une limitation de la masse, un choix judicieux des géométries. La tendance est d'employer au maximum la troisième méthode, autrement dit de fonctionner avec des systèmes à sécurité inhérente. Toutefois, les systèmes à sécurité non inhérente restent nombreux dans les installations nucléaires et le recours à l'une ou l'autre des deux premières méthodes devient nécessaire. Ces méthodes sont liées à la gestion des matières nucléaires.

Dans les installations où sont manipulées de petites quantités de matières fissiles, il est souvent possible d'assurer la sécurité nucléaire en admettant dans chaque salle une masse de matière fissile inférieure à la masse critique minimale. Pour des installations plus importantes, un tel contrôle, qui repose entièrement sur la gestion des matières, devient impossible.

L'auteur examine, dans le cas particulier d'une usine de traitement de combustibles irradiés par voie aqueuse (les combustibles sont de base d'uranium naturel), comment se présente la liaison entre gestion et sécurité nucléaire. Une telle usine comprend grossièrement trois parties au point de vue du contrôle de la criticité:

Dans la première partie, traitant des solutions assez diluées, le contrôle qui s'impose est celui de la concentration. Si ce contrôle est aisé (analyse et compteurs neutrons), il est difficile de s'assurer qu'il ne se forme pas de dépôts: bien que l'expérience ait montré qu'ils n'étaient pas tellement à craindre, le fait bien connu d'un bilan matières déficitaire reste un souci en matière de criticité. Dans cette partie, la gestion des matières, encore imparfaitement réalisée, est de première importance pour la sécurité.

La deuxième partie traite des solutions concentrées et des précipités humides. C'est le domaine de la géométrie sûre. La gestion des matières l'intéresse moins directement mais reste un facteur de sécurité important (en cas d'intervention, de nettoyage des appareils, etc.).

Un contrôle par la masse s'impose dans la troisième partie qui traite des produits secs. La gestion ne présente pas de difficulté particulière.

A ce schéma se superpose une partie moins spectaculaire mais qui sur le plan sûreté est de toute première importance: c'est la partie relative aux recyclages et au traitement des déchets. Il suffit de se reporter à la liste des accidents de criticité survenus aux Etats-Unis pour constater que sur six accidents, cinq ont trait plus ou moins directement au traitement des déchets. Si l'on examine plus en détail ces accidents, on constate que dans la plupart des cas, parmi les causes qui se conjuguent pour créer un accident de criticité, l'une d'elles est un défaut dans la gestion des matières fissiles.

Il est bon enfin de souligner l'importance «globale» de la liaison entre sécurité et gestion des matières nucléaires; en effet, si cette gestion n'est pas toujours la base du contrôle «primaire» de la criticité, elle reste indispensable tout au long d'un procédé, car la gestion des matières dans une partie d'usine influence la sécurité des parties voisines et tout spécialement la partie relative au traitement des déchets.
В первой части, где обрабатываются довольно разбавленные растворы, необходим контроль за концентрацией. Если этот контроль и удобен (анализ и нейтронные счетчики), то возникает трудность в уяснении того, что при этом не образуются отложения: хотя опыт и показал, что не следует бояться этих отложений, но хорошо известным из дефицитного материального баланса моментом является забота о критичности. В этой части обращение с материалами, еще не полностью достигнутое, имеет первостепенное значение для безопасности.

Во второй части обрабатываются концентрированные растворы и сырые осадки. Геометрия материалов имеет здесь основное значение. Обращение с материалами имеет меньшее значение, но остается существенным фактором безопасности (в случае принятия специальных мер, чистки аппаратуры и т.п.). Контроль по массе осуществляется в третьей части, где обрабатываются сухие продукты. Обращение не представляет собой какую-либо особую трудность.

По этой схеме на первое место выдвигается менее эффективный момент, который, однако, в плане безопасности приобретает первостепенное значение; этот момент относится к повторным циклам и к обработке отходов. Достаточно сослаться на перечень аварий, связанных с критичностью, имевших место в Соединенных Штатах, чтобы указать на то, что из этих аварий пять имеют более или менее прямое отношение к обработке отходов. Если мы более подробно изучим эти аварии, то отметим, что в большинстве случаев одной из причин, которые приводят к авариям, связанным с критичностью, являются недостатки в обращении с делящимися материалами.

Следует также подчеркнуть "глобальное" значение связи между безопасностью и обращением с ядерными материалами; действительно, если это обращение не всегда является основой "первичного" контроля за критичностью, то оно необходимо на протяжении всего процесса, поскольку обращение с материалами на одной части завода имеет отношение к безопасности соседних частей и, прежде всего, той части, где обрабатываются отходы.

LA ADMINISTRACION DE MATERIALES NUCLEARES Y SUS RELACIONES CON LA SEGURIDAD Y EL CONTROL DE LA CRITICIDAD, ESTUDIO DE LOS SISTEMAS DE SEGURIDAD NO INHERENTE. La producción, el empleo y la manipulación de ciertos materiales fisionables entrañan el riesgo de criticidad. El ingeniero tiene que evitar este riesgo del modo más económico posible y sin entorpecer demasiado las operaciones de producción o de transformación. Para ello puede hacer tres cosas: limitar la concentración de las soluciones, limitar la masa o elegir juiciosamente las geometrías. La tendencia es emplear lo más posible el tercer método, es decir, operar con sistemas de seguridad inherente. De todas formas, los sistemas de seguridad no inherente siguen siendo numerosos en las instalaciones nucleares y obligan a recurrir a uno u otro de los dos primeros métodos. Ambos van unidos a la administración de los materiales nucleares.

En las instancias donde los materiales fisionables se manipulan en pequeñas cantidades se puede con frecuencia garantizar la seguridad nuclear admitiendo en cada sala una masa de materiales fisionables inferior a la masa crítica mínima. En instalaciones más importantes esta forma de control, enteramente basada en la administración de los materiales, resulta imposible.

Examinemos las relaciones que existen entre la administración y la seguridad nuclear en el caso especial de una planta de tratamiento por vía acusa de combustibles irradiados (combustibles a base de uranio natural). Desde el punto de vista del control de la criticidad, una planta de esta clase puede dividirse en tres partes:

En la primera, donde se tratan soluciones bastante diluidas, lo que hay que controlar es la concentración. Este control es fácil (análisis y contadores neutrónicos) pero resulta en cambio difícil asegurarse de que no se forman depósitos: aunque la experiencia haya demostrado que no son muy peligrosos, el hecho bien conocido de un balance de materiales deficitario sigue siendo una preocupación en lo que respecta a la criticidad. En esta primera parte, la administración de los materiales, aún imperfecta, tiene una importancia primordial para la seguridad.

En la segunda parte se tratan soluciones concentradas y precipitados húmedos. Es la parte donde debe aplicarse una geometría segura. La administración de los materiales tiene menos interés directo pero sigue siendo un factor importante de seguridad (en caso de intervención, de limpieza de los aparatos, etc.).

En la tercera parte, donde se tratan productos secos, es preciso controlar la masa. La administración no presenta dificultades especiales.

A este esquema se superpone una parte menos espectacular pero que desde el punto de vista de la seguridad tiene muchísima importancia: es la parte donde se efectúan los reciclados y el tratamiento de los desechos. Basta examinar la lista de los accidentes de criticidad en los Estados Unidos para comprobar que de cada seis, cinco están más o menos directamente relacionados con el tratamiento de desechos. Si examinamos estos
accidentes con más atención comprobaremos que en la mayoría de los casos una de sus causas ha sido un defecto en la administración de los materiales fisionables.

Por último, conviene subrayar la importancia «global» de las relaciones que existen entre la seguridad y la administración de los materiales nucleares; en efecto, si esta administración no constituye siempre la base del control «primario» de la criticidad, sigue siendo indispensable a lo largo de todo un proceso, ya que la administración de los materiales en una parte de la planta repercute en la seguridad de las partes vecinas, en particular de la destinada al tratamiento de los desechos.

La producción, la mise en œuvre et la manutention de certaines matières fissiles comportent un type de danger particulier: le risque de criticité. L'art de l'ingénieur est d'écartier ce risque, au moindre prix, en apportant le moins de gêne possible à l'exploitation des installations. Pour écarter ce risque il existe, en première approximation, trois méthodes préventives: une limitation de la concentration des solutions en matière fissile, une limitation de la masse de la matière fissile, l'adoption d'une géométrie favorable pour les appareils de génie chimique. Les deux premières méthodes s'appliquent à des systèmes à sécurité non inhérente: la sécurité nucléaire de tels systèmes repose entièrement sur la gestion des matières fissiles. La troisième méthode de contrôle définit les systèmes à sécurité inhérente: la gestion des matières fissiles intéresse moins directement la sécurité de tels systèmes mais elle demeure - nous le montrerons dans la suite de ce mémoire - un facteur de sécurité important.

La gestion des matières fissiles et la sécurité nucléaire sont donc étroitement liées. Nous nous proposons de développer cet aspect dans le cas d'une usine de traitement par voie aqueuse de combustibles irradités (les combustibles sont à base d'uranium naturel) et dans le cas d'un atelier de métallurgie mettant en œuvre des matières fissiles concentrées.

I. SECURITE ET GESTION DANS UNE USINE DE TRAITEMENT DE COMBUSTIBLES IRRADIES

Une usine de traitement de combustibles irradités comprend grossièrement quatre parties du point de vue du contrôle de la criticité:
- La première partie traite des solutions peu concentrées en plutonium.
- La deuxième partie traite des solutions concentrées en plutonium et des précipités humides.
- La troisième partie traite des produits secs: poudres, métal.
- La quatrième partie est une partie moins spectaculaire mais qui, sur le plan sûreté, est de première importance: c'est la partie relative au traitement des produits de récupération.

A. Première partie de l'usine

La première partie de l'usine est constituée de systèmes à sécurité non inhérente dans lesquels le paramètre fondamental de contrôle de la criticité est la concentration en plutonium des solutions. Cette concentration doit être connue avec précision et doit toujours être inférieure à la concentration critique en milieu infini qui est de l'ordre de 7,8 g/litre pour une solution de plutonium-239 pur.
Dans cette partie d'usine, le meilleur contrôle de la criticité est le contrôle du procédé lui-même: ainsi dans un atelier d'extraction (en état d'équilibre) la valeur de la concentration en plutonium de la phase de sortie riche est liée au rapport des débits des phases entrante et sortante. Le contrôle des débits est donc la première opération de contrôle de la criticité. Il est nécessaire de compléter ce contrôle par un contrôle analytique et par un contrôle continu et instantané à l'aide de compteurs α ou de compteurs à neutrons. Notons ici que la conception de l'installation est d'une grande importance pour la sécurité nucléaire: par exemple il est possible dans un atelier d'extraction d'éviter les pics de concentration en plutonium grâce à une disposition spéciale des batteries de mélangeurs – décanteurs, telle que celle réalisée à l'usine de plutonium de la Hague. 

Mais pour garantir la sécurité nucléaire de tels systèmes, il ne suffit pas de maintenir la concentration des solutions en dessous de la valeur limite, il faut également prévoir les anomalies de fonctionnement pouvant provoquer des précipitations ou la présence de dépôts dans les appareils. C'est pourquoi on s'efforce d'effectuer un deuxième contrôle – dit contrôle secondaire – par la masse de plutonium mise en jeu dans les appareils. Cette façon de faire consiste à s'assurer que l'on n'introduit jamais dans un récipient plus de la masse critique minimale relative à ce récipient; on sait en effet que pour un récipient de géométrie donnée, la masse critique est minimale pour une certaine concentration en plutonium voisine de 30 g/litre. Or cette masse minimale dépend beaucoup de la forme du récipient et on s'est aperçu que, paradoxalement, on a souvent intérêt, dans une partie d'usine traitant des solutions peu concentrées, à adopter de grands appareils, ou plus exactement des appareils présentant une grande surface horizontale de dépôt (exemple: cuve cylindrique à axe horizontal de grande longueur): pour de tels récipients les masses critiques minimales peuvent en effet être importantes (de l'ordre de plusieurs kilogrammes de matière fissile). Bien entendu on cherchera à éviter ces incidents de précipitation, et à les détecter au plus tôt; il faut s'efforcer pour cela d'effectuer une gestion précise des matières fissiles entrant en jeu dans un atelier de fabrication, ce qui est un problème difficile lorsqu'on a affaire à des solutions: à Marcoule il n'a guère été possible jusqu'à présent de tenir un bilan à mieux de 3% près, ce qui ne permet pas de fonder la sécurité nucléaire sur les opérations de bilan. C'est pourquoi nous effectuons un contrôle périodique des batteries de mélangeurs-décanteurs grâce à un appareil mobile muni d'un compteur BF₃ capable de détecter des dépôts éventuels [1]. Notons que nous n'avons jamais relevé la présence de dépôts, que ce soit par ce mode de contrôle ou par les rinçages de fin de campagne.

En conclusion, la première partie d'une usine de traitement de combustibles irradiés comporte nécessairement des systèmes à sécurité non inhérente; l'adoption de systèmes à sécurité inhérente serait d'ailleurs d'un coût prohibitif étant donné les grands volumes de solutions traités. Il est toutefois possible, au stade de la conception, d'obtenir une certaine sécurité intrinsèque, par l'amélioration du procédé d'une part, par l'adoption de certaines formes d'appareils d'autre part. Mais c'est le contrôle de la concentration, c'est-à-dire la gestion des matières fissiles, qui est à la base de la sécurité.
B. Deuxième partie de l'usine

Si l'expérience montre que les systèmes à sécurité non inhérente sont parfaitement admissibles dans la première partie d'une usine de traitement de combustibles irradiés, il en va tout différemment dans la deuxième partie qui traite des solutions concentrées et des précipités humides: dans cette partie, il est nécessaire d'employer au maximum la géométrie favorable, c'est-à-dire de fonctionner avec des systèmes à sécurité inhérente. La définition et le perfectionnement des systèmes à sécurité inhérente constituent l'essentiel des recherches en matière de sûreté dans le domaine des risques de criticité; l'objet de ce mémoire n'est pas de développer cette question complexe, mais son importance est telle qu'il est nécessaire de l'évoquer brièvement, d'autant plus que la gestion des matières fissiles intéresse aussi de tels systèmes.

Quels sont les avantages de ces systèmes? Ce sont essentiellement la sécurité, la simplicité et, en définitive, quand ils sont bien adaptés au procédé, l'économie. De tels systèmes permettent en effet de supprimer un grand nombre de contrôles et de consignes qui représentent un travail supplémentaire pour l'exploitant et qui, de plus, sont sujets à des défaillances matérielles et humaines. Mais il serait dangereux de croire que la sécurité inhérente est une sécurité absolue: d'une part l'exploitant doit connaître les hypothèses qui sont à la base des calculs de ces systèmes, ce qui lui permet d'apprécier la marge de sécurité existante (cela est particulièrement valable pour l'interaction neutronique), d'autre part il doit se méfier des modifications apportées aux circuits: le contrôle de l'intégrité des systèmes à sécurité inhérente est un aspect important et souvent oublié du contrôle de la criticité dans une installation.

En quoi la gestion des matières fissiles intéresse-t-elle les systèmes à sécurité inhérente? Essentiellement en cas d'anomalies de fonctionnement: fuites, encroûtement des appareils, etc. De tels incidents nécessitent des interventions telles que des nettoyages: une bonne gestion permet d'apprécier les quantités de matières fissiles mises en jeu lors de ces interventions, ce qui est précieux pour le retraitement des produits de récupération. Ainsi, de même que, dans le cas d'un contrôle «primaire» par la concentration (première partie d'une usine), il faut assurer un contrôle «secondaire» par la masse, de même ce contrôle secondaire est utile dans le cas d'un contrôle primaire par la géométrie: dans les deux cas la caractéristique de ce contrôle est de jouer lors d'une anomalie de fonctionnement.

Si l'emploi d'une géométrie favorable est fortement souhaitable dans la partie d'une usine traitant des solutions concentrées ou des précipités humides, une telle partie peut malheureusement comprendre des systèmes à sécurité non inhérente, comme c'est le cas de l'usine d'extraction du plutonium de Marcoule par exemple. Ainsi, à Marcoule, nous effectuons deux opérations de précipitation dans des appareils de géométrie non favorable: décanteuses pendulaires, patouillettes de précipitation d'oxalate. Ces installations sont, par excellence, des systèmes à sécurité non inhérente, qui doivent d'ailleurs être remplacés prochainement par des installations beaucoup plus sûres. Dans ces installations peu favorables sur le plan sûreté, la sécurité nucléaire repose exclusivement sur un contrôle de la masse de matière fissile entrant en jeu dans chaque appareil. Personne n'ignore la
difficulté de mesurer avec précision la masse de matière fissile dans les solutions; c'est pourquoi nous cherchons, à Marcoule, à nous assurer la sécurité intrinsèque la plus grande possible, par exemple en utilisant les propriétés des colonnes échangeuses d'ions - situées en amont des patouillets de précipitation d'oxalate - de ne pas fixer plus de 300 g de plutonium. Mais pour ces systèmes à sécurité non inhérente, la sécurité repose en dernier ressort sur la stricte application de consignes administratives portant sur le contrôle visuel des appareils, leur rinçage, et la comptabilité de la matière fissile entrant en jeu à chaque cycle d'opérations. Il est certain que, lorsqu'on effectue un tel mode de contrôle, l'élément humain est prédominant; c'est pourquoi la consigne administrative doit faire ressortir clairement, d'une part le but visé, par exemple la masse à ne pas dépasser lors d'une opération, d'autre part les moyens mis en œuvre pour assurer la limitation imposée. De plus, il est nécessaire de s'assurer que la consigne a bien été comprise et qu'elle est exécutée correctement. Malgré toutes les précautions prises, une erreur humaine est toujours possible, surtout lorsqu'il s'agit d'opérations se répétant fréquemment; le responsable de la sécurité nucléaire doit toujours avoir le souci de cette possibilité d'erreur et prévoir une marge de sécurité suffisante pour pallier les erreurs prévisibles.

C. Troisième partie de l'usine

La troisième partie d'une usine de traitement de combustibles irradiés élabore des produits secs: poudres, telles que l'oxalate sec, l'oxyde et le fluorure de plutonium, ou bien plutonium métallique. Les installations correspondantes sont des systèmes à sécurité non inhérente, dans lesquels la sûreté nucléaire est assurée par un contrôle des masses de matières fissiles. Cette partie d'usine étant en fait un atelier de métallurgie mettant en œuvre des matières fissiles concentrées, nous l'évoquerons dans la dernière partie de ce mémoire.

D. Quatrième partie de l'usine: récupération des déchets de fabrication

Nous désirons mettre l'accent sur la partie d'une usine qui, d'après notre classification, est la dernière, et qui nous semble être de première importance sur le plan de la sûreté nucléaire: c'est la partie relative aux déchets ou sous-produits de fabrication et aux recyclages. On peut évaluer à environ 5% de la production d'une usine la quantité de plutonium à retraiter sous forme de déchets ou sous-produits, sans compter bien entendu les déchets éventuels de provenance extérieure: cette fraction est loin d'être négligeable et la variété des produits rencontrés est une source de difficulté et même de confusion si tout le soin désirable n'est pas apporté à leur stockage et à leur retraitement. On peut classer ces produits grossièrement en deux catégories: d'une part des sous-produits de routine: scories en provenance de l'atelier de métallurgie, échantillons de laboratoire; d'autre part les déchets occasionnels: papiers absorbants de nettoyage, gants, solution de rinçage, etc. Dans les deux cas il est nécessaire d'effectuer une gestion précise, même minutieuse, de la matière fissile; dans le premier cas cela est relativement aisé, car la gestion d'un atelier de métallurgie
est simple et les échantillons de laboratoire sont connus. Il n'en va pas de même dans le deuxième cas: c'est pourquoi nous cherchons à utiliser, pour les déchets solides, une méthode classique d'évaluation de la masse de plutonium fondée sur l'émission de photons $\gamma$ de 0,390 MeV par le plutonium-239; notre expérience n'est pas assez grande pour nous permettre d'estimer la valeur de la méthode. Soulignons que notre attention a été attirée sur l'importance du traitement des déchets en matière de sûreté nucléaire par l'analyse des accidents de criticité survenus aux États-Unis: sur six accidents, cinq en effet ont trait plus ou moins directement au traitement des déchets. Si nous examinons, à titre d'exemple, le dernier en date de ces accidents (récupération d'uranium enrichi, juillet 1964), nous constatons qu'il aurait suffi, pour éviter l'accident, d'étiqueter les bouteilles « sûres » renfermant la matière fissile; cette lacune relative à une opération très simple de gestion n'est certes pas la seule cause de l'accident; mais le fait est assez général: dans la plupart des cas, on constate que parmi les causes qui se conjuguent pour créer un accident de criticité, l'une d'elles est un défaut dans la gestion des matières fissiles.

II. SÉCURITÉ ET GESTION DANS LE CAS D'UN ATELIER DE METALLURGIE

Si le contrôle de la criticité est très varié dans une usine de traitement de combustibles irradiés et fait appel aux trois méthodes préventives ci-dessus, la sûreté nucléaire, dans un atelier de métallurgie mettant en œuvre des matières fissiles concentrées, est assurée de façon unique par une limitation de la masse. Cette limitation peut se faire soit à l'échelon de la cellule de fabrication soit à l'échelon de la boîte à gants. Dans ce dernier cas par exemple, une façon très simple de procéder consiste à indiquer sur la boîte la quantité de matière fissile autorisée et, en dessous de ce chiffre, la quantité réellement présente dans la boîte. Chaque entrée ou sortie de matière fissile oblige à un réajustement de la fiche de contrôle. Ce mode de contrôle minutieux, imposé par le risque de criticité, s'avère finalement très utile pour réaliser l'inventaire des matières fissiles présentes dans l'atelier.

On a parfois à mettre en œuvre, dans un atelier de métallurgie, des lots d'uranium enrichi à des taux d'enrichissement différents. Les normes de criticité variant suivant ces taux, il est important de vérifier qu'il n'y a pas confusion de lot. Une telle opération peut se réaliser grâce à un appareil de sécurité mis au point par le CEA [2]; cet appareil permet un contrôle rapide et non destructif de l'enrichissement en uranium-235 d'un combustible nucléaire quelconque.

Ce mémoire met en évidence l'importance, pour la sécurité nucléaire, à la fois de la conception de l'installation et de la gestion des matières fissiles dans l'installation; lors de la conception on cherchera à étendre au maximum, compte tenu des impératifs économiques, le domaine de la sécurité inhérente. Mais quelle que soit l'étendue de ce domaine, il est certain que les systèmes à sécurité non inhérente restent nombreux et finalement, c'est tout au long du procédé que la gestion des matières fissiles intéressera la sécurité, que ce soit comme contrôle primaire ou comme contrôle
secondaire. N'est-il pas paradoxal, en effet, de constater que les systèmes à sécurité inhérente - ou plus exactement des systèmes que l'on croyait tels - ont été à l'origine de nombreux accidents de criticité? C'est l'absence de gestion, liée à l'absence de contrôle de l'intégrité des systèmes sûrs, qui constitue la cause essentielle des accidents. C'est pourquoi nous pensons qu'une usine à sécurité intrinsèque parfaite n'existe pas. La sécurité nucléaire repose en définitive sur la compréhension et l'application par le personnel d'exploitation de consignes bien adaptées - et dans ces consignes la gestion des matières fissiles intervient de façon primordiale.

REFERENCES


DISCUSSION

J. SCHMETS: You say that you used alpha counters to determine the content of fissile materials in the "low-concentration" section of the Marcoule plant, and that they gave good accuracy. I wonder whether you did not encounter serious interference from other transuranic elements. Are the alpha counters capable of discrimination?

Y. FRUCHARD: There is in fact a certain amount of interference from other transuranic elements, such as plutonium-239, and it can be bothersome if one is dealing with irradiated fuels from different sources. This is not the case at Marcoule, however, where the irradiation to which the fuel has been subjected is well known. If the irradiation pattern is changed the counter has to be calibrated anew. To the best of my knowledge these counters are not capable of discrimination.

G.A. WELCH: You said that some odd waste materials from the operations at Marcoule are not yet being recovered. Can you tell me how you obtain fissile material balances without recovering the plutonium in these wastes, and how you determine the plutonium content of the wastes?

Y. FRUCHARD: Routine recovery products, such as slag from the metallurgical shops, are treated, as and when they are produced, in a device having unfavourable geometry; in this operation nuclear safety is guaranteed by a limitation on the mass of the plutonium. As to the odd waste products arising in the course of plant operations, such as absorbent cleaning papers, we prefer to postpone treatment of them until a calcinator can be put into operation; the ash will then be processed in a device having favourable geometry. In the meantime we estimate the mass of the plutonium in these wastes by gamma-spectrometry.

J.T. BYRNE: Do you use neutron absorbers such as Raschig rings containing boron to control criticality in your plutonium recovery plant?

Y. FRUCHARD: For a time we used cadmium plates immersed in the large vats which receive low-concentration plutonium solutions. However,
these plates did not permit the accurate homogenization of the solutions which is necessary for precise checks on concentration. This careful control of concentration is important for nuclear safety, moreover, in the equipment located further along in the cycle. The vats had other faults as well, and they have now been replaced by tanks without cadmium which we find to be satisfactory.

We are still using cadmium for containers with favourable geometry such as the flat reservoirs at Marcoule and the annular reservoirs at the plutonium plant in La Hague. So far we have made no regular use of boron; it has been used only once, in the form of boric acid, for a special purpose.
NUCLEAR MATERIALS MANAGEMENT AND ITS RELATIONSHIP TO NUCLEAR SAFETY AND CRITICALITY CONTROL

F.H. TINGEY
PHILLIPS PETROLEUM COMPANY, IDAHO FALLS, IDAHO, UNITED STATES OF AMERICA

Abstract — Résumé — Аннотация — Resumen

NUCLEAR MATERIALS MANAGEMENT AND ITS RELATIONSHIP TO NUCLEAR SAFETY AND CRITICALITY CONTROL. The preclusion of those conditions which would result in an inadvertent collection of fissionable material in quantity, geometry and environment to produce a neutron chain reaction is of fundamental importance to all those actively engaged in the nuclear industry. It is of particular concern to those individuals who are responsible for the handling, transfer and storage of the fissionable materials in the performance of their various tasks. These same people are part of the nuclear materials management (NMM) system. They are the individuals who are primarily responsible for generating and receiving the SS accountability data channelled through the NMM office and as such must depend upon that office for valid and timely information in discharging their responsibility.

Basically the responsibility for preventing inadvertent criticality in any area of plant operation lies with line supervision. In devising preventative measures at least three different types of problems need be considered. These are:

(a) The scientific or technical problems connected with evaluating the hazard presented by a proposed quality of fissile material in a given geometry and environment;
(b) The engineering problems presented by the restrictions dictated from the scientific or technical considerations;
(c) The purely administrative problems presented in implementing the restrictions and requirements imposed from the technical and engineering considerations.

To assist the supervisor in the successful discharge of this responsibility, some organizations provide certain groups or committees specialized in particular areas. Usually the technical problems are submitted to criticality specialists, commonly called a Nuclear Safety Committee, who are familiar with the experimental data of critical systems and can by theoretical calculation extend this experience to the problems at hand. Support in the engineering problems is given by plant engineering groups made cognizant of the requirements dictated by the technical considerations and familiar with the operational limitations. The administrative problems are solved through procedural restrictions placed by the supervisor.

The NMM office can be most valuable in further support to the supervisor in these activities. The primary responsibility of this office is to alert responsible supervision to potentially hazardous situations as indicated either from information made available through the routine processing of accountability data or from material balance calculations. Any information pertaining to criticality studies as requested from the NMM office must be accurate and precise. A vigorous inventory management policy must be pursued to lessen the problems presented by accumulation of fissile materials. Internal audit by the NMM staff of inventory holdings can also serve to verify adherence to safeguard procedures. A vigorous orientation programme can be implemented by the NMM staff in its dealing with those individuals handling nuclear materials to make them more aware of the hazards involved.

The integration of the NMM group into the general criticality safeguard problems at the NRTS has provided an effective system for assisting the line supervision in discharging this very important responsibility.

LA GESTION DES MATIERES NUCLEAIRES ET SES RAPPORTS AVEC LA SECURITE NUCLEAIRE ET LE CONTROLE DE LA CRITICITE. Il est essentiel pour tous ceux qui travaillent dans l'industrie nucléaire de savoir comment éviter que des matières nucléaires ne se trouvent rassemblées en quantités, selon une géométrie ou dans un milieu tels qu'elles puissent donner lieu à une réaction neutronique en chaîne. Ceci intéresse tout particulièrement ceux qui, dans l'exercice de leurs fonctions, sont responsables de la manipulation,
du transfert et de l’entreposage des matières fissiles. Ces mêmes personnes participent au système de gestion des matières nucléaires. Elles ont au premier chef la responsabilité de fournir et de recevoir les données comptables par l’intermédiaire du service de gestion des matières nucléaires; elles dépendent donc de ce service pour les renseignements précis dont elles ont besoin pour assumer leurs responsabilités.

C’est en premier lieu à la personne chargée de la surveillance générale du circuit qu’il incombe de veiller à ce qu’en aucun point les matières nucléaires ne puissent, par inadvertance, atteindre l’état critique. Pour mettre au point des mesures préventives, il faut tenir compte d’au moins trois catégories de problèmes, à savoir:

a) Les problèmes scientifiques et techniques liés à l’évaluation des risques que présente une certaine quantité de matières fissiles disposée selon une certaine géométrie dans un certain endroit;

b) Les problèmes matériels soulevés par les restrictions qu’imposent les considérations scientifiques ou techniques;

c) Les problèmes purement administratifs à résoudre pour respecter les restrictions et les conditions particulières imposées par les considérations techniques et matérielles.

Pour aider le responsable de la surveillance à s’acquitter de sa tâche, certaines organisations prévoient des groupes ou des comités spécialisés dans des domaines particuliers. Les problèmes techniques sont le plus souvent soumis à des spécialistes de la criticité, généralement constitués en comités de sécurité nucléaire, qui connaissent bien les données expérimentales des systèmes critiques et peuvent, par le calcul, exploiter cette expérience pour résoudre les problèmes qui se posent. En ce qui concerne les problèmes matériels, ils sont soumis à des groupes de techniciens que l’on met au courant des conditions particulières qu’imposent les facteurs techniques et qui connaissent bien les limites de fonctionnement de l’installation. Quant aux problèmes administratifs, le responsable de la surveillance se charge de les résoudre en réglementant les activités en conséquence.

Le service de gestion des matières nucléaires peut apporter une aide précieuse au responsable de la surveillance. C’est tout d’abord à ce service qu’il incombe d’alerter le service de surveillance en lui signalant les conditions présentant un danger potentiel qu’il a pu déceler par l’étude des données comptables ou par le bilan matières. Tous les renseignements demandés au service de gestion des matières nucléaires au sujet de la criticité doivent être exacts et précis. Un système efficace de contrôle de l’inventaire doit être appliqué pour limiter les problèmes que pose l’accumulation de matières fissiles. En faisant procéder par le personnel du service de gestion des matières nucléaires à une vérification interne des matières consignées dans l’inventaire, on peut également s’assurer que les mesures de sécurité sont bien appliquées. Le service de gestion des matières nucléaires peut aussi assurer la formation du personnel chargé de la manipulation des matières nucléaires afin de lui faire prendre davantage conscience des risques encourus.

En associant le service de gestion des matières nucléaires aux spécialistes de la prévention des états critiques, la National Reactor Testing Station (NRTS) dispose d’un système efficace pour faciliter la tâche du responsable de la surveillance du circuit matières.

ОБРАЩЕНИЕ С ЯДЕРНЫМИ МАТЕРИАЛАМИ И ЕГО СВЯЗЬ С ЯДЕРНОЙ БЕЗОПАСНОСТЬЮ И КОНТРОЛЕМ НАД КРИТИЧНОСТЬЮ. Для всех, активно работающих в области ядерной промышленности, чрезвычайно важно не допускать создания таких условий, которые могли бы привести к непредусмотренному скоплению расщепляющихся материалов в таких количествах, геометрии и окружении, которые могут вызвать целую реакцию нейтронов. Это особенно важно для тех, на ком лежит ответственность за погрузочно-разгрузочные работы, перемещение и хранение расщепляющихся материалов в ходе выполнения различных задач. Эти лица входят в систему административно-хозяйственного учета ядерных материалов. На обязанности этих лиц лежит в первую очередь ответственность за составление и получение данных статистической отчетности, направляемых через Управление по административно-хозяйственному учету ядерных материалов, и в качестве таковых при выполнении своих обязанностей они должны обращаться в указанное Управление за предоставлением им правильной и своевременной информации.

В основном ответственность за недопущение непредусмотренной критичности в любой зоне эксплуатации установки лежит на руководителе работ. При выработке предупредительных мероприятий следует учитывать по меньшей мере три различных вида проблем, а именно: а) проблемы научного или технического характера, связанные с определением степени опасности, возникающей в результате сосредоточения в определенной геометрии и окружении намечаемых количеств расщепляющихся материалов; б) технологические проблемы, вытекающие из диктуемых научными и техническими соображениями ограничений; в) чисто администра-
La administración de materiales nucleares y sus relaciones con la seguridad nuclear y el control de la crítica.

Para las personas que desempeñan funciones importantes en la industria nuclear es de suma importancia evitar las acumulaciones accidentales de material fisionable en cantidades, medios y disposiciones que puedan ocasionar una reacción neutónica en cadena. Esto concierne especialmente a los encargados de la manipulación, la transferencia y el almacenamiento de dichos materiales. Estas personas forman parte del sistema de administración de materiales nucleares y son fundamentalmente responsables de facilitar y recibir, por conducto de los servicios de administración, datos contables relativos a los materiales básicos y a los materiales fisionables especiales; para que puedan desempeñar satisfactoriamente sus funciones, es preciso que los servicios dispongan de información válida en el momento oportuno.

Básicamente, la prevención de los accidentes de crítica en cualquier etapa del funcionamiento de una planta es una de las funciones inherentes a la inspección de las operaciones.

Al proyectar las medidas preventivas hay que considerar, por lo menos, tres grupos distintos de problemas:

a) Los problemas científicos y técnicos relativos a la evaluación de los riesgos que presenta la acumulación de materiales fisionables en cantidades, disposiciones y medios determinados;

b) Los problemas de ingeniería que plantean las restricciones basadas en consideraciones de carácter científico o técnico;

c) Los problemas puramente administrativos que suscitan las limitaciones impuestas por cuestiones técnicas y de ingeniería.

Para facilitar el desempeño de las funciones de los inspectores algunas organizaciones han constituido comités especializados en determinados aspectos de la cuestión. Por lo general, los problemas técnicos se someten a especialistas en crítica, comúnmente agrupados en un comité de seguridad nuclear, quienes conocen los datos experimentales de los sistemas críticos y pueden por cálculo teórico extender ese conocimiento a los problemas específicos de la planta. Para los problemas de ingeniería se cuenta con la cooperación de los grupos de ingenieros, a los cuales se mantiene informados de las necesidades impuestas por consideraciones técnicas y de las limitaciones operativas. Los problemas administrativos se resuelven mediante restricciones de procedimiento dictadas por el inspector.

Los servicios de administración de materiales nucleares pueden facilitar en gran medida las tareas de inspección. En primer lugar, estos servicios tienen el deber de alertar a los inspectores cuando la información...
de que disponen gracias a la sistematización de los datos contables o a cálculos basados en el balance de materiales revela la existencia de una situación que puede resultar peligrosa. toda la información referente a cuestiones de criticidad facilitada por los servicios de administración de materiales nucleares debe ser exacta y precisa. Es necesario seguir una política rígida de administración de existencias a fin de limitar los problemas que suscita la acumulación de materiales fisionables. La comprobación interna de las existencias por el personal de los servicios de administración puede servir también para verificar si se aplican los procedimientos de salvaguardia. Este personal puede llevar a cabo una importante labor de orientación informando a los encargados de manipular los materiales fisionables sobre los peligros que sus actividades implican.

La orientación de los servicios de administración de materiales nucleares hacia los problemas relativos a la prevención de la criticidad aumenta considerablemente la eficacia de las inspecciones.

La orientación de los servicios de administración de materiales nucleares hacia los problemas relativos a la prevención de la criticidad aumenta considerablemente la eficacia de las inspecciones.

The preclusion of those conditions which would result in an inadvertent collection of fissionable material in quantity, geometry, and environment to produce a neutron chain reaction is of fundamental importance to all those actively engaged in the nuclear industry. It is of particular concern to those individuals who are responsible for the handling, transfer and storage of the fissionable materials in the performance of their various tasks. These people are a part of the nuclear materials management system. They are the individuals who are primarily responsible for generating and receiving the SS accountability data channelled through the nuclear materials management (NMM) office and as such must depend upon that office for valid and timely information in discharging their responsibilities.

Basically the responsibility for preventing inadvertent criticality in any area of plant operation lies with line supervision. In devising preventive measures, at least three different types of problems are presented. These are given in TID 7016 Rev. 1, USAEC Nuclear Safety Guide, as:

(a) The purely scientific problems connected with the conditions needed for a chain reaction. Such conditions are usually stated in terms of a number known as the critical or chain-reacting mass, being that quantity of fissionable material which is critical under the conditions stated. Although such problems usually can be precisely formulated, the quality of the information needed and the computational difficulties experienced in effecting a solution in a given case are such as to make the purely theoretical solutions to these problems relatively imprecise except in the simplest cases. Thus it is necessary to depend on experimental measurements of critical mass, and on short extensions of these by theory.

(b) In addition, there are the problems of an engineering type. These depend on the particular circumstance peculiar to the situation being considered. It is not sufficient to examine a process or configuration which would result from a normal course of events, but rather consideration must be given to the nature and consequence, with regard to criticality, of variations in conditions.

(c) The third type of problem is that usually classified as administrative. Consideration of the scientific and engineering aspects of a given nuclear safety problem usually results in certain conditions, restrictions, or requirements being imposed on the system. These restrictions are implemented through rules and procedures. It is necessary to consider these rules in detail in order to determine what violations of procedures, whether intentional or not, are possible, and what physical controls exist to minimize these violations.
To assist the supervisor in the successful discharge of his criticality responsibilities, some organizations provide certain groups or committees specialized in particular areas. Usually the technical problems are submitted to criticality specialists commonly called a Nuclear Safety Committee. The members of this committee are familiar with the experimental data of critical systems and can by theoretical calculation extend this experience to the problems at hand. Support in the engineering problems is given by plant engineering groups made cognizant of the requirements dictated by the technical considerations and familiar with the operational limitations. The administrative problems are solved through procedural restrictions placed by supervision.

The NMM office can be most valuable to supervision in support of these activities. First and foremost, this office has a responsibility to alert supervision to potentially hazardous situations as indicated either from information made available through the routine processing of accountability data, or from material balance summaries. This presupposes some knowledge by the NMM staff of process operating conditions and their relationship to nuclear safety. In the recovery of reactor fuel elements, for example, capacities of various measurement vessels, stream concentrations, normal process losses, in-plant hold-up, operating conditions, etc., should be and usually are a matter of common knowledge to the NMM staff, thus any deviation or departure from the expected as observed through the routine processing of the data can and should be called to the attention of responsible supervision.

At the USAEC Idaho Chemical Processing Plant (ICPP), since the NMM office is the principal user and interpreter of measurement data, it has become its responsibility through supervision to devise for each processing run a "dissolver operating line" for criticality control. This line defines the total allowed dissolver content when both random and systematic uncertainties of dissolver measurement and material residual content are taken into consideration. The limit determined for each dissolver is based upon criticality computations performed by the Nuclear Safety Committee as related to the particular solution involved, geometry of the vessel, moderation, and interaction with adjacent vessels. Such detailed considerations are not required for all dissolvers at ICPP, since some are nuclearly safe by geometry and concentration control and others are made safe through the batch by batch addition of a soluble neutron poison. As a result of the systematic uncertainties in the measurement generation scheme, the net effect of the operating line is to force a dissolver clean-out after a certain number of batches. Of course, if material is in fact accumulating in the dissolver, heel reduction steps are required by the line earlier than might otherwise be the case.

The plant material balance is fundamentally a safeguards as well as an accountability tool. The timeliness and validity of this calculation is closely related to process and criticality control. Material unaccounted for in a fuel reprocessing plant, for example, presents a problem of greater concern from the nuclear safety standpoint to the plant manager than it does from simply the represented financial loss. The subtle accumulation of material in a vessel whose content is not one of the components of the material balance can be disastrous.
Under the custodian system of NMM control, the NMM office maintains a record of each custodian's holdings. Thus he is aware of the quantity of material in the various repositories about the site and of any proposed changes in these inventories. Quantities less than a critical mass under any conditions of interaction can be made negligible through plant administrative control. However, when a custodian's material holdings approach critical mass sizes, the above may not be true. The NMM can call this to the attention of the responsible custodian and thus alert him to the potentiality of a criticality hazard. In so doing, competent review can be made before transfer and storage is actually accomplished. A particular example in this respect at the ICPP is the underwater storage of fuel elements in reactor canals and storage basins. This is a particularly sensitive criticality control area since arrays of items non-critical in air can become critical when immersed in water. To avoid such a contingency, a comprehensive review of each proposed storage condition for each type element is made before the actual storage. The review is based upon accountability records which are supplemented after the review by a detailed description and verification of the approved storage methods by the NMM staff. Any subsequent transfers or movements within the grid or array are made from the NMM records and are verified by a member of the NMM staff.

The NMM inventory system can easily be extended to encompass not only item and quantity accounting but location accounting as well. By this is meant the storage arrangement under which the SS material items are held. Usually a review is made as to the potential criticality problems of handling and storage. As a result of this review, procedures are defined and documented as to the mechanics of storage. With reference to these procedures, all inventories can then be verified for compliance with the established safeguard procedures as well as accounting practices. For example, at the National Reactor Testing Station (NRTS) a monthly listing of SS material holdings is provided to custodians. This listing contains, among other things, item identification and SS quantity. The custodian is required to physically verify the inventory and indicate by signature his concurrence with same. In addition, he is asked to indicate where each item is stored so that compliance with safe storage practices can be determined. Semi-annually the NMM staff makes an audit of each custodian and reports to management any violations of the established accountability requirements or of the approved practices with regard to storage and handling. Bases for review are Standard Practices for Storage and Handling compiled by each operating and technical group and reviewed by the Nuclear Safety Committee. Familiarity with the specific practice is required on the part of the NMM auditor before audit.

Another function of the NMM having a bearing on criticality safeguards is that of SS material inventory management. At the NRTS there are on inventory at any given time in excess of 10,000 individual fissile material bearing items not including that in the form of process solution. Many of these items are of the experimental variety and contain individually negligible quantities of material. However, the accumulation of such items does have criticality ramifications. Therefore, it is the interest of nuclear safety as well as good inventory practice to keep such inventories to a minimum. The NMM staff attacks this problem from two directions. First, by the
monthly listing of all items in inventory, NMM is in a position to determine for new procurements whether such an item is on inventory with another custodian. Since all requirements for SS material bearing items must be cleared through the NMM office, this prevents an unnecessary acquisition. Secondly, by examining the activity or use of all items on inventory, the NMM manager is in a position to recommend to management which items should be declared surplus and thus made available to other sites or the SS material recovered through chemical reprocessing. At the NRTS a listing of inactive items is supplied to custodian annually. He is required to justify in writing further retention of those items. Failure to do so results in the inclusion of these items in a SS surplus list and ultimate removal from the site.

Proper implementation and co-ordination of the accountability function with criticality safeguards requires an organizational structure which provides easy access to management and direct liaison with safeguards. This access and liaison is facilitated at the NRTS by the NMM group reporting directly to the plant manager through the Chairman of the Nuclear Safety Committee. Thus problems of mutual concern are automatically brought into proper focus.

It is apparent from the above that effective management of nuclear materials embraces far more than the simple accounting function as is usually assumed. An alert and aggressive NMM organization can render valuable service in many areas of operation, not the least of which is the avoidance of an inadvertent criticality.
APPLICATION OF REACTOR PHYSICS CALCULATION METHODS TO CRITICALITY CONTROL OF NON-INHERENTLY SAFE SYSTEMS IN FUEL PROCESSING PLANTS

M. RÄBER
UNITED NUCLEAR CORPORATION,
WHITE PLAINS, N. Y., UNITED STATES OF AMERICA

(presented by A. H. Kazi)

Abstract — Résumé — Аннотация — Resumen

APPLICATION OF REACTOR PHYSICS CALCULATION METHODS TO CRITICALITY CONTROL OF NON-INHERENTLY SAFE SYSTEMS IN FUEL PROCESSING PLANTS. The economics of fuel processing operations involving the handling of large quantities of fissionable materials can be significantly improved by utilizing somewhat detailed reactor physics calculation methods to establish criticality safe process parameters. These calculation techniques serve as an extension of critical experiment data to systems not specifically covered by such data and may be used to establish the safety of larger mass limits and process equipment dimensions than could be justified by conservative extrapolation of the available experimental data.

This paper describes some of the calculation techniques used for this purpose at United Nuclear Corporation. Both hand calculation techniques and computerized techniques are discussed in connection with highly enriched uranium alloy water systems, poison-wrapped cylinders, fixed poison sheets in unsafe geometry tanks, and safety arrays of unmoderated uranium materials; and generally useful data obtained with these methods are presented. Comparison of these calculation methods against experimental data, and the assumptions made in applying these methods to criticality safety work, are also discussed.
лов при больших размерах технологического оборудования, чем это можно было бы оправдать на основании тщательно рассчитанной экстраполяции имеющихся экспериментальных данных.

Дается описание некоторых вычислительных методов, применяемых с этой целью компанией "Юнайтед ньюклон копорейшн". Обсуждаются простые приемы расчетов вручную и методы расчетов на счетно-решающих устройствах относительно водяных систем с высокообогащенным ураном, цилиндров с оболочкой из поглотителя, фиксированных поглотительных пластин в емкостях небезопасной геометрии и безопасности целого ряда урановых материалов без замедлителя, и сообщаются полученные с помощью этих методов общеполезные данные. Данные, полученные этими методами сравниваются с экспериментальными данными, а также обсуждаются гипотезы, принятые при применении этих методов к проблемам обеспечения безопасности с точки зрения критичности.

APLICACION DE METODOS DE CALCULO DE FISICA DE REACTORES AL CONTROL DE LA CRITICIDAD DE SISTEMAS NO INHERENTEMENTE SEGUROS EN LAS PLANTAS DE TRATAMIENTO DE COMBUSTIBLE. La rentabilidad de las operaciones de tratamiento de combustible que requieren la manipulación de grandes cantidades de materiales fisiofables pueden mejorarse considerablemente mediante el empleo de métodos de cálculo relativamente minuciosos de física de reactores para dar a los parámetros del proceso valores seguros en lo que respecta a la criticidad. Esas técnicas de cálculo permiten extender los datos provenientes de experimentos críticos a sistemas no cubiertos específicamente por dichos datos y pueden usarse para establecer la seguridad de masas y equipos de dimensiones mayores que las que justificaría una extrapolación prudente de los datos experimentales disponibles.

En la memoria se describen algunos de los métodos de cálculo que con ese fin se usan en la United Nuclear Corporation. Se analizan tanto las técnicas manuales simples como las que se basan en el empleo de calculadoras electrónicas para los sistemas formados por aleaciones de uranio muy enriquecido y agua, los cilindros envueltos de venenos nucleares, las láminas de venenos nucleares en tanques de geometría peligrosa y la seguridad de los conjuntos de materiales de uranio no moderados, y se presentan algunos datos de utilidad general obtenidos con esas técnicas. Se establece una comparación entre los métodos de cálculo y los datos experimentales y se exponen las suposiciones de que se parte al aplicar dichos métodos a las tareas encaminadas a impedir la criticidad.

1. INTRODUCTION

The processing of large quantities of fissionable material presents an important potential hazard, that of accidental criticality. The necessity of providing adequate safeguards against such an occurrence frequently imposes severe restrictions on processing methods and equipment, and, hence, on process economics. These safeguards generally take the form of limitations on mass of fissionable material or limitations on the geometry and arrangement of process equipment. They are derived usually from experimental data that have been generated for this purpose. Although a considerable quantity of data is available, it by no means covers all situations of interest to the fuel processor. Highly conservative extrapolations of existing data must often be made, resulting in a relatively inefficient and less economical process.

At United Nuclear Corporation, the co-operative efforts of the Fuels Division and the Development Division have led to improvements in the efficiency and economics of many fuel processing operations through the use of detailed reactor physics calculation methods to establish criticality-safe process parameters. These calculation techniques, which have been developed primarily for use in reactor design, are used to establish the safety of larger mass limits and process equipment dimensions that could be justified by conservative extrapolation of existing experimental data.
The applications of reactor design techniques to criticality safety problems that are discussed in this paper are summarized in Table I. Simple recipes amenable to hand calculation have been developed to compute the reactivity of well-moderated uranium-water systems. Computerized techniques, such as multigroup transport theory, are utilized when simple recipes are inadequate. For example, they are used when highly absorbing poison regions must be treated explicitly, or when the neutron spectrum of the system is non-thermal and is a strong function of position.

The applications discussed in this paper deal specifically with the processing of highly enriched uranium (~90% $^{235}$U) because many difficult criticality problems arise in conjunction with this material. However, the techniques are also applicable, with some modification, to uranium of lower enrichment and to plutonium systems.

**TABLE I — APPLICATIONS OF REACTOR DESIGN METHODS TO CRITICALITY SAFETY PROBLEMS**

<table>
<thead>
<tr>
<th>Type of System</th>
<th>Purpose of Calculation</th>
<th>Calculation Method</th>
</tr>
</thead>
<tbody>
<tr>
<td>Well-moderated U solution or U-alloy- water mixture</td>
<td>Determine critical mass; determine $k_{\text{eff}}$ for safe masses</td>
<td>Modified one-group hand calculation</td>
</tr>
<tr>
<td>Poison-wrapped cylinders of U solution, cylinder diam greater than 5 in.</td>
<td>Determine minimum critical diameter; establish adequacy of specific poison wraps</td>
<td>Multigroup transport theory (DTF)</td>
</tr>
<tr>
<td>Fixed poisons in unsafe geometry vessel for U solution</td>
<td>Determine number of poison sheets needed to provide safety</td>
<td>Simplified control rod theory (modified area absorption)</td>
</tr>
<tr>
<td>Arrays of unmoderated U materials</td>
<td>Determine $k_{\text{eff}}$ for one unit; establish safety of array under conditions of partial moderation</td>
<td>Multigroup transport theory (DTF)</td>
</tr>
</tbody>
</table>

2. **PREDICTION OF REACTIVITY AND CRITICAL MASS FOR URANIUM SOLUTIONS AND URANIUM ALLOY-WATER MIXTURES**

2.1. **Uses of simple recipes for reactivity calculations**

Simple, modified one-group recipes for predicting the critical mass of highly enriched uranium in solutions and in alloy-water mixtures have been developed for convenient estimation of safe batch sizes for processing
equipment such as pickling and washing tanks, autoclaves, and storage racks.
In these recipes (described in section 2.2), non-homogeneous mixtures (in
which the uranium is in the form of plates or rods) are treated by approp-
riate application of disadvantage factors. Relatively few experimental data
are available for heterogeneous U-Al and U-Zr systems, and these data are
difficult to extrapolate directly to compositions and degrees of self-shielding
outside the range covered by experiment. If self-shielding in alloy pieces
is neglected, and criticality safety parameters are based on the relatively
abundant data for homogeneous solutions, these safety parameters will be
overly conservative. By taking self-shielding into account, the method con-
sidered here permits larger equipment dimensions or mass limits.

These recipes are useful in estimating \(k_{\text{eff}}\) values for individually safe
batches of fuel or pieces of equipment in order to establish safe spacings
between such units. The criteria developed at Oak Ridge [1, 2] for the safe
spacing of individually subcritical units are frequently used at United Nuclear
Corporation. According to these criteria, the safe spacing between a number
of fuel units is a function of the \(k_{\text{eff}}\)'s of the individual units and of the total
solid angle (interaction angle) subtended at the most central or most reactive
unit of the group by all other units (units shielded from the view of the cen-
tral unit are not included). The spacing is safe when the maximum inter-
action angle does not exceed the value given by the following relations:

\[
\Omega = 9 - 10 k_{\text{eff}}; \quad 0.3 \leq k_{\text{eff}} \leq 0.80
\]

\[
\Omega = 6.0 \text{ steradians for } k_{\text{eff}} < 0.3
\]

Method not applicable to units with \(k_{\text{eff}} > 0.80\)

where \(\Omega\) = interaction angle, steradians.

Interaction angles may be computed using the approximate formulas
given in Ref. [1] or the more exact curves for specific geometries presented
in Ref. [3].

2.2. Description of reactivity calculation method

The reactivity of a water-moderated system fuelled with highly enriched
uranium can be expressed as

\[
k_{\text{eff}} = \frac{\eta f}{1 + (L^2 + \tau)B^2_8} \quad (2)
\]

\[
f^{-1} = 1 + \sum_{i} \frac{A_{23}}{A_i} \frac{\sigma_{ai}}{\sigma_{25}} \left( \frac{W_i}{W_{25}} \right) F_i \quad (3)
\]

where \(\eta\) = regeneration factor, 2.07 for \(^{235}\)U, \(f\) = thermal utilization of fissile
material, \(L^2\) = thermal diffusion area, \(\text{cm}^2 \approx 8 (1 - f)\), \(\tau\) = age to thermal
energy (\text{cm}^2), \(B^2_8\) = geometric buckling (\text{cm}^{-2}), \(A\) = atomic or molecular weight,
\[ L^2 + \tau = M^2, \sigma_a = \text{microscopic absorption cross-section} \ (b), \ W = \text{mass} \ (g), \]

subscript 25 = 235U, subscript i = any material other than 235U, and

\[ F_i = \frac{\text{avg thermal neutron flux in component i}}{\text{avg thermal neutron flux in } 235\text{U}} = \text{disadvantage factor}. \]

Absorption cross-sections averaged over a room-temperature Maxwellian spectrum are used. A correction factor of 0.98 is applied to the 235U cross-section to account in part for its non-1/\(v\) behaviour. \(B\) depends on system geometry, and is computed using the usual formulas [4], with effective extrapolation distances of 2.5 cm for unreflected systems and 6.5 cm for systems surrounded by a thick water reflector. For uranium solutions, the \(\tau\) data presented in Ref. [5] may be used. Checks against critical experiment data on uranium solutions [6] show that this calculation model is slightly conservative (overpredicts \(k_{\text{eff}}\) by 0 to 2\%) for dilute solutions (H/235U > 1000) and is generally conservative by 2 to 6\% in \(k_{\text{eff}}\) for concentrated solutions. Illustrative check calculations are given in Table II.

**TABLE II — COMPARISON OF CALCULATION METHOD WITH CRITICAL EXPERIMENTAL DATA — URANIUM SOLUTIONS AND U-AL SLUGS IN WATER**

<table>
<thead>
<tr>
<th>(\text{UO}_2\text{F}_2) Solutions in Spherical Containers(^a)</th>
<th>(\text{Calculated }k_{\text{eff}})</th>
</tr>
</thead>
<tbody>
<tr>
<td>Diameter (cm)</td>
<td>(H/U^{235}) Atom Ratio</td>
</tr>
<tr>
<td>23.0</td>
<td>76.1</td>
</tr>
<tr>
<td>23.6</td>
<td>126.5</td>
</tr>
<tr>
<td>55.8</td>
<td>1112</td>
</tr>
<tr>
<td>55.8</td>
<td>1270</td>
</tr>
<tr>
<td>69.2</td>
<td>1393</td>
</tr>
</tbody>
</table>

**U-Al Slugs in Water\(^c\)**

<table>
<thead>
<tr>
<th>No. of Slugs</th>
<th>Edge-to-Edge Spacing (in.)</th>
<th>Metal/Water Volume Ratio</th>
<th>Calculated Disadvantage Factor</th>
<th>(k_{\text{eff}}) for Experimental Array, Modified One-Group Recipe</th>
</tr>
</thead>
<tbody>
<tr>
<td>162</td>
<td>0.38</td>
<td>0.92</td>
<td>1.78</td>
<td>1.028</td>
</tr>
<tr>
<td>150</td>
<td>0.50</td>
<td>0.72</td>
<td>1.88</td>
<td>1.042</td>
</tr>
<tr>
<td>162</td>
<td>0.63</td>
<td>0.58</td>
<td>1.98</td>
<td>1.052</td>
</tr>
<tr>
<td>215</td>
<td>0.75</td>
<td>0.48</td>
<td>2.08</td>
<td>1.057</td>
</tr>
</tbody>
</table>

(a) Data from [6]. U enriched to 93.2\% U\(^{235}\). Al container wall about 0.2-cm thick.
(b) See section 3.1
(c) Data from [8]. Slugs are 1.35-in. diameter \(\times\) 8.00-in. long; -7.6 wt\% U enriched to 93.3\% U\(^{235}\). Slugs are arranged on a square pitch in square or rectangular arrays, one tier high. Arrays are flooded and reflected by water.

Extending this type of calculation to heterogeneous uranium alloy systems requires the computation of disadvantage factors and the generation
of accurate $\tau$ data for mixtures of Al or Zr and water. Reasonably accurate
disadvantage factors in either slab geometry or cylindrical geometry can
be computed using the integral transport theory formulations of Theys and
Amouyal and Benoist (See Ref. [6]).

The $\tau$ data for Zr-water and Al-water mixtures have been computed
using the FORM code [7]. This is a FORTRAN version of the more familiar
MUFT code in use on United Nuclear's CDC-1604-A computer. Age to
thermal energy (0.625 eV) for various Al-water and Zr-water mixtures was
computed with FORM. For both systems, $\tau$ was found to be a linear function
of metal to water volume ratio, $V_m/V_{H_2O}$, up to a value of $V_m/V_{H_2O}$ of 1.60.
The $\tau$ could be expressed by:

$$\tau = 26.8 + 30.6 \left( \frac{V_m}{V_{H_2O}} \right) \text{ for Al-H}_2\text{O mixtures} \quad (4)$$

$$\tau = 26.8 + 19.5 \left( \frac{V_m}{V_{H_2O}} \right) \text{ for Zr-H}_2\text{O mixtures} \quad (5)$$

In using these expressions, the effect on $\tau$ of uranium present in dilute alloys
is neglected.

The above reactivity calculation method has been checked against cri-
tical experiment data on heterogeneous U-Al-water and U-Zr-water systems.
In all cases, the calculated values of $k_{eff}$ were found to be conservative,
sometimes by 5% or more. Illustrative check calculations are shown in
Table II. The corresponding safety factors on critical mass are in the range
of 30 to 50%. This allows margin to cover uncertainties when the calculation
method is extended to materials for which there are no experimental data.
In practice, it has been found possible to more than double previously ac-
cepted safe mass limits, based on homogeneous solution critical data, for
some uranium alloy pieces by taking self-shielding into account using this
calculation method. In all critical mass calculations, a large safety factor
($\sim 2.3$) is applied to calculate values to allow for double-batching accidents.

3. CRITICALITY SAFETY OF POISON-WRAPPED CYLINDERS

3.1. Uses of poison-wrapped cylinders

Equipment used for the chemical processing of fuels consists largely
of pipes and long cylinders such as storage tanks for uranium solutions,
and shells of shell and tube heat exchangers. Experiments with highly en-
riched uranium in solutions have established that a long 5-in.diameter cy-
linder cannot be made critical regardless of the uranium concentration in
solution and degree of reflection by water. Five inches is therefore ac-
cepted as an "always safe" diameter for solution containers. The safe use
of larger diameter piping or long cylinders with highly enriched uranium
generally requires control of uranium mass, uranium concentration, or
degree of reflection. Each of these controls is subject to failure through
human error or equipment malfunction, and, therefore, is less desirable
than the use of "always safe" geometry.
Long cylinders having inside diameters greater than five inches can be converted to "always safe" geometry by constructing them of, or wrapping them with, a nuclear poison. The safety of poison-wrapped cylinders has been investigated using the United Nuclear's DTF transport theory code. The DTF code, which is similar to the Los Alamos DTK code, is a one-dimensional multigroup programme for solving the neutron transport equation using the $S_n$ approximation. The DTF calculations were performed using the 16-group Hansen-Roach cross-section library [9]. The $S_4$ approximation was used, and anisotropic scattering by hydrogen was taken into account. This calculation model has been widely used at United Nuclear in the design of non-thermal reactors and has been checked against a variety of critical experiments, including Godiva, Topsy, and a number of ZPR-III assemblies [10]. Its ability to predict the reactivity of non-thermal systems to within about ±2% has been established.

The applicability of this model to uranium solution systems was established by applying it to critical experiment data on solution spheres and cylinders [6, 11]. The reactivity of these systems was predicted to within about ±1.5%. Some check calculation results are given in Table II.

### 3.2 Cylinders wrapped with cadmium

Experimental data are available for highly enriched UO$_2$F$_2$ solutions in stainless steel cylinders (1/16-in.wall) wrapped with 0.020 in.of cadmium and surrounded by a thick water reflector [6]. For each of several cylinder diameters, critical height was measured as a function of $^{235}$U concentration. The minimum critical heights are shown in Fig. 1. Using DTF calculations, in which the cylinder wall and cadmium liner were treated explicitly, these data have been extended to the case of an infinitely long cylinder. Critical inside diameter was calculated as a function of $^{235}$U concentration in solution. The minimum critical diameter was found to be 7.5 in. This is shown as the vertical asymptote in Fig. 1. Allowing for a safety factor, this calculation shows that cylinders having diameters up to 6.5 to 7 in.can be made "always safe" for solutions by wrapping them with a nuclear poison equivalent to 0.020 in of cadmium.

### 3.3 Borosilicate glass pipes

A piping material that normally contains a strong nuclear poison is borosilicate glass. Nominal 6-in.diameter borosilicate glass pipes are available commercially. DTF calculations were performed to determine whether or not the normal boron content of these pipes would be sufficient to make them "always safe" for uranium solutions. The standard pipes [12] are 6.0±1/32-in.ID and 6.656-in.OD. The glass has a density of 2.23 g/cm$^3$, and contains about 3.7 wt.% boron in the form of B$_2$O$_3$. The glass composition given in Ref. [13] was assumed.

The $k_{eff}$ was calculated as a function of uranium concentration in UO$_2$F$_2$ solutions contained in an infinitely long borosilicate glass pipe surrounded by a 15-cm water reflector. This reflector is a conservative representation of reflection effects likely to be encountered in practice. For example, these
effects may be due to nearby concrete walls or tanks of liquid, or due to accidental flooding of the area in which the pipe is located. A uranium enrichment of 93.5% and a pipe ID of $6\frac{1}{32}$-in. were assumed. A plot of $k_{eff}$ versus $^{235}U$ concentration yielded a maximum $k_{eff}$ of 0.83. This is sufficiently below 1.0 to ensure safety regardless of solution concentration or presence of a water reflector even after some allowance is made for possible non-conservative calculational bias. A similar set of calculations was performed for an unreflected pipe. The maximum $k_{eff}$ was found to be 0.70. This figure is useful in establishing safe spacings between adjacent pipes or between pipes and other pieces of equipment using the interaction angle criteria discussed in section 2.1. The maximum permissible interaction angle for a standard 6-in.pipe is 2.0 steradians.

In addition to a 50% increase in flow area over a 5-in.glass pipe, an economic bonus is obtained by using the standard 6-in.pipe because 5-in. pipes are non-standard items [12]. The latter are substantially more expensive and delivery times are longer.
### 3.4. Steel pipe wrapped with poison material

Ordinary schedule 40, 6-in. diameter steel pipe can be converted to an always safe vessel for uranium solutions by wrapping nuclear poison around the outside of the pipe. Cadmium would be a good choice of poison material from nuclear and economic considerations because it has adequate nuclear worth in thin sheets, it is relatively cheap, and it is easy to form into a wrap. Unfortunately, its low melting point of 609°F makes it undesirable for use in areas where the possibility of fire exists. Other suitable poison materials that are more fire-resistant include boron-steel and boral (B₄C in Al).

A series of DTF calculations was performed for a schedule 40, 6-in. diameter stainless steel pipe wrapped with boron-steel. These calculations accounted for the effects of the pipe wall and the steel in the poison wrap. The pipe was assumed to have an ID of 6.065 in., an OD of 6.625 in., and a composition of 71 wt.% Fe, 11 wt.% Ni, and 18 wt.% Cr. The wrap was assumed to be boron-stainless steel containing 1 wt.% natural boron (67 wt.% Fe, 14 wt.% Ni, 18 wt.% Cr). A wrap thickness of 5/16-in. was assumed, giving a boron surface density (g B/cm² x thickness in cm) of 0.063 g/cm². The maximum $k_{\text{eff}}$ for this pipe surrounded by a thick water reflector was calculated to be 0.89. This is sufficiently below 1.0 to ensure safety for all solutions, but it would be undesirable to have a $k_{\text{eff}}$ higher than 0.90. Therefore, if boron is to be used as a poison wrap, the boron surface density of the wrap should not be much less than 0.063 g/cm². For this pipe not reflected by water, the maximum $k_{\text{eff}}$ was calculated to be 0.80. The presence of almost 0.6 in. of steel around the solution limits the permissible interaction angle at a pipe to 1.0 sr. Since steel is a more efficient reflector than aluminium or thin sheets of cadmium, these reactivities should be conservatively applicable to pipes wrapped with the latter materials.

### 4. USE OF FIXED POISON IN NON-SAFE GEOMETRY VESSELS

It is often impractical to handle large quantities of uranium solution exclusively in small, safe geometry vessels. One method of assuring the nuclear safety of a relatively large vessel of unsafe geometry is to fix sheets of nuclear poison material into it. These sheets perform the same function as control rods in a reactor. This technique is more reliable than control of solution concentration or use of soluble poison where this is permissible. Suitable fixed poison materials include boron-steel, boral, cadmium sheet (suitably sandwiched between sheets of cladding material to provide structural strength), and borosilicate glass (particularly in laminated "safety" glass form). The choice of material depends on solution chemistry and relative cost.

Simple control-rod theory provides a means of computing the number of poison sheets required. The reactivity of a vessel containing uranium solution and poison sheets can be expressed as:

$$
(k_{\text{eff}})_{p} = k_{\text{eff}} \left[ 1 - \left( \frac{\Delta k}{k} \right)_{p} \right]
$$

(6)
where $k_{\text{eff}}$ is the reactivity of the vessel with no poison sheets and $(\Delta k/k)_p$ is the reactivity worth of the poison. For slab absorbers, $(\Delta k/k)_p$ can be calculated by a modification of the "absorption area" method [14, 15]:

$$
\left( \frac{\Delta k}{k} \right)_p = \frac{AR}{V} \left[ \frac{L'}{2} + \frac{T}{2} \right]
$$

where $A =$ total surface area of poison sheets (cm$^2$), $V =$ gross volume of vessel (cm$^3$), $T =$ thickness of poison sheet (cm), $L' =$ effective diffusion length for thermal neutrons in the solution, $L' \approx 0.7L$ ($L$ may be calculated per method of section 2.2), and $R =$ relative worth factor which accounts for absorption of epithermal neutrons.

The usual "absorption area" formula has been modified by the $R$ factor to include the effect on reactivity of the absorption of epithermal neutrons. $R$ is a function of the surface density of the absorber material. Empirical values of $R$, appropriate for well-moderated hydrogenous systems, are given in Ref. [15] for a number of poison materials. In the above formula, the actual volume of the poison material has been added to the "absorption area" in computing $(\Delta k/k)_p$. For criticality safety applications, the poison will occupy a significant portion of the vessel volume (perhaps as much as 25%). Therefore, neglecting the volume of poison would be overly conservative.

For solutions of highly enriched uranium, Eqs.(2), (6) and (7) can be combined to yield:

$$
\frac{A}{V} = \left[ 1 - \frac{(k_{\text{eff}})_p(1 + M^2B_0^2)}{\eta I} \right] \frac{1}{R \left( \frac{L'}{2} + \frac{T}{2} \right)}
$$

Since this recipe assumes that the poison sheets are black to thermal neutrons, it should not be used with a cadmium surface density less than 0.44 g/cm$^2$ (~0.020 in. of Cd) or with a boron surface density less than 0.04 g/cm$^2$. $R$ is 1.0 for boron and cadmium at these surface densities [15].

This recipe has been checked against critical experiment data on UO$_2$F$_2$ solutions in a tank containing boron sheets [16]. In these experiments, critical solution height was measured as a function of the number of sheets (~0.30 g B/cm$^2$; $R \approx 1.345$ from [15]) placed in the tank. The calculated values of $k_{\text{eff}}$ for these critical tests ranged between 1.00 and 1.056, as shown in Table III, indicating that the above recipe is slightly conservative.

5. SAFETY OF ARRAYS OF UNMODERATED URANIUM MATERIALS

Handling large quantities of fissionable materials in a given facility requires subdivision of this material into individually safe batches, and requires maintenance of safe separation distances between such batches. The safety of such arrays of fuel must be established under dry, flooded, and partially moderated conditions.
TABLE III — COMPARISON OF CALCULATION METHOD WITH CRITICAL EXPERIMENT DATA – URANIUM SOLUTION WITH POISON SHEETS

<table>
<thead>
<tr>
<th>No. of Poison Sheets</th>
<th>Measured Critical Solution Height (cm)</th>
<th>Calculated $(k_{eff})_p$</th>
<th>Calculated $(\Delta k/k)_p$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Unreflected Cores</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>0</td>
<td>12.4</td>
<td>0.996</td>
<td>0</td>
</tr>
<tr>
<td>1</td>
<td>12.9</td>
<td>0.999</td>
<td>0.0246</td>
</tr>
<tr>
<td>3</td>
<td>14.3</td>
<td>1.012</td>
<td>0.0740</td>
</tr>
<tr>
<td>4</td>
<td>15.0</td>
<td>1.014</td>
<td>0.0986</td>
</tr>
<tr>
<td>5</td>
<td>16.8</td>
<td>1.056</td>
<td>0.123</td>
</tr>
<tr>
<td>6</td>
<td>17.5</td>
<td>1.048</td>
<td>0.148</td>
</tr>
<tr>
<td>One-Half Reflected Cores</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>0</td>
<td>9.3</td>
<td>1.045</td>
<td>0</td>
</tr>
<tr>
<td>1</td>
<td>9.9</td>
<td>1.049</td>
<td>0.0246</td>
</tr>
<tr>
<td>3</td>
<td>10.4</td>
<td>1.018</td>
<td>0.0740</td>
</tr>
<tr>
<td>4</td>
<td>11.2</td>
<td>1.022</td>
<td>0.0986</td>
</tr>
<tr>
<td>5</td>
<td>12.0</td>
<td>1.028</td>
<td>0.123</td>
</tr>
<tr>
<td>6</td>
<td>12.8</td>
<td>1.025</td>
<td>0.148</td>
</tr>
<tr>
<td>7</td>
<td>13.7</td>
<td>1.028</td>
<td>0.173</td>
</tr>
<tr>
<td>8</td>
<td>15.0</td>
<td>1.037</td>
<td>0.197</td>
</tr>
<tr>
<td>10</td>
<td>17.6</td>
<td>1.032</td>
<td>0.246</td>
</tr>
</tbody>
</table>

(a) Tank cross-section was 30 in. x 60 in. Sheets were 59.5-in. long, 1/4-in. thick boral, and extended above the solution.

5.1 Reactivity of unmoderated $\text{UO}_2$ and U-Zr alloy units, and unmoderated arrays

The interaction angle criteria described in section 2.1 can be used to establish safe spacings for units of uranium materials in arrays, provided that values of $k_{eff}$ for individual units can be computed. It is frequently not possible to do this for unmoderated units with hand calculation recipes. More elaborate techniques, such as multigroup transport theory, must be utilized. The DTF code has been used for this purpose at United Nuclear. Figure 2 shows $k_{eff}$ values for unmoderated, unreflected spheres of $\text{UO}_2$ (93.5% enrichment) as computed with this code. These data are for spherical masses having a density of 75% of the theoretical density of $\text{UO}_2$. They may be conservatively applied to $\text{UO}_2$ powder in thin-walled containers where the bulk density of $\text{UO}_2$ is less than 75% of theoretical.

DTF calculations were also performed for unreflected infinite cylinders of U-Zr alloy (93.5% enriched). These cylinders may be used to simulate long shelves on which ingots or rolled pieces of such material are stored. Cylinder diameters up to 6 in. and uranium concentrations up to 25 wt.% were considered. The $k_{eff}$ values for these cylinders were all substantially below 0.3. According to the criteria discussed in section 2.1, the maximum permissible interaction angle at a shelf containing U-Zr alloy material of this concentration would be 6.0 Sr.
5.2. Safety under flooded conditions

To ensure the criticality safety of an array under conditions of accidental water flooding, each individual unit of the array should be subcritical when surrounded by an effectively infinite water reflector. A minimum surface-to-surface spacing of 12 in. between units is usually specified at United Nuclear Corporation, so that individual units would be completely decoupled under flooded conditions.

5.3. Safety of partially moderated arrays

When substantial quantities of unmoderated uranium material are stored in an area, consideration must be given to the effects of partial moderation on the reactivity of the array. A partial moderation condition is one in which there is enough water present between units of the array to influence the nuclear characteristics of the array significantly, but not enough to decouple the individual units. Such a condition might arise, for example, during fire-fighting operations by directing high pressure water jets at the fuel array, or through the use of the fog or foam generators that recently have become popular fire-fighting tools. Few experimental data exist on partially moderated arrays, and such systems generally cannot be treated exclusively by simple recipes.
A method of computing the reactivity of an array of identical subcritical units, with either air or liquid water between the units, is described in Ref. [3]. The $k_{\text{eff}}$ of the array is given by

$$ (k_{\text{eff}})_{\text{array}} = \frac{(k_{\text{eff}})_{\text{unit}}}{1 - (1 - U_f) \sum_i (ab)_i p_i \Omega_{fi}} $$

where $(1 - U_f) =$ neutron leakage probability for a single unit, $\Omega_{fi} =$ average fractional solid angle subtended at the central unit of the array by the $i$th unit (units shielded from view of the central unit are excluded), $p_i =$ neutron flux in the $i$th unit relative to that in the central unit of the array, as computed from elementary formulas for flux distribution in homogeneous reactors of various shapes, $(ab)_i =$ interaction effectiveness factor for the moderator between the $i$th and central units.

The quantity $(ab)$ is a measure of both the neutron-absorbing and moderating properties of the material between units. Data on $(ab)$ for liquid water [3] can be used with low-density water by replotting to give $(ab)$ as a function of the product of thickness and density. Application of this calculation method to critical experiment data on arrays of uranium solution containers, some with air between units and some with liquid water between units, are presented in Ref. [3]. Agreement with experiment is good (within ±5% in $k_{\text{eff}}$ for the array, and within about 0.25 in. in the critical spacing between units).

To obtain an indication of the validity of this method for arrays of unmoderated units with moderator between units, calculations of critical experiment data on arrays of highly enriched uranium metal cylinders enclosed in Plexiglas boxes were performed [17].

For each of seven arrays, $(k_{\text{eff}})_{\text{unit}}$ and $(1 - U_f)$ for an individual cylinder were computed using the simple recipes for a bare uranium metal unit given in Ref. [3]. These recipes were extended to a unit surrounded by a thin hydrogenous reflector by adding a reflector savings to the extrapolation length for a bare unit. Reflector savings for thin hydrogenous reflectors were obtained from experimental critical mass data on uranium metal spheres [18]. The check calculations are summarized in Table IV. The calculated values of $k_{\text{eff}}$ for the arrays were within ±4% of the measured value of unity except for two closely packed 8-unit arrays. The $k_{\text{eff}}$ values calculated for the latter arrays were about 1.09. This poor agreement was attributed largely to the approximation used in calculating $\Omega_f$ which breaks down for tight spacing. The approximation used results in an overestimate of $\Omega_f$, and hence of $(k_{\text{eff}})_{\text{array}}$, when the cylinders are closely spaced.

5.4. Application to partially moderated arrays of U-Zr alloy

The above calculation model was used to explore the effects of uniform partial moderation on arrays of long shelves of large U-Zr alloy pieces. Each shelf is represented as a 6-in.-diameter cylinder of alloy containing 25 wt. % U (93.5% enriched). The use of this relatively reactive cylinder should make the results of this calculation conservatively applicable to shelves with lighter loads.
TABLE IV — COMPARISON OF CALCULATION METHOD WITH CRITICAL EXPERIMENT DATA — ARRAYS OF U METAL CYLINDERS IN PLEXIGLAS BOXES

<table>
<thead>
<tr>
<th>Array</th>
<th>A</th>
<th>B</th>
<th>C</th>
<th>D</th>
<th>E</th>
<th>F</th>
<th>G</th>
</tr>
</thead>
<tbody>
<tr>
<td>Number of Units in Array</td>
<td>8</td>
<td>8</td>
<td>8</td>
<td>8</td>
<td>27</td>
<td>2</td>
<td>2</td>
</tr>
<tr>
<td>Array Geometry</td>
<td>Cubic</td>
<td>Cubic</td>
<td>Cubic</td>
<td>Cubic</td>
<td>Coaxial</td>
<td>Coaxial</td>
<td></td>
</tr>
<tr>
<td>Array Reflector (Hydrogenous)</td>
<td>None</td>
<td>None</td>
<td>None</td>
<td>None</td>
<td>None</td>
<td>None</td>
<td>None</td>
</tr>
<tr>
<td>Thickness of Plexiglas Box Around Units, cm</td>
<td>0.64</td>
<td>0.64</td>
<td>1.27</td>
<td>2.38</td>
<td>2.38</td>
<td>0.64</td>
<td>(a)</td>
</tr>
<tr>
<td>Surface-to-Surface Separation of Units, cm</td>
<td>4.082</td>
<td>4.239</td>
<td>6.619</td>
<td>10.239</td>
<td>16.289</td>
<td>12.9</td>
<td>10.9</td>
</tr>
<tr>
<td>Calculated Quantities</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$k_{\text{eff}}$ unit</td>
<td>0.815</td>
<td>0.815</td>
<td>0.843</td>
<td>0.888</td>
<td>0.888</td>
<td>0.960</td>
<td>0.991</td>
</tr>
<tr>
<td>$(1 - U_f)$</td>
<td>0.625</td>
<td>0.625</td>
<td>0.611</td>
<td>0.591</td>
<td>0.591</td>
<td>0.558</td>
<td>0.544</td>
</tr>
<tr>
<td>$\Sigma Q_{fi} P_i$</td>
<td>0.3122</td>
<td>0.3062</td>
<td>0.2187</td>
<td>0.1444</td>
<td>0.1362</td>
<td>0.0231</td>
<td>0.0286</td>
</tr>
<tr>
<td>$(k_{\text{eff}})^{array}$</td>
<td>1.098</td>
<td>1.090</td>
<td>1.038</td>
<td>0.992</td>
<td>0.986</td>
<td>0.977</td>
<td>0.998</td>
</tr>
</tbody>
</table>

(a) Entire space between units filled with Plexiglas.

![Graph](image)

FIG. 3. $k_{\text{eff}}$ and leakage probability for U-Zr alloy cylinder surrounded by infinite water reflectors.

The values of $(k_{\text{eff}})_{\text{unit}}$ and $(1 - U_f)$ for a single isolated cylinder surrounded by thick reflectors of water of different densities were computed with the DTF code. The reflector thicknesses assumed in these calculations varied from 15 cm for liquid water to 150 cm for very low density water. The results of these calculations are shown in Fig. 3.
A 5X5 array of these U-Zr alloy cylinders on a 21-in square pitch nearly meets the criteria for safety under dry and flooded conditions. The actual maximum interaction angle is 6.08 sr, slightly above the 6.0 sr permissible for a unit having a $k_{\text{eff}}$ less than 0.30. The reactivity of this 5X5 array with uniform partial moderation is shown in Fig. 4. In these calculations, $p_1$ was taken to be 1.0 for all units. This not only simplified the calculations, but also introduced a conservative bias, which is desirable when applying this technique to practical situations.

Figure 4 shows that the reactivity of the array increases sharply as interstitial moderator is added, with a peak $k_{\text{eff}}$ of 0.64 reached at a water density of 0.04 g/cm$^3$. Although this particular array is safe regardless of degree of partial moderation, it is difficult to extend this result to other arrays. The sharp rise in reactivity upon addition of a small quantity of moderator is probably characteristic of all arrays of unmoderated fuel units, and should be taken into account when arrays are designed.

![Graph showing reactivity of partially moderated array of U-Zr alloy cylinders](image)

**FIG. 4.** Reactivity of partially moderated array of U-Zr alloy cylinders

**REFERENCES**


DISCUSSION

J. SCHMETS: Figure 1 gives the critical diameter for an infinite cylinder as 7.5 in. What would be the criticality situation for the highest concentration if precipitation of UO$_2$F$_2$ occurred, for example as a result of cooling of the solution?

A.H. KAZI: A meaningful answer to that question would require detailed analysis of several factors which were not considered in the calculations reflected in Fig. 1. One would have to estimate the amount of fissile material which might conceivably be deposited on the wall of the cylinder. If solution were then still present in the centre of the cylinder, the system might still have a near-thermal spectrum, the cadmium would still be effective and there could be more self-absorption; reactivity would therefore not vary a great deal from the homogeneous case. If the centre of the cylinder were dry – and this could be prevented – one might reasonably suppose that the deposit of fissile material could give rise to a fast-spectrum system in which the cadmium would no longer be effective. I doubt whether in practice enough fissile material would accumulate to form a critical mass, and the cylinder would in any case allow a fairly high rate of fast neutron leakage.

The practical approach would be to do a series of calculations representing various conceivable situations and to apply a suitably conservative safety factor to the tube diameter. If the resultant diameter were too small to be acceptable, a better arrangement might require different poison material, for example boron steel strips. Various methods in reactor theory, such as the DTF code discussed in the paper, are well suited to this type of analysis.
ОБЕСПЕЧЕНИЕ ЯДЕРНОЙ БЕЗОПАСНОСТИ ПРИ РАБОТЕ С ДЕЛЯЩИМИСЯ ВЕЩЕСТВАМИ

КАМАЕВ А.В., КУЗНЕЦОВ Ф.М., ВЛАДЫКОВ Г.М., ДУБОВСКИЙ Б.Г.
ФИЗИКО-ЭНЕРГЕТИЧЕСКИЙ ИНСТИТУТ
ГОСУДАРСТВЕННОГО КОМИТЕТА ПО ИСПОЛЬЗОВАНИЮ АТОМНОЙ ЭНЕРГИИ СССР, ОБНИНСК СССР

Abstract — Résumé — Аннотация — Resumen

NUCLEAR SAFETY IN WORKING WITH FISSIONABLE MATERIALS. The paper discusses the ensuring of nuclear safety in working with uranium whose concentration is unknown and with a large amount of uranium solutions, using neutron absorbers. The minimum critical dimensions of systems containing uranium-235 together with the safe distances between tanks containing salts or uranium solutions are considered. Data are given relating to the study of absorbers in the form of rods, rod lattices and cylindrical baffles. The results given can be used in solving problems of nuclear safety arising in work with uranium solutions.

CONTROLE DE LA SÉCURITÉ NUCLEAIRE DANS LES TRAVAUX PORTANT SUR DES MATIÈRES FISSILES. Les auteurs étudient le contrôle de la sécurité nucléaire, à l'aide d'absorbeurs de neutrons, au cours d'opérations portant sur de l'uranium de concentration inconnue et sur de grands volumes de solutions d'uranium. Il parlent des dimensions critiques minimales des systèmes contenant de l'uranium-235 ainsi que des distances de sécurité à maintenir entre récipients remplis de sels ou de solutions d'uranium. Ils citent des données intéressant l'étude d'absorbeurs sous forme de barres, de réseaux de barres et de protections cylindriques.

Les résultats indiqués peuvent être utilisés pour résoudre les problèmes de sécurité nucléaire que posent les recherches sur les solutions d'uranium.

GARANTIAS DE SEGURIDAD NUCLEAR EN TRABAJOS CON MATERIALES FISIONABLES. Se examinan las garantías de seguridad nuclear cuando se trabaja con uranio de concentración desconocida y con un gran volumen de soluciones de uranio, con empleo de absorbeores de neutrones. Se estudia un sistema de dimensiones críticas mínimas, con una concentración de uranio-235, y la distancia segura entre los tanques que contienen sales o soluciones de uranio. Se citan datos sobre el estudio de los absorbeores en forma de barras, reticulados de barras y tabiques separadores cilíndricos.

Los resultados que se citan pueden utilizarse para resolver cuestiones de seguridad nuclear en trabajos con soluciones de uranio.

Работа с делящимися веществами включает несколько проблем, таких, например, как методы определения состава смесей, содержащих делящиеся вещества, методы определения количества делящихся веществ, методы хранения и перевозки, методы обеспечения радиационной безопасности, методы учета, экономические проблемы и т.п.
Однако на всех этапах работы с делящимися веществами в первую очередь должна рассматриваться проблема ядерной безопасности, т.е. исключение возможности возникновения неконтролируемой цепной реакции деления.

Для обеспечения ядерной безопасности необходимо знание критических параметров систем с делящимися веществами: критической массы и критических размеров. В настоящее время накоплено большое количество экспериментальных и расчетных данных о критических параметрах систем делящихся веществ, которые могут быть использованы при решении вопросов ядерной безопасности.

В публикуемом докладе рассмотрено обеспечение ядерной безопасности в двух случаях:
а) при работе с ураном неизвестной концентрации и
б) при работе с большими объемами растворов урана с применением поглотителей нейтронов.

1. ОБЕСПЕЧЕНИЕ ЯДЕРНОЙ БЕЗОПАСНОСТИ ПРИ РАБОТЕ С УРАНОМ НЕИЗВЕСТНОЙ КОНЦЕНТРАЦИИ

Рассмотрим обеспечение ядерной безопасности при работе с ураном-235. Так как концентрация урана неизвестна, ядерная безопасность обеспечивается ограничением емкостей, содержащих смеси или растворы урана. Обычно емкости выполняют либо в форме "бесконечных" цилиндров, (отношение H/D > 10), либо в форме "бесконечных" пластин (отношение l_1 / h > 10).

В таких случаях ограничивающим размером будет соответственно диаметр цилиндра D и толщина пластины h, которые должны быть меньше минимальных критических значений этих размеров.

В случае отсутствия указанных емкостей, работа с ураном может производиться в любой емкости, однако, при этом строго ограничивается объем смеси или раствора урана. Этот объем не должен превышать соответствен но минимального критического объема для металлического урана или раствора урана.

Из анализа экспериментальных и расчетных значений критических параметров систем делящихся веществ получены минимальные критические значения указанных параметров. Они приведены в табл.1 [1, 2].

Если уран находится в виде раствора, то следует брать минимальные критические параметры из табл.1, указанные для растворов; если в виде смеси с какими-либо веществами — минимальные критические параметры для металла.

Указанные в табл.1 минимальные критические объемы могут быть увеличены, если емкости, в которых находится уран, отличаются от сферических. На рис.1 показан критический объем для емкостей в форме цилиндра [2]. Если обогащение урана известно, то минимальные критические размеры могут быть увеличены. На рис.2 показаны зависимости минимальных критических размеров систем, содержащих растворы урана, от обогащения урана [2].

Следует помнить, что в табл.1 и на рис.1 и 2 приведены критические значения размеров емкостей. Для обеспечения ядерной безопасности эти размеры должны быть уменьшены на ~10%.
Таблица 1

Минимальные критические размеры систем, содержащих уран-235

<table>
<thead>
<tr>
<th>Металл</th>
<th>Растворы</th>
<th>( \text{UO}_2\text{F}_2 )</th>
<th>( \text{UO}_2(\text{NO}_3)_2 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>1. Диаметр бесконечного цилиндра, см</td>
<td>7,8</td>
<td>13,7</td>
<td>16,7</td>
</tr>
<tr>
<td>2. Толщина бесконечной пластины, см</td>
<td>1,5</td>
<td>4,3</td>
<td>6,9</td>
</tr>
<tr>
<td>3. Объем в форме сферы, л</td>
<td>1,2</td>
<td>6,3</td>
<td>8,4</td>
</tr>
</tbody>
</table>

Рис.1 Зависимость критического объема раствора в цилиндрической активной зоне от отношения высоты цилиндра к его диаметру:
- отношение \( \rho_h/\rho_c = 44,3 \); 
- отношение \( \rho_h/\rho_c = 320 \).

Если уран размещается в нескольких емкостях, то они должны быть расположены таким образом, чтобы исключить взаимодействие между ними. В табл. 2 указаны безопасные расстояния между емкостями, содержащими сухие соли или растворы урана, при размещении их в воздухе [3]. Бесконечные размеры цилиндров и пластин, указанных в табл. 2, приняты равными 6 м.

Аналогичный подход к обеспечению ядерной безопасности может быть принят также для урана-233 и плутония-239 с использованием соответствующих величин критических параметров.
ТАБЛИЦА 2

МИНИМАЛЬНЫЕ РАССТОЯНИЯ МЕЖДУ ЕМКОСТЯМИ БЕЗОПАСНОЙ ГЕОМЕТРИИ, СОДЕРЖАЩИМИ СУХИЕ СОЛИ ИЛИ РАСТВОРЫ УРАНА-235
(Плотность урана не более 3,2 кг/л. Расстояния указаны между ближайшими стенками емкостей).

<table>
<thead>
<tr>
<th>Емкость</th>
<th>Расстояние между двумя емкостями, см</th>
<th>Расстояние между соседними емкостями, расположенными в линию, см</th>
<th>Расстояние между соседними емкостями, расположенными в квадратной решетке, см</th>
</tr>
</thead>
<tbody>
<tr>
<td>Сфера объемом 4,8 л</td>
<td>30,5</td>
<td>30,5</td>
<td>30,5</td>
</tr>
<tr>
<td>Цилиндр диаметром 12,7 см</td>
<td>30,5</td>
<td>77,0</td>
<td>137,0</td>
</tr>
<tr>
<td>Пластина толщиной 3,5 см</td>
<td>30,5</td>
<td>215,0</td>
<td>-</td>
</tr>
</tbody>
</table>

Рис. 2 Зависимость минимальных критических размеров систем с растворами урана от обогащения урана:
1 - толщина пластины; 2 - объем; 3 - диаметр цилиндра.

2. ПРИМЕНЕНИЕ ПОГЛОТИТЕЛЕЙ НЕЙТРОНОВ ДЛЯ ОБЕСПЕЧЕНИЯ ЯДЕРНОЙ БЕЗОПАСНОСТИ

При хранении, переработке, а также на других операциях, часто возникает необходимость загрузки больших количеств растворов делящихся ве-
Рис. 3 Приращение критического объема раствора в цилиндрической сборке диаметром 40 см без отражателя при наличии центрального поглощающего стрежня:
1 - стрежень с кадмием; 2 - стрежень с карбидом бора.
δ = 1 мм, концентрация урана - 136 г/л. Критический объем без стрежня - 25,8 л.

ществ, превышающих критические величины, в различные емкости. Для обеспечения ядерной безопасности при этом в емкости необходимо вводить дополнительные поглотители нейтронов.

В качестве поглотителей нейтронов обычно используют бор и кадмий. Поглотители могут размещаться либо гомогенно, например в виде растворов солей бора и кадмия, либо гетерогенно в виде стержней, пластин и проч.

При гомогенном размещении эффективность поглотителей возрастает и, например, при концентрации бора, составляющей ~5% от концентрации урана, самоподдерживающаяся цепная реакция не может возникнуть в системах, содержащих водные растворы урана. Однако применение растворимых гомогенных поглотителей связано с определенными неудобствами: необходимостью очистки делящихся веществ от поглотителей в конце процесса, возможностью случайного вывода поглотителя из системы и связанной с этим необходимостью контроля за количеством поглотителя в системе.

Поэтому значительно удобнее применять поглотители, помещенные в герметическую оболочку из химически стойкого материала, например нержавеющей стали.

В ФЭИ проводилось изучение эффективности поглотителей нейтронов в водных растворах уранил-нитрата с 90% обогащением урана. Поглотители использовались в виде стержней, решеток стержней и цилиндрических перегородок.

Стержни представляли собой трубы из нержавеющей стали толщиной δ, в которые помещали либо порошок карбида бора с плотностью 1,25 г/см³ (бор естественный), либо трубу из кадмия толщиной 0,5 мм, заполненную водой. На рис. 3-6 приведены результаты экспериментов со стержнями и решетками стержней.

Цилиндрические перегородки представляли собой два коаксиальных цилиндра из нержавеющей стали, 6 мм зазор между которыми заполняли либо порошком карбида бора с плотностью 1,25 г/см³, либо кадмием тол-
Рис. 4 Критический объем раствора в цилиндрических сборках с боковым и нижним водяным отражателем в зависимости от диаметра цилиндра:
1 - с центральным поглощающим стержнем; 2 - без стержня; • - концентрация урана 72 г/л; ○ - концентрация урана 286 г/л. Диаметр борного стержня 50 мм, δ = 4 мм.

Рис. 5 Эффективность борных стержней, расположенных по окружности диаметром 19,2 см в цилиндрической активной зоне диаметром 40 см с боковым и нижним водяным отражателем:
1 - концентрация урана - 76 г/л; критический объем без стержней - 23,1 л; 2 - концентрация урана - 289 г/л, критический объем без стержней - 17,35 л. Диаметр стержней 50 мм, δ = 4 мм.
Рис. 6 Эффективность решетки борных стержней в цилиндрической активной зоне диаметром 110 см с боковым и нижним водяным отражателем:
- обогащение урана 90%; концентрация урана - 153 г/л (1), 75 г/л (2), 37,5 г/л (3); - обогащение урана 10%, концентрация урана 420 г/л.

щины 0,5 мм. Результаты экспериментов с такими перегородками показаны на рис.7. Кроме таких перегородок использовали еще кадмиево-водные перегородки. Это два коаксиальных цилиндра из нержавеющей стали зазор между которыми равен 50 мм. У стенок цилиндров внутри зазора помещали кадмий толщиной 0,5 мм и в зазор заливали воду. Результаты экспериментов с кадмиево-водными перегородками показаны на рис.8.

Приведенные результаты экспериментов по изучению эффективности поглотителей могут быть использованы для решения вопросов ядерной безопасности при работе с растворами урана.

Из рисунков видно, что одиночные поглощающие стержни не дают заметного увеличения критической загрузки урана. Стержни диаметром до 50 мм позволяют увеличить критический объем на 5-10%.

Хороший результат дает применение одиночных поглощающих стержней с целью увеличения минимальных критических размеров систем делящихся веществ. Например, при наличии борного стержня диаметром 50 мм в цилиндрической емкости, содержащей раствор уранил-нитрата, диаметр бесконечного цилиндра для таких растворов увеличивается с 16,7 до ~21 см (рис.4).

Ощутимый результат дает применение решетки стержней. Решетка из 6-7 стержней позволяет увеличить критический объем раствора почти вдвое,
Рис. 7 Эффективность борных и кадмевых перегородок в цилиндрической активной зоне диаметром 40 см. а) концентрация урана 239 г/л; б) концентрация урана 76 г/л.
● - борная перегородка; ▲ - кадмевая перегородка; 1 и 2 - с боковым и нижним водяным отражателем; 3 и 4 - без отражателя.

Рис. 8 Эффективность кадмево-водных перегородок в цилиндрической активной зоне диаметром 50 см с боковым и нижним водяным отражателем. Концентрация урана - 457 г/л:
● - перегородка с кадмием и водой; ▲ - перегородка с кадмием без воды; □ - перегородка без кадмия и без воды; X - перегородка без кадмия с водой.

а для раствора с концентрацией урана ниже 100 г/л - более, чем вдвое (рис. 5 и 6).
Решетка из 150 стержней дает возможность загрузить в цилиндрический бак диаметром 110 см и свыше 800 л раствора с концентрацией урана 150 г/л (рис. 6).
Применение цилиндрических поглощающих перегородок также позволяет заметно увеличить критическую загрузку урана.

ЛИТЕРАТУРА
ECONOMIC CONSIDERATIONS;
GOVERNMENT ACTIVITIES

(Session VI)
THE ECONOMIC BASIS FOR ESTABLISHING
THE NUMBER OF MEASUREMENTS ACCORDING
TO THE VALUE OF THE MATERIAL

H. H. PH. MOEKEN AND H. BOKELUND
EUROCHEMIC COMPANY, MOL, BELGIUM

Abstract — Résumé — Аннотация — Resumen

THE ECONOMIC BASIS FOR ESTABLISHING THE NUMBER OF MEASUREMENTS ACCORDING TO THE VALUE OF THE MATERIAL. The monetary value of a batch of material is determined by variables whose values are determined by measurement. The random error in the values found can be reduced by repeating the measurements. The uncertainty in the monetary value, which decreases with the repetition, and the total cost of the measurements, which increases, should be interrelated on economic grounds.

A criterion for establishing the number of measurements according to the monetary value of the material is proposed in this paper. The criterion is applied to uranium shipments. The analysis schemes suggested in the literature for the assessment of the value of uranium and plutonium batches are compared with those established according to the criterion.

CONSIDERATIONS ECONOMIQUES INTERVENANT DANS LA DETERMINATION DU NOMBRE DE MESURES A EFFECTUER EN FONCTION DE LA VALEUR DES MATIERES. La valeur monétaire d'un lot de matières dépend de variables déterminées par la mesure. L'erreur accidentelle peut être réduite par la répétition des mesures. Il convient de mettre en balance, du point de vue économique, la réduction de l'incertitude quant à la valeur monétaire et l'augmentation du coût total des mesures qui résulteront l'une et l'autre de la répétition.

L'auteur propose un critère pour la fixation du nombre des mesures en fonction de la valeur monétaire des matières. Il donne un exemple d'application de ce critère à propos d'envois d'uranium. Il fait également une comparaison entre les programmes d'analyse exposés dans les ouvrages pertinents pour la détermination de la valeur de lots d'uranium et de plutonium et les programmes établis conformément à ce critère.

ЭКОНОМИЧЕСКИЕ ОСНОВЫ ОПРЕДЕЛЕНИЯ КОЛИЧЕСТВА ИЗМЕРЕНИЙ В СООТВЕТСТВИИ СО СТОИМОСТЬЮ МАТЕРИАЛОВ. Денежная стоимость партии материала определяется переменными величинами, ценность которых определяется измерением. Случайные ошибки в найденных стоимостных величинах могут быть уменьшены путем повторения измерений. Неопределенность денежной стоимости, которая уменьшается при повторении измерений, и общая стоимость измерений, которая возрастает, должны быть взаимосвязаны на экономической основе.

Предлагается критерий определения количества измерений в соответствии с денежной стоимостью материала. Критерий применяется к грузам урана. Схемы анализа, предложенные в литературе для оценки стоимости партий урана и плутония, сравниваются со схемами, созданными в соответствии с данным критерием.

BASES ECONOMICAS PARA FIJAR EL NUMERO DE MEDICIONES NECESARIAS SEGUN EL VALOR DE LOS MATERIALES. El valor monetario de una partida de material está determinado por diversas variables cuyo valor se obtiene por medición. El error ocasional en los valores obtenidos puede reducirse repitiendo las mediciones. Por consideraciones económicas hay que establecer una correlación entre la incertidumbre acerca del valor monetario, que disminuye con esa repetición, y el costo total de las mediciones, que aumenta con ellas.

Los autores proponen un criterio para fijar el número de mediciones necesarias en relación con el valor monetario del material. Este criterio se aplica a las expediciones de uranio. Por último, comparan los sistemas de análisis descritos en las fuentes bibliográficas relativas a la determinación del valor de las expediciones de uranio y plutonio con los basados en el criterio expuesto.
INTRODUCTION

The monetary value of a batch of material is determined by variables such as weight, volume, concentration and isotopic composition whose values are determined by measurement. The random errors in the values found can be reduced by repeating the measurements. The uncertainty in the monetary value of the batch then decreases, but the total cost incurred for measurements increases. It is evident that these two figures should be interrelated on economic grounds.

Normally both the shipper and the receiver of a batch of valuable material establish the value independently. In the case of shipment of nuclear materials, there is a tendency for closer co-operation in this respect. One form of co-operation is the arrangement in which the weighing and sampling of the batch are carried out in the presence of the receiver [1]. Another kind of co-operation is practised for shipments of irradiated reactor fuel [2]. Here, for obvious reasons, the batch cannot be sampled before shipment to a reprocessing plant. An arrangement between both parties gives the reactor operator the right to be present during the sampling and the measurements, and to have access to all data produced for assessing the value of the shipment. This practice, which is also an economical one, requires agreement about the number of measurements acceptable to both parties.

ACCEPTABLE NUMBER OF MEASUREMENTS

In order to derive a criterion for an acceptable number of measurements we assume that many batches, having a true value \( V \), are shipped to a receiver. The random error of the shipper's determination of the value is \( S_s \) (standard deviation of the population of batch values relative to the true mean). The receiver may re-assay the batch in an attempt to obtain a more precise value, having a relative error \( S_r \). Both errors are assumed to be normally distributed.

The receiver may proceed according to the following policy. If the value quoted by the shipper is lower than his own, he accepts it; if the shipper's value is higher, he rejects it but arbitration is then required.

In the case of acceptance, which occurs for half the batches, the average value quoted by the shipper can be calculated as shown in the Appendix, being lower than the value \( V \) actually shipped. The receiver makes a profit and thus causes the shipper a loss. This profit per batch, averaged over all batches accepted or rejected, is equal to

\[
S_s V (2\pi(1 + S_r^2/S_s^2))^{-0.5}
\]

The receiver may be inclined to incur expenses for re-assay if his analysis costs per batch \( C_r \) are balanced by the profit so obtained. Thus

\[
C_r = S_s V \left[2\pi(1 + S_r^2/S_s^2)\right]^{-0.5}
\]

(1)
In the case of rejection, followed by arbitration, both parties have losses because of the arbitration costs, whatever reasonable arrangement has been made at this point. It is assumed that an arbiter can establish the true value and that the settlement between the two parties is made accordingly.

An alternative to the acceptance method described is that the shipper only should carry out the determination of the value of the batch. This value is then accepted by the receiver without re-assay. This method is obviously acceptable to the shipper, and is also acceptable to the receiver if the shipper's precision of the determination is satisfactory and if the receiver can be present during the measurements and have access to all relevant data. In this paper, the criterion for an acceptable assessment of the value, according to the alternative method, is that the shipper's error, the value of the batch and the hypothetical receiver's error and analysis cost corresponding to this error are interrelated according to Eq. (1).

REQUIRED ANALYSIS SCHEME

Case A: One main source of error

The first case discussed is when the error in the value of the batch is determined by the error in a single variable whose value is fixed by measurement. It is assumed that in their method of measuring the variable, the shipper and the receiver experience the same error, expressed by the relative standard deviation $s$ for a single measurement, and that for both parties the cost $c$ of a measurement is the same. The error of the shipper's determination of the value is reduced by $n$ repeat measurements according to

$$S_{t}^{2} = \frac{s^{2}}{n}$$

The number of measurements required to be made by the shipper for the case when the receiver hypothetically makes a minimum effort, namely one measurement, can be found from the criterion developed in the previous section. The ratio $S_{t}^{2}/S_{r}^{2}$ is then equal to $n$, because $S_{r}^{2} = s^{2}$. Thus the criterion can be written as

$$V = 2.5c (n + n^{2})^{0.5} s^{-1}$$

Figure 1 shows the relationship between $V$, $n$ and $s$ for $c = $10. The figure indicates for different values of the batch the number of measurements required to establish the value. This assessment should be acceptable to both parties. Introduction of other values of $c$ in the equation gives a graph with a set of lines, parallel to that of Fig. 1.

Case B: Two sources of error

The monetary value of a batch is often determined by the product of two variables whose values are fixed by measurement. As in case A, it is assumed that the shipper and the receiver have the same error in their
methods of measuring the two variables, and that the cost of a measurement is the same for both parties. The errors in the two kinds of measurements, expressed as relative standard deviations, are $s_1$, $s_2$, and the costs are $c_1$ and $c_2$ respectively. Repeat measurement by the shipper of the two variables, to total numbers of $n_1$ and $n_2$ respectively, reduces his error in the value determination according to

$$S_s^2 = s_1^2 / n_1 + s_2^2 / n_2$$

For the case when the receiver hypothetically makes a minimum effort, namely one measurement of each kind, the required number of shipper's
measurements as a function of the batch value can be derived from the criterion. The ratio \( S_1^2/S_i^2 \) is then equal to

\[
\frac{s_1^2 + s_2^2}{s_1^2/n_1 + s_2^2/n_2}
\]

Thus the criterion of the previous section can be transformed into

\[
V = 2.5 (c_1 + c_2) \left( \frac{s_1^2/n_1 + s_2^2/n_2 + s_1^2 + s_2^2}{s_1^2/n_1 + s_2^2/n_2} \right)^{0.5}
\]

or

\[
V = 2.5 (c_1 + c_2) \left( \frac{1/n_1 + p^2/n_2 + 1 + p^2}{1/n_1 + p^2/n_2} \right)^{0.5} s_1^2
\]

where \( p = s_2/s_1 \).

As an example, the relationship between \( V \), \( n_1 \), \( n_2 \) and \( s_1 \) is shown in Fig. 2 for the case where \( c_1 = $10 \), \( c_2 = $30 \) and \( p = 3 \). For particular analysis schemes, the different values of the batch are indicated. The analysis scheme for the establishment of a batch value that should be acceptable to both parties can be found from the graph. The combinations of \( n_1 \) and \( n_2 \) that give the shipper the required precision for the lowest analysis cost are used in the figure.

APPLICATION TO URANIUM SHIPMENTS

The monetary value of a batch of uranyl nitrate solution is determined by the product of the variables: weight or volume, uranium concentration and the price of a weight unit of uranium. This price is, however, a function of the fraction of uranium-235 by weight. The error in the weight or volume measurement and the sampling error are assumed to be negligible compared with the errors in the measurements of concentration and weight fraction of uranium-235. Thus, case B (two sources of error) applies.

The price of uranium hexafluoride for several values of the uranium-235 weight fraction \( w_5 \) has been tabulated in the United States Atomic Energy Commission price list [3]. The price of uranyl nitrate can be obtained by subtracting $5.60 per kg of uranium for conversion of nitrate to hexafluoride. The price for a particular value of \( w_5 \) can be found by linear interpolation between two adjacent \( w_5 \) values in the price table. Thus, the uranium price within each interval between published \( w_5 \) values can be expressed by a linear function \( f = a + bw_5 \), in which the values of \( a \) and \( b \) are different for each interval.

The relative error in the price setting can be estimated from the relative error in \( w_5 \) by means of the propagation of error formula, namely

\[
\frac{d(a + bw_5)}{a + bw_5} = b \frac{dw_5}{w_5} \times \frac{dw_5}{w_5} = r \frac{dw_5}{w_5}
\]

where \( r = bw_5/(a + bw_5) \) is the ratio of the two relative errors. The ratio \( r \) can be calculated for each interval in the given \( w_5 \) values.
Figure 3 shows the price of uranyl nitrate and the ratio $r$ as a function of the uranium-235 enrichment.

The application of the criterion for establishing the number of measurements can be demonstrated by the following numerical example. The relative errors and the unit costs of a measurement of the uranium concentration, $s_1$ and $c_1$, and of a measurement of uranium-235 weight fraction, $s_2$ and $c_2$, used in the calculation are

$$ s_1 = 0.2\% \quad c_1 = $10$$

and

$$ s_2 = 1\%, \ 0.5\% \ \text{and} \ 0.1\% \ \text{for natural, slightly enriched and highly enriched uranium respectively} \quad c_2 = $30$$
The ratio of the relative errors in the price setting and the uranium-235 weight fraction measurement and the price of uranyl nitrate, both as a function of the uranium-235 enrichment.

The relative error in the price setting, $s_2$, is thus $r s_1$, where $r$ can be taken from Fig. 3. The $r$ values used in the calculation are 4, 1.5 and 1 for the three uranium enrichments respectively, and the corresponding ratios $p$ in the criterion are thus 20, 3.75 and 0.5. A graph can now be constructed relating the value $V$, the type of uranium and the number of measurements $n_1$ and $n_2$ on the basis of the criterion.

Figure 4 shows the analysis schemes, to be carried out by the shipper of a uranium batch, which should be acceptable to the shipper and to the receiver.
DISCUSSION

The analysis schemes required according to the criterion proposed in this paper can be compared with the analysis schemes for uranium and plutonium batches suggested in the literature.

In a paper [4] originating from the Idaho Chemical Processing Plant, one concentration and one weight fraction measurement are suggested per batch of dissolved reactor fuel containing approximately 0.5 kg of highly enriched uranium (batch value $5000). The relative standard deviations of the measurement methods mentioned are 2% and 0.14% respectively. If the unit analysis costs of the two methods are assumed to be $50 and $50, the maximum value of a batch according to the criterion for an analysis scheme $n_1 = 1$ and $n_2 = 1$ is $17,600. The actual analysis scheme is therefore in agreement with the criterion.
Another proposal was published in an article by the Hanford Chemical Processing Department [5], which suggested a scheme of four concentration measurements for uranium and four for plutonium on a 3 t batch of dissolved reactor fuel containing $150,000 worth of uranium and $150,000 worth of plutonium. The precisions of the measurement methods, expressed as relative standard deviations, are 0.5% and 1.0% respectively. For a unit analysis cost of $50 and an analysis scheme of n = 4, the maximum values of a batch according to the criterion are $112,000 and $56,000 respectively. The criterion indicates that eight plutonium concentration measurements at least are justified.

In an extensive study [6] of the assessment of the content of plutonium nitrate solutions shipped between the Hanford Works and the Rocky Flat Plant, it is suggested that eight concentration measurements should be made on a batch containing 660 g of plutonium (batch value $6600). The maximum value of a batch according to the criterion on the basis of a measurement precision of 0.3% and a unit analysis cost of $15 can be calculated to be $106,000 for only one measurement. According to the criterion therefore only one measurement is required.

The shipment of purified uranium nitrate solutions from the Idaho Chemical Processing Plant has also been described [7]. Three concentration and three weight fraction measurements are carried out routinely on a batch of approximately 30 kg of highly enriched uranium (batch value $300,000). Assuming measurement errors of 0.1% and unit analysis costs of $10 and $30, the maximum value according to the criterion for the analysis scheme mentioned is $244,000, justifying the scheme.

No reasons have been published for the choice of these actual schemes. The criterion developed in this paper on economic grounds as a logical and consistent basis for the choice supports some of those actually in use, but not all of them. Evidently other reasons have influenced the choice.

ACKNOWLEDGEMENTS

Thanks are due to Professor Dr. H.C. Hamaker, of the Technological University, Eindhoven, and Dr. J. Mandel, National Bureau of Standards, Washington, D.C., for helpful discussions.

REFERENCES

THE MEAN VALUE OF $x_s$ IN THE CASE WHERE $x_s < x_r$

Assume that the differences of the shipper's and receiver's values from the true value $V$ are $x_s$ and $x_r$ respectively, and that they are normally distributed with relative standard deviations $S_s$ and $S_r$ respectively. According to the textbooks on statistics, the simultaneous distribution function of $x_s$ and $x_r$ is

$$f(x_s, x_r) = \frac{1}{2\pi S_s S_r V^2} \exp \left( -\frac{x_s^2}{2S_s^2 V^2} - \frac{x_r^2}{2S_r^2 V^2} \right)$$

The mean value of $x_s$ in the case where $x_s < x_r$ is equal to

$$\bar{x}_s = 2 \int_{-\infty}^{x_r} dx_r \int_{-\infty}^{+\infty} dx_s \times f(x_s, x_r) \times dx_s$$

The factor 2 is caused by the fact that the probability that $x_s < x_r$ is equal to 1/2.

$$\bar{x}_s = \frac{1}{\pi S_s S_r V^2} \int_{-\infty}^{+\infty} \exp \left( -\frac{x_r^2}{2S_r^2 V^2} \right) \times dx_r \int_{-\infty}^{x_r} x_s \times \exp \left( -\frac{x_s^2}{2S_s^2 V^2} \right) \times dx_s$$

$$= \frac{-S_s^2 V^2}{\pi S_s S_r V^2} \int_{-\infty}^{+\infty} \exp \left( -\frac{x_r^2}{2S_r^2 V^2} \right) \times \left[ \exp \left( -\frac{x_s^2}{2S_s^2 V^2} \right) \right]_{-\infty}^{x_r} \times dx_r$$

$$= \frac{-S_s^2}{\pi S_r} \times \pi^{0.5} \left( \frac{1}{2S_s^2 V^2} + \frac{1}{2S_r^2 V^2} \right)^{-0.5}$$

$$= -2S_s V (2\pi(1 + S_s^2/S_r^2))^{-0.5}$$
ECONOMIC CONSIDERATIONS IN THE MEASUREMENT OF NUCLEAR MATERIALS*

F.H. TINGEY
PHILLIPS PETROLEUM COMPANY, IDAHO FALLS, IDAHO, UNITED STATES OF AMERICA

Abstract — Résumé — Аннотация — Resumen

ECONOMIC CONSIDERATIONS IN THE MEASUREMENT OF NUCLEAR MATERIALS. It is implicit in accounting for source and special nuclear materials that measurements be taken of the material on inventory at various stages of processing. These measurements usually involve weights or volumes, concentrations, and mass assays obtained on samples from increments or batches. Each of these measurements admits of error.

In a given measurement scheme some latitude is afforded one in the allocation of the total effort to be expended in arriving at a material balance figure. For a given stated desired precision, various combinations of number of samples and number of analyses per sample may yield this precision. There is an obvious economic incentive to find the best combination. The economics of these measurement generation schemes — nested sampling and compositing of samples — are considered in this paper. Formulæ are derived to determine allocation of sample effort and analysis effort under each scheme. In addition are presented criteria for determining when compositing presents an advantage and also when a change to more expensive (or less expensive) measurement procedures is warranted.

CONSIDERATIONS D'ORDRE ECONOMIQUE RELATIVES A LA MESURE DES MATIERES NUCLEAIRES. Il va de soi que pour rendre compte des matières nucléaires brutes et spéciales, il faut effectuer aux divers stades des opérations des mesures des matières à l'inventaire. Dans ces mesures interviennent généralement des poids ou des volumes, des concentrations et des estimations de masse obtenus à partir d'échantillons prélevés sur des incrément ou sur des lots. Chacune de ces mesures prête le flanc à l'erreur.

Dans un programme donné de mesure, on jouit d'une certaine latitude pour déterminer l'effort global à déployer en vue d'arriver à un bilan matière chiffré. Pour obtenir une précision donnée, diverses combinaisons nombre d'échantillons/nombre d'analyses par échantillon peuvent donner satisfaction. Il y a un intérêt économique évident à trouver la meilleure combinaison. L'auteur étudie les aspects économiques de ces programmes de mesure — échantillonnage groupé et échantillonnage composite. Il tire de son étude des formulæ pour établir la part de l'échantillonnage et celle de l'analyse dans le cas de chaque programme. En outre, il indique les critères à appliquer pour déterminer si l'échantillonnage composite présente des avantages et si le choix de techniques de mesure plus onéreuses (ou moins onéreuses) se justifie.

ЭКОНОМИЧЕСКИЕ СООБРАЖЕНИЯ, СВЯЗАННЫЕ С ИЗМЕРЕНИЕМ ЯДЕРНЫХ МАТЕРИАЛОВ. Учет источников и специальных ядерных материалов предусматривает измерения при инвентаризации на различных этапах их обработки. Эти измерения обычно включают в себя вес или объем, концентрацию и количественный анализ массы, которые проводятся на образцах, взятых из инкрементов или партий. В каждом из этих измерений может быть допущена ошибка.

В данной схеме измерений допускается некоторая свобода, а именно свобода при выборе главного направления измерений, расширяемых при достижении балансового показателя материалов. Желаемой точности можно достичь путем различных комбинаций ряда образцов и ряда анализов каждого образца. Нахождение лучшей комбинации является очевидным экономическим стимулом. В данном докладе рассматривается экономическая сторона данных схем получения результатов измерений — отбор групп образцов и комбинация образцов. Выведены формулы для определения главного направления при работе с образцами и проведеняем анализов по каждой схеме. Кроме того, излагаются критерии того, когда комбинация дает преимущество, а также, когда оправдан переход к более (или менее) дорогостоящим методам измерений.

* This paper was not presented for discussion.
ASPECTOS ECONÓMICOS DE LA MEDICIÓN DE MATERIALES NUCLEARES. La contabilización de los materiales básicos y de los materiales nucleares especiales lleva consigo la necesidad de realizar mediciones del material inventariado en diversas etapas de su tratamiento. Estas mediciones incluyen, por lo general, determinaciones volumétricas, del grado de concentración y de masa, realizadas sobre muestras tomadas de tandas o partidas de material. Cada una de esas mediciones está sujeta a la posibilidad de error.

En un plan dado de mediciones se dispone siempre de un cierto margen para la distribución del trabajo que en conjunto ha de realizarse a fin de establecer en cifras el balance de los materiales. Para conseguir un determinado grado de exactitud, fijado como conveniente, cabe recurrir a diversas combinaciones del número de muestras empleado y del número de análisis por muestra. Es evidente el incentivo económico que encierra el encontrar la combinación óptima. El autor examina los aspectos económicos de estos planes de obtención de datos: muestreo sucesivo y mezcla de muestras de distintas partidas. Expone fórmulas para establecer, dentro de cada plan de mediciones, la distribución del esfuerzo total entre el trabajo de muestreo y el trabajo de análisis. Además, enuncia diversos criterios para determinar cuándo resulta conveniente emplear mezclas de muestras de diversas partidas así como en qué ocasiones está justificado recurrir a otros procedimientos más (o menos) costosos de medición.

1. INTRODUCTION

It is implicit in accounting for source and special nuclear material that measurements be taken of the material on inventory at various stages of processing. These measurements usually involve weights or volumes, concentrations, and mass assays obtained on samples from increments or batches. Each of these measurements admits to two different types of measurement error. These are commonly called random and systematic errors. Estimates of each of these for all types of measurements are essential if one is to estimate accurately the uncertainty to be associated with any result of these measurements.

In a given measurement scheme some latitude is afforded one in the allocation of the total effort to be expended in arriving at a material balance figure. For a given stated desired precision, various combinations of number of samples and number of analyses per sample may yield this precision. There is an economic incentive to find the best combination. Also through compositing of samples a significant economy can sometimes be effected.

It is the intent of this paper to investigate in some detail the economics associated with two fairly common measurement schemes and in so doing develop criteria by which economically optimum systems can be devised.

2. DEFINITION AND NOTATION

We shall have occasion to differentiate among eight types of measurement errors or sources of variation to be associated with a weight or volume, concentration, and mass assay scheme of data generation. In some instances some of these might be of negligible magnitude; however, for the sake of generality, all are included. It is implicit in what follows that valid estimates be available for each. For the sake of convenience we elect to use the coefficient of variation or relative standard deviation as the basic variation unit and at the risk of some ambiguity, choose to denote it simply
by the standard deviation symbol $\sigma$. In terms of the square of the relative standard deviation ($\sigma^2$), we define the relative variances to be

- $\sigma_{Vr}^2 = \text{relative variance of the random component of the volume or weight determination for each batch.}$
- $\sigma_{Cr}^2 = \text{relative variance of the concentration determination between samples of the same batch.}$
- $\sigma_{Ca}^2 = \text{relative variance of the random analytical concentration determination within a sample.}$
- $\sigma_{Mr}^2 = \text{relative variance of the mass assay determination between samples of the same batch.}$
- $\sigma_{Ma}^2 = \text{relative variance of the mass assay determination between preparations of the same sample.}$
- $\sigma_{Vd}^2 = \text{relative variance in the systematic component (bias) of the volume measurement. (This in essence would be reflected in the error to be associated with the volume calibration curve, for example).}$
- $\sigma_{Ca_b}^2 = \text{relative variance in the systematic component (bias) of the concentration measurement from one set (batch) of determinations to the next set.}$
- $\sigma_{Ma_b}^2 = \text{relative variance in the systematic component (bias) of the mass determination from one set (batch) of determinations to the next.}$

In addition, we will let:

- $A = \text{cost of obtaining a sample from a batch.}$
- $B = \text{cost of analyses including both concentration and mass for a given sample.}$
- $C = \text{cost of making a volume determination on a batch.}$

3. ECONOMIC CONSIDERATIONS IN NESTED SAMPLING AND ANALYSIS

3.1. Derivation of optimum number of samples and analyses per sample

The first case to be considered is that of measuring the total quantity under consideration in increments of $k$ batches where $m$ samples are removed from each batch and $n$ analyses made on each sample. Under this arrangement, assuming successive samples and analyses are as expensive as the first, the cost would be

$$\text{Cost} = Akm + Bkmn + kC$$ (1)
Suppose that we wish to take enough samples and make enough analyses so that the total quantity computed therefrom can be asserted to be within the fraction \( f \) of the true value. It can be shown that this is approximately equivalent to:

\[
(2) \quad t \left[ \frac{\sigma_{V_b}^2 + \sigma_{C_a b}^2 + \sigma_{M_{a b}}^2}{k} + \frac{\sigma_{C_{t r}}^2 + \sigma_{M_{t r}}^2}{k m} + \frac{\sigma_{C_{s t}}^2 + \sigma_{M_{s t}}^2}{k m n} \right]^{\frac{1}{2}} \leq f
\]

where the batch volumes, concentrations and assays are approximately the same, batch by batch, and \( t \) is the factor by which the estimated relative standard deviation of the total quantity estimate is multiplied to convert to a confidence interval. The factor \( t \) and associated confidence can be obtained for all practical purposes in this application from a tabulation of the normal probability curve.

We propose the optimum \( m \) and \( n \) to be those values which minimize the cost, Eq. (1), under the restriction given by Eq. (2).

For the sake of convenience of notation we shall let

\[
\sigma_{b_i}^2 = \sigma_{V_f}^2 + \sigma_{C_{a b}}^2 + \sigma_{M_{a b}}^2 + k\sigma_{V_b}^2
\]

\[
\sigma_{s_i}^2 = \sigma_{C_{t r}}^2 + \sigma_{M_{t r}}^2
\]

\[
\sigma_{a_i}^2 = \sigma_{C_{s t}}^2 + \sigma_{M_{s t}}^2
\]

In this notation the problem becomes one of minimizing

\[
\text{Cost} = A km + B km n + k C
\]

under the restriction

\[
\sigma_T^2 = \frac{\sigma_{b_i}^2}{k} + \frac{\sigma_{s_i}^2}{km} + \frac{\sigma_{a_i}^2}{km n} \left( \frac{f}{t} \right)^2 = r^2
\]

where \( r^2 > \frac{\sigma_{b_i}^2}{k} \)

Application of the differential calculus to this problem, using the method of Lagrange multipliers, and assuming equality, results in the solutions

\[
m = \frac{\sigma_s \left[ \sigma_s + \sigma_a \left( \frac{B}{A} \right)^{\frac{1}{2}} \right]}{k r^2 - \sigma_{b_i}^2}
\]

\[
n = \frac{\sigma_a}{\sigma_s} \left( \frac{A}{B} \right)^{\frac{1}{2}}
\]
with corresponding minimum cost of

\[
\text{Min. Cost} = \frac{1}{r^2 - \sigma_{b1}^2/k} \left\{ A^1 \sigma_a + B^1 \sigma_a \right\}^2 + kC
\]  

(7)

The practical requirement of integer solutions to Eq. (6) will result in a minimum cost differing somewhat from Eq. (7).

4. ECONOMIC CONSIDERATIONS IN COMPOSITE SAMPLING AND ANALYSIS

4.1. Derivation of optimum number of samples and analyses

Let us now consider the effect of compositing on the allocation of sample and analytical effort as well as total effort. One way to composite is by combining the batches themselves, i.e., several batches are transferred to a single container and then measured. Often, however, the case is such that physical considerations, such as limitations imposed by process equipment, or nuclear criticality as in nuclear fuel reprocessing, for example, precludes "batching" several batches. Thus any reduction in total analytical effort over that entailed in analysing a sample or samples from each batch requires compositing aliquots from the batches. In doing this, similar questions are raised: (a) How many samples should be taken from each batch? (b) How many analyses should be made on the composite of all samples? and in addition (c) What size aliquots should be taken from each batch so that the composite is representative of the "composite batch?"

The answer to the third question is intuitively clear: the volume of the aliquots should be in the same proportion as the volume of the corresponding batches. This is accomplished by making the volume of the total sample from the ith batch equal to the product of the ratio of the corresponding batch volume to the total of all batch volumes to which the composite is to apply and any convenient quantity representing the minimum total volume desired in the composite. Symbolically, this is summarized by the equation

\[
A_i = \left( \frac{V_i}{V_T} \right) Q
\]

where \( A_i \) = volume of all sample aliquots from the ith batch, \( V_i \) = volume of the ith batch, \( V_T \) = sum of the volumes of all batches to which the composite is to apply, \( Q \) = minimum total volume desired resulting from compositing all sample aliquots.

To answer the first two questions let us consider the following:

(a) From each batch (i), \( m \) samples each of size \( A_i/m \) are obtained.

(b) Assuming \( k \) batches, the \( mk \) samples so obtained are compositing into a single sample and blended into a homogeneous composition.

(c) From the composite \( n \) aliquots are withdrawn, each to be analysed for concentration of the given constituent.
(d) These $n$ results are then averaged and the average is multiplied by the total of all batch volumes to obtain the total quantity of the given constituent in all batches to which the composite applies.

It is not difficult to show that this procedure, on the average, will result in the true value for the total composition. However, the uncertainty with which this estimate is made in any given case by this procedure depends upon the magnitudes of the various measurement errors that are involved, the amount of total effort reflected in terms of numbers of samples and analyses, and the division of the total effort between sampling and analysis.

For approximately equal batch volumes, concentrations and assays, as is usually the case, the confidence interval construction on the total material contained in all batches as obtained by multiplying the sum of all volumes by the average concentration of the $n$ aliquots from the composite by the average assay of the $n$ aliquots is given under the above notation by

$$
\pm t \left\{ \sigma_v^2 + \sigma_{\text{Ca}}^2 + \sigma_{\text{Ma}}^2 + \frac{\sigma_{\text{Cr}}^2 + \sigma_{\text{Mr}}^2}{km} + \frac{\sigma_{\text{Va}}^2}{k} + \frac{\sigma_{\text{Ma}}^2}{n} \right\} \frac{1}{2}
$$

In this application we shall let

$$\sigma_{\text{bl}}^2 = \sigma_v^2 + \sigma_{\text{Ca}}^2 + \sigma_{\text{Ma}}^2 + \frac{\sigma_{\text{Va}}^2}{k}$$

and $\sigma_{\text{a}}^2$ and $\sigma_{\text{f}}^2$ be defined as before.

Then the optimum $m$ and $n$ are obtained by minimizing

$$\text{Cost} = mA + nB + kC$$

under the restriction

$$\frac{\sigma^2_v}{km} + \frac{\sigma_{\text{a}}^2}{n} + \sigma_{\text{bl}}^2 \leq r^2$$

where $r^2 > \sigma_{\text{bl}}^2$.

The minimizing solution for $m$ and $n$ can be shown to be

$$m = \frac{\sigma_{\text{a}}}{k} \left[ \sigma_{\text{a}} + \left( \frac{B}{A} \right)^{\frac{1}{2}} \sigma_{\text{a}} \right]$$

$$n = \frac{\sigma_{\text{a}}}{r^2 - \sigma_{\text{bl}}^2} \left[ \left( \frac{A}{B} \right)^{\frac{1}{2}} \sigma_{\text{a}} + \sigma_{\text{a}} \right]$$

which gives a minimum cost of

$$\text{Min. Cost} = \frac{(A^{\frac{1}{2}} \sigma_{\text{a}} + B^{\frac{1}{2}} \sigma_{\text{a}})^2}{r^2 - \sigma_{\text{bl}}^2} + kC.$$
5. WHEN TO COMPOSITE

For a given desired precision in the total measurement quantity in a given run Eqs. (7) and (11) provide the key by which one can determine whether compositing presents any economic advantage. It is not difficult to show from these equations that if any appreciable bias exists in any component of measurement and/or no reduction in cost of sampling and/or analysis is effected through the compositing procedures, it is never economically sound to composite. However, often compositing does effect a reduction in the effort required in one or more of the measurement steps. For example, in the recovery of fissile material from spent fuel elements, the nested sampling scheme requires decontamination by bench scale remote extraction processes of each sample before analysis. If one elects to include the decontamination costs as part of sampling, this could be a significant part of the unit sampling cost. Under the compositing scheme, where the compositing is done before decontamination, essentially a single decontamination is required and this cost when prorated over all samples would be negligible in the unit sampling cost. In terms of our model, therefore, under this condition a substantial reduction in $A$ would be expected under compositing. If we let $A_1$ be the cost of sampling (including sample preparation) under the nested procedure, and $A_2$ be the corresponding cost under the compositing scheme using the same desired measurement precision on the total for both cases, then by Eqs. (7) and (11) one should composite whenever

$$\left( \frac{r^2 - \sigma_b^2}{r^2 - \sigma_{b_i}^2 / k} \right)^{\frac{1}{2}} \leq \left( \frac{A_1 \sigma_i + B_1 \sigma_a}{A_2 \sigma_i + B_2 \sigma_a} \right)$$

6. LEVEL OF EFFORT

A fundamental problem in making measurements is that of determining the level of effort to be expended in a particular series. The derivation of a simple rule by which this can easily be determined has yet to be accomplished; however, once the desired measurement precision of a given quantity of material has been determined, Eqs. (7) and (11) are extremely useful in determining whether a change in procedure is justified. As an example, consider again the measurement of dissolver effluent in a fuel recovery plant. Many analytical techniques are available for measuring the uranium concentration. It is usually axiomatic that the precision of the measurement is directly related to the cost, the more precise technique being the more expensive. Let us suppose therefore that under consideration is a different analytical method which will reduce the relative analytical standard deviation defined as $\sigma_a$ in the above by an amount $\Delta \sigma_a$. Correspondingly the unit cost of analysis is increased by an amount $\Delta B$. The question is raised as to whether the change is economically justified. Regardless of whether the nested or composite scheme is used, it can be shown a decision in favour of increased effort (or even reduced effort with a co-
responding reduction in analytical cost) is economically sound whenever

\[ \frac{(B + \Delta B)}{B} < \left( \frac{\sigma_a}{\sigma_a + \Delta \sigma_a} \right)^2 \] (13)

This follows from the fact that the alternative is economically sound if

\[ \text{Cost} (\sigma_a + \Delta \sigma_a, B + \Delta B) < \text{Cost} (\sigma_a, B) \]

or for the nested case

\[ \frac{1}{r^2 - \sigma_{b_i}^2} \left\{ A^t \sigma_t + (B + \Delta B) \frac{1}{2} (\sigma_a + \Delta \sigma_a)^2 \right\} + kC < \frac{1}{r^2 - \sigma_{b_i}^2} \left\{ A^t \sigma_t + B \sigma_a \right\} + kC \] (14)

which reduces to

\[ (B + \Delta B)(\sigma_a + \Delta \sigma_a)^2 < B \sigma_a^2 \] or (13) (15)

An identical result would be obtained when the composite sampling scheme is used.

A similar treatment can be made for assumed changes in any of the parameters of Eqs. (7) and (11).

7. EXAMPLE

Suppose we desire to estimate the content of a series of 25 batches to 1% at the 95% confidence level, i.e. \( t = 2 \) where the following values for the relative errors and cost are assumed:

\[
\begin{align*}
\sigma_{V_t} & = 0.003 & \sigma_{C_{st}} & = 0.0015 \\
\sigma_{C_t} & = 0.003 & \sigma_{M_{st}} & = 0.001 \\
\sigma_{C_a} & = 0.010 & A_1 & = 25.00 \\
\sigma_{M_s} & = 0.002 & A_2 & = 4.00 \\
\sigma_{Mar} & = 0.001 & B & = 25.00 \\
\sigma_{V_b} & = 0.002 & C & = 2.00
\end{align*}
\]

We note from the above that

\[ k = 25 \]

\[ r = \frac{0.01}{2} = 0.005 \]
and calculate from Eqs. (3) and (9)

\[
\begin{align*}
\sigma_{b_1}^2 &= 112.25 \times 10^{-6} \\
\sigma_s^2 &= 13.0 \times 10^{-6} \\
\sigma_a^2 &= 101.0 \times 10^{-6} \\
\sigma_{b_2}^2 &= 7.61 \times 10^{-6}
\end{align*}
\]

To determine whether we should use the composite sampling scheme, we note from Eq. (12) that for this case

\[
\left( \frac{r^2 - \sigma_{b_1}^2}{r^2 - \sigma_{b_1}^2 / k} \right) = 0.921 > 0.841 = \frac{A^1 \sigma_s + B^1 \sigma_a}{A^1 \sigma_s + B^1 \sigma_a}
\]

Thus the criterion for compositing is satisfied. Application of Eq. (10) indicates that the minimum cost is realized when one sample from each batch is composited and seven analyses are made on the composite.

Let us now assume that an alternative analytical method at an increased cost of $15 per analysis is available, for which the relative combined analytical error is 0.008. Application of Eq. (12) to this situation still results in compositing as the most economical procedure; however, when we compute

\[
\frac{B + \Delta B}{B} = \frac{25.00 + 15.00}{25} = \frac{40}{25} = 1.6
\]

and

\[
\frac{\sigma_a^2}{(\sigma_a + \Delta \sigma_a)^2} = \frac{101.0 \times 10^{-6}}{(8 \times 10^{-3})^2} = \frac{101.0}{64} = 1.58
\]

and note from Eq. (13) that

\[
\frac{B + \Delta B}{B} = 1.6 > 1.58 = \frac{\sigma_a^2}{(\sigma_a + \Delta \sigma_a)^2}
\]

we see that a change is not justified.
ECONOMIC CRITERIA APPLIED TO NUCLEAR MATERIALS MANAGEMENT

W. J. SHELLEY AND M. N. KUEHN
MALLINCKRODT CHEMICAL WORKS,
ST. CHARLES, MISS., UNITED STATES OF AMERICA

Abstract — Résumé — Аннотация — Resumen

ECONOMIC CRITERIA APPLIED TO NUCLEAR MATERIALS MANAGEMENT. The management of nuclear materials must always be subsidiary to the primary purpose of nuclear material processing, i.e. the generation of power or the production and fabrication of end products. Therefore, those responsible for management of nuclear materials must be constantly responsive to the needs of the primary production purpose and fit the required systems to the process so as to secure the needs of nuclear materials management at optimum costs.

The nuclear materials management system must concern itself with careful examination of several factors that influence its costs. The control system evolved must complement the process, providing the lowest costs of personnel, analysis and minimum interruption of the operating process. The control system should be integrated with the process needs so that quantitative information derived is available promptly to those responsible for operating supervision.

The recording and reporting system should generate maximum subsidiary data. It should be compatible with the systems employed by suppliers and consumers and carry wherever possible additional information connected with the batches of nuclear material.

Data generated for the control of nuclear materials should only be that needed to ensure that no significant losses, theft, misappropriation or diversion occurs. Complementary data should be subject to the same rigid test of need as that applied to the nuclear material management data.

Procedures, practices, personnel and techniques have been continuously reviewed and revised to ensure the highest quality of nuclear material management performance. To ensure optimum costs balanced with adequate nuclear material control needs, some general rules have been evolved. It is all-important to determine the real needs for the recording and reporting of data. Real economies are attained by the assignment of nuclear materials management, production control and cost responsibilities to a single group. Reliance must then be placed upon such personnel to accept, understand and perform the work. Such personnel selected must be thoroughly and minutely trained in the importance of their activity.

Those responsible for the management of nuclear materials must be continually alert to the smallest kind of aberration in the quality and performance of the personnel, the process, the systems, procedures and techniques. Only through such alertness and willingness to revise, so as to secure improvement, does one secure the optimum balance of costs and needs.
Cette sélection rigoureuse des données de gestion des matières nucléaires en fonction de leur nécessité doit être également appliquée aux données complémentaires.

Les procédures, les méthodes, le personnel et les procédés sont continuellement soumis à des contrôles pour s'assurer que la gestion des matières nucléaires est effectuée dans les meilleures conditions possibles. Certains principes généraux ont été définis en vue de calculer les coûts au plus juste en fonction des exigences d'un contrôle efficace des matières nucléaires. Il est très important de déterminer les besoins réels en ce qui concerne l'enregistrement et la présentation des données. On peut réaliser de sérieuses économies en confiant à un seul service la responsabilité de la gestion des matières nucléaires, du contrôle de la production et de la détermination des coûts. Il faut alors faire confiance à ce personnel qui doit accepter, comprendre et assumer ces tâches. Ce personnel sélectionné doit recevoir une formation très poussée et être pleinement conscient de l'importance de ses activités.

Les responsables de la gestion des matières nucléaires doivent faire continuellement attention aux plus petites défaillances affectant la qualité et le rendement du personnel, des opérations, des systèmes, des procédures et des techniques. Ce n'est qu'au prix de cette vigilance et de cette attitudes à corriger pour perfectionner que l'on peut réaliser un équilibre optimal entre les coûts et le service à assurer.

Les procédures, les méthodes, le personnel et les procédés sont continuellement soumis à des contrôles pour s'assurer que la gestion des matières nucléaires est effectuée dans les meilleures conditions possibles. Certains principes généraux ont été définis en vue de calculer les coûts au plus juste en fonction des exigences d'un contrôle efficace des matières nucléaires. Il est très important de déterminer les besoins réels en ce qui concerne l'enregistrement et la présentation des données. On peut réaliser de sérieuses économies en confiant à un seul service la responsabilité de la gestion des matières nucléaires, du contrôle de la production et de la détermination des coûts. Il faut alors faire confiance à ce personnel qui doit accepter, comprendre et assumer ces tâches. Ce personnel sélectionné doit recevoir une formation très poussée et être pleinement conscient de l'importance de ses activités.
Al establecer un sistema de administración de materiales nucleares hay que estudiar con sumo cuidado los diversos factores que influyen sobre su costo. El sistema de control debe complementar el proceso con costos de personal y de análisis lo más bajos posible y con interrupciones mínimas. Debe ajustarse a las necesidades de las operaciones de forma que los responsables de la explotación puedan disponer sin demora de la información cuantitativa que proporcione.

El sistema de registro e información debe permitir obtener el mayor número posible de datos auxiliares. Debe ser compatible con los sistemas empleados por los proveedores y los consumidores y facilitar toda la información adicional posible respecto de las partidas de materiales nucleares.

Sólo hay que reunir los datos necesarios para poder tener la seguridad de que no se producen pérdidas significativas, robos, errores de asignación o diversiones de material. Los datos complementarios deben someterse a una rígida prueba de necesidad, igual a la impuesta a los destinados a la administración de los materiales nucleares.

Los procedimientos, las técnicas y el personal tienen que estar sujetos a inspección y revisión continuas a fin de asegurar la mejor administración posible del material. Se han establecido algunas normas generales para obtener un equilibrio entre el costo óptimo y las necesidades adecuadamente establecidas del control. Es de suma importancia determinar con exactitud las necesidades reales en lo que respecta al registro y la comunicación de los datos. Se logran considerables economías confiando a un solo grupo de personas la administración de los materiales nucleares, el control de la producción y la determinación de los costos. Esas personas deben tener una conciencia muy precisa de la importancia de su misión.

Los encargados de la administración de materiales nucleares tienen que estar constantemente atentos para poder descubrir cualquier anomalía o defecto en los procedimientos, las técnicas, el proceso de las operaciones y la forma en que el personal lleva a cabo su cometido. Esta atención y la voluntad de corregir los errores permitirán asegurar el equilibrio óptimo de los costos y las necesidades.

INTRODUCTION

During the 3rd International Conference on the Peaceful Uses of Atomic Energy in Geneva in 1964, Dr. Glenn T. Seaborg, Chairman of the United States Atomic Energy Commission, stated that it seemed appropriate that on the 25th anniversary of the discovery of nuclear fission the conference was meeting to inaugurate what might be known to future historians as the "Age of Nuclear Power". Dr. Seaborg then summarized for the conference the technological and scientific progress made in the first 25 years. This symposium on the management of nuclear materials seems appropriate also, since, with the advent of economic nuclear power, the emphasis of the industry will shift from scientific demonstration to business management. The future of the industry will continue to depend upon solid scientific and technological improvement, but will also depend upon astute business and materials management.

The demonstration of nuclear fission in 1939 and the subsequent development of an immense industry dealing in materials of high intrinsic and strategic value, whose uncontrolled release would endanger the entire modern industrial environment, presented those responsible with completely new demands for management information. The management of materials with high intrinsic value had, of course, been considered and established by those industries processing the noble metals and gem stones. Experience in the physics, chemistry, and security problems subsequently presented by the nuclear industry had never been developed on the scale required. In addition, the prohibition of scientific interchange between knowledgeable persons throughout the world applied equally to those with managerial responsibility. Techniques and procedures for the management of nuclear materials were developed by individual, small groups. It speaks well for
the management personnel responsible in those early pioneering days that none of the predicted possible catastrophies to our environment have occurred nor have been threatened through carelessness in handling large quantities of these extremely hazardous materials. It seems that a desirable result of this Symposium might well be that each of us resolve to assure future generations that such a catastrophe will never occur because of inattention to our management task.

The material accountancy procedures evolved in the early days of the nuclear industry have been improved and have become more effective. As with most human progress, it would be quite easy for us here today to criticize the techniques and procedures initially employed. Such should not be our purpose. Rather, we should ensure that the collective wisdom generated over the first 25 years in the management of these materials be organized in such a way as to provide a determinate philosophy for the next 25 years, with only nominal improvement of techniques and technology.

MALLINCKRODT PARTICIPATION

Mallinckrodt Chemical Works first became involved in the nuclear industry, at the request of the United States Government, by refining impure ores and concentrates to pure uranium dioxide in 1942. Materials produced in the spring of that year were used in the first demonstration of controlled nuclear fission at the University of Chicago in November 1942. Subsequent expansion in scope and size have maintained the Company's interest and position in the feed material section of the nuclear industry throughout the ensuing 23 years. During this entire period, nuclear material management techniques and procedures have been evolved, evaluated, improved, discarded and replaced to their present stage of application. The criteria applied to this evolution and the reason for such criteria are described in this paper.

DEFINITIONS

Throughout this paper several types of organizations will be discussed. The production organization directed by a general manager means the company, division or other entity responsible for processing radioactive material. The operating organization supervised by an operations manager means that portion of the production organization that is directly responsible for the operation of processing equipment to produce a radioactive or nuclear product. The nuclear materials management organization is primarily responsible for material accountancy.

NUCLEAR MATERIALS MANAGEMENT ORGANIZATION

The establishment of a nuclear materials management section in an organization processing radioactive materials is beset by some important questions, all related to the economics of that organization. As with the
establishment of any new section, nuclear materials management will cost money. Every competent manager wants to keep the costs of his operation as low as possible while obtaining a maximum amount of information for management purposes. Over the years our organization has considered a number of important questions regarding this function. They include the following:

1. Is the function of nuclear materials management one of control, such as that generally assigned to a controller's department for funds? Is it one of audit, that is, comparable to many staff groups that examine the activities of others against a set of standard procedures? Or is it one that actively participates in the day-to-day accomplishment of the primary purpose of the organization?

2. In addition to its primary function, can nuclear materials management contribute to the organization in a collateral sense because of its skill or information sources?

3. Since nuclear materials management is involved in the technical aspects of the process, should it include technical personnel, or should the technical staff established for other purposes provide the required service?

4. Nuclear materials management activities intrude into each processing operation of the organization and, therefore, significant funds are expended at its direction. Is the staff sufficiently capable of judging the appropriate level of such expenditure and evolving the optimum quantity and quality of activities?

5. Should the corporate responsibility for material accountancy be completely delegated to the nuclear materials management staff?

Each of these questions must receive a favourable answer and be continually reviewed as the situation changes.

Level of reporting

The production organization is responsible for the material in its possession. It must report to a regulatory body, accounting for its custodianship of the material, since it has a strategic value. It must protect its financial assets because of the high intrinsic value of the material in its possession. It is normally bound by a contractual relationship with its customers, not only for costs and funds but also for the quantity and quality of materials in its possession for each individual contract.

In most production organizations, those individuals reporting directly to the general manager represent a large part of the production organization's interest. As a consequence, personnel reporting to the general manager of a production organization generally represent sales, manufacturing, administrative activities, staff activities and technical efforts. The person responsible for nuclear materials management should report directly to the general manager or to his immediate subordinate. The alternate position is considered the minimum level for reporting. An immediate subordinate of the general manager may have nuclear materials management responsibility in addition to other support activities. It has been our experience that to secure the proper operating, staff, administrative and technical attention to the important function of nuclear materials management, the
nuclear materials manager must report directly to the general manager or his immediate subordinate.

Nature of function

The function of nuclear materials management can be described as narrowly or as broadly as the general manager desires. We believe that nuclear materials management includes elements of the audit function, the control function and an element of day-to-day participation in the work of the production organization. These various elements require extremely precise definition in the minds of all personnel to adequately secure the appropriate degree of co-operation and contribution to successful nuclear materials management. The activities within the responsibility of the nuclear materials manager may be further distributed to specific persons or groups. However, he should ensure that the jobs done by these persons do not go beyond the proper sphere of responsibility of the nuclear materials manager. The relationship between the nuclear materials manager and his immediate superior must be one of utmost candour and maximum mutual exchange. This relationship requires absolute trust in each other and an extremely high degree of judgment so as to avoid unnecessary administrative reaction due to variation in control techniques or reporting methods.

Collateral contribution

The data required for appropriate discharge of the responsibilities of nuclear materials management occurs in almost every function of the production organization. If examined closely, the data will be found to contain all the information relative to the flow of materials through such an installation. Since so many of the business aspects of the production organization are related to the flow of materials, certain economies can be realized by assigning to the nuclear materials management group collateral responsibilities in production control, budgeting, production scheduling, quality reporting, raw material acquisition and scheduling, production costing, and perhaps cost accounting and manpower scheduling. The nuclear materials management organization should not, however, be assigned too many collateral functions to the detriment of the nuclear materials management performance. We have found that the responsibility for material accountancy, production scheduling, raw materials scheduling, traffic, and production reporting fit very well under the supervision of a single person. The data required for one mutually supports the data required for the other. Spreading the work among more than one supervisor results in the unnecessary proliferation of supervisory interests. In this way, we have secured a high degree of economy in discharging the nuclear materials management responsibility, and at the same time giving the people assigned to this function wider activity.

Technical involvement

Material accountancy in the nuclear industry is intimately involved with technical skills. During the period of our experience, we have found that
the skills include those of analytical chemistry, materials handling, industrial hygiene, mechanical and chemical engineering, statistics and accountancy. Such a wide range of knowledge is extremely difficult to find in one person or train in one person to a level of competency required for adequate discharge of the nuclear materials management responsibility. Unnecessary separation of each of these many technical disciplines places an undue burden on the nuclear materials manager and his staff. Close integration of the needs of the nuclear materials management with process design engineering is a minimum requirement. Immediate availability of analytical chemists, statisticians, or mathematicians is of utmost importance. The advice of professional systems personnel is needed frequently. In the current organization we have generally provided, at some level, several of these skills. Such an arrangement has worked successfully in our case. Analytical chemistry and process engineering have generally been excluded from direct responsibility for nuclear materials management. The nuclear materials manager has generally had in his immediate staff, however, statistical, accounting and some engineering skills. His influence on the organizations primarily responsible for other skills is one of persuasion and, on occasion, is secured by the support of his superior or the general manager.

**Staffing**

With the wide range of possible responsibilities assignable to the nuclear materials management group, it becomes imperative that personnel selection be carefully considered. Since nuclear materials management includes an audit function, the individual manager or supervisor must possess a high degree of personal integrity. It is desirable, since he is operating in a staff function, to include personal characteristics of persuasiveness, resoluteness, patience and courage. This description can apply to many staff function managers, but in the nuclear industry, selection of the nuclear materials manager should be of primary importance. The best person available, possessing these characteristics, should be selected. His subordinate organization should be composed of people with significant quantities of these characteristics. The nuclear materials manager is subject to a wide range of influence that often seeks to compromise the principles he must support so as to achieve lower costs, higher operating efficiency, reduced record keeping and analytical work. Exemplary judgment as to the appropriate level of these activities in the context of the production organization is mandatory. As mentioned previously, the function itself must be flexible, and therefore his subordinate personnel must be alert, knowledgeable of the process, and constantly aware of their complementary role to the operations organization so as to serve their ultimate purpose of adequate nuclear materials management and the production organization. The person who seeks to establish the absolutes in systems or exactness is predestined to fail in his important role and secure only large quantities of data at extremely high cost to the production organization.
Delegation

The amount of responsibility for nuclear materials management delegated to the nuclear materials manager and his staff is uniquely dependent upon the relationship between the general manager and the nuclear materials manager. Given sufficient integrity, candour and trust by both parties, this delegation of responsibility can be almost complete with a minor amount of reporting to the general manager. As with any superior-subordinate relationship, optimum costs are secured when this delegation is as complete as possible. The appropriate balance of responsibility is within the province of each general manager. He should carefully examine his nuclear materials manager's qualifications, skills and personal characteristics. Since this is an important portion of the production organization's responsibility, he should not hesitate, at least initially, to insist on control information and reporting requirements in excess of that required of other similar staff positions. He must carefully tailor his attitude and behaviour in accordance with this important function so that the production organization, especially that portion responsible for operations, is continually aware that the general manager is interested and attentive to the needs for adequate nuclear material management.

TECHNIQUES AND PROCEDURES

So far we have only considered the initial organization of this important function. Once organized and the responsibilities assigned, it then becomes imperative that the nuclear materials manager establish certain techniques and procedures to secure the data, the personnel and the control required to discharge his responsibility properly. It is at this point that his relationship to the operating organization tends to be established. His insistence on excessive data can result in the complete disregard of his minimum needs. He must carefully relate his needs for information to the process he is controlling. He should ensure that sufficient analytical and statistical effort are exerted to provide him with reliable, quantitative data about the movement of materials. His control of the data flow surrounding the movement of material must be absolute. He must adopt the attitude that no materials move without proper documentation. Optimum low cost systems are not designed in an office remote from the processing operation; intimate knowledge of the operating organization is necessary to properly accommodate its needs. The nuclear materials manager must be intimately familiar with the process and the data requirements of the operating manager and his organization. His role in this regard is a complementary one rather than a subordinate or dominant one. It is desirable that he knows as much about that apparatus as those responsible for its operation.

Analytical and Statistical

The management of nuclear materials requires the analysis of a wide variety of solutions, solids and gases, or the contained radioactive components. The skills of the analytical chemist are tested and extended by
the complexity of chemical measurement of the processing materials of the nuclear industry, as best evidenced by the session spent on the subject in this Symposium. The application of these skills to the needs of the nuclear materials management is perhaps the largest single item of cost in this field of activity. The application of these skills should, therefore, be made in such a way to secure the maximum possible return. For instance, the nuclear materials manager should make use of the latest techniques of statistical evaluation to secure sufficient assurance of the measurement of radioactive materials without requiring excessive analytical work that would raise the cost of his efforts. The application of statistics need not apply only to the analytical work but should apply to the examination of all measurements. The nuclear materials manager would use statistics in selecting an optimum programme of sampling; establishment of appropriate lot sizes; the frequency of inventories; the frequency of weighing and check weighing; the accuracy of scales; the accuracy of sampling systems, the size of samples; and the reliability of testing procedures. The use of mathematics and statistics to measure the performance of equipment and personnel is a significant aid in securing optimum costs.

Control

The delegation to the nuclear materials manager should include authorization to move material only with his personnel in attendance. In many instances, this practice permits key personnel to be in charge of criticality supervision. On other occasions, the practice permits the isolation of an area that in the past has shown large discrepancies in material accountancy. The nuclear materials manager should insist upon such control only where absolutely necessary, using other methods of defining the cause of discrepancies wherever possible.

Personnel

The selection, training and supervision of nuclear materials management personnel is probably the most important factor in securing the most favourable cost system. As suggested earlier in describing the qualities of the nuclear materials manager, certain important personal characteristics are extremely vital in such personnel. However, the selection of the appropriate systems, procedures, and recording and reporting techniques is vital to personnel whose normal role is often repetitive and monotonous. The supervisor must exercise every skill of audit, work simplification, and clarity to provide records of sufficient interest to ensure attention. His training methods must seek to secure absolute accuracy and reliability. His relationships with subordinate personnel throughout his organization must be of the most candid to secure their acceptance of and adherence to his own strong principles. Supervision must be above reproach. Clear appraisal, corrective and disciplinary efforts must be applied promptly. The acceptance of sloppy, inefficient or inaccurate work weakens the entire system and should be corrected promptly. The production organization manager who secures a reliable nuclear materials management organization through the efforts of all subordinate supervision is indeed fortunate.
Recording and reporting

The recording and reporting function must be as simple and as directly correlatable to the movement of material as possible. Excessive pieces of paper and excessive data tend to destroy the effect of staff work of long standing. If possible, the needs of the operating supervision for production information should be recorded simultaneously with the information required for material accountancy. In this way, he secures the participation of the operating organization in producing accurate, reliable numbers for both purposes.

Data processing

Wherever possible, the nuclear materials manager should use those mechanical systems of data processing available to him. He should design manual systems for eventual mechanization wherever possible. It has been our experience that correctly designed manual systems are easily transferable to electronic data processing machinery or automatic data processing machinery.

The nuclear materials manager should try to ensure that his data will provide information required by operating supervision so as to minimize parallel recording and reporting systems. Often this practice requires that data for material accountancy should be provided more rapidly than the normal monthly frequency, but the results in additional cooperation, attention, and reliability are well worth this extra effort.

SUMMARY

The establishment of a nuclear materials management organization within a production organization and its subsequent operation requires the close attention of the entire management. The nuclear materials management organization must account for materials having high value, personnel hazards and an importance beyond those materials previously processed by most industrial organizations. Each production organization should carefully examine the importance of this new function to its organization and establish the level to which it should report. The characteristics of the function should be clearly established for all management personnel. Since this new function will concern itself with large amounts of data about the flow of materials, its skills and information should be widely applied to the needs of the general manager. The nuclear materials management should have available a wide range of technical skills to provide support and staff aid. The nuclear materials manager should carefully select those personnel who will report to him to ensure that the qualities of integrity, alertness and process knowledge are available to the people responsible for the successful operation of this function. The general manager of the production organization should thoroughly examine his willingness to delegate his organization's responsibility for nuclear materials management to a subordinate manager and, after balancing the factors involved, select the most favourable degree of delegation.
The nuclear materials manager should carefully assess his needs for analytical and statistical data, constantly adjusting them to the needs of the production organization. He must exercise the degree of control necessary but none in excess. His design of reporting and recording procedures should provide him with sufficient information to meet his needs but no more. Where possible, such procedures should be designed with simplicity, clarity and adaptability to mechanical or electronic data processing systems. He must select, discipline, evaluate and supervise the personnel contributing to this function with the highest degree of available managerial skill.

It has been our experience that the application of these criteria to the nuclear materials management function will secure the most favourable balance of cost, skill and assurance of adequate organization protection for this important responsibility.

**DISCUSSION**

J. SORNEIN: It would appear from Mr. Tingey's paper* on a materials control system for an enriched uranium chemical processing facility and from the paper which you have just presented, that there are two schools of thought on the relative positions which (1) operation, (2) laboratory and analysis, and (3) nuclear materials management ought to occupy within the organization.

Phillips Petroleum favour an arrangement whereby (3) is subordinate to (2), whereas you seem to support a system in which (1), (2) and (3) are all on a par. Is this correct?

W. J. SHELLEY: Our operation is primarily one of production, and our experience is that a nuclear materials management function which includes such activities as production scheduling and reporting gives the most satisfactory results. Other approaches we have tried have not worked as well, although for other types of organization, e.g. research organizations, they might possibly be as satisfactory.

---

* These proceedings (SM-67/23).
ECONOMIC MANAGEMENT OF A CENTRE FOR PROCESSING RADIOACTIVE RESIDUES. The paper shows the differences between the operations of an industrial undertaking and an installation for residue processing and describes the auditing system of an industry concerned with various phases of processing.

It shows that the auditing programme is necessarily and intimately connected with the general organization of the entire undertaking and that it is a useful tool of economic management. In this connection, the paper deals at length with the auditing principles used in arriving at

- the overall cost of the final product,
- the specific cost of each processing phase,

GESTION ECONOMIQUE D'UN CENTRE DE TRAITEMENT DE RESIDUS RADIOACTIFS. Après avoir exposé les différences existant entre les opérations effectuées par une entreprise industrielle et par une installation de traitement des résidus, le mémoire décrit la comptabilité d'une industrie comprenant plusieurs phases de traitement.

Il montre comment le plan comptable est nécessairement et intimement lié à l'organisation générale de l'ensemble de l'entreprise et est un outil utile de gestion économique.

A cette fin, le mémoire développe les principes comptables permettant d'établir:

- le prix de revient global du produit final obtenu,
- les prix de revient spécifiques de chaque phase de traitement partiel.

ЭКОНОМИЧЕСКИЙ АСПЕКТ ОБРАЩЕНИЯ С РАДИОАКТИВНЫМИ ОТХОДАМИ В ОБРАЗУЮЩЕМ ЦЕНТРЕ. После изложения различий, существующих между операциями на промышленном предприятии и на установке по обработке отходов, в исследовании приводятся данные об отчетности на промышленном предприятии с несколькими фазами обработки.

Показывается, насколько план отчетности необходим и тесно связан с общей организацией всего предприятия, а также является полезным для экономического управления.

В этой связи разрабатываются принципы отчетности, позволяющие установить:
пудель себестоимость полученной готовой продукции;
удельные себестоимости каждой фазы частичной обработки.

GESTION ECONOMICA DE UN CENTRO DE TRATAMIENTO DE DESECHOS RADIOACTIVOS. Después de exponer las diferencias que existen entre las operaciones efectuadas en una empresa industrial y en una instalación de tratamiento de desechos, el autor analiza la contabilidad de una industria que comprende diversas fases de tratamiento.

Muestra cómo el plan contable está necesaria e íntimamente vinculado a la organización general del conjunto de la empresa y es un instrumento útil de gestión económica.

Con este fin, desarrolla los principios contables que permiten establecer:

- el precio de costo global del producto final obtenido,
- los precios de costo específicos de cada fase de tratamiento parcial.

559
1. ORGANISATION GENERALE D'UN CENTRE DE TRAITEMENT

Un centre de traitement de résidus liquides et solides se compose en général de:
- une installation de traitement des effluents liquides,
- une installation de traitement et de stockage de résidus solides (les liquides et solides peuvent se subdiviser en différentes catégories d'après les types de traitement utilisés),
- une installation de décontamination,
- un laboratoire de contrôle et de recherches,
- un atelier,
- une section de transport.

Le tableau I schématise l'organisation générale d'un centre de traitement. Cette organisation ainsi que les méthodes de traitement dépendent de nombreux facteurs: débit des effluents liquides et quantités de résidus solides, activités, modes de transport, possibilités de rejet et doses maximales admises pour le rejet.

2. ORGANISATION COMPTABLE

A l'organisation générale se rattache une organisation comptable, dont le plan constitue l'armature essentielle. En réalité, ce plan comptable se confond souvent avec l'organisation comptable, car il matérialise en détail la technique comptable mise en œuvre.

L'organisation comptable comprend naturellement la conception des documents, des livres comptables à utiliser et des relevés, des «rapports comptables» et «rapports financiers» à établir. Certains documents, à l'origine purement comptables, peuvent être une source de renseignements techniques intéressants. C'est ainsi que l'étude des mouvements des stocks de résidus conduit tout naturellement à établir des rapports résidus/quantités traitées qui sont un moyen de juger scientifiquement la valeur d'un procédé de traitement.

Une attention particulière doit cependant être accordée à l'établissement de renseignements comptables facilitant le contrôle technique de l'installation. L'annexe I schématise les différents comptes qui découlent des fondements de l'organisation générale. Le plan comptable ne détaille que les comptes 7 et 8, comptes de frais et comptes de prix de revient respectivement, c'est-à-dire les comptes essentiels d'une comptabilité d'exploitation que l'on peut opposer à la comptabilité générale. Il faut signaler que les différentes techniques d'épuration pourraient être bien plus utilement comparées si le même plan comptable était appliqué dans toutes les exploitations.

3. OBJECTIFS DE LA COMPTABILITE D'EXPLOITATION

Les objectifs de la comptabilité sont fixés par les exigences essentielles d'une direction scientifique. La comptabilité d'exploitation a pour objectifs principaux:
- la connaissance des charges, des frais et des revenus ainsi que leur analyse et leur confrontation,
la connaissance des écarts entre les coûts et les prix de revient réels, d'une part, et les coûts ou les prix de revient préétablis d'autre part. Ces prix peuvent être en rapport avec une commande déterminée ou avoir trait à une période antérieure.

TABLEAU I

ORGANISATION GENERALE D'UN CENTRE DE TRAITEMENT

![Organigramme des diverses étapes de traitement des déchets](image)
En pratique, l'analyse des comptes d'exploitation devrait permettre de répondre à un grand nombre de questions relatives aux frais groupés selon différents critères, aux influences résultant d'une extension des installations, de la mise en application de nouvelles techniques, d'une activité plus intense, etc.

La comparaison des données comptables et des prévisions constitue un contrôle de gestion et un contrôle financier complémentaires. Ainsi pour ne citer qu'un exemple: «Il est d'un grand intérêt de comparer les coûts ou prix de revient réels à ceux qui devraient être obtenus dans des conditions préalablement définies comme normales. Si les coûts préétablis sont fondés sur des normes rationnelles de consommation de matière, de temps de travail, de prestations de services, les écarts qu'ils présentent avec les coûts réels ont une signification très utile pour l'orientation de la gestion. Convenablement analysés pour détecter les causes qui se rattachent, soit à des variations de quantités, soit à des variations de prix, soit à des modifications dans le niveau d'activité, ils permettent à la direction de situer les responsabilités engagées dans la recherche de la productivité et de la rentabilité.» [1]

4. PRIX DE REVIENT DES TRAITEMENTS SPECIFIQUES - PRIX DE REVIENT COMPOSES

En général, les opérations effectuées dans une entreprise industrielle consistent en l'achat de matières, la transformation de celles-ci, la vente des produits finals obtenus. Les opérations de transformations successives contribuent à accroître la valeur des matières mises en œuvre. La comptabilité industrielle doit permettre d'obtenir la valeur des produits commerciaux obtenus.

La situation est toute différente pour une installation de traitement de résidus radioactifs. Il ne s'agit plus ici d'obtenir un produit commercial, mais bien un produit qui, tout en répondant à des caractéristiques spécifiques bien déterminées, n'a aucune valeur.

Ces caractéristiques imposent des traitements spécifiques conduisant au produit final. Les opérations successives sont à la base d'une cumulation de dépenses de plus en plus élevées au fur et à mesure que le traitement avance. Une comptabilité qui ne pourrait fournir qu'une estimation «globale» d'une masse aussi complexe de dépenses serait absolument déficiente; on lui demande de donner, non seulement le prix de revient global ou composé du traitement, mais aussi le prix de revient des opérations intermédiaires, afin de permettre de juger et de comparer les différents modes de traitement. De plus, il est nécessaire, en vue de l'établissement du bilan de fin d'année, de connaître, à un stade quelconque des opérations de traitement, la valeur des frais jusqu'à la dernière phase des opérations.

Pour répondre à ces impératifs, on établit deux prix de revient.

4.1. Prix de revient des traitements spécifiques

Le plan comptable permet de calculer le prix de revient de chaque traitement spécifique, c'est-à-dire le prix de revient d'un traitement sans
tenir compte de ses antécédents ou de ses conséquents. L'établissement du prix de revient par type de traitement est extrêmement utile. En effet il permet:
- le contrôle de l'évolution des frais au cours du temps,
- l'examen des variations des prix de revient en fonction des quantités traitées,
- l'étude préliminaire éventuelle en cas de changement de machine ou de procédé de traitement,
- la détection rapide des variations de prix de revient, ainsi que des causes de ces variations,
- l'étude, par l'examen du rapport (PR final)/(PR par phase), de l'importance d'une des opérations de traitement par rapport au traitement dans son ensemble.

4.2. Prix de revient composé

Le prix de revient composé groupe les dépenses afférentes au traitement complet d'un type déterminé de résidu. Il comprend le coût du traitement spécifique et le coût de toutes les opérations subséquentes.

A titre d'exemple, le prix de revient composé des effluents chauds peut se schématiser comme indiqué au tableau II. Il comprend:
- le traitement spécifique en tant qu'effluents de ce type,
- le traitement et l'évacuation des boues chaudes produites par la première phase (insolubilisation, stockage provisoire, stockage définitif),
- le traitement et l'évacuation des effluents tièdes (traitement spécifique, insolubilisation des boues tièdes produites, stockage et déversement des eaux résiduaires).

5. FRAIS FIXES ET FRAIS VARIABLES

Les méthodes d'analyse des frais, dont nous avons donné les notions ci-dessus, sont insuffisantes pour permettre à la comptabilité d'atteindre certains objectifs voulus par la direction. Elles négligent, en effet, l'analyse des frais en fonction du temps ou en fonction des quantités produites.

Le montant des dépenses ramené à l'unité produite est influencé par le degré d'activité de l'entreprise. Cette influence résulte de ce que les dépenses ou les frais totaux de l'entreprise comprennent une partie fixe, indépendante de ces quantités.

Dire que les frais indirects ou les frais généraux sont des frais fixes et que les frais directs sont des frais variables selon les quantités produites est une affirmation trop simpliste. Ainsi, des frais fixes ne sont pas nécessairement invariables dans le temps; en fait leur ensemble varie par paliers. Nous allons examiner quelques enseignements que l'on peut tirer de l'analyse des frais fixes et des frais variables.
5.1. Coûts calculés en fonction du degré d'exploitation

Les frais totaux variables, proportionnels à la production, sont toujours imputés au prix de revient; les frais fixes ou à peu près fixes, quel que soit le degré d'activité (heures de travail, quantités traitées...) ne sont imputés totalement au prix de revient que pour autant que l'activité soit normale.

Si l'activité est inférieure à la normale, il n'est imputé au prix de revient qu'un pourcentage des frais fixes égal au pourcentage de la capacité de production réellement atteinte pendant la période considérée.

**TABLEAU II**

**PRIX DE REVIENT COMPOSE**

Effluents chauds

<table>
<thead>
<tr>
<th>Nature des dépenses</th>
<th>F/m³</th>
<th>%</th>
</tr>
</thead>
<tbody>
<tr>
<td>- Traitement spécifique (PRS)</td>
<td>...</td>
<td>...</td>
</tr>
<tr>
<td>- Traitement évacuation des boues (PRC de 1 m³ de boues): Volume de boues chaudes</td>
<td>...</td>
<td>...</td>
</tr>
<tr>
<td></td>
<td>Volume d'effluents chauds</td>
<td></td>
</tr>
<tr>
<td>- Traitement et évacuation des effluents tièdes (PRC de 1 m³ d'effluents tièdes)</td>
<td>...</td>
<td>...</td>
</tr>
<tr>
<td>PRC</td>
<td>...</td>
<td>...</td>
</tr>
</tbody>
</table>

Note: PRS = prix de revient du traitement spécifique
PRC = prix de revient composé

5.2. Frais différentiels [2]

Les frais différentiels sont ceux qui résultent d'un changement dans le degré d'activité ou dans les modes d'opération de l'entreprise; leurs montants peuvent être positifs ou négatifs. Ils peuvent représenter, soit le coût réel de l'exécution des opérations additionnelles, par exemple celui des unités produites, pour passer d'un degré d'activité à l'autre, soit la différence de coût d'opérations semblables, mais exécutées les unes avant le changement de méthode, les autres après.

Les frais différentiels unitaires sont appelés prix limites parce qu'ils marquent la limite du choix qui est laissé au chef d'entreprise dans ses décisions; ils sont souvent calculés hors comptabilité.
La connaissance et l’application de la méthode des différences sont indispensables à la direction pour choisir la solution la plus économique. Prenons un exemple:

La figure 1 montre la variation des coûts variables totaux (CVT), des coûts variables moyens (CVM) et des coûts variables marginaux (CVm) en fonction du degré d’activité. Elle fait ressortir les trois zones classiques au point de vue des prix de revient:
- la zone dégressive;
- la zone de stabilité ou zone optimale, marquée par les degrés 5 et 7; les prix unitaires restent sensiblement stables, ce sont les prix de revient les plus économiques;
- la zone progressive caractérisée par une augmentation du prix de revient unitaire.

L'exemple suppose une seule division interne alors qu'une installation de traitement en comporte plusieurs; il est donc utile de déterminer la zone optimale de production de chacune des sections de traitement.

Pour atteindre le rendement optimal, chaque section doit travailler à 100% de sa capacité. En pratique, il n'est pas possible d'atteindre cet objectif et cela pour plusieurs raisons:
- dépendance très importante de l'extérieur,
- nécessité de conserver une marge de sécurité suffisante,
- différence d'habileté entre le personnel de chaque section,
- erreur commise lors de la répartition des frais fixes entre les différentes sections,
- certaines sections sont la suite logique du traitement, d'autres dépendent plus de l'extérieur.

Néanmoins, l'étude de l'équilibre à maintenir entre les diverses sections peut conduire, par exemple, à agrandir ou modifier des installations, ou à modifier le type de fourniture de certains produits. C'est ainsi qu'actuellement, au Centre de Mol, l'insolubilisation des déchets solides et des boues s'effectue à l'aide de bitume fondu sur place. Ce bitume est reçu en fûts de 250 litres. Dans le cas d'un accroissement des quantités de matières solides à insolubiliser, il sera plus intéressant de faire venir le bitume fondu dans des réservoirs de plus grandes dimensions.

6. CONCLUSIONS

Le calcul des prix de revient de traitement spécifique et des prix de revient composés, et l'analyse des frais permettent de tirer une foule de conclusions intéressantes sur la marche d'une installation de traitement et son développement futur.

Il est extrêmement important de calculer les prix de revient avec précision: non seulement leur valeur est intéressante, mais également les documents dont ce calcul requiert l'établissement et qui peuvent servir de guide dans la gestion économique d'un centre de traitement de résidus.

ANNEXE I

<table>
<thead>
<tr>
<th>Classe</th>
<th>0 Comptes de capital</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>1 Comptes des valeurs immobilisées</td>
</tr>
<tr>
<td></td>
<td>2 Comptes des valeurs disponibles</td>
</tr>
<tr>
<td></td>
<td>3 Comptes des valeurs réalisables</td>
</tr>
<tr>
<td></td>
<td>4 Comptes des tiers débiteurs</td>
</tr>
<tr>
<td></td>
<td>5 Comptes des tiers débiteurs</td>
</tr>
<tr>
<td></td>
<td>6 Comptes de répartition et comptes d'ordre</td>
</tr>
<tr>
<td></td>
<td>7 Comptes de frais</td>
</tr>
</tbody>
</table>

|       | 70 Frais d'administration générale |
|       | 71 Frais de personnel |
72 Frais d’approvisionnement
73 Frais industriels généraux
74 Frais des services auxiliaires

740 Frais du service de contrôle
741 Frais du service atelier
742 Frais du service transport
743 Frais du service laboratoire de recherches

75 ...
76 Frais de la division des effluents liquides

760 Frais communs
761 Frais du service effluents froids

7610 Rémunérations et charges sociales
7611 Frais administratifs
7612 Frais de bâtiment
7613 Approvisionnements
7614 Frais de l'équipement
7615 Impôts et assurances
7616 Amortissements

762 Frais du service effluents tièdes
763 Frais du service effluents chauds
764 Frais du service déversement
765 Frais du service traitement et insolubilisation des boues

77 Frais de la division des déchets solides

770 Frais communs
771 Frais du service triage
772 Frais du service cisaillement
773 Frais du service compression
774 Frais du service incinération
775 Frais du service traitement et insolubilisation des cendres
776 Frais du service insolubilisation des déchets solides
777 Frais du service stockage provisoire
778 Frais du service stockage définitif

78 Frais de la division décontamination

780 Frais communs
781 Frais du service buanderie
782 Frais du service décontamination

80 Comptes de prix de revient composés

800 Prix de revient composé des effluents froids
8000 Approvisionnements
8001 Salaires directs
8002 Charges sociales sur salaires directs
8003 Application d'un quantum de frais d'administration générale
8004 Application d'un quantum de frais techniques généraux
8005 Frais de traitement ou quantum de tel service de traitement

801 Prix de revient composé des effluents tièdes
802 Prix de revient composé des effluents chauds
803 Prix de revient composé des boues
804 Prix de revient composé des cendres
805 Prix de revient composé des déchets solides
806 ... 
807 Prix de revient composé de la buanderie
808 Prix de revient composé du déversement

REFERENCES


DISCUSSION

G. JASNY: You alluded to the use of incremental costs. Do you use them in the routine analysis of the efficiency of your operations? And how in your accounting system do you consider or take into account the costs of material loss and crossover?

J. van LINT: The incremental costs method is not used in our routine accounting. It did, however, guide us in our choice of a new and more powerful solid waste press. This change, by virtue of a greater reduction in volume, enabled us to reduce considerably the cost of subsequent insolubilization and storage.

The materials we are treating are of negligible or no value; therefore our accounting system does not take material loss into account.

J. SORNEIN: In the light of our experience in France, I must say we rend to be wary of excessively detailed calculations of basic costs when an establishment is engaged in activities as numerous and various as those described by Mr. van Lint.

In such cases the total of fixed expenditure is much higher than that of proportional expenditure, and apportionment of the fixed expenditure is very difficult. We therefore often prefer to concentrate solely on the proportional expenditure and the differential costs of the various operations.

D.E. GEORGE: The development of a cost system based on sound measurements is very important, for the value of the material is so great
in relation to the cost of an operation that comparatively small variations in quantities will lead to large variations in unit cost. This demonstrates the need for a good nuclear materials management programme if a plant is to know its unit costs with any degree of accuracy.
ASPECTS ECONOMIQUES COMPARÉS DU TRAITEMENT ET DU STOCKAGE DES PRODUITS DE FISSION ISSUS DES USINES DE TRAITEMENT DE L’URANIUM NATUREL IRRADIE

L. THIRIET, P. LESUR ET R. GIRAUD,
COMMISSARIAT À L’ÉNERGIE ATOMIQUE, PARIS
ET
J. WANLIN,
SOCIETE SAINT-GOBAIN TECHNIQUES NOUVELLES,
PARIS, FRANCE

Abstract — Résumé — Аннотация — Resumen

COMPARISON OF THE ECONOMIC ASPECTS OF THE TREATMENT AND STORAGE OF FISSION PRODUCTS FROM INSTALLATIONS PROCESSING IRRADIATED NATURAL URANIUM. The paper summarizes the main policies adopted for the management of fission product effluents and briefly describes the essential technical features of the stainless steel storage installations (in acid medium) used in France, giving a breakdown of the costs involved. The dependence of these costs on the activity of the solutions stored and on their heat release is shown.

The second part of the paper discusses the cost of storage in terms of feasible tank size. The optimum size for such tanks and the rate at which they are placed in service are determined in relation to the characteristics of the fission products to be stored and to the respective capacities of the installations for processing irradiated natural uranium. It is shown that although storage costs depend on the assumptions made regarding the useful life of installations, rates of interest and operating costs, optimum policy decisions (as to size and rate of entry into service) are independent of these assumptions, being determined solely by the variation in the cost of tanks, according to size. These are the factors which enter into the optimized cost of storing these effluents indefinitely.

In the third part, this method of indefinite storage is compared with other possibilities of fission product management, e.g. vitrification and separation of Cs and Sr. The paper discusses the economics of the various methods and summarizes some possible long-term solutions.

СРАВНИТЕЛЬНЫЕ ЭКОНОМИЧЕСКИЕ АСПЕКТЫ ОБРАБОТКИ И ХРАНЕНИЯ ПРОДУКТОВ ДЕЛЕНИЯ, ПОЛУЧАЕМЫХ НА ЗАВОДАХ ПО ПЕРЕРАБОТКЕ ОБЛУЧЕННОГО ПРИРОД-
НОГО УРАНА. После упоминания основных методов обращения с продуктами деления в жидкем виде, дается краткое описание основных технических характеристик применяемых во Франции установок для хранения из нержавеющей стали (в кислой среде) и структуры стоимость этих установок. Показывается, как эта система зависит от активности находящихся на хранении растворов и от выделения ими тепла.

Во второй части исследуется стоимость их хранения в зависимости от размеров резервуаров, которые могут быть построены, и определяются размеры и максимальные пределы их использования с учетом характеристик подлежащих хранению продуктов деления и различных возможностей завода по переработке облученного природного урана. Показывается, что если стоимость хранения предположительно зависит от срока службы установок, процента на капитал и эксплуатационных расходов, то выбор оптимального метода независим от этого и связан лишь с законом изменения стоимости этих резервуаров с учетом их размеров. Таким путем получают оптимизированную стоимость бесконечного хранения жидких продуктов.

В третьей части с этим решением бесконечного хранения сравниваются другие возможные меры обращения с продуктами деления: остекловывание и выделение цезия и стронция. Обсуждается экономика различных методов хранения и намечаются возможные решения проблемы на длительный срок.

1. POLITIQUES DE GESTION DES PRODUITS DE FISSION

1.1. Considérations générales

Les produits de fission constituent un effluent fatal du traitement de l'uranium naturel irradié. Leur stockage est très onéreux et présente des risques inhérents à leur très forte activité. L'accumulation de ces déchets, qui croît avec le développement des installations nucléaires, exige un examen économique attentif des diverses politiques possibles de gestion. Nous les résumerons d'abord, avant d'évoquer 1er aspect économique de la solution adoptée actuellement: le stockage dans des réservoirs en acier inoxydable et la taille optimale des réservoirs à utiliser pour stocker une production donnée. Nous examinerons enfin des perspectives à moyen terme.

Le problème posé consiste à chercher les moyens de conserver pendant un temps très long et de la façon la plus sûre possible les solutions de produits de fission issues de la première étape de récupération des matières fissiles (U, Pu) qui suit la dissolution des barreaux de combustible irradié.
sortant de la pile. Ces solutions contiennent en pratique la totalité de l'activité du combustible irradié.

Devant les risques réels, mais impossibles à apprécier en ce moment, des méthodes envisagées, la tendance mondiale est de rechercher les procédés de stockage, impliquant éventuellement des traitements, les plus sûrs possibles. C'est ainsi que le stockage acide est considéré comme très sûr, mais que la fixation des produits de fission sous forme de verre non léchable est considérée comme encore plus sûre.

En outre, une autre préoccupation qui se fait jour est celle de la récupération des produits de fission, notamment du césium-137 et du strontium-90, pour une vente immédiate ou différée sur des marchés potentiels parfois importants. Trois politiques de gestion sont le plus souvent envisagées pour ces effluents: le stockage liquide dans des réservoirs en acier inoxydable, le stockage liquide avec séparation préalable du césium-137 et du strontium-90, la vitrification. La figure 1 schématishe ces trois politiques de gestion.

1.2. Stockage liquide dans des réservoirs en acier inoxydable

Une première étape consistera à réduire le volume de produits de fission à stocker par concentration, puis à les conserver dans des réservoirs étanches munis d'un système de réfrigération interne. Ils sont dotés d'un système de protection en cas de fuite (lèchefrite, piscine, bacs de béton...) et sont, de plus, soumis à des contrôles de sécurité. Leur programme de mise en service prévoit d'inclure dans l'installation un réservoir de sécurité permettant la réception immédiate du contenu du réservoir éventuellement défaillant.

Sur le plan pratique, seul le stockage liquide est en exploitation depuis un certain nombre d'années. A l'heure actuelle, les deux autres procédés sont, soit au stade pilote, soit tout au début du stade d'exploitation, et ne présentent encore qu'une très faible capacité de traitement. Cette constatation est illustrée en France par l'importance du stockage en acier inoxydable (en milieu acide), notamment celui du Centre de Marcoule.

1.3. Stockage liquide avec séparation préalable du césium et du strontium

Après dix ans environ, les activités du strontium-90 et du césium-137, radioéléments à vie longue (périodes: 28 et 30 ans respectivement), représentent l'activité quasi totale de l'effluent stocké. Une technique possible consiste donc à éliminer par voie chimique, d'une façon plus ou moins complète, ces deux éléments nocifs à long terme après concentration de l'effluent.

Il reste alors, d'une part à stocker l'effluent résultant qui pourra à long terme être repris par la station de traitement des effluents (STE), et d'autre part à stocker spécialement ou à mettre sous forme commerciale les sources de strontium et de césium après purification. Les débouchés commerciaux de ces deux radioéléments pourraient contrebalancer les investissements nécessaires à leur séparation.

On conserve la technique du stockage liquide à moyen terme, cette variante ayant à long terme un caractère de plus grande sécurité.
FIG. 1. Présentation schématique des 3 techniques de stockage à long terme des solutions de produits de fission
1.4. Vitrification

La solution concentrée de produits de fission est transformée en blocs de verre au cours des trois opérations suivantes: évaporation, calcination, fusion de la masse, qui peuvent s'effectuer soit successivement, soit simultanément, ou encore en groupant les deux premières.

Après avoir été évaporée et calcinée, la solution de produits de fission donne un calcanat qui se déverse dans un four de fusion où sont introduits les adjuvants nécessaires à la vitrification. Le verre, après affinage, s'écoule à la base du four et forme des blocs qui sont ensuite emmenés, gainés ou non, sur l'aire de stockage. Ces blocs sont empilés dans des cellules de béton suivant une certaine géométrie, le système de refroidissement étant assuré par une ventilation d'air en circuit fermé.

Nous poursuivons des études sur ces techniques, dont les principales difficultés sont actuellement:
- la nécessité de l'emploi d'une technologie de haute température pour un travail à haute activité,
- l'entraînement des poussières et d'éléments volatils (ruthénium-106 et césium-137) au cours de la cuisson, qui nécessite des installations supplémentaires d'épuration des gaz et de condensation des vapeurs,
- les installations de sécurité nécessaires pour la manipulation de telles activités (très haute activité spécifique des blocs de verre),
- la qualité du verre obtenu afin d'éviter toute contamination par léchage de l'eau (lixivation) et son homogénéité évitant son effritement à long terme.

Cette technique onéreuse exigera encore de nombreuses mises au point, la composition du verre étant liée à celle de l'effluent, mais elle présente l'avantage de permettre de stocker à long terme sous une forme stable et sous un volume réduit, c'est-à-dire avec une plus grande sécurité, les solutions concentrées de produits de fission de très haute activité.

2. ASPECTS TECHNIQUES ET ECONOMIQUES DES INSTALLATIONS DE STOCKAGE DES PRODUITS DE FISSION

2.1. Caractéristiques techniques essentielles de l'une de ces installations

Le principe du stockage liquide des produits de fission au Centre de Marcoule a été exposé au cours du Colloque sur le traitement et le stockage des déchets à forte radioactivité (Vienne, octobre 1962).

A l'heure actuelle, le stockage comporte essentiellement quatre cuves cylindriques à axe vertical, de 62 m³ utiles, enfermées dans quatre casemates de béton enterrées, étanches et ventilées. Un placage inox du fond et des parois assure une rétention équivalent à la capacité totale des cuves dans le cas de fuites catastrophiques.

CHAMBON, M., RODIER, J., "Concentration et stockage des produits de fission à l'usine d'extraction du plutonium de Marcoule", Treatment and Storage of High-Level Radioactive Wastes, IAEA, Vienna (1963) 43.
Le risque de contamination de la nappe phréatique a conduit à l'adoption de cette lèchefrite. La sécurité accrue qui en résulte a incité les exploitants à abandonner le type précédent de stockage au profit du nouveau; celui-là, cependant maintenu en état de service, tiendra lieu de dispositif de secours. Les lèchefrites sont équipées d'un détecteur de niveau; une alarme point bas permet de déceler l'écoulement de quelques litres de solutions.

La sélection des matériaux de construction a conduit à l'adoption d'un acier inoxydable 18 - 12 - Mo (C<0, 02%) pour les cuves (épaisseur 10 mm) et serpentins de refroidissement. Quant aux lèchefrites, elles sont façonnées avec de l'acier inoxydable 18 - 12 (C<0, 03%).

Nous allons examiner les diverses composantes de l'équipement des cuves et annexes en insistant principalement sur les réalisations nouvelles:

a) Les cuves

L'évacuation de la chaleur est assurée exclusivement par l'eau des serpentins de refroidissement. Normalement, cette eau déminéralisée et traitée circule dans des échangeurs à plaques alimentés par de l'eau industrielle à la température de 22°C. La capacité de refroidissement d'un tel système, cuve pleine, tous serpentins en parallèle, est de 175 000 frigories/h; la température des produits de fission est maintenue à 35°C. Il est possible de n'utiliser qu'une fraction de cette capacité par la mise en service d'une partie seulement des serpentins, ceux-ci comportant six circuits indépendants.

L'agitation des liquides est assurée par un pulseur central dont le rôle est de remettre en suspension les éventuels précipités qui pourraient se former et décanter.

Il existe également un dispositif de balayage de l'atmosphère de la cuve. L'alimentation en air comprimé de ces dispositifs est assurée par un poste de détente.

b) Les annexes

Circuits de refroidissement. Quatre pompes centrifuges de 70 m³/h appartenant aux anciennes installations ont été affectées aux circuits de quatre nouveaux réservoirs. Deux de ces pompes avaient été prévues pour le stockage alcalin et se trouvaient disponibles. Les deux autres assuraient la circulation de l'eau dans le condenseur du groupe frigorifique. Cette fonction a été supprimée par une alimentation directe du condenseur en eau industrielle.

Circuits de transfert. Les équipements de transfert des solutions actives initialement installés ont été réutilisés: il s'agit de transfert par relevage sous vide et mesureurs d'une part, par siphonnage et pompes de reprise d'autre part. En outre, les nouveaux réservoirs ont été équipés d'éjecteurs à vapeur.

Circuits de reprise des gaz. Les cuves sont équipées individuellement d'un cyclone dévésiculeur. Elles sont reliées à une installation commune comprenant: un condenseur, une colonne de lavage et un dispositif de sé-
chage des incondensables. Ceux-ci sont aspirés à travers des filtres absolus par un ventilateur (débit: 900 m$^3$/h) et refoulés vers les circuits d'extraction de la ventilation générale du bâtiment.

- **Prises d'échantillons.** Une installation nouvelle a été montée.

- **Commande et contrôle.** Une extension du tableau existant a été réalisée.

- **c) Les services généraux**

  Aucune extension n'a été nécessaire pour la production de vide, d'air comprimé, d'électricité (la vapeur est fournie par les services généraux de l'usine Plutonium). Quant à la ventilation des locaux dont les gaines ont été raccordées au réseau de l'installation primitive, elle a dû être renforcée par deux ventilateurs capables d'un débit unitaire de 3600 m$^3$/h affectés respectivement au soufflage et à l'extraction.

### 2.2. Structures de coût de ces installations

Les coûts des installations construites permettent d'évaluer une structure type des coûts d'une installation de stockage susceptible d'être réalisée dans l'avenir (tableau I).

En première approximation, la part revenant aux protections biologiques représente 5% des coûts directs, celle de l'évacuation de la chaleur (agitation + réfrigération) 15%, quant à celle concernant l'installation des réservoirs et des annexes, elle représente 80% des coûts directs.

On voit donc que l'activité spécifique influençant les protections aussi bien que le dégagement spécifique de la chaleur influençant les circuits de refroidissements - paramètres liés à la nature du combustible traité - ne représentent qu'une part relativement faible des investissements.

Pour obtenir des coûts minimaux de stockage en acier inoxydable, il faut tenir compte des frais d'exploitation et de renouvellement des réservoirs et optimiser la taille des installations. Nous allons évoquer maintenant cette recherche de la politique de mise en service optimale des réservoirs de stockage.

### 3. OPTIMISATION DES COUTS DE STOCKAGE DES PRODUITS DE FISSION DANS DES RESERVOIRS EN ACIER INOXYDABLE

#### 3.1. Considérations générales

Les frais d'investissement pour le stockage des produits de fission dans des réservoirs en acier inoxydable représentent l'essentiel du coût de stockage de ces effluents radioactifs. La taille du réservoir en sera donc le facteur déterminant. Il est commode d'exprimer les frais d'investisse-

---

2 Il faut noter que la protection biologique est essentiellement assurée par les structures béton de l'ouvrage. Le coût du génie civil est donc considérablement grevé par les impératifs de protection contre les rayonnements.
TABLEAU I

STRUCTURE DES COUTS D'UNE INSTALLATION DE STOCKAGE DES PRODUITS DE FISSION DANS DES RESERVOIRS EN ACIER INOXYDABLE

(250 m³ utiles)

<table>
<thead>
<tr>
<th>Catégorie</th>
<th>Coût en francs fr. a)</th>
<th>Pourcentage</th>
</tr>
</thead>
<tbody>
<tr>
<td>Génie civil</td>
<td>1300 000</td>
<td>19</td>
</tr>
<tr>
<td>Protections biologiques</td>
<td>300 000</td>
<td>5</td>
</tr>
<tr>
<td>Matériel, machines</td>
<td>2900 000</td>
<td>44</td>
</tr>
<tr>
<td>dont</td>
<td></td>
<td></td>
</tr>
<tr>
<td>(agitation PF b))</td>
<td>(640 000)</td>
<td>(10)</td>
</tr>
<tr>
<td>(réfrigération PF b))</td>
<td>(310 000)</td>
<td>(5)</td>
</tr>
<tr>
<td>Tuyauterie, robinetterie</td>
<td>1540 000</td>
<td>23</td>
</tr>
<tr>
<td>Electricité, contrôle</td>
<td>580 000</td>
<td>9</td>
</tr>
</tbody>
</table>

6600 000                   100

Architecte industriel       1400 000

TOTAL                     8 000 000

a) Taxes comprises (12% environ)
b) Produits de fission

ment des réservoirs en fonction de leur taille, comme on le fait habituel-
ment pour les usines de traitement des combustibles irradiés ou les usines
chimiques, par une loi de la forme

\[ \frac{I_2}{I_1} = \left( \frac{t_2}{t_1} \right)^\alpha \]

I₁ et I₂ représentant les frais d'investissement relatifs aux tailles t₁ et t₂,
\( \alpha \) étant un exposant spécifique du procédé utilisé.

L'étude de la variation des frais en fonction de la taille de chaque ca-
tégorie indiquée précédemment permet d'établir une valeur pondérée de \( \alpha \)
voisine de 0,5, se rapportant à l'ensemble de l'installation.

D'autre part, les frais d'exploitation relatifs à ces stockages sont géné-
ralement très limités. Le personnel d'exploitation nécessaire est en
effet peu nombreux, les consommations de réactifs et d'utilités assez réduites. Si l'on exprime ces frais sous forme de pourcentage des frais d'investissement, on arrive à une valeur de l'ordre de 3 à 4%.

Les frais de personnel varient en fonction de la taille selon une loi approximative de la forme

\[ \frac{M_2}{M_1} = \left( \frac{t_2}{t_1} \right)^{\alpha_1} \]

où

- \( M_1 \) et \( M_2 \) représentent les coûts de main d'œuvre relatifs aux tailles \( t_1 \) et \( t_2 \),
- \( \alpha_1 \) a une valeur relativement faible, de l'ordre de 0,3.

Les frais de réactifs et d'utilités varient par contre selon une loi proportionnelle à la taille.

Il en résulte, en première approximation, que ce pourcentage des frais d'exploitation, faible par rapport aux frais d'investissement, reste sensiblement du même ordre de grandeur lorsque la taille des réservoirs s'accroît.

La connaissance de cette loi de variation des frais d'investissement en fonction de la taille des réservoirs ne suffit pas à elle seule à déterminer la politique optimale de stockage en milieu liquide des produits de fission. Il s'agit en fait de résoudre un problème de stockage permanent au moindre coût total actualisé.

3.2. Taille optimale des stockages associés à une usine de flux de production constant

3.2.1. Position du problème

Etant donné un flux de production constant \( v \) (m³/an) issu d'une usine de taille donnée, il s'agit de savoir quelle est la taille optimale à donner au réservoir qui sera construit l'année 0 et la cadence des renouvellements.

Supposons qu'on prenne des décisions de construction une fois par an au plus. Le réservoir construit l'année 0 peut avoir une taille \( v, 2v, 3v \) et plus généralement \( Kv \). On supposera dans la suite de cette étude que les frais d'investissement d'un réservoir de taille \( Kv \) sont inférieurs à ceux de deux réservoirs de tailles \( K_1v \) et \( K_2v \) (avec \( K_1 + K_2 = K \)).

Cette hypothèse signifie que, dans le domaine des tailles possibles de réservoirs étudiés, il n'existe pas de contrainte technologique et qu'on n'est pas obligé d'accroître la taille des installations par duplication.

En rappelant que le coût des réservoirs en fonction de la taille est donné par une loi de la forme

\[ \frac{C_2}{C_1} = \left( \frac{t_2}{t_1} \right)^{\alpha} \]

avec \( \alpha = 0,5 \), et que les frais d'exploitation restent sensiblement du même ordre de grandeur lorsque la taille des réservoirs s'accroît, nous allons étudier successivement l'influence sur le coût du stockage des frais d'ex-
ploitation et du renouvellement de stockage à la fin de leur durée de vie, avant d'aborder le problème de la taille optimale de ces stockages.

3.2.2. Calcul des frais d'exploitation

Désignons par $E$ les frais d'exploitation annuels d'un réservoir de coût $I$ ($E$ représente un certain pourcentage $e$ de $I$), et par $d$ la durée de vie du réservoir. Jusqu'à la fin de sa durée de vie, les frais d'exploitation cumulés et actualisés s'élèvent à

$$
\sum E = \frac{E}{i} \left[1 - (1 + i)^{-d}\right] = If(i, d, e)
$$

$i$ étant le taux d'actualisation.

La dépense relative à la construction d'un réservoir est donc

$$I + If(i, d, e)$$

3.2.3. Renouvellement des réservoirs

Après sa durée de vie $d$, le réservoir doit être reconstruit. La dépense sera alors

$$I + If(i, d, e) + \frac{I + If(i, d, e)}{(1 + i)^2} + \ldots + \frac{I + If(i, d, e)}{(1 + i)^{dp}}$$

$p$ étant le nombre de renouvellements.

Pour une reproduction à l'identique, cette expression tend vers

$$C = [I + If(i, d, e)]g(i, d)$$

avec $g(i, d) = 1/[1 - (1 + i)^{-d}]$.

Il en résulte que deux politiques de construction de stockage de tailles différentes $t_1$ et $t_2$ correspondant à des coûts $I_1$ et $I_2$ seront caractérisées par les coûts

$$C_1 = [I_1 + I_2f(i, d, e)]g(i, d)$$

$$C_2 = [I_2 + I_2f(i, d, e)]g(i, d)$$

et que le classement de $C_1$ et $C_2$ (si $C_1 \leq C_2$ par exemple) sera le même que celui de $I_1$ et $I_2$.

Le problème de la taille optimale des stockages se pose dès lors indépendamment de la prise en compte des frais d'exploitation et du renouvellement des réservoirs à la fin de leur durée de vie, et même de la durée de vie supposée des réservoirs à condition qu'elle soit identique dans tous les cas comparés.
3.2.4. Taille optimale des stockages

Le problème de la recherche de la taille optimale du stockage à construire l'année 0 peut donc se présenter de la manière suivante: le réservoir pourra avoir une taille $v$, $2v$, ... et plus généralement $Kv$. On suppose que l'on ne prendra que des décisions annuelles et que la capacité du réservoir ne sera pas inférieure à $v$. Le volume optimal cherché sera alors de la forme $V = Kv$.

La loi de variation du coût d'investissement en fonction de la taille est de la forme

$$I_2 = I_1 \left( \frac{V}{v} \right)^\alpha = I_1 K^\alpha$$

La politique de stockage de volume $v$ (politique 1) avec reproduction à l'identique à la fin de la durée de vie des réservoirs s'écrit

$$C_1 = I_1 \frac{1+i}{i}$$

La politique de stockage de volume $Kv$ (politique 2) avec reproduction à l'identique s'écrit

$$C_2 = I_2 \left( \frac{1}{1-(1+i)^{-K}} \right)$$

La politique 2 sera meilleure que la politique 1 si

$$I_2 \left[ \frac{1}{1-(1+i)^{-K}} \right] < I_1 \left( \frac{1+i}{i} \right)$$

soit, comme $I_2 = I_1 K^\alpha$

$$\frac{K^\alpha}{1-(1+i)^{-K}} < \frac{1+i}{i}$$

L'étude de cette inéquation montre que l'optimum sera atteint pour $K = 18$ si $i = 0,07$ et $\alpha = 0,5$. Par conséquent, on devra construire un réservoir de taille $18v$, qui sera renouvelé tous les 18 ans.

3.2.5. Influence du réservoir de sécurité

En réalité, dans l'installation de stockage, il est prévu un certain nombre de réservoirs de sécurité. Nous prendrons l'hypothèse la plus largement acceptée, qui consiste à construire un réservoir de sécurité pour cinq réservoirs en service au maximum.

Le calcul effectué de la même manière montre alors que $K = 17$ et que par conséquent on devra construire un réservoir de taille $17v$ qui sera renouvelé tous les 17 ans.
3.2.6. Cas de l'obsolescence

Nous avons jusqu'à présent considéré le cas théorique d'un renouvellement de stockage à l'infini. Pratiquement, en fonction d'autres politiques de gestion des produits de fission, on peut envisager le stockage liquide d'une façon temporaire jusqu'à un horizon donné. Considérons les horizons suivants: $H_1 = 6$ ans, $H_2 = 10$ ans, $H_3 = 15$ ans, et examinons le choix optimal donné par les deux politiques de stockage dans chacun de ces cas:

<table>
<thead>
<tr>
<th>Horizon</th>
<th>Politique optimale</th>
</tr>
</thead>
<tbody>
<tr>
<td>6 ans</td>
<td>$K = 3$</td>
</tr>
<tr>
<td>10 ans</td>
<td>$K = 4$</td>
</tr>
<tr>
<td>15 ans</td>
<td>$K = 5$</td>
</tr>
</tbody>
</table>

On voit, comme on pouvait s'y attendre, que l'hypothèse d'une obsolescence à relativement moyen terme modifie considérablement les choix optimaux.

4. PERSPECTIVES A MOYEN TERME

Les coûts optimaux de stockage des produits de fission dans des réservoirs en acier inoxydable doivent permettre d'évaluer l'intérêt relatif d'autres politiques possibles de gestion.

Une remarque s'impose: Les coûts de stockage liquide sont bien connus alors que ceux de la séparation Cs-Sr et de la vitrification sont fondés sur de toutes premières estimations. Toutefois, en se référant à des études récentes en ce domaine, il est possible, en première approximation, d'analyser les différents facteurs qui concourent à la détermination d'une politique optimale de gestion.

En premier lieu, l'influence des programmes (tonnage annuel d'uranium à traiter) sur la taille des réalisations industrielles possibles, donc sur la déformation éventuelle des comparaisons des politiques possibles, doit être déterminée. Toutefois, l'exposant trouvé par la loi de variation du coût en fonction de la taille ($\alpha = 0,5$) relatif au stockage liquide en acier inoxydable et celui vraisemblable de la loi de croissance des investissements des deux autres procédés (vitrification et séparation Cs-Sr) ($\alpha = 0,4 - 0,5$) semblent indiquer que le classement économique relatif des procédés de stockage d'une part, et de vitrification ou de séparation d'autre part, ne sera pas modifié dans un très large domaine de variation des tailles possibles des réalisations.

Sur le plan de la sécurité, la vitrification est le procédé jugé actuellement le plus sûr; il présente par contre l'inconvénient de fixer sous une forme pratiquement irrécupérable les produits de fission éventuellement commercialisables.

On peut dès lors se demander si une solution mixte consistant à récupérer soit le Cs ou le Sr, soit ces deux radioéléments, ne pourrait pas être envisagée. Le produit le plus susceptible de trouver des débouchés
semble être le césium qui, par ailleurs, est le produit de fission le plus léchable donc le plus utile à séparer avant la vitrification.

Si l'on admet que les produits de fission seront fixés dans des verres, pour des raisons de sécurité, la récupération de ce césium avant vitrification acquiert alors un sens marginal du point de vue économique. Une première étude, pour des usines de traitement de taille moyenne (3 à 5 t/j), indique que la production marginale éventuelle de césium-137 avant vitrification permettrait d'obtenir le curie de césium-137 à 1,25 - 1,75 franc. La récupération éventuelle du strontium-90 par résines échangeuses d'ions selon un procédé voisin de celui de l'extraction du césium-137 mettrait probablement le curie de strontium-90 à un niveau analogue (1,25 - 1,75 franc).

En conclusion, si l'extrême fluidité actuelle du marché des produits de fission n'incite pas à prévoir dans l'immédiat la récupération du césium-137 et du strontium-90, il se dégage une tendance générale à considérer de plus en plus le stockage liquide des produits de fission comme une politique d'attente à moyen terme permettant de parfaire les techniques qu'on pourrait envisager à long terme impliquant un procédé de traitement et présentant une plus grande sécurité. Le choix entre les différentes politiques de gestion (vitrification, séparation ou solution mixte) pourra s'orienter lorsque les premiers éléments du futur marché des produits de fission commenceront à apparaître.
TECHNICAL SUPPORT ACTIVITIES OF A NUCLEAR MATERIALS MANAGEMENT PROGRAMME

S.C.T. McDowell
UNITED STATES ATOMIC ENERGY COMMISSION,
WASHINGTON, D. C., UNITED STATES OF AMERICA

Abstract — Résumé — Аннотация — Resumen

TECHNICAL SUPPORT ACTIVITIES OF A NUCLEAR MATERIALS MANAGEMENT PROGRAMME. The development of a nuclear materials management programme in the United States of America has recognized from its inception the value and need of strong technical support. The success of that programme has depended to a large extent on the development of a closely allied technical support effort. This effort has drawn on the technical competency of top governmental, industrial and academic consultants, in addition to that within the USAEC. Under the planning, development and administration of the USAEC's Division of Nuclear Materials Management, a broad spectrum of technical activities has evolved. These include:

(a) The establishment of an Advisory Committee for Standard Reference Materials and Methods of Measurement,
(b) Preparation and USAEC co-ordination with the National Bureau of Standards in the development of a series of uranium and plutonium chemical and isotopic standards,
(c) Research and development programmes designed to provide improved measurement techniques,
(d) Compilation and publication of a book of selected measurement methods for uranium and plutonium.

Each of these technical support activities is discussed in some detail, including the conditions that gave rise to their need and development, and their application to the USAEC's nuclear materials management programme.

Included is a discussion of the USAEC's Advisory Committee for Standard Reference Materials and Methods of Measurement, which was established to provide guidance to the nuclear materials management programme and recommend research and development activities. Resulting from these recommendations was a USAEC co-operative effort with the National Bureau of Standards for the development of chemical and isotopic standard reference materials of uranium and plutonium; particular attention is devoted to the results of that joint effort.

The need for research and development efforts in areas of mutual interest is examined, and the cooperation of other nations of the world is elicited in the development of internationally acceptable standards and methods of measurement.
L'auteur étudie également le rôle du Comité consultatif des étalons de matières nucléaires et des méthodes de mesure, qui a été créé pour donner des directives quant à la gestion des matières nucléaires et faire des recommandations concernant les activités de recherche et de développement. À la suite de ces recommandations, l'USAEC et le National Bureau of Standards ont travaillé en collaboration à l'établissement d'étalons chimiques et isotopiques d'uranium et de plutonium; une attention particulière est consacrée à l'étude des résultats de cet effort conjoint.

L'auteur dégage la nécessité d'une collaboration dans les recherches et les études concernant les domaines présentant un intérêt commun et souligne le rôle de la coopération avec les autres pays du monde en vue de la mise au point de normes et de méthodes de mesure applicables internationalement.

TEHNIČESKOE OBSPECHENIE POMOCI PROGRAMME KONTROLJA I UCHETA MATERIJALOVA.

При разработке программы контроля и учета ядерных материалов в США с самого начала признавались значение и необходимость ее технического обеспечения. Успех этой программы в значительной степени зависел от разработки тесно связанных мер по ее техническому обеспечению. При разработке этих мер учитывалась техническая компетентность консультантов высшей квалификации, работающих в государственных, промышленных и академических учреждениях, помимо технической компетентности консультантов Комиссии по атомной энергии США (КАЭ). Благодаря принятию Отделом контроля и учета ядерных материалов КАЭ мер в области планирования, разработок и административного руководства была разработана широкая программа технической деятельности.

Эта деятельность включает:

а) создание консультативного комитета по эталонным материалам и методам измерений;
б) подготовку и координацию КАЭ с Национальным бюро стандартов химических и изотопных стандартов урана и плутония;
в) программы исследований и разработок, предназначенные обеспечить усовершенствованные методы измерений;
г) составление и публикация книг, посвященных некоторым методам измерений, относящимся к урану и плутонию.

Несколько подробно обсуждается каждый из этих видов деятельности по техническому обеспечению, в том числе условия, приведшие к потребностям и необходимости расширения указанной деятельности, а также ее использование применительно к программе КАЭ по контролю и учету ядерных материалов.

Описывается дискуссия в Консультативном комитете по эталонным материалам и методам измерений КАЭ, который был создан для руководства программой контроля и учета ядерных материалов и дачи рекомендаций относительно деятельности в области исследований и разработок. На основе этих рекомендаций КАЭ предприняла совместные действия с Национальным бюро стандартов с целью разработки химических и изотопных эталонов урана и плутония; в докладе особое внимание уделяется результатам этих совместных усилий.

Рассматривается необходимость проведения исследований и разработок в тех областях, которые представляют общий интерес, и доказывается необходимость сотрудничества других стран в деле разработки общеприемлемых эталонов и методов измерений.

ACTIVIDADES DE APOYO TECNICO A UN PROGRAMA DE ADMINISTRACION DE MATERIALES RADIATIVOS. Cuando en los Estados Unidos se trató de establecer un programa de administración de materiales nucleares se reconoció en seguida la importancia y la necesidad de darle un firme apoyo técnico. El éxito de este programa ha sido en gran parte debido al esfuerzo llevado paralelamente a cabo con ese fin no sólo con los recursos de la Comisión de Energía Atómica sino también con el asesoramiento de eminentes especialistas de los servicios oficiales y de los medios industriales y docentes. Entre las actividades de apoyo técnico al programa planeadas y dirigidas por la División de Administración de Materiales Nucleares de la AEC cabe mencionar las siguientes:

a) Establecimiento de un comité consultivo sobre patrones y métodos normalizados de medición,
b) Preparación de una serie de patrones químicos e isotópicos de uranio y plutonio en cooperación con el National Bureau of Standards,
c) Ejecución de un programa de investigaciones y desarrollo para el mejoramiento de las técnicas de medición,
d) Redacción y publicación de un libro sobre métodos seleccionados de medición del uranio y el plutonio.

El autor analiza la mayor parte de estas actividades de apoyo técnico e indica cómo surgió su necesidad y la forma en que se aplican al programa de administración de materiales nucleares de la AEC. Expose además las funciones del comité asesor de la AEC sobre patrones y métodos normalizados de medición, creado para
INTRODUCTION

The inventory of nuclear materials of the US Atomic Energy Commission (USAEC) is valued in excess of $12,000 million and presents a formidable challenge to those of us engaged in nuclear materials management. A significant portion of this inventory is located at more than 200 domestic facilities and sites. However, we believe we have devised a nuclear materials management programme within the United States that is capable of meeting this challenge. The broad scope of that programme is detailed in another presentation at this Symposium [1]. I would like to review the technical support activities which have played an important role in its development.

These activities can be divided into three broad categories: nuclear standards, analytical services and research and development related to improved measurement and measurement evaluation techniques. I shall present the results of some of our efforts in these areas, and discuss some of our current activities.

NUCLEAR STANDARDS

Within the broad spectrum of nuclear standards, the USAEC's nuclear materials management programme has been active in two general areas. These are standards for measurement purposes, and general and administrative standards (or standards of practice sometimes referred to as engineering standards).

Standards for measurement purposes

Standard reference materials are a keystone of industrial development. The development of the nuclear industry has been no exception. However, because of the unique characteristics and infancy of that industry many of the early standard reference materials were developed by each corporate entity and were used and accepted only by that entity. In effect, these were internal working standards. Beginning in 1957, the USAEC, in co-operation with the National Bureau of Standards in Washington, D.C., undertook a joint programme to prepare, certify and distribute standard reference materials of plutonium and uranium that would have widespread acceptance [2]. The National Bureau of Standards is the recognized authority in the United States for the certification and distribution of standard reference materials. As a result of that programme there are now available for both domestic and foreign distribution a series of 16 uranium isotopic standards, one plutonium isotopic standard, and a chemical standard for each of those ele-
ments. These standards and their unit cost are shown in Table I. Additional standards are being developed to complete this set or to replace existing standards with more completely characterized materials. These standard reference materials enable analytical laboratories to reduce measurement uncertainties in their analytical procedures and provide the basis for resolving measurement differences between shippers and receivers. This in turn increases the probability of ensuring correct dollar transactions of the many nuclear materials now being exchanged in national and international commerce.

**General and administrative standards**

Such standards involve specifications, codes, symbols, nomenclature, procedures, records and reporting systems, and other standards of practice. Many of the R&D efforts reported subsequently in this presentation have resulted in improved nuclear standards within this general category. A specific example is the study of the measurement and shipment of plutonium nitrate that has added sufficiently to our knowledge to permit successful efforts at standardization, and obtain approval to meet regulatory requirements for shipments within the United States.

We have strongly encouraged the development and acceptance of specifications for nuclear materials management, nuclear materials colour codes, nomenclature, recording and reporting, and other practices. In this connection the USAEC has developed a single shipping or transfer form to document all transfers made by holders of nuclear materials within the United States. As a suggested further step, standardization and acceptance of a transfer document for use on an international scale would seem desirable. Such a standard form would ensure that each receiver of nuclear material would obtain sufficient information to meet his requirements. Also a standard form would have the advantage of a familiar format and size.

In another field, the USAEC, to achieve more economical handling and recovery of uranium and plutonium scrap, has developed criteria for segregating, packaging and labelling of these scrap materials. These criteria, based on years of USAEC operating experience, have formed the basis for establishing scrap classification standards for industrial use.

To provide technical guidance in the development of standard reference materials and approved methods of measurement for materials of special importance to atomic energy activities, an Advisory Committee was established by the US Atomic Energy Commission in March 1956, as the Committee for Uranium Isotopic Standards. Its reconstitution was approved in January 1958, under its present title, the Advisory Committee for Standard Reference Materials and Methods of Measurement, to reflect the broadened scope of its activities. The Committee reviews, evaluates and recommends means for providing standard reference materials (i.e., certified chemical and isotopic standards for uranium, plutonium, etc.) and approved methods of measurement for materials of special importance to atomic energy activities.
# TABLE I

**U.S. DEPARTMENT OF COMMERCE**

**National Bureau of Standards**

**PRICE LIST OF SPECIAL NUCLEAR STANDARD REFERENCE MATERIALS**

<table>
<thead>
<tr>
<th>NBS Standard Reference Material Number</th>
<th>Description</th>
<th>Certified for Plutonium Content</th>
<th>Unit</th>
<th>Price</th>
</tr>
</thead>
<tbody>
<tr>
<td>949-a</td>
<td>Plutonium metal</td>
<td>Pu</td>
<td>0.5g</td>
<td>$34.00</td>
</tr>
<tr>
<td>948</td>
<td>Plutonium sulfate hydrate</td>
<td>Pu-239  79.937  0.700  0.0334</td>
<td>0.25g</td>
<td>40.00</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Uranium Oxide (U₃O₈)</th>
<th>U-234</th>
<th>U-235</th>
<th>U-236</th>
<th>U-238</th>
<th>U</th>
</tr>
</thead>
<tbody>
<tr>
<td>U-005</td>
<td>0.023</td>
<td>0.483</td>
<td>0.0046</td>
<td>99.91</td>
<td>1.0g</td>
</tr>
<tr>
<td>U-010</td>
<td>0.0064</td>
<td>0.991</td>
<td>0.067</td>
<td>98.99</td>
<td>1.0</td>
</tr>
<tr>
<td>U-015</td>
<td>0.009</td>
<td>1.51</td>
<td>0.018</td>
<td>98.47</td>
<td>1.0</td>
</tr>
<tr>
<td>U-020</td>
<td>0.012</td>
<td>2.01</td>
<td>0.18</td>
<td>97.94</td>
<td>1.0</td>
</tr>
<tr>
<td>U-030</td>
<td>0.018</td>
<td>3.01</td>
<td>0.20</td>
<td>96.95</td>
<td>1.0</td>
</tr>
<tr>
<td>U-050</td>
<td>0.028</td>
<td>4.95</td>
<td>0.48</td>
<td>94.98</td>
<td>1.0</td>
</tr>
<tr>
<td>U-100</td>
<td>0.067</td>
<td>10.07</td>
<td>0.38</td>
<td>89.82</td>
<td>1.0</td>
</tr>
<tr>
<td>U-150</td>
<td>0.099</td>
<td>15.13</td>
<td>0.65</td>
<td>84.71</td>
<td>1.0</td>
</tr>
<tr>
<td>U-200</td>
<td>0.125</td>
<td>19.80</td>
<td>0.20</td>
<td>78.86</td>
<td>1.0</td>
</tr>
<tr>
<td>U-350</td>
<td>0.249</td>
<td>34.89</td>
<td>0.170</td>
<td>64.69</td>
<td>1.0</td>
</tr>
<tr>
<td>U-500,</td>
<td>0.512</td>
<td>49.38</td>
<td>0.755</td>
<td>50.03</td>
<td>1.0</td>
</tr>
<tr>
<td>U-750</td>
<td>0.593</td>
<td>75.12</td>
<td>0.252</td>
<td>24.03</td>
<td>1.0</td>
</tr>
<tr>
<td>U-800</td>
<td>0.660</td>
<td>80.07</td>
<td>0.246</td>
<td>19.02</td>
<td>1.0</td>
</tr>
<tr>
<td>U-850</td>
<td>0.64</td>
<td>84.98</td>
<td>0.37</td>
<td>14.00</td>
<td>1.0</td>
</tr>
<tr>
<td>U-900</td>
<td>0.77</td>
<td>90.10</td>
<td>0.33</td>
<td>8.80</td>
<td>1.0</td>
</tr>
<tr>
<td>U-930</td>
<td>1.08</td>
<td>93.27</td>
<td>0.205</td>
<td>5.44</td>
<td>1.0</td>
</tr>
</tbody>
</table>

Standards are available to AEC contractors, AEC or State licensees, and foreign governments which have entered an Agreement for Cooperation with the U.S. Government concerning the Civil Uses of Atomic Energy. The purchase request for these standards must be made on special forms obtainable free of charge from the Department of Commerce, National Bureau of Standards, Reference 310.04, Washington, D.C. 20234.

**Domestic orders must be placed on Form NBS-285 and sent in duplicate to the National Bureau of Standards.** Uranium standards will be shipped by prepaid parcel post. The plutonium standards will be shipped by Express, collect, from Washington.

**Foreign orders must be placed on Form NBS-285A and sent in quadruplicate to the Division of International Affairs, U.S. Atomic Energy Commission, Washington, D.C. 20545.** Uranium standards can be shipped post paid from the National Bureau of Standards, by International Parcel Post. Plutonium standards will be packed for overseas shipment and, unless otherwise advised, will be shipped by air freight, uninsured, FOB, Washington, transportation costs collect. Remittances in payment of foreign orders must be made payable to the National Bureau of Standards and are required in advance. These remittances must be drawn on a bank in the United States and payable at the standard rate of United States currency.
ANALYTICAL SERVICES

In addition to its contractor-operated laboratories, the USAEC has its own independently operated laboratory, the New Brunswick Laboratory (NBL) at New Brunswick, N. J. This laboratory specializes in analytical services, and research and development of new analytical methods and techniques.

The NBL also administers programmes to assist USAEC contractor laboratories in maintaining satisfactory analytical competence. One of these covers materials in the production flow such as UF₆, UO₃ and UF₄, and consists of an inter-laboratory exchange of samples on a continuing basis. Each participating laboratory receives periodic reports that present and discuss the results of the exchange, and each laboratory then makes whatever investigations appear warranted. At the present time, the NBL is also conducting a special programme to evaluate the measurement methods used in the nuclear fuel cycle [3]. This evaluation will serve as an indication of the present capabilities and limitations of the analytical methods, as they are actually used, and will provide laboratories with a basis for comparing their work with that of other analytical laboratories.

RESEARCH AND DEVELOPMENT RELATED TO IMPROVED MEASUREMENT AND MEASUREMENT EVALUATION TECHNIQUES

As improvement in all technical fields is dependent upon research, improvement in the nuclear materials management programme is sought through research into improved measurement techniques. Therefore, to channel technical efforts into the highest priority fields and to minimize duplication of effort, the USAEC's Division of Nuclear Materials Management has undertaken to sponsor as many of these research and development efforts as possible. Some of the projects recently completed or in progress are discussed below.

*Development of non-destructive assay techniques for determining special nuclear material content of unirradiated fuel plates, pellets, air filters and uranium-containing solutions; and examination of potentially useful techniques for assaying irradiated fuels [4]*

Because of the severe economic cost of destructively analysing fuel plates, pellets and various unique nuclear materials of a special nature associated with research and development efforts, there has been a long-standing need for reliable non-destructive assay techniques of acceptable precision and accuracy. While we have made significant advances in the development of such techniques, much remains to be accomplished. Gamma scintillation spectrometers have been developed that will permit determination to within 1-2% of total uranium and uranium-235 content of unirradiated fuel plates, pellets and solutions, and to within ±5-10% of the nuclear material content of air filters. The promising results of preliminary efforts in determining the nuclear material content of air filters have stimulated
much expanded research and development [5]. This non-destructive measurement technique is now being used to evaluate the economic recoverability of such air filters.

The USAEC's New Brunswick Laboratory has developed a gamma-attenuation technique for measuring the total uranium content of flat geometries such as fuel plates. Using low strength europium gamma sources, it is now possible, using the single channel analysers, to determine total uranium and uranium-235 independently (and, hence, isotopic assay) [6].

Non-destructive analysis of irradiated fuels presents a much more difficult obstacle. We have evaluated a number of proposed approaches to this problem. Each approach has had one characteristic in common: the research and development costs and development of a prototype are extremely costly. Any possibility of incorporating such features as portability or transportability appear remote, yet the problems associated with transporting hot fuels to and from such a test facility are only too well known.

Research and development areas that may offer promise include induced fission techniques, X-ray or neutron transmission, and solid-state gamma detectors. Some of the proposals for determining plutonium content are designed for measurements while the assemblies are still in the reactor by measuring resonance flux depression. Other proposals suggest determining the total fissile material content of irradiated fuel assemblies by activation of either the coolant or a substance dissolved in it.

It is most encouraging to hear of some of the promising developments in non-destructive assaying techniques that are being presented at this Symposium.

Compilation, publication and interlaboratory evaluation of methods for the measurement of source and special nuclear materials in the nuclear fuel cycle

The USAEC-sponsored publication, "Selected Measurement Methods for Plutonium and Uranium in the Nuclear Fuel Cycle", by R. J. Jones, was issued in January 1964. Many of the analytical methods contained therein have their reliability expressed as a coefficient of variation. These values were, in most cases, determined by a limited number of laboratories. For a publication of this type to be of the greatest use, it is highly desirable that such values, as well as statements regarding precision and accuracy, be determined on the basis of an inter-laboratory evaluation involving several laboratories. Such an evaluation will soon be completed by the USAEC's New Brunswick Laboratory through the co-operation and participation of the various USAEC laboratories and of qualified private industrial laboratories.

A study to establish measurement techniques, define handling procedures, and design and test shipping containers for plutonium nitrate solutions

Plutonium nitrate is one of the forms in which the U.S. Atomic Energy Commission will purchase privately produced plutonium. Historically, there have been differences between shipper and receiver of several percent in the
plutonium content of plutonium nitrate solutions. Therefore, in 1962, the USAEC undertook a study to determine the causes for, and find means to minimize, the shipper-receiver differences on shipments of plutonium nitrate. Another important aspect of this study was the development of a shipping container for plutonium nitrate that would permit ready sampling of its contents and would meet AEC regulatory requirements. Details of this programme are presented in another paper at this Symposium [7].

A study to define mixing parameters for restricted geometry process equipment (5 in. I.D. columns)

This work involves development of a basic understanding of the mechanisms of mixing of chemical solutions in the unusual tankage geometry made necessary by critical mass considerations. Solutions generated in a nuclear fuel processing plant vary both in composition and concentration with differing feed materials and with control of process operations. These solutions, which are stored in a safe-geometry-cylindrical tankage, cannot be satisfactorily sampled for fuel inventory until the tank contents are properly mixed. This programme was initiated to study the various parameters associated with mixing and sampling such solutions to determine optimum operating methods. The technology developed will provide criteria that can be used to determine more precise fuel inventories.

A comprehensive preliminary investigation has been made of the preliminary work in mixing and sampling of liquids contained in restricted geometry tankage. This investigation included a survey of published and unpublished literature, and consultation with personnel concerned with the inventory of nuclear fuel solutions at AEC plants and with recognized authorities on fluid mixing.

As a result of the preliminary investigation, an experimental programme was initiated to provide fundamental data of general interest. The experimental programme considered single and two in-parallel horizontal tanks of varying length-to-diameter ratios mixed by pump recirculation, a single vertical tank of varying height-to-diameter ratio mixed by air sparging, and a single horizontal tank mixed by a series-parallel flow pattern using pump recirculation. The accumulated data have been resolved into a defined mixing time as a function of flow rate for the systems considered.

A generalization which provides a means for predicting the mixing characteristics of any single tank system was developed from the single-pump, recirculated-tank mixing data. This generalization utilizes a combination of mixing models, as proposed in the literature, and the experimental data developed for mixing in straight pipelines and agitated tanks.

A continuation of both theoretical and experimental analyses is needed to extend mixing and sampling technology to encompass the complex multi-tank systems found in the nuclear fuel processing plants. Proposed work includes experimental and theoretical mixing studies on multi-parallel tank systems mixed by pump recirculation, and mixing and sampling studies for vertical air sparge tanks and for a horizontal pump-recirculated tank containing a solid-liquid mixture.

This study is still in progress and its results will be published upon completion.
An evaluation of the calculation of nuclear loss and plutonium production in nuclear reactor operations

A comprehensive evaluation of calculation procedures for power reactor fuel burn-up has been undertaken. Specifically, the study is designed to

(a) determine the capabilities and limitations of methods available at present for calculating the uranium-235 and plutonium content of nuclear power reactor fuel, not only on a total core basis, but also on an individual fuel assembly basis where zone or batch discharge schemes are envisioned; and

(b) develop means for independently testing the validity of such calculations (probably not to the same degree of precision), making maximum use of available physical data such as control rod positions or in-core instrumentation recordings.

The study will provide essential information concerning the agreement that may be expected between reactor calculations and dissolver measurements at chemical processors, and is reported in detail at this Symposium [8].

A study to recommend the most useful statistical techniques to evaluate material control indicators

The ultimate objective of the study (conducted by the Stanford Research Institute) is to develop an improved set of statistical procedures to monitor transfers and inventories of nuclear materials. The intermediate objectives are to

(a) assess the statistical methods now in use for evaluating the significance of the differences between a book inventory and a physical inventory, and those differences between the shipper's measurement and that of the receiver that occur on transfers of material;

(b) develop better methods, where possible, for evaluating these material control indices;

(c) develop a measure of the value of the improved measurements, data collection techniques or statistical analyses against the cost of obtaining such improvements;

(d) adapt or modify known techniques for optimum management of commercial inventories to the control of nuclear materials insofar as such techniques may be applicable; and

(e) prepare a handbook of standard methods of statistical analysis and inventory management, inclusive, of course, of any developments resulting from the study, that will be useful to the nuclear materials industries.

A programme to evaluate domestic and foreign analytical laboratories for use as referees or umpires in measurement disputes between the USAEC and foreign or domestic nuclear industries

Mainly as a result of domestic and foreign nuclear power programmes, the USAEC is entering into an increasing number of transactions with domestic private industry and foreign governments involving monetary payments for the sale, lease or purchase of plutonium and uranium. These payments
are based on measurements of both the quantity and quality of the materials involved.

Since differences in measurements are likely to arise between shippers and receivers, differences in monetary value also will be involved and, in some cases, could be of significant proportions. Thus, the USAEC felt the need for identifying and qualifying independent laboratories that could act as umpires in settling possible measurement disputes.

Although measurement differences requiring umpires have been relatively few so far, both the expanding domestic and foreign nuclear power programs and the advent within the United States of private ownership of nuclear fuel have emphasized the need for finding independent umpire laboratories.

Bibliography of nuclear materials management

In the United States, both within the USAEC and in private industry, nuclear materials management has become widely recognized as a specialized field. A survey of the literature and presentations made at several national nuclear materials management meetings revealed approximately 850 pertinent references in nuclear materials management. These references have been included in a bibliography of nuclear materials management for publication. Also, the specific reference category "nuclear materials management" is now listed in Nuclear Science Abstracts.

While the technical support activities discussed in this presentation were sponsored by the US Atomic Energy Commission, it should be recognized that the nuclear industry played a most significant role in their development. In addition to these major efforts, we are all aware of the interest and recognition that the nuclear field is devoting to the many facets of nuclear materials management. The growth of the industry, and the trend from government ownership to the private sector, with the increased emphasis on dollar costs, will result in even greater interest and efforts.

SUMMARY

Technical support activities are an integral part of any successful nuclear materials management programme. However, nuclear materials management is not unique to any one country, but rather is common to the entire nuclear industry. It is this characteristic that has engendered the interest and support of each of us in the United States, and may equally interest the International Atomic Energy Agency.

The magnitude of certain research and development efforts as, for example, the development of non-destructive assay techniques for determining the special nuclear material content of unirradiated and irradiated nuclear fuels, demands an effort commensurate with the magnitude of the dollar transactions now occurring. This need, and that of determining the nuclear content of heterogeneous scrap materials, represent the major gaps that now remain in the measurements picture within the nuclear fuel cycle.
Within the United States, the AEC in co-operation with the National Bureau of Standards has played a most significant part in the development of standard reference materials of interest to the nuclear industry. Other countries may have similar and possibly parallel programmes in hand. In addition, national organizations such as the Institute of Nuclear Materials Management, the American Nuclear Society and the American Standards Association, dedicated to the advancement of the nuclear industry, are rapidly developing and endorsing nuclear standards such as colour codes, scrap uranium classification guides, procedures and practices for transporting fissile materials, and specifications for nuclear materials management.

The need for co-ordinating such activities of the many countries represented at this Symposium and seeking international acceptance of nuclear standards and reference materials is a challenge that must be met. The international aspects and interests in these particular efforts may suggest that the International Atomic Energy Agency should play a leading role.

REFERENCES

[2] SHIELDS, W. R., "The evolution of the accuracy of isotopic analysis by thermal ionization from 2% to 0.02%", these Proceedings.
ADMINISTRATIVE CO-ORDINATION OF FISSILE MATERIAL MANAGEMENT AND ACCOUNTING IN THE U.K.A.E.A.

St. C. C. HOOD
UNITED KINGDOM ATOMIC ENERGY AUTHORITY, LONDON, UNITED KINGDOM

Abstract — Résumé — Аннотация — Resumen

ADMINISTRATIVE CO-ORDINATION OF FISSILE MATERIAL MANAGEMENT AND ACCOUNTING IN THE UNITED KINGDOM ATOMIC ENERGY AUTHORITY. The Authority are engaged as suppliers in fissile material production, distribution, recycle and reprocessing. As consumers, the Authority require fissile material for power reactors, a variety of prototypes, MTRs, zero-energy facilities and fuel development projects; and for other experimental and research purposes in laboratory quantities. Executive responsibility for these activities lies with the four Groups through which the Authority discharge these functions.

It has been found useful to keep these activities under review in specialized inter-Group Committees, with a common secretariat. These Committees:

(a) study all projects or proposals for work involving significant quantities of fissile material (plutonium and enriched uranium, other than natural U or U depleted in $^{235}$U) in the light of expected supplies over a number of years from all sources, including new production, scrap recovery and imports; and all uses including burn-up, losses and exports;

(b) recommend the optimum allocation of specific amounts for approved purposes in relation to other calls upon available supplies, and having regard to the economic issues involved;

(c) record and progress all approved allocations, and examine the nature, amount and purpose of all existing stockholdings in relation to current policies and objectives;

(d) record and study all losses of fissile material during fabrication or other processing and the measures taken to reduce them;

(e) assist in developing procedures and incentives to ensure that material is used economically and returned promptly.

Each Group has considerable autonomy in its day-to-day use of fissile material. The administrative machinery described above provides a means by which the Authority's scientists, engineers, accountants and administrators concerned with fissile material problems can operate collectively in a common frame of reference with a minimum of paperwork.

The paper is illustrated with a simplified flowsheet of the main flows of fissile material within the Authority for civil purposes during the year ending 31.3.65.

COORDINATION ADMINISTRATIVE DE LA GESTION ET DE LA COMPTABILITE DES MATIERES FISSILES DANS LES ETABLISSEMENTS DE L'AUTORITE DE L'ENERGIE ATOMIQUE DU ROYAUME-UNI. En tant que fournisseur, l'Autorité s'occupe de la production, de la distribution, du recyclage et du traitement chimique des matières fissiles. En tant que consommateur, elle a besoin de matières fissiles pour ses réacteurs de puissance, pour une série de prototypes, pour ses réacteurs d'essai des matériaux, pour ses réacteurs de puissance zéro et pour ses travaux de mise au point de combustibles; il lui en faut également, en petites quantités, pour ses expériences et ses recherches en laboratoire. L'Autorité confie à quatre groupes la direction de ces activités.

Elle a jugé utile de faire suivre régulièrement ces activités par des comités spécialisés intergroupes ayant un secrétariat commun. Ces comités ont les attributions suivantes:

a) Étudier tous travaux ou projets de travaux qui nécessitent l'utilisation de quantités importantes de matières fissiles (plutonium et uranium enrichi à l'exclusion de l'uranium naturel ou de l'uranium appauvi en uranium-235), en tenant compte de tous les approvisionnements prévus pendant un certain nombre d'années en provenance de toutes sources, y compris la production sur place, la récupération des déchets et les importations; en tenant compte aussi de toutes les utilisations, y compris la combustion, les pertes et les exportations:

597
b) recommander, compte tenu des autres besoins en matières disponibles et des impératifs économiques, la meilleure attribution possible de quantités déterminées destinées à des fins agréées;
c) enregistrer toutes les attributions approuvées de matières et suivre leur utilisation, et étudier la nature, les quantités et la destination de tous les stocks existants en tenant compte des directives en vigueur et des objectifs pursuivis;
d) enregistrer et étudier toutes les pertes de matières fissiles en cours de fabrication ou de traitement ainsi que les mesures prises pour les réduire;
e) aider à mettre au point des procédures et des méthodes permettant d’assurer que les matières sont utilisées de façon économique et renvoyées rapidement,

Chaque groupe dispose d’une assez large autonomie en ce qui concerne l’utilisation courante des matières fissiles. Le système administratif précédemment décrit permet aux savants et ingénieurs de l’Autorité, ainsi qu’aux comptables et administrateurs chargés de résoudre les problèmes relatifs aux matières fissiles, de travailler collectivement dans un cadre commun en ayant à établir un minimum de documents.

Le mémoire est illustré par un diagramme simplifié indiquant les principaux circuits d’opérations concernant des matières fissiles destinées à des fins civiles et traitées dans les établissements de l’Autorité au cours de l’année se terminant le 31 mars 1965,
Se ha encontrado conveniente someter todas las actividades mencionadas a la fiscalización de comités mixtos de estos Grupos, con una secretaría común. Dichos comités:

a) estudian todos los proyectos y propuestas que requieren el empleo de cantidades importantes de materiales fisionables (plutonio, uranio enriquecido, etc., con exclusión del uranio natural y del uranio empobrecido en \( ^{235}U \) teniendo en cuenta los suministros que durante un período de varios años se espera recibir de todas las fuentes — producción, recuperación de desechos, importación — y todas las cantidades empleadas, incluidos los materiales agotados, las pérdidas y las exportaciones;

b) recomiendan la mejor manera de asignar cantidades determinadas de materiales para fines aprobados, teniendo en cuenta el conjunto de las necesidades, el volumen de las existencias y los aspectos económicos de la operación;

c) registran y controlan todas las asignaciones aprobadas; en relación con los criterios y los objetivos establecidos, verifican la naturaleza de los materiales que se están utilizando, sus cantidades, y los fines a que se les destina;

d) registran y estudian todas las pérdidas de material fisionable que se producen durante los procesos de elaboración y tratamiento, así como las medidas adoptadas para evitar dichas pérdidas;

e) contribuyen a establecer procedimientos y a crear incentivos para lograr que los materiales se empleen con economía y se devuelvan con rapidez.

Cada Grupo goza de considerable autonomía por lo que respecta al empleo cotidiano de los materiales fisionables. Gracias a la estructura descrita, el personal técnico, los contables y los administradores que tienen a su cargo los problemas inherentes a los materiales fisionables pueden actuar colectivamente dentro de un sistema de referencias común y con un mínimo de dilaciones de carácter burocrático.

Ilustra la memoria un diagrama de las principales corrientes de materiales fisionables que con destino a fines civiles pasaron por las instalaciones y servicios de la Comisión durante el año que terminó el 31 de marzo de 1965.

1. INTRODUCTION

The United Kingdom Atomic Energy Authority produce, process, use and trade in fissile material in large amounts and for a very wide range of purposes. During the eleven years of their existence, a centralized allocation and accounting system has been evolved. The principal features of the system are described in this paper.

The Authority's functions are discharged by specialized Groups. Each Group is provided continuously with information about fissile material supplies, stocks and the requirements of other Groups. Representatives of all Groups are enabled to bring their expert knowledge and collective judgement to bear on all aspects of fissile material allocation, management and accounting. The centralized system provides a forum for their discussions and the rules and procedures by which control is exercised.

2. THE DEVELOPING USES OF FISSILE MATERIAL WITHIN THE AUTHORITY

When the U.K.A.E.A. were formed in 1954, they took over responsibility for a number of activities that were already going concerns. The Windscale piles, the diffusion plant at Capenhurst, and the associated reprocessing and finishing plants were already operating as producers of fissile material and the establishments at Harwell and Aldermaston were using this material for a variety of purposes (see Fig. 1).
FIG. 1. 1954: Primary flow of uranium and plutonium within the UK

NOTES:
solid lines show movement towards users,
broken lines show movement of residues towards recovery facilities.
It was decided from the outset that the Authority's responsibility for the development of atomic energy would be discharged most efficiently through an organizational structure which provided for functional devolution. Accordingly, separate Groups were to be maintained to deal on the one hand with the production and reprocessing of fuel and fissile material and the associated development work, and on the other with the conduct of research. Although there was to be close contact at all staff levels between these Groups, their day to day activities were to be controlled from within the Groups themselves.

This concept has never changed. During the last ten years, as the scale of the Authority's activities has widened, the number of Groups has increased from three to five, but each Group retains a large measure of autonomy in discharging its functions. Apart from the Engineering Group, who provide a common construction and engineering service, all Groups produce or use fissile material (see Fig.2).

2.1. The role of a centralized system

Although the principle of functional devolution was given full play, the Authority naturally retain ultimate responsibility for the Group's activities and also reserve a number of activities to be discharged by their London Office.

The central system of fissile material allocation and accounting, covering plutonium and enriched uranium, is one such activity. Demands upon fissile material have consistently outrun supply, and the task of adjudicating between the competing demands is essentially a matter for central decision. This task requires and involves common high standards for reporting stocks and usage within the Groups, and the central collection and dissemination of the information.

3. THE ALLOCATION SYSTEM

The Authority's procedure for allocating fissile material for civil use is administered by their Advisory Committee on Special Materials (A.C.S.M.) of which the Chairman is the Deputy Chairman of the Authority. This Committee reviews the supply capabilities of the Authority's production and reprocessing facilities, the claims upon them for defence purposes, the size and composition of existing stocks, and the extent to which supplies could be changed in character or quantity in the short and long terms. Against this background, the Committee consider in detail the case advanced by the Group concerned with each individual project for which fissile material is required in the forthcoming year, and more broadly the expected requirements over the ensuing five years. The sponsoring Group must demonstrate that the proposed project use will directly further the aims of the relevant sector of the overall R. and D. programme, that the amounts of fissile material requested are a practicable minimum and to the simplest possible specification, and that existing methods for recovery of fabrication residues.
FIG. 2. Primary flow of uranium and plutonium within the UK in 1964.
or irradiated fuel can be employed or the write-off of irrecoverable material borne on the cost of the project.

3.1. Annual review of requirements

The A.C.S.M. comprises senior representatives of the four Groups and the senior financial and economics officers of the Authority. Thus each Group is in a position to examine critically the claims of the other Groups for fissile material, and the Committee as a whole can help both to administer and to influence Authority research and development policy. Until the A.C.S.M. has recommended an allocation for a project and this has been approved by or on behalf of the Authority, no fissile material can be moved from stock or drawn from new production for that project. The A.C.S.M. reports to the Authority annually, recommending actual allocations for the next financial year and giving a comprehensive review of the prospective balance of the various materials over a six-year period.

Any new demands arising during the course of a year are considered by a standing Sub-Committee of the Advisory Committee, on which members of the main Committee are represented.

3.2. Fulfilment of allocations

When the allocations recommended in the annual reports have been approved by the Authority, their fulfilment is supervised by the Special Materials Accounting Committee (S.M.A.C.).

This Committee has the central responsibility for fissile material accounting control for all fissile material in the Authority's custody. In the civil field, the main objectives of the Committee are to verify the disposition of all the material, and to ensure that it is being used only for approved purposes and in the most economical way.

The Committee is under the chairmanship of the Authority's Principal Finance and Supplies Officer. The Groups are represented both by their accountants and by members of their technical staffs. The technical members help the Committee to ensure that material is being kept on the move towards the projects for which it is destined, that it is being used effectively, and that waste is being minimized; the accountants ensure that financial responsibility for stocks, usage and waste is properly apportioned between Groups and, within Groups, between projects.

The data collected for the Committee at quarterly- or six-monthly intervals is project-centred. The allocations made on the recommendations of the Advisory Committee are also directed to specific projects — for example, the development of fuel for the steam-generating heavy water reactor. The project management staff, who will have originated the request for the allocation, are responsible for ordering the material from the Production Group. (The role of the Production Group is explained in more detail in section 6.)
3.3. Progressing the allocations

Once material allocated to a given project has been drawn from stock or new production by the Production Group, its progress is kept under review by S.M.A.C. until it finally reaches the project management. The material may undergo fabrication and blending processes in some third Group, and at any stage some of the material may be lost in effluent or irrecoverable waste, or returned as fabrication scrap for chemical reprocessing. The original allocation will have been expressed in gross and net terms, the difference being the amount allowed for losses and fabrication allowance. The project management are entitled to receive the net allocation, although in some cases they may agree to accept less if the fabrication route proves to be unexpectedly wasteful, and in other cases they may ask to receive more if fabrication proves to be economical. Any such increase in the net allocation would have to be justified on its merits and approved through the executive machinery of the A.C.S.M.

Most allocations are delivered well within a single year, although a few may spread over two or more years if large quantities are required or if unusual or complex fabrication methods, sometimes drawing in two or more Groups, are involved. The Accounting Committee follow all such material stage by stage until it reaches its final destination with the project concerned.

The allocated material en route to projects at any given time is only part of the material that will be moving between the Groups, and only a small part of the total holdings of the Groups. Its particular importance is that it constitutes the current demand governing the deployment of production resources and that it is especially vulnerable to delay and fabrication or processing losses until it is handed over to the project management to whom it has been allocated.

Table I shows an extract from the summary form into which the returns from each Group are consolidated for consideration by the S.M.A.C. It can be seen that each reported receipt to one Group – marked R – is matched by a reported delivery – marked D – from some other Group. The entries marked S show the source from which the material originated. Normally the source is the Production Group, but occasionally, with the agreement of the Production Group, material is provided by some other Group which happens to have suitable material in stock and surplus to requirements. The full summary, of which Table I is an extract, covers all material allocated in the current year; there is a similar summary for material that was allocated in previous years but which is still en route to the project management.

4. REVIEW OF STOCKS AND USAGE

Once the material has reached the project to which it has been allocated, the problems of central control change from those of minimizing loss and delay to those of ensuring that the material is actually being put to good use. It is of course a function of efficient plant management, taken very seriously throughout the Authority, to account carefully for fissile material both for reasons of economy and because of possible criticality hazards, particularly
### Table I

**PROGRESS OF 1964/65 ALLOCATIONS**

**POSITION BY 19.9.1964**

<table>
<thead>
<tr>
<th>Ref.</th>
<th>Brief Title Of Project</th>
<th>Qty.</th>
<th>Grade or Enrichm'nt</th>
<th>Qty.</th>
<th>Grade or Enrichm'nt</th>
<th>Production Group</th>
<th>Research Group</th>
<th>Reactor Group</th>
<th>Weapons Group</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td><strong>PLUTONIUM</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Reactor Group</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>A</td>
<td>Fast Reactor Fuel Development</td>
<td>28</td>
<td>A</td>
<td>20</td>
<td>A</td>
<td>S 6.1</td>
<td>R 1.3</td>
<td>R 6.1</td>
<td>D 1.3</td>
</tr>
<tr>
<td></td>
<td>Research Group</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>B</td>
<td>Metallurgy Research</td>
<td>7</td>
<td>A</td>
<td>6</td>
<td>A</td>
<td>S 1.2</td>
<td>R 1.2</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Quantities in kg Plutonium

**Notes:**

1. Figures are illustrative.
2. A similar statement is prepared for U235 expressed in kg U235 (93%) in plants not designed as "ever-safe". Judicious central control procedures can however reinforce local management in both these respects.

The primary data for central accounting is again project-centred. All Groups are required to describe all the fissile material that they hold in any form or for any purpose at the end of each half year.

### 4.1. Project stocks and surplus stocks

Since civil material can be used only for purposes that are approved by the Advisory Committee, all these holdings are divided into three classes:

(a) **Project stocks**, including all the material which is actually in use in projects or is in transit to projects.

(b) **Surplus stock**, which in turn is divided into two categories:

Category 1. material which is in a usable form (e.g. unirradiated UO₂) but which has never yet been allocated or has been returned to stock after use in a project. For this and other purposes new material in process is disregarded until it passes the appropriate product accounting point in the production facility.
Category 2. material which has been returned from a project and is not in a usable form; and which is being or must be reprocessed or refabricated before it can be allocated for use in a project.

Table II is an extract, using illustrative figures, from the summary form into which all the Group stock reports are consolidated. The full summary shows on single sheets of paper for plutonium and $^{235}\text{U}$ respectively the holding of fissile material in use in every project, its approximate quality, and the amount of material surplus to requirements in Categories 1 and 2 at each of the four Groups.

4.2. Certification of stocks and usage

Both of the schedules illustrated in Tables I and II, that showing the progress of allocations and that showing the holdings of each Group, are based on returns personally certified by the Group Director or Managing Director or his Deputy. The form of the certificate commits the Director to the assertion that all material declared to be in Project Stocks is actually in use in or en route to a project, and that all the Surplus material within the custody of his Group has been accurately declared. This declaration directly associates the Directors with the centralized accounting system and assures their staff of the importance attached to scrupulous accuracy in their returns.

5. LOSSES AND INTER-GROUP FLOWS

The third and last class of standard Accounting Committee documents, shown as Table III below, fills in the areas not covered by the two detailed schedules and provides a matrix of all movements of material between Group and Group, and between Groups and external customers or collaborators. These Statements of Account analyse the losses of fissile material within each Group.

The certified schedules provide a basis for the future work of the Advisory Committee in recommending new allocations, and a means of checking on the progress of existing allocations. The Statements of Account, on the other hand, are used for a number of financial purposes. Their accuracy is safeguarded by a series of checks and by comparison with the certified Schedules:

(a) The total stock held by any Group must correspond with that declared for the total of all the Group's Projects and Surplus Stocks in the certified schedules mentioned above;

(b) The imports and exports of the whole matrix must be equal – i.e. shipper-receiver differences must be reconciled;

(c) The reported losses must be consistent with the implied shipper-receiver difference (test (2)) and with the net changes in total stock (test (1));

(d) All losses must be explained and accounted for.
### TABLE II

**AUTHORITY STOCKS OF PLUTONIUM AS AT 19.9.1964 IN kg (Pu)**

<table>
<thead>
<tr>
<th>Ref.</th>
<th>Brief Title of Project</th>
<th>GRADE A</th>
<th></th>
<th>GRADE B</th>
<th></th>
<th>Totals</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>RG</td>
<td>RG</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>A</td>
<td>Fast Reactor</td>
<td>29.38</td>
<td>9.23</td>
<td>11.71</td>
<td>50.32</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Fuel Development</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>B</td>
<td>Metallurgy</td>
<td>13.47</td>
<td>1.04</td>
<td>14.51</td>
<td>3.42</td>
<td>17.93</td>
</tr>
<tr>
<td></td>
<td>Research</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>PROJECT STOCKS</td>
<td>13.47</td>
<td>30.42</td>
<td>9.23</td>
<td>11.71</td>
<td>64.83</td>
</tr>
<tr>
<td></td>
<td></td>
<td>3.42</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>13.42</td>
<td></td>
<td>68.25</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Category</th>
<th>GRADE A</th>
<th></th>
<th>GRADE B</th>
<th></th>
<th>Totals</th>
</tr>
</thead>
<tbody>
<tr>
<td>Category I</td>
<td>10</td>
<td>20</td>
<td>8</td>
<td>8</td>
<td>28</td>
</tr>
<tr>
<td>Category II</td>
<td>12</td>
<td>15</td>
<td>20</td>
<td>1</td>
<td>2</td>
</tr>
</tbody>
</table>

**TOTAL** | 25.47 | 45.42 | 34.23 | 31.71 | 136.83 | 3.42 | 13.42 | 150.25 |

**Notes:**

i. Figures are illustrative.

ii. A similar statement is prepared for U235 expressed in kg U235 (93%)=.

iii. The grades shown above distinguish between material of different Pu 240 content.

**Key:-**

<table>
<thead>
<tr>
<th>RG</th>
<th>WG</th>
<th>PG</th>
<th>TRG</th>
<th>Totals</th>
</tr>
</thead>
<tbody>
<tr>
<td>RG</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>WG</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>PG</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>TRG</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

### 5.1. Accounting for losses

The most troublesome areas are of course those in which the data relate not to sitting stocks or to movements of new fuel, but to estimates of losses between successive accounting points either within or between Groups. The central accounting function in these areas is to isolate the significant losses and to encourage Groups to resolve shipper-receiver discrepancies.

### 5.2. Normal plant losses

As regards the isolation of losses, the Committee have approved the calculation of "normal plant losses" for each fissile material processing facility with which experience is sufficient to enable plant performance in this respect to be determined.
### TABLE III

**DETAILED STATEMENT OF ACCOUNT**

**CIVIL STOCKS 1964/65 OF PLUTONIUM (kg)**

<table>
<thead>
<tr>
<th>Stock Type</th>
<th>Opening Stock</th>
<th>Movements</th>
<th>Closing Stock</th>
<th>Movements</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>PRODUCTION GROUP STOCK</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>IN: New Material Feed</td>
<td>25.83</td>
<td>25.00</td>
<td>42.22</td>
<td></td>
</tr>
<tr>
<td>Other Groups</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>External</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>OUT: Other Groups</td>
<td></td>
<td>7.30</td>
<td>32.71</td>
<td></td>
</tr>
<tr>
<td>External</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Losses (see below)</td>
<td></td>
<td>0.30</td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>REACTOR GROUP STOCK</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>IN: Other Groups</td>
<td></td>
<td>1.30</td>
<td>28.89</td>
<td></td>
</tr>
<tr>
<td>External</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>OUT: Other Groups</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>External</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Losses (see below)</td>
<td></td>
<td>0.10</td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>RESEARCH GROUP STOCK</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>IN: Other Groups</td>
<td></td>
<td>1.20</td>
<td>26.99</td>
<td></td>
</tr>
<tr>
<td>External</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>OUT: Other Groups</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>External</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Losses (see below)</td>
<td></td>
<td>0.06</td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>WEAPONS GROUP STOCK</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>IN: Other Groups</td>
<td></td>
<td>6.10</td>
<td>45.42</td>
<td></td>
</tr>
<tr>
<td>External</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>OUT: Other Groups</td>
<td></td>
<td>1.30</td>
<td>45.42</td>
<td></td>
</tr>
<tr>
<td>External</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Losses (see below)</td>
<td></td>
<td>0.12</td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>U.K.A.E.A. TOTAL STOCK</strong></td>
<td>125.83</td>
<td>150.25</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

#### STOCKS

- Project: 60.95
- Surplus Category I: 9.95
- Surplus Category II: 54.92
- Total: 125.83

#### LOSSES

<table>
<thead>
<tr>
<th>Year</th>
<th>Downgrading</th>
<th>Decay or Burnup</th>
<th>Normal Plant Losses</th>
<th>Out of Balance</th>
<th>TOTAL</th>
</tr>
</thead>
<tbody>
<tr>
<td>1st half</td>
<td>P.G.</td>
<td>0.20</td>
<td>0.10</td>
<td>0.30</td>
<td>0.58</td>
</tr>
<tr>
<td>2nd half</td>
<td>T.R.G.</td>
<td>0.10</td>
<td>0.05</td>
<td>0.15</td>
<td>0.30</td>
</tr>
</tbody>
</table>

**Note:**

Figures are illustrative.
A similar statement is prepared for U235 expressed in kg U235 (93% w).
Proposals for the establishment of "normal plant losses" are discussed in the Committee, where the technical representatives of all Groups can comment upon the validity of the proposals in relation to the characteristics of the plant concerned, the nature of the throughput, and the length of the experience upon which the proposals are founded. In most cases, the "normal plant loss" can be distributed between specific operations in which routine or ad hoc methods are available for identifying component losses which are to be accepted as economically irrecoverable. In a complex cycle involving a large number of operations the internal imbalances between processes may be small but numerous and consistent, and several years may elapse before an overall normal plant loss can be determined and agreed. Throughout this period, the S.M.A.C. will receive full reports on the steps which are being taken to improve understanding of the loss mechanisms. The informed interest of the Committee in these studies encourages the establishments in their efforts. Once a normal plant loss is agreed, it is shown as a separate element in the total of reported losses suffered during each six-monthly accounting period.

5.3. "Unaccountable" losses

Each Group in turn is then required to satisfy the whole Committee that all the remaining "unaccountable" losses within the Group's responsibility are being investigated or should be written off as insignificant. Because fissile material which is lost must be paid for in full, whereas material in use as Project Stock is subject only to interest (hire charges) the Groups have a financial as well as a professional incentive to minimize losses.

It is, of course, theoretically possible for Project Managers incorrectly to estimate the amount of losses from Project Stocks, or the fissile content of material transferred to Surplus. In both cases the amount paid for the lost material would then be incorrect. However, most of the material taken into Surplus stock is promptly reprocessed and any necessary adjustment effected.

5.4. Shipper-receiver discrepancies

As regards shipper-receiver discrepancies, S.M.A.C. try to secure that, as far as possible, the accuracy of the data provided to them is guaranteed by some system of double-entry. Thus all movements between Groups are documented by both Groups. Explanations of discrepancies between shipper and receiver are usually obtained by discussion between the technical staff directly involved, and the financial consequences are agreed between their respective accountants. Where a serious discrepancy persists, the Committee may commission a special study. Sometimes the answer is simple - for example, a sender or shipper who includes permissible impurities, and a receiver who reckons only by fissile material atoms. Where heterogeneous material is transferred, the taking of representative samples may be difficult or impossible, and discrepancies must be resolved retrospectively by apportioning losses between the necessary processes and the original uses.
Sometimes independent check analyses or specially instrumented runs are conducted. A few years ago there was a persistent shipper-receiver discrepancy between AWRE and Windscale as to the amount of $^{235}\text{U}$ in certain residues. The residues were dissolved at Windscale and a "tracer" method was used to determine the volume of the solution by adding a known quantity of caesium-137. A sample could then be taken, the volume determined by analysis of caesium-137 and the $^{235}\text{U}$ concentration also determined by analysis. Thus the total amount of $^{235}\text{U}$ was calculated. This type of approach allowed AWRE staff to attend the dissolution and take samples for analysis at AWRE. In practice the AWRE staff were entirely satisfied with the Windscale results and did not exercise their right to analyse. This project resolved the difficulties between the two Groups. Elliott refers to this tracer method of determining volumes in his paper SM-67/53.

6. THE AUTHORITY STOCKPILE CONCEPT

The Authority Stockpile of fissile material is controlled by the Production Group as the agents of the Authority. It comprises all the fissile material which is in the books of the Authority, other than that which is in the Project Stocks for which individual Groups are responsible, and is therefore the sum at any time of the Category 1 and Category 2 Surplus stocks described at section 4.1 above. Most of the Surplus Stockpile is in the physical custody of the Production Group, but some surplus material is retained by the other Groups until the Production Group call it forward for recovery or direct it to a project in furtherance of an approved allocation. The Production Group keep a running record of the composition of the Surplus Stockpile, and are therefore in a position to supply material against allocations in the most economical way both as regards its physical form and as regards blending to the exact requirements of the customer.

6.1. Stock forecasting

This centralization of information about surplus stocks also enables the Production Group and, on occasions, the Reactor Group in respect of residues that can be called forward to their specialised facilities at Dounreay, to plan the timing of residue recovery campaigns so as to phase in residue recovery with new production where these two activities converge, e.g. in the plutonium finishing plant. Over a short run of, say a year, the planning extends to detailed scheduling. For the longer run of six years studied by the Advisory Committee, a stock forecast analysis is prepared annually as a guide to the prospective availability of re-usable and new material, after taking account of expected demands and the return of residues and re-usable material from Projects.

6.2. Financial incentives

Financial controls have been devised to ensure that the Stockpile gathers in all possible surplus material. All fissile material owned by the Authority
is revalued annually at the lower of cost or realizable value (or replacement cost, as appropriate). Interest is charged to the Stockpile on the total value of the material held. The Stockpile in turn charges interest to Projects in the form of a hire charge for the use of material. The hire charge provides for recovery of interest both on the material actually held by the projects, and on a nominal back-up stock. It ceases when the material is declared as surplus to Project needs. There is thus an incentive for each Project Manager to relinquish promptly fissile material that he no longer needs or which requires recovery.

7. LESSONS WE ARE LEARNING

Any system designed to control a multiplicity of activities like fissile material usage must operate in the area between rigid uniformity and excessive flexibility. If there is no room for exceptions and improvisation, the system is at best a bureaucratic paperchase justly resented by its victims. If on the other hand it concedes special treatment to every difficult case, then every case becomes difficult and the system is valueless as a control and even as a recording apparatus.

The system described in this paper is constantly changing in small ways because it is based on government by consent. This means that as the situation changes, pressures from technical, financial or administrative staff can be responded to if the system as a whole will clearly benefit. This is an advantage in many respects, but it can lead to the complacent conclusion that because the system operates without much friction it is therefore justified and useful per se.

Fissile material must always be safeguarded for health and safety reasons, and as required for political purposes. Beyond this point, control over allocation and use may be useful, if it is specialized to the needs of the organization concerned, but no control is justified unless on balance it benefits the organization. Obsessive care to reduce wasteful use of scarce material can lock up equally scarce skilled manpower and facilities in various ways. The emphasis should always be upon optimizing the use of resources. No one management discipline is suitable for this purpose, and the right blend is one that each organization must seek for itself.

DISCUSSION

S. KOPS: Do you maintain your financial and quantity records together or separately?

St. C. C. HOOD: The quantity records are an important source of information for the financial accounts, but largely as a cross-check on totals. The detailed financial charges for hiring, losses, etc. are assessed by the Production Group. The Group operates the Authority stockpile in accordance with approved allocations, and co-ordinates the associated financial and physical actions.

R.G. CARDWELL: You state in your paper that "All losses must be explained and accounted for". Does this mean that you do not recognize unaccounted-for differences of a low economic value?
St. C.C. HOOD: We do recognize "unaccountable" losses, including such losses in excess of "normal plant loss". The project holding the material in question is charged the full economic value of the amount lost.

R.G. CARDWELL: Do you experience any process gains in the system under the A.C.S.M. control, and do you make any attempt to balance these gains against your process losses to reach a total variation?

St. C.C. HOOD: Yes, when continuous processing is involved and campaigns do not end on the accounting dates.

J.J. DOWNING: Does the United Kingdom have a central organization which carries out physical inventories at the various installations?

St. C.C. HOOD: As I said in my oral presentation, the U.K.A.E.A. does not operate an independent auditing system for the physical inspection of inventories. The Authority is organized on a "federal" basis, and the "federated" Groups are represented at a high level on the committees which regulate the allocation and stocks of special materials and review explanations of out-of-balance losses. In our experience, this ensures a correspondingly high accuracy in the preparation of material accounts and records of movements between the Groups.

D.E. GEORGE: There is in fact a significant difference in approach to the problem of inspection between the United States of America and the United Kingdom, as is clear from comparing this paper with mine on the same subject*. The United Kingdom does not employ independent inspection on the plant site, whereas we regard it as the basis of our nuclear materials management programme.

F. RONTEIX: Can Mr Hood indicate orders of magnitude for some of the normal plant losses, e.g. in the case of uranium refining operations and fuel-element fabrication for power reactors at Springfield?

St. C.C. HOOD: The Committees described in the paper deal only with enriched uranium, whereas existing power reactors in the United Kingdom use natural uranium fuel. I do not have available details of normal plant losses for other applications.

---

* These proceedings (SM-67/59).
THE UNITED STATES ATOMIC ENERGY COMMISSION PROGRAMME OF NUCLEAR MATERIALS MANAGEMENT

D. E. GEORGE
UNITED STATES ATOMIC ENERGY COMMISSION, WASHINGTON, D.C., UNITED STATES OF AMERICA

Abstract — Résumé — Аннотация — Resumen

THE UNITED STATES ATOMIC ENERGY COMMISSION PROGRAMME OF NUCLEAR MATERIALS MANAGEMENT. Nuclear materials management as conceived by the US Atomic Energy Commission is defined and its development traced from 1943 to the present time. The general programme is outlined and its principal features discussed. Emphasis is placed on administration of those portions of the USAEC programme which pertain to the development and maintenance of quantity data, the establishment of internal controls and the evaluation of performance.

The current techniques whereby transfer data are recorded and processed within the USAEC are discussed in some detail, as are the techniques for presentation of material balance reports which periodically summarize the transactions and the results of operations. The techniques used by the USAEC to ascertain the effectiveness of the nuclear materials management programmes of its operating contractors are also discussed. In addition to material being held by contractors which operate USAEC-owned plants and laboratories, there are large quantities of special nuclear material held under a wide variety of financial and administrative arrangements, e.g., under lease, private ownership, under contract with the USAEC where the user is financially liable for losses, as well as those where the risk of losses is taken by the USAEC. This divergence of circumstances causes significant variation in the emphasis and approach used by the AEC and this variation is noted. A brief summary of unresolved problem areas concludes the presentation.

LE PROGRAMME DE GESTION DES MATIERES NUCLEAIRES APPLIQUE PAR L'USAEC. L'auteur définit la gestion des matières nucléaires telle qu'elle est conçue par l'USAEC et décrit son évolution depuis 1943 jusqu'à nos jours. Il expose le programme général et discute ses principales caractéristiques. Le mémoire souligne les aspects administratifs des parties du programme qui ont trait à la préparation et la tenue à jour des données relatives aux quantités, à l'application de contrôles internes et à l'évaluation de l'efficacité des mesures prises.

L'auteur commente en détail la façon dont les données concernant les transferts sont enregistrées et traitées à l'USAEC, ainsi que les procédés utilisés pour présenter la comptabilité matières sous forme de rapports périodiques récapitulant les transactions et les résultats des opérations. Il décrit également les moyens employés par l'USAEC pour déterminer l'efficacité des programmes de gestion des matières nucléaires appliqués par les entreprises travaillant pour son compte. En plus des matières détenues par les entrepreneurs qui dirigent les usines et laboratoires de l'USAEC, il y a de grandes quantités de matières nucléaires spéciales qui sont détenues selon des arrangements variés, par exemple location, propriété privée, contrats passés avec l'USAEC en vertu desquels l'utilisateur est financièrement responsable des pertes ou, inversement, prévoyant que l'USAEC assume les risques de perte. Du fait de cette diversité de conditions, l'USAEC prend des dispositions dont l'optique varie selon les éléments de chaque cas. L'auteur fait état de ces variations. Pour conclure, il note succinctement les domaines comportant des problèmes encore à résoudre.

ПРОГРАММА КОМИССИИ ПО АТОМНОЙ ЭНЕРГИИ США В ОБЛАСТИ ОБРАЩЕНИЯ С ЯДЕРНЫМИ МАТЕРИАЛАМИ. Определяется область обращения с ядерными материалами под руководством Комиссии по атомной энергии США и излагается история развития этой отрасли производства с 1943 года до настоящего времени. Намечается и обсуждается в общих чертах основная программа. Упор делается на осуществление тех частей программы КАЭ США, которые имеют отношение к разработке и хранению количественных данных, установлению внутреннего контроля и оценке качества исполнения.
Довольно подробно обсуждаются применяемые в настоящее время методы регистрации и обработки данных о передаче в рамках КАЭ, как, например, методы представления отчетов о балансе материалов, в которых периодически обобщаются переводы и результаты операций. Также обсуждаются методы, применяемые КАЭ для проверки эффективности программ подрядчиков в области обращения с ядерными материалами. Кроме материала, которым располагают подрядчики, эксплуатирующие установки и лаборатории, принадлежащие КАЭ, большое количество специального расщепляющего материала поступает в рамках очень разнообразных финансовых и административных мероприятий, например при сдаче в аренду, в случаях частной собственности, по контракту с КАЭ, когда потребитель несет финансовую ответственность за потери, а также по таким контрактам, когда ответственность за потери берет на себя КАЭ. Такие различия порождают значительные изменения в подходе к решению проблемы, применяемому КАЭ, и такие изменения отмечаются. В заключении доклада кратко перечисляются области нерешенных проблем.

EL PROGRAMA DE ADMINISTRACION DE MATERIALES NUCLEARES DE LA COMISION DE ENERGIA ATOMICA DE LOS ESTADOS UNIDOS. El autor expone los principios adoptados por la Comisión de Energía Atómica de los Estados Unidos para la administración de materiales nucleares y sigue la evolución de esos principios desde 1943 hasta el momento actual. Describe el programa general y analiza sus principales características, insistiendo especialmente en aquellas partes de dicho programa que se refieren a la obtención y al registro de datos cuantitativos, al establecimiento de un control interno y a la evaluación de los rendimientos.

Explica con cierto detalle las técnicas empleadas por la AEC para registrar y sistematizar los datos referentes a las transferencias, así como las técnicas de presentación de informes sobre el balance de materiales en los que se resumen periódicamente las transacciones y los resultados de las operaciones. Trata también de los métodos seguidos por la AEC para asegurarse de la eficacia de los programas establecidos por sus contratistas para la administración de los materiales nucleares. Además de las que se encuentran en poder de los contratistas que explotan plantas y laboratorios de la AEC, existen considerables cantidades de materiales nucleares especiales que son objeto de una gran variedad de arreglos de carácter financiero y administrativo: en arriendo, con el título de propiedad transferido, sujetos a contratos en virtud de los cuales el usuario es responsable económicamente de las pérdidas o a acuerdos en los que se estipula que las pérdidas recaen en la AEC. La memoria trata de la forma en que la AEC enfoca esas situaciones tan distintas y concluye con un breve resumen de los problemas que aún quedan por resolver.

1. INTRODUCTION

It might be stated that nuclear materials management is not a new discipline. In fact, materials management, or inventory control, or by whatever other designation the function is known in other countries, is a basic need of any efficient and successful organization. As applied to nuclear materials, perhaps this point was made most clearly when it was stated a few years ago that "The function of nuclear materials management is an ordinary part of business, surrounded by rather unordinary circumstances" [1].

2. DEFINITION

The nuclear materials management function is subject to many definitions and its scope depends largely upon the needs of the organizational entity concerned. Therefore, it is appropriate at the outset that I define for you the function and scope of this activity as it is employed by the United States Atomic Energy Commission:

"Nuclear materials management means the effective use of methods, procedures, and techniques for recording, reporting, analyzing, eva-
luating, adjusting, and regulating nuclear materials inventories to assure maximum efficiency and economy consistent with established national and corporate policies and goals. More specifically, nuclear materials management includes acquisition, use and disposal of materials so as to effect maximum economies in materials utilization and to minimize consumption and losses; development and maintenance of records, measurement and physical inventory procedures to provide quantity data for cost and financial control, production control, and health and safety; establishment of internal controls to guard against carelessness, theft and misappropriation and to assure compliance with managerial policy; determination of economic inventory levels and reorder quantities consistent with program requirements; and, determination of the economics of current recovery of scrap vis-a-vis storage for future recovery or discard.

Within this definition and scope, it is appropriate that I make clear what specific materials we in the United States include. We term as "nuclear materials" only those elements of matter which are fissionable or fusionable of from which a fissionable or fusionable element or material may be produced. Thus we include within this category the elements uranium (normal and depleted, as well as enriched), plutonium, thorium, uranium-233, deuterium, tritium, lithium enriched in lithium-6, and neptunium-237. I do not suggest that universal agreement could be obtained for this definition as I am sure that other governments would wish to add to or subtract from this list to better meet their individual needs.

3. HISTORY AND DEVELOPMENT

To provide you with a better understanding of our nuclear materials management programme, please permit me to trace its history and development very briefly. Its origin was in 1943 when preservation of every gram of enriched uranium and plutonium was important to our national security. In those days it was of vital importance that we maintained continuing and accurate knowledge of these materials to minimize losses and maximize utilization. Another important background point is that until 1954 all plants and laboratories in the United States which handled these nuclear materials were either owned by the United States government and operated for the government by contract or were privately owned but operated solely for the account of the government. In no instance did a contract operator invest his own funds in his special nuclear materials inventory nor was he held financially responsible for losses, except if such losses occurred as a result of corporate mismanagement.

From this beginning twenty-two years ago the USAEC nuclear materials management programme has grown until today it encompasses materials valued in excess of $12 thousand million, located at more than two hundred different sites. The value of these materials at the individual sites ranges from a few hundred dollars to several hundred million dollars. The types of facilities located at the sites where these materials are located include
gaseous diffusion plants for enriching uranium in the $^{235}$U isotope; nuclear reactor plants for the production of plutonium and tritium and the breeding of $^{233}$U; plants for fabricating fuel elements; nuclear power plants; plants for processing irradiated fuels; plants for the purposes of defense; research and development facilities ranging from the smallest research reactors to our large national nuclear research laboratories at Argonne, Brookhaven and Oak Ridge; and a large number of privately-owned facilities throughout the United States which perform nuclear research and development work under contract to the United States Atomic Energy Commission.

Subsequent to 1954, our government began leasing special nuclear material under conditions wherein the lessee is financially responsible for losses. Under such circumstances, the principal management responsibility for these nuclear materials is delegated to the lessee. And just last year our Congress authorized the private ownership of special nuclear materials within the United States. Our government has no direct responsibility for the management of these leased and privately-owned nuclear materials; however, for both of these categories the government through the AEC organization carries out other responsibilities under law. These other responsibilities result in the need to acquire nuclear materials data much the same as that required by the AEC in its own operations.

4. USAEC PROGRAMME

4.1. Discussion limitation and outline

Although earlier I defined quite broadly the scope of the nuclear materials management programme, this presentation will be limited to a discussion of those areas concerned primarily with the developing and recording of information on material quantities and the establishment of internal controls to assure continuing knowledge of the physical whereabouts of these nuclear materials quantities within a plant or complex of plants. Now with that background, I should like to discuss briefly four points:

(a) The philosophy and administration of the USAEC programme.

(b) The techniques used to transmit and report information on materials quantities.

(c) The techniques used to ascertain the effectiveness of the programme and to verify the materials information submitted.

(d) The areas in which we believe additional development is needed.

4.2. Philosophy and administration

Generally speaking, the direct involvement of the USAEC in atomic energy activities is limited to policy and programme development and contract administration, as it is the policy of the USAEC to implement its programmes by using contractors rather than directly by using employees of the government. These contractors are responsible for reaching the goals set forth in the contracts and are administered by the USAEC through regional offices. The manager of each such regional office has a small staff res-
ponsible for assuring that its contractors comply with USAEC policies and procedures for nuclear materials management. These policies and procedures are developed and disseminated by the USAEC's Division of Nuclear Materials Management, which reports to the USAEC General Manager and the five-man Atomic Energy Commission through the Assistant General Manager for Administration. These activities are performed by professional staffs which generally are integrated into a single organizational unit to assure the availability of all needed skills.

4.3. General principles

The USAEC has based its nuclear materials management programme on the same general principles and techniques as those used by private industry in the United States to control and account for highly valuable raw materials, in-process inventories and manufactured products. However, because of the variety of the materials processed, the wide range of complexities of the processes, and the dissimilarity of operations conducted by the different facilities, no attempt has been made by the USAEC to prescribe the detailed procedures to be used within the individual plants. Those procedures are the responsibility of the management of the individual facility, although they are subject to the approval by the manager of the USAEC regional office. Notwithstanding these wide variations in our operations, certain basic activities are required by the USAEC and are common to all; they vary only in degree. These basic activities are: the recording and reporting of transactions, inventories and losses; maintenance of adequate internal control, measurement, and physical inventory procedures; prompt and accurate detection and estimation of materials losses and loss mechanisms; and the institution of corrective measures to reduce materials losses.

4.4. Responsibility of plant management

The USAEC requires its local plant management to take all necessary actions to ensure that the locations of all government-owned nuclear materials are appropriately known and that these materials are utilized for the purpose for which they were acquired. In the performance of this responsibility the USAEC expects that its contractors will obtain and process the requisite minimum nuclear-materials-inventory control information which is necessary to permit prudent management to achieve safe, efficient and economical operations. In so doing, the USAEC expects such actions to be taken only after consideration of the value of the materials being controlled, the cost of such actions, and the value of the information to be obtained. The acquisition and use of such inventory control information for management decisions is conventional in American industry.

4.5. Strategic value

I think it appropriate at this time to set at rest any doubts one might have as to the importance the USAEC puts upon the strategic value of the material. The emphasis placed herein on the control measures appropriate
to the economic value of the material may not be construed as meaning that the USAEC no longer recognizes the strategic importance of the material. Rather, this criterion was adopted as an aid in judging the appropriateness of the control measures, for the USAEC has concluded that when it takes all measures which are appropriate for criticality, health and safety, and for the economic value of the material, additional control measures generally would result in little, if any, additional protection against unrecognized loss or unauthorized use. However, when the occasion arises, as is possible when handling materials under classified conditions, such additional effort as is necessary is, of course, provided.

4.6. Uses of data

The collected inventory control information provides the basis for determining how much material should be on hand at any location and, through inventoried, how much actually is on hand at that location. Thus, those responsible for plant management are made aware of any losses which may occur and where they are occurring, thus enabling necessary action to minimize future losses. In addition, the information provides a service to production control, cost and financial accounting, criticality control, health and safety control, and assistance in the control of scientific and engineering experiments.

4.7. Transfer documents

Although, as I have indicated, the USAEC assigns the responsibility for establishing and maintaining good nuclear materials inventory control and management to its local plant management, there are certain records areas in which the USAEC specifies the documentary requirements. One such area is that of the documentation of nuclear materials transfers. The USAEC requires that each transfer of nuclear material between discrete plant sites be recorded on a prescribed document with prescribed distribution of each such document to the shipper and receiver and to the respective USAEC regional offices. For material transfers between operating contractors in which neither the shipper nor the receiver is held financially liable for losses in transit, the USAEC requires that both the shipper and the receiver develop and record their own values for the nuclear material content of the shipments, independently of each other. For transfers in which either the shipper or the receiver may be held financially liable for losses, measurement differences are resolved prior to completion of the transfer document, and thus, only a single value for nuclear material content is reflected on the transfer document. Copies of these documents are forwarded independently by the shipper (at the time of shipment) and by the receiver (at time of receipt) to the respective USAEC regional office having responsibility for contract administration of the shipper and the receiver. A copy of this transfer form and its continuation sheet are attached as Fig. 1 and Fig. 2.
4.8. Material balance reports

Another such area in which the USAEC specifies the degree and kind of documentation is that involving the submission of periodic material balance reports summarizing all external receipts and shipments and other additions to or removals from the contractor's on-site inventories. Although the kind of information included in this document has always been prescribed, only recently has the USAEC adopted a prescribed form to be used. This is because we in the USAEC are now converting our nuclear materials information system to electronic data processing. Until last year, these material balance reports were submitted monthly but now they are required for submission to USAEC headquarters only semi-annually; however, they are submitted as frequently to the plant and USAEC regional managers as is deemed necessary by these managers to serve local operational control purposes. A copy of this material balance report form is attached as Fig. 3.

4.9. Indicators of effectiveness

Two "indicators" used by the USAEC and its operating contractors to judge whether the procedures employed by a plant facility are effective, are those known as "Material Unaccounted For" and "Shipper-Receiver Differences". "Material Unaccounted For" is the amount by which the quantity of material on hand as determined by physical inventories differs from that which the records show should be on hand after all data have been recorded. "Shipper-Receiver Difference" is the amount by which the quantity of material as measured by a shipper differs from the quantity of material as measured by a receiver. Material Unaccounted For and Shipper-Receiver Differences can occur in varying magnitudes for any of the following single or combined causes: (a) measurements uncertainties, (b) unknown process or other losses, (c) human errors, and (d) diversions or thefts. The USAEC uses control charts and other statistical tests for evaluating Material Unaccounted For and Shipper-Receiver Differences as an aid to management in judging whether investigative action is warranted for any single or recurring event and whether adequate inventory management control is being employed.

5. INSPECTIONS

5.1. Objectives

In addition, basic to any well-managed operation are programmes of inspection, the best known being those used for quality control and financial audits. Consistent with this management philosophy, the USAEC has a programme of surveillance and inspection to assure that the USAEC operating contractors have established and are maintaining effective nuclear materials management procedures. In implementing this activity, the USAEC conducts a comprehensive field examination, known as a "survey", of the nuclear materials management activities of each of its facilities at least once a year. The objectives of such a field survey are to determine, record, and report on the adequacy of the nuclear materials management procedures and prac-
FIG. 1. Nuclear materials transfer document
FIG. 2. Nuclear materials transfer document (continuation page)


FIG. 3. USAEC material balance report

jectives of the facility; to establish that the nuclear materials inventories are being utilized for the purposes intended by the USAEC; to ascertain whether the particular contractor's material balance reports present fairly the material inventories and results of operations; and to establish the degree to which the contractor has adhered to USAEC policies and procedures governing nuclear materials management. As part of this programme of surveillance, we encourage self-inspection or internal audit by the contractors who operate our plants for us.
5.2. Scope of inspections

Because of the highly technical nature of such diverse and complex operations as are found at laboratories, fuel fabrication plants, reactor plants and spent fuels processing plants, each survey is performed by a team of people trained in the professions appropriate to the survey mission, such as, reactor technologists, statisticians, physicists, chemical engineers, chemists and auditors. The actual size and professional make-up of the survey team, the scope and the details of the various measurement, audit and statistical tests will vary with the type, size, and complexity, as well as the numbers and types of transactions performed by the facility being surveyed. Nevertheless, a survey always encompasses the following examinations:

**Measurement system.** To evaluate the adequacy of the techniques of weighing, sampling, chemical and isotopic analyses, and calculations of reactor materials production and nuclear loss or consumption.

**Inventory procedures.** To evaluate the capability of the procedural system to routinely develop inventory data with an accuracy appropriate to the value of the nuclear materials.

**Independent test of inventory.** To obtain an independent opinion by survey personnel of the validity of the reported inventory quantities, including both the element and isotopic content, as distinct from merely ascertaining that containers, items and gross weight are present.

**Statistics.** To determine the appropriateness of the statistical principles as applied by the facility; the precision and accuracy of measurements made at the facility; and whether additional areas exist in which the application of statistical analysis would serve a useful nuclear materials control purpose. The survey also includes determination of the statistical reliability of the material balance report quantities.

**Internal control.** To evaluate the extent to which the facility has installed and maintains a system of checks and balances in the division of duties, designed so that the work of one person serves to verify the work of another.

**Audit of material records and reports.** To evaluate the integrity and accuracy of the materials accounting records and the correctness of the material balance reports prepared therefrom. The findings and recommendations resulting from each survey are reported in writing to USAEC management.

5.3. Inspection differences between USAEC-owned and privately-owned plants

At plants and laboratories owned by the government we are interested not only in verifying the nuclear materials inventory information but also in appraising the efficiency of such other nuclear materials management aspects
as prompt recovery of nuclear materials from scrap and residues, the development and application of improved measurements methods, the reduction of excess inventories and the use of more efficient management techniques. However, at plants and laboratories using nuclear materials which are leased from the government or are privately owned, USAEC interest is directed primarily to the verification of the nuclear materials inventory information. We do not attempt to evaluate those other aspects of nuclear materials management which relate to the economic use of the materials.

6. AREAS NEEDING DEVELOPMENT

6.1. Economic criteria

Until recently these economic aspects of the USAEC programme of nuclear materials management have been left to local USAEC and local USAEC contractor management at the plants and laboratories. This was done largely because rapid technological advances resulted in significant process changes and inventory requirements and because the urgency of certain programmes took precedence over other factors. Now, with the rapid increase in development of peaceful uses for nuclear materials, in competition with other forms of energy, greater emphasis and attention is being given to economic considerations. Now the USAEC is increasing its efforts to study and develop economic criteria on which to base management decisions relative to the recovery of nuclear materials from scrap inventories, the appropriate level of nuclear materials inventories, the appropriate re-order rates based on these economic considerations and the control of scrap generation in conversion and fabrication processes.

6.2. Electronic data processing

Another significant step being taken by the USAEC today is the conversion of all information on nuclear materials quantities within the United States from manual to electronic data processing. Although we have made a good start toward this goal we estimate it will take several years before this conversion is completed.

6.3. Technical problems

In recent years, development work in non-destructive testing of nuclear materials by gamma-scintillation spectrometers, an increase in the number of analytical samples submitted to independent laboratories, the distribution of standard reference materials, improved measurement methods and other technical advances have greatly enhanced the USAEC's knowledge of its resources and have provided improved measurement precision and accuracy. Although increased knowledge of the USAEC's nuclear material assets are being achieved continually, additional research and development efforts are still needed as there are numerous thorny technical problems which need solution. In general, these are associated with the development of better
methods of determining the nuclear-material content of scrap and residues, fuel elements both irradiated and unirradiated, in-process inventories, in-reactor fuel inventories, and the consumption and production of nuclear material in reactors. Many of the presentations during this Symposium are directed toward the solution of these problems.

7. CONCLUSION

This completes my discussion of the broader aspects of the programmes of nuclear materials management conducted by the United States Atomic Energy Commission. In closing, I should like to emphasize that because of the complexity of the processes and associated problems a high calibre of professional and scientific personnel is required. Thus, good management of nuclear materials is not obtained easily or cheaply, but failure to establish an effective programme can result in exorbitant costs.

During this meeting, representatives of the USAEC, its operating contractors and private industry in the USA will have discussed in some detail various aspects of nuclear materials management as practiced in individual plants and laboratories. Our representatives will also have discussed a number of specific problems which have arisen and the steps which have been taken toward their solution.

REFERENCE


DISCUSSION

J. SORNEIN: Hearing the last two papers in succession has left me a little confused. We were all probably under the impression, by this stage of the proceedings, that nuclear materials management in our various countries required more thorough organization than at present. Mr. Hood's paper might well have destroyed this impression, whereas yours supports it. What are your views on this?

D.E. GEORGE: The circumstances in the United Kingdom may differ sufficiently from those in the United States of America to justify the difference in approach which is apparent. However, the important thing is that inspection should be carried out by a group independent of that being inspected; it is not vital that the inspectors form part of a central agency. As I understand the position in the United Kingdom, they rely on physical inventories and inspections by the plant management; this is in my view not adequate.

St. C.C. HOOD: To a great extent, the difference in practice between the two countries results necessarily from the difference in the roles of the USAEC and the U.K.A.E.A. The Commission/contractor relationship is not present in the United Kingdom. In the U.K.A.E.A., the over-all structure is federal, with consumers and producers forming integral parts of
the organization. Because of this, the representatives of consumers and producers on the central committee system are integrated completely with the central as well as the local accounting system, and have common interests with both.

D.E. GEORGE: I recognize the importance of having each plant financially responsible, and agree that in this respect the United Kingdom approach gives a measure of assurance which is not usual in the United States of America. However, I doubt whether this is a completely satisfactory substitute for inspection by an independent group.

NAKAJIMA: Could you please define the term "quality control" as used in your paper?

D.E. GEORGE: My reference to quality control was made simply to demonstrate that scientists and technical personnel are not unaware of inspection procedures and requirements, so that those involved in nuclear materials management should not prove onerous.

M. J. HIGATSBERGER: We have heard that the USAEC has to manage nuclear materials valued at more than US $12 000 million. How much money is invested each year to improve nuclear materials management by new techniques and methods?

D.E. GEORGE: Since much of this research and development work is carried out by the plants as part of their normal business, and no cost totals are drawn up for this, the total invested cannot be exactly known. However, the USAEC's Division of Nuclear Material Management has a budget of US $500 00 for research and development.

J. SCHMETS: To revert to the question of independent inspections, there is at present a tendency in the United States of America to authorize the ownership of nuclear materials by private companies. In the extreme case of a large company owning materials and engaged in both the production and the reprocessing of fuel, do you not feel that the materials management will be particularly well done?

D.E. GEORGE: One could reasonably hope that having money invested in nuclear material will result in a desire to know what happens to that material. But this will not necessarily be so unless there is some independent group performing inspections to ensure that policies and procedures are complied with. Until there is completely free competition, private utilities will be able to pass excessive costs on to the consumers; they and their fabricators thus have at present no real incentive towards good nuclear materials management.
CHEMICAL AND ISOTOPIC ANALYSIS
(Session VII)
METHODS OF ANALYSING PLUTONIUM. Numerous problems of plutonium analysis are encountered in the management of production plants and waste recovery facilities. In both cases there are three main reasons for making the plutonium balance sheet as accurate as possible: to have information on the operation of the establishment, to check on movements of material between consignors and consignees, and to ensure safety.

It is therefore necessary to analyse plutonium at different stages of its production and recovery. This analysis applies to a considerable range of quantities and environments, and especially at the beginning of the process must be carried out in various, often inconvenient, circumstances. The numerous solutions in use at present are based on such techniques as chemistry, radiometry and mass spectrometry. The paper reviews the methods involved and discusses their advantages and disadvantages.

As an example of nuclear materials control, it describes a recent transaction between the United Kingdom Atomic Energy Authority and the Commissariat à l'énergie atomique in connection with RAPSODIE. This operation involved the delivery of 45 kg of plutonium as the oxide under a contract including a number of specifications which had to be checked by analysts of the two countries.

METHODES D'ANALYSE DU PLUTONIUM. De nombreux problèmes analytiques relatifs au plutonium se posent pour la gestion, soit d'une usine de production, soit d'une installation de récupération de déchets. Dans les deux cas, il est nécessaire d'en établir le plus exactement possible le bilan pour trois raisons principales: connaître le fonctionnement de l'unité, vérifier les mouvements entre expéditeur et destinataire, assurer la sécurité.

Aux différents stades de la production ou de la récupération du plutonium, il est donc nécessaire d'effectuer l'analyse de cet élément. Cette analyse s'applique à des quantités et des milieux très différents et doit, surtout en début de procédé, être réalisée en présence d'éléments variés et souvent gênants. De nombreuses solutions sont actuellement utilisées, faisant appel à des techniques telles que la chimie, la radiométrie, la spectrométrie de masse. Les méthodes correspondantes sont passées en revue et on effectue la critique de leurs avantages et leurs inconvénients.

Un exemple de contrôle des matières nucléaires est fourni par une transaction récemment effectuée pour RAPSODIE entre l'United Kingdom Atomic Energy Authority et le Commissariat à l'énergie atomique. Cette transaction concernait la livraison de 45 kg de plutonium sous forme d'oxyde, selon les termes d'un contrat qui comprenait un certain nombre de spécifications qui ont dû être vérifiées par les analystes des deux pays.

МЕТОДЫ АНАЛИЗА ПЛУТОНИЯ. Многие относящиеся к плутонию аналитические проблемы возникают в связи с управлением заводом по производству плутония либо установкой по регенерации отходов. В обоих случаях необходимо как можно точнее составить баланс, исходя из трех основных моментов: знать работу предприятия, проверить способ транспортировки от отправителя до получателя, обеспечивать безопасность.

На различных стадиях производства или регенерации плутония необходимо проводить анализ на этот элемент. Анализируются пробы, содержащие различные количества плутония в разнообразной среде и, особенно в начале процесса, в присутствии различных элементов, часто мешающих определению. Для решения таких аналитических проблем в настоящее время используются методы химии, радиометрии и масс-спектрометрии. Дается обзор соответствующих методов, критически рассматриваются их преимущества и недостатки.

Пример контроля за ядерными материалами приводится в договоре, недавно заключенном для реактора "Рапсодия" между Управлением по атомной энергии Соединенного Королевства и Комиссариатом по атомной энергии. Этот договор касается поставки 45 кг плутония в виде окиси в соответствии с условиями контракта, в котором приводится определенное количество спецификаций, подлежащих проверке специалистами-аналитиками обеих стран.
METODOS DE ANALISIS DEL PLUTONIO. En la administración de las plantas de producción o de las instalaciones de tratamiento de desechos el análisis del plutonio plantea numerosos problemas. En ambos casos es preciso establecer lo más exactamente posible su balance por tres motivos principales: para conocer el funcionamiento de la planta o de la instalación, para comprobar las transferencias entre el expedidor y el destinatario y para garantizar la seguridad.

Es pues necesario analizar el plutonio en todas las etapas de su producción o de su recuperación. Este análisis se efectúa con cantidades y en medios muy diversos y — sobre todo al iniciarse el proceso — en presencia de elementos que a menudo entorpecen la operación. Se han hallado varias soluciones basadas en procedimientos químicos, radiométricos o de espectrometría de masas. En la memoria se describen los correspondientes métodos y se examinan sus ventajas y sus inconvenientes.

Como ejemplo de control de materiales nucleares el autor cita una transacción reciente entre la United Kingdom Atomic Energy Authority y el Commissariat à l'Energie Atomique: se trataba de suministrar 45 kg de plutonio, en forma de óxido, para el reactor RAPSOIDE. Este suministro se efectuó con arreglo a un contrato en el que figuraban ciertas especificaciones que los analistas de ambos países tuvieron que verificar.

Dans le cadre du contrôle des matières nucléaires, l'analyse du plutonium a un rôle important à remplir. Elle doit permettre de répondre aux questions posées par la gestion d'usines de production, d'installations de récupération de déchets, d'unités de fabrication d'éléments combustibles. Cette gestion suppose, en particulier, l'établissement de bilans le plus exactement possible afin de connaître et contrôler le fonctionnement de l'unité, d'assurer la sécurité et de vérifier les mouvements de plutonium entre expéditeur et destinataire.

Les produits qu'il est nécessaire d'analyser sont très différents. Si l'on prend l'exemple d'une usine d'extraction de plutonium à partir de barreaux d'uranium irradiés, il faut, en début de procédé, doser le plutonium à l'état de traces en présence d'une quantité importante d'uranium et de produits de fission. En fin de procédé, il faut contrôler la pureté du plutonium produit, c'est-à-dire déterminer un pourcentage compris entre 99 et 100% et analyser les impuretés présentes. Pour doser le plutonium dans des domaines aussi variés, il est nécessaire d'utiliser des techniques elles-mêmes très différentes. L'application de ces techniques est rendue possible par le fait que le plutonium possède un large éventail de propriétés qui rendent utilisables, pour son analyse, des méthodes chimiques et électrochimiques, la radiométrie, la spectrographie. Ces méthodes, telles qu'elles sont utilisées en France à l'heure actuelle, vont être passées en revue.

1. METHODES CHIMIQUES ET INSTRUMENTALES

1.1. Méthodes de titrage par oxydoréduction

Les deux systèmes d'oxydoréduction utilisés en analyse sont les systèmes

Pu(III)/Pu(IV) (réversible)
Pu(IV)/Pu(VI) (irréversible)
a) Système Pu(III)/Pu(IV)

Le plutonium à doser est réduit à la valence III. Le titrage du plutonium (III) est ensuite effectué au moyen d'une solution oxydante. Les réducteurs les plus couramment utilisés pour obtenir le plutonium à la valence III sont: Ti(III), Cr(II) et l'amalgame de zinc. La solution titrée oxydante est une solution de sulfate cérique. Le point équivalent est déterminé au moyen d'un indicateur [1], par potentiométrie [2] ou par spectrophotométrie [3].

La méthode la plus couramment utilisée en France [4] consiste à réduire le plutonium en solution sulfurique-nitrique par le titane (III) en présence d'acide sulfamique. L'excès de titane (III) est détruit par les ions nitriques présents en solution. Le titrage est effectué par le sulfate cérique, le point équivalent déterminé par un indicateur coloré, l'orthophénantholine ferreuse, ou par potentiométrie.

Les ions gênants les plus fréquemment rencontrés sont le fer et l'uranium; cette méthode s'applique donc à des produits purs ou nécessite une correction si la teneur en impuretés est connue.

b) Système Pu(IV)/Pu(VI)

Le plutonium à doser est oxydé à la valence VI, puis réduit par une solution de fer (II) en excès qu'on titre en retour par une solution de sulfate cérique. Ce principe de réaction permet d'éviter l'interférence des éléments dont le système oxydoréducteur a un potentiel égal ou inférieur à celui du système Fe(II)/Fe(III), en particulier le fer lui-même et l'uranium.

Différents agents oxydants permettant d'obtenir le plutonium à la valence VI ont été décrits, en particulier l'acide perchlorique, le persulfate d'ammonium, l'oxyde argentique. Le principe du dosage en retour a été retenu par Metz et Waterbury [5], tandis que Larsen [6] titre le plutonium (VI) formé par une solution de fer (II) en utilisant une fin ampérométrique.

La méthode que nous utilisons consiste à oxyder le plutonium à la valence VI en solution nitrique, au moyen d'oxyde argentique. Le plutonium (VI) formé est réduit en présence d'acide sulfamique par une quantité connue de fer (II) en excès qu'on titre en retour par du sulfate cérique. Le point équivalent est déterminé par potentiométrie à intensité constante entre deux électrodes indicatrices de platine.


c) Performances des méthodes de titrage par oxydoréduction

Les deux méthodes décrites sont d'égale valeur et le choix doit être effectué en fonction de la nature de la solution à analyser, celle utilisant le système Pu(IV)/Pu(VI) étant tout naturellement indiquée pour le dosage
du plutonium en présence de quantités notables d'uranium. Elles présentent les intérêts suivants:
- utilisation de systèmes oxydoréducteurs rapides,
- possibilité de titrage en milieu nitrique – qui est un des acides le plus souvent utilisés en fabrication –, ce qui évite les fastidieuses opérations de changement de milieu,
- possibilité d'enregistrement du résultat,
- adaptation facile aux contrôles de routine,
- utilisation comme méthodes de haute précision sous réserve de certaines conditions opératoires: opérateur qualifié et entraîné, mesures des quantités de solution titrante au moyen de burettes calibrées ou par pesée, surveillance de la température, étalonnage des solutions titrantes par des produits de haute pureté. En prenant cet ensemble de précautions, les résultats donnés par les deux méthodes peuvent être obtenus avec une précision voisine de 0,1%.

Un des principaux facteurs influant sur la valeur du résultat est l'exactitude de l'étalonnage de la solution de sulfate céritique. Les produits retenus pour effectuer cet étalonnage sont l'oxalate de sodium et l'anhydride arséniux, qu'on peut se procurer avec une garantie de 99,9%, ainsi que l'étalon de plutonium fourni par le NBS. Les valeurs du titre obtenues au moyen de ces trois « standards » concordent de façon très satisfaisante.

1.2. Coulométrie à potentiel contrôlé

La coulométrie utilise les mêmes systèmes oxydoréducteurs que les méthodes de titrage par oxydoréduction, le volume ou le poids de la solution titrante étant remplacé par une quantité d'électricité. Kelley, Jones et Fisher [9,10] et Shults [11] ont publié un travail très abondant sur la coulométrie du plutonium et la mise au point d'intégrateurs perfectionnés.

La coulométrie a été appliquée en milieu sulfurique par réduction préalable du plutonium à la valence III au potentiel de +200 mV/ECS et mesure de l'oxydation du plutonium (III) en plutonium (IV) au potentiel de +800 mV/ECS. Les deux domaines explorés sont les suivants:

a) Quantités importantes de plutonium

La prise d'essai est comprise entre 20 et 100 mg de plutonium correspondant au meilleur domaine d'application du coulomètre ORNL/Q/2005×50 utilisé. Dans de bonnes conditions de marche, nous avons obtenu des reproductibilités de l'ordre de 0,1%, mais un léger doute subsiste au sujet de l'exactitude, les résultats obtenus au moyen du facteur électrique différant d'environ 0,2% de ceux donnés par l'étalon fourni par le NBS. En conclusion, la méthode est applicable concurremment au titrage par le cérium (IV), mais elle nécessite un appareillage et une surveillance de l'électronique plus compliqués.
b) Faibles quantités de plutonium

La coulométrie est largement utilisée pour le dosage du plutonium dans des solutions contenant 100 à 2000 fois plus d'uranium. En principe, le dosage pourrait être effectué directement, mais dans presque tous les cas l'uranium contient des impuretés à l'état de traces, le fer en particulier, dont le montant n'est pas négligeable par rapport à la faible quantité de plutonium. C'est la raison pour laquelle il est nécessaire d'effectuer une séparation.

La prise d'essai contient 1 à 2 mg de plutonium qu'on fixe en milieu nitrique 7N sur une résine anion type Dowex 1X4. Après lavage qui élimine la majeure partie de l'uranium et les éléments gênants, le plutonium est élué par une solution HNO₃ 0,5N/HF 0,01N. La solution est évaporée à fumées blanches en présence d'acide sulfurique, ajustée en milieu 1 ou 2N et dosée par coulométrie.

La méthode permet de doser le plutonium à environ 2% près, le soin principal devant porter sur la pureté des réactifs pour que les blancs soient négligeables devant la faible quantité d'électricité mesurée. Des résultats satisfaisants ont été obtenus sur des alliages U-Pu à faible teneur et la méthode a été également appliquée à l'analyse du plutonium dans l'uranium irradié. Dans ce dernier cas, l'inconvénient est que la séparation de l'uranium et des produits de fission doit être réalisée en enceinte αβγ, en raison des prises d'essai relativement élevées qui sont nécessaires. Ce dernier point fait préférer les méthodes d'analyse par dilution isotopique.

1.3. Spectrophotométrie par absorption

Les spectres d'absorption des différentes valences du plutonium présentent des pics utilisables en spectrophotométrie. Comme plusieurs valences coexistent souvent en solution, il est nécessaire d'ajuster le plutonium à une valence choisie, la plus facile à obtenir étant la valence III.

Le plutonium (III) présente deux pics, respectivement à 565 et 602 μm, ayant une valeur du coefficient d'extinction moléculaire assez voisine, de l'ordre de 37.

Une des méthodes les plus intéressantes est celle décrite par Phillips [12] qui applique au plutonium (III) la spectrophotométrie différentielle. Nous n'avons pas cherché à utiliser la spectrophotométrie comme méthode de haute précision; nous l'utilisons comme méthode de recoupement pour certains bilans où une précision de 1 à 2% est suffisante et où les solutions à analyser sont mal connues. En effet, les interférences sont peu nombreuses et l'on peut, en particulier, effectuer le dosage en présence de quantités notables de fer et d'uranium. Son inconvénient est de nécessiter un étalonnage au moyen de solutions connues de plutonium et d'être sensible à des variations de milieu et d'acidité. Elle garde néanmoins un grand intérêt en raison de sa facilité d'application.

2. MÉTHODES RADIOMETRIQUES

L'émission α du plutonium est l'une des propriétés utilisées depuis le plus longtemps pour son analyse en raison de sa grande sensibilité. À l'heure
actuelle encore, c'est une méthode très appréciée chaque fois que le pluto-
nium se trouve à l'état de traces et qu'on a besoin d'un contrôle rapide, par
exemple en début de procédé dans les usines d'extraction.

Malgré les perfectionnements apportés à la technique du comptage lui-
même, il semble difficile de le considérer comme autre chose qu'une mé-
thode pouvant donner une indication sur la marche d'un procédé et appli-
cable chaque fois qu'une grande précision n'est pas recherchée. De

nombreux facteurs doivent être pris en considération: la présence de sels
dissous, la composition isotopique du plutonium, la présence d'autres émet-
teurs α. La spectrométrie α ne résout que partiellement les problèmes
posés par ces deux derniers points et il est souvent nécessaire de faire pré-
céder le comptage d'une séparation chimique.

2.1. Concentration saline

Le plutonium à l'état de traces doit souvent être dosé en présence d'une

grande quantité de sels, comme c'est le cas par exemple dans de nombreux
effluents. Si l'on dépose et évapore la solution sur un support dans les con-
ditions normales, il se forme une croûte qui absorbe une partie du rayonne-
ment et fausse le résultat. On peut envisager deux façons d'y remédier:
- effectuer une séparation, ou
- effectuer le comptage en utilisant un scintillateur spécial de grande
  surface sur lequel on dépose directement la solution à doser en ajout-
tant du mergital [13].

2.2. Composition isotopique du plutonium - Autres éléments émetteurs α

Le plutonium comprend plusieurs isotopes dont la période est différente,
ce qui oblige à effectuer des mesures relatives si la qualité du plutonium
est constante, ou sinon nécessite l'emploi de la spectrométrie de masse.

Les autres émetteurs α principalement rencontrés sont, soit
des émetteurs à période plus longue mais présents à forte concentration, soit
des éléments peu abondants mais dont l'émission est importante vis-à-vis
de celle du plutonium-239. Le premier cas est celui de l'uranium, le se-
cond celui de l'américium.

Des séparations sont souvent nécessaires. Les deux procédés utilisés
sont
- l'extraction du plutonium (IV) par la thényoltrifluoroacétone (TTA)
en milieu nitrique 0,5 à 1N.
- la fixation du plutonium (IV) sur résine anion en milieu nitrique 7N.

3. SPECTROMETRIE DE MASSE

L'application de la spectrométrie de masse à l'analyse du plutonium
a un double but:
- connaître sa composition isotopique,
- déterminer sa teneur par une méthode de dilution isotopique [14-16].

La méthode de double dilution isotopique est utilisée en particulier pour
la détermination de rapports U/Pu dans les combustibles irradiés. On utilise
un traceur double, constitué d'un mélange de plutonium-242 et de plutonium-233, qu'on ajoute à la solution à doser. Le rapport U/Pu est donné par quatre analyses isotopiques:
- l'analyse isotopique du plutonium dans la solution à doser,
- l'analyse isotopique de l'uranium dans la solution à doser,
- l'analyse isotopique du plutonium dans le mélange solution + traceur,
- l'analyse isotopique de l'uranium dans le mélange solution + traceur.

L'analyse isotopique du plutonium nécessite une séparation chimique par fixation des complexes nitriques sur résine anion en raison de la trop grande quantité d'uranium présente.

Auparavant, la solution de traceur double est étalonnée par rapport à une solution connue contenant l'uranium et le plutonium dans un rapport voisin de celui à doser, et fabriquée par exemple à partir d'uranium et de plutonium électroraffinés.

La précision obtenue par la méthode de double dilution isotopique est de l'ordre de 1 à 2%. La mise en œuvre est coûteuse, longue et délicate. Elle demande beaucoup de soin pour éviter des contaminations isotopiques, beaucoup de temps puisque plusieurs analyses isotopiques sont nécessaires et que chaque détermination est elle-même la somme d'un grand nombre de mesures. A cela s'ajoute le temps passé pour les séparations chimiques et pour les calculs. Malgré ces inconvénients, elle présente un très grand intérêt pour l'analyse du plutonium pour les deux raisons suivantes:
- elle est actuellement la meilleure méthode de dosage de traces et peut être appliquée à des calculs de bilans,
- elle ne nécessite, pour sa mise en œuvre, que des prises d'essai très faibles (5 µg) qui la rendent facilement applicable à l'analyse de solutions fortement irradiées.

4. ANALYSE DES IMPURETÉS DANS LE PLUTONIUM

Bien que le dosage des impuretés dans le plutonium puisse sembler s'éloigner du problème de la gestion des matières nucléaires, il est utile de l'inclure dans les méthodes d'analyse, car
- il représente un pourcentage important du travail analytique effectué,
- il peut servir d'élément de calcul ou de recoupement, pour l'estimation de la teneur en plutonium dans le plutonium métal,
- il donne des indications sur la qualité du plutonium au point de vue de ses propriétés nucléaires.

L'analyse des impuretés fait généralement appel à des méthodes instrumentales dont la plus répandue est la spectrographie d'émission parce qu'elle permet de doser dans le même temps un grand nombre d'éléments. On connaît trois méthodes de spectrographie d'émission:
- «Copper Spark» [17], effectuée directement sur une solution chlorhydrique de plutonium; ce procédé est de moins en moins utilisé à cause de l'interférence des raies du plutonium qui limite la sensibilité;
- «Graphite Spark», effectuée sur une solution nitrique après séparation du plutonium par la TTA;
- la «distillation par entraîneur» [18], effectuée sur l'oxyde de plutonium.
DOSAGE DES IMPURETÉS DANS LE PLUTONIUM

<table>
<thead>
<tr>
<th>Élément dosé</th>
<th>Méthode</th>
<th>Sensibilité ppm</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ag</td>
<td>Spectrographie d'émission</td>
<td>&lt;1</td>
</tr>
<tr>
<td>Al</td>
<td>Spectrographie d'émission</td>
<td>20</td>
</tr>
<tr>
<td>B</td>
<td>Spectrographie d'émission</td>
<td>&lt;1</td>
</tr>
<tr>
<td>Be</td>
<td>Spectrographie d'émission</td>
<td>5</td>
</tr>
<tr>
<td>C</td>
<td>Microvolumétrie</td>
<td>20</td>
</tr>
<tr>
<td>Ca</td>
<td>Spectrographie d'émission</td>
<td>10</td>
</tr>
<tr>
<td>Cd</td>
<td>Spectrographie d'émission</td>
<td>1</td>
</tr>
<tr>
<td>Cl</td>
<td>Potentiométrie après séparation sur résine cation</td>
<td>4</td>
</tr>
<tr>
<td>Cr</td>
<td>Spectrographie d'émission</td>
<td>5</td>
</tr>
<tr>
<td>Cu</td>
<td>Spectrographie d'émission</td>
<td>5</td>
</tr>
<tr>
<td>F</td>
<td>Spectrophotométrie par décoloration du complexe zirconium, Eriochromecyanine R après séparation par pyrohydrolyse</td>
<td>&lt;5</td>
</tr>
<tr>
<td>Fe</td>
<td>Spectrophotométrie par l'orthophénanthroline Spectrographie d'émission</td>
<td>20</td>
</tr>
<tr>
<td>K</td>
<td>Spectrographie d'émission</td>
<td>1</td>
</tr>
<tr>
<td>Li</td>
<td>Spectrographie d'émission</td>
<td>1</td>
</tr>
<tr>
<td>Mg</td>
<td>Spectrographie d'émission</td>
<td>10</td>
</tr>
<tr>
<td>Mn</td>
<td>Spectrographie d'émission</td>
<td>10</td>
</tr>
<tr>
<td>Mo</td>
<td>Spectrographie d'émission</td>
<td>5</td>
</tr>
<tr>
<td>N</td>
<td>Microvolumétrie après séparation par méthode Kjeldahl</td>
<td>10</td>
</tr>
<tr>
<td>Na</td>
<td>Spectrographie d'émission</td>
<td>50</td>
</tr>
<tr>
<td>Ni</td>
<td>Spectrographie d'émission Spectrophotométrie par diméthylglyoxime nickelique</td>
<td>10</td>
</tr>
<tr>
<td>Np</td>
<td>Polarographie après séparation sur résine anion</td>
<td>50</td>
</tr>
<tr>
<td>O</td>
<td>Spectrographie d'émission (méthode Fassel)</td>
<td>100</td>
</tr>
<tr>
<td>P</td>
<td>Spectrographie d'émission</td>
<td>50</td>
</tr>
<tr>
<td>Pb</td>
<td>Spectrographie d'émission</td>
<td>10</td>
</tr>
</tbody>
</table>
C'est la méthode la plus couramment utilisée en raison du nombre élevé d'éléments qu'elle permet de doser.

D'autres procédés sont également utilisés, le plus courant étant la spectrophotométrie par absorption; viennent ensuite la microvolumétrie, la polarographie, la fluorimétrie. L'application de ces méthodes est destinée, soit à l'analyse des éléments non dosables à l'état de traces par spectrographie d'émission, c'est le cas par exemple de l'uranium et du thorium, soit à servir de recoupement pour l'étalonnage des méthodes spectrographiques, soit à une analyse isolée quand une seule impureté est recherchée.

Dans la plupart des cas, les analyses d'impuretés sont précédées d'une séparation. Ce procédé permet de se débarrasser de la majeure partie du plutonium, de faciliter sa récupération et d’éviter l’interférence fréquente du plutonium dans la méthode d'analyse choisie. Les séparations les plus couramment utilisées sont:

- la fixation des complexes nitriques ou chlorhydriques du plutonium sur résine anion [19],
- l'extraction par solvant: thénoyl trifluoroacétone (TTA) et trioctylphosphine oxyde (TOPO) [20, 21].

La liste des impuretés dosées par ces différents procédés est résumée dans le tableau I.

**TABLEAU I (suite)**

<table>
<thead>
<tr>
<th>Élément dosé</th>
<th>Méthode</th>
<th>Sensibilité ppm</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sb</td>
<td>Spectrographie d'émission</td>
<td>10</td>
</tr>
<tr>
<td>Sn</td>
<td>Spectrographie d'émission</td>
<td>1</td>
</tr>
<tr>
<td>Si</td>
<td>Spectrographie d'émission</td>
<td>10</td>
</tr>
<tr>
<td>Th</td>
<td>Spectrophotométrie avec thoron après séparation sur résine anion en milieu chlorhydrique</td>
<td>30</td>
</tr>
<tr>
<td>Tl</td>
<td>Spectrographie d'émission</td>
<td>50</td>
</tr>
<tr>
<td>U</td>
<td>Polarographie, Fluorimétrie</td>
<td>100, 10</td>
</tr>
<tr>
<td>Zn</td>
<td>Spectrographie d'émission</td>
<td>50</td>
</tr>
<tr>
<td>Zr</td>
<td>Spectrographie d'émission</td>
<td>50</td>
</tr>
</tbody>
</table>

**REFERENCES**

ANALYTICAL METHODS FOR URANIUM CONCENTRATION MEASUREMENTS

J. P. HIGHFILL AND C. J. RODDEN
NEW BRUNSWICK LABORATORY,
UNITED STATES ATOMIC ENERGY COMMISSION,
NEW BRUNSWICK, N. J., UNITED STATES OF AMERICA

Abstract — Résumé — Аннотация — Resumen

ANALYTICAL METHODS FOR URANIUM CONCENTRATION MEASUREMENTS. A survey of analytical procedures for the determination of uranium, as performed for NMM in the United States of America, is presented. Methods are outlined for the measurement of the element in a variety of materials, i.e. ores, concentrates, uranium metal, alloys, ceramics, compounds of uranium, scrap processing solutions, residues, and waste stream products. It is not intended as a complete résumé dealing with the subject, but it does offer measurement methods believed to give precise and accurate results of a high order. Because of the monetary value of the materials, and the transfer activities from one installation to another, involving payments or credits, burn-up charges, use charges, etc., it is essential that such methods are used.

Methods of analysis to a large extent are dictated by the types of material to be analysed. The use of gravimetric methods are reviewed pertaining to product materials, which are generally defined as uranium metal, or compounds of the metal, such as oxides, halides, or nitrates. A pyro-hydrolysis technique is included under this heading. Non-volatile metallic impurities are determined spectroscopically, and the gravimetric results are corrected accordingly. Volumetric procedures, the "work horse" methods for determining uranium, are thoroughly explored. The technique is applicable to all types of material, providing the uranium available for measurement is present in milligram quantities. Due to the valence states of uranium, reduction-oxidation schemes are particularly attractive. Dissolution problems, separation of interfering elements, reduction steps, and oxidation titrations of reduced uranium are discussed.

The application of certain spectrophotometric and fluorometric procedures for analysing low-grade materials are included. Various separation steps incorporated in the procedures before the determination of uranium are reviewed. Along these lines the utilization of differential colorimetry is examined for determining uranium in impure material. Development of measurement methods in the fields of coulometry, gamma-ray absorptiometry, and X-ray analysis are discussed, with emphasis placed on the limits and applicability of these techniques.

Finally, the subject of standard reference materials is dealt with along with information regarding analytical evaluation programmes conducted in the United States for the purpose of obtaining better analytical data.

METHODES D'ANALYSE POUR LA MESURE DE CONCENTRATIONS D'URANIUM. Les auteurs donnent un aperçu des méthodes d'analyse utilisées aux États-Unis pour le dosage de l'uranium dans le cadre de la gestion des matières nucléaires. Ils exposent les méthodes appliquées pour le dosage de cet élément dans des matières très différentes: minerais, concentrés, uranium métal, alliages, céramiques, composés d'uranium, solutions pour le traitement des déchets métalliques, résidus et produits du circuit déchets. Le mémoire ne vise pas à donner un aperçu complet de la question, mais il présente des méthodes de mesure qui semblent devoir donner d'excellents résultats quant à leur précision et leur exactitude. Étant donné le prix des matières et leur transfert d'une installation à l'autre, ce qui implique des versements ou des avoirs, le règlement des quantités irradiées et des frais d'utilisation, etc., il est indispensable de recourir à ces méthodes.

Les méthodes d'analyse dépendent dans une large mesure du genre de matériau à analyser. Les auteurs passent en revue les méthodes gravimétriques appliquées aux produits finis, généralement constitués par de l'uranium métal ou des composés de ce métal, tels que les oxydes, les halogénures ou les nitrates. Un procédé de pyrohydrolyse est inclus dans ce groupe. Les impuretés métalliques non volatiles sont dosées par spectroscopie et les résultats gravimétriques rectifiés en conséquence. Les auteurs décrivent dans le détail les méthodes volumétriques, méthodes à grand rendement utilisées pour la détermination de l'uranium. Elles sont applicables à tous les types de matières, à condition que l'uranium à doser soit présent en quantités de l'ordre du milligramme. Étant donné les états de valence de l'uranium, les procédés par réduction et oxydation sont
АНАЛИТИЧЕСКИЕ МЕТОДЫ ИЗМЕРЕНИЙ КОНЦЕНТРАЦИИ УРАНА. Дается обзор аналитических методов определения урана, применяемых в соответствии с программой обращения с ядерными материалами в США. Описываются методы измерения элемента в различных материалах, т.е. в рудах, концентрах, металлургическом уране, сплавах, керамике, соединениях урана, растворах после обработки скрапа, остатках и продуктах отходов. Этот обзор не является полным резюме по данному вопросу, однако в нем описываются методы измерений, которые как полагают, дают точные и правильные результаты высокого порядка. Такие методы необходимо использовать ввиду стоимости материалов и их передачи из одной уставновки в другую, что связано с платежами или кредитами, стоимостью выгоревшего топлива, арендной платы и т.д.

Методы анализа в значительной степени определяются типами исследуемого материала. Рассматривается использование гравиметрических методов в отношении продуктов производства, которые обычно определяются как металлический уран или соединения этого металла, как, например, окислы, галоидные соединения или нитраты. Дается описание методов пиро-гидролиза. Спектроскопическим способом определяются нелетучие металлические примеси и соответственно корректируются гравиметрические результаты. Тщательно изучаются волнометрические процедуры, являющиеся основными методами определения урана. Этот метод применяется к материалам всех типов при условии, что подлежащей измерению уран присутствует в миллиграммовых количествах. Благодаря валентному состоянию урана восстановительно-окислительные схемы представляют особый интерес. Обсуждаются проблемы растворения, разделение интерферирующих элементов, ступени восстановления и окислительные титрования восстановленного урана.

Описывается также применение некоторых спектрофотометрических и флуориметрических процедур анализа низкосортных материалов. Рассматриваются различные ступени разделения в процедурах, предшествующих определению урана. В соответствии с этими принципами изучается использование дифференциальной калориметрии для определения урана в неоцищенных материалах. Обсуждаются разработка методов измерений в области кулон-метрии, гамма-абсорбционной, а также рентгеновского анализа, причем особое внимание обращается на ограничение и применимость этих методов.

Наконец, рассматривается вопрос об эталонах совместно с информацией относительно программ по анализической оценке, которые осуществляются в США с целью получения более точных аналитических данных.

METODOS ANALITICOS PARA DETERMINAR LA CONCENTRACION DEL URANIO. Los autores estudian los procedimientos analíticos de determinación del uranio empleados en los Estados Unidos por los servicios de administración de materiales nucleares. Describen sucintamente algunos métodos para la medición de dicho elemento en una gran variedad de materiales: minerales, concentrados, uranio metálico, aleaciones, productos cerámicos, compuestos, soluciones de tratamiento de residuos, productos de desecho. No pretenden dar un resumen exhaustivo de los métodos de medición, pero presentan algunos cuyos resultados son sumamente precisos. Esta condición es esencial, ya que los materiales de que se trata son muy costosos y se transfieren de una instalación a otra, lo que supone desembolsos, aperturas de crédito, gravámenes por grado de combustión, uso, etc.

Los métodos de análisis dependen de gran medida del tipo de material. En la memoria se examinan los métodos gravimétricos, empleados cuando se trata de materiales elaborados, generalmente uranio metálico y compuestos: óxidos, haluros, nitratos. Para estos materiales se emplea también una técnica pirohidrolítica. Las impurezas metálicas no volátiles se determinan por espectroscopía y los valores obtenidos sirven para corregir los resultados del análisis gravimétrico. La memoria trata detenidamente de los procedimientos volumétricos, que son los más empleados para la determinación del uranio. Esta técnica es aplicable a materiales de toda.
1. INTRODUCTION

Introductory remarks seem to be advisable before starting the discussion of uranium measurements as practiced in the United States of America. They will be limited to selected methods and techniques that have proved to be workable and capable of giving precise and, to the best of our knowledge, accurate measurements. For a more comprehensive study of uranium analyses, there have been numerous review articles and books on this subject [1-4], one of the most recent being Ref. [5].

The Nuclear Materials Management group of the USAEC utilize the services of the laboratory in the following five categories:

1. Analyses of source and special nuclear materials for accountability purposes;
2. Development, improvement, and evaluation of analytical methods relative to materials used in nuclear technology;
3. Co-ordination of work programmes designed for providing standard samples and recommending methods of analyses;
4. Umpire, or referee analysis; and
5. Training of NMM personnel in specialized analytical techniques.

The measurement of uranium is of major importance under all five points and covers the entire uranium field; therefore, the analyst is confronted with the analysis of ores, concentrates, product materials, fuel in the form of alloys or ceramics, waste stream products, uranium metal, and samples from enriched uranium recovery programmes. Because of the monetary value of these materials it is essential that only proven methods for uranium measurements be employed. Since the international exchange of enriched uranium is constantly increasing, it would be highly desirable if methods acceptable to all parties concerned could be agreed upon.

2. MEASUREMENT METHODS

2.1. Gravimetric methods

The first type of analysis to be dealt with is the gravimetric procedures. They are applicable for the measurement of uranium in the following materials: UO₂, UO₃, UF₄ and UF₆. The products are of such purity that chemical separations of impurities are not necessary. Non-volatile metal
impurities are determined spectroscopically and the final weight of U₃O₈ is corrected accordingly.

In the case of the oxides of uranium the weighed samples are generally pre-ignited at 500°C for 2 h, then ignited to constant weight at 900°C. A pyrohydrolysis technique is employed for the fluoride compounds of uranium. For UF₄ steam is passed over the sample for 1 h at 850°C, followed by an ignition to constant weight at 900°C. In assaying UF₅ a hydrolysis and an evaporation to dryness step necessarily precedes the pyrohydrolysis and ignition to constant weight, as directed for UF₄.

The chief problem with the gravimetric type of analysis concerns the temperature of ignition that will result in stoichiometric U₃O₈. From work performed at New Brunswick it appears that at sea level or thereabouts the temperature should be 850 to 900°C. Work at the Los Alamos Laboratory, which has an altitude of 7000 ft, indicates that a temperature of 750°C is more suitable. Temperatures in excess of those given result in an oxygen deficient compound, whereas temperatures lower than those listed yield oxides containing more oxygen than the theoretical for U₃O₈.

Of the various analytical approaches for the determination of uranium, the gravimetric procedures are more uniform in detailed instructions, and are generally accepted as correct procedures for product materials. This stems from the simplicity of the methods plus the high order of precision attained. Tables I and II illustrate typical six-month summaries of gravimetric results obtained from an evaluation programme routinely conducted by the USAEC. In addition, precision data for the gravimetric assays of these compounds are listed.

Because of the lack of standard reference samples, gathering accuracy data presents a problem. To obtain an estimate of accuracy we analysed several of the product materials both gravimetrically and by a precise volumetric method. Samples weighing 5-10 g were put into solution, converted to sulphate solutions, the uranium reduced by means of a Jones reductor, aerated to a uranium(IV) potentiometric endpoint, a slight excess of potassium dichromate was added over that theoretically required for oxidizing the uranium to the U(VI) state, and finally the excess dichromate was titrated potentiometrically with standard ferrous sulphate.

An example of work along this line is given for UF₄ in Table III. In addition the U₃O₈ residues obtained by the pyrohydrolysis-gravimetric procedure were analysed by the same high precision method, and the results are included in Table III.

2.2. Volumetric methods (visual indicator)

The use of volumetric procedures for the measurement of uranium are next considered. They are used more often than any other technique in the United States. By variations or modifications in the separation steps for interfering elements the methods are adaptable for macro determinations of uranium, regardless of the composition of the material.

The methods can be classified into three categories: (1) the reduction of uranium to the quadrivalent state and titrating with a standard oxidizing solution, (2) titration of uranyl salts or the bivalent radical UO₂⁺ with a precipitating agent, such as a standard phosphate, and (3) precipitation of a
TABLE I
TOTAL METAL IN ORANGE OXIDE
(Values in per cent)

<table>
<thead>
<tr>
<th>Month</th>
<th>Lab 1</th>
<th>Lab 2</th>
<th>Lab 3</th>
<th>Lab 4</th>
<th>Average</th>
<th>Standard deviation</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Production samples</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>November</td>
<td>82.48</td>
<td>82.50</td>
<td>82.46</td>
<td>82.48</td>
<td>82.48</td>
<td>0.02</td>
</tr>
<tr>
<td>December</td>
<td>81.96</td>
<td>81.95</td>
<td>81.90</td>
<td>81.92</td>
<td>81.91</td>
<td>0.07</td>
</tr>
<tr>
<td>January</td>
<td>82.57</td>
<td>82.60</td>
<td>82.56</td>
<td>82.57</td>
<td>82.58</td>
<td>0.02</td>
</tr>
<tr>
<td>February</td>
<td>82.33</td>
<td>82.32</td>
<td>82.32</td>
<td>82.32</td>
<td>82.32</td>
<td>0.01</td>
</tr>
<tr>
<td>March</td>
<td>82.29</td>
<td>82.29</td>
<td>82.27</td>
<td>82.28</td>
<td>82.28</td>
<td>0.01</td>
</tr>
<tr>
<td>April</td>
<td>82.48</td>
<td>82.51</td>
<td>82.46</td>
<td>82.44</td>
<td>82.47</td>
<td>0.03</td>
</tr>
<tr>
<td>Average</td>
<td>82.35</td>
<td>82.36</td>
<td>82.31</td>
<td>82.34</td>
<td>82.34</td>
<td></td>
</tr>
<tr>
<td>&quot;Known&quot; samples</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>November</td>
<td>82.46</td>
<td>82.47</td>
<td>82.43</td>
<td>82.44</td>
<td></td>
<td></td>
</tr>
<tr>
<td>December</td>
<td>82.44</td>
<td>82.46</td>
<td>82.42</td>
<td>82.42</td>
<td></td>
<td></td>
</tr>
<tr>
<td>January</td>
<td>82.42</td>
<td>82.43</td>
<td>82.39</td>
<td>82.40</td>
<td></td>
<td></td>
</tr>
<tr>
<td>February</td>
<td>82.43</td>
<td>82.42</td>
<td>82.46</td>
<td>82.41</td>
<td></td>
<td></td>
</tr>
<tr>
<td>March</td>
<td>82.43</td>
<td>82.44</td>
<td>82.40</td>
<td>82.42</td>
<td></td>
<td></td>
</tr>
<tr>
<td>April</td>
<td>82.45</td>
<td>82.44</td>
<td>82.43</td>
<td>82.42</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Average</td>
<td>82.44</td>
<td>82.44</td>
<td>82.42</td>
<td>82.42</td>
<td>Overall average: 82.43</td>
<td></td>
</tr>
<tr>
<td>Standard deviation</td>
<td>0.01</td>
<td>0.02</td>
<td>0.03</td>
<td>0.01</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

uranium compound, such as the 8-hydroxyquinoline derivative and titration of the precipitating agent that is carried down. The last two categories are rarely, if ever, employed in the United States.

The reduction of uranium to the U(IV) oxidation state followed by a titration to the U(VI) state is by far the most acceptable and will be the only one discussed. The oxidation potentials of the principal valence states of uranium should be borne in mind. They are U(III), U(IV) and U(VI). Because of the relatively low oxidation potential of the U(IV)-U(VI) couple, the element can be quantitatively converted to either state at will. The alternate use of both valences permits many separations that would be otherwise impossible. Titrations usually involve a pre-reduction to U(IV) followed by an oxidation titration to U(VI).

There are four major steps to be considered using these methods, they are: (1) dissolution of sample, (2) separation steps for the removal of
INTERFERING ELEMENTS, (3) REDUCTION OF URANIUM, AND (4) TITRATION OF REDUCED URANIUM WITH A STANDARD OXIDANT.

2.2.1. DISSOLUTION [6]

The dissolution of sample material is the first concern. The method of attack is dependent on the nature of the material. This varies from watersoluble uranium compounds to extremely complex uranium fuels.

Probably the most common reagent for the dissolution of uranium metal and many of its alloys, as well as concentrates, is nitric acid. Zirconium and niobium are sometimes added to uranium to improve its irradiation- and corrosion-resistant properties. Nitric acid or aqua regia readily reacts with uranium, but does not dissolve zirconium or niobium. This results in leaving a finely divided residue that has been known to react explosively. The addition of hydrofluoric acid or a soluble fluoride to a nitric acid dissolution

<table>
<thead>
<tr>
<th>Month</th>
<th>Lab 1</th>
<th>Lab 2</th>
<th>Lab 3</th>
<th>Average</th>
<th>Standard deviation</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Production samples</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>November</td>
<td>76.43</td>
<td>76.40</td>
<td>76.45</td>
<td>76.43</td>
<td>0.03</td>
</tr>
<tr>
<td>December</td>
<td>76.16</td>
<td>76.15</td>
<td>76.19</td>
<td>76.17</td>
<td>0.02</td>
</tr>
<tr>
<td>January</td>
<td>75.98</td>
<td>75.96</td>
<td>76.03</td>
<td>75.99</td>
<td>0.04</td>
</tr>
<tr>
<td>February</td>
<td>75.99</td>
<td>76.03</td>
<td>76.05</td>
<td>76.02</td>
<td>0.03</td>
</tr>
<tr>
<td>March</td>
<td>75.96</td>
<td>75.99</td>
<td>76.04</td>
<td>76.00</td>
<td>0.04</td>
</tr>
<tr>
<td>April</td>
<td>76.00</td>
<td>75.97</td>
<td>75.99</td>
<td>75.99</td>
<td>0.02</td>
</tr>
<tr>
<td><strong>Average</strong></td>
<td>76.09</td>
<td>76.08</td>
<td>76.12</td>
<td>76.10</td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Month</th>
<th>Lab 1</th>
<th>Lab 2</th>
<th>Lab 3</th>
<th>Average</th>
<th>Standard deviation</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>&quot;Known&quot; samples</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>November</td>
<td>75.92</td>
<td>75.92</td>
<td>75.99</td>
<td></td>
<td></td>
</tr>
<tr>
<td>December</td>
<td>75.93</td>
<td>75.94</td>
<td>75.95</td>
<td></td>
<td></td>
</tr>
<tr>
<td>January</td>
<td>75.93</td>
<td>75.94</td>
<td>75.95</td>
<td></td>
<td></td>
</tr>
<tr>
<td>February</td>
<td>75.94</td>
<td>75.93</td>
<td>75.95</td>
<td></td>
<td></td>
</tr>
<tr>
<td>March</td>
<td>75.92</td>
<td>75.93</td>
<td>75.96</td>
<td></td>
<td></td>
</tr>
<tr>
<td>April</td>
<td>75.93</td>
<td>75.93</td>
<td>75.95</td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>Average</strong></td>
<td>75.93</td>
<td>75.93</td>
<td>75.96</td>
<td>Overall average: 75.94</td>
<td></td>
</tr>
<tr>
<td><strong>Standard deviation</strong></td>
<td>0.01</td>
<td>0.01</td>
<td>0.02</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
**TABLE III**

**URANIUM CONTENT OF UF$_4$**
(Values in per cent)

<table>
<thead>
<tr>
<th>Pyrohydrolysis: gravimetric method (2-g sample)</th>
<th>New Brunswick high precision method (5 to 10-g sample)</th>
</tr>
</thead>
<tbody>
<tr>
<td>75.88</td>
<td>75.877</td>
</tr>
<tr>
<td>75.86</td>
<td>75.878</td>
</tr>
<tr>
<td>75.87</td>
<td>75.866</td>
</tr>
<tr>
<td>75.86</td>
<td>75.87</td>
</tr>
<tr>
<td>75.86</td>
<td>75.868</td>
</tr>
<tr>
<td><strong>Average 75.86</strong></td>
<td><strong>Average 75.874</strong></td>
</tr>
</tbody>
</table>

**U residue from pyrohydrolysis analysed by precision method**

<table>
<thead>
<tr>
<th>U in U$_3$O$_8$ residue</th>
<th>U$_3$O$_8$ (calculated)</th>
</tr>
</thead>
<tbody>
<tr>
<td>84.797</td>
<td>99.995</td>
</tr>
<tr>
<td>84.804</td>
<td>100.003</td>
</tr>
<tr>
<td>84.784</td>
<td>99.979</td>
</tr>
<tr>
<td><strong>Average 84.795</strong></td>
<td><strong>99.992</strong></td>
</tr>
</tbody>
</table>

of Zircaloy prevents the accumulation of these residues and eliminates the hazards with nitric acid.

Hydrochloric acid reacts very rapidly with uranium and some of the product material to form the uranium(IV) chloride and a black oxide residue that has been described as a hydrated oxide of U(III) and U(IV). The common practice when using this reagent is to add an oxidizing agent only after the rapid reaction with HCl ceases, such as sodium chlorate, hydrogen peroxide, or bromine.

Aqua regia, as mentioned previously, is very often used in dissolution problems. It is particularly effective with uranium-stainless steels, and uranium alloyed with platinum metals. Stainless steel-UO$_2$ fuels very often contain 1-3% silicon. In such cases we add hydrofluoric and perchloric acids to the solution after decomposition with aqua regia, and evaporate the solution to strong fumes of HClO$_4$. By so doing we are able to volatilize the major portion of the silica and remove the aqua regia all in one operation.

Uranium metal, some of its alloys and compounds, dissolve very readily and sometimes quite spectacularly in hot perchloric acid (70%). We do not recommend its use except under close supervision and access to fume hoods that are designed for this acid.
Sulphuric acid does not react with uranium materials at moderate temperatures or concentrations. It does play a very important role in several volumetric methods in that, once dissolution of the material has been accomplished by other means, the solutions are converted to a sulphate by the addition of H₂SO₄ and evaporating to strong SO₃ fumes for subsequent separation steps.

Alkali and acid fusions are very often resorted to in breaking up materials that resist attack by acid treatments. It is a common practice when dealing with material of unknown composition first to treat the sample with an acid or combination of acids and then, if a residue remains, to fuse the insoluble material with an alkali after filtration and ignition of the unreacted material.

2.2.2. Interfering elements

The second major step in volumetric procedures is the elimination of interfering elements. Since there is a reduction of U(VI) to U(IV) preceding the titration with a standard oxidant, it is imperative to eliminate other elements that are reduced and interfere by also consuming the oxidant. Examples of such elements are iron, titanium, vanadium, and niobium. Other elements, such as arsenic, antimony, molybdenum, copper, cadmium, and platinum tend to plate out on the reductor and consequently to interfere in the complete reduction of the uranium. It would be a time-consuming task to give details regarding the numerous techniques that are employed to accomplish the separation of these metals. They include precipitation, solvent extraction, complexing and ion-exchange methods. The discussion will be limited to three steps that have proved to be acceptable, regardless of the composition of the material.

The first of these is to precipitate the second group metals as insoluble sulphides. After converting the dissolution sample, as previously described, to a sulphate solution it is saturated with hydrogen sulphide. The precipitated sulphides are separated by filtration. H₂S gas or the use of thioacetamide may be used. The latter breaks down to hydrogen sulphide as one of its by-products when introduced to a solution of prescribed pH and volume concentration. If the analyst has assurance of the absence of group (II) metals in the test material the sulphide step can and should be omitted.

To continue with the analysis, the filtrate from the sulphide precipitation is boiled to free it from excess H₂S. Sulphuric acid is added to bring the concentration to approximately 10%. After cooling to 5°C potassium permanganate is added drop-wise until the solution remains pink upon stirring, indicating that the uranium is present as uranium(VI). The solution is transferred to a separatory funnel, a cupferron solution is added. After shaking, the metal cupferrates are extracted with chloroform and the organic layer is drained off. The extraction of the aqueous phase with fresh portions of chloroform is repeated until the organic extract is colourless. The aqueous layer is returned to the original beaker and evaporated to strong SO₃ fumes. Nitric and perchloric acids are added and the fuming step is repeated several times for the complete removal of cupferron decomposition products. This is necessary since nitrates and nitriles are reduced to hydroxylamine and other compounds that consume the oxidant in the titration step.
In the case of samples containing macro quantities of iron, e.g. stainless steel-UO₂ fuels, the cupferron treatment for its removal would be unsatisfactory. In such cases mercury-cathode electrolysis is particularly effective for eliminating large amounts of iron, chromium, nickel and the second group metals from a 5% sulphuric acid uranium solution. A commercial mercury-cathode electrolysis apparatus featuring a moving body of mercury, which decreases the time required for the electrolysis to one hour or less, is available. Unfortunately, from our experience its use does not eliminate the last traces of iron and we have found it necessary to follow up such a separation with a cupferron-chloroform extraction.

Before leaving the separation techniques, the use of ion-exchange methods should be mentioned as they are being employed by many laboratories. Generally, uranium as a sulphate complex is separated as an anion from a great number of other metallic ions by its absorption on a strong base anion-exchange resin such as Dowex-1 or Amberlite LRA-400.

2.2.3. Reduction of U(VI) to U(IV)

The reduction of uranium in the purified test solution is the next major step to consider. In the United States this is usually accomplished with amalgamated zinc or lead. Occasionally chromous or titaneous ion, or electrolytic methods are used. With zinc or electrolytic reductions a mixture of trivalent and tetravalent uranium is obtained; however, the U(III) is easily oxidized to the U(IV) state by a simple aeration step. It is of interest to note that with lead it has been stated that no trivalent uranium is present, and HCl solutions can be used in the reductor. Sulphate cannot be used because a film of lead sulphate is formed over the reagent, which inhibits the reduction of uranium.

In most instances, our laboratories employ the Jones reductor. This is largely a matter of choice since both zinc and lead are capable of giving excellent performances.

Figure 1 is a sketch of the Jones reductor used by us. The tube is connected to a vacuum source through a side arm equipped with a three-way stopcock. This regulates the rate at which the solution and washings are passed through the reductor. Usually a 3% zinc amalgam is used, although there may be instances where amalgams as high as 10% are preferable. As previously mentioned, a 15-min aeration period of the reduced solution must be carried out following the reduction with zinc.

2.2.4. Titration U(IV) to U(VI)

The final step in the volumetric procedure is the titration of U(IV) to U(VI). This is accomplished by one of several oxidants using either an internal indicator or a potentiometric endpoint. The oxidants most preferred by us are potassium dichromate or ceric sulphate, the choice again seems to be one of personal preference. Potassium dichromate has the advantage of being obtainable as a very pure salt and is considered a primary standard in volumetric work. The salt is water soluble and its solution is exceptionally stable. The preparation of ceric sulphate solution is troublesome because of solubility problems. The effect of the indicator (sodium diphenylamine
sulphonate) used with dichromate is more pronounced than the indicator (ferroin) used with ceric sulphate. This will be enlarged upon later in the discussion. In using potassium dichromate as the oxidant the reduced uranium(IV) solution is treated with a 4% solution of ferric chloride, followed by a phosphoric-sulphuric acid mixture and 8 drops of sodium diphenylamine sulphonate indicator before titrating with 0.027 N potassium dichromate. The reaction of tetravalent uranium with dichromate is slow and sluggish, but ferric iron reacts very readily with uranium(IV) to form ferrous iron and hexavalent uranium.

$$\text{UO}_2^+ + 2\text{Fe}^{3+} + \text{H}_2\text{O} \rightarrow \text{UO}_2^{2+} + 2\text{Fe} + 2\text{H}^+$$

In turn, the ferrous iron is oxidized rapidly and quantitatively by dichromate. The purpose of the addition of phosphate is for complexing the unreacted ferric as well as the ferric iron formed by the titration. As mentioned, the oxidant is 0.027 N and 1 ml is equivalent to 3.75 mg U₃O₈. We attempt to base our sample weights so that 40–100 ml of dichromate solution will be required to complete the titration.
It was pointed out that a standardization of the dichromate with National Bureau of Standards \( \text{U}_3\text{O}_8 \) is required for establishing the uranium titre of the solution. When a volume of 40-100 ml of dichromate is used, an indicator titration blank correction is applied. This is determined by titrating 190 mg of standard \( \text{U}_3\text{O}_8 \) and then titrating double this amount. The titration blank is calculated as twice the volume used for the first titration minus the volume used for the second titration. The volume of dichromate is corrected for this blank both in the standard and in the unknown. The correction is necessary since reduction-oxidation indicators of this type consume minute amounts of the oxidant.

The disturbing factor with the indicator when less than 40 ml of dichromate is required, stems from the fact that the uranium titre varies with the volume of dichromate used. The variations are tied in with the indicator since no similar effect is noted with potentiometric endpoints. The action of the indicator is described as an irreversible oxidation of the diphenylamine to diphenyl benzidine. After the addition of the dichromate is started, a part of the diphenyl benzidine can be further oxidized before all the amine is converted. The reaction occurs only in the presence of ferrous iron and is dependent on the amount of ferrous iron titrated, which in turn is dependent on the uranium concentration of the sample. Therefore, when less than 40 ml of 0.027 N potassium dichromate is used the uranium titre is obtained by titrating essentially the same weight of standard \( \text{U}_3\text{O}_8 \).

2.2.5. Results (precision and accuracy)

In regard to the precision and accuracy of the volumetric methods recent analytical studies have been made for evaluating the same in connection with the publication of "Selected Measurement Methods for Plutonium and Uranium in the Nuclear Fuel Cycle" [3] compiled by the US Nuclear Materials Management group.

Due to the lack of standard reference materials accurate information is difficult to obtain. Due to this handicap we were directed to prepare and circulate to a number of co-operating laboratories, synthetic solutions corresponding to the composition of many of these materials. The National Bureau of Standards black oxide (\( \text{U}_3\text{O}_8 \)) served as the source of the uranium. In turn uranium values were assigned to each preparation. From the analytical data gathered by this means it was possible to make statistical evaluations. Examples of these evaluations are given in Tables IV to VI.

2.3. Volumetric methods (potentiometric)

There has been a considerable effort carried out in recent years for employing potentiometric techniques to detect endpoints in uranium volumetric procedures. In such cases the potential developed by the substance to be determined contained in a suitable galvanic cell is utilized to measure the quantity of substance present. An excellent review of this work is given in Ref. [5]. Various oxidants, temperature conditions, electrode systems, and measurement equipment are discussed. Similar preliminary dissolution, separation of interfering elements, and reduction steps are required to those used for visual methods.
TABLE IV

VOLUMETRIC POTASSIUM DICHLOROMATE DETERMINATION OF URANIUM IN URANIUM NITRATE SOLUTION

<table>
<thead>
<tr>
<th>Laboratory</th>
<th>Reported values (g U/g soin)</th>
<th>Average</th>
<th>Coefficient of variation (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>0.055368 0.055192</td>
<td>0.055280</td>
<td>0.22</td>
</tr>
<tr>
<td>W</td>
<td>0.055175 0.055315</td>
<td>0.055245</td>
<td>0.18</td>
</tr>
<tr>
<td>R</td>
<td>0.055247 0.055228</td>
<td>0.055238</td>
<td>0.02</td>
</tr>
<tr>
<td>LA</td>
<td>0.05528 0.05519 0.05522</td>
<td>0.05523</td>
<td>0.08</td>
</tr>
<tr>
<td>B</td>
<td>0.055180 0.055206 0.055186</td>
<td>0.055191</td>
<td>0.02</td>
</tr>
<tr>
<td>D</td>
<td>0.05504 0.05499 0.05497</td>
<td>0.05500</td>
<td>0.07</td>
</tr>
<tr>
<td>VB</td>
<td>0.05484 0.05513 0.05494</td>
<td>0.05497</td>
<td>0.27</td>
</tr>
<tr>
<td>Z</td>
<td>0.054804 0.054856 0.054929</td>
<td>0.054863</td>
<td>0.11</td>
</tr>
<tr>
<td>F</td>
<td>0.05466 0.05472 0.05465</td>
<td>0.05478</td>
<td>0.28</td>
</tr>
<tr>
<td>E</td>
<td>0.05467 0.05475</td>
<td>0.05471</td>
<td>0.12</td>
</tr>
<tr>
<td>U</td>
<td>0.05472 0.05466</td>
<td>0.05469</td>
<td>0.08</td>
</tr>
<tr>
<td>CA</td>
<td>0.054110 0.054099</td>
<td>0.054104</td>
<td>0.01</td>
</tr>
</tbody>
</table>

Standard deviation within laboratories: 0.000087
Coefficient of variation within laboratories: 0.16%
Standard deviation between laboratory averages (s): 0.00034
Coefficient of variation between laboratory averages (v): 0.62%
Overall average of laboratory averages (X): 0.054968
Prepared value: 0.055106
Difference ±95% confidence limits: -0.000138 ± 0.000207
Relative difference ±95% confidence limits (d): -0.3 ± 0.4%

Summary: Number of laboratories (n): 12
Average (X): 0.05497
Precision (s): 0.00034
Accuracy (v): 0.6%

* Each laboratory was requested to report at least one more figure than usual
** Coefficient of variation (%) is 100 × standard deviation per average

For specific types of material, potentiometric methods have been successfully developed. As a general purpose technique difficulty has been encountered. In many instances, elevated temperature conditions required
TABLE V
VOLUMETRIC POTASSIUM DICHROMATE DETERMINATION OF URANIUM IN URANIUM-ALUMINIUM ALLOYS

<table>
<thead>
<tr>
<th>Laboratory</th>
<th>Reported values (g U/g soin)*</th>
<th>Average</th>
<th>Coefficient of variation (%)**</th>
</tr>
</thead>
<tbody>
<tr>
<td>E</td>
<td>0.00798 0.00800</td>
<td>0.00799</td>
<td>0.18</td>
</tr>
<tr>
<td>V8</td>
<td>0.00795 0.00797</td>
<td>0.00796</td>
<td>0.18</td>
</tr>
<tr>
<td>F</td>
<td>0.007969 0.007950</td>
<td>0.007960</td>
<td>0.17</td>
</tr>
<tr>
<td>D</td>
<td>0.007928 0.007943</td>
<td>0.007934</td>
<td>0.15</td>
</tr>
<tr>
<td>Z</td>
<td>0.007908 0.007920 0.007916</td>
<td>0.007915</td>
<td>0.08</td>
</tr>
<tr>
<td>B</td>
<td>0.007932 0.007892</td>
<td>0.007912</td>
<td>0.35</td>
</tr>
<tr>
<td>R</td>
<td>0.007910 0.007913</td>
<td>0.007912</td>
<td>0.03</td>
</tr>
<tr>
<td>CA</td>
<td>0.007912 0.007912 0.007908</td>
<td>0.007911</td>
<td>0.03</td>
</tr>
<tr>
<td>U</td>
<td>0.00787 0.00793</td>
<td>0.00790</td>
<td>0.54</td>
</tr>
<tr>
<td>LA</td>
<td>0.007885 0.007888 0.007885</td>
<td>0.007886</td>
<td>0.02</td>
</tr>
<tr>
<td>A</td>
<td>0.0078814 0.0078863</td>
<td>0.007884</td>
<td>0.04</td>
</tr>
</tbody>
</table>

Standard deviation within laboratories: 0.000017, 0.21%
Coefficient of variation within laboratories: 0.000033, 0.42%
Standard deviation of laboratory averages (s): 0.007924
Coefficient of variation of laboratory averages (v): ±0.000028 ± 0.42%
Overall average of laboratory averages (X): 0.007924
Prepared value: 0.007896
Difference ± 95% confidence limits: ±0.000028 ± 0.000022
Relative difference ± 95% confidence limits (d): ±0.35 ± 0.28%

Summary: Number of laboratories (n): 11
Average (X): 0.007924
Precision (s): 0.000028
Accuracy (d): ±0.35 ± 0.28%

* Each laboratory was requested to report at least one more figure than usual
** Coefficient of variation (%) is 100 x standard deviation per average

for the titration and the care required for the prevention of electrode poisoning have presented major problems.

The Oak Ridge National Laboratory developed a high temperature potentiometric method for the routine determination of uranium in U-Al alloys [3]. A uranyl sulphate solution of the sample is prepared, and the uranium is
### Table VI

**VOLUMETRIC POTASSIUM DICHROMATE DETERMINATION OF URANIUM IN URANIUM-STAINLESS STEEL ALLOY**

<table>
<thead>
<tr>
<th>Laboratory</th>
<th>Reported values (g U/g soln)*</th>
<th>Average</th>
<th>Coefficient of variation (%)* **</th>
</tr>
</thead>
<tbody>
<tr>
<td>W</td>
<td>0.011145 0.011156</td>
<td>0.011150</td>
<td>0.07</td>
</tr>
<tr>
<td>R</td>
<td>0.011141 0.011150</td>
<td>0.011146</td>
<td>0.05</td>
</tr>
<tr>
<td>LA</td>
<td>0.01109 0.01105 0.01110</td>
<td>0.01108</td>
<td>0.23</td>
</tr>
<tr>
<td>VB</td>
<td>0.01107 0.01105</td>
<td>0.01106</td>
<td>0.13</td>
</tr>
<tr>
<td>F</td>
<td>0.01104 0.01102 0.01109</td>
<td>0.01105</td>
<td>0.29</td>
</tr>
<tr>
<td>Z</td>
<td>0.011026 0.011080 0.011036</td>
<td>0.011047</td>
<td>0.26</td>
</tr>
<tr>
<td>B</td>
<td>0.011100 0.010986 0.010998</td>
<td>0.011028</td>
<td>0.57</td>
</tr>
<tr>
<td>CA</td>
<td>0.011013 0.010998</td>
<td>0.011006</td>
<td>0.10</td>
</tr>
<tr>
<td>A</td>
<td>0.010977 0.011018</td>
<td>0.010998</td>
<td>0.26</td>
</tr>
<tr>
<td>M</td>
<td>0.01084 0.01085</td>
<td>0.01084</td>
<td>0.06</td>
</tr>
<tr>
<td>U</td>
<td>0.01081 0.01083</td>
<td>0.01082</td>
<td>0.13</td>
</tr>
</tbody>
</table>

Standard deviation within laboratories: 0.000031
Coefficient of variation within laboratories: 0.28%
Standard deviation between laboratory averages (s): 0.00011
Coefficient of variation between laboratory averages (v): 1.0%
Overall average of laboratory averages (X): 0.011009
Prepared value: 0.01115
Difference ± 95% confidence limits: -0.000146 ± 0.000070
Relative difference ± 95% confidence limits (d): -1.3 ± 0.6%

**Summary**
- Number of laboratories (n): 11
- Average (X): 0.01101
- Precision (s): 0.00011
- (v): 1.0%
- Accuracy (d): -1.3 ± 0.6%

* Each laboratory was requested to report at least one more figure than usual
** Coefficient of variation (%) is 100 x standard deviation per average

Reduced to U(IV) with Cr(II). Excess Cr(II) is titrated with standard ferric sulphate solution to the first potentiometric endpoint. (The point of maximum potentialal change per unit volume of titrant.) The U(IV) is then titrated to U(VI) with standard ferric sulphate solution. Oxidation of all the U(IV) to U(VI) is indicated by the second potentiometric endpoint (the
FIG. 2. Reaction cell

endpoint will be approximately 300 mV). Titration under an atmosphere of carbon dioxide minimizes the possibility of air oxidation of the U(IV). The coefficient of variation for a single determination for this method is 0.1%. Figure 2 is a diagram of the reaction cell employed in the procedure.

In regard to the various oxidants used in volumetric analysis it is desirable to standardize them with compounds of known uranium content. In the United States a black oxide (U₃O₈) identified as the National Bureau of Standards #950, is used nationally for this purpose. At the present time there is no international standard for uranium analytical purposes. It is a problem that the IAEA has been confronted with for some time and if solved it will be a step of major importance.

The use of uranium metal for a standard has been proposed by some of our leading authorities. At one time, uranium metal was produced in the United States that contained less than 200 ppm of metal impurities. Due to its purity, the homogeneity of the metal, and the fact that its use would not involve stoichiometric problems, this would appear to make it a better standard than our present use of U₃O₈. Unfortunately, this metal is no longer produced since it was found that the presence of certain metal impurities were desirable from a metallurgical viewpoint. It is possible that
other nations are currently producing similar high purity uranium that could be utilized by the international agency in their standard studies for uranium measurements.

2.4. Other uranium methods and techniques

Certain other methods for the determination of uranium either as a major constituent or a minor constituent are dependent on the separation of the uranium before the determination. The methods most commonly used after separation employ spectrophotometric and fluorometric procedures for the final determination.

The methods of separation are quite numerous but generally consist of a solvent extraction procedure or one employing ion-exchange resins. The methods of separation are covered in Ref. [5].

2.4.1. Spectrophotometric methods

The spectrophotometric methods usually employ one of the following reagents:

<table>
<thead>
<tr>
<th>Reagent</th>
<th>Molar absorbancy index</th>
</tr>
</thead>
<tbody>
<tr>
<td>Peroxide</td>
<td>900 - 1200</td>
</tr>
<tr>
<td>Diethyldithiocarbamate</td>
<td>4050</td>
</tr>
<tr>
<td>Thiocyanate, aqueous</td>
<td>3770</td>
</tr>
<tr>
<td>Thiocyanate, aqueous acetone</td>
<td>3900</td>
</tr>
<tr>
<td>Thiocyanate, butyl cellusolve</td>
<td>5500</td>
</tr>
<tr>
<td>Oxine</td>
<td>3000 - 6500</td>
</tr>
<tr>
<td>Thoronol</td>
<td>16 400</td>
</tr>
<tr>
<td>Dibenzoylmethane</td>
<td>19 000</td>
</tr>
<tr>
<td>Arsenazo-III (U-IV)</td>
<td>100 000</td>
</tr>
<tr>
<td>1-(2 Pyridylazo) 2-naphthol</td>
<td>23 000</td>
</tr>
</tbody>
</table>

Of these reagents the ones used to the greatest extent are basic peroxide, dibenzoylmethane and arsenazo III. For waste stream products and residues a direct spectrophotometric determination after separation of the uranium is employed while for the determination of major amounts of uranium a differential spectrophotometric method is applied. Indicative of the differential technique is a procedure as used by the Goodyear Atomic Corporation for the determination of uranium in scrap dissolver solutions.

The analysis consists of a separation of uranium by ion exchange and then the determination of the separated uranium by a differential colorimetric procedure, using the colour developed by the basic peroxide method. Aliquots of the sample containing 30 to 60 mg of uranium are converted to the nitrate form and the uranium is absorbed on an anion-exchange resin (Dowex 1X-8) from a concentrated aluminium nitrate medium. This separates the uranium from most interferences except thorium and zirconium, which are removed by washing the resin with strong hydrochloric acid. The uranium is then eluted from the resin with very dilute nitric acid.
The aliquot containing the purified uranium is treated with sodium hydroxide and hydrogen peroxide to develop the yellow peruranate complex. A weighed aliquot of a standard solution containing about the same amount of uranium as the sample is treated in a similar manner. The uranium is determined by measuring the small differences between the optical densities of the standard and unknown solutions using the standard techniques of differential colorimetry. High concentrations of sulphate (more than 2 M) and phosphate (more than 1 M) interfere with the separation, but they can be removed by suitable preliminary separations. The uranium is determined by this procedure within 0.1 to 0.2 mg of the quantity originally taken, so that the precision of the method is about ±0.5%.

2.4.2. Fluorometric methods

Fluorescent methods for the determination of uranium are only applied when trace amounts of uranium are present and where a not too precise value is needed. In most instances a fusion method employing a mixture of sodium fluoride and lithium fluoride or a mixture of sodium fluoride, sodium carbonate and potassium carbonate are used. The choice of flux appears to be optional. The precision varies widely and in certain respects depends on the operator. While coefficients of variations have been reported that range from 0.7 to 20%, our experience has shown a coefficient of about 5% with the sodium carbonate, potassium carbonate-sodium fluoride flux.

2.4.3. Coulometric method

Coulometry, in which one measures the number of coulombs passing through an electrode during a titration, has been used to a considerable extent for the determination of uranium. Controlled potential coulometry has been used for irradiated dissolver solutions, after a separation from interfering elements, and directly on solution of alloys such as uranium-aluminium. Certain impurities can be tolerated. The sample size is small, usually 2 to 10 mg, and a precision of 0.1 to 0.5% is usually attained. The method is rapid and can be recommended where applicable.

2.4.4. X-ray spectrography or fluorescent methods

Another rapid method for the determination of uranium is one employing X-ray spectrography or fluorescent methods. The method is applied to both solids and solutions. It can be used for determining the homogeneity of fuel plates. The accuracy of the method is a function of the quality of the standards used for calibration, which should be similar to the sample, and the reproducibility of the sample prepared. In general the precision is comparable to that with wet chemical methods and is dependent on the counting statistics. The minimum amount of uranium measured is 0.05% with a range to 20%. Solution methods are generally to be preferred to the ones employing solids. In routine control work as many as 60 analyses per 8-h day have been performed with an accuracy of ±1%.
REFERENCES


DISCUSSION

J.T. BYRNE: From Tables I and IV of your report it appears that the gravimetric determination of uranium in "orange oxide" is about 10 times more precise (coefficient of variation < 0.06%) than the volumetric determination of uranium in uranyl nitrate solutions (coefficient of variation = 0.62%). Is this an inherent difference in the two methods themselves?

J.P. HIGHFILL: If you examine Table IV you will note that eleven laboratories co-operated in this evaluation programme. Examination of the individual results indicates that some laboratories were much more adept with the volumetric methods than others. Actually, at New Brunswick Laboratory (NBL) the precision of data gathered by the volumetric methods compares very favourably with that of data obtained gravimetrically.

J. ARTAUD: In connection with uranium gravimetry, you mention differences between NBL and Los Alamos for the oxide stoichiometry. Can you give figures for these differences? This could be of interest to foreign users of the standard oxide NBX 950.

J.P. HIGHFILL: I did not intend to imply that there was a difference between NBL and Los Alamos in regard to U₃O₈ results obtained gravimetrically. I stated that at sea level or thereabouts a temperature of 850-900°C was used for obtaining stoichiometric U₃O₈, while at Los Alamos (altitude 700 ft) a temperature of 750°C was more suitable.
METHODS FOR THE DETERMINATION OF PLUTONIUM IN SPENT REACTOR FUELS, PLUTONIUM METAL, ALLOYS AND COMPOUNDS

C.F. METZ AND G.R. WATERBURY
LOS ALAMOS SCIENTIFIC LABORATORY, LOS ALAMOS, N. MEX., UNITED STATES OF AMERICA

Abstract — Résumé — Аннотация — Resumen

METHODS FOR THE DETERMINATION OF PLUTONIUM IN SPENT REACTOR FUELS, PLUTONIUM METAL, ALLOYS AND COMPOUNDS. The monetary value, as well as the radiological and nuclear hazards of plutonium, and the necessity for monitoring recovery processes for their efficiency all contribute to the necessity for measuring the plutonium content of plutonium-containing materials essentially from the time the element is produced, and continue as long as it exists. Unique problems encountered in dissolver solutions such as the dangerously high radioactivity levels and the chemical complexity have influenced the methods for separating the plutonium before it can be determined by the radiochemical techniques designed for this purpose. The final measurement usually involves alpha counting. Methods of chemical separation of plutonium from uranium and fission products and final measurement of the plutonium are discussed. Problems and methods of handling intensely radioactive solutions also are discussed.

Plutonium, whether it be in the form of metal, solid compounds, alloys (for future power reactor use) or solutions, may be measured by one of several accurate and precise chemical methods. These include potentiometric methods in which Pu(III) is quantitatively oxidized to the (IV) state of oxidation, with the completeness of the reaction being indicated either potentiometrically or photoelectrically, amperometric titration of Pu(VI) with Fe(II) to an amperometric endpoint, constant-current and constant-potential coulometric titration techniques, potentiometric titration of excess Fe(II) following reduction of Pu(VI) to (IV) and spectrophotometric methods. These are discussed from the standpoint of accuracy and precision and also from the standpoint of application to various materials. Experience in the use of applications of ion exchange separations of plutonium from interfering elements are included for the applications of the above methods to plutonium alloys.

Applications of the sealed-tube technique to the dissolution of difficult soluble refractory plutonium materials are discussed with specific applications to refractory oxides and alloys. Analytical techniques required for the characterization of high-purity plutonium metal are reviewed. These apply to the analysis of plutonium metal having a total impurity content of less than 200 ppm. Methods for non-metallic, interstitial, and spectrographic (metallic) impurities are included.

MODES DE DOSAGE DU PLUTONIUM DANS LE COMBUSTIBLE IRRADIE, LE PLUTONIUM METALLIQUE, LES ALLIAGES ET LES COMPOSES. Etant donné la valeur marchande du plutonium, ainsi que les risques radiologiques et nucléaires qu'il implique et la nécessité de contrôler le rendement des méthodes de récupération, il est nécessaire de doser la teneur des matières qui en contiennent, à partir du moment où le plutonium est produit et pendant toute la durée de son utilisation. L'utilisation des solvants pose des problèmes extrêmement ardu, notamment à cause de la très dangereuse radioactivité et de la complexité chimique; c'est pourquoi on préfère séparer le plutonium avant qu'il puisse être dosé par les méthodes radiochimiques conçues à cette fin. La mesure finale implique généralement le comptage alpha. Les auteurs écrivent les méthodes chimiques utilisées pour séparer le plutonium de l'uranium et des produits de fission, ainsi que le dosage du plutonium, et précisent comment on peut résoudre les problèmes posés par la manipulation des solutions de très haute activité.

Le plutonium, sous forme de métal, de composés solides, d'alliages (pour réacteurs de puissance) ou de solutions, peut être dosé par l'une des méthodes exactes et précises qui ont été mises au point. On peut citer les méthodes potentiométriques qui font intervenir l'oxydation quantitative de Pu(III) de manière à obtenir Pu(IV), l'achèvement de la réaction étant indiqué par des mesures potentiométriques ou photoélectriques;

* Sponsored by the United States Atomic Energy Commission.
le titrage amperométrique de Pu(VI) avec Fe(II) jusqu'à une limite amperométrique, les méthodes de titrage coulombométriques à courant constant et à potentiel constant; le titrage potentiométrique de l'excédent de Fe(II) après réduction de Pu(VI) en Pu(IV); enfin les méthodes spectrophotométriques. Les auteurs comparent ces mesures au point de vue de leur exactitude et de leur précision, et de leurs applications à différentes matières. Ils donnent les résultats de l'expérience acquise avec les procédés par échange ionique utilisés pour séparer le plutonium des éléments générants lors de l'application des méthodes énumérées plus haut à des alliages de plutonium.

Ils montrent comment on peut utiliser la méthode du tube scellé à la dissolution des matières contenant du plutonium qui sont réfractaires et difficilement solubles, notamment les oxydes et alliages réfractaires. Ils passent en revue les procédés d'analyse permettant de caractériser le plutonium métal extrêmement pur. Ces derniers s'appliquent à l'analyse de plutonium métallique dont la teneur en impuretés est inférieure à 0,001%. Ils signalent aussi les méthodes applicables aux impuretés non métalliques, interstitielles et spectrographiques (métalliques).

MÉTHODES OПРЕДЕЛЯНИЯ ПЛУТОНИЯ В ОТРАБОТАННОМ РЕАКТОРНОМ ТОПЛИВЕ, МЕТАЛЛИЧЕСКОМ ПЛУТОНИИ, ЕГО СПЛАВАХ И СОЕДИНЕНИЯХ. Высокая стоимость, а также радиологическая и ядерная опасности, связанные с plutonium, и необходимость контролю за процессами регенерации топлива в целях их эффективности диктуют необходимость измерения содержания plutonium в его соединениях, по существу, с момента его получения и в течение всей его жизни. Уникальные проблемы, которые возникли в связи с растворами растворителей, как например, опасно высокий уровень радиоактивности и сложный химический состав, оказали влияние на методы разделения plutonium до того, как его можно определить с помощью радиохимических методов, предназначенных для этой цели. Окончательное измерение обычно связано со счетом альфа-частиц. Обсуждаются методы химического разделения plutonium от урана и продуктов деления, а также окончательное измерение plutonium. Обсуждаются также проблемы и методы обращения с высокоактивными растворами. Количество plutonium, независимо от того, находится ли он в виде металла, твердых соединений, сплавов (для использования в будущих энергетических реакторах) или растворов, можно измерить с помощью одного из нескольких верных и точных химических методов. Сюда входят потенциометрические методы, когда Pu (III) количественно окисляется до (IV валентного) состояния окисления, причем завершение реакции определяется потенциометрическим или фотоэлектрическим способом, амперометрическое титрование Pu (VI) с помощью Fe (II) до амперометрической границной точки, кулонометрические методы титрования при постоянном токе и постоянном потенциале, потенциометрическое титрование избыточного Fe (III) после восстановления Pu (VI) до (IV) и спектрофотометрические методы. Все эти методы обсуждаются с точки зрения верности и точности, а также с точки зрения применения к различным материалам. Описывается опыт применения разделения plutonium с помощью ноносеменных смол от побочных элементов в целях его применения к плутониевым сплавам.

Обсуждается применение метода герметических труб к растворению труднорастворимых жаропрочных плутониевых материалов, в частности жаропрочных окислов и сплавов. Рассматриваются аналитические методы, необходимые для характеристики металлического плутония высокой чистоты. Эти методы применяются для анализа металлического плутония, обееве содержание примесей в котором составляет менее 200 частей на миллион. Описывается также метод анализа неметаллических, промежуточных и спектрографических (металлических) примесей.

MÉTODOS PARA LA DETERMINACIÓN DEL PLUTONIO EN COMBUSTIBLES NUCLEARES AGOTADOS, PLUTONIO METÁLICO, ALEACIONES Y COMPUERTOS. El elevado valor del plutonio, los riesgos radiológicos y nucleares que le son inherentes y la necesidad de comprobar la eficacia de los procesos de regeneración obligan a determinar la concentración de este elemento en todos los materiales que lo contienen y, esencialmente, desde que dichos materiales se producen hasta que dejan de existir. Los singulares problemas que por su complejidad química y por su peligrosa radiactividad plantean las soluciones disolventes han inducido a emplear métodos para la separación del plutonio antes de que pueda ser determinado por las técnicas radioquímicas ideadas para este fin. En la memoria se exponen algunos métodos para separar químicamente el plutonio del uranio y de los productos de fisión, y para la medición final del plutonio. Se examinan también los problemas que suscita la manipulación de soluciones de elevada radioactividad y los métodos empleados para resolverlos.

El plutonio —sea en forma de metal, de compuestos sólidos, de aleaciones para reactores de potencia o en solución— puede ser medido por diversos métodos químicos exactos y precisos. Entre ellos cabe mencionar

METZ and WATERBURY
los métodos potenciométricos con los que el Pu(III) se oxida cuantitativamente hasta Pu(IV) y se determina potenciométrica o fotoelectricamente el final de la reacción, la valoración amperométrica del Pu(VI) con Fe(II), técnicas de valoración cumbiomiétrica de corriente constante y potencial constante, valoración potenciométrica del exceso de Fe(II) después de reducir el Pu(VI) a Pu(IV) y métodos espectrofotométricos. En la memoria se discute la exactitud y precisión de esos métodos y la posibilidad de emplearlos con diversos materiales. Por ser de interés para su aplicación, se expone también la experiencia adquirida en el empleo del intercambio iónico para separar el plutonio de otros elementos interferentes.

Se explica la forma en que puede emplearse la técnica del tubo sellado para disolver materiales de uranio difícilmente solubles – en particular las aleaciones y los óxidos refractarios. Por último, se describen las técnicas analíticas necesarias para la caracterización del plutonio metálico de alto grado de pureza. Esas técnicas se emplean cuando el material contiene menos de 200 partes por millón de impurezas. También se indican métodos para determinar impurezas no metálicas, intersticiales y espectrográficas (metálicas).

1. INTRODUCTION

The successful management, or control, of such valuable and hazardous nuclear material as plutonium must rely heavily on methods of analysis of various process solutions arising during the separation of plutonium from irradiated reactor fuel, and also analysis of the final separated product. All subsequent work with plutonium, either research or manufacturing activities, likewise must rely heavily on methods of analysis for plutonium, its alloys and compounds.

This paper deals with those methods of analysis currently in use in the United States of America in the production of plutonium and also some of those used in the research and developmental programs involving plutonium metal, its alloys and compounds. Most of these methods have been developed in the United States. Allotted space did not permit complete coverage of all methods of analysis involving plutonium. However, an attempt was made to include those most widely used. No attempt has been made to give adequate coverage to those analytical methods that originated outside the United States. It is hoped some of these will be discussed in other papers to be included in this Symposium.

2. CONTROL ANALYSIS IN PLUTONIUM PRODUCTION

The two United States processes currently in use for the recovery of plutonium from irradiated production reactor fuels are known as Redox [23] and Purex [16]. Figures 1 and 2 [2] show schematically the various steps in each of these processes and also where samples are taken for plutonium measurements. Table I [2] shows a summary of the plutonium analytical programs for both recovery processes. Process sample analyses serve principally to provide plant inventory data for the plutonium, accurate knowledge of plant efficiencies, stream composition data important for efficiency control, and plutonium concentration data for nuclear safety reasons.

As indicated in Table I, alpha counting is the technique used in nearly all the control analysis in plutonium production. It is routinely used for lean plutonium solutions such as waste streams, and also for highly radioactive solutions so that small aliquots may be analysed. Direct alpha counting (without previous separation of the plutonium) is the simplest, and is
used for such samples as the metal solution-blend streams (R-1) and (P-1), the code-contamination feed (R-2) and (P-2), and the feed to the plutonium recovery section (R-8) and (P-8). As in all radiochemical methods, the specific activity of the radionuclide being counted must be known. This is usually the case in plutonium production samples. If not, it must be calculated.
from isotopic distribution data obtained by mass spectrometer analyses. It is beyond the scope of this paper to discuss isotopic analysis. Corrections must also be made for all other alpha emitters, such as $^{241}$Am and $^{242}$Cm, if present.

In solutions containing high salt or high fission product concentrations and low plutonium concentrations, a separation of the plutonium is necessary prior to alpha counting. Both lanthanum trifluoride carrying and thenoyl-trifluoroacetone (TTA) extraction methods are employed. Waste streams and uranium stream samples, shown in Figs. 1 and 2 by (R-3), (P-3), (R-6) and (P-6), are typical examples.

In the Redox process, streams in the plutonium-recovery cycle (low uranium) and also the product, are usually analysed by an X-ray absorption method. Several other methods also are applicable to plutonium-rich solutions of this nature.

### 2.1. Methods usually applied to dissolver and feed solutions

The determination of plutonium at this point, (Samples (R-1) and (P-1), Table I) establishes the amount produced in the reactor and is important, when used in conjunction with samples (R-10) and (P-9) (Table I), to establish the overall recovery efficiency of the process. These solutions, intensely radioactive, mostly from their fission product contents, are analysed for plutonium by direct alpha counting. Samples of dissolver solutions (R-1), (P-1), (R-2), and (P-2) (Table I), are taken in a shielded facility by remote pipetting, and diluted with nitric acid to a known volume. The size of the sample aliquot is dependent on the activity of the dissolver solution from a shielding standpoint and on the alpha activity from the instrument capability standpoint. The diluted solution is, at this point, removed from the shielded facility and the remainder of the analysis carried out without shielding. An accurate volume of the diluted and stirred solution, to provide 5000 to 30 000 alpha disintegrations per minute, is pipetted on to a counting disc previously ringed with Zapon, evaporated by infra-red heating, flamed to dull red, cooled, and counted.

Corrections for $^{241}$Am and $^{242}$Cm are made on another aliquot by separating these two elements from plutonium by carrying them on cerium trifluoride, preparing a counting disc in the usual manner, and counting them together. For this separation the plutonium must first be oxidized with dichromate to the (VI) oxidation state which does not co-precipitate with cerium trifluoride.

Although it seems to be standard practice to use the direct alpha counting techniques [11] for measuring the plutonium in dissolver solutions, it is clear from Table I that these solutions are high in dissolved salts. Self absorption on the counting disc always exists, resulting in some negative bias. In order to correct this situation, previous extraction of the plutonium with TTA [31] or a double extraction using hexone and TTA should be used [24].

The techniques of alpha counting are too well known to require extended comment. Gas proportional counters, scintillation counters, and low geometry vacuum counters have been used satisfactorily. For some samples, tolerance of beta activity up to $10^8$ beta counts per minute is a requirement of the counting equipment.
<table>
<thead>
<tr>
<th>Sample</th>
<th>Approximate composition</th>
<th>Usual method</th>
</tr>
</thead>
<tbody>
<tr>
<td>Metal-solution blend</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Sample</td>
<td>Usual method</td>
<td></td>
</tr>
<tr>
<td>Metal-solution blend</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Redox</td>
<td>Redox</td>
<td></td>
</tr>
<tr>
<td>Purex</td>
<td>Purex</td>
<td></td>
</tr>
<tr>
<td>Sample</td>
<td>Usual method</td>
<td></td>
</tr>
<tr>
<td>Feed to codecontamination</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Various column waste streams throughout the entire process</td>
<td>Extraction, alpha counting</td>
<td></td>
</tr>
<tr>
<td>Feed to partitioning</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Feed to uranium recovery</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Uranium product</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Uranium trioxide product</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Feed to plutonium recovery</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Feed to second-cycle plutonium recovery</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Plutonium product</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Plutonium-metal product</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Condensates and cooling water</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
2.2. Methods usually applied to process solutions

These include various column waste streams throughout the processes, such as R-3 and P-3 (Table I), and the purified uranium recovered (R-6) and (P-6). These solutions contain relatively high concentrations of fission products and of uranium, and particularly low concentrations of plutonium. In these cases, it becomes necessary to extract the plutonium away from other constituents before its measurement by alpha counting.

The TTA extraction method [31] is very widely used. In this method, 100 µl of dissolver solution is diluted to 10 ml with 1 M nitric acid, and 500 µl of this solution is further diluted to 1.5 ml with 2 M nitric acid. The plutonium in this final aliquot is quantitatively reduced to the (III) oxidation state with hydroxylamine, then oxidized to the (IV) state with sodium nitrite. To this solution is then added a 0.5 M solution of TTA in xylene which quantitatively extracts only the plutonium. A known volume of the TTA solution is pipetted on to the counting disc, evaporated to dryness, flamed to redness, and alpha counted in the usual manner. No corrections to the alpha count are necessary because other alpha emitters such as americium, curium, and neptunium (V) remain in the aqueous phase. When applied to a series of dissolver solutions, the relative standard deviation for a single determination was 3 to 5%. Analysis of a standard plutonium solution gave a recovery greater than 99.5%.

An improvement has been made in this method whereby the extraction is accomplished with a combination of hexone and TTA, resulting in better decontamination from fission products. As developed by Maeck et al. [24], the plutonium is first separated from the remainder of the dissolver solution by a two-cycle extraction step. It is oxidized to the (VI) oxidation state with permanganate, extracted as the tetra-alkylammonium trinitrate complex into methyl isobutyl ketone from an acid-deficient salting solution. The plutonium is reduced to the (III) oxidation state with hydroxylamine and stripped from the organic phase into 3 M nitric acid. It is then oxidized to the (IV) state with nitrite, and quantitatively extracted into TTA-xylene. Overall recovery of the plutonium was 98.8% with a relative standard deviation of 0.8%. The plutonium was determined by alpha counting. The need for uranium and americium-curium corrections is thereby eliminated. This is one advantage of the method.

Worthy of note is an X-ray absorptiometric method [21] which is applied to the measurement of plutonium in the feed to the second-cycle plutonium recovery step in the Redox process. This solution has a relatively high concentration of plutonium and is also 0.9 M in aluminium nitrate and 0.2 M in nitric acid. The method applies best to a solution containing one atomic species of high atomic number. See section 3.6.

2.3. Other methods applicable to dissolver or process solutions

On paper, at least, any of the methods listed in section 3 can be used for determining plutonium in dissolver or process solutions, providing prior quantitative separation from the remaining constituents is accomplished. In a practical sense, however, this is not the case. Only those methods
requiring small samples have possibilities of successful application because it is not practical to handle remotely, behind heavy shielding, those quantities of highly radioactive solutions required to yield, quantitatively, multimilli-gram amounts of plutonium.

Among those methods requiring small-size samples which have actually been applied to the determination of plutonium concentrations in dissolver solutions following prior separation of the plutonium are:

(a) Isotopic dilution - mass spectrometer [32]. To the sample of dissolver solution is added a known amount of $^{242}$Pu (and $^{233}$U if U is analysed for). The plutonium (and uranium) are oxidized to their (VI) oxidation states with permanganate, in which form they are selectively extracted as the tetra-propylammonium complexes into methyl isobutyl ketone (hexone). The organic phase is separated, tetrahexylammonium iodide is added to it, and the reduced uranium and plutonium are stripped into hydrochloric acid. By the careful selection of the concentration of hydrochloric acid, the uranium-to-plutonium atom ratio in the extract can be chemically controlled. For example, if the ratio in the original solution is 250 to 1, a final ratio of 12.6 to 1 is obtained when 4 M hydrochloric acid is used. To the aqueous phase are then added a few drops of hydrogen peroxide and nitric acid and the solution is evaporated to fumes. The plutonium (and uranium) are determined by multiple filament mass spectrometry in which the singly charged metal ions are measured. From the knowledge of the volumes used, and the isotopic ratios of the spike, both plutonium isotopic composition and the plutonium concentration of the dissolver solution can be calculated. One significant advantage of the method is that quantitative extraction of the plutonium from the dissolver solution is not required. Hence a possible source of bias is eliminated. Neither is a clean separation from the uranium required. As little as 10^{-9} g of plutonium on the filament may be analysed; practical considerations, however, suggest that approximately 10^{-7} g actually be used. A uranium-to-plutonium ratio between 10 and 25 to 1 on the filament gives excellent results. The relative standard deviation of a single determination of plutonium concentration was reported to be 0.7% for an isotopic population of $^{239}$Pu = 92.69%; $^{240}$Pu = 6.74%, and $^{241}$Pu = 0.567%.

(b) Controlled potential coulometric titration [13]. The plutonium is separated by anion exchange from the remaining constituents of a Hanford-type dissolver solution. The plutonium is absorbed in the (IV) oxidation state from 8 M nitric acid on to Dowex 1X4 resin. It is then eluted with 0.5 M nitric acid - 0.02 M hydroxylamine, and titrated. For the titration, the cell is equipped with a platinum mesh electrode. The sample is preoxidized, then reduced, the reduction step being used for the actual analysis. Standard dissolver solutions do not exist with which a comparison might be made. Analyses made on Hanford-type dissolver solutions by direct alpha counting and by coulometric titration showed that the relative standard deviation of the counting was about 2% as compared with 1.5% for the titration technique. Coulometric techniques have not found wide acceptance.

(c) Potentiometric titration, Cr(II) reduction-Ce(IV) oxidation [12]. (See section 3.2 (a).) This method has been applied to the determination of Pu, following its separation from Scrupp dissolver solutions. The Pu is extracted from the 2 M nitric acid dissolver solution into 1 M TTA con-
taining di-isopropyl ether, and then stripped into 10 M hydrochloric acid. It is then reduced with 0.2 M potassium iodide and passed through Dowex-1 to remove the U. Traces of iron are removed by mercury cathode electrolysis. Titrations of plutonium samples as small as 100 μg were reported to be unbiased and showed relative standard deviations from 0.5 to 2%.

(d) Spectrometric methods. Several spectrophotometric methods have been developed, some of which are quite sensitive. (See section 3.4.) None of these has found application to the direct determination of plutonium in dissolver solutions of production reactor fuels for obvious reasons. However, any of the more sensitive of these methods may be used providing a clean-cut separation of the plutonium from the uranium and fission products is first accomplished. A few applications of spectrophotometry to dissolver solutions from specific reactors have been reported [22, 25].

(e) X-ray fluorescence. This technique has not found acceptance for the routine analysis of dissolver solutions, nor of production process solutions. However, a method [10] has been developed whereby an analysis may be made on a sample containing a total amount of plutonium in the range of 0.4 to 0.8 mg. When applied to synthetic samples a relative standard deviation of less than 1% was reported. As in the case of all X-ray emission techniques, the matrix affects the results and must always be known and reproducible. Prior separation of the plutonium from the remaining constituents of any solution would be necessary to obtain a precision of less than 1%.

Limited applications of some methods have been made to the direct determination of plutonium in dissolver solutions (without prior separation of the plutonium). Included are the following methods:

(f) Controlled potential coulometric titration [40]. In early work by Scott and Peekema, a controlled potential coulometric technique similar, but not identical, to that reported in section 2.3 (b) was applied to the direct measurement of plutonium in dissolver solutions. The reproducibility of this method was 1% at the 95% confidence level.

(g) Scanning coulometric method. More recently, Propst [38] developed a highly sensitive method for determining plutonium in dissolver solutions which does not require a prior separation of the plutonium. The method makes use of an electroanalytical instrument called a scanning coulometer. It records coulomb-potential curves of reversible processes under nearly equilibrium conditions. The circuit features a novel system for producing a scan rate that is an inverse function of the electrolysis current. Compensator networks provide automatic corrections for the continuous faradic current and the charging of the electrical double layer. It is reported that plutonium may be determined at the 2 to 5 μg level with a relative standard deviation of 1%. The typical solution of irradiated nuclear fuel used was 1.4 M in uranium and 0.5 to 1.0 M in HNO₃; 25-μl aliquots containing 3.3 μg of Pu gave the above precision. Limited experience with this method indicates that it may be rather successful for this purpose.
3. DETERMINATION OF PLUTONIUM IN METAL, SOLUTIONS, ALLOYS, AND COMPOUNDS

3.1. Preparation of samples

Plutonium metal and most of its alloys are oxidized readily in air, especially in the presence of traces of moisture; therefore, pre-treatment before analysis is required to remove surface contaminants. The preferred cleaning method for large regularly shaped pieces is mechanical polishing, either with a file or abrasive cloth, to expose a silvery metallic surface. This cleaning should be done at a slow rate, to prevent heating of the sample, and in an inert atmosphere, if possible. Small or irregularly shaped pieces of the metal and some alloys may be polished electrolytically in a 20% potassium carbonate electrolyte by making the sample the anode and using a tantalum cathode and a 22.5-V dry-cell battery as a current source. The current flow is kept to the minimum that will expose a bright metallic surface, then the sample is washed immediately with water, ethanol, and methyl chloroform, in that order, and dried briefly in air or an inert atmosphere. Plutonium compounds generally do not require pre-treatment, except for drying of stable materials prior to dissolution. Suitable solvents for some plutonium-containing materials are given in Tables II and III.

Removal of interfering ions often is required before analysis of plutonium-containing solutions. Addition of sulphuric acid, or nitric and perchloric acids, and repeated evaporations to strong fumes is recommended to eliminate volatile ions, such as nitrate or fluoride, and to destroy organic material. Undesired cations are removed by mercury cathode electrolysis from dilute sulphuric acid solutions, or by an anion-exchange resin separation based upon selective adsorption of plutonium from 7.2 M nitric acid solution.

3.2. Electrometric methods

The electrometric techniques applied to the determination of plutonium include potentiometric, coulometric, and amperometric titrations, polarography, and chronopotentiometry. Potentiometric or amperometric titrations generally are used for the most precise determinations; applications of polarography and chronopotentiometry are very limited. Polarography has been used to measure the reduction-oxidation potentials of plutonium in solution [8, 14], and more recently, preliminary work has shown that square-wave polarography, using the plutonium (IV)-(III) couple in nitric or hydrochloric acids, is applicable to the determination of micromolar concentrations of plutonium [20]. The chronopotentiometric behaviour of plutonium (III), (IV), and (VI) at a platinum electrode in various mineral acids was investigated to determine possible applications to analysis of plutonium [35]. Well-defined chronopotentiograms were obtained for electro-oxidation of plutonium (III) in perchloric acid, but analytical methods based upon this technique have not been developed.
### TABLE II

**SOLVENTS FOR PLUTONIUM METAL AND ALLOYS**

<table>
<thead>
<tr>
<th>Type sample</th>
<th>Suitable solvents</th>
</tr>
</thead>
<tbody>
<tr>
<td>Alloys with:</td>
<td></td>
</tr>
<tr>
<td>Al, Bi, Cd, Ce, Cr, Co, Fe, Ga, In, La, Mg, Mn, Mo, Ni, Si, Sn, Ti, Th, U, V, or Zn.</td>
<td>3 to 12 M HCl or HClO₄</td>
</tr>
<tr>
<td>Cu, Hg, Rh</td>
<td>HCl-HNO₃ (300°C) a</td>
</tr>
<tr>
<td>Ru, or Ru, Mo, Zr, Ce, La, Nd, and Fe</td>
<td>HCl-HClO₄ (boiling) b or HCl-HNO₃-H₂SO₄ d and NaNO₃-NaOH fusion c</td>
</tr>
<tr>
<td>Platinum metals</td>
<td></td>
</tr>
<tr>
<td>Nb, Si, Ta</td>
<td>HCl-HF</td>
</tr>
<tr>
<td>Nb, Ta, Zr</td>
<td>HCl and bisulphate fusion or HCl-trace HF</td>
</tr>
<tr>
<td>Al or Al and Si</td>
<td></td>
</tr>
<tr>
<td>Ce-Co, Ce-Co-Mn, or Ce-Ni</td>
<td>Hg catalysed HNO₃</td>
</tr>
<tr>
<td>High purity metal and metal containing such impurities as:</td>
<td>12 M HCl</td>
</tr>
<tr>
<td>Al, B, Ca, Cd, Cs, Ce, Cr, Co, Fe, La, Li, Mg, Mn, Mo, Ni, K, Sr, Na, Si, Sn, Ti, Th, U, V, Zn, Zr</td>
<td>3 to 12 M HCl, other halogen acids also</td>
</tr>
</tbody>
</table>

a Rhodium requires the sealed-tube method.  
b RuO₄ distilled.  
c Alternate acid and fusion treatments.  
d Wichers' sealed-tube method [45].

### 3.2.1. Potentiometric methods

The potentiometric titration methods are based upon either the oxidation of plutonium (III) or the reduction of plutonium (IV) to the (VI) oxidation state which is stabilized by complexing with sulphate ion. Generally, elimination of interfering anions, such as fluoride and nitrate, by a preliminary fuming operation is prerequisite to successful titration. Determinations involving the plutonium (VI)-(IV) couple have the potential advantage of greater sensitivity, because of the two electron change, but concern for high reliability predominates in selection of sample size, usually 5 to 200 mg of plutonium.
### TABLE III

**SOLVENTS FOR PLUTONIUM COMPOUNDS**

<table>
<thead>
<tr>
<th>Compounds:</th>
<th>Suitable solvents</th>
</tr>
</thead>
<tbody>
<tr>
<td>PuF$_3$, PuF$_4$</td>
<td>Hot conc. HNO$_3$, HClO$_4$, or H$_2$SO$_4$</td>
</tr>
<tr>
<td>Pu(IO$_3$)$_3$, Pu(VO$_3$)$_4$, Pu$_2$(C$_2$O$_4$)$_3$, Pu(C$_2$O$_4$)$_2$, PuO$_4$$^-$$\times$H$_2$O, BiPuH(C$_2$O$_4$)$_4$, PuBiO$_3$, NaPuO$_3$(C$_2$H$_2$O$_2$)$_3$</td>
<td>Hot conc. HNO$_3$, HClO$_4$, or H$_2$SO$_4$</td>
</tr>
<tr>
<td>PuCl$_3$, PuCl$_4$, Pu(NO$_3$)$_4$, Pu(SO$_4$)$_2$, Pu(CIO$_4$)$_2$, PuO$_2$Cl$_2$, PuO$_2$(NO$_3$)$_2$, PuO$_2$(ClO$_4$)$_2$</td>
<td>Water, followed by dilute acid to prevent hydrolysis</td>
</tr>
<tr>
<td>PuO$_2$</td>
<td>HF-HNO$_3$, H$_2$SO$_4$, HN$_4$HF; fusion, then hot HNO$_3$; Na$_2$O$_2$ fusion, then hot HNO$_3$; HCl-HClO$_4$ (300°C)$^a$, HBr (teflux)</td>
</tr>
<tr>
<td>Double chloride, nitrate, or perchlorate salts with Ca, Cs, Ce, La, Li, Mg, K, Sr, or Na</td>
<td>3 to 12 M HCl; 1 to 6 M HNO$_3$</td>
</tr>
<tr>
<td>Pu peroxides</td>
<td>HCl or HNO$_3$ with Mn(II), Ce(IV), Sn(II), or K(I)</td>
</tr>
<tr>
<td>Pu carbides</td>
<td>HCl, HNO$_3$, HClO$_4$, or H$_2$SO$_4$; 15 M HNO$_3$ - 6 M HF (teflux) if difficulty soluble</td>
</tr>
<tr>
<td>Pu nitrides</td>
<td>Dilute HCl, HNO$_3$, HClO$_4$, or H$_2$SO$_4$</td>
</tr>
</tbody>
</table>

$^a$ Wichers' sealed-tube method [45].

For the same reason, weight burettes or accurate screw-driven syringe burettes generally are used for taking aliquots and for adding titrants.

In one of the first reliable titration methods [29], an excess of titanium (III) is added to reduce the plutonium quantitatively to the (III) oxidation state. Addition of cerium (IV) solution produces sharp endpoints for the oxidation, first of the excess titanium (III), and second, of the plutonium (III). In titrating 4 to 6 mg of plutonium, a relative standard deviation of 0.15% is possible routinely. Iron, chromium, vanadium, platinum, and gold cause interference.
Chromium (II) is used as the reductant in a microtitration method [34] for determining 0.1 to 0.2 mg of plutonium. An aliquot of the sample solution is fumed with sulphuric acid, and a slight excess of chromium (II) is added. The excess chromium (II) and then the plutonium (III) are titrated with cerium (IV). The two endpoints are detected by using a bimetallic gold-platinum electrode system, and the titration curve is plotted by a recorder. The relative standard deviation of a single determination is 0.5%. Iron, uranium, vanadium, platinum, and gold interfere. For application of this method to the determination of 0.2 to 0.4 mg/ml of plutonium, following separation from irradiated uranium fuel slugs, see section 2.2.

Titration methods in which the plutonium is reduced with zinc amalgam have the advantage of requiring detection of only one endpoint. In a very reliable method [1, 29], the sulphuric acid solution of the sample is stirred for 30 to 60 minutes in contact with liquid zinc amalgam in a special microtitration cup. The amalgam is withdrawn through a capillary sealed to the bottom of the cup, and the plutonium (III) is titrated with cerium (IV) using a platinum-calomel electrode system to detect the endpoint. For a single determination of 25 mg of plutonium, the relative standard deviation is 0.06%. Iron, chromium, vanadium, molybdenum, titanium, gold, and platinum interfere.

In another method [37], amalgamated zinc in the form of a Jones reductor is used as a reductant for plutonium in 4 M sulphuric acid solution. The plutonium (III) is titrated with standard potassium dichromate solution, using polarized gold electrodes to indicate the endpoint. A relative standard deviation of 0.04% was reported for a single determination of 70 to 140 mg of plutonium. Metals, except chromium, that interfere in the above method also cause interference here. This method was applied to the determination of plutonium (following its separation) in plutonium oxide-uranium oxide, and in plutonium-uranium carbide fuel elements. Separation was accomplished by a cation-exchange resin method in which plutonium and uranium were adsorbed on Dowex-50 resin from 0.3 M nitric acid-0.05 M hydroxylamine solution. The uranium was eluted with 0.3 M sulphuric acid-0.05 M hydroxylamine, then plutonium was eluted with 6 M nitric-0.05 M sulphamic acid. Reported recovery of plutonium was greater than 99% and the relative standard deviations of the determinations were in the range of 0.1 to 1.0% [36].

In the most precise potentiometric method [44], at least 180 mg of plutonium are oxidized with fuming perchloric acid to plutonium (VI) which is reduced to plutonium (III) and (IV) by adding a slight excess of iron (II) solution. Titration of the plutonium (III) and excess iron (II) with cerium (IV), using a platinum-calomel electrode system, completes the determination, which has a relative standard deviation of 0.02%. Chromium, gold, manganese, and vanadium interfere.

A variation of this method was proposed recently [28] in which argentic "peroxide" in 1 M sulphuric acid oxidizes the plutonium to the (VI) state, and 0.50 N potassium dichromate solution is the oxidant used in the backtitration of excess iron (II). With this system a smaller break in potential is obtained at the endpoint. Preliminary tests indicate a relative standard deviation of 0.3%. The effect of various ions on the determination was not reported.
3.2.2. Amperometric methods

Plutonium may be determined quite precisely by titration of plutonium (VI) with iron (II) in which the endpoint is detected amperometrically with a rotating platinum electrode [41]. The plutonium is oxidized to the (VI) oxidation state with argentic oxide, the excess oxidant is destroyed by heating, and then the plutonium (VI) is titrated to the (IV) oxidation state by the addition of standard iron (II) sulphate. Relative standard deviations are 0.05% for sample sizes of 10 to 20 mg and 0.4% for 0.2 mg of plutonium. For the smaller quantity, there is a negative bias of 0.6%, but there is no significant bias if the sample size is greater than 10 mg. Metals that interfere with this method include cerium, chromium, manganese, and vanadium.

3.2.3. Coulometric methods

Constant current and controlled potential coulometry have been shown to apply to the determination of plutonium in dissolver solutions, process solutions, and product plutonium. Constant current methods were developed first, but the advent of simple and reliable instruments for controlled potential coulometry led to greater use of this latter technique. The constant current coulometric titrations involve redox reactions of the plutonium (III)-(IV) and (VI)-(IV) couples. In methods involving the plutonium (III)-(IV) couple, plutonium is reduced electrolytically in nitric acid-sulphamic acid solution or by adding a slight excess of chromium (II) or titanium (III) ions [5-7]. Then the plutonium (III) is titrated coulometrically with electrolytically generated bromine or cerium (IV), using a potentiometric detection of the endpoint.

Wide acceptance did not follow the introduction of these methods because of experimental difficulties that include large reagent blanks caused by impurities, and oxidation of plutonium (III) by anodic products diffusing into the main titration compartment of the titration cell. Some of these methods are tentative and are based upon limited investigations that have not been continued.

A method [7] involving the plutonium (VI)-(IV) couple seems to be more reliable and is not influenced by iron impurities. This method, which applies to the determination of 0.003 to 10 mg of plutonium, starts with the oxidation of the plutonium to the (VI) oxidation state with permanganate and bismuthate. The manganese dioxide that is formed is destroyed with formaldehyde or sodium azide, and the plutonium (VI) is titrated to plutonium (IV) with electrolytically generated iron (II) sulphate. The endpoint is determined potentiometrically. Relative standard deviations are 5% and 1% in determining 0.003 mg and 1 mg of plutonium, respectively. Separation of the plutonium by a lanthanum fluoride carrier co-precipitation eliminates interference from most elements except cerium and oxygen.

Controlled potential coulometric determinations [39, 40, 42, 43] utilize titration cells that have either a mercury-working electrode, with a citrate-buffered aluminium sulphate electrolyte, or a platinum-working electrode with 1 M hydrochloric, nitric, or perchloric acid as the electrolyte. Recently, phosphate electrolytes were reported to be satisfactory [26]. The coulometric oxidation of plutonium (III) to (IV) is the preferred reaction in
the mercury cell, but either the coulometric oxidation of plutonium (III) or reduction of plutonium (IV) is a satisfactory electrode reaction in the platinum cell. Relative standard deviations of these determinations increase from 0.1% for samples greater than 2 mg to 1% for 0.05 mg. Anions that must be removed by fuming prior to the titration include nitrite, thiocyanate, peroxide, and fluoride. Judicious choice of potential and electrolyte prevents interference by most metal ions.

The simplicity of controlled potential coulometry in determining plutonium in complex solutions is evident in a typical application [42] to a plutonium "product" solution. An aliquot containing 5 mg of plutonium is warmed with nitric acid, then the plutonium is reduced coulometrically in a perchloric-sulphamic acid electrolyte. Following this pre-treatment, the integrated current to oxidize the plutonium to the (IV) oxidation state is measured and related to plutonium concentration. Small amounts of iron and uranium do not interfere. As mentioned previously, controlled potential coulometry may be applied to reactor fuel dissolver solutions, either directly or following anion-exchange resin separation of the plutonium. However, this technique has not been adopted as a routine method.

3.3. Volumetric methods

One of the earliest methods [18] for determining plutonium consisted of reduction of plutonium to the (III) oxidation state in a Jones reductor and subsequent titration to plutonium (IV) with standardized cerium (IV) solution. Ferrous o-phenthroline was used as a colour indicator in the visual detection of the endpoint. This method was modified recently to include photometric detection of the colour change of the indicator [4]; this change improved the precision of the method to a relative standard deviation of 0.03% when employing a 0.5-g sample.

3.4. Spectrophotometric methods

Applications of spectrometric methods have been limited because of difficulties caused by multiplicity of plutonium oxidation states, each of which has a characteristic spectrum and reacts differently with colour-forming reagents, and by the low molar absorptivities of the plutonium ions. However, spectrophotometry has the advantage, compared with radiochemical methods, of not requiring a knowledge of the isotopic population of the plutonium.

These methods are based upon measurement of the absorbance of the plutonium ions directly, or of coloured complexes formed with some reagent. Colour-forming reagents generally are used in methods for determining microgram quantities of plutonium. Sodium alizarin sulphonate forms with plutonium (IV) a complex that has a molar absorptivity of 9870 when measured at 530 nm; this method applies to the determination of 2 to 5 mg Pu/ml with relative standard deviations not greater than 2% [17]. This reagent is not selective nor particularly sensitive, and the method has not found wide acceptance.
More recently, a method was developed in which the absorbance of the plutonium (IV)-arsenazo I complex, formed in 0.4 M perchloric acid solution, is measured at a wavelength of 600 nm [9]. The large molar absorptivity of this complex, 22,000, makes the method very sensitive; 20 to 100 μg of plutonium in a 10-ml volume are determined with relative standard deviation of 6%. At the 0.4 M acidity, the interference caused by uranium at lower acidities is virtually eliminated, but several other metals cause interference. Separation of the plutonium by anion-exchange resin or fluoride precipitation are incorporated into the method to reduce the number of interfering ions.

Larger concentrations of plutonium (from 145 to 170 mg per gram of solution) may be determined directly by reducing the plutonium with ascorbic acid and measuring the absorbance of the plutonium (III) at 562 nm. A high-density reference prepared from the perchlorate salts of chromium (III), cobalt (II), and copper (II) is the reference solution. Careful control of temperature and other variables, and use of weight burettes, make this method precise; a relative standard deviation of 0.11% was reported [3]. Application has been made to nitrate recovery solutions and to plutonium metal dissolved in hydrochloric acid.

3.5. Radiochemical methods

Radiochemical methods may be employed if high precision is not required. They are applied to an extremely wide range of plutonium concentrations, and with adequate separation of the plutonium from other constituents, application is made to a very great variety of solutions. The most useful application is to dilute solutions and arises because quantities as small as 10^{-10} g can be measured reliably, a capability frequently exploited in biological work.

The simplest condition in which plutonium is encountered is in a solution of one of its salts without other dissolved salts being present. In such cases, following proper dilution, if necessary, a suitable aliquot is transferred to a counting plate, dried under controlled conditions, flamed to a dull red heat, and alpha counted. In case the sample contains other dissolved but non-radioactive salts, the direct evaporation technique may still be used, providing the ratio of dissolved salts to plutonium is not too great. The quantity of salts on the counting plate should not exceed 100 to 200 μg.

If the dissolved salt content is too great and interferes too seriously for the precision required, the plutonium frequently is separated by co-precipitation with lanthanum fluoride. The lanthanum ion is added in specific amounts in the form of lanthanum nitrate, and fluoride ion is added in the form of hydrofluoric acid or ammonium fluoride. Plutonium (III) and (IV) are carried quantitatively by the lanthanum trifluoride precipitate. The solution is centrifuged, and the precipitate is slurried with nitric acid and transferred to a counting plate for drying and counting in the usual manner. Correction is made for the self-absorption caused by carrier. Other precipitating or carrying agents include bismuth phosphate, lanthanum hydroxide, zirconium phosphate, and thorium pyrophosphate; the oxalates of thorium, uranium(IV), bismuth, and lanthanum; and the iodates of cerium, thorium, and zirconium.
In general, ion-exchange or liquid-liquid extraction techniques are preferred for separating plutonium from irradiated reactor fuel solutions. Anion-exchange resin systems employ either nitric acid or hydrochloric acid solutions for the loading solutions, and elution of the plutonium is accomplished with dilute acid, or complexing or reducing agents such as fluoride or hydroxylamine. Several cation-exchange resin systems also have been reported for this purpose. However, liquid-liquid extraction techniques are less time-consuming than ion-exchange techniques and, in addition, the latter frequently separate the element into a volume of solution too large to handle conveniently. For these reasons liquid-liquid extraction methods are preferred to anion-exchange techniques.

One excellent extraction method for determining plutonium employs 0.5 M TTA in xylene as the extractant [31]. Plutonium (IV) is extracted from 1 M nitric acid and then stripped from the organic phase into 10 M nitric acid. A second extraction with TTA removes traces of gamma activity. Other useful extractants include tri-isooctyl amine in xylene, cupferron, and mixtures of TBP and TTA. Following the selected separation method, an aliquot of the plutonium solution is evaporated, or the plutonium is electro-deposited as a compound, on a counting disc, and the appropriate radioactivity measurements are made. Radiochemical methods generally are not highly precise, but they are selective, rapid, and very sensitive.

3.6. X-ray methods

3.6.1. Absorption

For assaying plutonium in high purity samples, an X-ray absorption method has been developed [21]. Using a "criss-cross" technique in a split-beam X-ray photometer, with optimum conditions of a 12-cm cell path and a plutonium concentration of 36 g/litre in a solution of known acidity, a relative standard deviation of 0.3% was achieved. This method works best for clean solutions. For impure solutions, the plutonium is separated by solvent extraction with TBP. Then the organic phase is analysed for plutonium by X-ray absorption using a fluorescent-screen type X-ray photometer. This method has a relative standard deviation of 0.5% in determining 5 to 70 mg of plutonium per millimetre of solution [19]. However, it should be pointed out that this absorption method measures total absorption of all solutes, solvents, and cell. The method has not gained wide acceptance. This is probably due, at least in part, to the non-availability of a commercial instrument.

Recently, a newer technique [15], X-ray absorption edge analysis, has been applied to improve the selectivity for the determination of plutonium. In this method secondary X-rays, generated by irradiating a niobium-molybdenum metal target with X-rays from a tungsten target X-ray tube, are transmitted through the sample solution and through standard solutions. The relative absorptions by the sample solution, and by the standard solutions, of these two X-rays, one located closely on each side of an absorption edge of plutonium, are used to calculate the plutonium concentration. For determining plutonium concentrations in alloys which vary from 30 to 100%, the
relative standard deviations vary from 0.91 to 0.55%. Only radon, radium, neptunium, and americium have absorption edges between the X-rays used in this method.

3.6.2. Fluorescence methods

In addition to the example cited in section 2.3(e), another X-ray emission method has been recommended for determining plutonium in refractory oxides [27]. The sample, to which thorium has been added as an internal standard and to reduce matrix effects, is fused with potassium pyrosulphate, cooled to form pellets, and the X-rays of thorium and plutonium are measured. The relative standard deviation in determining 1 to 10 mg of plutonium is reported to be 3.2%, but this can be reduced to 0.6% by polishing the pellets before analysis.

4. ANALYSIS OF SUPER-PURE PLUTONIUM

A complete analysis of super-pure plutonium metal consists of determining all known impurities, using the most sensitive methods available, and of assaying for the plutonium content using the most reliable of the assay methods. Recent developments in purifying plutonium by electro-refining from molten salt baths have led to almost routine production of very pure metal [33]. Various analytical techniques are required for the analysis of the super-pure metal made by this process. Approximately thirty impurities are determined by spectrochemical methods. These methods utilize two general techniques: (a) carrier distillation to effect partial separation of the impurities from the plutonium by selective volatilization of the impurities and suppression of the plutonium spectrum; in this technique a blend of the ignited sample with gallium oxide, silver chloride, or sodium fluoride is excited in a cratered electrode; and (b) separation of the plutonium from the impurities by ion-exchange resin, fluoride precipitation, or extraction methods, and evaporation of an aliquot of the impurity solution on to the electrode for arcing [30].

Spectrophotometric, physicochemical, or radiochemical methods are applied to determine those elements for which emission spectrography is not applicable or too insensitive [30]. Spectrophotometric methods often are used to determine Ce, Fe, Hf, Mo, N, Nb, P, Ta, Th, U, W, and Zr. Methods exist also for the following elements: B, Cd, Co, Cr, Ga, Mn, Ni, Pt, Rh, Ru, S, Si, Ta, and Zn. Physicochemical methods are applied to the determinations of oxygen, carbon, fluoride, and chloride. Radiochemical methods are used in determining americium and neptunium.

The assay of super-pure plutonium requires extreme care to obtain a reliability that makes the results meaningful. The best methods have relative standard deviations of 0.02 to 0.03%. This variance is greater than the total of the impurity concentrations, and a large number of determinations are required to obtain an average assay value of sufficient reliability. Of the methods described previously, those which have been applied to the assay of super-pure plutonium are: amperometric titration of
plutonium (VI) with iron (II), potentiometric titration in which plutonium is reduced to the (III) oxidation state with zinc amalgam and then titrated with cerium (IV), and potentiometric titration of plutonium (VI) with iron (II). All the high-precision methods described in this paper are suitable. Although detailed descriptions of methods for analysis of super-pure metal perhaps are not necessary for analysts familiar with this field, it is worthy of note that present methods for the determination of trace amounts of impurities in super-pure metal permit the meaningful analysis of metal having a total impurity content of the order of 100 ppm. Methods for the pp 10\textsuperscript{9} range of impurities remain to be developed.

REFERENCES

[27] MENIS, O. et al., Analyst. Chem. 35 (1963) 1049.
DISCUSSION

J. T. Byrne: In section 1.2 of his paper* Mr. Regnaud reports a difference of 0.2% between the ORNL 2005 x 50 controlled potential coulometer and the NBS Pu standard. Do you know of any United States laboratories finding this same difference?

C. F. Metz: No, I know of no other case. I should like to know whether there are any.

---

* These proceedings (SM-67/19).
PRACTICAL GAMMA COUNTING OF UNIRRADIATED URANIUM-235

P. COLSMANN
BROOKHAVEN NATIONAL LABORATORY, UPTON, N.Y., UNITED STATES OF AMERICA

Abstract — Résumé — Аннотация — Resumen

PRACTICAL GAMMA COUNTING OF UNIRRADIATED URANIUM-235. During the fabrication of reactor fuel elements it is necessary to have assurance regarding the accuracy of the fabricator's uranium assignment within the specified tolerances. Destructive analysis of random samples is both expensive and time-consuming. Where the uranium-bearing components are suitable for gamma counting, a non-destructive method of assay can be used with greater efficiency and equal accuracy.

The particular method described was used for checking fuel cores of nominal 30 wt. % enriched uranium in aluminium measuring about two inches square by 0.080 in. and 0.160 in. thick. The equipment was a basic NaI scintillation counter equipped with a single-channel analyser. The analyser, however, was operated with a very wide window covering both the 90-keV and 184-keV peaks characteristic of uranium-235. In practice, the threshold level and the window opening were adjusted to give the optimum maximum count rate as indicated by a ratemeter. The counting of a fuel core was then performed with the NaI crystal essentially unshielded and located several inches above the fuel core.

The counting time was adjusted to yield a total count in the range of $10^5$ to $10^6$ in order to minimize the counting error. Effects due to variations in the counting geometry and to non-uniform uranium distribution were minimized by the relatively large separation of the crystal from the fuel core. Effects due to shifting of analyser window were minimized by use of a wide opening. To compensate for possible non-uniform uranium distribution through the thickness of a fuel core, each core was counted on both sides. The total count obtained in this manner was directly proportional to the uranium-235 content of the fuel core.

In application, the counting equipment was set up in the fabricator's plant and a number of production fuel cores were counted. The plotting of the total counts against the fabricator's uranium-235 assignment revealed an unexpected error in the fabricator's system. This error was revealed on a qualitative basis before any destructive analyses were made to obtain quantitative correlation.

Mention is also made of three other applications of the same equipment. With the addition of a crystal traversing device and an output recorder, finished fuel plates were scanned to determine uranium distribution while a total count of the plate at the same time gave an indication of the total uranium content. Some unknown samples were assayed by the standard method of measuring self-absorption and comparing with a known uranium-235 standard. The addition of a well-type crystal allowed for assaying dilute uranium-235 solutions with sufficient accuracy for waste sampling and criticality evaluation.
стержня производили с помощью полностью неэкранированного кристалла NaJ, расположенного оптимальный максимум скорости счета, указанный интенсиметром. Затем счет топливного

на практике пороговый уровень и отверстие окошка регулировались так, чтобы обеспечить с очень широким окном, охватывающим пиковые характеристики урана-235 при 90 и 184 кэв.

Время счета было отрегулировано таким образом, чтобы получить общий счет в пределах от $10^5$ до $10^6$ с тем, чтобы уменьшить ошибки счета. Эффекты, вызываемые колебаниями в геометрии счета и неоднородным распределением урана, были сведены до минимума за счет сравнительно большого удаления кристалла от топливного стержня. Эффекты, вызываемые смещением окна анализатора, были сведены до минимума за счет использования широкого открытия окна. Чтобы компенсировать возможное неоднородное распределение урана в топливном стержне, каждый стержень обсчитывался с двух сторон. Полученный таким образом общий счет был прямо пропорционален урану-235, содержащемуся в топливном стержне.

На заводе-изготовителе было установлено счетное оборудование и изготовленные многочисленные топливные стержни были подвергнуты счету. Показания общих счетов в зависимости от назначения урана-235 изготовителем вскрыли неожиданную ошибку в системе изготовителя. Эта ошибка была обнаружена на качественной основе до того, как были проведены анализы с разрушением образца для получения количественной корреляции.

Упоминаются также три других вида применения этого же оборудования. Используя устройство с кристаллами и регистратором мощности, изготовленные топливные пластины подвергались сканированию для определения распределения урана, в то время как общий счет пластин за это же время показывал общее содержание урана. Несколько неизвестных образцов были проанализированы с помощью стандардного метода измерения самоАборбции, и сравнины с известными стандартами урана-235. Использование кристалла канального типа позволило провести с достаточной точностью анализ разбавленных растворов урана-235 с тем, чтобы произвести оценку критичности.
tomadas al azar lleva mucho tiempo y es muy costoso. Cuando los componentes que contienen uranio se prestan al recuento gamma se puede usar con más eficiencia e igual exactitud un método de ensayo no destructivo.

El método descrito en la memoria se utilizó para verificar núcleos combustibles de aleación de aluminio y uranio con un contenido nominal, en peso, del 30% de uranio enriquecido, cuyas dimensiones eran de unas 2 pulg³ por 0,80 y 0,160 pulg de espesor. El equipo consistía básicamente en un contador de centelleo con un cristal de NaI y un analizador monocanal. El analizador funcionó con una ventana muy amplia que permitía abarcar tanto el pico de 90 keV como el de 184 keV, característicos del uranio-235. En la práctica, el umbral de medición y la abertura de la ventana se ajustaron para lograr el recuento máximo y óptimo por unidad de tiempo, con arreglo a las indicaciones de un integrador. La verificación de cada núcleo se llevó a cabo con el cristal de NaI prácticamente sin blindaje y colocado a varias pulgadas de distancia.

Se reguló el tiempo de recuento para obtener un valor total del orden de $10^5$ a $10^6$ y reducir así el error mínimo. La separación relativamente grande entre el cristal y el núcleo de combustible redujo al mínimo los efectos debidos a las variaciones en la geometría del recuento y a la falta de uniformidad en la distribución del uranio; los ocasionados por el desplazamiento de la ventana del analizador se redujeron también al mínimo empleando una abertura amplia. Para compensar la posible falta de uniformidad en la distribución del uranio en el sentido del espesor de los núcleos se hizo el recuento por ambos lados. El recuento total de esta forma obtenido fue directamente proporcional al contenido de uranio-235.

El equipo de recuento, instalado en la planta, se utilizó para medir una serie de núcleos combustibles de la cadena de fabricación. La representación gráfica de los valores obtenidos, comparados con la asignación de uranio-235 calculada por el fabricante, puso de manifiesto un error inesperado en el sistema seguido en la planta. Este error fue descubierto sobre una base cualitativa antes de hacer los análisis destructivos necesarios para obtener una correlación cuantitativa.

En la memoria se mencionan otras tres aplicaciones del equipo descrito. Agregándole un dispositivo de desplazamiento transversal del cristal y un registrador de salida se exploraron placas de combustible, con objeto de determinar la distribución del uranio y de obtener al mismo tiempo, mediante el recuento total de la placa, el contenido total de dicho elemento. Se analizaron algunas muestras desconocidas con el procedimiento que consiste en medir la autoabsorción y compararla con un patrón conocido de uranio-235. La adición de un cristal de pozo permitió analizar soluciones diluidas de uranio-235 con una precisión suficiente para el muestreo de desechos y la evaluación de la criticidad.

The primary purpose behind the gamma counting technique to be described was to check on a reactor fuel fabricator's $^{235}\text{U}$ weights as assigned to fuel plates in production. Since no practical method was available to verify the $^{235}\text{U}$ content of fuel elements after delivery, it was necessary to perform some sort of verification during production at the fabricator's plant. The use of a destructive technique on random samples had been used in the past, but this method is both expensive and time consuming considering the relatively small coverage provided.

Since the particular fuel alloy involved was aluminium with a nominal 30 wt.% fully enriched uranium, the counting of the gamma rays from the $^{235}\text{U}$ provided the possibility of a non-destructive assay method. To be practical for inspection purposes, however, the technique had to be simple and fast enough to be performed at the fabricator's plant without undue interference with production. It was assumed that suitable standards could be obtained for use in quantitative comparison with production fuel plates.

As a start, the gamma-counting equipment pictured in Fig. 1 was purchased. It consists basically of two separate single-channel analyser systems with a single timer and a single ratemeter. The dual system was acquired to speed up the inspection by allowing the counting of two items at one time and to provide a safety factor in the event of one system failing. The equipment is constructed of solid state components in modular form and has complete flexibility for the interchange of modules and the varying of connections. The ratemeter can be used to monitor either system as de-
THE RATEMETER, WHILE USED WITH THE RECORDER FOR FUEL PLATE SCANNING, HAS BEEN INDISPENSIBLE FOR RAPIDLY MAKING DESIRED ANALYSER SETTINGS AND FOR QUICK CHECKING OF BACKGROUND AND GENERAL COUNTING LEVELS.

The lead bricks with holes of one square inch area are used when making absolute U-235 measurements of an unknown versus a standard foil. The results obtained are in grams/square inch for convenience.

FIG. 1. Dual single-channel analyser with ratemeter and recorder.

sired and provides an easy means of adjusting the analyser settings. It also provides a count rate output for recording purposes. The separate counting systems can be controlled by the timer, either singly or together, or they can be controlled manually, or by an automatic external switch.

In addition to checking on the $^{235}\text{U}$ total weight, it was also desired to check the uniformity of the $^{235}\text{U}$ distribution in a fuel plate. For this purpose, the scanning device pictured in Fig. 2 was constructed and a chart recorder was added to receive the output of the count ratemeter. The scanning device was provided with a synchronous-motor drive to ensure a constant traversing speed. All other features, such as the crystal holder, the lead collimator and automatic stops were adjustable.

To keep the counting system reasonably portable, heavy lead shielding around the crystal was not desirable. Since most counting would be performed with a relatively low background, a brass tube was made to hold the integral crystal and photomultiplier tube with about 0.125 in of lead filling the section immediately around the crystal. The bottom of the crystal was
open and flush with the bottom edge of the brass holder. The holder itself was simply clamped in a ring stand and positioned as desired.

Initial attempts to count some $^{235}\text{U}$ samples were performed with the analyser centred on the 184 keV gamma peak characteristic of $^{235}\text{U}$ and with a narrow window of a few volts. This arrangement, however, posed problems due to shifting of the peak settings and the relatively low count rates obtained. To eliminate these problems, the threshold was lowered and the window was opened. The adjustments were made arbitrarily using the count rate meter for an indication of when the optimum count rate was achieved. The final settings, which were standardized upon for this particular equipment, were a threshold of about 40 keV and a window of about 400 keV, but no attempts were made to obtain accurate calibrations.

In addition to providing a large increase in the count rate, these settings minimized the effects due to shifting, even after initial start-up of the systems. Besides reducing the required counting time, it was possible to proceed with actual counting within a half hour after start-up. Considering the fact that the equipment was to be transported periodically to the fabricator's plant and be set up for short periods, these were significant advantages. The increased count rate from the $^{235}\text{U}$ was accompanied by an increase in the background count rate. However, since the proposed method of inspection was to be based upon the total counts obtained, this increase posed no problem as long as the background remained essentially constant. The statistical variations in the background count rate had little effect on the total count rate and there was a net decrease in the overall statistical variation at the higher count rate. As a result, increased accuracy was gained together with a shorter counting time.
Having established the counting method, the next problem was to decide what would be counted and to establish the geometry. While it would be possible to count a finished fuel plate prior to assembly into a fuel element, the large size, about 3 in wide by 2 ft long, posed problems for a suitable geometry. Before being made into clad fuel plates, the uranium-aluminium alloy was available in production as a fuel plate core about 2 in by 2.5 in by 0.160 in thick. These fuel cores offered reasonable counting geometry and were readily available in the production line. The only precaution required for handling was to place the cores in thin plastic bags to prevent the spread of radioactive contamination.

To eliminate the need for accurate positioning of the cores during counting, the crystal was raised to a height of eight or nine inches. At this height, the crystal could "see" the whole core and minor displacements in core positioning did not significantly affect the total count. While the use of this approach allowed simple hand positioning of the core, caution was required to ensure no change in the general surroundings which would cause scattering of the gamma rays from the core. The use of a pair of plastic stops for positioning of the cores was found to give a 2% increase in the counts observed. It was also found that an individual who stood next to the crystal gave a 4% increase. While the basic counting geometry was very simple, it was important that nothing be moved or changed in the immediate surroundings during the counting of a series of cores.

The counting technique and associated geometry described so far had been arrived at with the use of some samples of uranium on hand, since actual production cores were not yet available. The lack of such cores meant that quantitative standard cores could not be obtained before the first visit to the fabricator. However, since the total core counts obtained with the system described should be directly proportional to the $^{235}$U content of the cores, it was decided to use an alternative approach. If the fabricator's $^{235}$U weight assignments for the cores were accurate, a plot of his $^{235}$U weights versus the core counts should yield a straight line. While the lack of known standards prevented obtaining quantitative results, this approach would at least permit a qualitative check on the fabricator's results. To obtain a representative total count, each core was counted for two minutes on both sides. In only a few instances did these two counts vary significantly, indicating some possibility of non-uniformity in the $^{235}$U distribution through the thickness of a core. The two counts were then added to give a total count in the range of $5 \times 10^5$ to $10^6$. At this level, the counting error was estimated to be about $\pm 0.2\%$.

The results of the first ten cores counted at the fabricator's plant are shown in Fig. 3. These cores were selected at random from different $^{235}$U weight ranges in order to achieve a reasonable spread in the data. As can be seen, the data for six of the cores gave a very good straight line, as anticipated. The fabricator, who also used a gamma counting technique, rechecked the four cores which did not fall on the line. With his revised data, two cores fell on the line as shown but the other two remained essentially unchanged. The two corrections were apparently due to human error. A check of radiographs of the other unchanged cores, however, revealed the uranium to be non-uniformly distributed. The fabricator's assay system was determined to be affected significantly by this non-uniformity.
The particular fuel element being made by the fabricator had side plates with one half the $^{235}\text{U}$ content of the central plates. The first cores counted had been approximately 0.160 in thick and were for use in the central fuel plates. For the side plates, the fabricator had similar cores which were half as thick, about 0.080 in, and with half of the $^{235}\text{U}$ content. A number of these thin cores were counted at random from the production line. The initial data obtained are shown as circles in Fig. 4. Unlike the data for the thick cores, it was not possible to draw a straight line through these points. It was finally determined that a series of parallel straight lines could be approximated through these points and that these lines appeared to be related to the actual core thickness.

At this point a detailed review of the fabricator's assay system revealed an error causing discrepancies due to variations in core thickness. Similar discrepancies had not been evident in the $^{235}\text{U}$ assignment to the thick cores because with these the thickness variations had less effect. The discovery of this error necessitated not only a revision in the fabricator's procedure for $^{235}\text{U}$ assignment but also a re-evaluation of all fuel cores and plates that had already been produced. The fabricator's revised data for the thin cores inspected are shown as crosses in Fig. 4 with a solid line drawn through the best points. With the exception of three points, the agreement is fairly close. Some variation is to be expected since the production technique used by the fabricator was not as precise as the counting procedure used for inspection.
It should be noted here that the results obtained up to this point in the inspection had been achieved on a qualitative basis without the use of standards. These results were based upon the total count obtained being directly proportional to the $^{235}\text{U}$ content by minimizing the effects of other variables. The normal variations of core thickness and uranium distribution to be expected in production did not appear to affect the accuracy significantly. The effects introduced when using thicker cores or higher weight percents of $^{235}\text{U}$ and when cladding is present were not investigated.

To provide a quantitative check, some production cores with a known $^{235}\text{U}$ content were needed. Four such cores were obtained, two for destructive analysis and two for counting standards. Prior to the destructive analysis, the four cores were carefully compared by repeated gamma counting to establish their relative $^{235}\text{U}$ contents. Because of dissolution problems, two cores were chemically analysed and only one core could be kept as a standard instead of two as planned. The $^{235}\text{U}$ content of this one core was calculated from its gamma counting relationship to each of the two cores destructively analysed. The two results obtained were within 0.3% of each other and the average was assigned as the $^{235}\text{U}$ content of the standard. The use of this single standard in conjunction with the previously established counting technique now provides an accurate and simple non-destructive inspection procedure for use at the fabricator's plant.

With the $^{235}\text{U}$ content of the fuel plates under control, an attempt was made to determine the uniformity of the $^{235}\text{U}$ distribution by gamma scanning.
finished flat fuel plates. The same counting equipment was used with the addition of the scanning device and chart recorder previously mentioned. As can be seen from Fig. 2, the same brass tube crystal holder is used by placing it in the traversing head. The crystal is adjusted to be as close to the fuel plate as is permitted by the one-half inch thick lead collimator. The opening in this collimator is a slit which can be adjusted from fully closed to about one inch wide. An additional 0.5 in of lead shielding was placed on each side of the crystal to shield it from radiation in the direction of the fuel plate length. No lead was used on the front or back sides of the crystal. The traversing head was provided with adjustable automatic stops in both directions, mainly to protect the drive mechanism.

Two microswitches were added to the front of the traversing head. One, which was activated by two simple stops, controlled a scaler so that a total count could be taken during a scan. This total count is indicative of the total \( ^{235}\text{U} \) content of the fuel plate. The other microswitch is activated by tack heads in a ruler and is used to produce marker blips on the right hand side of the chart recording. These marker blips allow direct correlation of the chart readings with corresponding positions on the fuel plate.

With this scanner it was possible to detect an overall variation, such as a gradient in the uranium distribution and some relatively large local perturbations. In practice, the interdependence of the traversing speed, the collimator width, the count rate and the ratemeter time constant places a practical limit on the percentage change in \( ^{235}\text{U} \) content detectable over a local area. For instance, in the radiographic inspection of a fuel plate a high concentration of \( ^{235}\text{U} \) in a spot is acceptable if it represents less than a 5% increase in \( ^{235}\text{U} \) over a 0.125 in diameter area. With a collimator slit 0.5 in wide, the area "seen" by the crystal is well over one square inch so that such a spot would not register in the scanning. The use of a very narrow slit increases the relative effect of such local spots, but the corresponding loss in count rate necessitates the use of a longer time constant to reduce the statistical variations. The narrow slit also means that such local spots are "seen" by the crystal for a shorter time which, when coupled with the longer time constant required for counting, nullifies the initial relative gain from the narrower slit. The only real gain in sensitivity has to be made by reducing the traversing speed.

Figure 5 shows the results of a typical plate scan with a 0.5 in collimator slit. For display purposes, the trace of a scan at a long time constant is superimposed on the same trace of the same scan at a short time constant. While the longer time constant results in some loss of response, especially at the beginning of the trace, it offers much less statistical variation. For the 10^5 counts/min count rate shown, the approximate variation expected at the short time constant of 0.5 sec is ± 3% and at the long time constant of 5.0 sec, ± 1%. The many possibilities of this scanning procedure were not investigated for the lack of time and standard plates. Similar work [1] on fuel plate scanning indicates that it is possible to obtain the total plate \( ^{235}\text{U} \) content and the relative uranium distribution in one operation.

As mentioned in the beginning, the primary purpose of this investigation was to develop a practical non-destructive technique which would ensure the quality of delivered fuel elements for use in a reactor. It is obvious, how-
FIG. 5. Sample gamma scan of a nominal 5 g ($^{235}$U) fuel plate. For both traces, collimator slit 0.5 in. wide and full scale represents $10^5$ counts/min (trace A: 0.5 sec time constant; trace B: 5.0 sec time constant)

ever, that the requirements for good nuclear material management were achieved at the same time. It was logical, therefore, to extend the use of this equipment to other unirradiated uranium-235 as a non-destructive means of inventory verification.

For many years there has been use of a technique to determine the $^{235}$U content of uranium foils by measurement of the self-absorption factor at the 184 keV gamma peak and comparing the corrected gamma count of the unknown foil to that of a known $^{235}$U standard. The single channel analyser is readily adapted to this technique by setting the analyser on the 184 keV peak and using a narrow window. Since the size of most items to be analysed is different than the standard used for comparison, it was decided to use a set-up which would yield results directly in grams of $^{235}$U per square inch. This was accomplished by using a 1.5 in thick lead brick with a hole of one square inch in area bored through the centre. In practice, two such bricks are superimposed with their holes aligned. The unknown and the standard are placed between the bricks for comparative counting. For measurement of the self-absorption factor of the unknown, the standard is placed under the bottom brick where it acts simply as a source of radiation and is counted with and without the unknown in place between the bricks.

The results obtained by this method were exceptionally good. A 0.002 in 93% enriched uranium metal foil about 2.5 in in diameter was available with a known $^{235}$U content. This foil was first used to examine some 93% enriched uranium-aluminium alloy consisting of 10 wt.% uranium, 0.058 in thick, and 20 wt.% uranium, 0.065 in thick. Four pieces of each alloy, about two inches square, were compared to the standard $^{235}$U foil to obtain an average value of grams $^{235}$U per square inch. Two pieces of each alloy were then destruc-
tively analysed. The gamma data had indicated values of 0.283 and 0.574 g-(\textsuperscript{235}U)/in\textsuperscript{2}. The corresponding values by destructive chemistry were 0.276 and 0.572 g(\textsuperscript{235}U)/m\textsuperscript{2}.

The total \textsuperscript{235}U content of an available flat fuel plate was then determined by measuring the grams \textsuperscript{235}U per square inch at one and two inch intervals along the length and then integrating these values over the fuel area as measured from a radiograph of the plate. As an exercise, the \textsuperscript{235}U concentration at each point was obtained by comparing to both the standard \textsuperscript{235}U metal foil and to a piece of the 0.572 g/in\textsuperscript{2} uranium-aluminium alloy foil. The comparable values obtained using these two different standards agreed within one percent or less in all cases. The final \textsuperscript{235}U total value calculated for the fuel plate was 16.1 g as compared to the fabricator's value of 15.9 g.

The addition of a well-type crystal was then made to provide for the examination of the \textsuperscript{235}U content of solutions, using the same gamma counting equipment and techniques. The use of a well-type crystal permitted a standard size glass vial to be used as a sample holder and gave a fixed counting geometry. To obtain some reference samples, a solution was made with a concentration of 0.08 g \textsuperscript{235}U/mL by dissolving a known quantity of 93% enriched uranyl nitrate in a known volume of water. The solution was then diluted to provide a series of lower concentrations. A set of vials was filled with 5 ml aliquots making six reference samples with total \textsuperscript{235}U contents ranging from 0.01 to 0.40 g. Another set of vials was filled with one millilitre aliquots making five samples with total \textsuperscript{235}U contents ranging from 0.003 to 0.08 g. These samples were then gamma counted using a narrow window at the 184 keV gamma peak. The results were not linear because of increased self-absorption at the higher concentrations. Analyses of unknown solutions must, therefore, be related to samples of comparable \textsuperscript{235}U concentration and the unknown can generally be diluted as required for this purpose. While this method of assaying for \textsuperscript{235}U is subject to inaccuracies due to the possible presence of unknowns and variations in self-absorption, it is useful in many instances for purposes of inventory verification and nuclear safety control.

In summary, the use of the basic counting equipment described provides a rapid and non-destructive means for the measurement or the approximate mensuration of \textsuperscript{235}U contents. The main problem with its use is the adaptability of an item to the counting requirements but this is generally not a serious restriction. The specific applications reviewed in this paper are typical and indicative of the simplicity, portability and flexibility of the equipment and the accuracy of the results obtainable. Further details on various techniques are available in the references listed [1-8].

REFERENCES

DISCUSSION

C. F. METZ: It may be of interest that at Los Alamos we have developed a non-destructive technique for measuring uranium-235 in rod-type elements. The instrument used consists of a sodium iodide thallium-activated scintillator detector. Matched phototubes are connected in parallel and the total current is integrated. The read-out is from a digital voltmeter. Ten seconds suffice to measure the uranium-235 in a fuel element. Quantitative measurements have a relative standard deviation of 0.3%. This new technique is being reported this week at a meeting of the Society for Applied Spectroscopy. It would have been reported here but time limitations did not make it possible. I assume an abstract will appear in Nuclear Science Abstracts.

P. COLSMANN: This technique is certainly practical, but it involves problems of geometry, i.e. size and shape of fuel element, and it is costly and difficult to obtain a standard element for reference purposes. So far we have found it more practical to check the uranium-235 content of a single fuel plate. We hope to establish a simplified version of your technique to serve as secondary check for delivered fuel elements.

C. L. A. BUECHLER: I wonder whether any of the participants have experience in the application of this technique to more complex geometries than flat plates or cylindrical rods.

J. T. BYRNE: In response to Mr. Buechler's question, I can say that the 184-keV $\gamma$-peak provides a useful measurement for uranium in a cold, enriched-uranium scrap recovery plant. Reference [2]* for Mr. Colsmann's paper (SM-67/47) describes this method, which has been used at Rocky Flats for about 10 years. It has proved very economical and sufficiently precise. The sample is dissolved and the solution counted in a well crystal (NaI(Na)).

A. J. FUDGE: I would like to point out another use of this technique of non-destructive determination of uranium. Before a non-destructive determination of burn-up by gamma spectrometry can be carried out, it is necessary to know the original distribution and amount of fissile material in the fuel element — information which is provided by this technique. It is to be hoped that it will be extended to all fuel elements to be irradiated.

---

* USAEC Rpt TID-7581.
It does not seem to be generally known that this technique is also applicable to the non-destructive determination of plutonium-239. The composite peak at 384 keV is used, but it should be remembered that the abundance of the gamma rays is much less than for uranium-235.

D. E. GEORGE: I should also like to recall that in paper SM-67/37 Dr. McDowell alluded to the use of the gamma spectrometer to measure uranium-235 in several sizes of filter. Perhaps Mr. Highfill would also comment on this application.

J. P. HIGHFILL: New York Operations Office first worked with 8-in absolute air filters and later with 12-in filters. I believe the uranium-235 results obtained from gamma-ray work agreed within 5-10% with the results obtained by destructive methods.

Work has recently been completed on 24-in air filters, but I have not seen the data from this job.
AN APPRAISAL OF ANALYTICAL METHODS FOR PLUTONIUM AND THEIR APPLICATIONS TO THE ANALYSIS OF NUCLEAR MATERIALS

G. W. C. MILNER AND G. PHILLIPS
ATOMIC ENERGY RESEARCH ESTABLISHMENT, HARWELL, BERKS., UNITED KINGDOM

Abstract — Résumé — Аннотация — Resumen

AN APPRAISAL OF ANALYTICAL METHODS FOR PLUTONIUM AND THEIR APPLICATIONS TO THE ANALYSIS OF NUCLEAR MATERIALS. A variety of methods is available for the determination of the plutonium content of nuclear materials. For milligram amounts of plutonium the available methods include differential spectrophotometry using the colour of Pu(III), gravimetry based on PuO₂, gamma counting, and redox methods including potentiometric and amperometric titrations and controlled-potential coulometry. Alpha counting, isotopic dilution, and polarographic methods are suitable for microgram amounts. Some methods are more suitable than others for certain types of sample and the analyst is faced with difficult selection problems to achieve the best results. The advantages and limitations of the above methods are described in the light of a varied experience with them over many years at AERE, together with comments on accuracy, precision, sensitivity and features of special interest. Some of the methods cannot be applied without some separation of the plutonium from other sample constituents, and comments are made from experience in the use of anion exchange and reverse-phase chromatographic techniques for this with emphasis on their suitability for radioactive samples. Consideration is given to the many problems that have arisen in the successful application of these methods to the analysis of plutonium alloys, ceramics and cermets in various systems containing uranium, thorium, iron, chromium, molybdenum, cerium and cobalt. Difficulties occurring in the dissolution of samples and in the adjustment of the plutonium to the required valency state are described. The respective merits of dissolving in mixtures of common mineral acids and with the aid of fusion techniques are commented upon with examples. Outlines of procedures, together with analytical results for typical samples, are given for the analysis of Pu-U, Pu-Ce-Co and Pu-U-Mo alloys, Pu-U oxides and carbides, and Pu-U carbide cermets with Fe, Mo and Cr. These materials have arisen from metallurgical research and development programmes for new nuclear fuels.

EVALUATION DES METHODES ANALYTIQUES DE DOSAGE DU PLUTONIUM ET DE LEUR APPLICATION A L'ANALYSE DES MATIERES NUCLEAIRES. Il existe plusieurs méthodes de dosage de la teneur en plutonium des matières nucléaires. Pour les quantités de l'ordre du milligramme, les méthodes utilisables sont la spectrophotométrie différentielle fondée sur la couleur de Pu(III), la gravimétrie fondée sur PuO₂, le comptage gamma et les méthodes de réduction/oxydation comprenant les titrages potentiométriques et ampèremétriques et la coulombométrie à potentiel constant. Pour les quantités de l'ordre du microgramme, le comptage alpha, la dilution isotopique et les méthodes polarographiques sont à utiliser. Certaines méthodes conviennent mieux que d'autres à des types déterminés d'échantillons et l'analyste soucieux d'obtenir les meilleurs résultats se heurte à un choix difficile. Les auteurs exposent les avantages et les inconvénients des méthodes citées tels qu'ils se sont dégagés de l'expérience acquise au cours des années à l'Atomic Energy Research Establishment, et ils discutent l'exactitude, la précision, la sensibilité de ces méthodes, et d'autres caractéristiques présentant un intérêt particulier. Certaines méthodes ne peuvent être utilisées si l'on n'a, dans une certaine mesure, séparé le plutonium des autres constituants de l'échantillon et le mémoire commente l'expérience acquise avec l'échange d'anions et les procédés de chromatographie à phase inversée utilisés à cette fin, en insistant surtout sur la mesure dans laquelle cette méthode convient aux échantillons radioactifs. Les auteurs étudient en outre les nombreux problèmes qui se sont posés lors de l'application (d'ailleurs couronnée de succès) de ces méthodes à l'analyse des alliages de plutonium, des céramiques et des cermets dans différentes combinaisons contenant de l'uranium, du thorium, du fer, du chrome, du molybdène, du cérium et du cobalt. Ils exposent les difficultés de la dissolution des échantillons et de la réduction du plutonium à l'état de valence voulu, ainsi

691
ОЦЕНКА АНАЛИТИЧЕСКИХ МЕТОДОВ ОПРЕДЕЛЕНИЯ ПЛУТОНИЯ И ИХ ПРИМЕНЕНИЕ ДЛЯ АНАЛИЗА ЯДЕРНЫХ МАТЕРИАЛОВ. Существуют различные методы определения содержания плутония в ядерных материалах. Для определения мириаграммовых количеств применяют методы, включая дифференциональную спектрофотометрию с использованием цвета Pu (III), гравиметрию, основанную на PuO₂, гамма-счет и методы окисления-восстановления с потенциометрическим и амперометрическим титрованием и контролируемой потенциалиметрией. Альфа-счет, изотопное разбавление и полярографические методы вполне пригодны по сравнению с другими для некоторых образцов, лицо, производящее анализ, сталкивается с трудными проблемами выбора метода для получения наилучших результатов. Преимущества и недостатки вышеуказанных методов описываются в свете разнообразного опыта, накопленного при работе с ними в течение многих лет в атомном научно-исследовательском центре в Харуэлле, а также излагаются замечания о точности, четкости, чувствительности и конструктурских особенностях, представляющих особый интерес. Некоторые методы невозможно применить без определенного отделения плутония от других составных частей образца. В связи с этим излагаются замечания на основе опыта по использованию анионного обмена и хроматографического способа обратной фазы, причем особое внимание обращается на их пригодность для радиоактивных образцов. Уделяется внимание многим проблемам, которые возникают при использовании этих методов для анализа сплавов плутония, карбидов и карбидных керметов в различных системах, содержащих уран, торий, железо, хром, молибден, церий и кобальт. Описываются трудности, возникающие при растворении образцов и при подгонке плутония к требуемому состоянию валентности. В докладе на примерах комментируются соответствующие преимущества растворения в смеси обычных минеральных кислот с помощью плавления. Излагаются процедуры с одновременным описанием аналитических результатов для типичных образцов с целью использования их для анализов сплавов Pu-U, Pu-Се-Co и Pu-U-Mo, окисей и карбидов Pu-U карбидных керметов Pu-U с добавлением Fe, Mo и Cr. Эти материалы получены на основе металлургических исследований и программных разработок для нового ядерного топлива.

METODOS ANALITICOS DE DETERMINACION DEL PLUTONIO Y SU EMPLEO EN EL ANALISIS DE MATERIALES NUCLEARES. — Para determinar el contenido de plutonio en los materiales nucleares existen diversos métodos. Si se trata de cantidades del orden del miligramo se pueden emplear la espectrofotometría diferencial por el color del Pu(III), la gravimetría basada en el PuO₂, el recuento gamma y métodos de oxidorreducción como las valoraciones potenciométricas o amperimétricas y la coluomembrética de potencial controlado. Si se trata de microgramos son preferibles el recuento alfa, la dilución isotópica o las técnicas polarográficas. Teniendo en cuenta que unos métodos son más adecuados que otros para ciertos tipos de muestra, el analista tiene que resolver un difícil problema de selección a fin de obtener los mejores resultados posibles. Los autores exponen las ventajas y las limitaciones puestas por el uso de esos métodos y dan sus limitaciones, sus sensibilidades y otras cuestiones de especial interés. Como algunos de esos métodos exigen la separación previa del plutonio, los autores estudian el empleo de las técnicas de intercambio aniónico y de cromatografía en fase inversa, y en particular su conveniencia para el análisis de muestras radiactivas. Examinan los muchos problemas que han surgido al analizar por estos métodos aleaciones, productos cerámicos y cermetes de plutonio en diversos sistemas que contienen uranio, torio, hierro, cromo, molibdeno, cerio y cobalto. La memoria trata además de las dificultades que se encuentran al disolver las muestras y al llevar el plutonio al estado de valencia deseado. Comenta con ejemplos las ventajas y los inconvenientes de disolver las muestras en mezclas de ácidos inorgánicos corrosivos o mediante técnicas de fusión. Los autores exponen someramente los procedimientos de análisis y dan los resultados obtenidos con muestras típicas de aleaciones de Pu-U, Pu-Се-Co y Pu-U-Mo, óxidos y carburados Pu-U, y cermetes carburados de Pu-U con Fe, Mo y Cr. Estos materiales han sido surgiendo de los programas de investigación metalúrgica y de desarrollo de nuevos combustibles nucleares.
1. INTRODUCTORY DISCUSSION

Investigations are carried out at A.E.R.E. on newer types of nuclear fuel containing plutonium, and these require analytical support, particularly for the determination of plutonium contents. When work first started about ten years ago, investigations were limited to binary alloys of plutonium with such metals as uranium, aluminium, iron, etc. The required precision depended on the plutonium content, decreasing approximately linearly from ±10% at the 1% Pu level to ±0.10% at the 99% Pu level. Several analytical methods were available for plutonium at that time, including potentiometric titration, gravimetry, alpha counting and direct spectrophotometry, but none of these could be applied directly to all the alloys under investigation. For example, uranium and iron interfered with the potentiometric titration using the Pu$^{3+}$-Pu$^{4+}$ couple. Gravimetry based on ignition to PuO$_2$ was only applicable after the quantitative separation of plutonium from other sample constituents, and alpha counting needed improvement to give an acceptable precision. In 1955 differential spectrophotometry was shown at A.E.R.E. to be an excellent technique for the analysis of uranium-base alloys for uranium content [1]. Although this technique lacked sensitivity, it was particularly suitable for alloy analysis owing to its freedom from interferences, and as a consequence its applicability to the direct determination of plutonium in alloys was studied by Phillips [2]. It was found to be satisfactory for the direct analysis of a variety of binary alloys, but the separation of plutonium was necessary for some samples and an anion exchange procedure proved to be suitable for this purpose [3].

Although metallurgical studies have been diversified over recent years to include more complex alloys, ceramics and cermets, the differential spectrophotometric method for plutonium has been applied wherever possible. With such systems, however, some independent checking of results has been required and, for this purpose, investigations have been carried out on newer and more absolute methods, including controlled-potential coulometry and potentiometric titration based on the Pu(VI)-Pu(IV) system. These latter methods have also proved to be more suitable for the analysis of samples requiring the use of sulphuric acid to dissolve them.

For materials containing plutonium as a minor constituent (<1% Pu), alpha counting was chosen initially and found to be satisfactory for most samples. More recently, the need has arisen for results of higher precision and for check analyses by different methods, and so investigations have been carried out with such techniques as isotopic dilution analysis and AC polarography.

Many of the above developments are considered in subsequent sections of this paper, together with typical results for the analysis of various materials.

2. METHODS FOR MACRO-AMOUNTS OF PLUTONIUM IN METALLURGICAL SAMPLES

2.1. Spectrophotometric methods

The spectrophotometric method for measuring the valency states of plutonium is well known [4], and the determination of the total plutonium con-
tent can be readily achieved by an extension of this involving adjustment of all valencies to a single state. Ashley and Allison [5] reported such a determination by measuring the peak at 476 nm for Pu(IV) in 3M HNO₃, but unfortunately this peak proved to be particularly sensitive to acidity. From work at A.E.R.E., it was concluded that complete reduction of plutonium to the trivalent state would be easier to carry out than adjustment to the tetravalent state, and that trivalent plutonium would be less sensitive to changes in acidity than the tetravalent species. A method using the 603 nm peak for Pu(III) was tested by taking spectrophotometric measurements on solutions against a solvent blank, but the precision and accuracy of the results was not usually better than ±1% [6].

A detailed study of differential spectrophotometry was carried out in an effort to achieve some improvement, and conditions were established to produce results to within ±0.1% [2]. Of the two major absorption peaks for Pu(III) at 565 and 603 nm respectively, the former was chosen as being the more suitable for differential measurements owing to the lower temperature coefficient and the lower susceptibility to changes in anion concentration at this wavelength. The temperature of sample and standard solutions was controlled to within 25 ± 0.5°C to obtain the best precision, but in routine application of the method the need to thermostat solutions in a glove box proved very inconvenient. The practice of measuring the slope of the calibration graph for each determination has now been adopted with satisfactory results [7]. In this the standard and sample solutions are allowed to equilibrate to the temperature of the glove box before measurement. Although differential spectrophotometry results in a convenient method, at least 30 mg of plutonium are required for each sample, together with an adequate supply of pure plutonium for the preparation of reference standards.

Differential spectrophotometry has been applied successfully to the determination of the plutonium content of alloys (binary and ternary), and ceramic materials. Details are given below.

Pu-Al. This binary alloy system consisted of fuel element plates containing 20% plutonium in aluminium. For this ratio of aluminium to plutonium, there is a slight reduction in the extinction coefficient for Pu(III). Most determinations were based therefore on the separation of plutonium by anion exchange from 8M HNO₃ before spectrophotometry, although some direct determinations were carried out using plutonium standards containing the requisite amounts of aluminium [8]. The spectrophotometric results were compared against those obtained by potentiometric titration of Pu³⁺ to Pu⁴⁺ using standard ceric sulphate solution [12] with the good agreement shown in Table I.

Pu-Mo-U and Pu-Ce-Co. The ternary alloys consisted of Pu-Mo-U with uranium as the major constituent and Pu-Ce-Co with plutonium contents ranging from 15–80%. In the analysis of all samples, a suitable weight was dissolved and then plutonium was separated from other constituents by the anion exchange procedure. For Pu-Mo-U alloys [9], a sample containing 50–150 mg of plutonium was dissolved in 3.5 ml 5.5M HCl followed by the addition of 3.5 ml HNO₃ (sp.gr. 1.42) to complete the solution. After dilution to 15 ml with 8M HNO₃, the solution was passed through a 2 ml column
### TABLE I
RESULTS FOR PLUTONIUM IN 20% Pu-Al FUEL PLATES

<table>
<thead>
<tr>
<th>Sample No.</th>
<th>Analytical method for plutonium</th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Differential spectrophotometry</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Potentiometric titration</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Pu (%)</td>
<td>c. v. (%)</td>
<td>No. of determins</td>
<td>Pu (%)</td>
<td>c. v. (%)</td>
<td>No. of determins</td>
</tr>
<tr>
<td>1</td>
<td>19.54</td>
<td>0.15</td>
<td>4</td>
<td>19.43</td>
<td>0.20</td>
<td>4</td>
</tr>
<tr>
<td>2</td>
<td>19.08</td>
<td>0.15</td>
<td>4</td>
<td>19.11</td>
<td>0.30</td>
<td>4</td>
</tr>
<tr>
<td>3</td>
<td>19.38</td>
<td>0.25</td>
<td>4</td>
<td>19.36</td>
<td>0.30</td>
<td>4</td>
</tr>
</tbody>
</table>

### TABLE II
THE ANALYSIS OF Pu-U-Mo AND Pu-Ce-Co ALLOYS

<table>
<thead>
<tr>
<th>Pu-U-Mo</th>
<th>Composition by analysis (%)</th>
<th>Total (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sample No.</td>
<td>Pu</td>
<td>Mo</td>
</tr>
<tr>
<td>1</td>
<td>11.6 ± 0.1</td>
<td>5.7 ± 0.05</td>
</tr>
<tr>
<td>2</td>
<td>11.8 ± 0.1</td>
<td>5.5 ± 0.05</td>
</tr>
<tr>
<td>3</td>
<td>11.6 ± 0.1</td>
<td>5.6 ± 0.05</td>
</tr>
<tr>
<td>4</td>
<td>6.5 ± 0.05</td>
<td>17.1 ± 0.15</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Pu-Ce-Co</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Sample No.</td>
<td></td>
</tr>
<tr>
<td>1</td>
<td>26.4 ± 0.25</td>
</tr>
<tr>
<td>2</td>
<td>84.5 ± 0.25</td>
</tr>
<tr>
<td>3</td>
<td>15.4 ± 0.15</td>
</tr>
<tr>
<td>4</td>
<td>50.0 ± 0.25</td>
</tr>
</tbody>
</table>

of Deacidite FF anion exchange resin. A similar procedure was used for the Pu-Ce-Co alloys with slightly larger volumes of 5.5M HCl and concentrated HNO₃ [10]. Results for the analysis of typical alloys are given in Table II and all results are quoted with the range of uncertainty at the 1σ level. The values for plutonium were not checked by an independent method, but the satisfactory totals for all constituents were taken as good evidence of the validity of these results.
PuO₂. In studies with ceramic materials, work has been concentrated on oxide and carbide systems. The analysis of oxide samples has produced some problems owing to difficulties in the dissolution of PuO₂, especially high-fired materials. Although PuO₂ can be dissolved in HNO₃+HF mixtures, the procedure is time consuming and uncertain. Recent work [11] has resulted in the development of a fairly rapid sinter technique for the dissolution of samples prior to the application of differential spectrophotometry. In this method 100 mg of PuO₂ are intimately mixed with 0.5 g Na₂O₂ in a platinum crucible and heated at 400-420°C for 10 min. After the addition of 0.5 g NaOH, heating is continued for a further 10 min. Then the melt is extracted with water and acidified to give 50 ml of solution, molar in HCl and 5% in NH₂OH HCl. The plutonium content is determined by differential measurements at 565 nm using 4 cm cells. Very satisfactory results have been obtained with this method on PuO₂ specimens ignited at 850°C and 1550°C respectively. Typical results were 99.81% Pu for PuO₂ at 850°C (c.v. 0.3%) and 99.85% for PuO₂ ignited at 1550°C (c.v. 0.2%). This method is also applicable to the dissolution and analyses of PuO₂-UO₂ materials.

(U₀.₈₅Pu₀.₁₅)-C. The dissolution of actinide carbides in mineral acids, although less difficult than for the corresponding oxides, is complicated by the formation of soluble and insoluble organic products. Attempts to determine plutonium directly in such solutions by differential spectrophotometry have proved unsuccessful. The only satisfactory approach to this problem is dependent on the dry oxidation of each sample by slowly heating to 850°C in air, followed by dissolution in HNO₃ with the addition of a few drops of HF. Sample solutions are then evaporated to remove HF and as much of the HNO₃ as possible, before dilution to known volumes with M HCl containing 5% NH₂OH HCl. A series of spectrophotometric determinations on a selected batch of crushed arc-melted material gave a precision of 0.76%, which is not as good as can be expected from a differential spectrophotometric method. However, these results were confirmed by further series of analyses by other techniques including controlled-potential coulometry and potentiometry (see Table III), and it was concluded that the poor precision was a consequence of inhomogeneity in the sample. For the control

<table>
<thead>
<tr>
<th>Method of analysis</th>
<th>Pu content (%)</th>
<th>Coefficient of variation</th>
<th>No. of determinations</th>
</tr>
</thead>
<tbody>
<tr>
<td>Differential spectrophotometry</td>
<td>13.96</td>
<td>0.76</td>
<td>6</td>
</tr>
<tr>
<td>Potentiometric titration</td>
<td>14.02</td>
<td>0.93</td>
<td>6</td>
</tr>
<tr>
<td>Controlled-potential coulometry</td>
<td>14.04</td>
<td>1.3</td>
<td>5</td>
</tr>
</tbody>
</table>

1 Coefficient of variation.
analysis of production material of this type, the differential spectrophotometric method is preferred because it is direct and fairly quick.

2.2. Electrochemical methods

2.2.1. Potentiometric titrations

Two possible procedures exist for the potentiometric titration of plutonium, the first being based on the oxidation of Pu(III) to Pu(IV) and the second on the reduction of Pu(VI) to Pu(IV). Titrations in the first category have received much attention and satisfactory results have been obtained at A.E.R.E. by using chromous sulphate as the reductant followed by titration with ceric sulphate [12]. There is interference from many elements, including uranium and iron, and consequently the method is only applicable to favourable systems.

Titrations in the second category are more attractive because of the 2-electron change involved. A suitable procedure involves quantitative oxidation to the Pu(VI) state, followed by the addition of ferrous iron in a slight excess and titration of this excess with ceric sulphate to a potentiometric endpoint. It is only in recent times that a convenient oxidant has become available, and argentic oxide is suitable for this purpose because any excess can be destroyed readily by warming the solution. In a high precision method developed at A.E.R.E. [13], ferrous sulphate solution is added by weight, and the amount in excess is then titrated by adding standard ceric sulphate automatically from an Agla microburette to a potentiometric endpoint. The main advantage of this approach is the non-interference of such elements as uranium and iron. Interferences do occur, however, particularly from elements which are oxidized by argentic oxide, such as chromium, manganese and cerium.

The oxidation procedure consists of the addition of three successive 25 mg amounts of argentic oxide at 5 min intervals to 5 ml of sample solution, 1M in H\textsubscript{2}SO\textsubscript{4} and 0.02M in Ag\textsubscript{2}SO\textsubscript{4}. From our experience the most reproducible results are obtained with freshly precipitated argentic oxide\textsuperscript{2} and by destroying any in excess by warming to \(\sim 90^\circ\text{C}\) for 5 minutes. For the reduction of Pu(VI) with ferrous iron, the acidity is increased to 3M in H\textsubscript{2}SO\textsubscript{4} for the best results.

The precision and accuracy of the method for amounts of plutonium in the range 2-10 mg has been determined on suitable aliquots of a standard solution prepared from high-purity plutonium metal, and results are given in Table IV. In applied analysis this method has been used so far to check the results for plutonium obtained by other methods for uranium-plutonium carbides (see Table III) and plutonium carbide-chromium cermets (see also Table VI).

\textsuperscript{2} Mix 100 ml solution containing 3 g potassium persulphate with 1 g AgNO\textsubscript{3} in 10 ml H\textsubscript{2}O. After allowing to settle for 1 h, separate the precipitate with a centrifuge, wash with water and then slurry into a petri dish for drying in a desiccator.
TABLE IV

RESULTS FOR PLUTONIUM BY POTENTIOMETRIC TITRATION
BASED ON Pu(VI) TO Pu(IV) REDUCTION

<table>
<thead>
<tr>
<th>Experiment</th>
<th>Weight taken (mg)</th>
<th>Weight found by analysis (mg)</th>
<th>Recovery (%)</th>
<th>Coefficient of variation (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>2.4525</td>
<td>2.4533</td>
<td>100.03</td>
<td>0.09</td>
</tr>
<tr>
<td>2</td>
<td>4.7211</td>
<td>4.7179</td>
<td>99.93</td>
<td>0.06</td>
</tr>
<tr>
<td>3</td>
<td>9.4048</td>
<td>9.4163</td>
<td>100.12</td>
<td>0.11</td>
</tr>
</tbody>
</table>

2.2.2. Coulometric methods

Although constant-current coulometry was the first coulometric technique to be used for the determination of plutonium [14], controlled-potential coulometry has proved to be a simpler and more specific technique for this element when using the reversible Pu⁴⁺-Pu³⁺ couple. The determination of milligram amounts of plutonium in several mineral acid solutions (e.g. HCl, H₂SO₄, HClO₄ and HNO₃) has been studied to test the accuracy and reproducibility of the method with a coulometer built at A.E.R.E. [15]. The best results were obtained in sulphuric acid solutions, and under these conditions reduction of plutonium was found to be quicker with sulphur dioxide than by electrochemical means. The determination then involved a short pre-titration at +0.30 V versus a saturated calomel electrode (SCE) down to background current, followed by quantitative oxidation at +0.70 V versus SCE using the decrease of the cell current to follow the progress of the determination. Under normal circumstances a time of 10-20 min is needed for the current to decrease exponentially to a low value (~10 μA), and the precision for the determination of 4 mg amounts of plutonium is ±0.25% (c.v. based on 12 replicates).

Iron is the main interfering element in sulphuric acid solutions, the Eₒ for the Fe³⁺-Fe²⁺ couple being +0.42 V versus SCE compared with +0.50 V versus SCE for the Pu⁴⁺-Pu³⁺ couple. The behaviour of many other elements in this determination can be predicted from electrochemical data (Eₒ values), and U, Cr, Mo, Ti, Th and Be have been tested and found to cause no interference. In the analysis of samples containing iron, therefore, plutonium should be separated before determining its concentration coulometrically. The anion exchange separation for plutonium from 8M HNO₃ is not applicable in this case, since degradation products from the resin (Deacidite FF) resist destruction by wet oxidation and then interfere in the coulometry by poisoning the platinum working electrode. A satisfactory separation procedure involves the use of reverse phase chromatography with columns of KEL-F containing tri-n-butyl phosphate. A column is prepared from 1.5 g of KEL-F powder and 1.5 ml of tri-n-butyl phosphate in 25 ml of carbon
tetrachloride. After the evaporation of the carbon tetrachloride, the powder is slurried into a column with 5.5M HNO₃. The sample solution in 5.5M HNO₃ + 0.5M HCl with plutonium in the tetravalent state is put on the column, iron passes through, whereas plutonium and uranium are retained and then recovered in 20 ml 1M H₂SO₄ before coulometry.

In recent years controlled potential coulometry has been applied successfully to the determination of plutonium in ceramics and cermets. Although samples contained various other elements, none of them contained iron and so the direct procedure could be applied. The main problems in this work involved the dissolution of samples and the preparation of solutions suitable for coulometry. Further details for typical systems are given below:

**PuC-Mo.** Controlled-potential coulometry resulted in a direct procedure for the analysis of samples containing from 7 to 27% molybdenum. The dissolution procedure involved heating 100 mg of sample with 6M HCl and with the careful addition of 8M HNO₃ to avoid the formation of suspended organic matter. After dilution to 50 ml with 8M HNO₃, aliquots containing ~4 mg Pu were evaporated to fumes of H₂SO₄ after addition of 2 ml H₂SO₄ (sp.gr. 1.84) followed by the dropwise addition of HNO₃ (sp.gr. 1.42) to destroy soluble organic matter which would otherwise poison the working electrode. Results for plutonium on typical samples are given in Table V, and the very satisfactory totals for the complete sample analyses confirm the validity of these figures. Molybdenum contents were obtained by differential spectrophotometry on separate aliquots of sample solution, and carbon contents by a combustion procedure [16] on separate sample weights.

<table>
<thead>
<tr>
<th>Composition by analysis (%)</th>
<th>Sample No. 1</th>
<th>Sample No. 2</th>
<th>Sample No. 3</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pu</td>
<td>89.1</td>
<td>77.9</td>
<td>66.7</td>
</tr>
<tr>
<td>Mo</td>
<td>7.2</td>
<td>16.2</td>
<td>26.56</td>
</tr>
<tr>
<td>C</td>
<td>3.68</td>
<td>5.58</td>
<td>6.59</td>
</tr>
<tr>
<td>Total</td>
<td>99.98</td>
<td>99.68</td>
<td>99.85</td>
</tr>
</tbody>
</table>

**PuC-Cr.** Controlled-potential coulometry was selected for the determination of the plutonium content of this type of cermet because of the non-interference of chromium [17]. The development of a direct procedure for samples containing from ~40 to 80% Pu was dependent upon the dissolution of samples followed by the rapid destruction of organic matter. Unfortunately, samples were found to be resistant to attack by mineral acids, but sintering with sodium peroxide proved to be more effective. For samples in powder form,
the sample (100 mg) was mixed with 1 g sodium peroxide in a platinum crucible and then sintered for one hour at 500°C. Lumpy samples were oxidized first by ignition at 850°C before mixing with sodium peroxide. In all cases complete solution was achieved on extracting the sinter with water and then 8M HNO₃, the final solution being 2 to 3M in HNO₃ in a volume of 50 ml. An aliquot containing ~4 mg Pu was taken from each solution, and then evaporated to fumes of sulphuric acid after the addition of 1 ml H₂SO₄ (sp.gr. 1.84), 1 ml of 0.1M potassium pyrophosphate and 50 mg sodium sulphite. The residue was dissolved in 1M H₂SO₄, and after transferring the solution to a coulometer cell, the electrolysis was carried out at +0.285 V and +0.685 V versus SCE, respectively. Potassium pyrophosphate was added to prevent precipitation of chromium at the fuming stage.

Results and total recoveries for typical samples are given in Table VI. With sample No.1, the plutonium content was checked independently by potentiometric titration (argentie oxide method). Values of 76.0% and 76.1% were obtained and these confirmed the results by coulometry. Chromium contents were determined by potentiometric titration after separation of plutonium by anion exchange, whilst carbon contents were obtained by combustion analysis. The following coefficients of variation were obtained for the determination of the major constituents – 0.25% for Pu, 0.2% for Cr and 0.5% for C respectively.

### Table VI

**ANALYTICAL RESULTS FOR PuC-Cr SAMPLES**

<table>
<thead>
<tr>
<th>Composition by analysis (%)</th>
<th>Sample No. 1</th>
<th>2</th>
<th>3</th>
<th>4</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pu</td>
<td>76.1</td>
<td>66.7</td>
<td>42.6</td>
<td>75.9</td>
</tr>
<tr>
<td>Cr</td>
<td>17.4</td>
<td>28.7</td>
<td>39.7</td>
<td>16.1</td>
</tr>
<tr>
<td>C</td>
<td>2.9</td>
<td>2.1</td>
<td>17.4</td>
<td>7.9</td>
</tr>
<tr>
<td>O₂ + N₂</td>
<td>3.3</td>
<td>2.3</td>
<td>&lt;0.02</td>
<td>0.02</td>
</tr>
<tr>
<td>Total</td>
<td>99.7</td>
<td>100.3</td>
<td>99.7</td>
<td>99.9</td>
</tr>
</tbody>
</table>

(Pu, Th)C₂. Samples containing about 20% Pu proved to be readily dissolved by nitric acid, but difficulties occurred on attempting to destroy organic matter by wet oxidation. Evaporation of solutions to fumes of sulphuric acid resulted in the precipitation of thorium sulphate with consequent bumping during the wet oxidation procedure. The use of perchloric acid instead of sulphuric acid overcame the bumping difficulty, but it caused the oxidation of the plutonium to the hexavalent state. Sodium sulphite was employed to reduce the plutonium to the trivalent state so as to avoid the slow electrochemical reduction. The procedure was tested on synthetic solutions corresponding to a composition of 20% PuC₂ in ThC₂ prepared from plutonium
metal and thorium dicarbide, and resulted in a mean recovery of 100.1% for plutonium with a coefficient of variation of 0.24%. In the analysis of typical samples, satisfactory agreement was obtained for plutonium by controlled-potential coulometry and by differential spectrophotometry as shown in Table VII.

### TABLE VII
RESULTS FOR PLUTONIUM IN PLUTONIUM-THORIUM CARBIDES

<table>
<thead>
<tr>
<th>Sample No.</th>
<th>Mean result for Pu content (% by weight)</th>
<th>Coulometry</th>
<th>Differential spectrophotometry</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>18.4</td>
<td>18.5</td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>17.4</td>
<td>17.35</td>
<td></td>
</tr>
</tbody>
</table>

3. METHODS FOR MINOR AMOUNTS OF PLUTONIUM IN METALLURGICAL SAMPLES

3.1. Alpha counting

Plutonium consists of isotopes of mass numbers 238 to 241, together with insignificant amounts of higher isotopes, but the first three isotopes are the most important alpha emitters and their alpha energies and specific activities have been reported \[18\]. The $^{241}$Pu undergoes $\beta$ decay to produce the alpha emitter $^{241}$Am, and the contribution of this isotope to the total count is dependent upon the time that has elapsed since the chemical separation of the plutonium. The specific activities are such that sources containing from 0.01 to 0.10 $\mu$g plutonium are suitable provided that losses due to self absorption are minimized by keeping the amount of solid material on a 1 in. diameter source to less than about 50 $\mu$g. Satisfactory sources can be prepared by pipetting 0.05 or 0.10 ml of a sample solution onto a tray, adding a spreading agent, and then evaporating to dryness under an infra-red heater.

It is the usual practice to prepare and count a number of sources from the same solution and to calculate the result from the mean count. There are two possible ways of counting, either it is done absolutely with a known geometry and with a knowledge of the specific activity, or it is carried out comparatively with standard sources prepared from known amounts of plutonium with an isotopic composition the same as that for the plutonium in the samples. In the comparative method it is important to ensure that the equilibrium between $^{241}$Pu and $^{241}$Am has not been disturbed. The precision of a determination is influenced by the reproducibility of source preparation. Personal factors in pipetting aliquots and in evaporating them to form evenly-spread sources are important and can only be evaluated individually. With practice it is possible to prepare a single source with a reproducibility (c.v.) in the range 0.5 to 1.0%, and from this the number of sources necessary
to give a definite precision in the analysis of an unknown sample can be established.

This technique has been employed extensively at A.E.R.E. to check the plutonium contents of a series (400 in number) of plutonium-uranium alloys containing 0.25% Pu. The plutonium content of individual alloys was required to be known to within ±0.005% (3σ), and that of the whole series to within ±0.001 (3σ). In assessing the applicability of alpha counting, a typical sample and a standard were dissolved and two series of sources were prepared for counting. The reproducibility of these sources was 0.75% (c.v.) and on this basis an analysis based on one sample and one standard source would be correct to within ±0.0078% (3σ) at the 0.25% Pu level. Some improvement was needed therefore to meet the specification, and this was achieved by increasing the number of sources for standard and sample to six, giving limits of ±0.005% (3σ), due allowance being made for the degrees of freedom. The accuracy of results by alpha counting was checked by applying this method to alloys that were simultaneously analysed by differential spectrophotometry following the anion exchange separation of plutonium. The results for several samples are given in Table VIII, and the good agreement confirms the accuracy of the counting method.

TABLE VIII

RESULTS FOR THE ANALYSIS OF 0.25% Pu–U ALLOYS

<table>
<thead>
<tr>
<th>Sample No.</th>
<th>Alpha counting</th>
<th>Plutonium content (%)</th>
<th>Differential spectrophotometry</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.244</td>
<td>0.244</td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>0.241</td>
<td>0.244</td>
<td></td>
</tr>
<tr>
<td>3</td>
<td>0.246</td>
<td>0.244</td>
<td></td>
</tr>
<tr>
<td>4</td>
<td>0.243</td>
<td>0.244</td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>0.242</td>
<td>0.244</td>
<td></td>
</tr>
<tr>
<td>6</td>
<td>0.244</td>
<td>0.244</td>
<td></td>
</tr>
</tbody>
</table>

The average plutonium content for the whole series was determined by analysing a bulk sample prepared by dissolving a representative sample from each fuel rod and combining the solutions. Ten dilutions were made from the bulk solution and twelve sources were prepared from each dilution to attain the required precision. The mean reproducibility of the alpha count rates of the sources, as measured on each batch, was 0.75% (c.v.). From this a result at the 0.25% level of plutonium would be correct to within ±0.0026% (3σ) based on one batch of twelve sources and within ±0.0008% (3σ) based on all ten batches. This fulfilled the required specification. The plutonium content of the bulk solution was also determined independently by the isotopic dilution method and the results are given in Table IX.
TABLE IX
RESULTS FOR THE ANALYSIS OF BULK SOLUTION
FOR 0.25% Pu-U ALLOY

<table>
<thead>
<tr>
<th>Method of analysis</th>
<th>Plutonium (%)</th>
<th>Precision</th>
</tr>
</thead>
<tbody>
<tr>
<td>Alpha counting</td>
<td>0.2483</td>
<td>±0.0008 (3 σ)</td>
</tr>
<tr>
<td>Isotopic dilution</td>
<td>0.2490</td>
<td>±0.0027 (3 σ)</td>
</tr>
</tbody>
</table>

3.2. Isotopic dilution analysis

This technique is particularly suitable when the quantity of plutonium is restricted, since it can give precise results for microgram amounts. It consists of adding a known quantity of tracer (preferably $^{242}$Pu) to the sample, and after isotopic exchange by successive oxidation and reduction of the plutonium, some of the mixed element is separated by an anion exchange procedure for mass spectrometric examination. The total content of a specific nuclide of plutonium in the sample can then be calculated from the isotopic composition of the mixture. A mean coefficient of variation of 0.17% has been obtained at A.E.R.E. for plutonium concentrations in the range 0.1 to 1 μg/ml by taking 30–50 individual scans in each isotopic analysis with a solid source mass spectrometer [19].

The plutonium mass spectrum must be free from interferences due to other elements. For example, $^{241}$Am could interfere with the determination of $^{241}$Pu, but this is prevented by separating plutonium by anion exchange immediately before mass analysis. The small $^{238}$Pu content (~0.005%) is best determined by alpha pulse analysis because of difficulty in identifying its peak in the presence of the usual background level of $^{238}$U. Contamination is an important factor in chemical manipulations with such small amounts of plutonium, and the procedure must be designed to minimize this effect.

3.3. Polarography

AC polarography has been found to be suitable for the determination of plutonium in the concentration range from $4 \times 10^{-6}$ M to $4 \times 10^{-4}$ M on using a peak at ~0.42 V versus a mercury pool anode from a citrate supporting electrolyte at pH 4.5 [20]. This peak is produced by plutonium in either the trivalent or the tetravalent state because of the reversibility of the Pu$^{4+}$/Pu$^{3+}$ couple. Tetravalent plutonium is stable in the citrate supporting electrolyte, whereas the trivalent species undergoes oxidation. The peak for the tetravalent complex is therefore preferred for analytical work, but unfortunately interference can be caused by a few elements, including iron. An anion exchange procedure can be employed for separating interfering elements from plutonium, and this method has been applied to the determination of the plutonium content of uranium-plutonium alloys containing from 0.25 to 1.0% Pu to within ±1.5%. Although polarography is not as sensitive or as convenient as alpha counting, it does have the merit of not requiring a knowledge of the isotopic composition of the plutonium.
REFERENCES


DISCUSSION

J.T. BYRNE: In section 2.2.2. of your paper you state that anion-exchange separation for Pu is not applicable prior to coulometry because degradation products of the Deacidite FF resist destruction by wet oxidation and poison the platinum working electrode. Mr. Regnau (SM-67/10) and Dr. Metz (SM-67/33) both recommend anion-exchange separation (with Dowex 1 X 4) for this purpose. Do you suppose your difficulty is due to the use of Deacidite FF rather than Dowex 1 X 4?

G.W.C. MILNER: Our experience has been limited to the use of the resin Deacidite FF, because of its ready availability in the United Kingdom. However, Dr. Shults of the Oak Ridge National Laboratory has reported serious interference by degradation products resulting from Dowex 1 X 4 in the coulometric determination of plutonium*. After recovering plutonium from Dowex 1 X 4 with dilute HNO₃-HF solutions, Shults claimed to be able to eliminate the effects of organic matter by evaporating to fumes of sulfuric acid before titrating coulometrically. We have had limited success applying the Shults method after separations with Deacidite FF. Satisfactory results for plutonium, however, were only obtained on using columns prepared from new batches of resin.

* Talanta 10 (1963) 646.
C. F. METZ: We have had occasional trouble with organic material in concentrated hydrochloric acid of reagent grade. Your experience with organic material coming from an ion-exchange resin is interesting and, I think, important. We have not done much with controlled potential coulometry at Los Alamos, but I am glad to hear about your experience.

Do you consider that coulometric techniques are more sensitive to the presence of organic material than the potentiometric technique?

G. W. C. MILNER: Controlled potential coulometry is definitely more sensitive than potentiometric titrations to the presence of organic material in solution. This is because a much greater surface of platinum is in contact with the plutonium sample solution in coulometry.

Organic matter can cause interference in most electroanalytical techniques, particularly polarography.

F. REGNAUD: Perhaps I can add that plutonium coulometry after separation on ion-exchange resins has given us no trouble as far as interference from organic materials is concerned. The elution solution is heated with sulphuric acid to white fumes, and the blanks we have measured give an electrolysis current less than 10 μA.

I agree with you, however, that ion-exchange resins should not be used when the analytical method which follows is polarography.
METHODS USED FOR ISOTOPIC ANALYSIS OF URANIUM AND PLUTONIUM. Isotopic analysis is commonly done by mass spectrometry. Use is made of two methods differing technologically according to the kind of ion source used in the measuring equipment. The paper describes the operational techniques and the manner in which the results are calculated in the two methods. The precautions necessary for accurate analysis of uranium-234 and uranium-236 are given.

Surface-ionization sources can be used for working with small quantities of material: samples of the order of a milligram. Uranium analysis requires no complex chemical preparation and the measurements are absolute. Accuracy is limited: 1% accuracy is easily obtained for uranium-235 but not so easily for uranium-234 and uranium-236. Gaseous sources are supplied with uranium hexafluoride. An accuracy of ± 5 × 10^-7 can be reached in relative measurements. With this technique a higher analysis rate can be obtained than with the surface-ionization method, but quantities of the order of 1 gram are required and chemical treatment is necessary to convert the product into UF₄. A given apparatus can operate only in a particular range of concentrations.

Isotopic analysis of plutonium is generally done only by the surface-ionization method because it is desirable to handle only very small quantities of radioactive products (hundreds of a microgram). Solutions of plutonium containing uranium require chemical separation. The accuracy of analysis is of the order of 1%. Measurement of small quantities of heavy isotopes is generally done when the abundance exceeds 0.01%.
подготовки и представляют собой абсолютные измерения. Точность ограничена. Она достигает 1%, легко полученного для урана-235 и более трудно для урана-234 и урана-236. Газовые источники снабжаются шестифтористым уралом. В сравнительном измерении точность достигает ±5·10⁻⁴. Этот метод открывает более широкие возможности для аналитов по сравнению с предыдущим. Однако для работы необходимо иметь количество порядка грамма, и к тому же требуется химическая обработка для превращения продукта в шестифтористый уран — каждый прибор может работать только в пределах специально подготовленной концентрации.

Изотопический анализ плутония проводится в общем лишь по методу поверхностной ионизации с тем, чтобы манипулировать только очень небольшими количествами радиоактивных продуктов (сотые доли микрограмма). Содержащие уран растворы плутония требуют химического разделения. Точность анализов составляет порядка 1%. Измерение небольших количеств тяжелых изотопов производится в общем тогда, когда распространенность превышает 0,01%.

**METODOS EMPLEADOS PARA EL ANALISIS ISOTOPICO DEL URANIO Y DEL PLUTONIO.** Los análisis isotópicos se efectúan corrientemente por espectrometría de masas. Se emplean dos métodos que se diferencian técnicamente por la distinta naturaleza de las fuentes iónicas con que están equipados los aparatos de medición.

En la memoria se describen las técnicas operativas y la manera de calcular los resultados según ambos métodos. También se indican las precauciones que hay que tomar para poder analizar con precisión el uranio-234, y el uranio-236.

Las fuentes de ionización de superficie permiten trabajar con cantidades muy pequeñas de producto; muestras del orden del miligramo. Los análisis del uranio no exigen una preparación química compleja; se trata de mediciones absolutas. La precisión es limitada: llega al 1% con facilidad para el uranio-235, y más difícilmente para el uranio-234, y el uranio-236. Las fuentes gaseosas se alimentan con hexafluoruro de uranio. La exactitud llega a ±5·10⁻⁹ en medición comparativa. Esta técnica permite efectuar los análisis a un ritmo más rápido que la anterior. De todas maneras hay que disponer de cantidades del orden del gramo y es preciso un tratamiento químico para convertir el producto en UF₆. Cada aparato sólo puede trabajar dentro de unos límites de concentración determinados.

En general, el análisis isotópico del plutonio se efectúa exclusivamente por ionización de superficie, ya que así sólo hay que manipular cantidades muy pequeñas de producto radiactivo (centésimas de microgramo). Las soluciones de plutonio que contienen uranio exigen una separación química. La precisión de los análisis es del orden del 1%. La medición de cantidades pequeñas de isótopos pesados se efectúa en general cuando la abundancia excede del 0,01%.

Les qualités nucléaires des matières radioactives sont liées aux propriétés de chacun des isotopes des éléments constitutifs. Si l'analyse chimique est nécessaire pour s'assurer des propriétés des matières utilisées, la connaissance de leur composition isotopique est tout aussi essentielle.

Si l'on excepte les combustibles nucléaires élaborés à partir d'uranium naturel dont la composition isotopique est très sensiblement la même quelle que soit l'origine du minerai, la connaissance de la composition isotopique est liée à toutes les étapes de la gestion des matières de base, en particulier:

- aux différentes étapes de la fabrication des combustibles nucléaires enrichis en uranium-235: traitement chimique puis métallurgique;
- à l'évaluation des matières nucléaires contenues dans les résidus et dans les uraniums appauvris après irradiation;
- à la détermination des matières fissiles consommées (uranium-235) et produites (plutonium) au cours de l'irradiation d'un combustible.

L'analyse isotopique permet également de déterminer par dilution isotopique

---

1 L'uranium naturel contient 0,726 atomes d'uranium-235 pour 100 atomes de métal.
- la teneur en plutonium d'un uranium irradié,
- la teneur en uranium-233 d'un thorium irradié.

La complexité des calculs concernant la formation du plutonium augmente au cours de l'irradiation, au fur et à mesure que s'accumulent les produits de fission. Le dosage du plutonium formé permet de vérifier expérimentalement les calculs théoriques pour un taux de combustion (burn-up) donné.

La méthode la plus couramment utilisée et la plus précise pour la mesure de la concentration des isotopes est la spectrométrie de masse. La quantité totale d'échantillon dont il faut disposer pour effectuer l'analyse est très faible. Une analyse isotopique s'effectue couramment avec un dépôt de quelque $10^{-7}$ g d'uranium ou de quelque $10^{-8}$ g de plutonium. L'avantage, en particulier pour l'étude des combustibles irradiés, est indéniable.

Le principe d'une analyse isotopique par spectrométrie de masse est connu. Le composé préparé à partir du métal étudié, oxyde ou fluorure, est vaporisé puis ionisé. Les ions sont accélérés par un champ électrique, puis déviés par un champ magnétique qui sépare dans l'espace les faisceaux d'ions de même charge suivant leur masse, ce qui permet de les collecter séparément et d'en mesurer l'intensité. Les courants d'ions de chaque masse sont proportionnels à la concentration de l'isotope correspondant dans l'échantillon.

Nous décrivons successivement l'analyse isotopique de l'uranium et du plutonium, puis l'application de l'analyse isotopique au dosage du plutonium dans l'uranium.

1. L'ANALYSE ISOTOPIQUE DE L'URANIUM

Deux méthodes sont utilisées, qui se différencient par la nature différente des sources d'ions qui équipent les appareils de mesure: source à ionisation de surface et source à ionisation de gaz.

1.1. Méthode par ionisation de surface

Les analyses isotopiques sont effectuées à l'aide d'un spectromètre de masse. Le secteur magnétique est de 90° et les trajectoires des ions ont 30 cm de rayon de courbure.$^2$

Cet appareil est équipé d'une source à thermo-ionisation se composant de deux parties distinctes:
- la source d'ions proprement dite;
- le pied de source où sont disposés trois filaments: deux sont utilisés indifféremment pour déposer l'échantillon et placés de part et d'autre du filament central où s'effectue l'ionisation. Ce pied de source aisément amovible est changé à chaque analyse. Le filament central ionisant en rhénium est chauffé à 2500°C environ. La température des filaments latéraux où s'effectue l'évaporation varie selon la nature du produit à analyser, pour l'uranium elle se situe au voisinage de 1400°C.

$^2$ MS 5 - Associated Electrical Industries, Grande-Bretagne.
Les dépôts d'uranium (2 · 10^{-7} g) sont effectués sur filaments de tantale. L'échantillon, reçu sous forme de métal ou d'oxyde, est mis en solution nitrique d'une concentration en uranium de 1 mg/ml. Le dépôt s'effectue à partir de cette solution diluée. Un appareil constitué par une seringue orientable a été spécialement étudié pour obtenir des dépôts de très faibles quantités d'échantillon qui soient reproductibles.

Les courants d'ions sont détectés au moyen d'un multiplicateur d'électrons suivi d'un amplificateur à courant continu et d'un enregistreur à plume. Le multiplicateur comporte dix étages; il travaille couramment avec un gain de 5000 pour une tension appliquée de 1800 V. Dans de telles conditions, on peut mesurer des courants d'ions dont l'intensité est comprise entre 10^{-12} A (pic principal) et 10^{-16} A. La tension d'accélération des ions reste constante. Le défilement des masses est obtenu par variation cyclique du champ magnétique.

Les mesures sont faites sur une série de dix enregistrements successifs du spectre considéré; elles sont reportées sur des cartes perforées. Les moyennes des hauteurs de pics relatifs à chaque isotope sont utilisées pour le calcul des concentrations isotopiques. Une calculatrice électronique (IBM 7090) effectue le calcul de l'erreur statistique affectée à la concentration de chaque isotope. Il lui est demandé de plus de sortir les valeurs individuelles présentant des écarts supérieurs à une valeur donnée - cette valeur est fonction de la précision relative recherchée; ceci permet d'attirer l'attention sur une valeur individuelle aberrante et d'en déceler rapidement l'origine: erreur de lecture des hauteurs de pics ou instabilité de l'appareil.

En analyse courante, la précision relative est comprise entre 7\% et 1\% pour l'isotope 235. Elle est de 2\% pour les isotopes 234 et 236 quand leur concentration est de l'ordre de 0,05\%.

Une technique particulière est utilisée pour la mesure précise des isotopes rares 234 et 236. Elle consiste à comparer dans une première analyse l'isotope 235 à l'isotope 238 puis, dans une seconde analyse effectuée à partir du même dépôt d'uranium, à élever la température du filament latéral de façon à obtenir un courant d'ions 235 maximal. Les pics 234 et 236 sont alors comparés au pic 235.

La sensibilité est ainsi considérablement accrue et la précision reste acceptable. La mesure de l'abondance de l'isotope 234 dans l'uranium naturel a été trouvée dans ces conditions égale à 0,0054\% ± 0,0001.

1.2. Analyse par ionisation de gaz

Cette méthode est surtout intéressante pour mesurer de faibles variations de concentration d'uranium-235 [1]. On utilise dans ce cas la spectrométrie de masse de l'hexasfluorure d'uranium (UF₆) qui permet une haute précision dans la comparaison des teneurs isotopiques de deux échantillons (environ 0,05\%).

La principale difficulté de cette méthode provient de la quantité de matière relativement élevée nécessaire, en comparaison de ce qui est suffi-

\[3\] De cette quantité seule une fraction de l'ordre de quelques milligrammes est consommée, mais la totalité est nécessaire pour des raisons technologiques.
sant pour des analyses par ionisation de surface (grammes au lieu de micro-
grammes). Dans le cas d'un combustible irradié, une décontamination
poussée de la solution d'échantillon est nécessaire. En outre, dans le cas
où une irradiation élevée a provoqué un fort appauvrissement en uranium-235,
un étalonnage est nécessaire pour conserver la précision.

1.2.1. Traitement chimique

L'uranium (environ 1 g) est converti en hexafluorure. S'il provient
d'un combustible irradié, il est séparé du plutonium et des produits de
fission par une extraction au solvant organique puis converti en oxyde $\text{U}_3\text{O}_8$
par calcination à l'air. L'oxyde est ensuite fluoré par réaction avec le
trifluorure de cobalt $\text{CoF}_3$ [2].

$$\text{U}_3\text{O}_8 + 18 \text{CoF}_3 \rightarrow 3 \text{UF}_6 + 18 \text{CoF}_2 + 4\text{O}_2$$

La réaction dure 2 h à 500°C. Elle est effectuée dans une installation
métallique résistant à la corrosion par le fluor.

La pureté chimique de l'UF$_6$ obtenu est d'une grande importance. Si
elle est insuffisante, le résultat de l'analyse est affecté d'une erreur im-
portante. La purification est effectuée par distillation sous vide.

1.2.2. Analyse isotopique

L'appareil utilisé pour les analyses isotopiques d'UF$_6^4$ est équipé d'une
source où les ions sont produits à l'aide d'un faisceau d'électrons. On
utilise pour l'analyse les ions UF$_5^-$, qui sont les plus abondants. Ils sont
accélérés sous 2500 V puis déviés par un secteur magnétique à 90° avec
des trajectoires de 20 cm de rayon de courbure.

Le spectromètre de masse à UF$_6$ utilise la technique de double collec-
tion pour mesurer les rapports isotopiques. Un ensemble de deux collec-
teurs sélectionne, d'une part le courant d'ions de l'isotope 235, et d'autre
part la somme des courants d'ions provenant des autres isotopes. L'in-
formation lue sur l'appareil est

$$L = KR \quad \text{avec} \quad R = \frac{n_5}{1 - n_5}$$

où $n_5$ est la concentration atomique de l'uranium-235. $K$ est un coefficient
qui dépend de l'appareil et qu'on élimine en comparant l'échantillon à me-
surer à un échantillon de référence.

$$\alpha = \frac{L_1}{L_2} = \frac{R_1}{R_2}$$

Si la valeur de $R_2$ est connue, $R_1$ peut en être déduit.

---

4 SMU 500 spécialement étudié pour le CEA par la Société Française CSF.
Si l'échantillon 1 provient d'un combustible irradié et l'échantillon 2 du même combustible avant irradiation, $\alpha$ est la valeur de l'appauvrissement.

En réalité, une complication est introduite par ce qu'on appelle «l'effet de mémoire» [3,4], qui doit être corrigé.

L'hexafluorure d'uranium possède la propriété de se déposer sur les surfaces internes du spectromètre de masse. La composition isotopique de l'uranium déposé dépend des compositions isotopiques des échantillons introduits précédemment dans l'appareil. Quand l'échantillon d'UF₆ est introduit, un échange isotopique se produit et, en général, la composition isotopique de l'échantillon est altérée.

Par exemple, si deux produits différents sont introduits alternativement, la composition isotopique du dépôt tend vers une valeur comprise entre les valeurs correspondantes des deux produits - chacun de ceux-ci va échanger avec le dépôt et sera altéré en sens inverse. Finalement $\alpha_M$ (valeur mesurée de $\alpha$) sera telle que:

$$|\alpha_M - 1| < |\alpha_E - 1|$$

où $\alpha_E$ est la valeur exacte de $\alpha$.

L'erreur due à l'effet de mémoire est couramment égale à plusieurs fois la dispersion normale des mesures. Pour la corriger on utilise un facteur M calculé à partir de

$$\alpha_E - 1 = M(\alpha_M - 1)$$

M est toujours plus grand que 1. Les valeurs usuelles sont comprises entre 1,03 et 1,10.

On observe que si la concentration en uranium-235 du produit n'est pas trop élevée, la valeur de M est indépendante de $\alpha_E$; ainsi si la valeur de M a été déterminée pour un couple d'échantillons de valeurs connues, la même valeur est valable pour un autre couple. Le principe de la correction de mémoire consiste à mesurer M en analysant un couple dont la valeur $\alpha_E$ est connue.

Les standards $\alpha$ sont préparés à partir des standards isotopiques d'uranium enrichi provenant du National Bureau of Standards. En prenant l'uranium naturel comme produit de référence, la préparation est suffisamment précise dans l'intervalle $0.9 < \alpha < 3$. Dans ce domaine de concentration, M peut être mesuré avec une bonne précision et les mesures d'appauvrissement peuvent être effectuées dans le même domaine de concentration. La correction de l'effet de mémoire tend vers 0 si $\alpha$ tend vers 1. Plus $\alpha - 1$ est petit, meilleure est la précision. Si une série d'analyses est faite sur un ensemble d'échantillons irradiés dans des conditions voisines, le meilleur moyen pour obtenir une bonne précision est de comparer les échantillons et de mesurer avec un soin particulier l'appauvrissement de l'un d'entre eux.

2. ANALYSE ISOTOPIQUE DU PLUTONIUM

Les analyses isotopiques du plutonium sont effectuées à l'aide d'un spectromètre de masse équipé d'une source à ionisation de surface, selon
une technique analogue à celle qui a été décrite pour l'uranium. Les solutions de plutonium sont manipulées en boîte à gants étanche, maintenue en légère dépression. Une analyse se fait en déposant quelques centièmes de microgramme de plutonium en solution nitrique diluée sur un filament de rhénium.

Dans le cas fréquent où le plutonium se trouve en faible quantité dans une solution d'uranium, il est nécessaire d'effectuer une séparation chimique qui élimine la plus grande partie de l'uranium. La méthode mise au point à cette fin [5, 6] utilise les résines échangeuses d'ions; sur résine anion, les complexes nitriques de plutonium (IV) sont fixés sélectivement et peuvent ainsi être séparés de l'uranium et des produits de fission. Une réduction ultérieure du plutonium à la valence III par le chlorhydrate d'hydroxylamine permet d'éluer le plutonium.

Une séparation U/Pu totale n'est pas indispensable, mais la quantité d'uranium résiduel doit être suffisamment faible (rapport U/Pu de l'ordre de 25) pour qu'on obtienne une émission thermo-ionique du plutonium stable.

L'analyse est conduite comme pour l'uranium. On enregistre les pics de masse des isotopes 239, 240, 241 et si nécessaire 242. La précision est comprise entre 7% et 1% pour les isotopes les plus abondants. En analyse courante les isotopes dont la concentration est inférieure à 0,01% ne sont pas enregistrés.

3. APPLICATION AU DOSAGE DU PLUTONIUM DANS L'URANIUM

Les études concernant les taux de combustion des combustibles (burn-up), et plus simplement les calculs concernant la production de plutonium peuvent être vérifiés par le dosage du plutonium dans le combustible irradié. La détermination précise du plutonium peut être effectuée par dilution isotopique, méthode qui fait appel à l'analyse isotopique de l'uranium et du plutonium.

3.1. Principe de la dilution isotopique à double traceur

Un combustible nucléaire après irradiation est encore constitué pour l'essentiel d'uranium. Les deux constituants principaux sont l'uranium et le plutonium qui pourraient être dosés séparément par dilution isotopique simple. On préfère utiliser une solution contenant les deux traceurs, uranium et plutonium.

3.1.1. Dilution isotopique simple

Pour doser dans un échantillon un élément possédant plusieurs isotopes on utilise un «traceur» c'est-à-dire une solution où l'élément à doser est présent en quantité connue, dans le même état chimique que dans l'échantillon; la composition isotopique de l'élément contenu dans le traceur est choisie très différente de celle de l'élément présent dans l'échantillon.
La méthode consiste à mélanger, en proportions connues, l'échantillon et le traceur. Après homogénéisation de la répartition des isotopes, la composition isotopique du mélange dépendra :

a) des quantités d'élément respectivement présentes dans l'échantillon et le traceur,

b) des proportions dans lesquelles le mélange a été effectué,

c) des compositions isotopiques de l'élément dans le traceur et l'échantillon.

La composition isotopique du mélange sera intermédiaire entre celle de l'échantillon et celle du traceur. Pour doser par exemple l'uranium dans une solution échantillon, on utilise une solution traceur contenant de l'uranium-233 en quantité connue.

On mesure la composition isotopique de l'uranium de l'échantillon et celle du traceur. On mélange des volumes connus de traceur et d'échantillon et on mesure la composition isotopique du mélange.

L'uranium-233 du traceur a été « dilué » par l'uranium de l'échantillon dans une proportion qui dépend des volumes mélangés et des compositions isotopiques des solutions avant mélange. Connaissant toutes ces grandeurs avant et après mélange on peut calculer la quantité d'uranium contenue dans l'échantillon.

3.1.2. Dilution isotopique avec double traceur

Si l'on désire seulement déterminer le rapport des masses de deux éléments en solution (par exemple U/Pu) on peut utiliser un traceur double contenant les deux éléments correspondants. La dilution isotopique a lieu simultanément pour l'uranium et le plutonium au moment où l'on mélange la masse unique du traceur double à l'échantillon. Les compositions isotopiques des deux éléments sont modifiées par le mélange, mais de façon telle que la relation existant entre le rapport des masses des éléments dans l'échantillon et la composition isotopique des éléments dans le mélange est indépendante des masses mélangées, ce qui permet d'éviter la mesure précise des quantités d'indicateur et d'échantillon que l'on utilise.

Le traceur double employé pour déterminer le rapport U/Pu contient de l'uranium-233, isotope n'existant pas dans l'uranium naturel, qui est obtenu par irradiation neutronique du thorium-232 et du plutonium-242 préparé par irradiation prolongée (plusieurs années) de plutonium-239 dans un réacteur à haut flux. Chaque essai nécessite 0,2 . 10^{-6} g de plutonium-242; 1 mg de plutonium-242 est une quantité théoriquement suffisante pour 5000 déterminations. Dans ces conditions, on calcule le rapport U/Pu de l'échantillon au moyen de la formule suivante, dans laquelle les rapports isotopiques sont exprimés en atomes:

\[
\left( \frac{^{238}\text{U}}{^{239}\text{Pu}} \right)_E = \left( \frac{^{233}\text{U}}{^{242}\text{Pu}} \right)_T \frac{^{238}\text{U}}{^{233}\text{U}} M - \frac{^{238}\text{U}}{^{233}\text{U}} T \nonumber
\]
Les indices E, T, M désignent respectivement l'échantillon, le traceur et le mélange.

Étalonnage du traceur double

Le rapport \((^{233}\text{U}/^{242}\text{Pu})_T\) doit être connu avec précision; il ne peut être mesuré directement par pesée des éléments car on dispose en général de trop faibles quantités d'uranium-233 et de plutonium-242 et leur pureté chimique n'est pas connue.

La détermination précise de ce rapport est obtenue par dilution isotopique à l'aide d'une solution synthétique dite «étalon», préparée par pesée d'uranium naturel et de plutonium de pureté chimique connue (tableau I).

**TABLEAU I**

**ÉTALONNAGE DU TRACEUR**

<table>
<thead>
<tr>
<th></th>
<th>(\left(\frac{^{238}\text{U}}{^{235}\text{U}}\right)_M)</th>
<th>(\left(\frac{^{239}\text{Pu}}{^{244}\text{Pu}}\right)_M)</th>
<th>(R = \left(\frac{^{233}\text{U}}{^{242}\text{Pu}}\right)_T)</th>
</tr>
</thead>
<tbody>
<tr>
<td>a</td>
<td>3,54 \pm 0,02</td>
<td>9,61 \pm 0,09</td>
<td>5080</td>
</tr>
<tr>
<td>b</td>
<td>3,55 \pm 0,02</td>
<td>[ \begin{array}{c} 9,61 \pm 0,09 \ 9,61 \pm 0,09 \end{array} ]</td>
<td>5073</td>
</tr>
<tr>
<td>c</td>
<td>3,52 \pm 0,02</td>
<td>0,54 \pm 0,09</td>
<td>5074</td>
</tr>
<tr>
<td>d</td>
<td>3,38 \pm 0,02</td>
<td>[ \begin{array}{c} 9,22 \pm 0,09 \ 9,08 \pm 0,09 \ 9,11 \pm 0,09 \end{array} ]</td>
<td>5112</td>
</tr>
</tbody>
</table>

Valeur moyenne de \(R\) : \(\bar{x} = 5072\)

Note: Quatre mélanges étalon-traceur a, b, c, d ont été effectués. Certaines analyses isotopiques de plutonium (3e colonne) ont été reproduites plusieurs fois sur un même mélange; les résultats sont groupés sous une même accolade.

La composition de l'étalon est la suivante: uranium métal: 186,8 g; plutonium métal: 0,102 g, dissous dans 1000 ml d'acide nitrique 8N.

3.2. Préparation des échantillons et analyse isotopique

Les rapports U/Pu rencontrés dans les combustibles irradiés varient entre 200 et 10 000.
La mise en solution par l’acide nitrique concentré a lieu en boîte à gants ou en boîte à pinces avec protection de plomb si l’échantillon contient des produits de fission. A partir de telles solutions, on détermine le rapport U/Pu par dilution isotopique ainsi que la composition isotopique du plutonium présent et éventuellement celle de l’uranium. On mélange des volumes déterminés de solution d’attaque et de traceur. Ces volumes n’ont pas besoin d’être connus avec précision ce qui est l’avantage essentiel du double traceur. On mesure la composition isotopique de l’uranium et du plutonium après mélange. L’uranium peut être déposé directement sur filament et l’analyse est conduite comme au paragraphe 1.1. (analyse de l’uranium par ionisation de surface).

Par contre, pour le plutonium il est nécessaire d’effectuer une séparation chimique puisqu’on se trouve en présence d’uranium (cf. par. 2.).Le traitement chimique utilisé [7] permet, outre une séparation U/Pu convenable, d’obtenir une répartition homogène des isotopes dans les mélanges traceur-échantillon [8]. Les quantités de plutonium utilisées sont de l’ordre de 2·10^{-6} g par essai. La quantité déposée sur filament varie entre 2 et 5·10^{-8} g. Des comptages sont effectués sur les filaments pour contrôler la quantité de plutonium qui s’y trouve déposée.

On trouvera au tableau I les valeurs des rapports isotopiques mesurés au cours de l’étalonnage du traceur; elles permettent d’évaluer la reproductibilité de la méthode. Le calcul statistique montre que, pour une probabilité de 95%, la valeur moyenne vraie est comprise entre les limites $\bar{x}\pm 25$, soit une précision relative de 0,5%.

Pour les analyses courantes effectuées sur des combustibles irradiés, on se contente d’effectuer deux mélanges échantillon-traceur à partir d’une même solution d’attaque, ce qui conduit à une précision de l’ordre de 1%.

4. PRECISION ET REPRODUCTIBILITE DES ANALYSES ISOTOPIQUES

Nous avons rassemblé dans le tableau II les valeurs des précisions et des reproductibilités pour les différentes mesures isotopiques dont il a été question dans ce rapport. On remarquera que précision et reproductibilité ont des valeurs numériques souvent identiques. C’est la reproductibilité qui, en fait, détermine la précision. Cela est surtout vrai pour un ensemble analytique comme celui qui est mis en œuvre pour le dosage du plutonium. La reproductibilité du traitement chimique domine l’ensemble de l’analyse, plus que ne le font les fluctuations des mesures au spectromètre de masse. Une étude très serrée a été nécessaire [7] pour assurer cette reproductibilité du traitement chimique.

Dans le domaine des mesures d’appauvrissement, le phénomène dominant est le phénomène de «mémoire» (cf. par. 1.2.) dû à la modification de la composition isotopique du gaz analysé au contact des parois. Dans ce cas, la reproductibilité peut être meilleure que la précision, car la mesure peut être entachée d’une erreur systématique.

Les analyses isotopiques courantes sur spectromètre à ionisation de surface ont une précision de l’ordre de 7%. La reproductibilité des analyses est assez remarquable. Les analyses d’un standard enrichi à 93%
## TABLEAU II

PRECISION ET REPRODUCTIBILITE

<table>
<thead>
<tr>
<th>Mesures</th>
<th>Précision</th>
<th>Reproductibilité</th>
<th>Domaine de concentration</th>
</tr>
</thead>
<tbody>
<tr>
<td>Composition isotopique du plutonium</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$^{240}\text{Pu}/^{239}\text{Pu}$</td>
<td>0,7%</td>
<td>0,7%</td>
<td>$241\text{Pu}/^{239}\text{Pu} &gt; 1%$</td>
</tr>
<tr>
<td>$^{241}\text{Pu}/^{239}\text{Pu}$</td>
<td>1,0%</td>
<td>1,0%</td>
<td>$241\text{Pu}/^{239}\text{Pu} &lt; 1%$</td>
</tr>
<tr>
<td></td>
<td>2%</td>
<td>2%</td>
<td>Limite de sensibilité courante 0,01%</td>
</tr>
</tbody>
</table>

| Dosage du plutonium | 1% | 1% | $300 < \text{U/Pu} < 15,000$ |

| Composition isotopique de l'uranium | | |
| | 0,7% | 0,7% | $235\text{U}/^{238}\text{U} \approx 0,7\%$ (concentration naturelle) |
| | 1,0% | 1,0% | $0,2% < 235\text{U}/^{238}\text{U} < 0,7\%$ |

| Appauvrissement de l'uranium | | |
| | 0,05% | 0,03% | $1 > \alpha > 0,9$ |
| | 0,1% | 0,07% | $0,9 > \alpha > 0,6$ |

effectué à cinq ans d'intervalle sur un même appareil se sont trouvées identiques à la précision des mesures.

5. CONCLUSION

Les techniques décrites dans ce rapport permettent d'effectuer les contrôles qui concernent de façon générale la concentration des isotopes contenus dans les matières nucléaires. La nature du contrôle, la précision demandée, le délai exigé entre la demande et la réponse orienteront le choix vers les différentes techniques d'analyse isotopique par ionisation de surface, ionisation de gaz ou dilution isotopique.

Ces techniques se caractérisent cependant par l'importance des moyens nécessaires, tant en personnel qu'en matériel, qui se justifie par la précision des résultats qu'elles permettent d'obtenir.
REFERENCES

ISOTOPIC MEASUREMENT OF URANIUM BY MASS SPECTROMETRY

G.F. KAUFFMAN AND C.D. TABOR
GOODYEAR ATOMIC CORPORATION, PIKETON, OHIO,
UNITED STATES OF AMERICA

Abstract — Résumé — Аннотация — Resumen

ISOTOPIC MEASUREMENT OF URANIUM BY MASS SPECTROMETRY. The growing application of atomic energy creates a wider need for precise and accurate knowledge of the isotopic composition of uranium. This information is particularly of great importance in the accountability and transfer of enriched uranium for reactor and research applications involving millions of dollars worth of fissionable materials. Reliable isotopic measurements are also necessary to ensure compliance of fuel element compositions with the reactor design specifications and to permit calculation of process and fuel burn-up losses.

Mass spectrometry methods, which far surpass the capabilities of other methods, were developed for very precise isotopic determinations. These methods, "Single Standard" and "Double Standard", involve the comparison of measurements of an unknown sample to similar measurements on known standards. Use of the "Double Standard" method eliminates the effects of instrument bias, thus permitting isotopic determinations with precisions (95% limit of error) of the order of ± 0.02% of the values. Accuracies are limited only by the knowledge of the standard values used, which are referenced to the series of uranium isotopic standards available from the US National Bureau of Standards.

The mass spectrometers are also useful for the absolute determination of isotopic composition of uranium, especially in forms other than UF$_6$. Thermal ionization techniques using high-resolution (approximately 12-in. radius) spectrometers permit the absolute isotopic characterization of the minor isotopes (i.e. those less than 10 wt.%) with an accuracy and precision of about 0.5% of the values per analysis. These analyses are particularly useful in calibrating highly enriched and highly depleted uranium for subsequent use as blending materials in an isotopic standards programme.

Both relative and absolute isotopic measurement methods are described as well as their application in the accountability and operational analytical programmes. These applications include: (1) the analysis of umpire samples to resolve discrepancies between other laboratories, (2) the analysis of product to assure specifications of shipments, and (3) the analysis of uranium in various forms to establish accountability for receipts.

DOSAGE ISOTOPIQUE DE L'URANIUM PAR SPECTROMETRIE DE MASSE. Avec le développement des applications de l'énergie atomique, on a de plus en plus besoin de connaître avec précision et exactitude la composition isotopique de l'uranium. Ces données sont particulièrement importantes pour la comptabilité et pour le transfert de l'uranium enrichi destiné aux réacteurs et à la recherche, car il s'agit là de matières fissiles dont la valeur se chiffre par millions de dollars. Des mesures isotopiques fiables sont également nécessaires pour s'assurer qu'un élément combustible a bien la composition correspondant aux spécifications du réacteur considéré et pour permettre le calcul des pertes résultant du traitement et de l'irradiation.

On a mis au point, pour ces dosages isotopiques très précis, des méthodes par spectrométrie de masse dont les possibilités sont de loin supérieures à celles des autres méthodes. Ces méthodes, dites à « étalement unique » et à « double étalement », consistent à comparer les mesures faites sur un échantillon de composition inconnue avec des mesures identiques faites sur des étalements de composition connue. La méthode du « double étalement » élimine l'erreur systématique due aux instruments de mesure, ce qui permet des dosages isotopiques avec des précisions (limite d'erreur de 95%) de l'ordre de ± 0,02%.

L'exactitude n'est fonction que de celle des valeurs témoins utilisées, lesquelles dérivent d'une série d'étalements isotopiques d'uranium obtenus du National Bureau of Standards des Etats-Unis.

Les spectromètres de masse sont également utiles pour le dosage absolu des isotopes de l'uranium, notamment sous les formes autres que UF$_6$. Les méthodes d'ionisation thermique utilisant des spectromètres à haute résolution (rayon d'environ 30 cm) permettent le dosage isotopique absolu des isotopes les moins abondants (c'est-à-dire ceux dont la concentration en poids est inférieure à 10%) avec une précision et une exactitude
ИЗМЕРЕНИЕ ИЗОТОПНОГО СОСТАВА УРАНА С ПОМОЩЬЮ МАСС-СПЕКТРОМЕТРИИ. В связи с тем, что атомная энергия находится все большее применение, возрастает необходимость в получении точных и правильных данных об изотопном составе урана. Эти данные имеют особенно большое значение в деле учета и передачи обогащенного урана для применения в реакторе и научных исследованиях, что связано с использованием делящихся материалов на миллионы долларов. Надежные измерения изотопного состава также необходимы для обеспечения соответствия состава топливных элементов конструкции реактора, а также для расчета потерь при обработке и выгорании топлива.

Для очень точных измерений изотопного состава разработаны масс-спектрометрические методы, которые намного превосходят возможности других методов. Эти методы - "одинаковый этalon" и "двойной этalon" - основаны на сравнении между измерениями неизвестного образца с аналогичными измерениями, сделанными на известных эталонах. Использование метода "двойного эталона" устраняет влияние ошибок прибора, что дает возможность произвести измерение изотопного состава с точностью (95% предела ошибки) порядка ±0,02% значений. Точность ограничена только данными об используемых значениях эталона, которые стандартизованы в соответствии с серий изотопных эталонов урана, получаемых от Национального бюро стандартов США.

Масс-спектрометры являются также полезными для абсолютного измерения изотопного состава урана, особенно в формах, иных чем UF₆. Методы тепловой ионизации, при которых используются спектрометры с большей разрешающей способностью (приблизительно 12-дюймовый радиус), позволяют давать абсолютную изотопную характеристику небольших количеств изотопов (т.е. тех, которые составляют менее 10 вес%) с точностью приблизительно 0,5% значений на один анализ. Эти анализы являются особенно полезными при калибровке высокоточного и сильно обогащенного урана для последующего их использования в качестве добавок при осуществлении программ, связанных с изотопными эталонами.

Описываются методы относительного и абсолютного измерения изотопного состава, а также их применение в целях учета и рабочего анализа. Эти методы применяются при: 1) анализе контрольных образцов для устранения расхождений измерений различных лабораторий, 2) анализе продукта с целью его соответствия требованиям транспортировки и 3) анализе урана в различных формах для его учета при защите.

MEDICION DE LOS ISOTOPOS DEL URANO POR ESPECTROMETRIA DE MASAS. El empleo cada vez mayor de la energía atómica exige un conocimiento muy preciso de la composición isotópica del urano. Este conocimiento tiene particular importancia cuando se trata de contabilizar transferencias de urano enriquecido para reactores o trabajos de investigación, ya que en estos casos el valor del material transferido puede ascender a millones de dólares. También es necesario conocer exactamente la composición isotópica del urano para cerciorarse de que los elementos combustibles se ajustan a las especificaciones previstas en el diseño del reactor y para poder calcular las pérdidas producidas durante la elaboración del combustible o las debidas a su agotamiento.

Para lograr determinaciones isotópicas sumamente precisas se han ideado métodos de espectrometría de masas muy superiores a los ya conocidos. Estos métodos - «de patrón único» y «de doble patrón» - se basan en una comparación de los resultados obtenidos con la muestra analizada y con la muestra conocida. El método de doble patrón elimina los efectos de la desviación sistemática de los instrumentos y permite determinaciones isotópicas de una precisión (límite de error de 95%) del orden de ±0,02% de los valores. La exactitud está sólo limitada por el conocimiento de los valores de los patrones utilizados, que se ajustan a la serie de patrones isotópicos del urano distribuidos por el National Bureau of Standards de los Estados Unidos.

Los espectrómetros de masas sirven también para determinar la composición isotópica absoluta del urano, especialmente cuando no se encuentra en forma de UF₆. Las técnicas de ionización térmica con empleo de espectrómetros de gran resolución (radio de 12 pulg aproximadamente) permiten la caracterización absoluta de los isótopos de menor importancia -los que constituyen menos de un 10% en peso- con una exactitud y pre-
cision de un 0.5% de los valores obtenidos por análisis. Estos análisis son particularmente útiles para calibrar el uranio muy enriquecido y el uranio muy agotado que se utilizarán como materiales de mezcla para programas de patrones isotópicos.

En la memoria se describen algunos métodos relativos y absolutos de medición isotópica y sus aplicaciones en programas de contabilidad y control analítico de operaciones. Estas aplicaciones son: a) el análisis de muestras de arbitraje para resolver discrepancias entre laboratorios, b) el análisis de los productos para comprobar las especificaciones de las remesas y c) el análisis del uranio en diversas formas para facilitar la contabilización de las partidas recibidas.

INTRODUCTION

Mass spectrometers have been closely associated with isotopic measurements since 1919 when Aston used his mass spectrograph to obtain conclusive evidence of the existence of the isotopes of neon. In atomic energy, mass spectrometers have been used extensively both to separate isotopes on a production basis and to measure isotopic composition of uranium and plutonium. In the latter application, mass spectrometers have assumed a predominant position because of their precision and accuracy.

The increasing demand for uranium, at various stages of enrichment for power reactors and research applications throughout the world, has re-emphasized the need for precise and accurate isotopic abundance measurements. This need is obvious when one considers the multi-million dollar inventories currently involved with leases of uranium and, in the fast approaching future, the large monetary exchanges to be involved in the sale of enriched uranium. Differences in isotopic abundance measurements between a shipper and receiver of uranium material result in substantial differences in the calculated monetary value of the shipment, and can best be prevented through the proper use of the most precise and accurate analytical methods available. Reliable isotopic measurements are also necessary for efficient operation of gaseous diffusion plants, for assuring compliance of fuel element compositions with the reactor design specifications, for permitting calculation of fuel burn-up losses, and for uranium-235 accountability in all processing operations.

Since the demonstrated precision and accuracy capabilities of mass spectrometry surpass those of other methods, mass spectrometry has become the most valuable technique available to determine the isotopic composition of uranium.

INSTRUMENTATION

The entire scope of mass spectrometry, including its history, theory, instrument descriptions and applications to various problems in physics and chemistry has been well documented [1-12]; therefore, only a brief description will be presented of the basic principles of mass spectrometry and of the specific detection systems and ion sources used for uranium analyses.
Principles of mass spectrometry

The basic principles involved in mass spectrometry include ion formation, collimation, acceleration, separation, collection and measurement. In Fig. 1 (which depicts a mass spectrometer with an electron bombardment ion source) the sample molecules are introduced into the source as a gas and ionized by collision with electrons emitted from the hot filament. A mass spectrometer with a thermal ionization source differs in that the sample is first deposited as a solid on the filament and is then evaporated from the filament surface as ions. The electrostatic focusing system of the source collimates the positive ions and accelerates them into the magnetic analyzer. Ion separation is accomplished in the analyzer as the ions follow circular paths having radii proportional to the square root of their mass-to-charge \((m/e)\) ratio. Ion collection is accomplished by placing collector targets in suitable positions to intercept the separated ion beams. The collector currents are then amplified and recorded.

**Ion beam collection and measurement**

Two basic techniques are used to measure the ion beams after they have been separated in the magnetic analyzer. In the first method, the individual ion currents are measured. The accelerating voltage or the magnetic field strength is varied to move, or sweep, the ion beams across a collector plate having a narrow slit. As each beam falls through the slit, one after another, each impinges upon another collector plate located behind the slit. The current caused by each ion beam as it impinges upon the second plate is amplified and recorded. The magnitudes of the recorded currents for the various ion beams are functions of the isotopic concentrations of the sample. Analyses made in this manner are absolute in nature and standards are not required as references. However, the analytical precision is limited to
about ± 0.5%/determination; and systematic errors, if present, bias the analyses.

In the second method, ratio measurements are made by centring one of the ion beams on the slit of the first collector plate so that the ions of that beam impinge on the second collector plate. All other ion beams of interest impinge upon the first plate. Each collector plate is connected to a separate amplifier, and a null detection system incorporating a bridge network is used to measure the ratio of the two currents. The resistance ratio (r) obtained is a function of the isotopic mole ratio of the sample being measured; e.g., if the $^{235}\text{U}$ ion beam is centred on the collector slit, the resistance ratio measured will be a function of the $^{235}\text{U}$ mole ratio $N_5 / (1 - N_5)$. Since simultaneous detection of the two signals is accomplished, the analytical precision of this method is better by one or two orders of magnitude. However, the numerical value of the measured resistance ratio does not provide a measurement on an absolute basis and must be related to values obtained from similar measurements on known standards. In these relative analyses, biases tend to cancel since systematic errors affect the measurements of both the unknown sample and the known standards.

**Ion sources**

Since most mass spectrometers used for making isotopic abundance measurements are first-order, direction-focusing instruments sensitive to differences in ion momentum, the ion sources must provide ions that are essentially mono-energetic. Both the electron bombardment source and the thermal ionization source provide ions having a small energy spread; each has distinct advantages and disadvantages.

In the electron bombardment source, the sample, which is introduced as a gas, is ionized by collision with electrons emitted from a hot filament (see Fig. 1). The ion beam has excellent stability and mass spectrometers using electron bombardment sources are readily adaptable to automatic operation because of the relative ease with which the gaseous samples (usually UF₆) are introduced. Comparative measurements to known standards are easily made thus permitting very precise and accurate analyses. The primary disadvantage of this source is a phenomenon known as memory that basically is caused by contamination of the source parts with the UF₆ sample as it is introduced into the source, thereby adding bias to succeeding analyses.

The thermal ionization source is extensively used for absolute analyses of uranium samples and is of particular value in analyses involving the use of isotopic dilution techniques [13]. The sample, either gas, liquid or solid, is applied to the sample filaments as a solution, evaporated to dryness, and the filament assembly is installed in the source. When the filaments are heated during the subsequent analysis the sample is evaporated as positive ions. Memory effects are much less pronounced than with the electron bombardment source. Its primary disadvantages are that sample introduction is more difficult and the ion formation rate varies with time.
RELATIVE ANALYTICAL PROCEDURES

General

Two procedures, the "double-standard method" and "single-standard method" used for making relative isotopic abundance determinations on UF₆, involve use of an electron bombardment, dual collector, mass spectrometer. Figure 2 is a schematic diagram of a sector-type (60°) spectrometer including the automatic features incorporated into the instrument at the Portsmouth Area gaseous diffusion plant.

Before the double- and single-standard methods are explained, however, a brief description of sample preparation, applicable to both methods, is needed. The sample and standard containers are attached to the manifold sample ports. Since contaminant gases, e.g. air, hydrogen fluoride and Freon, will cause bias, considerable effort is exerted to purify all UF₆ samples. Purification is accomplished by pumping off residual vapours after the UF₆ is condensed at about -60°C and/or by venting a fraction of the vapours after the UF₆ has been liquified at +70°C. After sample purification, the desired sequence of operations (i.e. the order of sample and standard introduction into the mass spectrometer) is scheduled on a programmer unit. For each entry, pneumatically actuated valves are automatically controlled by the programmer to admit and remove the sample (or standards) as required; a leak valve automatically regulates the sample flow; the accelerating voltage is automatically adjusted so that the ion beam of primary interest is centred on the slit; and the resistance ratio is nulled and recorded by the automatic read-out system.

Double-standard method

The double-standard (interpolative) method is designed to minimize the effects of memory. The method consists of analysing a sequence of two standards along with the sample. Standard A has a $^{235}\text{U}$ mole ratio $N_5/1 - N_5$ lower than that of the sample $X$, and standard B has a mole ratio that is higher. The ratio of mole ratios of the standard pair is normally about 1:3. A closely timed, symmetrical sequence of six introductions is used for each determination — each sequence beginning and ending with the low standard (A X B B X A) [14-16].

Two and a half minutes is allowed for each entry in the sequence — one minute for stabilization, another minute for data recording, and thirty seconds for pumpout before the next entry in the sequence is admitted. During the first minute of each sample introduction, the flow through the leak valve into the source is adjusted to a predetermined value. The resistance ratio ($r$) is recorded throughout the entire two minutes to provide a visual record of the stabilization period as well as data for the measurement. This procedure is followed until completion of the six-step sequence. The chosen sequence should be used consistently for a series of analyses so that one determination ends with, and the next determination begins with, the same standard. If different standards are needed to bracket the next sample, a conditioning sequence of the new standards, e.g., ABBBBA or AABBA, should be used to meet this requirement.
Average resistance ratios, or scalers, are calculated for the A-standard ($r_A$), the B-standard ($r_B$), and the unknown sample ($r_X$). The differences in scalers between the high and low standards and between the sample and low standards are calculated. The difference in mole ratios between the standards ($\Delta H = H_B - H_A$) is calculated.

The ratio of differences ($R_D$) is defined as $r_X - \frac{r_A}{r_B} - r_A$ and the mole ratio ($H_X$) of the sample is calculated from the interpolative equation

$$H_X = R_D \Delta H + H_A$$  \hspace{1cm} (1)

The weight percent of the isotope of interest is calculated from the relationship

$$\text{Weight percent} = \frac{100}{100 + H_X}$$  \hspace{1cm} (2)

The small residual bias associated with this method is no larger than $\pm 0.02\%$ and even this may be corrected by making "end-point" determinations. The end-point controls are additional measurements with one of the standards, A or B, substituted for the unknown in the analytical sequence. Thus, for an end-point measurement, one of the sequences A A B A A or A B B B B A is followed and the results are used to make corrections for residual bias in the measurements on the unknown.

**Single-standard method**

The single-standard method involves the direct comparison of an unknown sample to a known standard. A single determination is composed of six alternate entries of the standard and sample ($AXAAXA$) or ($BXBXBX$); the timing involved in the sequence is the same as that used for the double-standard method. The resistance ratio of the first standard entry is discarded to make the sample-standard interaction uniform. The three sample entry ratios and the remaining two standard entry ratios are averaged and the observed ratio of resistance ratios ($R_O$) is calculated.

$$R_O = \frac{r_X}{r_S}$$  \hspace{1cm} (3)

where $r_X$ is either $r_A$ or $r_B$. Memory corrections must be made since the bias associated with this method is about $\pm 0.3\%$. The corrections are made by the following equations:

$$M = \frac{W_T - 1}{R_O - 1}$$  \hspace{1cm} (4)

where $M$ is the memory factor of the instrument and $W_T$ is the known ratio of mole ratios $H_A/H_B$ of a pair of standards A and B, and $R_O = r_A/r_B$ from independent measurements;

$$W_C = R_O M - (M - 1)$$  \hspace{1cm} (5)
FIG. 2. Block diagram of sector-type mass spectrometer.
where $W_C$ is now the calculated ratio of mole ratios for the sample with $R_0 = \frac{r_x}{r_S}$. The mole ratio of the sample ($H_X$) is then calculated by the following equation, and the $^{235}\text{U}$ content of the sample is then calculated from Eq. (2):

$$H_X = W_C H_5 = H_5 \left( R_0 M - (M - 1) \right)$$

(6)

where $H_5$ is the mole ratio of the standard.

**Discussion of errors**

The measured resistance ratio ($r_X$) is usually assumed to be proportional to $N_5 / 1 - N_5$. Unfortunately, the actual relationship between the resistance scaler readings and the sample mole ratios is much more complex, and instrument characteristics such as drop out, resolution, memory and measuring system non-linearity contribute to the complexity of the relationship.

The mass spectrometers used for relative isotopic ratio measurements do not completely separate or resolve individual ion beams, and when the $^{235}\text{U}$ beam is centred on the slit, some of the other isotopes also fall through the slit. This means that the ratio $N_5 + g (1 - N_5) / (1 - N_5) (1 - g)$ is measured rather than the desired ratio $N_5 / 1 - N_5$ ($g$ equals the fraction of the other isotopes that fall through the slit). The magnitude of this error can be reduced if the collector slit is narrowed; however, when the slit is narrowed, not all of the $^{235}\text{U}$ beam falls through the slit and the ratio $d N_5 / 1 - d N_5$ is measured ($d$ equals the fraction of $^{235}\text{U}$ that falls through the slit). The magnitudes of calculated deviations of resistance ratios from the sample mole ratios are shown in Table I. The errors shown here have been purposely exaggerated to more clearly illustrate the non-linearity of the relationship between the sample mole ratio and the measured resistance ratio. In practice and as a compromise, the slit is generally adjusted so that 95-97% of the $^{235}\text{U}$ beam and only about 0.01% of the other beams go through the slit.

If the resistance ratios versus the sample mole ratios are plotted, a curve similar to that shown in Fig. 3 is obtained.

It is evident from this figure that the true ratio of mole ratios $N_2 / N_1$ is not equal to the observed ratio of resistance ratios, $r_2 / r_1$, but is equal to $r_2 - r_0 / r_1 - r_0$. However, the application of the instrument memory factor ($M$) compensates for this error since a similar error is involved in the determination of the memory factor. This is true, of course, only if the resolution of the instrument has not changed since $M$ was evaluated. The additive term, $r_0$, that is present in the measured resistance ratios does not cause an error in the double-standard method since $R_0$ is the ratio of differences and the additive term is essentially removed by subtraction in both the numerator and the denominator.

The true memory of a mass spectrometer may be demonstrated by first conditioning an instrument, over a period of several hours, with a UF$_6$ sample containing $^{233}\text{U}$. Measurements are then made on a UF$_6$ sample not containing $^{233}\text{U}$. The ratio $N_3 / 1 - N_3$ is monitored during each entry of the new sample and is representative of the amount of $^{233}\text{U}$ that is exchanging
TABLE I

DEVIATIONS OF RESISTANCE RATIOS

<table>
<thead>
<tr>
<th>Weight Percent $^{235}U$</th>
<th>Mole Ratio</th>
<th>$^{90}%$ $^{235}U$ Through Slit</th>
<th>Percent Deviation</th>
<th>$^{100}%$ $^{235}U + 0.1%$ Other Isotopes Through Slit</th>
<th>Percent Deviation</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.500</td>
<td>0.00509</td>
<td>0.00458</td>
<td>-10.0</td>
<td>0.00610</td>
<td>+20.0</td>
</tr>
<tr>
<td>1.000</td>
<td>0.01023</td>
<td>0.00920</td>
<td>-10.1</td>
<td>0.01124</td>
<td>+9.9</td>
</tr>
<tr>
<td>5.000</td>
<td>0.05331</td>
<td>0.04772</td>
<td>-10.5</td>
<td>0.05436</td>
<td>+1.2</td>
</tr>
<tr>
<td>15.00</td>
<td>0.1787</td>
<td>0.1580</td>
<td>-11.6</td>
<td>0.1799</td>
<td>+0.66</td>
</tr>
<tr>
<td>35.00</td>
<td>0.5453</td>
<td>0.4654</td>
<td>-14.7</td>
<td>0.5468</td>
<td>+0.28</td>
</tr>
<tr>
<td>60.00</td>
<td>1.5189</td>
<td>1.1868</td>
<td>-21.9</td>
<td>1.5214</td>
<td>+0.16</td>
</tr>
</tbody>
</table>

FIG. 3. Resistance ratios versus sample mole ratios

with the $UF_6$ sample being introduced. Although the $N_3/1 - N_3$ ratio falls off very rapidly during the first entries it still is detectable after weeks of normal operation and clearly demonstrates the sample interaction called memory. The bias due to this sample interaction is also included in the correction factor, $M$, used in the single-standard method. In the double-standard method the effects of memory are reduced, to a large extent, by subtraction.
**Precision and accuracy of relative methods**

In the relative methods described, an unknown sample is compared or related to a known standard or standards. In the double-standard method, the ratio of differences, \( R_D \) or \( \frac{r_X - r_A}{r_S - r_A} \), is measured. In the single-standard method the observed ratio, \( R_O \) or \( \frac{r_X}{r_S} \), is measured and then corrected by the application of the instrument correction factor, \( M \), obtained by making similar measurements on known standards.

If the variances (\( V \)) associated with the measurements of \( R_D \), \( R_O \) and \( M \) are known, the analytical precisions of the two methods may be estimated from Eqs. (7) and (8) derived from the working equations (6) and (1) shown below:

\[
\begin{align*}
\text{Single standard} & \\
(6) & \quad H_X = H_S [R_OM - (M-1)] \\
(7) & \quad V_{H_X} = (H_S M)^2 V_{R_O} + [H_S (R_O - 1)]^2 V_M \\
\text{Double standard} & \\
(1) & \quad H_X = R_D \Delta H + H_A \\
(8) & \quad V_{H_X} = (\Delta H)^2 V_{R_D}
\end{align*}
\]

Under routine conditions the variance of measurements of \( R_O \) is about \( 50 \times 10^{-10} \) and the variance of measurements of \( R_S \) is about \( 50 \times 10^{-8} \) for isotopic mass spectrometric measurements made at Goodyear Atomic Corporation. However, when the usual standard spread of 30% is used for the double-standard method, the coefficient \( (\Delta H)^2 \) is only about 1/15 of the value of the single standard coefficient \( (H_S M)^2 \). For special tests requiring better analytical precision, the instrument operating parameters are more closely controlled and the variance of \( R_O \) is about \( 36 \times 10^{-10} \) and the variance of \( R_D \) is about \( 10 \times 10^{-6} \).

In the single-standard method, however, the correction factor, \( M \), is dependent on several variables, and the variance of \( M \) is relatively high—normally of the order of \( 200 \times 10^{-6} \). Even under optimum conditions the variance of \( M \) is of the order of \( 6 \times 10^{-6} \). However, in the single-standard method the variance component affected by \( V_M \) disappears when \( R_O = 1 \) and, for this reason, the single-standard method becomes more precise when \( R_O \) is very close to unity.

Using either of these relative methods, analytical precisions of better than 0.1% determination are obtained routinely, and the accuracy of each analysis is dependent primarily on the accuracy of the standard values.

**ABSOLUTE PROCEDURES**

**General**

Accurate absolute analyses may be made only if all the ions of a particular \( m/e \) value and none of the ions of adjacent beams go through the slit. The high resolving power required is obtained by producing high-energy ions (8 to 20 kV), using a large ion path radius (12-15 in.), and using a vacuum system capable of maintaining the analyser tube at a pressure in the \( 10^{-8} \) Torr region. Either single or multiple thermal filament assemblies can be used.
but most uranium isotopic analyses are made using a triple-filament assembly. If the analysis time is an important factor, a vacuum lock system can be used to facilitate changing the filaments without introducing a high pressure in the ion chamber. However, the ultimate vacuum obtained with such a system is generally not so low and, since the resolution of the instrument is directly dependent upon the operating pressure, the quality of the analyses is somewhat less.

Uranium samples are prepared for analysis by first converting the sample to uranyl nitrate by addition of dilute nitric acid. Small drops of this uranyl nitrate solution are successively placed on the centre of the side filaments of a triple filament assembly [14] and evaporated to dryness by connecting the filaments to a low-voltage source. After sufficient sample has been deposited, the voltage is increased until the sample turns yellow then orange, indicating formation of UO$_3$. The filament assembly is then installed in the ion source. When operating pressure is achieved, the filament currents are adjusted to produce positive ions from the uranium contained in the sample.

Absolute isotopic abundance measurements are made by sweeping (i.e., varying) the accelerating voltage or magnetic field strength in such a manner as to obtain a scan of the individual ion beams from the lower through the higher mass isotopes. The sweep is then reversed to obtain a scan of the ion beams in the opposite direction. Generally, eight such scans — lower through higher mass and reverse — are made per filament loading. Assuming a linear intensity drift for each scan, averaging the ion peak heights will give reasonably accurate measurement for calculation of the isotope ratio or abundance percentages. In these measurements, the uncertainty concerning the percent concentration of the principle isotope is associated with the measurement of the minor isotopes, and as the concentrations of the minor isotopes become smaller, the uncertainty of the principle isotope also becomes smaller. In general, the precision of analyses by this method is equivalent to ±0.5% of the minor isotopes per filament loading.

If somewhat better accuracy is desired, the sample may be spiked with a known amount of $^{233}$U isotope; i.e., weighed amounts of the sample and $^{233}$U oxides are combined for analysis permitting comparison of other isotope peaks to the $^{233}$U peak. Should complete isotopic characterization of the sample be required and should the best precision and accuracy be desired for each isotope, the $^{234}$U and $^{236}$U isotopes are determined by using the thermal ionization instrument and the $^{235}$U or $^{238}$U isotope is determined by a relative procedure using the electron bombardment spectrometer.

**The determination of uranium concentration**

To establish accountability for receipts of impure samples such as waste solutions or dissolver solutions, it is necessary to know both the isotopic and the total uranium concentrations. The use of the isotope $^{233}$U spike technique offers the unique advantage of permitting the determination of both total uranium content and uranium isotopic distribution of the sample with a single analysis. The analysis must be one of the absolute measurement type wherein a scan of the peaks of the isotopes, $^{233}$U – $^{238}$U, is made by varying either the high voltage or the magnetic field strength. The ratio
of the sum of the other peak heights to the height of the $^{235}$U peak, multiplied by the quantity of $^{233}$U added to the sample, gives the quantity of uranium in the sample. Since the total quantity of sample to which the $^{233}$U was added is known, the uranium concentration in the original sample can be calculated. Also, since the $^{233}$U spike should contain insignificant amounts of the naturally occurring isotopes, the $^{235}$U-$^{238}$U isotopic distribution in the original sample should be the same as that observed in the mixture, and the peak heights for these isotopes in the mixture can be used to calculate the isotopic ratios for the sample.

APPLICATIONS

_Umpire analyses_

When uranium material is transferred from one plant or processor to another, any monetary exchange or accountability credit is based on the amount of the fissionable $^{235}$U isotope involved. In these transactions, isotopic concentration or purity differences between the parties are resolved by umpire laboratory analyses. The Goodyear Atomic Corporation has acted in this capacity to resolve both isotopic concentration and purity differences; typical results are presented in Tables II and III.

### Table II

**UMPIRE ISOTOPIC CONCENTRATION ANALYSES**  
(Weight Percent $U^{235}$)

<table>
<thead>
<tr>
<th>Sample</th>
<th>Weight Percent $U^{235}$</th>
<th>Lab A</th>
<th>Lab B</th>
<th>Goodyear</th>
</tr>
</thead>
<tbody>
<tr>
<td>$UO_2$</td>
<td></td>
<td>1.600</td>
<td>1.663</td>
<td>1.600 EB*</td>
</tr>
<tr>
<td>Uranyl nitrate</td>
<td></td>
<td>90.76</td>
<td>93.18</td>
<td>90.82 EB</td>
</tr>
<tr>
<td>hydrate</td>
<td></td>
<td>89.20</td>
<td>89.73</td>
<td>89.20 EB</td>
</tr>
<tr>
<td>Dissolver solutions</td>
<td></td>
<td>87.27</td>
<td>87.48</td>
<td>87.27 EB</td>
</tr>
<tr>
<td></td>
<td></td>
<td>17.39</td>
<td>11</td>
<td>17.36 EB</td>
</tr>
<tr>
<td></td>
<td></td>
<td>85.62</td>
<td>77</td>
<td>85.73 TI</td>
</tr>
<tr>
<td></td>
<td></td>
<td>90.28</td>
<td>81.07</td>
<td>90.32 TI</td>
</tr>
</tbody>
</table>

*Electron bombardment gas instrument  
†Thermal ionization mass spectrometer

*Product analyses*

The mass spectrometer double-standard method is used for control of various product streams from the Portsmouth diffusion plant. Some measurements are shown in Table IV that indicate results obtained on one
### TABLE III

**UMPIRE PURITY ANALYSES — DISSOLVER SOLUTIONS**  
*(Grams of \( \text{U} \) per Gram of Sample)*

<table>
<thead>
<tr>
<th>Lab A</th>
<th>Lab B</th>
<th>Chemical</th>
<th>Spectrometer*</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.00053</td>
<td>0.00050</td>
<td>0.00054</td>
<td>0.00056</td>
</tr>
<tr>
<td>0.00065</td>
<td>0.00072</td>
<td>0.00066</td>
<td>0.00067</td>
</tr>
<tr>
<td>0.00084</td>
<td>0.00077</td>
<td>0.00083</td>
<td>0.00084</td>
</tr>
<tr>
<td>0.00092</td>
<td>0.00084</td>
<td>0.00088</td>
<td>0.00090</td>
</tr>
<tr>
<td>0.00112</td>
<td>0.00106</td>
<td>0.00109</td>
<td>0.00111</td>
</tr>
<tr>
<td>0.00189</td>
<td>0.00104</td>
<td>0.00180</td>
<td>0.00190</td>
</tr>
<tr>
<td>0.00942</td>
<td>0.00909</td>
<td>0.00950</td>
<td>0.00942</td>
</tr>
<tr>
<td>0.03346</td>
<td>0.03163</td>
<td>0.03345</td>
<td>0.03332</td>
</tr>
<tr>
<td>0.08246</td>
<td>0.07575</td>
<td>0.08102</td>
<td>0.08102</td>
</tr>
<tr>
<td>0.08427</td>
<td>0.07937</td>
<td>0.08445</td>
<td>0.08430</td>
</tr>
</tbody>
</table>

*Using the isotopic spike technique.

### TABLE IV

**PRODUCT CONTROL ANALYSES**

<table>
<thead>
<tr>
<th>Date Analyzed</th>
<th>Weight Percent</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>( \text{U}^{234} )</td>
</tr>
<tr>
<td>3/25/63</td>
<td>0.8899</td>
</tr>
<tr>
<td>3/29/63</td>
<td>0.8904</td>
</tr>
<tr>
<td>4/01/63</td>
<td>0.8905</td>
</tr>
<tr>
<td>4/08/63</td>
<td>0.8896</td>
</tr>
<tr>
<td>4/15/63</td>
<td>0.8897</td>
</tr>
<tr>
<td>4/22/63</td>
<td>0.8905</td>
</tr>
<tr>
<td>4/29/63</td>
<td>0.8908</td>
</tr>
<tr>
<td>5/13/63</td>
<td>0.8904</td>
</tr>
<tr>
<td>5/20/63</td>
<td>0.8902</td>
</tr>
<tr>
<td>Mean</td>
<td>0.8902</td>
</tr>
<tr>
<td>LE/det*</td>
<td>± 0.001</td>
</tr>
<tr>
<td>Percent LE/det</td>
<td>± 0.106</td>
</tr>
</tbody>
</table>

* Limit of error per determination at the 95 percent confidence level.

Control sample analysed repetitively. Precision and accuracy at other \( \text{U}^{235} \) concentrations are consistently better than 0.1% of the \( \text{U}^{235} \) values. In these
### TABLE V

**THERMAL IONIZATION ANALYSES OF DISSOLVER SOLUTIONS**

<table>
<thead>
<tr>
<th>Date</th>
<th>Type*</th>
<th>Weight Percent</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>U²³⁵</td>
</tr>
<tr>
<td>July</td>
<td>1</td>
<td>0.995</td>
</tr>
<tr>
<td>Aug.</td>
<td>2</td>
<td>0.995</td>
</tr>
<tr>
<td>Sept.</td>
<td>2</td>
<td>0.996</td>
</tr>
<tr>
<td>Oct.</td>
<td>1</td>
<td>0.997</td>
</tr>
<tr>
<td>Nov.</td>
<td>1</td>
<td>0.996</td>
</tr>
<tr>
<td>Nov.</td>
<td>2</td>
<td>0.997</td>
</tr>
<tr>
<td>Dec.</td>
<td>2</td>
<td>0.994</td>
</tr>
<tr>
<td>Dec.</td>
<td>1</td>
<td>0.997</td>
</tr>
<tr>
<td>Jan.</td>
<td>1</td>
<td>0.993</td>
</tr>
<tr>
<td></td>
<td></td>
<td>0.995</td>
</tr>
</tbody>
</table>

Mean: 0.995

**LE/detm:** 0.0027

**% LE/detm:** 0.27%

---

*Sample type 1 is a pure uranium solution having a concentration of 0.01 gU/g sample. Sample type 2 contains uranium, molybdenum, and zirconium and has a concentration of 0.005 gU/g sample.

### TABLE VI

**THERMAL IONIZATION ANALYSES OF URANYL NITRATE HYDRATE SAMPLES**

<table>
<thead>
<tr>
<th>Analysis Number 1</th>
<th>January</th>
<th>February</th>
<th>March</th>
<th>April</th>
<th>May</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>3.936</td>
<td>3.938</td>
<td>3.940</td>
<td>3.938</td>
<td>3.941</td>
</tr>
<tr>
<td></td>
<td>3.957</td>
<td>3.944</td>
<td>3.956</td>
<td>3.939</td>
<td>3.934</td>
</tr>
</tbody>
</table>

Mean: 3.942

**LE/Detm. ± 0.016**

**% LE/Detm. ± 0.408**
measurements, the collector slit is adjusted for the $^{238}\text{U}$ analyses so that approximately 97% of the $^{238}\text{U}$ beam falls through the slit. However, for the $^{234}\text{U}$ and $^{236}\text{U}$ analyses, the slit is narrowed and only about 60% of the beam drops through. The $^{234}\text{U}$ concentrations are obtained by comparing the $N_4 / (1 - N_4)$ ratio of the sample to the ratios of two bracketing standards. The $^{236}\text{U}$ concentrations are calculated from the ratio of the $^{234}\text{U}$ and $^{236}\text{U}$ ion peaks of the individual samples.

Accountability for receipts

Uranium materials received for processing may be in the form of UF$_6$, U$_3$O$_8$, UO$_2$, uranyl nitrate hydrate or uranium solution. To establish accountability for these receipts, the materials must be analysed for weight percent $^{235}\text{U}$. When the material is in the form of UF$_6$ the sample is compared to isotopic standards on an electron bombardment gas instrument, and the limit of error of a single determination, at the 95% confidence level, will range from about ± 0.1% to about ± 0.01%, depending upon the concentration. However, when the samples are received in a form other than UF$_6$ they must either be converted to UF$_6$ for analyses on a gas instrument or, more conveniently, they may be analysed on a thermal-ionization mass spectrometer.

Realistic estimates of the analytical precision and bias associated with routine thermal ionization analyses have been obtained at the Goodyear Atomic Corporation by evaluating data reported for a special inter-laboratory control programme using both control and production samples. Each month, as part of the programme, three uranium solution and two uranyl nitrate hydrate (UNH) samples were received for analysis. About one half of the solutions were pure uranium and one half were impure solutions. The $^{235}\text{U}$ concentrations ranged from 2 to 93%. The UNH samples ranged from about 3 to 7 wt. % $^{235}\text{U}$.

In April 1965, the results of this programme were reported and, at this time, the identity of the control samples was given. The values reported by Goodyear Atomic for the control samples, and the precision statements are given in Tables V and VI.

REFERENCES

THE EVOLUTION OF THE ACCURACY OF ISOTOPIC ANALYSIS BY THERMAL IONIZATION FROM 2% TO 0.02%

W.R. SHIELDS
NATIONAL BUREAU OF STANDARDS,
WASHINGTON, D.C., UNITED STATES OF AMERICA

Abstract — Résumé — Аннотация — Resumen

THE EVOLUTION OF THE ACCURACY OF ISOTOPIC ANALYSIS BY THERMAL IONIZATION FROM 2% TO 0.02%. A detailed study of the parameters that affect the precision, both internal and external, of an isotopic analysis has been made. The magnitude of the effects of some of these parameters on the analysis of transuranic elements is shown.

The calibration of mass spectrometers at many laboratories is ineffective because of the failure to consider the many different components that enter into the total bias of the analytical system. Most laboratories take cognizance of the physical bias factors associated with mass spectrometers but are unaware of the biases that are introduced in the sample preparation procedures. The usual calibration reduces the size of only a few of the bias terms and leads to erroneous statistical calculations of the accuracy of an analysis. For example, the intercomparison of two identical samples, e.g. shipper receiver samples on a container, frequently shows significant disagreement between results which may be partly or wholly due to differences in sample preparation and/or analytical procedure.

The effects of pH, concentration and composition or purity on an analysis are not fully understood, if indeed they are acknowledged to exist at all. However, investigations at the National Bureau of Standards in conjunction with the Uranium Standards Program have shown that each of these parameters does have an effect. Moreover, by changes in the analytical method or by modification of the mass spectrometer, the magnitudes of the effects can be altered from a point at which the bias terms are generally additive to one at which they are compensating. Thus, provable accuracy statements are contingent on the identification and control of all bias factors, including those involved in sample preparation and analytical procedure as well as those involving the collector system and measuring circuit. The measuring circuit is of prime importance since those in use at most laboratories have a variable bias component of the order of 0.5% which must be removed before the effects of many of the other bias components can be detected. A simple solution to this problem is the use of an expanded scale recorder such as the one developed at the National Bureau of Standards.

The product of this research is a table of the best estimates of the isotopic compositions of 16 uranium isotope standards distributed by the National Bureau of Standards. The current level of analytical proof of these values will be given along with a discussion of the work (in progress) that is necessary to change the analytical precision statements to accurate statements.

DEGREG D'EXACTITUDE DE L'ANALYSE ISOTOPIQUE PAR IONISATION THERMIQUE: PASSAGE DE 2% A 0,02%. Les paramètres qui influent sur la précision, tant interne qu'externe, d'une analyse isotopique ont fait l'objet d'une étude minutieuse. L'auteur fait ressortir l'ampleur des effets que certains de ces paramètres ont sur l'analyse des éléments transuraniens.

L'étalonnage des spectromètres de masse dans de nombreux laboratoires est en fait inefficace, étant donné que l'on ne prend pas en considération les nombreuses composantes de l'erreur systématique totale du système d'analyse. La plupart des laboratoires tiennent compte des composantes de l'erreur systématique qui sont imputables à la physique des spectromètres, mais ils ne se rendent pas compte des erreurs attribuables aux méthodes de préparation des échantillons. L'étalonnage habituel ne réduit que quelques-unes des composantes de l'erreur, ce qui fausse les calculs statistiques portant sur l'exactitude de l'analyse. Par exemple, la comparaison de deux échantillons identiques, tels que les échantillons prélevés par l'expéditeur et le destinataire d'un envoi, révèle bien souvent une différence assez importante entre les divers résultats, laquelle peut résulter en partie, sinon exclusivement, des différences de préparation des échantillons ou de la méthode d'analyse.

Les effets que le pH, la concentration et la composition ou le degré de pureté exercent sur l'analyse ne sont pas très bien compris, si tant est que l'on en reconnaisse l'existence. Cependant, les recherches effectuées
par le National Bureau of Standards conjointement avec les organismes chargés du programme relatif aux étalons d'uranium ont démontré que chacun de ces paramètres a un effet. En outre, on peut, en modifiant la méthode d'analyse ou le spectromètre de masse, arriver au point où, d'une manière générale, les termes de l'erreur systématique se compensent au lieu de s'ajouter. Ainsi, on ne peut démontrer l'exactitude que dans la mesure où l'on a pu identifier et réduire toutes les composantes de l'erreur, y compris celles qui interviennent dans la préparation des échantillons et dans la méthode d'analyse, ainsi que celles qui intéressent le système collecteur et le circuit de mesure. Ce dernier est d'une importance primordiale car les circuits utilisés dans la plupart des laboratoires comportent un élément d'erreur systématique variable de l'ordre de 0,5% qu'il faut éliminer avant de pouvoir déceler les autres composantes de l'erreur. On peut résoudre ce problème d'une manière assez simple en utilisant un enregistreur à échelle élargie tel que celui qui a été mis au point par le National Bureau of Standards.

Cette recherche a abouti à l'établissement d'un tableau indiquant les meilleures estimations des compositions isotopiques de 16 étalons d'isotopes d'uranium qui ont été distribués par le National Bureau of Standards. Le mémoire précise le degré de rigueur actuel de la preuve analytique de ces valeurs et commente l'étude - actuellement en cours - qui est nécessaire pour transformer les énoncés de la précision d'analyse en énoncés de l'exactitude.

ПОСТЕПЕННОЕ ПОВЫШЕНИЕ ТОЧНОСТИ ИЗОТОПНОГО АНАЛИЗА ТЕПЛОВОЙ ИОНИЗАЦИЕЙ С 2% ДО 0,02%. Подробно изучали параметры, отражающиеся на внутренней и внешней точности изотопных анализов. Показывается степень влияния этих параметров на анализ трансурановых элементов.

Калибрование масс-спектрометров во многих лабораториях часто неэффективно из-за недостаточного учета многих различных компонентов, определяющих общую погрешность определенной системы анализа. Большая часть лабораторий отдает себе отчет в вызывающих погрешность физических факторах, связанных с масс-спектрометрами, но они упускают из вида погрешности, возникающие в ходе подготовки проб. В этом случае обычные методы калибровки уменьшают величину только некоторых из факторов погрешностей, что приводит к ошибочным расчетам точности анализа. Примером такого рода проблемы может служить взаимное сравнение двух тождественных проб, как, например, проб, взятых из того же самого контейнера соответственно отправителем и получателем, которые нередко указывают на значительные расхождения между результатами анализа, частично или полностью, вызываемые различиями в способе приготовления проб или в методе анализа, или в том и другом.

Влияние pH, концентрации и состава или степени его чистоты на результаты анализа еще не полностью понимается, если вообще отдается отчет в существовании этих факторов. Однако проведенные в Национальном бюро стандартов исследования в связи с программой урановых стандартов показали, что каждый из этих параметров оказывает определенное воздействие. Более того, посредством изменения метода анализа или внесения изменений в масс-спектрометр величина эффектов может измениться таким образом, что отдельные погрешности будут прибавляться к тому, что они должны компенсировать. Таким образом допустимые утверждения относительно правильности анализа зависят от выявления и исправления всех факторов погрешностей, присущих подготовке проб и методам анализа и связанных с системой выборки проб и системой промеров. Система промеров имеет первостепенное значение, так как применяемые в большинстве случаев лабораторией системы страдают переменной погрешностью порядка 0,5%, и эта погрешность должна быть устранена, прежде чем удастся установить другие факторы, от которых зависит погрешность. Простым решением этой проблемы может быть использование регистрирующего устройства с развёркой шкал, аналого-тактового устройства, разработанного в Национальном бюро стандартов.

Результатом этого исследования является таблица, содержащая наиболее достоверные данные об изотопном составе шестнадцати урановых стандартов, рассылаемых Национальным бюро стандартов. Дается указание о степени точности анализа, достигнутой в настоящее время, и вместе с тем обсуждается программа работы (проводимой в настоящее время), необходимой для того, чтобы заменить указания о точности анализа указанием о его правильности.

AUMENTO DE LA EXACTITUD DE LOS ANALISIS ISOTOPICOS POR IONIZACION TERMICA, DEL 2% AL 0,02%. Se ha hecho un detenido estudio de los parámetros que influyen en la precisión, tanto interna como externa, de los análisis isotópicos. En la memoria se indica la gran influencia que algunos de esos parámetros tienen en el análisis de los elementos transuránicos.
La calibración de los espectrómetros de masas que se lleva a cabo en muchos laboratorios resulta ineficaz debido a que no se toman en consideración las numerosas componentes que intervienen en la desviación sistemática total del método de análisis. En la mayor parte de los laboratorios se conocen los factores físicos que determinan dicha desviación en los espectrómetros de masas pero se ignoran las desviaciones introducidas en los procedimientos que se siguen para la preparación de las muestras. La calibración corriente sólo reduce algunas de las condiciones de la desviación y conduce a cálculos estadísticos erróneos sobre la exactitud de los análisis. Por ejemplo, la comparación de dos muestras idénticas, tales como las que de un mismo recipiente se envían al expedidor y al destinatario, pone a menudo de manifiesto discrepancias significativas entre los resultados, que pueden ser en parte, si no enteramente, debidas a diferencias en la preparación de las muestras o en el procedimiento analítico, o en ambos a la vez.

Suponiendo que se reconozca su existencia, no se comprenden aún completamente los efectos que sobre un análisis tienen el pH, la concentración y la composición o pureza. No obstante, las investigaciones realizadas en el National Bureau of Standards en relación con el programa de patrones de uranio muestran que cada uno de esos parámetros tiene un efecto. Además, introduciendo algunos cambios en el método analítico, o modificando el espectrómetro de masas, es posible alterar la magnitud de los efectos desde un punto en el que las condiciones de la desviación sistemática suelen ser aditivas hasta otro punto en el que dichas condiciones se compensan. Así, la probabilidad de los enunciados de exactitud depende de la medida en que se hayan identificado y controlado todos los factores de desviación sistemática, tanto los inherentes a la preparación de la muestra y al procedimiento de análisis como los que intervienen en el sistema colector y en el circuito de medición. Este último es de primordial importancia, ya que en los circuitos usados en la mayor parte de los laboratorios existe una componente variable de la desviación sistemática, del orden de 0,5%, que es preciso eliminar para poder detectar los efectos de muchas de las otras componentes. Este problema se resuelve sencillamente empleando un registrador de escala amplificada como el que se utiliza en el National Bureau of Standards. El resultado de esa investigación es una tabla que contiene las mejores estimaciones de la composición isotópica de dieciséis patrones de isótopos de uranio distribuidos por el National Bureau of Standards. En la memoria se indica el nivel general de la prueba analítica de esos valores y se exponen los trabajos necesarios para poder transformar los enunciados de precisión estadística en enunciados de exactitud. Estos trabajos están en curso de realización.

The utilization of mass spectrometers by the nuclear materials management groups generally falls into two categories: relative gas analysis, such as UF₆, and the pseudo-absolute analysis of solids by surface or thermal ionization. The aim of this paper is to show that the adaptation and application of some of the analytical procedures and instrumentation used in relative gas analyses can significantly increase the precision and accuracy of the solids analyses. Many parameters which have similar effects in both types of analysis have long been recognized and controlled by the workers in gas analysis, but have been almost uniformly ignored by the workers doing solids analysis.

Figure 1 is a pictogram showing the interrelationships of some very old experimental data taken on the Uranium Standards. The data were supplied by laboratories in both Europe and the United States of America and show a very interesting first partitioning of the variance components which constitute the uncertainty statement of an isotopic analysis using surface or thermal ionization. All the data have theoretically been corrected for all known biases and each bit of information has a calculated analytical precision statement of better than ± 0.25%. A very cursory examination of Fig. 1 reveals two obvious bias factors: one due to the difference in the mode of ionization, evidenced by the average 1.1% bias of the single filament - D.C. amplifier results; and the other due to the mode of collecting and measuring the ions, evidenced by the average 0.55% bias superimposed on the results of both modes of ionization when a multiplier is used. And,
FIG. 1. Experimental $^{235}\text{U}/^{238}\text{U}$ versus NBS best estimate
finally, superimposed on both of these systematic errors, is a random error or average limit of error of bias of approximately 0.5%. The random error is made up of two components, the most important of which is due to the strip chart recorder. Manufacturers of strip chart recorders have for years supplied the information that new recorders have possible errors of 0.1% (full scale) in dead zone and 0.25% (full scale) in linearity. Still, the recognition that these parameters are the limiting factor (0.5% limit of error of bias) on the precision of most isotopic analyses is, unfortunately, not widespread. To circumvent this limitation all recorders used at the National Bureau of Standards are supplied with an expanded-scale circuit (Fig. 2). This circuit is very similar to the circuits used on many UF₆ gas instruments and its effective operation requires only that ten wire wound resistors remain constant indefinitely and that the reference potential remain constant for at least a few minutes. Installation of the expanded-scale circuits did not require any modification of the existing amplifiers, either in the mass spectrometer or the recorder, and they have performed very well for five years. The only necessary maintenance has been an occasional adjustment of the slidewire shunt to balance the effects of slidewire wear and maintain the resistance at 110 ohms.

FIG. 2. Recorder with expanded scale circuit
With the use of an expanded scale recorder several lower level bias factors become apparent, all of which are operative in the source region of the mass spectrometers and are involved in the chemistry and physics of ionization and not the ion optics directly. The first major variability factor to be observed was that arising from pressure variations. This effect became readily apparent when results from analyses performed after an all-night pump-down were compared with those obtained after a pump-down of only a few minutes. This variability component was removed by the installation of a liquid nitrogen cold finger directly in the source region; thereby maintaining a much better vacuum in the source region at all times. The next error component to be identified and removed was the variability due to variations in the temperature needed to achieve the standard ion beam intensities with different samples. For pure samples, the two major factors which appear to determine the final temperature of analysis are the pH of the sample solution and the filament heating pattern used. The pH effect is removed by running all samples of all transuranic elements from a 10% nitric acid solution. The temperature effect which arises from variations in the filament heating procedure are very interesting and make it possible to "dial-in" a bias. The data in Tables I, II, and III were taken in order to demonstrate the level at which filament bias usually controlled by the analyst and to show the magnitude of the effect introduced by variations in the heating pattern.

### Table I

**COMPARISON OF THE CALCULATED $^{235}$U/$^{238}$U RATIO TO THE OBSERVED $^{235}$U/$^{238}$U RATIO FOR HIGH TEMPERATURE ANALYSES OF SYNTHETIC URANIUM SAMPLES**

<table>
<thead>
<tr>
<th>Group 1</th>
<th>Group 2</th>
<th>Group 3</th>
</tr>
</thead>
<tbody>
<tr>
<td>Approximate ratio 1:9</td>
<td>Approximate ratio 1:1</td>
<td>Approximate ratio 9:1</td>
</tr>
<tr>
<td>Mixture</td>
<td>Ratio of ratios</td>
<td>Mixture</td>
</tr>
<tr>
<td>1</td>
<td>0.99857</td>
<td>1</td>
</tr>
<tr>
<td>2</td>
<td>0.99852</td>
<td>2</td>
</tr>
<tr>
<td>3</td>
<td>0.99786</td>
<td>3</td>
</tr>
<tr>
<td>4</td>
<td>0.99830</td>
<td>4</td>
</tr>
<tr>
<td>5</td>
<td>0.99783</td>
<td>5</td>
</tr>
<tr>
<td></td>
<td>0.99822</td>
<td></td>
</tr>
</tbody>
</table>

Average bias for high temperature analysis = 0.99794

Table I shows the results of "high temperature" analyses of fifteen synthetically prepared uranium samples. The results are given as the ratio of the theoretical value divided by the experimental value so that an intercomparison of all fifteen results can be made without interpolation. The
heating pattern used for this type of analysis is to take the sample filaments directly up to a high temperature after the ionizing filament temperature has been set. For the sample size and pH used at the Bureau, this corresponds to a current of 2.5 A in the side filaments. The final sample filament currents are in the range of 2.6 to 2.9 A for an ion signal of about $10^{-10}$ A. The results show that the bias is constant at approximately 0.25%. This particular heating pattern is very similar to that used for routine analyses performed on the Bureau instruments equipped with D.C. amplifiers.

Table II shows the results of "low temperature" analyses of the same fifteen samples. Again the bias is constant but in this case it amounts to approximately 0.5%. The shift is brought about solely by the change in the filament heating pattern with a consequent slight change in the final temperature of the sample filament. The procedure in this case is to heat the side filaments much more slowly. After the centre filament temperature has been set, the current in the side filaments is brought up to about 1.8 A and a final current of 2.2 - 2.4 A gives the standard $10^{-10}$ A ion current. This particular heating pattern is very similar to that used in routine analyses performed on instruments equipped with ion multipliers, where it is very important that too large a signal does not strike the first dynode.

**TABLE II**

**COMPARISON OF THE CALCULATED $^{235}$U/$^{238}$U RATIO TO THE OBSERVED $^{235}$U/$^{238}$U RATIO FOR LOW TEMPERATURE ANALYSES OF SYNTHETIC URANIUM SAMPLES**

<table>
<thead>
<tr>
<th>Group 1</th>
<th>Group 2</th>
<th>Group 3</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Approximate ratio</strong></td>
<td><strong>Approximate ratio</strong></td>
<td><strong>Approximate ratio</strong></td>
</tr>
<tr>
<td>1:9</td>
<td>1:1</td>
<td>9:1</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Mixture</th>
<th>Ratio of ratios</th>
<th>Mixture</th>
<th>Ratio of ratios</th>
<th>Mixture</th>
<th>Ratio of ratios</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.99564</td>
<td>1</td>
<td>0.99584</td>
<td>1</td>
<td>0.99597</td>
</tr>
<tr>
<td>2</td>
<td>0.99523</td>
<td>2</td>
<td>0.99476</td>
<td>2</td>
<td>0.99590</td>
</tr>
<tr>
<td>3</td>
<td>0.99574</td>
<td>3</td>
<td>0.99521</td>
<td>3</td>
<td>0.99619</td>
</tr>
<tr>
<td>4</td>
<td>0.99468</td>
<td>4</td>
<td>0.99567</td>
<td>4</td>
<td>0.99586</td>
</tr>
<tr>
<td>5</td>
<td>0.99520</td>
<td>5</td>
<td>0.99563</td>
<td>5</td>
<td>0.99608</td>
</tr>
<tr>
<td></td>
<td>0.99534</td>
<td></td>
<td>0.99542</td>
<td></td>
<td>0.99600</td>
</tr>
</tbody>
</table>

Average bias for low temperature analysis = 0.99562

Table III shows the results of ten consecutive analyses in which the heating patterns were alternated. The table clearly demonstrates that the results obtained with each heating pattern are distinct and reproducible. In essence, these and other experiments at the National Bureau of Standards show that the analyst can control the level of bias over about a 1% range.
TABLE III

ALTERNATE HIGH TEMPERATURE AND LOW TEMPERATURE ANALYSES OF SYNTHETIC 9:1 URANIUM SAMPLES

<table>
<thead>
<tr>
<th>Sample number</th>
<th>Observed (^{235}\text{U}/^{238}\text{U}) pattern</th>
<th>Analytical (^{235}\text{U}/^{238}\text{U}) pattern</th>
<th>Corrected (^{235}\text{U}/^{238}\text{U})</th>
<th>Calculated (^{235}\text{U}/^{238}\text{U})</th>
<th>Calc. (^{235}\text{U}/^{238}\text{U}) Corr.</th>
</tr>
</thead>
<tbody>
<tr>
<td>3</td>
<td>9.08386 LT</td>
<td></td>
<td>9.044</td>
<td>9.048</td>
<td>1.0004</td>
</tr>
<tr>
<td>2</td>
<td>9.02307 HT</td>
<td></td>
<td>9.004</td>
<td>9.004</td>
<td>1.0000</td>
</tr>
<tr>
<td>1</td>
<td>9.02257 LT</td>
<td></td>
<td>8.983</td>
<td>8.979</td>
<td>0.9996</td>
</tr>
<tr>
<td>5</td>
<td>8.97484 HT</td>
<td></td>
<td>8.956</td>
<td>8.950</td>
<td>0.9993</td>
</tr>
<tr>
<td>4</td>
<td>9.05800 LT</td>
<td></td>
<td>9.018</td>
<td>9.023</td>
<td>1.0006</td>
</tr>
<tr>
<td>3</td>
<td>9.06294 HT</td>
<td></td>
<td>9.050</td>
<td>9.048</td>
<td>0.9998</td>
</tr>
<tr>
<td>2</td>
<td>9.03922 LT</td>
<td></td>
<td>9.000</td>
<td>9.004</td>
<td>1.0004</td>
</tr>
<tr>
<td>1</td>
<td>9.00090 HT</td>
<td></td>
<td>8.982</td>
<td>8.979</td>
<td>0.9997</td>
</tr>
<tr>
<td>4</td>
<td>9.06001 LT</td>
<td></td>
<td>9.020</td>
<td>9.023</td>
<td>1.0003</td>
</tr>
<tr>
<td>5</td>
<td>8.97193 HT</td>
<td></td>
<td>8.953</td>
<td>8.950</td>
<td>0.9997</td>
</tr>
</tbody>
</table>

HT -- Denotes high temperature analysis. The correction factor for HT is 0.99794
LT -- Denotes low temperature analysis. The correction factor for LT is 0.99562

The best external precision is obtained using the heating pattern which results in an average bias of about 0.25%. It is important to note that these analytical patterns can only be reproduced on instruments which are equipped with liquid nitrogen cold traps in the source regions and, even then, only on instruments which have about the same ion transmission efficiency.

Early in 1964, the National Bureau of Standards certified a plutonium isotope standard. Since the quantitative chemistry of plutonium has not been developed to the point where synthetic mixtures can be made, this standard was certified relative to uranium. The fact that plutonium reacts in exactly the same manner as uranium during mass spectrometric analysis and has exactly the same biases for equivalent heating patterns lends credence to this relative certification.

SUMMARY

At the beginning of the paper there was a reference to the correlation of the problems and analytical procedures when doing isotopic analysis by the
TABLE IV

TABLE OF "BEST ESTIMATE" VALUES

COVER LETTER

As many of you may know, the uranium isotopic standards have been in process of re-evaluation for the past two years. 235U and 238U values are being re-checked using mixtures, prepared at the National Bureau of Standards from high-purity 235U and 238U furnished by the Atomic Energy Commission, as calibration standards. The minor isotopes are being re-determined by "spiking" with high-purity 234U, also furnished by the Atomic Energy Commission. 235U values for standards U005 through U350 and 238U values for standards U750 through U930 have been obtained at both Oak Ridge and Goodyear Atomic by oxide dilution and UF6 analysis with a general agreement well within 0.1% of the amount present. As work on each standard is completed during the year, new certificates will be issued to replace the present provisional certificates, and copies furnished to those who have received samples.

The following table of "best estimate" values is not to be construed as an NBS certification, but is a tabulation of the present best estimate value for each isotope. The pooled analysis of variance on the data used to compile the table yields the following general analytical uncertainty statements: (to + limit for known sources of bias) (1) 234U and 238U, to at least 0.1% of the amount; (2) 235U to 238U ratio, to at least 0.1%; and (3) minor isotope values are self consistent to at least 0.25% of the amount present or to 1.0 ppm, whichever is larger.

<table>
<thead>
<tr>
<th>Standard</th>
<th>234U</th>
<th>235U</th>
<th>236U</th>
<th>238U</th>
<th>235U/238U</th>
</tr>
</thead>
<tbody>
<tr>
<td>U005</td>
<td>0.0022</td>
<td>0.4896</td>
<td>0.0047</td>
<td>99.5035</td>
<td>0.004920</td>
</tr>
<tr>
<td>U010</td>
<td>0.0054</td>
<td>1.0038</td>
<td>0.0068</td>
<td>98.9840</td>
<td>0.010141</td>
</tr>
<tr>
<td>U015</td>
<td>0.0085</td>
<td>1.5322</td>
<td>0.0164</td>
<td>98.4429</td>
<td>0.015564</td>
</tr>
<tr>
<td>U020</td>
<td>0.0125</td>
<td>2.0938</td>
<td>0.0165</td>
<td>97.9277</td>
<td>0.020813</td>
</tr>
<tr>
<td>U030</td>
<td>0.0190</td>
<td>3.0459</td>
<td>0.0204</td>
<td>96.9147</td>
<td>0.031429</td>
</tr>
<tr>
<td>U050</td>
<td>0.0279</td>
<td>5.0089</td>
<td>0.0480</td>
<td>94.9152</td>
<td>0.05277</td>
</tr>
<tr>
<td>U100</td>
<td>0.0676</td>
<td>10.1381</td>
<td>0.0377</td>
<td>89.7016</td>
<td>0.11363</td>
</tr>
<tr>
<td>U150</td>
<td>0.0993</td>
<td>15.3062</td>
<td>0.0659</td>
<td>84.5286</td>
<td>0.18108</td>
</tr>
<tr>
<td>U200</td>
<td>0.1248</td>
<td>20.0134</td>
<td>0.0120</td>
<td>79.6498</td>
<td>0.25127</td>
</tr>
<tr>
<td>U350</td>
<td>0.2500</td>
<td>35.1924</td>
<td>0.1678</td>
<td>64.3898</td>
<td>0.54655</td>
</tr>
<tr>
<td>U500</td>
<td>0.5183</td>
<td>49.6979</td>
<td>0.7555</td>
<td>49.7083</td>
<td>0.99979</td>
</tr>
<tr>
<td>U750</td>
<td>0.5922</td>
<td>75.3589</td>
<td>0.2497</td>
<td>23.7992</td>
<td>3.1664</td>
</tr>
<tr>
<td>U800</td>
<td>0.6564</td>
<td>80.2782</td>
<td>0.2446</td>
<td>18.8208</td>
<td>4.2654</td>
</tr>
<tr>
<td>U850</td>
<td>0.6439</td>
<td>85.1382</td>
<td>0.3707</td>
<td>13.8472</td>
<td>6.1484</td>
</tr>
<tr>
<td>U900</td>
<td>0.7779</td>
<td>90.1955</td>
<td>0.3329</td>
<td>8.6837</td>
<td>16.3748</td>
</tr>
<tr>
<td>U930</td>
<td>1.0806</td>
<td>93.3368</td>
<td>0.2027</td>
<td>5.3799</td>
<td>17.3492</td>
</tr>
</tbody>
</table>

UF6 method or solids by thermal ionization. Several obvious correlations may be made:

(1) Emission or signal intensity: An analyst operating a UF6 instrument would never adjust his emission regulator within an analysis to achieve his standard operating intensity. The average analyst doing thermal ionization
work probably has as part of his analytical procedure that he should operate over as wide a range of temperatures and ion intensity as possible to cancel out various bias effects. This effect possibly cancels out some of the bias due to the recorder but replaces it with an even larger error due to the heating pattern and temperature.

(2) Composition of sample (pH): The analyst doing the UF₆ analysis knowing that any impurity in his gas will introduce error insists that his sample and reference sample be identical in composition. The average analysis of a solid sample is done with almost complete disregard for purity or pH. A case in point might well be the shipper-receiver samples. An example of this would be the comparison of a neutral solution of uranium or plutonium and a strong acid solution. A current of 2.2 or 2.3 A on the sample filaments is very hot for a neutral solution and the data produced would probably not have a significant bias. The same temperature for an acid solution would be a "low-temperature" analysis with possibly a 0.5% bias high for the lighter isotopes.

(3) Time: The analyst doing the gas analysis has an infinite sample but must contend with sample exchange (memory), and for this reason controls time. For the solids analysis exchange is not too important but sample consumption has the same effect, causing a change in the observed ratio with time, so again time is important and data should be taken symmetrically about some fixed point of time.

The table of best estimate values for the U standards (Table IV) with its cover letter is included as a general summary of the entire effort.

DISCUSSION

C. A. BENNETT: I would like to compliment Mr. Shields on the excellence of his statistical designs. Obviously he realizes that if you tell the statistician all your troubles, the chances of getting the correct prescription are greatly increased.

I would like to ask whether the bias correction which has made possible the greatly increased precision is unique to each instrument and whether it is reasonably stable with time?

W. R. SHIELDS: The bias correction does have to be made separately for each instrument; it is directly related to the transmission efficiency, average sample size used, etc., but it remains very constant for the particular instrument.

M. J. HIGATSBERGER: I should just like to remark that with ion sources based on surface ionization there is preferential evaporation of the higher isotope. The effect is temperature-dependent and approximately proportional to the square root of mass. Further discrimination occurs with electron multipliers; here again the secondary electron emission is in first approximation proportional to the square root of the isotopic mass.
BURN-UP AND PRODUCTION

(Sessions VIII and IX)
PLUTONIUM PRODUCTION AND BURN-UP CALCULATION METHODS FOR LIGHT-WATER MODERATED REACTORS

G.A. SOFER, A.H. KAZI, J.R. TOMONTO AND P. BUCK
UNITED NUCLEAR CORPORATION,
WHITE PLAINS, N. Y., UNITED STATES OF AMERICA

Abstract — Résumé — Аннотация — Resumen

PLUTONIUM PRODUCTION AND BURN-UP CALCULATION METHODS FOR LIGHT-WATER MODERATED REACTORS. Data are presented on the production, utilization and burn-up of plutonium fuel in light-water moderated power reactors. The physics of plutonium fuel is compared with the physics of uranium-235-bearing fuel. A key problem in the analysis of plutonium-fuelled systems is the treatment of the plutonium-239 and plutonium-241 resonances at about 0.3 eV, and the plutonium-240 resonance at 1 eV. A relatively simple method has been developed at United Nuclear Corporation which satisfactorily predicts the reactivity of existing 1.5 wt. % \( {\text{PuO}_2-\text{UO}_2} \) critics. This method is based on a multigroup Amoyal and Benoist type disadvantage factor calculation, and a Wigner-Wilkins spectrum calculation, both performed up to 1.855 eV. More precise spectrum calculations are performed using the UNC-THERMOPILE Monte Carlo machine programme.

Measured burn-up data from the Hanford Plutonium Recycle Test Reactor is satisfactorily calculated using existing cross-sections and the FUEL portion of the relatively simple FUELMOVE code. A more sophisticated burn-up code in use at United Nuclear Corporation is TRANSLUX.

Parametric data are presented for natural \( \text{UO}_2 \) fuel enriched with \( \text{PuO}_2 \), which could be used in a 500-MW(e) zirconium-clad PWR. Burn-up comparable to an all-uranium system is obtainable from a \( \text{PuO}_2-\text{UO}_2 \) mixture with equal initial total fissile enrichment. With appropriate optimization, a smaller initial excess reactivity is expected for a discharge burn-up equal to an all-uranium system. The initial excess reactivity may be made lower by optimized selection of water-to-fuel ratio and rod size, which strongly influence the plutonium-240 absorption and its variation with burn-up.

METHODE DE CALCUL DE LA PRODUCTION ET DE LA COMBUSTION DU PLUTONIUM DANS LES REACTEURS MODERES A L'EAU LEGERE. Les auteurs présentent des données sur la production, l'utilisation et le combustible du plutonium dans des réacteurs de puissance modérés à l'eau légère. Ils comparèrent la physique du combustible au plutonium à celle du combustible à l'uranium-235. L'analyse des systèmes fonctionnant au plutonium pose le problème capital du traitement des résonances du plutonium-239 et du plutonium-241 à 0.3 eV environ et de celle du plutonium-240 à 1 eV. Une méthode relativement simple mise au point à l'United Nuclear Corporation permet de prédir de façon satisfaisante la réactivité d'assemblages critiques existants contenant 1.5\% en poids de \( \text{PuO}_2-\text{UO}_2 \). Cette méthode est basée sur un calcul à plusieurs groupes faisant intervenir un facteur de désavantage du type Amoyal et Benoist et un calcul du spectre de Wigner-Wilkins, tous deux possibles jusqu'à 1.855 eV. Des calculs plus précis du spectre sont réalisés au moyen du programme automatique UNC-THERMOPILE de Monte-Carlo.

Des données relatives au taux d'irradiation mesuré dans le réacteur d'essai de recyclage du plutonium de Hanford peuvent être élaborées de façon satisfaisante à l'aide des sections efficaces existantes et de la partie « combustible » du code FUELMOVE qui est relativement simple. TRANSLUX est un code plus compliqué applicable à l'irradiation; il est aussi utilisé par la United Nuclear Corporation.

Les auteurs présentent des données paramétriques concernant le combustible à \( \text{UO}_2 \) naturel enrichi avec \( \text{PuO}_2 \) qui pourrait être utilisé dans un réacteur à eau sous pression de 500 MW(e), à gaines de zirconium. Un mélangé de \( \text{PuO}_2-\text{UO}_2 \) permet d'obtenir une irradiation comparable à celle d'un système tout uranium, le taux d'enrichissement total initial en matière fissile étant le même. Grâce à une optimisation appropriée, on peut s'attendre à un moindre excédent initial de réactivité pour une irradiation au déchargement égale à celle d'un système tout uranium. On peut réduire l'excédent initial de réactivité par un choix optimal du rapport eau-combustible et de la taille des barres qui ont une forte influence sur le pouvoir absorbant du plutonium-240 et sur ses variations en fonction de l'irradiation.
МЕТОДЫ РАСЧЕТА ПРОИЗВОДСТВА И ВЫГОРАНИЯ ПЛУТОНИЯ ДЛЯ РЕАКТОРОВ С ЗАМЕДЛИТЕЛЯМИ ИЗ ОБЫКНОВЕННОЙ ВОДЫ. Представлены данные по производству, использованию и выгоранию плутониевого топлива в энергетических реакторах с замедлителями из обыкновенной воды. Физика плутониевого топлива сравнивается с физикой топлива, содержащего уран-235. Ключевой проблемой при анализе систем с плутониевым топливом является обработка резонансов плутония-239 и плутония-241 примерно при 0,3 эв и резонанса плутония-240 при 1 эв. На фирме "Юнайтед ньюклер корпорейшн" разработан относительно простой метод, который удовлетворительно предсказывает реактивность существующих критических смесей с 1,5 вес% PuO2 - UO2. Данный метод основан на расчете многогруппового расчета коэффициента помех типа Амойяла и Беноиста и на расчете спектра Вигнера-Уилкса, причем то и другое используется до 1,855 эв. Более точные расчеты спектра производятся с помощью машинной программы UNC - THERMOPILE при использовании метода Монте-Карло.

Измеренные данные выгорания в Хэнфордском испытательном реакторе с повторным использованием плутония удовлетворительно вычисляются путем использования существующих сечений и доли FUEL относительно простого кода FUELMOVE. Более сложным кодом выгорания, используемым фирмой "Юнайтед ньюклер корпорейшн", является код TRANSLUX.

Параметрические данные представлены для природного топлива UO2, обогащенного PuO2, которое может быть использовано в реакторе, охлаждаемом водой под давлением, с циркониевой оболочкой, мощностью 500 МВт (эл). Выгорание, сравнимое с полностью урановой системой, получается из смеси PuO2 - UO2 с одинаковым начальным общим обогащением делящихся веществ. С соответствующей оптимизацией меньшая начальная избыточная реактивность ожидается для разрядного выгорания, равного полностью урановой системе. Начальная избыточная реактивность может быть уменьшена оптимальным выбором отношения воды-топливо и размера стержней, что сильно влияет на поглощение плутония-240 и его изменение по мере выгорания.

МЕТОДЫ ПАРАМЕТРИЧЕСКИЕ ДЛЯ РАСЧЕТА ПРОИЗВОДСТВА И ВЫГОРАНИЯ ПЛУТОНИЯ В РЕАКТОРАХ МОДЕРИРОВАННЫХ ПОГЛОЩАЮЩЕЙ АГАДУ БЕЗОПАСНОЙ ВОДЫ. En esta memoria se presentan datos sobre la producción, el empleo y el grado de combustión de plutonio en reactivos de potenciación moderados por agua ligera. La física del combustible de plutonio se compara con la física del combustible muy alto en el 239Pu y del 238Pu alrededor de 0,3 eV y del 239Pu (1 eV). La United Nuclear Corporation ha preparado un método de medición relativa al cálculo del factor de desventaja y en un cálculo del espectro según Wigner-Wilkins, efectuados ambos hasta 1,855 eV. Se pueden hacer cálculos más precisos del espectro utilizando un programa de Monte Carlo en la calculadora UNC-THERMOPILE.

Los datos sobre el grado de combustión proporcionados por el Reactor de ensayo de Hanford de recirculación de plutonio se calculan satisfactoriamente utilizando las secciones eficaces conocidas y la parte FUEL de la clave FUELMOVE relativa al cálculo. En la United Nuclear Corporation se utiliza la clave TRANSLUX, que es más compleja, para calcular el grado de combustión.

Los autores presentan datos parámetros para el combustible de UO2 natural enriquecido con PuO2 que podría utilizarse en forma de elementos revestidos de circonio en un reactor PWR de 500 MW(e). Con una mezcla de PuO2-UO2 que presente el mismo enriquecimiento inicial total de elementos fisionables puede obtenerse un grado de combustión comparable al alcanzado con un combustible exclusivamente de uranio.

Con una optimización apropiada se prevé un exceso inicial de reactividad más pequeño para un grado de combustión final equivalente al alcanzado con un sistema exclusivamente de uranio. El exceso inicial de reactividad puede disminuirse por selección óptima de la proporción agua-combustible y tamaño del elemento que influye fuertemente en la absorción del 239Pu y en su variación con el grado de combustión.
1. INTRODUCTION

1.1. Incentive for plutonium recycle in thermal spectrum power reactors

The economics of nuclear power in the foreseeable future will depend, to an increasing degree, on the use which can be made of plutonium. The principal reason for this is growing plutonium production in power reactors. In the USA, eight to ten tons of plutonium will be available by 1972 (see Fig. 1) [1-3]. In the UK, the plutonium stock is estimated at 10 t by 1972 and 35 t by 1980 [4]. A second factor which makes plutonium utilization increasingly important in the USA is the implementation in 1969 to 1971 of the fuel private ownership law. By that time, the economics of nuclear power can no longer count on a USAEC-guaranteed plutonium buy-back price.

Indications are that plutonium can be used most economically in fast spectrum breeder reactors [5]. However, these reactors are still develop-
mental, and installation of commercial fast reactors is not expected to commence before the late 1970's or early 1980's. Therefore, it is appropriate to investigate the utilization of plutonium in thermal spectrum power reactors and thus provide at least an interim use for plutonium pending its optimum utilization in fast spectrum reactors.

In considering the use of plutonium in thermal reactors, five general types of fuel are of interest:

1. A mixture of plutonium and uranium where either depleted or natural uranium is intimately mixed with plutonium.
2. Spiked (or concentrated) plutonium fuel in a uranium reactor. In this form of utilization plutonium fuel is interspersed with uranium fuel, but each is kept segregated.
3. Batch loading. In this form of utilization an entire core region is loaded with plutonium fuel.
4. Plutonium burner reactors, or the so-called Phoenix fuel concept, represents a potential utilization of plutonium which does not involve uranium. The fuel is made entirely of plutonium. This form of utilization is not suitable for utility-type power reactors.
5. Mixtures of plutonium and thorium may be of interest eventually.

Analyses performed by Eschbach [6] and others [7, 5] indicate that the value of fissile plutonium for typical light-water recycle applications is in the range $9 to $11.50/g when uranium 90% enriched in $^{235}$U is worth $12/g.

1.2. Characteristics of plutonium recycle physics analysis

1.2.1. Plutonium isotope chains

In a uranium-fuelled reactor, plutonium is formed through neutron capture in $^{238}$U. In a plutonium-fuelled reactor, the considerably larger quantity of plutonium present makes the physics more complex than that of a uranium-fuelled reactor. Plutonium fuel contains two fissionable isotopes, $^{239}$Pu and $^{241}$Pu, and the $^{240}$Pu absorption cross-section is very much larger than that of $^{238}$U [281 as opposed to 2.7 b at 0.025 eV, R.I. (∞) of 8600 versus 268 b].

1.2.2. Plutonium low-energy resonances

Determination of reaction rates in a plutonium system is complicated by the fact that $^{239}$Pu, $^{240}$Pu, and $^{241}$Pu all have large low-lying resonances in the energy range 0.1 to 2.0 eV (see Fig. 2). A small error in the representation of neutron spectrum in the 0.1- to 2-eV resonance range could cause a large error in the reactivity characteristics of a plutonium system. Therefore, accurate representations of spectra and cross-sections in this range are necessary for the useful analysis of plutonium-fuelled systems. The smooth, nearly 1/v variance of the $^{235}$U cross-section in the thermal and epithermal region makes the spectral representation of a $^{235}$U system amenable to more approximate methods.

In systems with appreciable plutonium concentrations, it is necessary to consider thermal inelastic scattering in H$_2$O. In the usual water-moderated reactor computation, scattering from water is treated as though
water were a gas. The only concession to the fact that H$_2$O is a liquid has been to assign an effective mass to the gas. In actuality, neutrons may scatter from H$_2$O and exchange energy with the molecule in amounts corresponding to multiples of the fundamental vibrational or rotational energy levels of the molecule. These inelastic scatterings represent relatively large changes in neutron energy, and, in the presence of large energy dependent absorptions, can produce a thermal spectrum significantly different from the one which would be computed on the basis of the gas model with an effective mass. A similar, but much less significant, effect is inelastic scattering in the UO$_2$ lattice.

1.2.3. Nuclear properties of parasitic capture products

The nuclear characteristics of the parasitic capture products ($^{236}$U, $^{240}$Pu, $^{242}$Pu, $^{241}$Am) are important in evaluating the reactivity of plutonium-uranium fuels (see Table I). Because of the large $\alpha$ of $^{239}$Pu, the yield of $^{240}$Pu per gram of $^{239}$Pu destroyed is $\sim 80\%$ greater than the yield of $^{236}$U per gram of $^{235}$U destroyed. Uranium-236 is a mild parasitic absorber ($\sigma_\text{th} = 6$ b), whereas $^{240}$Pu has a high cross-section ($\sigma_\text{th} = 281$ b, R.I. = 8600 b), and forms $^{241}$Pu which, in turn, has a high fission cross-section and a large $\alpha$.

The cross-sections and ratios of cross-sections between different nuclides vary significantly with energy.
### TABLE I — THERMAL GROUP CONSTANTS OF URANIUM AND PLUTONIUM NUCLIDES (AT 2200 METERS/SEC)

<table>
<thead>
<tr>
<th>Constant</th>
<th>$^{235}$U</th>
<th>$^{238}$U</th>
<th>$^{238}$Pu</th>
<th>$^{239}$Pu</th>
<th>$^{240}$Pu</th>
<th>$^{241}$Pu</th>
<th>$^{242}$Pu</th>
</tr>
</thead>
<tbody>
<tr>
<td>Absorption cross section, barns</td>
<td>678</td>
<td>6</td>
<td>2.7</td>
<td>1015</td>
<td>281</td>
<td>1370</td>
<td>30</td>
</tr>
<tr>
<td>Fission cross section, barns</td>
<td>577</td>
<td>—</td>
<td>—</td>
<td>741</td>
<td>0.03</td>
<td>1030</td>
<td>—</td>
</tr>
<tr>
<td>Capture cross section, barns</td>
<td>101</td>
<td>6</td>
<td>2.7</td>
<td>(268)*</td>
<td>274</td>
<td>281</td>
<td>(8600)*</td>
</tr>
<tr>
<td>Ratio of capture to fission, $\alpha$</td>
<td>0.175</td>
<td>—</td>
<td>—</td>
<td>0.370</td>
<td>—</td>
<td>0.330</td>
<td>—</td>
</tr>
<tr>
<td>Neutrons per fission, $\nu$</td>
<td>2.442</td>
<td>—</td>
<td>—</td>
<td>2.898</td>
<td>—</td>
<td>3.06</td>
<td>—</td>
</tr>
<tr>
<td>Probability of fission per neutron absorbed, $1/(1+\alpha)$</td>
<td>0.851</td>
<td>—</td>
<td>—</td>
<td>0.730</td>
<td>—</td>
<td>0.752</td>
<td>—</td>
</tr>
<tr>
<td>Neutrons produced per neutrons absorbed, $\eta=\nu/(1+\alpha)$</td>
<td>2.077</td>
<td>—</td>
<td>—</td>
<td>2.115</td>
<td>—</td>
<td>2.30</td>
<td>—</td>
</tr>
<tr>
<td>Energy/fission, Mev</td>
<td>194</td>
<td>—</td>
<td>—</td>
<td>201</td>
<td>—</td>
<td>205</td>
<td>—</td>
</tr>
<tr>
<td>Energy/neutron absorbed, Mev</td>
<td>166</td>
<td>—</td>
<td>—</td>
<td>147</td>
<td>—</td>
<td>149</td>
<td>—</td>
</tr>
<tr>
<td>$^{235}$U produced/neutron absorbed, $\alpha/(1+\alpha)$</td>
<td>0.15</td>
<td>—</td>
<td>—</td>
<td>—</td>
<td>—</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td>$^{238}$Pu produced/neutron absorbed, $\alpha/(1+\alpha)$</td>
<td>—</td>
<td>—</td>
<td>—</td>
<td>0.27</td>
<td>—</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td>$^{242}$Pu produced/neutron absorbed, $\alpha/(1+\alpha)$</td>
<td>—</td>
<td>—</td>
<td>—</td>
<td>—</td>
<td>—</td>
<td>—</td>
<td>0.25</td>
</tr>
</tbody>
</table>

*Infinite dilution resonance integral

---

2. CHARACTERISTICS OF CURRENT UNITED NUCLEAR PLUTONIUM RECYCLE PHYSICS METHODS

2.1. Neutron slowing down

The principal tool for slowing down calculations is FORM [8], supplemented by HRG [9]. The effective resonance integral for $^{238}$U is calculated using the SDKR code [10], which uses a correlation based on Hellstrand's UO$_2$ data [11] and the Dancoff-Ginsburg factor treatment by Fukai [12]. Since the high-energy resonances tend to be narrow, their net absorption is low relative to low-energy resonances. As a result, when reasonable values of self-shielding factors are included for $^{238}$U resonances, the homogeneous representation is a good one. In this case, the fast flux above ~2 eV can be expected to show little spatial variation and small self-shielding except right at the resonance energy. In a FORM calculation, an iterative procedure is followed to determine a self-shielding or "L" factor for $^{238}$U or
that is consistent with the resonance integral as calculated from data such as Hellstrand's [11] or Nichols' [13]. The L factor corrects for heterogeneity and Doppler broadening. Experimental data on $^{239}\text{Pu}$ and $^{240}\text{Pu}$ resonance integrals are still inadequate. Some data are available on the $^{240}\text{Pu}$ resonance integral in PuAl-D$_2$O lattices [13]. Self-shielding factors can be calculated using theoretical models such as the RIC [14] or GROUSS [9] codes.

2.2. Neutron thermalization

The principal tool used for neutron thermalization calculations is TEMPEST [15]. With TEMPEST, one can average over a Wigner-Wilkins, Wilkins, or Maxwellian spectrum, or a combination of any two of these (e.g., Wilkins below a selected transition energy $E_T$ and Wigner-Wilkins above $E_T$). The thermal cut-off energies normally used are either 0.625 eV or 1.855 eV. The 1-eV $^{240}\text{Pu}$ resonance is thus included in either the slowing down or in the thermalization calculation.

Thermal disadvantage factors are obtained using either a one-group or multigroup Amouyal–Benoist calculation. One-group Amouyal–Benoist calculations are made with the AMBER code [10]. Multigroup Amouyal–Benoist calculations are made with the TEAM code [16] which calculates disadvantage factors at each of the 291 energy points of the TEMPEST cross-section library.

For more detailed spectrum calculations, the UNC–THERMOPILE Monte Carlo programme is being developed [17].

As noted previously (Fig. 2), both $^{239}\text{Pu}$ and $^{241}\text{Pu}$ have large resonances at about 0.3 eV. In this energy range, the spectrum changes rapidly, so that a perturbation in this region can have a large effect on reaction rates. In addition, $^{240}\text{Pu}$ has a large resonance at 1 eV. The Maxwellian spectrum, which is satisfactory for many uranium lattices which have an approximately $1/\nu$ spectrum, is not adequate to predict the spectrum in the presence of these resonances. Table II lists thermalization models of major interest. With the two methods in current use at United Nuclear (Wigner-Wilkins and Monte Carlo) it is possible to treat all problems of interest adequately.

If the cells of the reactor lattice are small, simple diffusion theory is likely to be inadequate. The problem is to represent the angular variation of the neutron flux. The various multigroup methods available can be characterized by their treatment of the angular variable. Spherical harmonic expansion is applied typically up to the $P_3$ approximation and including the double-P approximation [18, 19]. Discrete ordinate methods are most generally used in the form of the $D_{SN}$ method [20, 21]. A number of solutions have been developed to the integral transport equation [22–24] and use is made of Monte Carlo methods [17, 25, 26].

A basic difficulty in applying multigroup transport methods to cell calculations (whether uranium- or plutonium-fuelled) is the cylindrical cell approximation. The boundary conditions presently are not properly treated in a numerical cylindrical transport cell calculation. This leads to an error in the thermal utilization [29].
### TABLE II – THERMALIZATION CODES

<table>
<thead>
<tr>
<th>Model Name</th>
<th>Type</th>
<th>Code Name</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Maxwellian</td>
<td>1/v spectrum</td>
<td>TEMPEST* (SOFOCATE)</td>
<td>[15]</td>
</tr>
<tr>
<td>Wilkins</td>
<td>Heavy sphere moderation</td>
<td>TEMPEST* (SOFOCATE)</td>
<td></td>
</tr>
<tr>
<td>Wigner-Wilkins</td>
<td>Unit sphere moderation</td>
<td>TEMPEST* (SOFOCATE)</td>
<td></td>
</tr>
<tr>
<td>Nelkin</td>
<td>$H_2O$ inelastic scatter-</td>
<td>THERMOS – BNL</td>
<td>[22]</td>
</tr>
<tr>
<td></td>
<td>tering, space energy separability</td>
<td>THERMOS – ANL</td>
<td>[27]</td>
</tr>
<tr>
<td>Modified Nelkin</td>
<td>$H_2O$ and $UO_2$ inelas-</td>
<td>EPITHERMOS</td>
<td>[28]</td>
</tr>
<tr>
<td></td>
<td>tic scattering, space energy separability</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Nelkin</td>
<td>$H_2O$ inelastic scatter-</td>
<td>SPECTRUM V</td>
<td>[23]</td>
</tr>
<tr>
<td></td>
<td>tering</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Monte Carlo</td>
<td>Potentially exact</td>
<td>MARC</td>
<td>[26]</td>
</tr>
<tr>
<td></td>
<td></td>
<td>DRAM</td>
<td>[25]</td>
</tr>
<tr>
<td></td>
<td></td>
<td>THERMOPILE*</td>
<td>[17]</td>
</tr>
</tbody>
</table>

*In use at United Nuclear

One approach of practical utility is the so-called "white" boundary condition used in the $DS_N$ method. The outward rays of the angular fluxes are treated correctly, but the backscatter into the cell is treated isotropically [30].

2.3. Nuclide depletion

Routine survey burn-up calculations are made using the FUELMOVE [31] or CANDLE code [32]. More detailed burn-up calculations are made using the TRANSLUX code [33]. TRANSLUX combines standard depletion analysis and one-dimensional $DS_N$ transport theory (DTF) [21].

This code permits the use of a multigroup treatment in the near-thermal plutonium resonance region using the $DS_N$ approximation. Running time comparable to diffusion theory calculation can be obtained with a $DS_2$ treatment.

The depletion calculation in a plutonium-fuelled core does not present any difficulties once the slowing-down and thermalization problems are solved. If spectrum changes are significant during burn-up, spectrum
weighted cross-sections are re-evaluated at all or at selected burn-up time steps.

3. CALCULATION OF HANFORD PuO$_2$-UO$_2$ CRICALS

3.1. Results of PuO$_2$-UO$_2$ critical analyses from several laboratories

A fair amount of critical and approach-to-critical data has been generated at Hanford in support of the Plutonium Recycle Program [9, 34]. Of principal interest to plutonium recycle are the PuO$_2$-UO$_2$ criticals. The physical characteristics of these 1.5 wt. % PuO$_2$ lattices are summarized in Table III. The hexagonal pitch of the lattices studied ranges from 0.55 to 0.93 in. corresponding to a range in H/Pu atom ratio of 230 to 1169.

**TABLE III – PHYSICAL CHARACTERISTICS OF HANFORD PuO$_2$-UO$_2$ CRICALS [34]**

<table>
<thead>
<tr>
<th>Fuel Rod OD, in.</th>
<th>0.426</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fuel OD, in.</td>
<td>0.372</td>
</tr>
<tr>
<td>Clad Thickness (Zr-2), in.</td>
<td>0.027</td>
</tr>
<tr>
<td>Fuel Height, in.</td>
<td>48.5</td>
</tr>
<tr>
<td>PuO$_2$ in UO$_2$, w/o</td>
<td>1.5</td>
</tr>
<tr>
<td>U$^{235}$ (depleted) in UO$_2$, w/o</td>
<td>0.22</td>
</tr>
<tr>
<td>Pu in PuO$_2$, w/o</td>
<td></td>
</tr>
<tr>
<td>Pu$^{239}$</td>
<td>91.41</td>
</tr>
<tr>
<td>Pu$^{240}$</td>
<td>7.83</td>
</tr>
<tr>
<td>Pu$^{241}$</td>
<td>0.73</td>
</tr>
<tr>
<td>Pu$^{242}$</td>
<td>0.03</td>
</tr>
<tr>
<td>Weight of PuO$_2$-UO$_2$ per rod, g</td>
<td>826.6</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Lattice Spacing in.</th>
<th>(a)</th>
<th>(b)</th>
<th>(c)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.55</td>
<td>0.71</td>
<td>0.90</td>
<td></td>
</tr>
<tr>
<td>H/Pu atom ratio</td>
<td>230</td>
<td>567</td>
<td>1077</td>
</tr>
<tr>
<td>Number of Rods per Criticality</td>
<td>1478</td>
<td>484</td>
<td>452</td>
</tr>
<tr>
<td>Experimental Geometric Buckling, m$^{-2}$</td>
<td>48.0</td>
<td>78.5</td>
<td>60.9</td>
</tr>
<tr>
<td>Approximate Diameter of Critical Lattice, in.</td>
<td>22.4</td>
<td>16.4</td>
<td>20.4</td>
</tr>
</tbody>
</table>

Calculations of $k_{eff}$ performed for these criticals at UNC, WAPD, and ANL are compared in Fig. 3. In the UNC-I calculations, the non-thermal spectrum is calculated using FORM and $^{238}$U and $^{240}$Pu L-factors based on Hellstrand's and Nichols' correlations respectively. The thermal spectrum to 0.625 eV is determined using a Wigner-Wilkins TEMPEST calculation.
Disadvantage factors are determined at each TEMPEST energy point using the method of Amouyal and Benoist (TEAM code) [33]. The maximum to minimum range in $k_{\text{eff}}$ is 1.5%. In the UNC-II calculations, the thermal spectrum to 1.855 eV is determined from a Wigner–Wilkins TEMPEST calculation, using TEAM multigroup disadvantage factors. The $^{240}\text{Pu}$ resonance at 1 eV is thus treated in the thermal spectrum calculation. The non-thermal spectrum above 1.855 eV is determined using FORM with the $^{238}\text{U}$ L-factor determined from the Hellstrand correlation, and $L$ for $^{239}\text{Pu}$ and $^{240}\text{Pu}$ is taken to be equal to unity. The $^{239}\text{Pu}$ thermal cross-sections used in the UNC calculations are a compromise between the Leonard and Sher data ($\eta = 2.118$, $\nu = 2.886$, $\alpha = 0.363$ at 2200 m). The maximum to minimum range in $k_{\text{eff}}$ is 0.9%.

The WAPD calculations [35] make use of a modified MUFT-SOFOCATE model. The non-thermal spectrum (>0.625 eV) is computed using MUFT-IV with appropriate L-factors. The $^{240}\text{Pu}$ L-factor used by WAPD is within $\sim$1% of the $^{238}\text{U}$ L-factor based on Hellstrand's correlation. The thermal spectrum is determined by a Wigner–Wilkins SOFOCATE calculation. The two WAPD calculations shown in Fig. 3 illustrate the effect of using the Leonard cross-sections [36] and a cross-section set consistent with the data compiled by Sher [37]. The maximum to minimum range in $k_{\text{eff}}$ for the Sher data is 1.25%.

The ANL calculations are based on a GAM-1 and SOFOCATE model [38]. The $^{239}\text{Pu}$ cross-sections were adjusted to normalize the results to a $k_{\text{eff}}$ of 1.001 for the 0.90-in. lattice ($\alpha = 0.383$). The maximum to minimum range in $k_{\text{eff}}$ is 0.8%.
The results of Fig. 3 show that light-water PuO$_2$-UO$_2$ critical data can be predicted to within $<1\% \Delta k/k$ using existing cross-section data and the relatively simple Wigner-Wilkins model. At least in the present case it is not necessary to go to a space-energy separable thermalization model such as THERMOS [22, 27]. In analyses on the PuO$_2$-UO$_2$ criticals performed at WAPD [39], an average percentage difference of 0.1% in $\eta$ was obtained between THERMOS and a SOFOCATE Wigner-Wilkins calculation, using identical cross-sections.

3.2. Effect of neutron thermalization model on calculated reactivity of PuO$_2$-UO$_2$ criticals

A number of $k_{\text{eff}}$ calculations using several thermalization models are summarized in Table IV. The Wigner-Wilkins model gives the closest agreement with experiment. The Wilkins and combined Wilkins and Wigner-Wilkins models yield somewhat poorer agreement. The Maxwellian model is entirely unsatisfactory. A number of spectrum calculations for the 0.55-in. lattice are compared in Fig. 4. The Maxwellian flux is almost negligible in the plutonium resonance regions, and, hence, the Maxwellian model grossly underpredicts absorptions in $^{239}$Pu and $^{240}$Pu. This leads to a large overprediction in reactivity when the $^{240}$Pu resonance is included in the thermal group, especially for relatively undermoderated lattices (Table IV, cases with 1.855 eV thermal cut-off). Table V compares flux-weighted cross-sections for the 0.55 and 0.90-in. lattices using the Maxwellian and Wigner-Wilkins models. The effective $^{240}$Pu absorption cross-section to 1.855 eV for the 0.55-in. lattice is 842.5 b using the Wigner-Wilkins model, and only 229.8 b with the Maxwellian model. With a thermal cut-off of 0.625 eV, the Maxwellian model underpredicts $k_{\text{eff}}$ (see Table IV). In this case, it is seen from Table V that while the Maxwellian model underpredicts absorption in the $^{238}$Pu resonance, it overpredicts smooth thermal absorption of $^{235}$U, $^{240}$Pu, and $^{241}$Pu. The above effects are considerably less for the 0.90-in. lattice since, in this case, the flux at the 0.3 eV resonance is only $\sim50\%$ of what it is for the 0.55-in. lattice (see Fig. 4).

From Table IV, the multigroup disadvantage factor treatment of the $^{239}$Pu resonance is seen to reduce $k_{\text{eff}}$ by 0.6 to 1.2% (AMBER versus TEAM, 0.625 eV cut-off). The spectra in Fig. 4 show that the use of multigroup disadvantage factors reduces the flux depression in the resonance regions. As is seen from the data of Table V, the self-shielding factor $"g"$ in the resonance regions is small, thus decreasing absorptions and increasing the flux over the one-group disadvantage factor treatment. In the case of the $^{239}$Pu resonance, this effect decreases the calculated reactivity.

The heavy-gas Wilkins model does not adequately account for the flux depression at the resonance energies. The existence of these flux depressions is verified by the recent spectrum measurements on the Zenith plutonium core No. 7 [4].

The calculated $k_{\text{eff}}$ is quite sensitive to the value of the $^{240}$Pu resonance integral, particularly for the tight lattice where a significant neutron density is found in the region of $^{240}$Pu resonance (see Fig. 5).
<table>
<thead>
<tr>
<th>Pitch, in.</th>
<th>AMBER (a)</th>
<th>TEAM (c)</th>
<th>TEAM</th>
<th>TEAM</th>
<th>TEAM</th>
<th>TEAM</th>
<th>Thermal Cutoff of 0.625 ev</th>
<th>Thermal Cutoff of 1.855 ev</th>
<th>Pu\textsuperscript{238}</th>
<th>Pu\textsuperscript{240}</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.55</td>
<td>-1.5</td>
<td>-1.4</td>
<td>-2.0</td>
<td>-2.1</td>
<td>-1.8</td>
<td>-0.1</td>
<td>-1.9</td>
<td>-1.5</td>
<td>18.5</td>
<td>4005</td>
</tr>
<tr>
<td>0.71</td>
<td>+1.2</td>
<td>0.0</td>
<td>+0.2</td>
<td>+0.2</td>
<td>-0.7</td>
<td>+0.7</td>
<td>+0.5</td>
<td>+0.1</td>
<td>20.5</td>
<td>4288</td>
</tr>
<tr>
<td>0.90</td>
<td>+2.3</td>
<td>+0.1</td>
<td>+0.4</td>
<td>+1.1</td>
<td>-1.2</td>
<td>+0.8</td>
<td>+0.9</td>
<td>+0.7</td>
<td>21.5</td>
<td>4407</td>
</tr>
</tbody>
</table>

(a) One-group Amouyal-Benoist disadvantage factor.
(b) Wilkins (W) spectrum below 0.1 ev and Wigner-Wilkins (WW) above 0.1 ev.
(c) Multigroup Amouyal-Benoist disadvantage factors.
(d) Based on Hellstrand's correlation – L U\textsuperscript{238} of 0.626, 0.504, and 0.390 for 0.55, 0.71, and 0.90 lattices respectively.
(e) Based on Nichols' correlation – L Pu\textsuperscript{240} of 0.582, 0.570, and 0.568 for 0.55, 0.71, and 0.90 lattices respectively when cutoff is 0.625 ev. When cutoff is 1.855 ev, L Pu\textsuperscript{240} equals 1.0.
4. PLUTONIUM RECYCLE TEST REACTOR BURN-UP DATA ANALYSIS

Burn-up experiments have been conducted on plutonium-aluminium alloy fuel elements irradiated in Hanford's Plutonium Recycle Test Reactor [9, 40]. The data obtained indicate that the isotopic content as a function of burn-up is essentially independent of the location of the sample within the fuel cluster. Since the fuel clusters were widely separated, most of the neutron thermalization took place in the D$_2$O moderator region. These conditions make the experiments ideal for testing effective plutonium cross-sections without the complexity of the thermalization effects in closely packed lattices.

The reported burn-up data have been analysed at United Nuclear using the zero-dimensional FUEL burn-up option of the FUELMOVE code [31]. In order to obtain appropriate flux-weighted number densities, local thermal flux disadvantage factors for individual fuel rods were calculated using the Amouyal-Benoist method. Separate FORM-TEMPEST calculations were performed to generate few-group constants for the fuel element cluster and the surrounding moderator region. These constants were used in a two-group, two-region diffusion calculation (AIM-6 code) to determine flux-averaged homogenized number densities for materials in the PRTR supercell.
TABLE V — FLUX-WEIGHTED EFFECTIVE MICROSCOPIC CROSS SECTIONS FOR TWO MIXED OXIDE PLUTONIUM LATTICES

\( g = \text{self-shielding factor, } \frac{\phi_F}{\phi_{\text{cell}}} \)

<table>
<thead>
<tr>
<th>Lattice Pitch 0.55 in., H/Pu Atom Ratio 230</th>
<th>Lattice Pitch 0.90 in., H/Pu Atom Ratio 1077</th>
</tr>
</thead>
<tbody>
<tr>
<td>Wigner-Wilkins Flux Spectrum</td>
<td>Maxwellian Flux Spectrum</td>
</tr>
<tr>
<td>0 to 0.625 ev</td>
<td>0 to 1.855 ev</td>
</tr>
<tr>
<td>0.625 ev</td>
<td>1.855 ev</td>
</tr>
</tbody>
</table>

| \( \bar{g}_f^{\alpha} (U^{235}) \) | 405.8 | 341.6 | 528.5 | 528.3 | 450.6 | 427.5 | 491.2 | 491.2 |
| \( \bar{g}_f^{\alpha} (U^{239}) \) | 346.0 | 291.1 | 451.8 | 451.8 | 384.9 | 365.1 | 419.9 | 419.9 |
| \( \bar{a}_\text{eff} (U^{235}) \) | 0.173 | 0.173 | 0.170 | 0.170 | 0.171 | 0.171 | 0.170 | 0.170 |
| \( \bar{g}_f^{\alpha} (Pu^{239}) \) | 939.0 | 776.5 | 862.7 | 862.7 | 831.5 | 786.1 | 804.6 | 804.6 |
| \( \bar{g}_f^{\alpha} (Pu^{240}) \) | 642.2 | 531.1 | 625.7 | 625.7 | 589.4 | 557.4 | 583.1 | 583.1 |
| \( \bar{a}_\text{eff} (Pu^{239}) \) | 0.462 | 0.462 | 0.379 | 0.379 | 0.411 | 0.410 | 0.380 | 0.380 |
| \( \bar{g}_f^{\alpha} (Pu^{240}) \) | 203.6 | 842.5 | 229.8 | 229.8 | 204.8 | 412.5 | 214.1 | 214.1 |
| \( \bar{g}_f^{\alpha} (Pu^{241}) \) | 1023.3 | 842.3 | 1192.7 | 1192.7 | 1056.5 | 996.9 | 1109.7 | 1109.7 |
| \( \bar{g}_f^{\alpha} (Pu^{241}) \) | 778.0 | 641.3 | 912.5 | 912.5 | 807.0 | 761.8 | 849.4 | 849.4 |
| \( \bar{a}_\text{eff} (Pu^{241}) \) | 0.315 | 0.313 | 0.307 | 0.307 | 0.309 | 0.309 | 0.306 | 0.306 |
| \( \bar{g}_f \) | 0.914 | 0.923 | 0.929 | 0.929 | 0.868 | 0.872 | 0.874 | 0.874 |

\( (g_f)_{0.3 \text{ ev}} \) | 0.663 | 0.542 |
\( (g_f)_{1.06 \text{ ev}} \) | 0.400 | 0.291 |
The supercell average number densities were then used in FORM-TEMPEST calculations to generate input constants for the FUEL code. The thermal flux disadvantage factors thus calculated for the 1.82 wt. % Pu case are 1.042 for the local fuel pin and 1.326 for the supercell, yielding an over-all disadvantage factor of 1.38. Using a simpler homogenization method, an over-all disadvantage factor of 1.68 was obtained. The two methods predicted isotopic concentrations within 1% of each other. Agreement with experiment was quite good and insensitive to the homogenization method. Two different cross-section sets were used. FUEL III utilizes a set very close to the data of Sher [37], while FUEL IV uses the data of Leonard [36]. A slightly better agreement is obtained with the Leonard data. The FUELMOVE code calculates thermal cross-sections (0 to 0.625 eV) based on 2200 m cross-sections using 1/v energy dependence modified by a Breit-Wigner single-level resonance treatment. These energy dependent cross-sections are flux-weighted using a Wilkins thermalization spectrum to obtain effective thermal cross-sections.

The effective resonance cross-sections are obtained by a resonance escape probability formulation using effective resonance integrals derived from infinite dilution resonance integral data (BNL-325). The resonance reaction rates are normalized such that the effective resonance cross-sections, when multiplied by the thermal energy flux, yield the reaction rates of all energies above 0.625.

Results obtained for another set of PRTR fuel clusters [40], containing 2.68 wt. % Pu in aluminium and 16.4 at. % $^{240}$Pu initially, are shown in Fig.6.
Again, agreement of calculation with experiment is good, indicating proper relationship between capture and fission cross-sections of individual plutonium isotopes, and between cross-sections of different isotopes. The variation of effective one-group cross-sections for key isotopes as a function of burn-up is shown in Fig. 7 for the 2.68 wt. % Pu set of burn-up data.
5. PLUTONIUM BURN-UP IN LARGE PWR's

Although the necessity for plutonium recycle in thermal power reactors is clear, there also are certain advantages in the use of plutonium in such reactors which provide special design and operational benefits. Foremost among these advantages is the behaviour of the plutonium isotope chain. Bred plutonium always contains some $^{240}\text{Pu}$ formed from $^{239}\text{Pu}$ by neutron capture. Since $^{240}\text{Pu}$ has a high absorption cross-section, any of it present at the beginning of core life will act as a poison, reducing reactivity and later will add reactivity as $^{241}\text{Pu}$ is formed. In addition, the shift in spectrum away from the 1-eV $^{240}\text{Pu}$ resonance as depletion progresses reduces the effective absorption in $^{240}\text{Pu}$ and tends to slow down the reactivity decrease with burn-up. These effects combine to reduce the amount of control required at beginning of core life to yield a given core burn-up.
To investigate the effects of plutonium recycle in thermal power reactors, reactivity and burn-up calculations have been performed at United Nuclear for natural UO$_2$ fuel enriched with PuO$_2$ which could be used in a 500 MW(e), Zr–clad PWR. A total fissile enrichment \( \left( \frac{^{235}U+^{239}Pu+^{241}Pu}{U+Pu} \right) \) of 3.0% was used, corresponding to the $^{235}$U enrichment required to produce 24 000 to 25 000 MWd/t in an equilibrium cycle Zr–clad PWR. Other characteristics assumed are summarized in Table VI.

**TABLE VI — DESIGN CHARACTERISTICS FOR 3% Pu REACTOR STUDIES**

<table>
<thead>
<tr>
<th>Characteristic</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Rod Diameter to Clad Thickness Ratio</td>
<td>20.0</td>
</tr>
<tr>
<td>Amount of Stainless Steel Structure, v/o</td>
<td>2.0</td>
</tr>
<tr>
<td>Density of (PuU)$_2$O$_2$ Fuel, g/cm$^3$</td>
<td>9.82</td>
</tr>
<tr>
<td>Water Temperature, °F</td>
<td>566</td>
</tr>
<tr>
<td>Water Density, g/cc</td>
<td>0.726</td>
</tr>
<tr>
<td>Type of Fuel</td>
<td>Vibratory compacted</td>
</tr>
<tr>
<td>Type of Lattice Geometry</td>
<td>Square</td>
</tr>
<tr>
<td>Fraction of U$^{235}$ in Fuel, %</td>
<td>0.7</td>
</tr>
<tr>
<td>Fraction of Fissile Pu in Fuel, %</td>
<td>2.3</td>
</tr>
</tbody>
</table>

The isotopic composition of the plutonium was taken to be: 59.5% $^{239}$Pu, 22.5% $^{240}$Pu, 13.6% $^{241}$Pu, and 4.4% $^{242}$Pu. These values are typical for fuel discharged after ~ 24 000 MWd/t U from a Zr–clad PWR. The parameters varied in this study were water-to-fuel ratio and rod diameter. Three rod sizes, 0.3, 0.45, and 0.6-in. diameter were studied at six water-to-fuel ratios from 1.25 to 2.5. The $k_{\infty}$ at beginning of life for the 18 combinations of rod size and water-to-fuel ratio were calculated with the analytical procedure used for the Hanford criticals (see section 3). These beginning of life $k_{\infty}$ values are plotted in Fig. 8 as a function of rod diameter with water-to-fuel ratio as a parameter.

From Fig. 8, the cases having sufficient reactivity at beginning of life were selected for burn-up calculations with the FUEL option of the FUELMOVE code [31]. The $^{240}$Pu resonance integral was self-shielded in the FUELMOVE code according to Nichols' correlation [13].

Figure 9 shows the variation of $k_{\infty}$ versus burn-up for three rod sizes at a water-to-fuel volume ratio of 2.0 along with a curve for a 3% enriched, Zr–clad $^{235}$U core for comparison. Note that beginning of life $k_{\infty}$ calculated by FUELMOVE is different from those shown in Fig. 8 which were calculated by FORM-TEMPEST. It is apparent that the plutonium cores can achieve burn-ups comparable to those obtainable with the reference 3% enriched uranium core. The PuO$_2$–UO$_2$ fuel appears to be capable of achieving such burn-ups with less reactivity control required at beginning of life. Figure 9 also shows the effect of rod diameter on $k_{\infty}$. At the beginning of life, the $^{240}$Pu resonance (and also the $^{238}$U resonances) is more self-shielded in the larger rod sizes, and consequently, $k_{\infty}$ is higher. As burn-up progresses, the $^{240}$Pu burns out more slowly in the larger rods, as shown...
in Fig. 10, and consequently $k_\infty$ decreases faster than in the small rods. All three rod sizes at $V_W/V_F = 2.0$ yield essentially the same burn-up when $k_\infty$ drops to 1.00. The figure shows that the smaller initial control requirement for a given burn-up is obtained with the smallest rods.

The effect of rod size on key nuclear parameters at beginning of life is shown in Table VII. Of particular interest is the variation in the effective cross-sections of plutonium isotopes, $\bar{\sigma}$, with rod size compared with $^{235}$U and $^{238}$U. It will be noted that $\bar{\sigma}^{239}$ and $\bar{\sigma}^{241}$ are substantially higher than $\bar{\sigma}^{235}$ for this case, and that the effect of rod size does not influence $\bar{\sigma}^{239}$ appreciably.

The variations of effective one-group cross-sections with flux time are shown in Fig. 11. This figure clearly indicates the effect of depletion on $^{240}$Pu absorption cross-section. The FUELMOVE cross-sections given are the sum of thermal and effective epithermal cross-sections, which, when multiplied by the thermal flux ($0 - 0.625$ eV), account for reaction rates over all neutron energies. They are not directly comparable with the TEAM-TEMPEST cross-sections given in Table VII.
FIG. 9. $k_{\infty}$ versus burn-up for different rod sizes (3% enrichment, PuO$_2$-UO$_2$ fuel compared with UO$_2$ fuel) - $V_W/V_F = 2.0$

FIG. 10. Fuel atom number densities as function of flux time (PuO$_2$-UO$_2$ 3% enriched fuel)
<table>
<thead>
<tr>
<th></th>
<th>Case 1, OD = 0.3 in.</th>
<th>Case 2, OD = 0.45 in.</th>
<th>Case 3, OD = 0.6 in.</th>
</tr>
</thead>
<tbody>
<tr>
<td>D</td>
<td>0.3868</td>
<td>0.37595</td>
<td>0.36681</td>
</tr>
<tr>
<td>$\Sigma_a$</td>
<td>0.15712</td>
<td>0.14510</td>
<td>0.13494</td>
</tr>
<tr>
<td>$\nu\Sigma_f$</td>
<td>0.23244</td>
<td>0.21705</td>
<td>0.20317</td>
</tr>
<tr>
<td>$\eta_f$</td>
<td>1.4794</td>
<td>1.4959</td>
<td>1.5056</td>
</tr>
<tr>
<td>$\bar{\sigma}_{235}^{235}$</td>
<td>213.2</td>
<td>202.8</td>
<td>193.0</td>
</tr>
<tr>
<td>$\bar{\sigma}_{236}^{235}$</td>
<td>180.6</td>
<td>171.9</td>
<td>163.6</td>
</tr>
<tr>
<td>$\bar{\sigma}_{236}^{235}$</td>
<td>0.1805</td>
<td>0.1792</td>
<td>0.1797</td>
</tr>
<tr>
<td>$\bar{\sigma}_{238}^{235}$</td>
<td>1.02</td>
<td>0.97</td>
<td>0.92</td>
</tr>
<tr>
<td>$\bar{\sigma}_{239}^{233}$</td>
<td>738.6</td>
<td>683.9</td>
<td>635.4</td>
</tr>
<tr>
<td>$\bar{\sigma}_{239}^{235}$</td>
<td>479.3</td>
<td>445.0</td>
<td>414.3</td>
</tr>
<tr>
<td>$\bar{\sigma}_{233}^{233}$</td>
<td>0.5410</td>
<td>0.5369</td>
<td>0.5337</td>
</tr>
<tr>
<td>$\bar{\sigma}_{240}^{238}$</td>
<td>507.1</td>
<td>426.6</td>
<td>368.0</td>
</tr>
<tr>
<td>$\bar{\sigma}_{241}^{239}$</td>
<td>647.1</td>
<td>612.1</td>
<td>579.4</td>
</tr>
<tr>
<td>$\bar{\sigma}_{241}^{239}$</td>
<td>489.9</td>
<td>464.0</td>
<td>439.7</td>
</tr>
<tr>
<td>$\bar{\sigma}_{242}^{241}$</td>
<td>0.3209</td>
<td>0.3192</td>
<td>0.3177</td>
</tr>
<tr>
<td>$\bar{\sigma}_{242}^{242}$</td>
<td>8.39</td>
<td>7.89</td>
<td>7.44</td>
</tr>
<tr>
<td>$\bar{\sigma} (\bar{\sigma}<em>F / \bar{\sigma}</em>{cell})$</td>
<td>0.8637</td>
<td>0.8010</td>
<td>0.7638</td>
</tr>
</tbody>
</table>
The effect of water-to-fuel ratio on the variation of $k_e$ with burn-up is shown in Fig. 12. Note that a lower excess reactivity at beginning of life is required to yield a given burn-up with a lower water-to-fuel ratio. This effect is due partially to an increased conversion ratio of $^{238}$U to $^{239}$Pu, and partially to the increased effect of spectrum shift during burn-up on $^{240}$Pu absorption, at lower water-to-fuel ratios.

It is concluded from the above that burn-up comparable to an all-uranium system is obtainable from a PuO$_2$-UO$_2$ mixture with equal initial total fissile enrichment. With appropriate optimization a smaller initial excess reactivity is expected for a discharge burn-up equal to an all-uranium system. The initial excess reactivity may be made lower by optimized selection of water-to-fuel ratio and rod size which strongly influence the $^{240}$Pu absorption and its variation with burn-up.
FIG. 12. $k_{\infty}$ versus burn-up for different $V_{H_2O}/V_{Fuel}$ ratios (3% enriched PuO$_2$-UO$_2$ fuel)

REFERENCES

W. J. WRIGHT: In the introduction to this paper, you state that the Phoenix fuel concept is not suitable for utility-type power reactors. Could you elaborate on this?
A. H. KAZI: I was thinking of utility-type power reactors in the 500-1000-MW(e) range. As it stands, the Phoenix concept involves very high power densities and very high fuel inventories. A system of this sort could be used for economic utility-type reactors but it would not be very suitable. The Phoenix concept is mainly of interest for small or medium-sized power applications such as power units for maritime or space propulsion, where very long core lifetimes are required.
INVESTIGATION OF METHODS OF CALCULATING HEAVY-ISOTOPE CONTENT OF OPERATING FUEL ELEMENTS

M.F. VALERINO AND Z.R. ROSZTOCZY
COMBUSTION ENGINEERING, INC., WINDSOR, CONN., UNITED STATES OF AMERICA

Abstract — Résumé — Аннотация — Resumen

INVESTIGATION OF METHODS OF CALCULATING HEAVY-ISOTOPE CONTENT OF OPERATING FUEL ELEMENTS. A review is given of the calculational techniques used for predicting heavy-isotope content of fuel elements. The practical capabilities and limitations of the particular methods used are discussed. In particular, attention is directed to those calculations performed for purposes of fuel accountability.

Calculation of heavy-isotope content for fuel accounting represents a simpler problem than for predicting reactor characteristics at later stages of core life. Several factors are responsible, namely that, for the purpose of fuel accounting, (1) control-rod positions and fuel-management procedures actually used are known, (2) detailed spatial variations of heavy-isotope concentrations do not have to be explicitly defined but, instead, only average values for a fuel assembly or a group of assemblies are required, and (3) other input data may be available, as for example the results of in-core flux-wire measurements and/or of fuel-assembly activity measurements. As a result, much larger mesh spacings can be employed in the geometrical representations of the 3-dimensional spatial variations. An assessment is made of the accuracy associated with the use of such simplifications in various geometrical representations of several power reactor types. In-core instrumentation and/or fuel-assembly gamma scans used in several operating power reactors have proved susceptible to reasonably accurate interpretation in terms of the relative elementwise power output. This suggests that relatively simple methods of computing burn-up and production can be evolved based on such measurement results; the possibilities of this approach are explored.
Расчет содержания тяжелых радиоизотопов для учета ядерного топлива является более простой проблемой, чем заблаговременное определение характеристик реактора на дальнейших стадиях работы его активной зоны. Это объясняется целым рядом факторов, а именно тем, что для учета ядерного топлива имеются благоприятные предпосылки: 1) позиции регулирующих стержней и фактически применяемые процедуры учета топлива известны, 2) нет необходимости точного определения детальных пространственных изменений концентраций тяжелых радиоизотопов, а вместо этого требуется только знание средних величин для топливных сборок или групп сборок и 3) могут быть другие исходные данные, как, например, результаты измерений потока внутри активной зоны при помощи проволочных приборов или измерения активности топливных сборок. Вследствие этого допустимо применение значительно более крупной сетки для графического представления трехмерных пространственных изменений. Дается оценка увеличения погрешности в результате допущения таких упрощений для различных графических представлений реакторов различных типов. Установленные в активной зоне приборы или же гамма-сканирование топливных сборок, используемые на целом ряде действующих энергетических реакторов, или и те и другие, оказались способными давать достаточное точное представление об относительном выходе мощности от каждого тепловыделяющего элемента. Это говорит о возможности выработки на основании результатов таких измерений относительно простых методов расчета выгорания и энерговыделения; исследуются возможности такого подхода.
operating data. These differences are extremely difficult to identify; and, when identified, their overall effect on prediction results is not easily assessed.

Fuel depletion calculations are generally performed during reactor design primarily for the purpose of: (1) determining that sufficient excess reactivity is available; and, (2) verifying that the permissible hot-spot factors are not exceeded. The information obtained on isotopic composition is considered to be of secondary importance, inasmuch as it generally turns out that actual reactor operation is different from that conceived. Calculations for fuel accounting purposes can best be performed during and following reactor operation where advantage can be taken of the additional data accrued, such as actual control-rod positions, total energy production, measured flux and power distributions, etc.

This paper presents the results of work performed for the US Atomic Energy Commission under Contract AT(38-1)-207, Task Agreement No. VII. The investigation is aimed at exploring the feasibility of developing simple methods for testing the results of nuclear loss and production calculations in power reactors, using the maximum amount of actual reactor operating data as input. The problem of isotopic-content determination divides into two general areas. One area involves the determination of the exposure level of the individual fuel assemblies. The second area is the determination of the detailed burn-up and production of the heavy isotopes as a function of exposure level. Estimations of accuracy of predictions in these two general areas can best be made by comparison of available calculational and experimental results. A significant number of experimental measurements have been made to serve as suitable bases for such comparisons.

2. BASIC CALCULATIONAL TECHNIQUES

The various techniques vary considerably with regard to complexity of calculations required. The simplest calculation is concerned with only the average fuel isotopic composition over the entire core volume as it varies during the irradiation exposure. For this purpose, the reactor core is effectively treated as a single point with average leakage properties and average absorptive and multiplicative nuclear parameters. The nuclear processes are analysed on the basis of multigroup theory with the neutron spectra corresponding to an average spectra for the reactor core. The most complicated calculations are concerned with the detailed spatial variations of the fuel isotopic composition and hence involve the use of three-dimensional multigroup calculations. In conjunction with the gross three-dimensional treatment, two-dimensional calculations are used to obtain the fine structure variations in isotopic composition among the individual fuel rods of a fuel assembly. Such calculations require extensive machine computations and are extremely expensive and time consuming to perform.

Various simplifications can be introduced to reduce the costs of the computations. The three-dimensional problem can be reduced to a series of two- and one-dimensional calculations. Two-dimensional fuel-burn-up calculations are performed for various axial zones of the reactor; each calculation gives the isotopic distribution, across the reactor core at the axial zone
considered, as a function of average fuel exposure in the given axial zone. The results of the two-dimensional zonal calculations are then stitched together by means of a single one-dimensional calculation performed for each value of core average fuel exposure. The foregoing synthesis technique reduces the required machine calculations to a somewhat more practical level.

Further simplifications of the problem are possible. An extremely effective simplification is cylindricization of the core cross-section. Generally, the core layout, rod patterns, and fuel reloading scheme (e.g. out-in zonal refuelling) permit cylindricization with consequent reduction to a two- or a one-dimensional treatment. Because of the geometrical distortion introduced, the comparable locations of the individual fuel assemblies in the cylindricized core cannot be identified. However, the fuel assemblies can be grouped into batches and each batch approximately identified as an annular ring in the cylindricized core. Hence, the cylindricization approximation is most meaningful for fuel accounting purposes when "annular" batches of fuel assemblies are discharged at a time.

With cylindricization of the core, various alternate approaches are possible. The cylindrical core can be directly analysed by two-dimensional R-Z depletion calculations wherein the actual rod positions (through simulation with equivalent distributed poisons) are taken into account during the burn-up (provided their withdrawal pattern reasonably well preserves azimuthal symmetry, which generally is the case). Another approach is to apply the synthesis technique but utilizing, instead, one-dimensional burn-up analysis for each chosen axial zone of the core.

The problem can be still further simplified to one purely in the one-dimensional domain. Analysis is performed for the reactor cylindricized cross-section with no detailed consideration of the variations in the axial direction. Partially inserted control rods are accounted for by adjustment in the strength of the equivalent distributed poisons introduced in the various annular rings of the core cross-section. This method has the important advantage of simplicity of analysis; however, its use to obtain meaningful results is dependent on whether reasonably appropriate axially averaged nuclear constants can be developed. The next order of simplification reduces the problem to a zero-dimensional one, i.e. to one where the reactor core is effectively treated as a single point.

The foregoing successive orders of approximation reduce the problem from an actual three-dimensional to a final zero-dimensional one. It is possible, however, to considerably reduce complexity of treatment by considering fewer neutron energy groups, while retaining the three-dimensional geometry. By use of only a single neutron energy group and of a coarse mesh description of the three-dimensional geometry layout, the calculations are simple enough to be performed on a routine basis.

Various nuclear codes are available for performance of calculations on fuel depletion and heavy isotope build-up. The various reactor designers often use different codes to perform the same type of calculation; these codes were, in many cases, independently developed for the designer's own use and so have different code names. Although differences exist between the codes which perform similar type calculations, the differences are
generally on the details of the calculations rather than on the method of approach.

The following nuclear codes are frequently used in the United States of America. Zero-dimensional multigroup depletion codes: CINDER, CRUMB, GAD, LEOPARD, TONG. One-dimensional multigroup depletion codes: CANDLE, CNCR, GROM, FEVER, SIZZLE, ZIP. Two-dimensional multigroup depletion codes: ASSAULT, DDB, TURBO. Three-dimensional one-group codes: FLARE. Three-dimensional multigroup depletion codes: DRACO, TNT-1, TNT02. Depletion systems: DEPLETION, HARMONY. For detailed descriptions of these codes, Refs. [1-18] should be consulted.

3. UTILIZATION OF IN-CORE AND OUT-OF-CORE MEASUREMENTS

The promising measurement techniques used in operating power reactors for obtaining fuel assembly relative power outputs and, in some cases, also axial power distributions are described.

3.1. Coolant enthalpy rise measurements in individual fuel assemblies

In several power reactors, the fuel assemblies are housed in individual flow channels which confine the coolant flow to the fuel assembly. Measurements are taken during power operation of the coolant flow for each assembly and of the coolant-inlet and individual coolant-outlet temperatures. Hence, the measurement results directly give the individual assembly integrated exposure values. The overall accuracy depends on the individual measurement accuracies and on the relative magnitude and accuracy of estimation of the coolant heat losses. With all or a sufficient number of representative channels monitored, the individual channel outputs can be normalized to the more accurately measured total power output.

In the Carolinas-Virginia reactor, the reactor core comprises a lattice array of U-tubes, each of which contains two fuel assemblies (one in each leg of a U-tube). Instrumentation is provided to measure the coolant flow rate and temperature rise across the individual U-tubes. These data provide a continuing indication of the relative energy production from each U-tube. The division of measured U-tube thermal power between the two fuel assemblies in the U-tube is obtained by means of PDQ calculations. The foregoing data, when combined with the normal plant measurements of total thermal power output (including experimental measurement of the total heat losses to the moderator), permit determination of assemblywise power outputs and integrated exposures.

In the Hallam reactor, measurements are taken of coolant-inlet and outlet temperatures and of the positions of the individual fuel channel orifices, which are adjustable under power operation. The overall accuracy is crucially dependent on the accuracy of the orifice-position indications and of the orifice flow calibration curves. The average coolant temperature rise across the core is 450° F so that errors of as much as 5° F in the individual temperature measurements result in only 1% uncertainty. The main discrepancy is introduced through the flow measurements. It is estimated that the measured relative power factors are good to ± 10%.
In the Piqua reactor, the individual outlet temperatures are measured continually by a single thermocouple located in the outlet coolant stream of each fuel assembly. Individual fuel-assembly powers are found by rationing the individual flow-weighted coolant-temperature rises. The average coolant temperature rise across the core is only about 46°F, and the estimated accuracy of the temperature measurements is ± 4°F. Hence, the uncertainty in the relative power factors associated with the temperature measurements is ± 9%. The flow rate is determined on the basis of the fixed channel orifice setting and is believed known to about ± 10%. When statistically combined, the sum of the errors due to the individual temperature and flow-rate measurements amounts to ± 14%. The heat loss to the moderator is relatively small and so its contribution is negligible.

3.2. In-core flux-wire and ion-chamber measurements

Indirect indications of assemblywise power outputs are obtainable by use of in-core flux wires or miniature ion chambers. Preferably, the flux wires should run along the axis of each fuel assembly or each representative of symmetrical assemblies. More convenient and practical locations for installation of ion chambers can be found by limiting the required flux indications to be representative of individual supracells rather than of individual fuel assemblies. A supracell could, for example, be defined as a 2 × 2 array of fuel assemblies which may or may not include a central cruciform control rod.

If the flux mapping is sufficiently extensive, e.g. each representative of symmetrical fuel assemblies contains at least one flux wire, the flux-wire activity readings can be interpreted in terms of the relative assemblywise power output. This interpretation requires performance of X-Y, PDQ or TURBO calculations for a unit-cell representation of the fuel assembly and associated channel water. The capabilities of this approach are amply demonstrated by results of this type of an analysis on Yankee Core I [19]. This approach is vastly simpler than the use of direct calculations alone and undoubtedly leads to better accuracy of overall results.

The Dresden, Humboldt Bay and Big Rock Point Reactors employ in-core ion chamber system consists of vertical strings of ion chambers which are located so as to measure the relative average power density in suitably defined supracell regions of the core. In Humboldt Bay, eight strings are used which are located at eight of the supracell boundary corners, where a supracell consists of a cruciform control rod and its four surrounding fuel assemblies. Each ion chamber string consists of three miniature fission chambers, one at each of three elevations, and a calibration tube for insertion of flux wires. This system permits monitoring of the neutron flux at 24 discrete points in the core. Dresden contains 16 ion chamber strings with 4 fission chambers per string. Big Rock Point contains 8 strings with 3 fission chambers per string. The in-core readings are used to calculate the relative power of each fuel assembly. The procedure involves interpolations between in-core locations to obtain fuel assembly-average neutron fluxes. It takes into account the various core variables which influence the flux distributions, including control-rod position, steam voids, and reflector effects. Another experimental technique employed at Humboldt Bay utilizes
in-core miniature ion chambers at the sides of the fuel assemblies to measure the fission product gamma fluxes. The results of the in-core gamma scans were in good agreement with the results of the in-core neutron flux measurements.

3.3. Out-of-core gamma flux scanning

In the Yankee Core Evaluation Program [20] gross gamma scans of individual fuel assemblies were made to determine the degree of correlation existing between relative gross gamma activity and relative burn-up (as obtained from in-core data and calculations). Variations in cobalt content of the fuel-rod clad introduced a variable activity source which is not correlated with burn-up and, hence, which interfered with straightforward interpretation of the measurements. The relative gamma activity values generally agreed with the relative burn-up values to within ±16%. This accuracy of indication may be adequate for checking purposes if other methods of determination are not possible or practical.

For measurement of the mid-life power distributions in the Indian Point Reactor, a technique has been developed by Babcock and Wilcox to measure the characteristic gamma-ray activities emitted by the 12.8-d Ba-La fission product chain [21]. Because of the relatively short half-life of the fission products measured, the relative assembly activity is essentially proportional to the relative assembly power just before reactor shut-down. The experimentally inferred relative power distributions (both assemblywise and axially along the length of the assemblies) agreed well with analytically derived distributions, thus providing a check on the accuracy of the measurements [22].

4. ASSEMBLYWISE POWER OUTPUTS AND FUEL EXPOSURES

Of special interest here are the measurements of assemblywise power distributions and, in a few cases, of assemblywise exposure distributions made in several operating power reactors, notably Yankee Core I, Indian Point, Dresden, Humboldt Bay and RWE-Kahl. Calculations have been performed by the reactor designers; both calculated and measured results are presented to provide an indication of the reliability of the calculational techniques used. The results of calculations performed at Combustion Engineering (CEND) for Yankee Core I are included.

4.1. Yankee Core I

An extensive in-core instrumentation system was installed in Yankee Core I to measure the power density and exposure distributions throughout life. Analytical determinations were also made for comparison with the measurement results [19]. The primary analytical tool used by Westinghouse was the TURBO depletion code run with two neutron groups in X-Y geometry; the nuclear code, FLARE, which handles only a single neutron energy group, was run in X-Y geometry at CEND.
In the analytical studies, it was possible to use experimental power densities and rod configurations from the actual operation. By using a fine mesh description in one quadrant of Yankee Core I (which possessed quarter-core symmetry), the core was represented quite accurately in the TURBO study. A coarse mesh description with nodal points placed only at the centres of the individual fuel assemblies was used in the FLARE study. The analyses were made for an axial slab of the core. The power output of the slab relative to the power output of the total core was obtained from the flux-wire data.

The analytical and experimental power distributions are compared in Figs. 1-3 at values of MWd/t U ranging from 1385 to 7755. The agreement between calculated and experimental values of relative assembly power outputs is generally good. Some of the differences between analytical and experimental values are attributable to scatter in the experimental data. Another source of discrepancies is due to the fact that symmetrically located control rod groups were periodically interchanged during operation of Yankee Core I, whereas in the TURBO and FLARE problems, no interchanges were made.

The burn-up distributions at the end of 7755 MWd/t U average exposure are compared in Fig. 4. The agreement between analytical and experimental burn-ups is excellent. Most discrepancies are well within the range of interchange effects and estimated experimental error (5-10%). The experimental burn-ups were found by a time integration of the discrete power distribution measurements; this integration smoothed out the scatter in the power data. The good agreement obtained demonstrates the validity of the TURBO and FLARE burn-up results.

4.2. Dresden, Humboldt Bay and RWE-Kahl

Gamma scan measurements have been made by General Electric as part of start-up test programmes on the Dresden, Humboldt Bay and Kahl boiling-water reactors. These measurements, which were performed at reactor shut-down, permit very detailed power distribution determinations which are not possible during reactor operation.

Comparisons were made by General Electric between measured and calculated power distributions [23-24]. The calculations were performed with the coarse mesh, three-dimensional, one-group nuclear code, FLARE. The calculated results were found to be in reasonably good agreement with the experimental values. Some statistics on the comparisons between calculated and experimental data are given in Table I. The estimated standard deviation of the experimental data from Dresden and Humboldt is 7%; hence, the standard deviation of the error between calculations and measurements is nearly the same as the standard deviation of the experimental measurements.

4.3. Indian point

As indicated in section 3.3, the gamma scan tests performed by Babcock and Wilcox utilized a technique which measured the distribution of the characteristic gamma rays of the barium-lanthanum fission product chain along the length of individual fuel elements. Calculations were also performed by Babcock and Wilcox for comparison with the experimentally derived power
FIG. 1. Analytical and experimental power distributions for axial zone of Yankee Core I at 1385 MWd/t U average exposure

FIG. 2. Analytical and experimental power distributions for axial zone of Yankee Core I at 5694 MWd/t U average exposure
FIG. 3. Analytical and experimental power distributions for axial zone of Yankee Core I at 7755 MWd/t U average exposure

FIG. 4. Analytical and experimental exposure distributions for axial zone of Yankee Core I at 7755 MWd/t U average exposure
TABLE I

| REACTOR       | APPROX. CORE CONDITION | \( |\Delta| \) | \( \sigma \) |
|---------------|------------------------|--------|--------|
| DRESDEN       | CLEAN                  | 8.0    | 9.6    |
| HUMBOLDT      | CLEAN                  | 4.7    | 5.7    |
| RWE-KAHL      | CLEAN                  | 4.8    | 6.5    |
| RWE-KAHL      | 2000 MWD/MTU           | 2.7    | 3.2    |
| RWE-KAHL      | 4000 MWD/MTU           | 2.9    | 3.7    |
| INDIAN POINT  | MIDLIFE                | 7.7    | 8.7    |

\( |\Delta| \) - IS THE AVERAGE PERCENTAGE VALUE OF THE ABSOLUTE DIFFERENCE BETWEEN INDIVIDUAL CALCULATED AND MEASURED VALUES

\( \sigma \) - IS THE STANDARD DEVIATION (%) OF THE DIFFERENCE BETWEEN CALCULATED AND MEASURED VALUES

distributions. The calculational method used is basically of the synthesis type in that it combines the results of two-dimensional X-Y calculations by means of one-dimensional axial calculations.

Figure 5 gives the average power output of each element, normalized to the average of unity in the core [25]. The greatest deviation between calculated and measured elementwise power output is 15.2%. Some statistics on the comparisons between calculated and experimental data are included in Table I.

5. CALCULATED AND MEASURED ISOTOPIC COMPOSITIONS

At present, the bulk of the experimental data are generally limited to exposure values of less than about 16 000 MWD/t U with a few data points from Yankee Core I extending into the 16 000 to 25 000 MWD/t U range. Heavy-isotope measurement programmes are being carried out on the Shippingport, Yankee Core I and Dresden reactors. Measurements have been conducted in the Vallecitos reactor. The available data and comparisons with calculated results are presented.

5.1. Sources of data and descriptions of analyses

Isotopic analyses have been made of 30 fuel rods removed from the natural-uranium blanket region of the Shippingport reactor at the end of the first and second seed lives [26]. The depletion ranged from 2500 to 16 000 MWD/t U. The isotopic analyses gave the average concentration in each fuel rod of the uranium and plutonium isotopes and of the fission product \( ^{137} \text{Cs} \) (used as exposure indicator).

The calculational technique employed involves: (a) a three-dimensional, three-group diffusion theory calculation of the gross flux variations over the
core volume; (b) a two-dimensional, three-group diffusion theory calculation of the fine structure of the flux variations over the individual fuel assemblies; (c) a superposition of the gross and fine structure fluxes; and (d) a one-point depletion calculation [26]. The nuclear code, TNT-1, was used for the three-dimensional calculations, PDQ for the two-dimensional calculations, and CINDER for the one-point depletion calculations. The calculational technique is a highly sophisticated one which provides for considerable detail both in geometrical representation and in nuclear data utilization. Hence, the comparison results presented in section 5.2 should give an indication of the probable best prediction accuracy obtainable at the present time.

Isotopic composition measurements have been performed of slightly enriched UO₂ fuel irradiated in the Vallecitos Boiling Water Reactor (VBWR) [27]. The fuel exposures ranged from 100 to 3200 MWd/t. Reference [28] reports these same data and, in addition, gives data on one fuel sample which attained an exposure of 7000 MWd/t in VBWR. Irradiated fuel samples from Dresden in the exposure range of 100 to 3200 MWd/t were also analysed using the same techniques as for the Vallecitos fuel samples, and these data are included in Ref. [28]. The fuel pellets analysed were taken from several positions within the reactor cores.

A multiregion fuel depletion code was utilized at General Electric for calculating the depletion and production of the heavy-isotopes corresponding to the measured burn-up. Details of the method are outlined in Ref. [28].

Following removal of fuel assemblies from Yankee Core I, a programme was initiated at Westinghouse to evaluate the characteristics of the spent fuel by destructive analysis. Fuel burn-up distribution, uranium depletion and plutonium build-up were among the parameters studied. Altogether, more than 200 pellets were analysed.

To ascertain the accuracies of predicting isotopic concentrations, Westinghouse performed four-group TURBO calculations in a manner consistent with the Yankee Core design methods [29]. Simplified four-group calculations were performed at CEND to determine the feasibility of intro-
ducing fairly gross simplifications in the determination of isotopic fuel composition. The procedure used at CEND was briefly as follows:

(a) A homogenized core composition was assumed with criticality maintained during fuel burn-up by variation of homogeneous core poisons. Four-group constants were derived for the central fuel-rod lattice cell of the Yankee Core I fuel assemblies so as to provide direct comparisons with the isotopic composition data obtained for fuel rods in the asymptotic neutron spectrum. FORM was used to obtain the cell constants for the above-thermal energy groups; except for $^{235}\text{U}$ and $^{240}\text{Pu}$, the L-factors for all isotopes were taken as unity. The L-factor for $^{235}\text{U}$ was determined by use of the nuclear code, QUERY, with lattice-interaction effects included. The L-factor for $^{240}\text{Pu}$ was based on the infinite mass approximation of the effective resonance integral and was taken to be concentration dependent. For the thermal group, TEMPEST was used to determine the Wigner-Wilkins spectrum for the homogenized unit cell; the thermal disadvantage factor was obtained from a cylindrical-geometry P-3 calculation of the heterogeneous unit cell using thermal-neutron constants appropriate to the Wigner-Wilkins spectrum.

(b) The zero-dimensional burn-up code, CRUMB, was used to determine $^{235}\text{U}$ depletion and Pu isotopic build-up as a function of average fuel exposure. With the single point treatment, all detailed spatial effects of fuel burn-up are disregarded.

5.2. Comparison results

The Yankee Core I data are compared with the results of the Westinghouse and CEND analyses in Figs. 6 and 7 wherein plutonium isotopic composition and Pu/U mass ratio, respectively, are plotted against $^{235}\text{U}$ depletion. The data scatter is due to experimental uncertainties in both the plutonium and $^{235}\text{U}$ concentration measurements. The available results of the four-group TURBO calculations extend only to about 16 000 MWD/t U. Over the range investigated, the TURBO results agree quite well with the data, the major discrepancy occurring for $^{242}\text{Pu}$. The measured and CEND-calculated compositions at 14 000 and 23 500 MWD/t U average exposure are more conveniently compared in Table II.

Comparison results for all available data are presented in Figs. 8 to 14 as plots of [calculated - measured/measured] $\times$ 100 versus exposure. Insufficient detail was available on the calculational results of Westinghouse on Yankee Core I; in particular, the relationship between $^{235}\text{U}$ depletion and exposure is needed to obtain the calculated plutonium composition as a function of exposure. In view of the importance of the Westinghouse results as one piece of information for assessing the general order of accuracy of calculations and in view of the excellent agreement between the CEND and Westinghouse calculated results on Pu/U mass ratio versus $^{235}\text{U}$ depletion (Fig. 7), the CEND calculated relationship between $^{235}\text{U}$ depletion and exposure was used for further interpretation of the Westinghouse calculated results. Hence, these particular comparison results should be considered as tentative.

The results of Figs. 8 to 14 reflect both the uncertainties in the individual measurements and in the calculations. The percentage differences between calculation and measurement fluctuate appreciably from fuel sample to fuel
What is important for fuel accounting is not the prediction of isotopic content in individual fuel pellets but rather the prediction for a batch of fuel assemblies which contains a large number of pellets. Hence, the individual differences between calculation and measurement obtained for the fuel pellets should be averaged to obtain the uncertainty of prediction corresponding to a batch of fuel assemblies. Of necessity it is assumed that the fuel pellets analysed from any one reactor constitute a representative sample; hence, a simple averaging procedure is used with no weighting factors applied. The results of the averaging process are indicated in Figs. 8 to 14 wherein values of the standard deviation of the difference between calculated and measured values are tabulated.

Preliminary calculated and measured isotopic inventories on a core-total basis are available for the end of Yankee Core I life, which corresponds to 8470 MWd/t U average exposure [30]. These results indicate that $^{235}$U depletion and total Pu content can be predicted to within 3.2 and 2.4%, respectively, of the measured values. These differences are of the order of one-half of the differences indicated in Figs. 8 to 14.
6. ESTIMATED PREDICTION ACCURACIES

Plant measurements of total power output have accuracies of ± 1.5 to 3%, with accuracies close to ± 1.5% being more usual. The accuracy of determining average exposure of a batch relative to average total-core exposure depends on the size of the batch. In a small reactor core, such as BONUS, for which about 20 fuel assemblies would be discharged at any one time, this error is estimated to be less than about ± 5%; for a large power reactor such as Dresden with greater than 100 or so fuel assemblies discharged, the error is less than about ± 2%. Statistical combination of the uncertainties (using a more probable accuracy value of ± 1.8% in total power output) leads to an overall uncertainty in absolute average exposure value of a discharge batch of ± 2.7% for a large batch (≥ 100 fuel assemblies) and of ± 5.3% for a small
FIG. 8. Comparison of measured and calculated $^{235}$U depletion in analysed fuel samples

TABLE II

MEASURED AND CEND-CALCULATED(9)
ISOTOPIC COMPOSITIONS IN YANKEE CORE I

<table>
<thead>
<tr>
<th>MWD/MTU</th>
<th>14,000</th>
<th>23,500</th>
</tr>
</thead>
<tbody>
<tr>
<td>AVERAGE OF MEASUREMENTS</td>
<td>CALCULATED</td>
<td>AVERAGE OF MEASUREMENTS</td>
</tr>
<tr>
<td>$^{235}$U ATOM DEPLETION, %</td>
<td>37.5</td>
<td>38.5</td>
</tr>
<tr>
<td>Pu/U MASS RATIO, %</td>
<td>0.76</td>
<td>0.75</td>
</tr>
<tr>
<td>Pu ISOTOPIC COMPOSITION (ATOM %)</td>
<td>Pu-239</td>
<td>Pu-240</td>
</tr>
<tr>
<td>78.0</td>
<td>14.0</td>
<td>7.2</td>
</tr>
<tr>
<td>75.3</td>
<td>16.2</td>
<td>7.4</td>
</tr>
<tr>
<td>69.5</td>
<td>16.2</td>
<td>12.3</td>
</tr>
<tr>
<td>64.1</td>
<td>21.5</td>
<td>11.3</td>
</tr>
</tbody>
</table>

(a) ZERO-DIMENSIONAL CALCULATIONS FOR FUEL RODS IN THE ASYMPOTIC NEUTRON FLUX OF THE VARIOUS FUEL ASSEMBLIES

batch (~20 fuel assemblies). The corresponding errors in isotopic composition depend on the conversion ratio and total exposure. For conversion ratios in the range of those for Dresden and Yankee (which represent the usual range in present-day power reactors), the uncertainties in isotopic composition corresponding to the uncertainties in batch exposure are approximately as given in Table III.

Uncertainties in the computed conversion ratio lead to about ± 5% uncertainty in total plutonium content; the corresponding uncertainty in $^{235}$U depletion is about ± 1.4%. When the uncertainties in the Pu isotope cross-
<table>
<thead>
<tr>
<th>SYMBOL</th>
<th>SOURCE OF MEASUREMENT</th>
<th>SOURCE OF CALCULATION</th>
<th>STANDARD DEVIATION (%) BETWEEN CALC AND MEAS</th>
</tr>
</thead>
<tbody>
<tr>
<td>○</td>
<td>VBWR</td>
<td>GE</td>
<td>4.8</td>
</tr>
<tr>
<td>○</td>
<td>YANKEE 1</td>
<td>CEND (0-DIMENS CRUMB)</td>
<td>10.1</td>
</tr>
</tbody>
</table>

FIG. 9. Comparison of measured and calculated $^{235}$U content in analysed fuel samples

<table>
<thead>
<tr>
<th>SYMBOL</th>
<th>SOURCE OF MEASUREMENT</th>
<th>SOURCE OF CALCULATION</th>
<th>STANDARD DEVIATION (%) BETWEEN CALC AND MEAS</th>
</tr>
</thead>
<tbody>
<tr>
<td>○</td>
<td>VBWR</td>
<td>GE</td>
<td>9.1</td>
</tr>
<tr>
<td>□</td>
<td>SHIPPINGPORT</td>
<td>BETTIS</td>
<td>5.7</td>
</tr>
<tr>
<td>△</td>
<td>DRESDEN</td>
<td>GE</td>
<td>6.0</td>
</tr>
<tr>
<td>×</td>
<td>YANKEE 1</td>
<td>WESTINGHOUSE</td>
<td>5.2</td>
</tr>
<tr>
<td>○</td>
<td>YANKEE 1</td>
<td>CEND (0-DIMENS CRUMB)</td>
<td>8.7</td>
</tr>
</tbody>
</table>

FIG. 10. Comparison of measured and calculated $^{239}$Pu content in analysed fuel samples

sections are included, the overall uncertainties for constant exposure are increased to about ± 7.0% for total Pu content and about ± 2.0% for $^{235}$U depletion.
FIG. 11. Comparison of measured and calculated $^{240}$Pu content in analysed fuel samples

FIG. 12. Comparison of measured and calculated $^{241}$Pu content in analysed fuel samples
FIG. 13. Comparison of measured and calculated $^{239}$Pu content in analysed fuel samples.

<table>
<thead>
<tr>
<th>SYMBOL</th>
<th>SOURCE OF MEASUREMENT</th>
<th>SOURCE OF CALCULATION</th>
<th>STANDARD DEVIATION (%) BETWEEN CALC AND MEAS</th>
</tr>
</thead>
<tbody>
<tr>
<td>○</td>
<td>VBWR</td>
<td>GE</td>
<td>9.2</td>
</tr>
<tr>
<td>□</td>
<td>SHIPPINGPORT</td>
<td>BETTIS</td>
<td>6.3</td>
</tr>
<tr>
<td>△</td>
<td>YANKEE I</td>
<td>WESTINGHOUSE (2-DIMENS TURBO)</td>
<td>7.0</td>
</tr>
<tr>
<td>□</td>
<td>YANKEE I</td>
<td>CEND (0-DIMENS CRUMB)</td>
<td>9.0</td>
</tr>
</tbody>
</table>

FIG. 14. Comparison of measured and calculated total Pu in analysed fuel samples.

Statistical combination of the uncertainties associated with the exposure level of the discharge batch with the uncertainties associated with the conversion ratio and isotopic cross-sections at constant exposure gives an
TABLE III

ESTIMATED UNCERTAINTIES IN ISOTOPIC COMPOSITION DUE TO UNCERTAINTIES IN EVALUATION OF BATCH EXPOSURE

<table>
<thead>
<tr>
<th>Uncertainty</th>
<th>U-235 Depletion</th>
<th>Total Pu Content</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>MWD/MTU</td>
<td>MWD/MTU</td>
</tr>
<tr>
<td>LARGE BATCH</td>
<td></td>
<td></td>
</tr>
<tr>
<td>(&gt;100 ASSEMBLIES)</td>
<td>2.4%</td>
<td>2.0% 1.8%</td>
</tr>
<tr>
<td>SMALL BATCH</td>
<td>(~20 ASSEMBLIES)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>4.6% 3.9% 3.5%</td>
<td>1.9%</td>
</tr>
</tbody>
</table>

TABLE IV

ESTIMATED OVERALL UNCERTAINTIES IN PREDICTED ISOTOPIC COMPOSITIONS

<table>
<thead>
<tr>
<th>Total Uncertainty</th>
<th>U-235 Depletion</th>
<th>Total Pu Content</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>MWD/MTU</td>
<td>MWD/MTU</td>
</tr>
<tr>
<td>LARGE BATCH</td>
<td></td>
<td></td>
</tr>
<tr>
<td>(&gt;100 ASSEMBLIES)</td>
<td>3.2% 2.8% 7.3%</td>
<td>7.1%</td>
</tr>
<tr>
<td>SMALL BATCH</td>
<td>(~20 ASSEMBLIES)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>5.0% 4.4% 7.9%</td>
<td>7.3%</td>
</tr>
</tbody>
</table>

estimate of the overall uncertainties involved. These overall uncertainties are tabulated in Table IV.

The overall uncertainty values are consistent, in order of magnitude, with the standard-deviation values arrived at from calculated and measured isotopic compositions of fuel samples taken from a number of operating power reactors.

7. GENERAL CONCLUSIONS

The evaluation performed herein indicates that use of the FLARE code provides the best compromise for calculating relative assemblywise fuel exposures for use in fuel accounting. FLARE is simple and inexpensive to run on a routine basis and is flexible enough to handle the variety of control-rod and refuelling management schemes considered for use in present power reactors. In conjunction with FLARE, a zero-dimensional multigroup code must be used to generate the relationships between isotopic concentration and fuel exposure corresponding to the average neutron spectra prevailing over the various core regions.
REFERENCES


[25] BACCOCK AND WILCOX COMPANY, "Consolidated Edison Indian Point Reactor - Core 'A' Gamma Scan Results", USAEC Docket No. 50-3 (1964).
DISCUSSION

C. BENNETT: Are the standard deviations quoted in your paper computed about zero error or about the average of the measurements? I am referring especially to the case of an obviously biased group of errors as shown, for example, in Fig. 11 of the paper.

M. F. VALERINO: The "standard deviations" are computed about zero error and not about the average of the measurements.

F. RONTEIX: In your paper you give examples of comparisons between calculated burn-ups on the one hand and measured plutonium contents or isotopic compositions on the other. What methods were used for the measurements and analyses and how accurate were they?

M. F. VALERINO: The methods used were the standard techniques used in such measurement laboratories as the New Brunswick Laboratory and the Tracerlab Laboratory, viz. mass-spectrometry isotopic dilution and X-ray fluorescence. With reference to the errors associated with fuel-pellet handling and processing, the over-all measurement error involved for $^{235}$U depletion and $^{239}$Pu content was about 4%. The calculation techniques are described in the paper and the relevant references are given.
BURN-UP CALCULATIONS FOR THE BROOKHAVEN GRAPHITE RESEARCH REACTOR FUEL ELEMENTS

P. COLSMANN
BROOKHAVEN NATIONAL LABORATORY, UPTON, N. Y., UNITED STATES OF AMERICA

Abstract — Résumé — Аннотация — Resumen

BURN-UP CALCULATIONS FOR THE BROOKHAVEN GRAPHITE RESEARCH REACTOR FUEL ELEMENTS.

Fuel burn-up calculations for the Brookhaven Graphite Research Reactor involve a distribution of the thermal megawatt days of operations to the fuel elements in proportion to the average thermal neutron flux at their location in the reactor. The megawatt days so assigned can be converted to equivalent uranium-235 consumption when needed.

The original fuel loading for the BGRR was neutral uranium and a single calculation was performed on each fuel element upon discharge from the reactor. A subsequent change to a fully enriched uranium-235 fuel element, however, introduced complications. The average loading of enriched uranium involves about 4800 individual elements, each occupying four different reactor positions during its term in the reactor. The total term for a central channel element is about one year as against six to eight years for an element in a peripheral channel. With the large number of individual fuel elements involved and the approximately monthly small changes needed for operation, it was necessary to resort to a computer programme to follow the burn-up of all the elements on the reactor continuously. Both this and other functions of the computer programme are discussed in the paper.

To date, uranium has been recovered from two batches of spent fuel. On the first, involving 3674 elements discharged from the reactor over a period of 4.9 years, the recovery figures were 5.5% higher than the calculated total of 32.3 kg uranium-235. On the second batch, involving 1296 elements discharged from the reactor over a period of one year, the recovery figures were 2.3% higher than the calculated figures of 10.8 kg uranium-235.

This relatively close agreement seems to indicate that the assumptions made to simplify the programme are acceptable and that the results of the programme are satisfactory for our particular accounting and operating requirements.

CALCUL DU TAUX DE COMBUSTION DANS LE REACTEUR DE RECHERCHE AU GRAPHITE DE BROOKHAVEN. Le calcul du taux de combustion dans le reacteur de recherche au graphite de Brookhaven necessite une repartition des megawatt-jour thermiques entre les elements combustibles proportionnelle au flux moyen de neutrons thermiques a l'emplacement occupé par les elements dans le reacteur. Les megawatt-jours ainsi repartis peuvent être, si nécessaire, convertis en consommation équivalente d’uranium-235.

La charge initiale de combustible dans le réacteur de Brookhaven était de l’uranium naturel et un seul calcul a été effectué sur chaque élément combustible déchargé. L’utilisation ultérieure d’éléments d’uranium-235 totalement enrichi a compliqué les choses. La charge moyenne d’uranium enrichi comprend environ 4800 éléments, chacun occupant quatre positions différentes pendant son séjour dans le réacteur. Le séjour total d’un élément placé dans un canal central est d’environ 1 an tandis qu’il est de 6 à 8 ans pour un élément placé dans un canal de la périphérie. Étant donné le nombre important d’éléments combustibles et les légères modifications auxquelles il faut procéder tous les mois environ, il est nécessaire d’utiliser un programme d’ordinateur pour suivre continuellement la combustion de tous les éléments dans le réacteur. Le mémoire traite les différents aspects de ce programme.

A l’heure actuelle, l’uranium a été récupéré dans deux lots de combustible irradié. Dans le premier, qui comprenait 3674 éléments déchargés du réacteur au cours d’une période de 4,9 années, la quantité récupérée a été supérieure de 5,5% à la quantité totale calculée qui était de 32,3 kg d’uranium-235. Dans le second lot, qui comprenait 1296 éléments déchargés au cours d’une période d’un an, la quantité récupérée a été supérieure de 2,3% à la quantité totale calculée qui était de 10,8 kg d’uranium-235.
Le fait que ces chiffres concordent relativement bien semble indiquer que les hypothèses retenues pour simplifier le programme sont acceptables et que les résultats de ce programme sont satisfaisants pour les besoins particuliers de la comptabilité et des opérations de l’installation considérée.

РАСЧЕТЫ ВЫГОРАНИЯ ТОПЛИВНЫХ ЭЛЕМЕНТОВ ДЛЯ ГРАФИТОВОГО ИССЛЕДОВАТЕЛЬСКОГО РЕАКТОРА В БРУКХЕЙВЕНЕ. Расчеты выгорания топливных элементов для Брукхейвенского графитового исследовательского реактора (БГИР) выполнены с учетом распределения мегаватт-дей тепловой энергии в топливных элементах, пропорционального распределению среднего потока тепловых нейтронов в месте их нахождения в реакторе. Определенные таким образом мегаватт-дни можно преобразовать в случае необходимости в эквивалентное потребление урана-235.

Первоначально реактор загружали природным ураном, и производили один расчет каждого топливного элемента после выгрузки топлива из реактора. Однако последующий переход к топливным элементам, полностью обогащенным U-235, привел к осложнениям. Средняя загрузка обогащенного урана включает приблизительно 4800 отдельных элементов, каждый из которых занимает четыре различных положения в реакторе во время своего срока работы. Общий срок работы элемента в центральном канале составляет приблизительно один год, в то время как срок службы элемента в периферийном канале составляет 6-8 лет. Ввиду большого числа отдельных топливных элементов и необходимости проведения приблизительно раз в месяц небольших необходимых для работы перестановок пришлось прибегнуть к составлению программы для счетно-решающего устройства с тем, чтобы постоянно следить за выгоранием всех элементов в реакторе. Обсуждаются эта и другие функции программы счетно-решающего устройства.

К настоящему времени уран регенерировал из двух партий отработанного топлива. В первой партии, состоящей из 3674 элементов, выгруженных из реактора за период 4,9 года, количество извлеченного топлива на 5,5% превышало общее расчетное количество, составляющее 32,3 кг урана-235. Во второй партии, состоящей из 1296 элементов, выгруженных из реактора за период в один год, количество извлеченного топлива на 2,3% превышало расчетное количество, составляющее 10,8 кг урана-235.

Это сравнительно близкое согласие, по-видимому, указывает на то, что предположения, сделанные в целях упрощения программы, являются приемлемыми и что результаты программы удовлетворяют конкретные потребности по учету и эксплуатации.

CALCULO DEL GRADO DE COMBUSTION DE LOS ELEMENTOS DEL REACTOR DE INVESTIGACION MODERADO POR GRAFITO DE BROOKHAVEN. Para calcular el grado de combustión de los elementos combustibles del reactor de investigación moderado por grafito de Brookhaven (BGRR) es preciso distribuir entre estos elementos los MW térmicos-día de operación proporcionalmente al flujo medio de neutrones térmicos en el lugar que ocupan dentro del reactor. Los MW-día así asignados pueden ser convertidos en consumo equivalente de uranio-235.

La primera carga del BGRR fue de uranio natural y al descargar los elementos combustibles se efectuó un cálculo para cada uno de ellos. Pero la adopción posterior de elementos combustibles de uranio totalmente enriquecido en el isotopo 235 introdujo algunas complicaciones. La carga media de uranio enriquecido consta de unos 4800 elementos, cada uno de los cuales ocupa cuatro posiciones distintas durante el tiempo que permanece dentro del reactor. El tiempo total de permanencia de un elemento combustible en un canal central es aproximadamente de un año en un canal periférico, de seis a ocho años. Dado el gran número de elementos combustibles y los pequeños cambios que la explotación del reactor exige casi mensualmente, resultó necesario recurrir a un programa de cómputo electrónico para poder seguir de manera continua el grado de combustión de todos los elementos combustibles contenidos en el reactor. La memoria trata de los diversos aspectos de este programa.

Hasta la fecha se ha recuperado uranio de dos tandas de combustible agotado. Por lo que respecta a la primera, que contenía 3674 elementos descargados del reactor durante un periodo de 4,9 años, las cifras correspondientes a la recuperación excedieron en 5,6% del total calculado de 32,3 kg de uranio-235. En cuanto a la segunda, con 1296 elementos descargados del reactor durante un periodo de un año, las cifras excedieron en un 2,3% del total calculado de 10,8 kg de uranio-235.

Esta concordancia relativamente estrecha parece indicar que las suposiciones hechas para simplificar el programa son aceptables y que los resultados de éste satisfacen las necesidades que crea la contabilidad y la explotación del BGRR.
To understand properly the problems involved in calculating the burn-up of fuel for a particular reactor, it is necessary to be acquainted with the general reactor configuration and operation. The Brookhaven Graphite Research Reactor is a uranium-fuelled, graphite-moderated, air-cooled reactor of relatively low specific power. It is basically a 7.6-m graphite cube with horizontal fuel channels on a 20.3-cm lattice spacing. One feature is that this cube is in two halves with a gap of about 12 cm between the halves. The cooling air enters at this gap and flows out at right angles to the gap through the fuel channels in each half of the graphite.

As originally designed, this reactor was fuelled with natural uranium metal sealed in finned aluminium tubes about 3.35 m long. Each fuel channel had two such elements, one on each side of the gap. The total reactor loading was in the range of 1400 elements or about 53.5 metric tons of natural uranium. The maximum power level was 30 MW (thermal heat), which gave a maximum central thermal neutron flux of about $5 \times 10^{12} \text{n cm}^{-2} \text{s}^{-1}$. With this relatively low specific power, the life of the fuel elements was many years and there was not much reloading. The burn-up of the $^{235}\text{U}$ and the plutonium produced were hand calculated upon discharge of a fuel element. Knowing the dates of charging and discharging gave the total megawatt days (MWd) of operation accumulated on the reactor while an element was in the reactor. Two basic assumptions were made in determining the portion of these megawatt days to be assigned to a particular fuel element. The first was that the heat production, measured in terms of megawatt days, was directly proportional to the thermal neutron flux distribution in the fuelled area of the reactor. The second was that this thermal neutron flux approximated a cosine distribution. On this basis, each fuel channel in the graphite was assigned a channel factor to be used in assigning burn-up. This channel factor was in essence the portion of the power supplied by a fuel element while in that particular channel. The number of megawatt days assigned to a fuel element in this manner was then converted to an equivalent uranium burn-up and plutonium produced.

No time will be spent in discussing the natural uranium fuel calculations since they were primarily a ready approximation and, because of reprocessing difficulties, no useful verification was ever made. Primary accountability for this type of fuel at the time was maintained on the total uranium involved by piece count rather than on the change of contents due to burn-up in the reactor. The brief outline of the natural uranium elements here is mainly for historical reference.

After about 6.5 years of operation on natural uranium fuel, there was a change over to a new type of fully enriched (approx. 93% $^{235}\text{U}$) uranium fuel element. This new element consisted of three aluminium-clad U-Al alloy plates about one-fourth the length of the natural uranium element, and contained a nominal 14.25 g of $^{235}\text{U}$. The use of this new fuel element offered an increase in the available thermal neutron flux, centrally from $5 \times 10^{12}$ to $2 \times 10^{13} \text{n cm}^{-2} \text{s}^{-1}$, along with a drop in the reactor power level from 30 to 20 MW. While both these features were desirable improvements, the increased handling of the new fuel elements was a complication.

Four of the enriched uranium fuel elements were used to replace one of the natural uranium elements, and the reactor loading was changed
numerically from about 1400 to 4800 fuel elements. In addition, since the centrally located elements next to the gap were in the highest neutron flux, they tended to burn-up faster necessitating a cycling of the elements in a channel towards these central positions to achieve uniform consumption. The enriched uranium elements, therefore, start at the outer end of a channel and occupy four different positions in the channel before being finally discharged from a central position next to the air gap.

The time pattern for this cycling of the enriched uranium element towards the centre of a channel is a function of the location of that channel in the reactor. For a centrally located channel, a single fuel element might remain about three months in each position and be in the reactor for one year. In a channel at the edge of the loading, a fuel element might remain up to two years in each position for a total of six to eight years in the reactor. These estimates were based upon attaining an average of about 40% burn-up of the $^{235}$U atoms in each fuel element.

The reactor loading with the enriched fuel elements presented a relatively complicated pattern of burn-up which, along with the large number of elements involved, made any type of hand calculation impractical. In addition, the simple cosine distribution, previously assumed for the thermal neutron flux distribution, was no longer usable. Actual flux distributions in the new loading had to be approximated from flux traverse measurements. While it was still assumed that the heat production, measured in terms of megawatt days, was directly proportional to the thermal neutron flux distribution, the assignment of channel factors had to be modified. With all these complications, it was necessary to develop a data processing procedure to provide the periodic burn-up information required for operation of the reactor and for nuclear material management.

Because the complete cycling of individual fuel elements varies from one to several years, it is obvious that fuel usage is a continuous rather than a batch process, so that successive burn-up computations are interrelated. The only verification that can be made concerning these computations is an eventual comparison of the assigned and recovered values of uranium contents in the discharged fuel elements over a representative period of time. Any errors, therefore, may not be detected for some time and could be difficult to rectify.

While the present system is on a magnetic tape computer, the only equipment initially available utilized punched cards for data processing. These punched cards imposed limitations on the amount of information that could be processed and their manipulation placed limits on the flexibility of the programme. It was possible, however, to process the large number of fuel elements involved which could not have otherwise been processed by hand. The basic procedure was to have a card for each element in the reactor and to process these cards periodically through a programme that updated the burn-up for each element and at the same time made any changes required in the location of an element due to charging, shifting or discharging operations. In practice, a computation has to be made each time there is any change in fuel element location.

Since the recording of changes in fuel location is simply a mechanical operation, the main point of interest here is the computation of the burn-up
assignment. As mentioned previously, the basic computation involves prorating the reactor heat production, measured in megawatt days, to all the elements in the loading in accordance with their appropriate position factors. The programme for performing this proration was set up to use as a common denominator the MWd value for a position factor of unity. Since the position factors are determined by normalizing the thermal flux distribution, a unity position factor channel would be in the maximum flux. In present practice this common denominator for a particular period is arrived at by dividing the total reactor accumulated MWd by the sum of the position factors for all occupied positions.

Initially, however, the changeover from natural to enriched uranium fuel was characterized by a "two zone" loading. As the natural uranium elements were removed from the central fuel channels, these channels were reloaded with enriched uranium fuel elements. A separation ring of one to two empty channels in width was maintained as a buffer. Over a period of 1.3 yr, the size of the central enriched uranium zone was increased in stages while the surrounding natural uranium zone was proportionately decreased. Since there was no practical way of measuring or estimating the separate power contribution from each fuel zone, the required MWd value for unity position factor could not be arrived at from the measured reactor power.

For the early burn-up programmes it was necessary, therefore, to calculate this value from the fission rate using assumed conditions of thermal neutron flux and $^{235}\text{U}$ fuel content. This calculation was performed on a cumulative basis for successive periods in terms of a single centrally located fuel element. This method appeared valid as long as the central fuel channels represented unity position factor and there was essentially no cycling of fuel elements; these conditions existed during the initial loading and subsequent growth of the enriched uranium fuel zone.

The factor for a particular fuel element is a function of its three-dimensional displacement from the geometric centre of the reactor loading. One of these displacements is represented by the four possible positions in one half of a fuel channel. The other two displacements are simply the horizontal and vertical co-ordinates of the fuel channel location in a plane through the centre of the reactor and perpendicular to the channels. In practice, the vertical and horizontal thermal neutron flux distributions in this central plane are each normalized to unity to give a set of relative factors for the vertical and horizontal channel co-ordinates. The product of these factors for any set of co-ordinates yields a channel factor. Similarly, from the flux distribution along a channel, a relative factor is obtained for each of the four possible fuel element positions in either half of the channel.

The assignment of all these factors is based on the assumption that the thermal neutron flux distribution curves used are relatively uniform throughout the reactor. Actual neutron flux traverses are made during the operation of the reactor by activation of a long wire, such as copper, inserted through the fuel loading. The number of locations that permit such measurements are relatively few, so that local flux perturbations may not be detected. The factors used in the burn-up computation are, therefore, a best approximation for the period concerned. Gross changes in the overall flux distribution patterns are compensated for by periodic flux measurements.
and corresponding factor changes. In the early burn-up computations, during which the enriched uranium fuel was reaching an equilibrium pattern, new position factors were introduced as often as available, roughly at six-month intervals.

Because of these various complications, the burn-up calculation of the enriched uranium fuel since the initial loading in 1957 has involved some modifications. In review, there are essentially four phases which can be summarized. The first phase covered the 490-d period of the "two zone" loading, at the end of which there were 2035 fuel elements in the reactor loading. During this phase, there was no way of verifying the burn-up assigned because of the two zone problem. The second phase covered the next 378 d during which the same method of computation was used, but the loading was all enriched uranium. During this second phase, it was possible to compare the total MWd burn-up assigned to the loading, as calculated on the basis of following a central element, with the measured power accumulated in the reactor operation records. This comparison indicated that the calculated power was 1.5% lower than the measured power. This fairly close agreement gave confidence to the calculations performed during the first phase.

The reactor loading, during the second phase, had been increased to a total of 3695 elements and only 40 elements had been discharged. Starting with the third phase, the basing of the MWd value for a unity position factor on the following of a central element was beginning to lose its validity and actually was not necessary since the measured reactor power could be assigned to the whole loading. This value during the third phase, therefore, was calculated by dividing the MWd measured by reactor operations by the sum of the position factors of all occupied positions. This latter summation was not adaptable to the particular punch card data processing equipment in use and was, therefore, approximated manually. With the use of these approximations, a comparison over the third phase, lasting 259 d, showed the assigned MWd burn-up to be 3.3% lower than the measured power.

The fourth and final phase began in January 1960 with a change from the use of a punch card to magnetic tape system. In the punch card system, two cards were required for each element during a computation, one for the input information and a blank to receive the output. The manipulation of approximately 8000 or more cards for each run was relatively tedious and slow. Also, the limited capacity of a single card restricted the program flexibility by requiring the manual approximation of the sum of the occupied position factors. The magnetic tape system eliminated all these problems. Besides performing the complete computation from a minimum of input data in a matter of minutes, it simultaneously provides a great deal of selective output information as required by nuclear material management or for reactor operations.

A magnetic tape carries a record of all fuel element positions whether empty or occupied. For each position occupied by a fuel element, the tape carries the element serial number, the date it entered that position, where it came from (i.e., direct charge or previous position), and the total accumulated MWd of burn-up assigned. This tape, which represents the reactor
loading as of a certain date, is processed in a computer programme using additional tapes for the input of position factor constants and operational data. The computer processes the information to yield a new tape representing the new status of the reactor loading. Desired output information appears on a secondary tape and is subsequently printed out for use. The tapes representing the reactor loading are filed so that re-runs may be made if necessary and once a year a reactor loading tape is completely printed out as a file record.

Since the reactor loading must be updated whenever fuel elements are moved, the programme is run after each routine reactor shut-down, or about every three to four weeks. During these shut-downs, about 50 to 100 fuel elements may be replaced and three times that number shifted. For nuclear material management purposes, the printed output includes separate records of all elements charged, shifted and discharged as shown by the samples in Figs. 1, 2 and 3. Figures 1 and 2 are typical of the information carried on the magnetic tape. Figure 3 shows the conversion performed by the computer on the MWd figure to equivalent per cent burn-up and uranium consumption and content. For operational purposes, there is a printout of the

<table>
<thead>
<tr>
<th>Q CHAN</th>
<th>CHAN</th>
<th>CHAN</th>
<th>CODE</th>
<th>CUR</th>
<th>PRE</th>
<th>PLS</th>
<th>PLS</th>
<th>DAY</th>
<th>SERIAL</th>
<th>MWD</th>
</tr>
</thead>
<tbody>
<tr>
<td>0100</td>
<td>0211</td>
<td>0301</td>
<td>0304</td>
<td>0307</td>
<td>0402</td>
<td>0508</td>
<td>0608</td>
<td>0705</td>
<td>0705</td>
<td>0705</td>
</tr>
<tr>
<td>0.996</td>
<td>0.493</td>
<td>0.967</td>
<td>0.918</td>
<td>0.785</td>
<td>0.933</td>
<td>0.643</td>
<td>0.736</td>
<td>0.572</td>
<td>0.572</td>
<td></td>
</tr>
<tr>
<td>C</td>
<td>C</td>
<td>C</td>
<td>C</td>
<td>C</td>
<td>C</td>
<td>C</td>
<td>C</td>
<td>C</td>
<td>C</td>
<td></td>
</tr>
<tr>
<td>7</td>
<td>8</td>
<td>7</td>
<td>7</td>
<td>7</td>
<td>7</td>
<td>7</td>
<td>7</td>
<td>7</td>
<td>7</td>
<td></td>
</tr>
<tr>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td></td>
</tr>
<tr>
<td>5384</td>
<td>5384</td>
<td>5384</td>
<td>5384</td>
<td>5384</td>
<td>5384</td>
<td>5384</td>
<td>5384</td>
<td>5384</td>
<td>5384</td>
<td></td>
</tr>
<tr>
<td>68JR</td>
<td>1TNT</td>
<td>370G</td>
<td>20NT</td>
<td>11LZ</td>
<td>7IKK</td>
<td>32PR</td>
<td>35PB</td>
<td>33SS</td>
<td>38PR</td>
<td></td>
</tr>
<tr>
<td>0.</td>
<td>0.</td>
<td>0.</td>
<td>0.</td>
<td>0.</td>
<td>0.</td>
<td>0.</td>
<td>0.</td>
<td>0.</td>
<td>0.</td>
<td></td>
</tr>
</tbody>
</table>

FIG. 1. Sample printout of fuel elements charged into the BGRR on day No. 5384

status of all channels with one or more empty positions, as shown by the sample in Fig. 4. It is also possible to print out all channels which exceed certain burn-up criteria as desired. These listings allow the reactor operators to plan the future fuel cycling requirements.

Figure 5 is a sample of the summary sheet provided by the computer. As indicated by the first item, "Number of Loading Errors", the computer
programme will detect and record any accidental errors in the input information. Malfunctions of the computer or tape defects, however, can only be detected by checking the totals on the sheet to be sure that all operations were properly performed.

The burn-up programme involves only a few basic mathematical computations in comparison with the capabilities of the computer. The bulk of the effort performed by the computer is in correctly processing the data and, for this reason, the problems of programming involved mostly ensuring a sufficient number of testing steps to prevent erroneous operations. For instance, the computer will reject an instruction to charge a loaded position or to discharge or shift out of an empty position. In such instances it prints out the erroneous information separately for analysis and records an error on the summary sheet.

The burn-up information as carried in the computer programme is the single cumulative MWd figure. When specific burn-up information is required for an element, such as at the time of final discharge, this MWd figure is converted into equivalent per cent burn-up of the $^{235}\text{U}$ atoms, $^{235}\text{U}$ consumed, $^{235}\text{U}$ fissioned, $^{235}\text{U}$ remaining and uranium remaining, on the basis of calculated conversion ratios and assigned standard weights for the uranium and $^{235}\text{U}$ contents of a fuel element. While most of the $^{235}\text{U}$ is consumed by thermal fission, approximately 20% of that amount is at the same time converted to the isotope $^{238}\text{U}$ by neutron capture. The initial $^{235}\text{U}$ content of an element is, therefore, reduced by the total of the amounts of $^{235}\text{U}$ fissioned and converted to $^{238}\text{U}$. The initial uranium content, however, is reduced only by the amount of $^{235}\text{U}$ fissioned.

### Table: Reactor Fuel Inventory

<table>
<thead>
<tr>
<th>CHAN</th>
<th>CHAN</th>
<th>FACT</th>
<th>CODE</th>
<th>CUR</th>
<th>POS</th>
<th>PRE</th>
<th>POS</th>
<th>DAY</th>
<th>SERIAL</th>
<th>MWD</th>
</tr>
</thead>
<tbody>
<tr>
<td>0100</td>
<td>0.996</td>
<td>SI</td>
<td>5</td>
<td>7</td>
<td>5384</td>
<td>31</td>
<td>HP</td>
<td>0.906</td>
<td></td>
<td></td>
</tr>
<tr>
<td>0100</td>
<td>0.996</td>
<td>SI</td>
<td>3</td>
<td>5</td>
<td>5384</td>
<td>71</td>
<td>ED</td>
<td>2.149</td>
<td></td>
<td></td>
</tr>
<tr>
<td>0100</td>
<td>0.996</td>
<td>SI</td>
<td>1</td>
<td>3</td>
<td>5384</td>
<td>24</td>
<td>DT</td>
<td>3.415</td>
<td></td>
<td></td>
</tr>
<tr>
<td>0100</td>
<td>0.996</td>
<td>SI</td>
<td>2</td>
<td>4</td>
<td>5384</td>
<td>73</td>
<td>DS</td>
<td>3.415</td>
<td></td>
<td></td>
</tr>
<tr>
<td>0100</td>
<td>0.996</td>
<td>SI</td>
<td>4</td>
<td>6</td>
<td>5384</td>
<td>69</td>
<td>ED</td>
<td>2.149</td>
<td></td>
<td></td>
</tr>
<tr>
<td>0100</td>
<td>0.996</td>
<td>SI</td>
<td>6</td>
<td>8</td>
<td>5384</td>
<td>36</td>
<td>HP</td>
<td>0.996</td>
<td></td>
<td></td>
</tr>
<tr>
<td>0211</td>
<td>0.493</td>
<td>SI</td>
<td>5</td>
<td>7</td>
<td>5384</td>
<td>48</td>
<td>CL</td>
<td>1.066</td>
<td></td>
<td></td>
</tr>
<tr>
<td>0211</td>
<td>0.493</td>
<td>SI</td>
<td>3</td>
<td>5</td>
<td>5384</td>
<td>58</td>
<td>BX</td>
<td>2.067</td>
<td></td>
<td></td>
</tr>
<tr>
<td>0211</td>
<td>0.493</td>
<td>SI</td>
<td>1</td>
<td>3</td>
<td>5384</td>
<td>JJ</td>
<td>25</td>
<td>3.017</td>
<td></td>
<td></td>
</tr>
<tr>
<td>0211</td>
<td>0.493</td>
<td>SI</td>
<td>2</td>
<td>4</td>
<td>5384</td>
<td>JK</td>
<td>20</td>
<td>3.017</td>
<td></td>
<td></td>
</tr>
<tr>
<td>0211</td>
<td>0.493</td>
<td>SI</td>
<td>4</td>
<td>6</td>
<td>5384</td>
<td>75</td>
<td>DM</td>
<td>2.067</td>
<td></td>
<td></td>
</tr>
<tr>
<td>0211</td>
<td>0.493</td>
<td>SI</td>
<td>6</td>
<td>8</td>
<td>5384</td>
<td>66</td>
<td>DW</td>
<td>1.066</td>
<td></td>
<td></td>
</tr>
<tr>
<td>0301</td>
<td>0.967</td>
<td>SI</td>
<td>5</td>
<td>7</td>
<td>5384</td>
<td>47</td>
<td>TP</td>
<td>0.879</td>
<td></td>
<td></td>
</tr>
<tr>
<td>0301</td>
<td>0.967</td>
<td>SI</td>
<td>3</td>
<td>5</td>
<td>5384</td>
<td>23</td>
<td>EV</td>
<td>2.092</td>
<td></td>
<td></td>
</tr>
<tr>
<td>0301</td>
<td>0.967</td>
<td>SI</td>
<td>1</td>
<td>3</td>
<td>5384</td>
<td>23</td>
<td>DT</td>
<td>3.338</td>
<td></td>
<td></td>
</tr>
<tr>
<td>0301</td>
<td>0.967</td>
<td>SI</td>
<td>2</td>
<td>4</td>
<td>5384</td>
<td>75</td>
<td>DS</td>
<td>3.338</td>
<td></td>
<td></td>
</tr>
<tr>
<td>0301</td>
<td>0.967</td>
<td>SI</td>
<td>4</td>
<td>6</td>
<td>5384</td>
<td>16</td>
<td>EE</td>
<td>2.092</td>
<td></td>
<td></td>
</tr>
<tr>
<td>0301</td>
<td>0.967</td>
<td>SI</td>
<td>6</td>
<td>8</td>
<td>5384</td>
<td>17</td>
<td>IN</td>
<td>0.879</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

**FIG. 2.** Sample printout of fuel elements shifted in the BGR on day No. 5384
The conversion of burn-up in terms of MWd to equivalent $^{235}\text{U}$ fissioned and consumed is by use of a simple ratio determined for the particular reactor involved. For the BGRR the conversion factors used are 1.055 g of $^{235}\text{U}$ fissioned per MWd and 1.255 g of $^{235}\text{U}$ consumed per MWd. These factors are based on the assumption of 300 MeV of energy as heat per thermal fission in the reactor and an alpha factor of 0.19 for the conversion of $^{235}\text{U}$ to $^{236}\text{U}$. The standard weights for the uranium and $^{235}\text{U}$ contents of a single fuel element are based on the average contents of the elements as received from the fuel fabricator. These standard weights, which are introduced as constants in the computer programme, have been adjusted occasionally over the years to compensate for variations in the uranium contents of new fuel received. Since the burn-up information carried by the computer is in terms of MWd, this type of adjustment is easily made and while it results in a small change in the overall reactor uranium content, the effect per element is negligible.

Since the initial loading of the enriched uranium fuel elements in January of 1957, over eight years have elapsed and approximately 6400 enriched uranium fuel elements have been cycled through the reactor and discharged. To date, a total of 4970 of these elements from the first eight years have been reprocessed for recovery of the uranium. The first batch consisted of 3674 elements from the first six years involving all four phases of the burn-up calculations discussed. About 2000 of these elements were in the reactor during the first phase involving the "two zone" loading. The total $^{235}\text{U}$ content of this batch was calculated to be 32,344 g and the recovered value reported was 34,140 g. The total uranium content was calculated to be 39,483 g and the recovered value reported was 41,310 g. Compared with the recovered values, therefore, the calculated values were 5.3% low for $^{235}\text{U}$ and 4.4% low for uranium. The comparisons made during the second and third phases had indicated that the calculated burn-up assignment was low compared with that measured by reactor operations. The recovery values in turn indicated that the calculated values had been too high. However, because of the uncertainties introduced by the two-zone loading, this discrepancy could not be considered as significant.

The second batch of elements reprocessed consisted of 1296 elements discharged during the seventh year, so that these had been in the reactor mostly during the fourth phase or present method of computation. The total $^{235}\text{U}$ content of this second batch was calculated to be 10,841 g and the recovered value reported was 11,099 g. The total uranium content was calculated to be 13,418 g and the recovered value reported was 13,422 g. Compared with the recovered values, the calculated value was 2.3% low for $^{235}\text{U}$ and essentially equal for uranium. While this comparison would indicate that the calculated burn-up assignment for this batch was high, the per cent differences are too close to be significant.

This relatively close agreement between the calculated and recovered values, obtained over a representative period of time, seems to indicate that the many assumptions made and the operational data used were valid, and that the results of the computer programme are satisfactory for both the operational and accounting requirements.
**DISChARGED ELEMENTS**

**FIG. 3.** Sample printout of fuel elements discharged from the BGRR on day No. 5384

<table>
<thead>
<tr>
<th>Q CHAN</th>
<th>CHAN CODE</th>
<th>CUR</th>
<th>PRE</th>
<th>DAY</th>
<th>SERIAL</th>
<th>MWD</th>
<th>PCT</th>
<th>U235</th>
<th>U235</th>
<th>U235</th>
<th>URAN</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 0100</td>
<td>0.996 SI</td>
<td>1</td>
<td>2</td>
<td>3</td>
<td>5132</td>
<td>180X</td>
<td>4.595</td>
<td>40.599</td>
<td>5.767</td>
<td>5.767</td>
<td>4.846</td>
</tr>
<tr>
<td>1 0100</td>
<td>0.996 SI</td>
<td>1</td>
<td>2</td>
<td>3</td>
<td>5132</td>
<td>258X</td>
<td>4.595</td>
<td>40.599</td>
<td>5.767</td>
<td>5.767</td>
<td>4.846</td>
</tr>
<tr>
<td>1 0211</td>
<td>0.473 SI</td>
<td>1</td>
<td>1</td>
<td>3</td>
<td>4814</td>
<td>KG 6</td>
<td>5.553</td>
<td>49.045</td>
<td>6.988</td>
<td>5.856</td>
<td>7.267</td>
</tr>
<tr>
<td>1 0211</td>
<td>0.473 SI</td>
<td>1</td>
<td>2</td>
<td>4</td>
<td>4814</td>
<td>L37</td>
<td>5.553</td>
<td>40.045</td>
<td>6.568</td>
<td>5.856</td>
<td>7.267</td>
</tr>
<tr>
<td>1 0301</td>
<td>0.987 SI</td>
<td>1</td>
<td>1</td>
<td>3</td>
<td>5132</td>
<td>38AF</td>
<td>4.505</td>
<td>39.792</td>
<td>5.654</td>
<td>4.751</td>
<td>8.582</td>
</tr>
<tr>
<td>1 0301</td>
<td>0.987 SI</td>
<td>1</td>
<td>2</td>
<td>4</td>
<td>5132</td>
<td>74OU</td>
<td>4.505</td>
<td>39.792</td>
<td>5.654</td>
<td>4.751</td>
<td>8.582</td>
</tr>
<tr>
<td>1 0304</td>
<td>0.811 SI</td>
<td>1</td>
<td>1</td>
<td>3</td>
<td>5132</td>
<td>43AY</td>
<td>4.609</td>
<td>40.721</td>
<td>5.783</td>
<td>4.891</td>
<td>8.451</td>
</tr>
<tr>
<td>1 0304</td>
<td>0.811 SI</td>
<td>1</td>
<td>2</td>
<td>4</td>
<td>5132</td>
<td>26C6</td>
<td>4.609</td>
<td>40.721</td>
<td>5.783</td>
<td>4.891</td>
<td>8.451</td>
</tr>
<tr>
<td>1 0307</td>
<td>0.785 SI</td>
<td>1</td>
<td>1</td>
<td>3</td>
<td>5076</td>
<td>55C2</td>
<td>4.616</td>
<td>40.770</td>
<td>5.793</td>
<td>4.868</td>
<td>8.443</td>
</tr>
<tr>
<td>1 0307</td>
<td>0.785 SI</td>
<td>1</td>
<td>2</td>
<td>4</td>
<td>5076</td>
<td>65GU</td>
<td>3.825</td>
<td>33.784</td>
<td>4.690</td>
<td>4.034</td>
<td>9.435</td>
</tr>
<tr>
<td>1 0402</td>
<td>0.933 SI</td>
<td>1</td>
<td>1</td>
<td>3</td>
<td>5132</td>
<td>128G</td>
<td>4.564</td>
<td>40.226</td>
<td>5.715</td>
<td>4.803</td>
<td>8.520</td>
</tr>
<tr>
<td>1 0402</td>
<td>0.933 SI</td>
<td>1</td>
<td>2</td>
<td>4</td>
<td>5132</td>
<td>80BY</td>
<td>4.564</td>
<td>40.226</td>
<td>5.715</td>
<td>4.803</td>
<td>8.520</td>
</tr>
<tr>
<td>1 0606</td>
<td>0.643 SI</td>
<td>1</td>
<td>1</td>
<td>3</td>
<td>5031</td>
<td>L016</td>
<td>4.624</td>
<td>40.843</td>
<td>5.803</td>
<td>4.876</td>
<td>8.432</td>
</tr>
<tr>
<td>1 0606</td>
<td>0.643 SI</td>
<td>1</td>
<td>2</td>
<td>4</td>
<td>5031</td>
<td>G015</td>
<td>4.624</td>
<td>40.843</td>
<td>5.803</td>
<td>4.876</td>
<td>8.432</td>
</tr>
<tr>
<td>1 0705</td>
<td>0.736 SI</td>
<td>1</td>
<td>1</td>
<td>3</td>
<td>5076</td>
<td>RAG26</td>
<td>4.617</td>
<td>40.783</td>
<td>5.794</td>
<td>4.869</td>
<td>8.441</td>
</tr>
<tr>
<td>1 0705</td>
<td>0.736 SI</td>
<td>1</td>
<td>2</td>
<td>4</td>
<td>5076</td>
<td>HP57</td>
<td>4.617</td>
<td>40.783</td>
<td>5.794</td>
<td>4.869</td>
<td>8.441</td>
</tr>
<tr>
<td>1 1003</td>
<td>0.572 SI</td>
<td>1</td>
<td>1</td>
<td>3</td>
<td>4906</td>
<td>TP67</td>
<td>5.777</td>
<td>51.027</td>
<td>7.250</td>
<td>6.093</td>
<td>6.985</td>
</tr>
<tr>
<td>1 1003</td>
<td>0.572 SI</td>
<td>1</td>
<td>2</td>
<td>4</td>
<td>4906</td>
<td>TL28</td>
<td>5.777</td>
<td>51.027</td>
<td>7.250</td>
<td>6.093</td>
<td>6.985</td>
</tr>
</tbody>
</table>
## Reactor Fuel Inventory

### Reactor Day 5384

**Channels Containing Empty Positions**

<table>
<thead>
<tr>
<th>Q CHAN</th>
<th>CHAN</th>
<th>FCT</th>
<th>CODE</th>
<th>CUR</th>
<th>PRE</th>
<th>NO.</th>
<th>SERIAL</th>
<th>MWD</th>
<th>PCT</th>
<th>U235</th>
<th>U235</th>
<th>U235</th>
<th>URAN</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 0201</td>
<td>0.983</td>
<td>O.C</td>
<td>7</td>
<td>0</td>
<td>5160</td>
<td>14JE</td>
<td>0.790</td>
<td>6.942</td>
<td>0.992</td>
<td>0.634</td>
<td>13.243</td>
<td>14.451</td>
<td></td>
</tr>
<tr>
<td>1 0201</td>
<td>0.983</td>
<td>S.I</td>
<td>5</td>
<td>7</td>
<td>5160</td>
<td>436K</td>
<td>1.904</td>
<td>14.920</td>
<td>2.390</td>
<td>2.408</td>
<td>11.846</td>
<td>13.276</td>
<td></td>
</tr>
<tr>
<td>1 0201</td>
<td>0.983</td>
<td>S.I</td>
<td>3</td>
<td>5</td>
<td>5160</td>
<td>82RA</td>
<td>3.344</td>
<td>29.890</td>
<td>4.247</td>
<td>3.569</td>
<td>9.989</td>
<td>11.715</td>
<td></td>
</tr>
<tr>
<td>1 0201</td>
<td>0.983</td>
<td>F.P</td>
<td>0</td>
<td>0</td>
<td>000000</td>
<td>000000</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td></td>
</tr>
<tr>
<td>1 0201</td>
<td>0.993</td>
<td>S.I</td>
<td>4</td>
<td>6</td>
<td>5160</td>
<td>06FY</td>
<td>3.344</td>
<td>29.890</td>
<td>4.247</td>
<td>3.569</td>
<td>9.989</td>
<td>11.715</td>
<td></td>
</tr>
<tr>
<td>1 0201</td>
<td>0.993</td>
<td>S.I</td>
<td>6</td>
<td>8</td>
<td>5160</td>
<td>07FE</td>
<td>1.904</td>
<td>15.870</td>
<td>2.390</td>
<td>2.008</td>
<td>11.846</td>
<td>13.276</td>
<td></td>
</tr>
<tr>
<td>1 0201</td>
<td>0.993</td>
<td>O.C</td>
<td>8</td>
<td>0</td>
<td>5160</td>
<td>40AH</td>
<td>0.790</td>
<td>6.942</td>
<td>0.992</td>
<td>0.834</td>
<td>13.243</td>
<td>14.451</td>
<td></td>
</tr>
<tr>
<td>1 0308</td>
<td>0.722</td>
<td>O.C</td>
<td>7</td>
<td>0</td>
<td>4906</td>
<td>74EH</td>
<td>1.361</td>
<td>12.025</td>
<td>1.708</td>
<td>1.436</td>
<td>12.527</td>
<td>13.848</td>
<td></td>
</tr>
<tr>
<td>1 0308</td>
<td>0.722</td>
<td>O.C</td>
<td>5</td>
<td>0</td>
<td>4906</td>
<td>1269</td>
<td>1.708</td>
<td>15.594</td>
<td>2.216</td>
<td>1.862</td>
<td>12.019</td>
<td>13.422</td>
<td></td>
</tr>
<tr>
<td>1 0308</td>
<td>0.722</td>
<td>O.C</td>
<td>0</td>
<td>0</td>
<td>4906</td>
<td>020V</td>
<td>1.828</td>
<td>16.145</td>
<td>2.294</td>
<td>1.928</td>
<td>11.941</td>
<td>13.356</td>
<td></td>
</tr>
<tr>
<td>1 0308</td>
<td>0.722</td>
<td>F.P</td>
<td>0</td>
<td>0</td>
<td>000000</td>
<td>000000</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td></td>
</tr>
<tr>
<td>1 0308</td>
<td>0.722</td>
<td>S.I</td>
<td>2</td>
<td>4</td>
<td>5272</td>
<td>044R</td>
<td>3.419</td>
<td>33.735</td>
<td>4.793</td>
<td>4.028</td>
<td>9.442</td>
<td>11.256</td>
<td></td>
</tr>
<tr>
<td>1 0308</td>
<td>0.722</td>
<td>S.I</td>
<td>4</td>
<td>6</td>
<td>5272</td>
<td>48CC</td>
<td>2.554</td>
<td>22.366</td>
<td>3.206</td>
<td>2.694</td>
<td>11.029</td>
<td>12.590</td>
<td></td>
</tr>
<tr>
<td>1 0308</td>
<td>0.722</td>
<td>S.I</td>
<td>6</td>
<td>8</td>
<td>5272</td>
<td>33FX</td>
<td>1.316</td>
<td>11.621</td>
<td>1.651</td>
<td>1.388</td>
<td>12.584</td>
<td>13.897</td>
<td></td>
</tr>
<tr>
<td>1 0308</td>
<td>0.722</td>
<td>O.C</td>
<td>9</td>
<td>0</td>
<td>5272</td>
<td>70TC</td>
<td>0.304</td>
<td>2.689</td>
<td>0.382</td>
<td>0.321</td>
<td>13.853</td>
<td>14.963</td>
<td></td>
</tr>
</tbody>
</table>

**Fig. 4.** Sample printout of channels with one or more empty fuel positions
### CHANNEL DATA

<table>
<thead>
<tr>
<th>Description</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Run # 103</td>
<td></td>
</tr>
<tr>
<td>Date</td>
<td>14 May 1965</td>
</tr>
<tr>
<td>Number of loading errors</td>
<td>0</td>
</tr>
<tr>
<td>Number of channels with fuel</td>
<td>615</td>
</tr>
<tr>
<td>Number of channels with empty positions</td>
<td>25</td>
</tr>
<tr>
<td>Number of new channels charged</td>
<td>0</td>
</tr>
<tr>
<td>Number of channels exceeding burnup criteria</td>
<td>232</td>
</tr>
</tbody>
</table>

### ELEMENT DATA

<table>
<thead>
<tr>
<th>Description</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Number of elements burnup updated only</td>
<td>4873</td>
</tr>
<tr>
<td>Number of new elements charged</td>
<td>92</td>
</tr>
<tr>
<td>Number of elements shifted</td>
<td>276</td>
</tr>
<tr>
<td>Number of elements discharged</td>
<td>92</td>
</tr>
<tr>
<td>Number of elements in new loading</td>
<td>4873</td>
</tr>
</tbody>
</table>

### OUTPUT DATA

<table>
<thead>
<tr>
<th>Description</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sum of MWD of discharged elements</td>
<td>441.778</td>
</tr>
<tr>
<td>Sum of MWD of elements in new loading</td>
<td>10202.640</td>
</tr>
<tr>
<td>Grams U-235 in new loading</td>
<td>56564.791</td>
</tr>
<tr>
<td>Grams uranium in new loading</td>
<td>63719.511</td>
</tr>
<tr>
<td>Sum of element factors in new loading</td>
<td>768.229</td>
</tr>
</tbody>
</table>

### INPUT DATA

<table>
<thead>
<tr>
<th>Description</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>BU. CRITERION 1 (7 OR 8)</td>
<td>2.250</td>
</tr>
<tr>
<td>BU. CRITERION 2 (5,3,1,2,4,OR 6)</td>
<td>5.900</td>
</tr>
<tr>
<td>BU. CRITERION 3 (7+5+3 OR 4+6+8)</td>
<td>5.100</td>
</tr>
<tr>
<td>N-S FACTORS</td>
<td></td>
</tr>
<tr>
<td></td>
<td>0.257</td>
</tr>
<tr>
<td></td>
<td>0.274</td>
</tr>
<tr>
<td></td>
<td>0.274</td>
</tr>
<tr>
<td></td>
<td>0.265</td>
</tr>
<tr>
<td></td>
<td>0.265</td>
</tr>
<tr>
<td></td>
<td>0.204</td>
</tr>
<tr>
<td></td>
<td>0.204</td>
</tr>
<tr>
<td>Input sum of element factors</td>
<td>768.651</td>
</tr>
<tr>
<td>Input sum of megawatt/day</td>
<td>396.390</td>
</tr>
</tbody>
</table>

**FIG. 5.** Sample of computer summary for day No. 5384
DETERMINATION OF PLUTONIUM-239 CONTENT IN NUCLEAR FUEL BY NEUTRON TRANSMISSION TECHNIQUE

F. PUTZ, T. AL-KHAFAJI AND P. WEINZIEL
ÖSTERREICHISCHE STUDIENGESELLSCHAFT FÜR ATOMENERGIE, GES. m. b. H., VIENNA, AUSTRIA

Abstract — Résumé — Аннотация — Resumen

DETERMINATION OF PLUTONIUM-239 CONTENT IN NUCLEAR FUEL BY NEUTRON TRANSMISSION TECHNIQUE. In the present paper a procedure for the determination of the plutonium-239 content in nuclear fuel is discussed. The suggested method takes advantage of the fact that a neutron beam passing through a sample containing an appreciable amount of plutonium-239 is highly attenuated in the energy region which is at the lowest resonance at 0.3 eV. In this energy region the total microscopic cross-section of plutonium-239 may be assumed to surpass the total microscopic cross-sections of the other materials contained in the sample, e.g. in an irradiated fuel element. Therefore, the sensitivity of the neutron transmission to changes of the plutonium-239 concentration is increased in the energy range around 0.3 eV. But in discriminating plutonium-239 from the other constituents of irradiated fuel one encounters difficulties because the concentrations of the depleted fuel and the fission products are unknown in general. Therefore, some simplification is necessary. For the calculation of the attenuation of the neutron beam one can, for instance, try to replace all the materials but plutonium-239 by a single fictitious absorber. For the total cross-section of this absorber a reasonable energy dependence, e.g. 1/v-behaviour, is assumed (the validity of this approximation must be examined by experiments). In this way the problem is reduced to the determination of two constants. This determination can be made by means of two transmission measurements; one of these is performed with well-thermalized neutrons, the other one with neutrons of energies covering a small range in the area of 0.3 eV. In both cases the neutron attenuation is calculated by numerical integration as a function of the two unknown constants. The latter ones can be determined by comparison with the results of the measurements.

DOSAGE DU PLUTONIUM-239 DANS DU COMBUSTIBLE NUCLEAIRE PAR LA METHODE DE LA TRANSMISSION DES NEUTRONS. Le mémoire expose une méthode de dosage du plutonium-239 dans le combustible nucléaire. Cette méthode exploite le fait qu'un faisceau de neutrons traversant un échantillon contenant une quantité appreciable de plutonium-239 est fortement atténué dans la région d'énergie correspondant au niveau de résonance le plus bas, soit 0,3 eV. Dans cette région, on peut considérer que la somme des sections efficaces microscopiques du plutonium-239 est supérieure à celle des sections efficaces microscopiques des autres matières contenues dans l'échantillon, en l'occurrence l'élément combustible iradié. En conséquence, les variations de la concentration de plutonium-239 influent davantage sur la transmission des neutrons dans la bande d'énergie de 0,3 eV. Toutefois, pour «separer» le plutonium-239 des autres composants du combustible iradié, on se heurte à des difficultés, car les concentrations du combustible appauvri et des produits de fission ne sont généralement pas connues. Il faut donc simplifier le problème. Pour calculer l'atténuation du faisceau de neutrons, on peut essayer, par exemple, de remplacer toutes les matières - à l'exception du plutonium-239 - par un seul absorbeur fictif. Pour la section efficace totale de cet absorbeur, il faut admettre un certain rapport avec l'énergie, par exemple comportement en 1/v (la validité de cette approximation doit être contrôlée par l'expérience). Le problème se réduit ainsi à la détermination de deux constantes. Cette détermination peut se faire à l'aide de deux mesures de la transmission; l'une est effectuée avec des neutrons amenés à l'état thermique, l'autre avec des neutrons d'une énergie située dans une bande étroite aux alentours de 0,3 eV. Dans les deux cas, l'atténuation du faisceau est calculée par intégration numérique en fonction des deux constantes inconnues. Ces dernières peuvent être déterminées par comparaison avec les résultats des mesures.

ОПРЕДЕЛЕНИЕ СОДЕРЖАНИЯ ПЛУТОНИЯ-239 В ЯДЕРНОМ ТОПЛИВЕ МЕТОДОМ ПРОПУСКАНИЯ НЕЙТРОНОВ. Обсуждается метод определения содержания плутония-239 в ядерном топливе. В предлагаемом методе используется то явление, что пучок нейтронов, проходя через образец, в котором содержится значительное количество плутония-239, сильно ослабляется в диапазоне энергии, который лежит на уровне наименьшего резонанса при энергии 0,3 эв. В этом диапазоне энергии можно предположить, что полное микроскопическое
DETERMINACIÓN POR TRANSMISIÓN NEUTRONICA DEL PLUTONIO-239 CONTENIDO EN COMBUSTIBLES NUCLEARES. Los autores describen un procedimiento para determinar el plutonio-239 contenido en combustibles nucleares. Este procedimiento está basado en el hecho de que, al atravesar una muestra de material que contenga cantidades apreciables de plutonio-239, un haz neutrónico sufre una atenuación muy marcada en la región energética de menor resonancia a 0,3 eV. Se puede suponer que en esa región la sección eficaz microscópica total del plutonio-239 es mayor que las de todos los demás elementos contenidos en la muestra (un elemento combustible irradiado, por ejemplo). Por tanto, la sensibilidad de la transmisión neutónica a las variaciones de la concentración del plutonio-239 aumenta en el intervalo energético situado alrededor de 0,3 eV. Pero cuando se trata de distinguir el plutonio-239 de los demás constituyentes del combustible irradiado se tropieza con dificultades porque, en general, se desconocen las concentraciones del combustible agotado y de los productos de fisión. Es necesario, pues, simplificar un poco. Por ejemplo: para calcular la atenuación del haz neutrónico se puede tratar de sustituir todos los materiales menos el plutonio-239 por un solo absorbente ficticio. Se supone que la sección eficaz total de ese absorbente depende razonablemente de la energía; que obedece, por ejemplo, a la ley 1/v (la validez de esta aproximación debe comprobarse experimentalmente); Así, el problema se reduce a la determinación de dos constantes, lo que puede efectuarse con dos mediciones de la transmisión: una mediante neutrones bien termalizados, y la otra con neutrones cuyas energías cubran un pequeño intervalo alrededor de 0,3 eV. La atenuación neutónica se calcula en ambos casos por integración numérica en función de las dos constantes desconocidas. Estas últimas pueden determinarse por comparación con los resultados de las mediciones.

INTRODUCTION

In some cases it would be very desirable to obtain information on the 239Pu content of irradiated fuel elements, especially of the natural uranium or low enrichment type. Typical cases where information on 239Pu content is of interest are:

(a) At the entrance of a reprocessing line a check of the 239Pu content of the elements to be treated would be very useful.

(b) An inexpensive determination of 239Pu content would be helpful to decide whether or not the further use of irradiated fuel elements would be practical.

(c) A determination of the average conversion ratio in this type of fuel would be very important for the economy of this fuel cycle. From a knowledge of the original content of 235U, the number of total fissions that occurred in the elements (obtained e.g. from a γ-spectroscopic 137Cs determination) and the 239Pu content, this average conversion factor can be deduced in principle.
Conventional techniques to determine the \(^{239}\text{Pu}\) content (radiochemistry and mass analysis) are destructive and very costly, and therefore it is worthwhile to look for a simple non-destructive procedure.

The utilization of neutron transmission measurement seems to be one possible way for the discrimination of \(^{239}\text{Pu}\). The absorption cross-section of \(^{239}\text{Pu}\) shows a marked resonance in the epithermal energy region, the peak being at 0.296 eV (Fig. 1). Neutrons that pass through a sample containing an appreciable amount of \(^{239}\text{Pu}\) are strongly attenuated in the resonance region because the high cross-section increases the sensitivity of the neutron attenuation to changes of the \(^{239}\text{Pu}\) concentration. How to take advantage of this fact in determining the \(^{239}\text{Pu}\) content in nuclear fuel will be the subject of the following preliminary considerations.

**BASIC PRINCIPLE**

Suppose a slab containing \(^{239}\text{Pu}\) is placed in a well-collimated beam of neutrons (Fig. 2). The intensity of the neutrons is measured by a suitable detector after passing through the sample. For the further derivations the detector is assumed to be a \(^{239}\text{Pu}\) fission chamber. Its response to incident neutrons, the energy of which lies between \(E_1\) and \(E_2\), is expressed by

\[
R = A \int_{E_1}^{E_2} \phi(x_D, E)\sigma_f^{49}(E)dE
\]

(1)

where \(\phi(x_D, E)\) is the energy-dependent flux at the position \(x_D\) of the detector and \(\sigma_f^{49}(E)\) is the microscopic fission cross-section of \(^{239}\text{Pu}\). The proportionality factor \(A\) depends on the parameters of the detector. For the flux \(\phi(x_D, E)\) we find

\[
\phi(x_D, E) = \phi_0(E) \times \exp \left[ -\int N^{49}(x)\sigma_f^{49}(E) + \sum_i N^i(x)\sigma_i^f(E) \right] dE
\]

(2)

The symbols used in (2) have the following meaning:

- \(\phi_0(E)\): unperturbed flux of the incident neutrons on the surface of the sample at \(x = 0\)
- \(N^{49}(x)\): number of \(^{239}\text{Pu}\) atoms per unit volume
- \(N^i(x)\): number of atoms of the \(i\)-th kind contained in the sample per unit volume
- \(\sigma_f^{49}(E)\): total microscopic cross-section of \(^{239}\text{Pu}\)
- \(\sigma_i^f(E)\): total microscopic cross-section of the \(i\)-th kind of atoms.

In deriving formula (2) the assumption is made that a collision within the sample leads to the removal of the neutron from the beam. If we introduce the abbreviations

\[
C_f^{49}(t) = \int_0^t N^{49}(x)dx, \quad C_i^f = \int_0^t N^i(x)dx
\]

(3)
(2) can be rewritten as

$$\phi(x_0, E) = \phi_0(E) \exp \left\{ - \left[ C^{49} \sigma^{49}_1(E) + \sum_i C^i \sigma^i_1(E) \right] \right\}$$

The quantities $C^{49}$ and $C^i$ are clearly the total numbers of the corresponding kind of nuclei that are within a cylinder having a unit cross-section and a length $t$ parallel to the $x$-direction. Substitution for $\phi(x_0, E)$ in (1) then gives

$$R = AJ$$

where

$$J = \int_{E_1}^{E_2} \phi_0(E) \exp \left\{ - \left[ C^{49} \sigma^{49}_1(E) + \sum_i C^i \sigma^i_1(E) \right] \right\} \sigma^{49}_1(E) dE$$
For the response $R_0$ of the detector to the unperturbed neutron beam we have

$$R_0 = A \int_{E_1}^{E_2} \phi_0(E) \sigma^{49}_f(E) dE = AJ_0$$

and thus the transmission $T = R/R_0$ can be written

$$T = \frac{J}{J_0}$$

If the spectrum of the neutron beam and the concentrations of all the materials contained in the sample except that of $^{239}$Pu are known, the transmission $T$ may be calculated as a function of $C^{49}$ by numerical computation of $J$ and $J_0$. Then for a measured value of $T$ the corresponding value of $C^{49}$ may easily be determined. In practice the situation is more complex since in an irradiated fuel, for instance, the concentration of the fission products are unknown in general. In this case an approximation must be made. One possible way to simplify the problem is to assume that all the materials together (except $^{239}$Pu) give rise to the same effect as a single fictitious $1/\nu$-absorber. This involves the replacement of the term $\Sigma C\sigma_i(E)$ by $C^f/\sqrt{E}$ so that formula (6) takes the simple form

$$J = \int_{E_1}^{E_2} \phi_0(E) \exp \left\{ - \left[ C^{49}\sigma^{49}_f(E) + C^f/\sqrt{E} \right] \right\} \sigma^{49}_f(E) dE$$
Preliminary calculations show that the sensitivity of the transmission $T$ to relative changes $\Delta C^{49}/C^{49}$ is not very high for low values of $C^{49}$ but rises with increasing concentration of $^{239}$Pu, the more rapidly the smaller the energy interval $(E_1, E_2)$, which includes the resonance peak at 0.3 eV. This latter condition may be satisfied by means of time-of-flight techniques or proper filters.

Since the transmission $T$ now depends on two variables, $C^{49}$ and $C^f$, an additional measurement is needed to be able to determine the $^{239}$Pu content of the sample. It is promising to perform this second transmission measurement with well-thermalized neutrons having a Maxwellian energy distribution. In this region the absorption cross-section of $^{239}$Pu also shows almost a $1/\nu$-behaviour and therefore the dependence of the transmission on $C^{49}$ and $C^f$ will be quite different from the dependence in the resonance region. In low-enriched fuel the total cross-section for thermal neutrons is expected to deviate from the $1/\nu$-behaviour, so that the replacement of all the materials other than $^{239}$Pu by a $1/\nu$-absorber seems doubtful. In these cases another possible approach to the problem is the use of time-of-flight technique. Since this technique enables measurements to be performed with nearly mono-energetic neutrons the formulas (6) and (7) for $J$ and $J_0$ will reduce to simple expressions without integration over the energy. This makes it possible to introduce a fictitious capture that does not vary as $1/\nu$ but is characterized by a function $f(\sqrt{\nu})$ depending on $N$ unknown constants $C_1 \ldots C_N$. The $N+1$ constants $C^{49}, C_1 \ldots C_N$ may be calculated from the results of transmission measurements at $N+1$ points within an energy range that includes the $^{239}$Pu resonance at 0.296 eV.

NUMERICAL RESULTS

The two integrals $J_0$ and $J$ were evaluated numerically using the IBM-7040 computer at the Technical University, Vienna. The results of these calculations are shown in Figs. 3 and 4.

The curves of Fig. 3 represent the transmission $T_{th}$ for thermal neutrons as a function of $C^{49}$ and $C^f$. The neutron spectrum was assumed to be a Maxwellian one and the limits of the integrals was set at $E_1 = 0.001$ eV and at $E_2 = 0.5$ eV.

Figure 4 shows the transmission $T_e$ in the epithermal energy range as a function of the same variables. In these calculations a $1/E$-flux was assumed and the limits $E_1$ and $E_2$ were equal to 0.1 eV and 0.5 eV respectively. Neutrons with energies below and above the $^{239}$Pu resonance at about 0.3 eV had to be cut off, and therefore the use of a samarium filter (0.05 cm thickness) and an indium filter (0.15 cm thickness) was assumed. These two materials are well suited for this purpose, as can be seen in Fig. 1 where their total microscopic cross-sections are shown.

If two measured values $T_{th}$ and $T_e$ are given, the corresponding solutions of $C^{49}$ and $C^f$ may easily be determined from the diagrams of Figs. 3 and 4 by graphical methods.

FINAL REMARKS

The procedure described in the preceding sections represents a simple tool allowing the determination of $^{239}$Pu content in nuclear fuel without de-
stroying the sample. A definite answer, however, to the question whether or not the method described here may be practical cannot be given before some further investigations have been made. The next step should be to justify the approximations made in deriving the formulas by comparison with experiments. In addition, the influence of 241Pu must be studied. Unfortunately, 241Pu shows a resonance of the absorption cross-section (even if it is not very high) almost at the same energy as 239Pu.

It should be mentioned that a detailed mathematical treatment of the general case (arbitrary shape and inhomogeneous distribution of the materials) shows that it is impossible to give an expression for T that is a function of $C^{49} = \int N^{49}(x, y, z)dx dy dz$. The neutron beam, therefore, must be sufficiently small to neglect any spatial dependence on the co-ordinates perpendicular to the beam direction. The change of the neutron spectrum because of these two filters was determined exactly.

Finally, the expected sensitivity of the transmission measurement in the resonance region of 239Pu at 0.296 eV should be demonstrated by a rough estimate. The fractional change of the transmission due to the change of $C^{49}$ is given by

$$\frac{\Delta T}{T} = -\bar{\sigma}^{49} \cdot \Delta C^{49}$$

where $\bar{\sigma}^{49}$ is the average value of the total cross-section of 239Pu within the energy range under consideration. From a 239Pu content of $C^{49} \approx 10^{21}$/cm$^2$, which corresponds to an enrichment of 1%, and a thickness of the sample
of $t = 2 \text{ cm}$, one gets for $\Delta C^{49}$ corresponding to a fractional change of 1%: $\Delta C^{49} \approx 10^{19}/\text{cm}^2$. With $\sigma_t^{49} \approx 3 \times 10^{-21} \text{ cm}^2$ it follows $\frac{\Delta T}{T} \approx -3\%$, which is quite promising for the experimental realization of the procedure suggested in this paper.

**DISCUSSION**

J. T. BYRNE: Have you made any estimate of the precision with which neutron transmission differences might be measured with this method?

P. WEINZIERS: As an experimental physicist I would not care to make any statement on the question of error at the present stage when there are still so many problems to be solved. I have already drawn attention to the estimate of sensitivity, which is promising.
OPERATING EXPERIENCE WITH THE SEMICONDUCTOR GAMMA-COMPTON SPECTROMETER TO DETERMINE THE BURN-UP AND BURN-UP HISTORY OF NUCLEAR FUEL

M.J. HIGATSBERGER, H. HICK, K. RUMPOLD, P. WEINZIERL AND A. BURTSCHER
OSTERREICHISCHE STUDIENGESELLSCHAFT FUR ATOMENERGIE,
Ges. m. b. H., VIENNA, AUSTRIA

Abstract — Résumé — Аннотация — Resumen

OPERATING EXPERIENCE WITH THE SEMICONDUCTOR GAMMA-COMPTON SPECTROMETER TO DETERMINE THE BURN-UP AND BURN-UP HISTORY OF NUCLEAR FUELS. The quantitative reliability of non-destructive burn-up determination by means of gamma-spectroscopy using a semiconductor-detector in a Compton spectrometer has been tested. An MTR-type fuel element was irradiated under known conditions with inserted monitor wires. Neutron-physical evaluation of these monitors led to a burn-up value of 37.6 mg ± 7% uranium-235 for this fuel element. Non-destructive burn-up determination with the Compton spectrometer yielded 36.1 mg ± 15% uranium-235. A rough calculation of the reactor operating group based on thermal reactor power measurements and core position weight factors resulted in a burn-up value of 40 mg ± 30% uranium-235.

During a routine run of one week a highly irradiated fuel plate cut into 18 separate pieces was measured. A comparison of these results with radiochemical and mass analytical burn-up determination carried out later will be presented. A test run covering 18 irradiated MTR-type fuel elements required another week of routine operation of the Compton spectrometer.

By comparing relative intensities of γ-energies related to fission products of different half-lives further information on burn-up history of the fuel under investigation can be extracted.

RESULTATS OBTENUS AVEC LE SPECTROMÈTRE COMPTON À SEMI-CONDUCTEURS UTILISE POUR DÉTERMINER LE TAUX ET LE PROCESSUS DE COMBUSTION DES COMBUSTIBLES NUCLEAIRES. On a utilisé un spectromètre Compton pourvu d'un détecteur à semi-conducteurs pour déterminer sans destruction, par spectroscopie gamma, la sûreté du point de vue quantitatif des calculs du taux de combustion. On a étudié la combustion, dans des conditions déterminées, d'un élément combustible MTR dans lequel on avait inséré des fils de contrôle. L'analyse physique de ces derniers par les neutrons a permis d'évaluer à 37,6 mg ± 7% d'uranium-235 le taux de combustion de l'élément. En procédant à une analyse non destructive avec un spectromètre Compton, on a obtenu un taux de combustion de 36,1 mg ± 15% d'uranium-235. D'après un calcul approximatif effectué par les opérateurs du réacteur à partir des mesures de la puissance thermique du réacteur et des coefficients de pondération selon la position des éléments dans le cœur, le taux de combustion obtenu est de 40 mg ± 30% d'uranium-235.

Au cours d'une série d'expériences effectuées pendant une semaine, on a mesuré le taux de combustion d'une plaque de combustible fortement irradiée découpée en 18 morceaux. Les auteurs comparent ces résultats à ceux d'évaluations ultérieures du taux de combustion par la radiochimie et la spectrographie de masse. Le spectromètre a fonctionné constamment pendant une autre semaine pour permettre d'effectuer une série d'essais portant sur 18 éléments combustibles MTR irradiés.

En comparant l'intensité relative du rayonnement gamma des produits de fission de périodes différentes, on peut obtenir des renseignements complémentaires sur le processus de combustion des combustibles étudiés.

ОПЫТ ЭКСПЛУАТАЦИИ ПОЛУПРОВОДНИКОВОГО КОМПТОН-СПЕКТРОМЕТРА ПРИ ОПРЕДЕЛЕНИИ СТЕПЕНИ ВЫГОРАНИЯ И ПРОЦЕССА ВЫГОРАНИЯ В ЯДЕРНЫХ ТОПЛИВНЫХ ЭЛЕМЕНТАХ. Проверена надежность в количественном отношении метода определения степени выгорания с помощью гамма-спектроскопии при использовании полупроводникового детектора в Комптон-спектрометре без разрушения материала. Топливный элемент типа МТР облучали при заданных условиях с введенными прутковыми мониторами. Нейтронно-физическая оценка этих мониторов привела к величине степени выгорания в размере 37,6 мг ± 7%
HIGATSBERGER et al.

uran-235 for this fuel element. In determining the degree of burning without destroying the material with the help of the Compton-spectrometer, a value of 36.1 mg ± 15% of uran-235. With a first approximation, performed by the reactor group, based on measurements of the nuclear reactor's power and weight coefficients of the active zone, a value of burning 40 mg ± 30% of uran-235.

During normal operation for one week a highly irradiated fuel plate was cut into 18 separate pieces. Will be presented the results of comparison of these results with the results of the radiochemical and mass-analytical determination of the degree of burning. For the check of 18 irradiated fuel elements of type MTR was another week of normal operation of the Compton-spectrometer.

By comparing the relative intensities of the energy of gamma-rays, associated with the products of the fission with different half-lives, you can get more information about the degree of burning and the process of depletion of the fuels studied.

INTRODUCTION

Gamma radiation emitted from fission products of spent nuclear fuel contains much information that can be elicited without destroying the fuel elements. But the extraction of such information from the complex spectrum is difficult and requires a high resolution gamma-spectrometer. Because of the lack of a high-resolution instrument suitable for routine use in an industrial nuclear installation, only small fraction of the available information has been extracted in earlier work. We have developed a new type of gamma-spectrometer, using a lithium-drifted silicon detector in a Compton-coincidence arrangement. An operating instrument was demonstrated at the Governmental Scientific Exhibition of the 3rd UN International Conference on the Peaceful Uses of Atomic Energy (September 1964). This paper reviews some measured data obtained with the prototype semiconductor Compton-spectrometer in comparison with destructive analytical work.

INFORMATION CONTAINED IN THE GAMMA-SPECTRUM

During irradiation of a fuel element many fission products of different half-lives are formed with known fission yields. Their characteristic
gamma-spectra (intensities and energies of lines) are also known as well as their half-lives. By suitable analysis of the complex gamma-spectrum of a fuel element (measured with a suitable spectrometer) the absolute and relative isotopic fission products can be distinguished.

In nuclear fission the number of fission isotopes produced is determined by

$$\frac{dN_i}{dt} = \gamma_i \Sigma_f \phi + \sum_j \gamma_{ij} N_j + \sum_k \epsilon_{ik} N_k \phi - \lambda_i N_i - \sigma_i N_i \phi$$

where $\gamma_i$ stands for fission yield of $N_i$, $\Sigma_f$ for the macroscopic cross-section, $\phi$ for the thermal neutron flux, $\epsilon_{ik} N_k$ for the number of atoms produced from the $k$ isotope. The terms $\lambda_i N_i$ and $\sigma_i N_i \phi$ represent decay of the $i$ isotope and the isotope lost by neutron capture respectively. The better the spectrometer the more fission products may be determined quantitatively and the more independent equations are obtained.

**Total burn-up**

The information of primary interest is the total burn-up. The burn-up definition used here is the total number of fissions that have occurred in a fuel element. This information may be obtained by measuring the absolute abundance of only a single radioactive isotope. But for the evaluation it is necessary to know the time factor governing production and decay of the measured fission product (i.e. the reactor operation history). If the reactor operation history is not known, the determination of burn-up may become involved. Even for a given irradiation history considerable errors may be caused by the varying flux-distribution caused by core poisoning, decreasing fuel density, plutonium production, changed core configurations, etc.

Only if a fission product is chosen whose half-life exceeds the usual irradiation and cooling period by an order of magnitude, then without any knowledge of the reactor operation history the time factor can be estimated with sufficient accuracy.

Only one $\gamma$-emitting fission product is known that meets these requirements: $^{137}$Cs with $\sim 30$ yr half-life and a single $\gamma$-peak at 661 keV. The measurement of this line is difficult because of the neighbourhood of much stronger $\gamma$-lines (zirconium-niobium-95), unless excessive cooling times are acceptable. Our spectrometer has sufficient resolution to measure the amount of caesium-137 after a few weeks cooling only.

Spectrometers of lower resolution (as, for instance, NaI spectrometers) allow only a burn-up determination based on a measurement of isotopes such as

- cerium-praseodymium-144 (284 d, 2190 keV) or
- zirconium-niobium-95 (65 + 35 d, 728 - 759 + 768 keV) or
- barium-lanthanum-140 (12.8 d, 159 + 643 + 1597 keV).

The above-mentioned difficulties regarding the time factor uncertainty apply to all these isotopes.
IRRADIATION HISTORY

If a set of fission products of different half-lives has been measured, a set of equations is obtained that enables a check to be made whether an assumed irradiation history is compatible with the experimental results or not. Usually a compatible group of irradiation histories may be sufficiently narrowed by physical requirements to fit the mathematical solutions such as limits on irradiation time, reactor power etc.

The evaluation normally requires a computer programme especially for a treatment of error propagation. So far only a few cases have been dealt with. It is advisable to choose fission products of considerably varying half-lives. The unambiguousness is achieved in complicated cases, if measurements are taken at time intervals allowing a marked decay of the shorter-lived isotopes.

By measuring the relative amounts of fission products, information on fission product release may be obtained. If the spatial distribution of selected fission products is scanned, leaks, voids and cracks may be located in the fuel element. Also information on the physico-chemical behaviour of the fission products during irradiation may be inferred from such measurements inaccessible by other means.

SEMICONDUCTOR COMPTON-SPECTROMETER

Principle and construction

The principle and construction of our instrument has been described in detail elsewhere [1, 2]. Figure 1 shows the Geneva Demonstration Prototype Unit that uses a vacuum pump, liquid nitrogen cooling and conventional versatile electronics. By simple technical improvements (sealed vacuum system, thermo-electric cooling to -30°C, special electronic systems) the size is reduced considerably and a transportable instrument may be built either as a battery and/or plug-in a.c.-operated unit. Figure 2 shows the principal features of the instrument (1, fuel element; 2, 4, 5, collimator; 3, Al-tank; 6, semiconductor; 7, NaI crystal; 8, photomultiplier; 9-16, accessories and framework). Figure 3 shows a scanning mechanism adapted to measure MTR fuel elements in the Seibersdorf ASTRA reactor [3].

Spectrometer performances

A gamma-spectrometer is characterized by its efficiency, its energy resolution and its response function. The efficiency of the spectrometer had to be made as low as $5 \times 10^{-10}$ in order not to exceed the useful coincident count rate of 50 counts/s. The variation of efficiency with energy is low, which is very convenient for analysis of widespread spectra. Figure 4 shows a bromine-82 spectrum that was used to determine the variation of efficiency with energy.

The energy resolution is 13 keV full width at half maximum, rather independent of energy and should be improvable to <10 keV by more careful
FIG. 1. Coincidence Compton spectrometer for the determination of burn-up and burn-up history in nuclear fuels (demonstration model for the 3rd UN International Conference on the Peaceful Uses of Atomic Energy, in Geneva, 1958)

FIG. 2. Sketch of the spectrometer head
The response function is almost ideal: a nearly gaussian peak, followed by a low tail. Figure 5 shows a caesium-137 spectrum. A figure of merit for the quality of a response function is the gaussian peak fraction, i.e. the area under the gaussian peak divided by the integral over
the whole pulse-height distribution. The semiconductor Compton-spectrometer has the highest fraction of all concurring spectrometers and is therefore best suited for the analysis of complex spectra. The area below a single caesium-137 peak has been found to be reproducible within statistically determined limits of 1.3\% for measurements within 4 h without changing electronic settings. Reproducibility is probably still better, but this has not yet been investigated.

During a non-stop run of 4 d a similar reproducibility could be obtained, but this does not occur regularly. Peak drifts were ±1 channel with 2.2 keV/channel using a TMC-256 multichannel analyser.

OPERATION EXPERIENCE

Test on quantitative reliability [4]

After the instrument was completed the first and main aim was to prove the quantitative reliability of a burn-up determination on an actual fuel element. Already during December 1963 an unirradiated MTR-type partial fuel element of the ASTRA reactor was prepared with 17 monitor wires, and irradiated. After irradiation the monitor wires were removed and their activity was measured by means of a wire-scanner. Evaluation by neutron-physical methods yielded the total neutron flux-dose distribution, and by further evaluation the number of fissions that had occurred within the fuel element was obtained. The quantity of $^{235}$U consumed was found to be 36.1 mg ± 7\% (σ).

After a year of cooling (needed because the irradiation time was only one day) the fuel element was measured with the Compton-spectrometer. To
avoid inadmissible high activities of the monitor-wires, the burn-up of the fuel element was limited to a very low value (36.1 mg \(^{235}\text{U}\) consumed; total loading 95.61 g \(^{235}\text{U}\)). Therefore the count-rate achieved with the Compton-spectrometer was so low that a total measuring time of 12 h was needed to obtain sufficient statistics. Frequent calibration runs were done in between to eliminate the possible influence of long-term drifts.

The measured spectrum is shown in Fig. 6. The height of the caesium-137 peak was compared with the peak height obtained by a calibration run with a calibrated standard source. In this way the amount of caesium-137 in the fuel element could be determined, and corresponds to a consumed \(^{235}\text{U}\) amount of 37.4 mg. The estimated error is \(\sigma \approx 10\%\). The good agreement shown in Table I shows that the estimated error of the non-destructive burn-up determination may be a rather pessimistic assumption. It seems proved that the spectrometer is also able to determine accurately the total burn-up for extremely low active probes, i.e. for very small burn-up values.

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{spectrum.png}
\caption{Y-spectrum of a calibrated element (partial MTR element)}
\end{figure}

**Measurements on an irradiated BGRR fuel plate**

During a service contract\(^1\) with the International Atomic Energy Agency, an irradiated BGRR fuel plate cut into 18 pieces had to be measured by the Compton-spectrometer non-destructively, and then some of the parts were

\(^1\) IAEA Service Contract SC 810 (1) 1964.
TABLE I
AMOUNT OF $^{235}$U CONSUMED

<table>
<thead>
<tr>
<th>Neutron physical result</th>
<th>Compton spectrometer result</th>
</tr>
</thead>
<tbody>
<tr>
<td>36.1 mg</td>
<td>37.4 mg</td>
</tr>
<tr>
<td>$\sigma \approx 7%$</td>
<td>$\sigma \approx 10%$ (estimated)</td>
</tr>
</tbody>
</table>

FIG. 7. Sum spectrum of MTR fuel plate (total of 18 pieces)

analysed destructively by chemical and mass spectrometric methods. In this way an even more stringent control than the one described previously was possible.

The arrangement shown in Fig. 3 was used for the Compton-spectrometer measurements. To avoid contamination of the pool water, each fuel piece had to be enclosed in a tight plastic bag. The remote insertion of these air-filled bags into the special scanning mechanism under water was more difficult than scanning a full element. The sum spectrum of all 18 pieces is shown in Fig. 7. It can be observed from the relative strengths of the
zirconium-niobium-95 peaks and the caesium-137 peak that the cooling time of these samples is about one year.

Table II shows the various results and their comparison. Column 1 shows the fragment number, columns 2-4 give the Compton-spectrometer results. The error due to counting statistics is between 3.5 and 6.5% (σ). Column 5 is the total uranium content as determined by chemical analysis (σ~1.5%). Columns 6 and 7 are the $^{235}\text{U}$ and $^{236}\text{U}$ abundance as obtained by mass spectroscopy (σ~1%). By multiplying columns 5 and 6 one obtains the amount of $^{235}\text{U}$ remaining (column 9) and by combining this with the amount of $^{235}\text{U}$ consumed (column 4) one can calculate the percentage of $^{235}\text{U}$ depletion (column 10). When the initial isotopic fuel composition (93.7% $^{235}\text{U}$, 0% $^{236}\text{U}$) is known one can calculate the $^{235}\text{U}$ depletion based on the mass spectrometric measurements. The result is shown in column 12 (σ~1%). Comparison of columns 8 and 9 gives the error of the Compton-spectrometer results, because the other errors involved are almost negligible.

The largest deviation of 11.6% is found for pieces 3 and 4, which are endpieces of the original fuel plate. The mean deviation is 6.4%. The deviation of the mean values, however, is only 0.5%.

The comparison proves two things:
(a) While the mean deviation between the Compton-spectrometer results and the chemical and mass analytical determination for the single pieces amounts to σ = 6.4%, the mean values for the entire fuel plate agrees within 0.5%. This indicates that the deviations in the results for the single pieces are largely due to caesium-137 migration.
(b) For highly active fuel also the Compton-spectrometer allows quick and accurate non-destructive burn-up determination, but the amount of caesium-migration causes in the fuel type under investigation a mean error of σ = 6.4% for determining local burn-up by caesium-137 measurements.

Further experience

During the 1964 Christmas shut-down of the ASTRA reactor the opportunity for the total core to be available was used for measurements after 3 weeks cooling. Because of the short cooling time and a non-optimal improvised collimator geometry, the peak-to-background ratio was low and 2.5 h measuring time per fuel element had to be taken. The total core of 16 standard elements was measured within 8 d. Because of accidental distortion of the collimator geometry before the calibration run, evaluation of data and comparison with the reactor log-book calculations were not yet possible.

CONCLUSION

A simple non-destructive instrument to determine burn-up and burn-up history of nuclear fuel is highly desirable for the economic operation of nuclear reactors. It could serve both fuel manufacturers and users. Such an instrument would greatly facilitate objective inventory control. Since 1962 a semiconductor Compton spectrometer has been developed and cons-
### TABLE II

COMPARISON OF RESULTS

<table>
<thead>
<tr>
<th>No.</th>
<th>Compton spectrometry</th>
<th>Chemical analysis</th>
<th>Mass spectrometry</th>
<th>Measured data only</th>
<th>Unchecked input data</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>( ^{235}U )</td>
<td>( ^{238}U )</td>
<td>( ^{239}U )</td>
<td>( ^{233}U )</td>
<td>( ^{239}U )</td>
</tr>
<tr>
<td></td>
<td>fissioned (mg)</td>
<td>converted (mg)</td>
<td>consumed (mg)</td>
<td>total (mg)</td>
<td>remaining (mg)</td>
</tr>
<tr>
<td>1</td>
<td>108.0</td>
<td>18.4</td>
<td>126.4</td>
<td>289.9</td>
<td>82.0</td>
</tr>
<tr>
<td>2</td>
<td>115.8</td>
<td>15.3</td>
<td>133.1</td>
<td>292.9</td>
<td>84.0</td>
</tr>
<tr>
<td>3</td>
<td>96.5</td>
<td>16.7</td>
<td>115.2</td>
<td>232.9</td>
<td>78.0</td>
</tr>
<tr>
<td>3 + 4</td>
<td>187.4</td>
<td>31.8</td>
<td>219.2</td>
<td>496.9</td>
<td>88.0</td>
</tr>
<tr>
<td>4</td>
<td>88.9</td>
<td>15.1</td>
<td>104.0</td>
<td>233.9</td>
<td>78.0</td>
</tr>
<tr>
<td>5</td>
<td>122.2</td>
<td>29.8</td>
<td>152.0</td>
<td>354.9</td>
<td>88.0</td>
</tr>
<tr>
<td>6</td>
<td>98.1</td>
<td>16.7</td>
<td>114.8</td>
<td>234.9</td>
<td>88.0</td>
</tr>
<tr>
<td>7</td>
<td>83.2</td>
<td>14.1</td>
<td>97.3</td>
<td>234.9</td>
<td>78.0</td>
</tr>
<tr>
<td>8</td>
<td>158.2</td>
<td>21.5</td>
<td>179.7</td>
<td>369.9</td>
<td>88.0</td>
</tr>
<tr>
<td>9</td>
<td>103.9</td>
<td>17.7</td>
<td>121.6</td>
<td>239.9</td>
<td>88.0</td>
</tr>
<tr>
<td>10</td>
<td>127.1</td>
<td>15.9</td>
<td>143.0</td>
<td>239.9</td>
<td>88.0</td>
</tr>
<tr>
<td>11</td>
<td>109.0</td>
<td>16.5</td>
<td>125.5</td>
<td>239.9</td>
<td>88.0</td>
</tr>
<tr>
<td>12</td>
<td>160.6</td>
<td>27.3</td>
<td>187.9</td>
<td>363.9</td>
<td>88.0</td>
</tr>
<tr>
<td>13</td>
<td>77.4</td>
<td>13.2</td>
<td>90.6</td>
<td>230.9</td>
<td>88.0</td>
</tr>
<tr>
<td>14</td>
<td>158.2</td>
<td>31.5</td>
<td>189.7</td>
<td>320.9</td>
<td>88.0</td>
</tr>
<tr>
<td>15</td>
<td>119.6</td>
<td>20.9</td>
<td>139.9</td>
<td>319.9</td>
<td>88.0</td>
</tr>
<tr>
<td>16</td>
<td>160.9</td>
<td>17.1</td>
<td>118.0</td>
<td>231.9</td>
<td>88.0</td>
</tr>
<tr>
<td>17</td>
<td>101.9</td>
<td>17.3</td>
<td>119.2</td>
<td>229.9</td>
<td>88.0</td>
</tr>
<tr>
<td>18</td>
<td>20.0</td>
<td>3.6</td>
<td>24.6</td>
<td>47.6</td>
<td>88.0</td>
</tr>
</tbody>
</table>

| Plate | 1730 | 294 | 2924 | 3314.5 🅔 | 82.8 🅔 | 2744 | 42.5 🅔 | 36.3 🅔 | 42.5 🅔 | 36.1 🅔 |

|     | 2    | 3    | 4    | 5    | 6 | 7 | 8 | 9 | 10 | 11 | 12 | 13 |

\( a \) = calculated value based on analysis of randomly selected individual pieces

\( b \) = mean value
constructed at the Seibersdorf reactor Centre, and demonstrated at the 3rd UN International Conference on the Peaceful Uses of Atomic Energy. The instrument underwent stringent tests on fuel samples and fuel elements of extremely low and extremely high burn-up values. The measured data with the spectrometer have been independently checked by destructive chemical and mass-spectroscopic analyses. A neutron physical method also has been used for cross checks. It has been proved that the Compton spectrometer is a powerful tool for non-destructive burn-up determination with an accuracy comparable to any other method. The Compton spectrometer was also superior to the photopeak spectrometer and the anti-Compton spectrometer. With proper mechanical scanning arrangements quick and accurate information is obtainable on burn-up, burn-up history and fuel migration.

REFERENCES


DISCUSSION

N.C. RASMUSSEN: What nuclear data are required in this analysis?
M.J. HIGATSBERGER: The fission yield constant of $^{137}$Cs is the most important item required. We also need the capture-to-fission cross-section ratio and the gamma-absorption constant for the relevant fuel-element geometry.
THE NON-DESTRUCTIVE MEASUREMENT OF BURN-UP BY GAMMA-RAY SPECTROSCOPY*

N. C. RASMUSSEN, J. A. SOVKA AND S. A. MAYMAN**
MASSACHUSETTS INSTITUTE OF TECHNOLOGY, CAMBRIDGE, MASS., UNITED STATES OF AMERICA

Abstract — Résumé — Аннотация — Resumen

THE NON-DESTRUCTIVE MEASUREMENT OF BURN-UP BY GAMMA-RAY SPECTROSCOPY. A method of non-destructive analysis of spent reactor fuel using a lithium-drifted-germanium gamma-ray spectrometer is described. The spent fuel studied was MTR type fully enriched elements from the MIT reactor. Detailed gamma-ray spectra of fuel elements having cooling times between 1½ weeks and 3½ years are reported. The relative intensities of gamma rays from three fission products, caesium-137, caesium-134 and zirconium-95, are given as a function of position within one carefully studied element having a cooling time of nine months. A computer method for using ratios of these gamma-ray intensities to determine average flux, irradiation time and burn-up is described. These results are compared to the known conditions in the MIT reactor and are in good agreement within the ±10% error assigned. A major source of this error arises from uncertainties in the nuclear data used. A brief comparison is made with previous work done at this laboratory using a bent quartz crystal gamma-ray spectrometer.

* The research reported in this paper was sponsored by the Air Force Cambridge Research Laboratories, Office of Aerospace Research, under contract AF19(604)-7492. This work was done in part at the MIT Computation Center.

** Now at Whiteshell Nuclear Research Establishment, Pinawa, Man., Canada.
MEDICION NO DESTRUCTIVA DEL GRADO DE COMBUSTION POR ESPECTROSCOPIA GAMMA. Los autores describen un método de análisis no destructivo del combustible agotado utilizando un espectrómetro gamma con un elemento sensible de litio activado por germanio. El combustible estudiado fue del tipo MTR con elementos totalmente enriquecidos del reactor del Massachusetts Institute of Technology. En la memoria se informa sobre los espectros gamma detallados de elementos combustibles enfriados durante un período, comprendido entre una semana y media y tres años y medio. Las intensidades relativas de los rayos gamma de tres productos de fisión cesio-137, cesio-134, circonio-95 se dan en función de la posición dentro de un elemento cuidadosamente escogido con un tiempo de enfriamiento de nueve meses. Se describe un método de cálculo para utilizar las razones de esas intensidades gamma a fin de determinar el flujo medio, el tiempo de irradiación y el grado de combustión. Esos resultados se comparan con las condiciones que se dan en el reactor del MIT y su error está dentro del límite previsto de ± 10%. La inseguridad en los datos nucleares empleados contribuye significativamente en ese error. Se establece una breve comparación con otros trabajos hechos anteriormente en este laboratorio utilizando un espectrómetro gamma provisto de un cristal de cuarzo curvo.

INTRODUCTION

The possibility of determining information about the irradiation history of spent reactor fuel by gamma-ray spectroscopy has been recognized for some time. A number of laboratories have reported measuring the intensity of the total unresolved gamma-ray spectrum and successfully relating these measurements to such parameters as fuel burn-up and core power distribution [1-3]. However, considerably more information about the performance of the fuel is potentially available if amounts of various fission products can be determined by measuring the intensity of the gamma-rays associated with each of them. This type of analysis requires gamma-ray spectrometers of relatively high energy resolution because of the large number of closely-spaced gamma-rays emitted by the fission products [4-6]. The following paper describes non-destructive studies of spent Massachusetts Institute of Technology reactor (MITR) fuel using two different types of high resolution gamma-ray spectrometers: (a) a bent quartz crystal spectrometer of the Cauchois type, and (b) a lithium-drifted germanium gamma-ray detector.

The purpose of these studies has been to determine what information can be obtained about irradiated fuel with each instrument, and to compare the relative merits of each. The first part of the paper describes experiments using the lithium-drifted germanium detector, the results obtained, and their interpretation, while the second part describes experiments with the bent crystal.

The accuracy of the results is evaluated by comparing them to the known operating conditions in the MITR. A brief discussion of the possible application of this method to spent fuel from other types of reactors is included.
STUDIES WITH A LITHIUM-DRIFTED GERMANIUM GAMMA-RAY SPECTROMETER

Description of experimental apparatus

The Ge(Li) detector used in these experiments was fabricated at this laboratory by techniques similar to those described in the literature [7]. The measured depletion depth was 3.5 mm with an area of 1.6 cm$^2$. The detector was stored and operated at liquid nitrogen temperatures (77°K) using the vacuum dewar shown schematically in Fig. 1. A full charge of 1.35 litres of liquid nitrogen gave typical hold times of ~60 h at a vacuum of 10$^{-6}$ mm Hg maintained by a 1 litre/s ion pump mounted on the dewar.

![Diagram of vacuum chamber and electronics](image)

FIG. 1. Schematic diagram of vacuum chamber and electronics for use with Li-drifted germanium γ-ray detectors

The detector was operated with a reverse bias of between 100 to 200 V. The charge collected was fed to a low noise, charge-sensitive preamplifier (ORTEC Model 103XL or TENNELEC 100). The output of the preamplifier went to a biased amplifier (ORTEC Model 203) and then to a 1024-channel pulse-height analyser (Nuclear Data Model ND160). The noise level of the electronic system as determined with a precision pulser was equivalent to a spread of 3 keV in the energy of the output pulse.

Figures 2 and 3 show the spectra of $^{137}$Cs and Th$^{11}$C obtained with the spectrometer. The energy resolution for the $^{137}$Cs 662-keV gamma-ray was 4.7 keV full width at half maximum (FWHM), and for Th$^{11}$C the FWHM was 8 keV at 2614 keV. The double escape peak shown in the Th$^{11}$C spectrum is the result of pair production interactions in which both annihilation photons escape. Because the detector is small the escape probability is large so this peak is considerably larger than either the total absorption or the single escape peak.

The intrinsic efficiency of the detector as a function of energy is shown in Fig. 4. The points were obtained with calibrated radioactive point sources and a calculated geometry factor.

The fuel element scanning was carried out at the spent fuel storage pool of the MITR, with the experimental arrangement shown in Fig. 5.
element, shown in Fig. 6, was positioned horizontally 6 ft below the surface of the pool. The detector was mounted on a carriage that could be moved either parallel or normal to the axis of the fuel element. An air-filled tube extended from near the bottom of the dewar through the water to the surface of the element. Steel and lead collimators with aperture diameters varying between 1/16 and 1/4 in. could be inserted into the ends of the tube to reduce counting rates to acceptable levels. The scanning tube could be reproducibly positioned with an accuracy of better than ±1/16 in.

**Experimental results**

A number of MITR fuel elements having different irradiation histories and different cooling times have been studied. Figure 7 shows the energy spectra of four elements having cooling times ranging from 1.5 weeks to 3 1/2 yr. The fission product responsible for each of the prominent peaks is labelled. The relative position of the curves to each other is arbitrary, and was chosen only to show how the prominent features of the spectrum change with time.

An enlarged view of the region between 500 keV and 800 keV for an element with 9 months' cooling is shown in Fig. 8. The six fission products that have readily identifiable peaks in this region are $^{134}$Cs, $^{106}$Ru, $^{137}$Cs, $^{144}$Pr, $^{95}$Zr and $^{95}$Nb.

From Fig. 7 it can be seen that most of the peaks in the 600 to 800 keV region cannot be resolved after 1.5 weeks of cooling because the background from short-lived fission products is still too high. By following the decay of an element over a period of about 2 months it was found that a cooling period of 4 to 6 weeks is required before a reasonably accurate determination of the intensities of the fission product peaks in this region is possible.
The actual determination of the number of counts in a peak was done with a computer code by subtracting a suitably averaged background from the total counts under the peak. The accuracy of this method was checked using composite sources containing known ratios of gamma emitters. The measured intensity of a gamma-ray peak superimposed on a background provided by another source was compared to the measured intensity of that gamma-ray without any background present. It was found that the method gave a standard deviation of the intensity that was never more than a factor of two larger than the standard deviation caused only by counting statistics. Since for most prominent peaks the error from counting statistics was 1-2\%, the error in the intensity was less than 5\%.
FIG. 4. Intrinsic full peak efficiency of lithium-drifted germanium detector No. 9-19.1, 3.5 mm thick 200 V bias, 77°K (12 Nov. 1964)

FIG. 5. Schematic diagram of apparatus for gamma-ray scanning of MITR spent-fuel elements
Figure 9 shows the results of a scan made along the length of an element. The relative activity of three fission products, $^{137}$Cs, $^{134}$Cs, $^{95}$Zr, is plotted as a function of position for an element having a cooling time of 9 months.

Figures 10 and 11 show results of transverse scans in a direction perpendicular to the axis of the element. The spatial resolution obtained is shown in Fig. 11, where the large fluctuations in the $^{137}$Cs activity with position are a result of viewing different amounts of a fuel plate as the collimator is moved. Fuel plate dimensions are given in Fig. 6.

Interpretation of results

Fission product activities

The properties of fission products of interest are summarized in Table I. The relations governing their rate of accumulation can be divided into three types. The first type includes fission products that are produced directly in the fission process or are the result of precursors of half lives much
shorter than their own. The rate of accumulation of activity of this type of fission product in fully enriched fuel run at steady power is

$$\frac{d(N_A \lambda_A)}{dt} = N_{25} \alpha_{25}^f \phi \lambda_A Y_A - N_A \lambda_A - N_A \sigma_A \phi \lambda_A$$  \hspace{1cm} (1)$$

where $N_{25}$ is the number of $^{235}$U atoms present

$\alpha_{25}^f, \sigma_{25}$ = fission and absorption cross-section of $^{235}$U

$N_A$ = number of atoms of type A

$Y_A$ = fission yield of A (includes short-lived precursors)

$\phi$ = neutron flux

$\lambda_A$ = radioactive decay constant of A.

The first term on the right hand side represents production by $^{235}$U fission, the second term radioactive decay of A and the third term loss due to neutron capture. In most cases the third term can be neglected. This equation applies to the production of $^{137}$Cs, $^{95}$Zr, $^{140}$Ba, $^{166}$Ru and $^{144}$Ce.
FIG. 8. Gamma-ray spectrum of MITR fuel element 2M19 after 9 months cooling, germanium lithium-drift detector No. 9·19·2, 3.8 mm depletion depth, 1.6 cm² 170 V bias, 77°K run D4 (5/5/65)

$^{144}\text{Pr}$ and $^{140}\text{La}$ also have half lives short enough so that in most cases they have the same activity as their parents.

The second type includes fission products, such as $^{95}\text{Nb}$, which are daughters of the first type that are not in secular equilibrium with their parents. The activity of such nuclides as a function of irradiation time at constant flux is

$$\frac{d(N_B \lambda_B)}{dt} = N_A \lambda_A \lambda_B - N_B \lambda_B^2 - N_B \sigma_B \phi \lambda_B$$  \hspace{1cm} (2)

where subscript $A$ refers to the parent and $N_A \lambda_A$ can be obtained from Eq. (1).

The third type includes radioactive nuclides produced by neutron reactions on the fission products. The only case of interest here is $^{134}\text{Cs}$, which is produced by an $(n, \gamma)$ reaction on the $^{133}\text{Cs}$ that is the stable end product of the mass 133 fission product chain. The rate of accumulation of such a nuclide is given by

$$\frac{d(N_c \lambda_c)}{dt} = N_a \sigma_A \gamma \phi \lambda_c - N_c \lambda_c^2 - N_c \sigma_c \phi \lambda_c$$  \hspace{1cm} (3)
where $\sigma_{A}^{\gamma}$ is the cross-section for the $(n, \gamma)$ reaction and $N_A$ is obtained from Eq.(1), since in this case $\lambda_A = 0$.

Determination of flux and irradiation time

The above equations were solved for the conditions of the MITR. This solution required a knowledge of the mode of operation. In this case the reactor was operated at a constant power for 95 h each week and was shut down for the remaining 73 h. The effect of $^{235}$U burn-up was also included. The values of $\sigma_{25}^{\gamma}$ and $\sigma_{25}^{\gamma}$ were chosen to be consistent with the known neutron spectrum of the MITR. These solutions were used to obtain expressions for $R_1 = \text{activity}^{137}\text{Cs}/\text{activity}^{134}\text{Cs}$ and $R_2 = \text{activity}^{137}\text{Cs}/\text{activity}^{95}\text{Zr}$. $R_1$ and $R_2$ depend upon $\phi$ and $t$ in different ways. A simultaneous solution of the expressions for $R_1$ and $R_2$ gives unique values for $\phi$ and $t$. The expression is transcendental so the solution was done graphically.

The steps in the solution are the following. Plots of $R_1$ and $R_2$ versus $\omega$ (where $\omega = \phi t$) are made for various assumed values of $\phi$. Typical plots are shown in Figs. 12 and 13. The measured values of $R_1$ and $R_2$ are then obtained from Fig. 9 and corrected for decay since removal from the reactor flux. This requires knowing the cooling time, which was obtained from the records. Corrections for detector efficiency and self absorption
FIG. 10. Results of transverse scan showing $^{137}$Cs, $^{95}$Zr ($\gamma$-724 keV) and $^{134}$Cs ($\gamma$-605 keV) activities as a function of position across fuel element 2M19 after 9 months' cooling time.

must also be made. A plot is then made of $\phi$ versus $\omega$ for the experimental values of $R_1$ and $R_2$ and the intersection of these two plots (see Fig. 14) gives unique values of $\phi$ and $\omega$ and hence also $t$.

Although the fraction of time the reactor was operated each week was obtained from the records, it is also possible to verify the value from the experimentally determined ratio of zirconium/niobium activities. In this experiment what was actually measured was the ratio of the activity of the 724-keV zirconium gamma-ray to the combined activities of the 758-keV and 766-keV gamma-rays from zirconium and niobium respectively. The experimental value was 0.143. The value calculated assuming operation at constant power with no shut down was 0.212 and the value calculated assuming operation at constant power but for only 95 h/week was 0.144, in good agreement with the measured value.

The values of the flux as a function of position along the element obtained by the above method are plotted as squares in Fig. 16. The circles represent an independent determination of the flux using a cobalt wire [10]. The results have not been normalized and represent independent absolute flux determinations.

The value of irradiation time obtained was $4.65 \times 10^7$ s. The reactor records gave a value of $4.48 \times 10^7$ s. These results use a value of 41.2%
FIG. 11. Results of transverse scan using 0.125 in. diam. aperture in Pb collimator showing comparison between measured relative $^{137}$Cs activity as a function of position with calculated fraction of fuel plate area seen by detector through aperture.

FIG. 12. Calculated ratio of $^{137}$Cs activity to $^{134}$Cs activity in spent MITR fuel at removal from core as a function of neutron exposure at constant flux.
FIG. 13. Calculated ratio of $^{137}\text{Cs}$ activity to $^{95}\text{Zr}$ activity in spent MITR fuel at removal from core as a function of neutron exposure at constant flux.

FIG. 14. Curves of neutron flux versus neutron exposure for activity ratios $R_1$ and $R_2$ indicating form of solution for flux and exposure.
<table>
<thead>
<tr>
<th>Fission product</th>
<th>Gamma-ray energy (MeV)</th>
<th>Half period</th>
<th>Fission yield (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>Thermal fission</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>U²³⁵</td>
</tr>
<tr>
<td><strong>⁵⁵Zr</strong></td>
<td>0.724, 0.758</td>
<td>65 d</td>
<td>6.27</td>
</tr>
<tr>
<td><strong>⁹⁵Nb</strong></td>
<td>0.766</td>
<td>35 d</td>
<td></td>
</tr>
<tr>
<td><strong>¹⁰⁶Ru</strong></td>
<td>2.66, 2.40, 2.10, 1.55</td>
<td>1.0 y</td>
<td>0.38</td>
</tr>
<tr>
<td><strong>¹⁰⁶Rh</strong></td>
<td>1.05, 0.624, 0.607, 0.513</td>
<td>30 s</td>
<td></td>
</tr>
<tr>
<td><strong>¹³⁴Cs</strong></td>
<td>0.605, 0.796</td>
<td>2.3 y</td>
<td></td>
</tr>
<tr>
<td><strong>¹³³Cs</strong></td>
<td>none</td>
<td>stable</td>
<td>6.00</td>
</tr>
<tr>
<td><strong>¹³⁷Cs</strong></td>
<td>0.662</td>
<td>30 y</td>
<td></td>
</tr>
<tr>
<td><strong>¹⁴⁰Ba</strong></td>
<td>3.00, 2.520, 1.586</td>
<td>13 d</td>
<td>6.44</td>
</tr>
<tr>
<td><strong>¹⁴⁰La</strong></td>
<td>0.92, 0.82</td>
<td>40 h</td>
<td></td>
</tr>
<tr>
<td><strong>¹⁴⁴Ce</strong></td>
<td>2.186, 1.50, 0.697</td>
<td>285 d</td>
<td>5.62</td>
</tr>
<tr>
<td><strong>¹⁴⁴Pr</strong></td>
<td>0.610</td>
<td>17 min</td>
<td></td>
</tr>
</tbody>
</table>
for the branching ratio of the $^{95}$Zr 724-keV gamma-ray, obtained from an independent measurement using the lithium-drifted germanium detector.

Determination of burn-up

Figure 15 shows the calculated fraction of $^{235}$U atoms remaining per initial $^{235}$U atoms, $N_{25}/N_0$, as a function of total flux time $\omega$. The average value of $N_{25}/N_0$ for the element was then obtained using the measured values of $\omega$. The results of this calculation gave a value of 42.5%±10% of initial $^{235}$U fissioned. A two-group depletion code calculation with absolute flux normalized to the total reactor power production gave 34.4%±15% $^{235}$U fission [8].

![Graph showing relative axial flux distribution within fuel element 2M19](image)

**FIG. 15.** Relative axial flux distribution within fuel element 2M19 as obtained by method described in section II. 3.2. Included are results of independent measurements using cobalt wire for a similar element from Ref. [10]

Thermal measurements made during reactor operations on other elements indicate that the burn-up code tends to predict about 10% too few fissions. If this correction were applicable the agreement would be very good.

Determination of cooling time

Although the cooling time (i.e. time since last irradiated) is usually known from records, it is always possible to estimate it and in some cases determine it quite accurately from relative intensity of peaks in the spectrum. From Fig. 7 it is clear that the spectrum changes in obvious ways as the element cools. By comparing the measured spectra to a series of such curves a rough estimate of cooling time is always possible.
FIG. 16. Number of $^{235}$U atoms remaining per initial $^{235}$U atom, $N_{235}/N_{0}$, as a function of neutron exposure, calculated for MITR fuel.

In cases where the reactor has been run at steady power for times long compared to 65 d the $^{95}$Zr and $^{95}$Nb will be in equilibrium. In this case the ratio of the area under the combined $^{95}$Nb - $^{95}$Zr peak to the area under the isolated $^{95}$Zr peak will vary with cooling time, as shown in Fig. 17. Thus the measured ratio changes significantly during about the first 4 months and could be used to determine the cooling time. The above curve will not be correct if the reactor operation has been intermittent.

BENT CRYSTAL SPECTROMETER STUDIES

The 6-m bent quartz crystal spectrograph [9] used in previous experiments [4] was modified by replacing the photographic film by a movable
slit counter. A well-shielded NaI scintillation counter was placed behind a lead collimator on the focal circle. This instrument was capable of resolving the fission product spectrum in the energy range from 80 keV to about 500 keV. Because of the very rapid decrease of efficiency with energy no lines were detected above 500 keV. Two of the most prominent gamma-rays with reasonably long half lives were ones at 133 keV and 145 keV from $^{144}\text{Ce}$ and $^{141}\text{Ce}$ respectively. Both of these peaks were studied in an attempt to understand the problems associated with the use of the spectrometer.

One advantage of this instrument is that it was possible to resolve the lines at even the shortest cooling times obtainable (18 h). Figure 18 shows the signal-to-background levels as cooling time is increased for the 133-keV $^{144}\text{Ce}$ gamma-ray. The principal problem encountered with measurements made at such short times was the rapid change in background that made comparison of different measurements difficult. This problem disappears for cooling times of one month or more.

![Figure 18](image_url)

**FIG. 18.** Count rate of $^{144}\text{Ce}$ 133 keV gamma-ray for different cooling times

Because of considerable difficulties in determining the absolute efficiency of the spectrometer no absolute determination of burn-up was attempted. The intensities of the $^{144}\text{Ce}$ peaks in different elements were compared and were found to be approximately proportional to the calculated burn-up. However, considerably more work would be required before this method
could successfully be used to actually determine burn-up. Because the results from the lithium-drifted germanium detectors seemed so much more promising, work with this instrument has not been continued.

CONCLUSIONS

The investigations using a germanium (lithium) spectrometer have shown that it is possible to determine considerable information about the irradiation history of the spent fuel. In particular the flux time, flux, irradiation time, cooling time, spatial flux distribution within the element, and operating mode can be determined from ratios of fission product gamma-ray activity. None of these results depends upon an absolute intensity measurement. The studies of the MITR fuel indicate that these parameters can be determined, after suitable cooling time, to accuracies of ±10% or better. Inaccuracies due to self absorption and counter efficiency corrections are small because the gamma-rays of interest have roughly the same energy.

Although the results were obtained for fully enriched fuel from the MITR the method should be applicable to other types of fuel. In particular the $^{106}$Rh peak may be extremely useful in low enrichment fuel because of the difference in yield, of about a factor of 10, between $^{235}$U and $^{239}$Pu thermal fission. It should be possible to infer the ratio of $^{239}$Pu to $^{235}$U fissions and therefore conversion ratios.

The bent crystal spectrometer was able to resolve the gamma-ray spectrum below 500 keV even after little or no decay time. This was not possible with the germanium (lithium) spectrometer. The results of the bent crystal measurements will not yield the same information as the germanium (lithium) measurements because the identifiable fission products do not possess the appropriate nuclear characteristics. In particular neither $^{137}$Cs nor $^{134}$Cs give lines in this region. The bent crystal spectrometer therefore is considerably less useful for measurements of this type.

ACKNOWLEDGEMENTS

The authors wish to thank Dr. H.W. Kraner for extensive help in detector development. We also wish to thank Professor T.J. Thompson and the staff of the MITR for their suggestions and assistance.

REFERENCES


DISCUSSION

M. J. HIGATSBERGER: I should like to point out that burn-up history can also be predicted with the Compton spectrometer. One has to choose a suitable number of fission products with variable half-lives and make at least two measurements with a time interval in between.

N. C. RASMUSSEN: We have not tried this method.

A. J. FUDGE: I think that a word of caution should be added in connection with the use of caesium in any form as a monitor for burn-up in any burn-up determination. This and the previous paper have both dealt with MTR plate-type fuel elements irradiated at low temperatures. Anyone who has been concerned with power-producing reactors or any high-temperature system will have observed the marked migration which takes place in such cases. This is bound to cause errors because distribution through the sample will not be homogeneous or uniform. In cases of this sort, one has to use other monitor elements which are not likely to migrate.

N. C. RASMUSSEN: If migration occurred, this would present a real difficulty with a method like the one we have used. It certainly does occur in some cases. There may be ways of getting round this difficulty, such as averaging over the whole element, but this is bound to give rise to other difficulties. Of course, if the cladding is ruptured then the case is hopeless. I think that the chance of reducing this 10% error to something like 1% is fairly remote. One might expect a value of, say, 5%, but the kind of accuracy that has been spoken of in connection with other problems discussed at this Symposium just cannot be expected from this kind of method. The advantage of the method is that it can be used quickly and easily at a time when there is no figure available from other methods. It may be useful for that kind of thing. It could perhaps be of value to the Agency from the point of view of inspection or checking other methods but I agree that considerable difficulties can be expected.

C. L. A. BUECHLER: It is hard to imagine that this sort of method could be applied quickly and easily to the Yankee fuel elements, for example. Just moving those fuel elements is by no means quick or easy.

N. C. RASMUSSEN: I meant of course that it is only easy when compared to the destructive methods.

P. WEINZIERL: I must disagree with the remarks made by Mr. Fudge. Paper SM-67/7 does actually give additional relevant information on these BGR fuel-plate investigations but Mr. Higatsberger did not have time to include it in his oral presentation. For the individual pieces, the mean deviation between the gamma-spectrometry $^{137}$Cs determinations and the radiochemical and mass-analytical results was about 6%. If, however, the
results are added up and integrated over the whole element, an agreement of about 0.5% is obtained. We think that the effects of the migration of caesium — or its precursor — can clearly be seen in these results. If the canning of the element is intact, the scanning of the element as a whole gives a correct result.

A. J. FUDGE: I think that one of the big difficulties here is being overlooked. When migration occurs, it does not always occur in the way one wants it to. If one takes the whole fuel assembly, one can of course measure the total amount of activity from $^{137}$Cs, but unfortunately a pretty dense material is involved. So far, consideration has only been given to thin plates, but it seems unlikely that fuel will in fact be used in this way in power reactors. It will certainly be used in plate form but it is more likely to be used in rod form. Now when migration occurs in rods, it always does so in an outward direction, so that a very much larger signal is obtained than would otherwise be the case because there is less attenuation of the gamma-radiation. This is something, of course, which just has to be taken into consideration in the case of rods. One does not have a homogeneous material to measure; the material is distributed in an unknown way and so no correction can be made for it. I think that this is the sort of problem that will arise when these investigations are extended to power reactors. We have run into them already. The same difficulties with $^{137}$Cs have cropped up in the United States of America as well. Radiochemically, this isotope is just about the ideal marker. In practice, in the case of elements that have been cycled in power reactors, one can obtain any answer one likes with it.

M. J. HIGATSBERGER: All I can say is that if caesium does not work for a particular type of configuration, one just takes fission products with high energies, for instance barium or lanthanum. We have in fact already done this. I would refer you to the paper we presented at the third Geneva Conference.*

Three years ago everybody said it would be impossible to carry out a non-destructive test on nuclear fuel with gamma spectroscopy. It was even said that MTR elements could not be measured with a reasonable degree of accuracy. Well, we have shown that this method does work on the Brookhaven type and we will show that it works with power-reactor fuel as well.

P. WEINZIERL: In connection with Mr. Fudge's remarks on attenuation in fuel with uneven distributions, I should like to mention the following calculation results. If in a 16-plate MTR fuel element a 10% variation of burn-up — and thus $^{137}$Cs distribution — is assumed and if one scans the element from both sides, averages and applies the absorption correction for the case of an even distribution of burn-up, an error of only 0.8% is introduced into the result of the measurement.

N. C. RASMUSSEN: I think that is clear. Unless the elements are really large, the absorption correction is not a big number and the problem may not be too difficult.

G. WELCH: At Dounreay we have been examining mixed oxide fuels after irradiation at high temperatures. The nature of the migration of

* 3rd UN Int. Conf. PUAE, A/CONF.28/P/399,
fission-product caesium has been investigated by measuring the isotopic ratios of caesium from different parts of the fuel pins. These results have shown that there is some migration of fission-product caesium but most of the apparent migration occurs as the precursor xenon. Therefore, if caesium is to be used as a measure of burn-up in nuclear fuels there must be no leakage or diffusion of the gaseous xenon out of the fuel during irradiation.

We have also considered the possibilities of ruthenium-106 as an indicator of plutonium burn-up. On the basis of the few results observed so far we have been quite unable to balance the ruthenium found with the amount that should have been present. However, I wish Dr. Rasmussen luck with the work he is doing in this connection.
NON-DESTRUCTIVE METHODS FOR DETERMINING
BURN-UP IN NUCLEAR FUEL

W.J. McCONNAGLE
ILLINOIS INSTITUTE OF TECHNOLOGY, CHICAGO, ILL.,
UNITED STATES OF AMERICA

Abstract — Résumé — Аннотация — Resumen

NON-DESTRUCTIVE METHODS FOR DETERMINING BURN-UP IN NUCLEAR FUEL. Non-destructive methods for quantitative measurement of burn-up in nuclear reactor fuel elements are useful and desirable. The ideal method for fuel assay would be one that requires no special information about the neutron spectra, radiation history, or cooling time.

The irradiated fuel element contains a record of the fuel burn-up. This record is in the form of radioactive and stable isotopes resulting from the fission process. Unfortunately, in the non-destructive as well as the destructive fuel assay methods, the neutron spectrum, irradiation history, and cooling period influence this record. Likewise, the lack of precise nuclear data, such as values of nuclear cross-sections, affects any calculations that can be made. Another difficulty in the non-destructive assay is the presence of high radiation fields which contribute to the "noise" background of the measurements. The development of useful and realistic standards is difficult. The non-destructive burn-up methods do serve a useful purpose especially when an approximate value of burn-up is required quickly and economically even though in the present state of the art they lack the desired precision and accuracy.

Several non-destructive methods for determining burn-up have been used, are being evaluated, or have been proposed. Various types of spectrometers including the bent crystal, magnetic Compton, Compton coincidence, and scintillation have been used to analyse the gamma radiation from the radioactive material formed during the fission process. Other non-destructive methods include foil activation, neutron transmission, activation analysis, measurement of capture gamma rays, and the measurement of prompt and delayed neutrons. The basic principles of each of the above instruments and methods, their sensitivities and their limitations will be reviewed.

Non-destructive methods using stable isotopes produced during the fission process are proposed. In the use of stable isotopes, detailed irradiation history is of secondary importance and the cooling time is unimportant. Also, there is more precise nuclear data for the stable fission products. Of the stable isotopes produced during the fission process, zirconium, molybdenum, ruthenium, and neodymium appear to be the most useful. The proposed non-destructive methods using stable isotopes will be discussed.

METHODES NON DESTRUCTIVES D'ÉVALUATION DU TAUX DE COMBUSTION DANS LE COMBUSTIBLE NUCLEAIRE. Il est à la fois utile et souhaitable d'utiliser des méthodes non destructives pour procéder à la mesure quantitative du taux de combustion des éléments combustibles d'un réacteur nucléaire. L'idéal serait de pouvoir analyser le combustible à l'aide d'une méthode ne nécessitant pas de renseignements particuliers sur les spectres des neutrons, le schéma d'irradiation ou le temps de refroidissement.

Les isotopes radioactifs et les isotopes stables résultant du processus de fission qui sont présents dans un élément combustible irradié caractérisent son irradiation. Malheureusement, que l'analyse soit effectuée au moyen de méthodes destructives ou non destructives, les résultats obtenus varient en fonction du spectre de neutrons, du schéma d'irradiation et du temps de refroidissement. De plus, l'absence de données nucléaires précises, comme les valeurs des section efficaces, influe sur tous les calculs qui peuvent être effectués. L'analyse non destructive est également gênée par la présence de champs de rayonnements intenses qui augmentent le bruit de fond. Il est difficile d'établir des normes utiles et réalisées. Bien que, dans l'état actuel de la technique, les méthodes non destructives n'ient pas toute la précision et l'exactitude voulues, elles présentent néanmoins un grand intérêt, notamment dans les cas où il faut obtenir rapidement et économiquement une valeur approximative du taux de combustion.

Plusieurs méthodes non destructives d'évaluation du taux de combustion sont actuellement appliquées, à l'étude ou en projet. Plusieurs types de spectromètres sont utilisés pour la mesure du rayonnement gamma émis par les matières radioactives qui se forment au cours du processus de fission, en particulier le spectro-
метод, не требующий специальных данных о нейтронном спектре, истории радиации или времени охлаждения.

Облученные топливные элементы сами по себе являются носителями информации о степени выгорания топлива. Эта информация содержится в форме радиоактивных и стабильных изотопов, образующихся в процессе деления. К сожалению, как при методах анализа топлива без разрушения образца, так и с разрушением образца нейтронный спектр, история радиации и период охлаждения влияют на эту информацию. Точно так же недостаток точных ядерных данных, таких, как значения ядерных поперечных сечений, влияет на любые вычисления, которые можно было бы произвести. Другой трудностью при анализах без разрушения образца является наличие полей высокой радиации, которая вносит дополнительный "шум" в фон при измерениях. Разработка полезных и реальных стандартов является трудным делом. Методы анализа степени выгорания без разрушения образца особенно полезны тогда, когда требуется быстро и экономично, даже при существующем положении с их недостатком, получить желательную точность приблизительной величины выгорания.

Использовалось несколько методов для определения степени выгорания без разрушения образца; ведется их оценка или они предлагаются. Использовались различные типы спектрометров, в том числе спектрометры с изогнутым кристаллом, магнитный комптоновский спектрометр, комптоновский спектрометр на совпадения и сцинтилляционный спектрометр для анализа гамма-излучения радиоактивного материала, образующегося во время процесса деления. Другие методы анализа без разрушения образца включают активацию фольги, прохождение нейтронов, активационный анализ, измерение захвата гамма-лучей и измерение мгновенных и запаздывающих нейтронов. Рассматриваются основные принципы каждого из вышеуказанных методов и приборов, их чувствительность и их пределы.

Предлагаются методы анализа без разрушения образца с использованием стабильных изотопов, образующихся во время процесса деления. При использовании стабильных изотопов детальная история радиации приобретает второстепенное значение, а время охлаждения вообще не имеет значения. К тому же имеются более точные данные для стабильных продуктов распада. Из стабильных изотопов, полученных в процессе деления, наиболее полезными являются цирконий, молибден, рутений и неодим. Обсуждаются предлагаемые методы анализа без разрушения образца с использованием стабильных изотопов.

METODOS NO DESTRUCTIVOS PARA DETERMINAR EL GRADO DE COMBUSTION DE LOS ELEMENTOS COMBUSTIBLES NUCLEARES. Los métodos no destructivos son de gran utilidad para medir cuantitativamente el grado de combustión. El método ideal sería el que no requiriese datos especiales sobre los espectros neutrónicos, las irradiaciones precedentes o el tiempo de enfriamiento.

Los elementos combustibles irradiados llevan en cierto modo el registro de su grado de combustión. Este registro consiste en el conjunto de los isótopos radiactivos y estables resultantes del proceso de fisión. Desgraciadamente, tanto con los métodos no destructivos como con los destructivos el espectro neutrónico, las irradiaciones precedentes y el tiempo de enfriamiento influyen sobre dicho registro. Análogamente, la falta de datos nucleares precisos tales como las secciones eficaces nucleares repercute en los cálculos que pueden efectuarse. Otra de las dificultades de la determinación no destructiva consiste en la presencia de intensos campos de radiaciones que contribuyen a "ruido" de fondo de la medición. La preparación de patrones eficaces es difícil. Aunque en el estado actual de la técnica carezcan de la precisión deseada, los métodos no destructivos para determinar el grado de combustión resultan útiles sobre todo cuando es preciso obtener rápidamente valores aproximados sin que el costo de la operación resulte excesivo.
Para determinar el grado de combustión se han empleado, se están estudiando o se han propuesto diversos métodos no destructivos. Se han empleado varios tipos de espectrómetro -el de cristal curvo, el magnético de Compton, el de coincidencia de Compton y el de centelleo- para analizar la radiación gamma proveniente del material radiactivo formado durante el proceso de fisión. Entre otros métodos no destructivos cabe mencionar la activación de láminas, la transmisión neutónica, el análisis por activación, la medición de rayos gamma de captura y la medición de neutrones rápidos y retardados. En la memoria se exponen los principios básicos de cada uno de los instrumentos y de los métodos mencionados y se formulan observaciones sobre su sensibilidad y sus inconvenientes.

El autor propone métodos no destructivos basados en el empleo de isótopos estables producidos durante el proceso de fisión. Con el empleo de esos isótopos los datos relativos a las irradiaciones precedentes tienen una importancia secundaria y los relativos al tiempo de enfriamiento no tienen ninguna. Además, para los productos estables de fisión se dispone de datos nucleares más precisos. Entre los isótopos estables producidos durante el proceso de fisión, los más útiles parecen ser el circonio, el molibdeno, el níquel y el neodimio. En la memoria se analizan todos los métodos propuestos.

1. INTRODUCTION

The importance of an adequate determination of burn-up in reactor applications is hardly be over-estimated. This data is of prime importance in establishing the validity of the reactor constants which are an integral part of core design, reactor performance, and core life. This, of course, is related to the economics of the reactor operation. Non-destructive means for burn-up determinations of nuclear fuel are useful for the economic operation of power reactors, and serve both fuel manufacturers as well as the utilizers. An error in the over-prediction of the reactor core life could possibly result in severe economic loss to the reactor designer and manufacturer.

One of the fundamental quantities required in a fuel research programme is the burn-up of the fuel test specimens. This quantity is required to correlate the changes taking place in the fuel with temperature, neutron flux and integrated neutron flux. Burn-up determinations are needed for correlating grain growth, thermal expansion, fission gas release, the fission product migration with fuel temperature and reactivity changes with integrated neutron irradiation.

The term burn-up has a rather broad meaning in that it includes both depletion of a fissionable species ($^{235}$U depletion), and the energy produced by a fuel (number of fission events). The latter quantity, which is of interest here, can be expressed conveniently as atom per cent fission which is defined as the number of fission events per 100 initial heavy element atoms. This is related to the energy released (i.e. MWD/t) by a constant.

$$\text{Burn-up} (%) = \frac{\text{Total atoms of uranium fissioned}}{\text{Total atoms of uranium present initially}} \times 100$$

As $^{235}$U is burned up, it is partially replaced by $^{239}$Pu. As irradiation proceeds, $^{235}$U and $^{238}$U are burned up by fission and neutron capture according to the following scheme.

$$^{235}\text{U} + n = \text{fission products} + \text{neutrons}$$

$$^{235}\text{U} + n = ^{236}\text{U}$$
235U + n = fission products + neutrons
236U + n = 239U → 239Np → 239Pu
238Pu + n = fission products + neutrons

The irradiated fuel itself has or contains a record of the burn-up in the form of fission products. The problem is to utilize the information. One of the difficult problems in the assay of irradiated fuel arises from the intensely high radioactivity present in the fuel element.

An irradiated reactor fuel element emits a great variety of nuclear gamma rays and X-rays. Therefore, most of the commonly used radiation detection systems are unable to uniquely resolve the various energies. Using a high-resolution bent-crystal quartz gamma-ray spectrograph, the spectrum from an irradiated MIT reactor fuel element has been resolved into about 70 different components [1,2]. Most of these lines represent gamma ray transitions in the various fission fragments. The fission product source of most of these gamma rays have been identified. In addition to the gamma rays, a series of lines characteristic of uranium X-rays has been identified and some of the plutonium X-rays have also been observed. This information is shown in Table I.

The time factors governing production and decay of the various fission products are generally not accurately known, even if the reactor operation history and core configuration are given. This is due to the variation of flux distribution caused by core poisoning, decreasing fuel density, and in low enrichment fuels, increasing plutonium content. In principle the time factors are necessary in order to calculate fission density from a measurement of the intensities of certain selected fission product gamma lines. It can be closely estimated, however, if a fission product is chosen, the half life of which exceeds the maximum possible irradiation cooling period by an order of magnitude.

2. METHODS

Methods used or proposed for the non-destructive determination of burn-up are (1) activation of special foils or wires attached to the surface of the fuel element; (2) spectrum analysis using either the Compton magnetic spectrometer, or the bent-crystal diffraction spectrometer, or the Compton coincidence spectrometer; (3) gamma scanning; (4) activation analysis; (5) measurement of prompt and delayed neutrons; (6) capture gamma ray measurements; (7) neutron transmission; (8) slowing-down spectrometer; and (9) stable isotopes. Each of these techniques will be discussed separately.

2.1. Activation

In the activation technique, a uranium or plutonium foil or cobalt wire is attached on the surface of a fuel element. The foil is removed after discharge of the element from the reactor. The quantities of 235U burned out and plutonium produced are determined by the analysis of the alpha spectra of the foil. Because of low activity of 235U, the foil must be enriched for
TABLE I
OBSERVED $\gamma$- AND X-RAYS
E < 500 keV

<table>
<thead>
<tr>
<th>$\lambda$ (X-units)</th>
<th>E (keV)</th>
<th>Source</th>
<th>$\lambda$ (X-units)</th>
<th>E (keV)</th>
<th>Source</th>
</tr>
</thead>
<tbody>
<tr>
<td>25.418</td>
<td>486.76</td>
<td>La-140</td>
<td>124.316</td>
<td>99.524</td>
<td>Pu Ka$_2$ X-ray</td>
</tr>
<tr>
<td>24.912</td>
<td>496.65</td>
<td>Ru-103</td>
<td>135.791</td>
<td>91.114</td>
<td>Nd-147</td>
</tr>
<tr>
<td>37.639</td>
<td>328.71</td>
<td>La-140</td>
<td>152.750</td>
<td>80.998</td>
<td>Xe-133</td>
</tr>
<tr>
<td>40.589</td>
<td>304.82</td>
<td>Ba-140</td>
<td>154.326</td>
<td>80.171</td>
<td>I-131</td>
</tr>
<tr>
<td>42.190</td>
<td>293.26</td>
<td>Ce-143</td>
<td>154.433</td>
<td>80.115</td>
<td>Ce-144</td>
</tr>
<tr>
<td>40.574</td>
<td>249.58</td>
<td>Xe-135</td>
<td>155.420</td>
<td>79.606</td>
<td>Ce-144</td>
</tr>
<tr>
<td>54.210</td>
<td>228.23</td>
<td>Te-132</td>
<td>170.198</td>
<td>72.694</td>
<td>Ce-144</td>
</tr>
<tr>
<td>68.351</td>
<td>181.013</td>
<td>Mo-99</td>
<td>176.199</td>
<td>70.219</td>
<td>Ce-144</td>
</tr>
<tr>
<td>76.086</td>
<td>162.611</td>
<td>Ba-140</td>
<td>177.678</td>
<td>69.634</td>
<td>Sm-153</td>
</tr>
<tr>
<td>82.654</td>
<td>149.690</td>
<td>Te-131</td>
<td>179.620</td>
<td>68.881</td>
<td>Ce-144</td>
</tr>
<tr>
<td>85.102</td>
<td>145.384</td>
<td>Ce-141</td>
<td>185.003</td>
<td>66.877</td>
<td>Ce-144</td>
</tr>
<tr>
<td>88.096</td>
<td>140.443</td>
<td>Mo-99</td>
<td>185.463</td>
<td>66.711</td>
<td>La-140</td>
</tr>
<tr>
<td>92.704</td>
<td>133.462</td>
<td>Ce-144</td>
<td>207.971</td>
<td>59.491</td>
<td>Sb or Te-127</td>
</tr>
<tr>
<td>94.405</td>
<td>131.057</td>
<td>Ba-140</td>
<td>215.871</td>
<td>57.314</td>
<td>Ce-143</td>
</tr>
<tr>
<td>102.720</td>
<td>120.448</td>
<td>Nd-147</td>
<td>231.949</td>
<td>53.341</td>
<td>Ce-144</td>
</tr>
<tr>
<td>106.263</td>
<td>116.432</td>
<td>Pm-151</td>
<td>232.519</td>
<td>53.210</td>
<td>Te-132</td>
</tr>
<tr>
<td>107.218</td>
<td>115.395</td>
<td>U K X-ray</td>
<td>33.946</td>
<td>I-131</td>
<td></td>
</tr>
<tr>
<td>108.013</td>
<td>114.546</td>
<td></td>
<td>43.517</td>
<td>I-131</td>
<td></td>
</tr>
<tr>
<td>108.229</td>
<td>114.317</td>
<td>U K$_3$ X-ray</td>
<td>111.161</td>
<td>U K$_3$</td>
<td></td>
</tr>
<tr>
<td>110.589</td>
<td>111.878</td>
<td></td>
<td>112.062</td>
<td>U K$_3$</td>
<td></td>
</tr>
<tr>
<td>113.067</td>
<td>109.426</td>
<td>La-140</td>
<td>125.686</td>
<td>U K$_1$</td>
<td></td>
</tr>
<tr>
<td>116.577</td>
<td>106.131</td>
<td>Te-129</td>
<td>130.698</td>
<td>U K$_2$</td>
<td></td>
</tr>
<tr>
<td>118.001</td>
<td>104.850</td>
<td></td>
<td>208.571</td>
<td>W K$_2$</td>
<td></td>
</tr>
<tr>
<td>119.906</td>
<td>103.184</td>
<td>Sm-153</td>
<td>213.382</td>
<td>W K$_2$</td>
<td></td>
</tr>
<tr>
<td>121.237</td>
<td>102.052</td>
<td></td>
<td>213.382</td>
<td>W K$_2$</td>
<td></td>
</tr>
<tr>
<td>122.437</td>
<td>101.051</td>
<td>Np K$_1$ X-ray</td>
<td>213.382</td>
<td>W K$_2$</td>
<td></td>
</tr>
<tr>
<td>123.746</td>
<td>99.982</td>
<td>Ce-144</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

the $^{235}$U isotope. In cases of enriched fuel produced by the diffusion process, $^{234}$U is also present. The background of the $^{234}$U peak has a disturbing effect on the resolution of both $^{235}$U and $^{238}$U.
Some difficulties with the technique are that: (1) the uranium and plutonium must be "canned" or "clad" before being put in the reactor, which means additional work; (2) the resolution of alpha spectra is low; (3) the enriched uranium must be used to obtain sufficient alphas; (4) $^{238}$U is produced; (5) a resolution of 1% is required; (6) the high beta-background exerts an influence on the alpha spectra; (7) there is self absorption.

Cobalt monitors have been used to determine burn-up [3]. A cobalt-60 monitor was used because of the need for a detector with long half-life. Pure cobalt was not used because of corrosion. The large absorption cross-section of the monitor would disturb the flux and the activity of the monitor would be too great for convenient manipulation and measurement of the activity. Consequently, an aluminium-cobalt alloy wire is used.

Table II shows the energies and activity of the various alpha isotopes of interest.

2.2. Spectrum analysis

Another method of assaying irradiated fuel is to measure the intensity of one of the several gamma lines from a fission product of known fission yield. Owing to the extremely complicated spectrum, a high resolution instrument is needed for this analysis: Groshev and Demidov [4,5] have used a magnetic Compton spectrometer for such an investigation. Van Antwerp [6], Motz [7,8,9], and Lee Whiting [10-14] discuss the construction and operation of the Compton magnetic spectrometer in great detail. The experience of Groshev, Adyasevich and Demidov [15] indicates that the order of magnitude of the resolution of such an instrument is as shown in Table III. In the Compton magnetic spectrometer the gamma rays from the irradiated material are converted to electrons by Compton scattering in a special converter foil placed in the spectrometer. The spectrometer is designed to measure the momentum and distribution of electrons ejected from the foil into a prescribed solid angle by the incident photon beam.

The collimated photon beam impinges on a foil (such as beryllium) positioned in the gap between the pole pieces of an electromagnet. Electrons and positrons are ejected from the foil with an energy and angular distribution which depends on the incident photon energies, the foil thickness and the atomic number of the foil. For a given magnetic field in the pole face gap, electrons at a particular momentum range are focused at the exit slit. However, because of the interposition of the defining baffle, only a small fraction of these electrons, i.e. the electrons contained in a small solid angle defined by the baffle opening, reach the detector.

Such an instrument permits the evaluation of photon energies in a range extending from approximately 0.2 to 12 MeV with an accuracy of 2%. Also, the photon intensity for a specific energy can be measured to an accuracy of 10%. The price paid for higher resolution is a decrease in efficiency. For example in a typical magnetic spectrometer (in the energy range of 0.2 to 12 MeV) a monoenergetic photon flux of the order of 100 photons/cm$^2$ will produce approximately one detectable event.

The Compton electrons are generated uniformly throughout the foil, and upon emergence have travelled distances in the foil ranging from zero to the total thickness of the foil. Thus, with a monoenergetic source of
TABLE II

ALPHA SPECTRUM

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Energy (MeV)</th>
<th>Disintegrations per sec per mg</th>
</tr>
</thead>
<tbody>
<tr>
<td>U-234</td>
<td>4.763</td>
<td>$2.25 \times 10^5$</td>
</tr>
<tr>
<td>U-235</td>
<td>4.200 (4%)</td>
<td>802</td>
</tr>
<tr>
<td></td>
<td>4.40 (83%)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>4.47 (3%)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>4.58 (10%)</td>
<td></td>
</tr>
<tr>
<td>U-236</td>
<td>4.499</td>
<td>$2.4 \times 10^5$</td>
</tr>
<tr>
<td>U-238</td>
<td>4.180</td>
<td>12.4</td>
</tr>
<tr>
<td>Pu-239</td>
<td>5.099 (11%)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>5.137 (20%)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>5.150 (69%)</td>
<td></td>
</tr>
<tr>
<td>Pu-240</td>
<td>5.118 (24%)</td>
<td>$8.6 \times 10^6$</td>
</tr>
<tr>
<td></td>
<td>5.162 (76%)</td>
<td></td>
</tr>
</tbody>
</table>

TABLE III

RESOLUTION OF COMPTON MAGNETIC SPECTROMETER

<table>
<thead>
<tr>
<th>Gamma Ray Energy (MeV)</th>
<th>Half Width at Half Maximum = Per Cent</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.48</td>
<td>4.15</td>
</tr>
<tr>
<td>0.78</td>
<td>2.0</td>
</tr>
<tr>
<td>1.14</td>
<td>1.45</td>
</tr>
<tr>
<td>2.74</td>
<td>1.15</td>
</tr>
</tbody>
</table>

gamma rays, the electrons leaving the beryllium foil in the forward direction will have an energy distribution. An irreducible source of error in the Compton spectrometer is the plural scattering of electrons on passage through the foil.

Van Antwerp [6] used film for his detector as well as solid state detectors and scintillation detectors. He also used "beta filters" placed directly in front of the film. The purpose of the beta filters was to remove the very low energy electrons incident at the film holder. These filters also reduced the light sensitivity of the film. He also used intensifiers made of either scintillating plastic or plain plastic.
Another way of analysing the spectrum is with a bent-crystal diffraction spectrometer. The crystal diffraction spectrometer of the Dumont type has very good resolution, but requires extremely strong sources, and the construction is not simple. The total efficiency of a diffraction spectrometer is inherently very low (10^{-8} to 10^{-10}) the efficiency decreasing with increasing energy.

In the study by Rasmussen and Cohen [1, 2] previously mentioned, about 70 energies were observed, ranging from 50 to 500 kV; among these were the Kα lines of plutonium and uranium which could be easily identified. Rasmussen and Cohen used a six-metre bent-crystal spectrometer similar to that described by Kazi [16]. The uranium (Kα-1) X-ray has an energy of 98.442 keV and the plutonium (Kα-1) X-ray has an energy of 103.71 keV. The six-metre spectrograph can resolve these energies. Line intensities were measured using a sodium iodide scintillation detector mounted on a carriage which could be moved along the focal circle. The two Kα radiations previously mentioned were separated by 1.6 cm on the spectrograph focal circle. The background radiation was discriminated by using a relatively small sodium iodine crystal (2 cm in diameter by 2 mm thick) and a single-channel pulse-height analyser following the detector.

Johnson and Vrablik [17], and Kristiansen-Rogeberg [18] have used a crystal spectrometer to evaluate burn-up. Their results indicate that the burn-up must be rather high to detect the presence of plutonium and the cooling time must be relatively short.

The Compton recoil spectrometer has been used for burn-up determinations. The Compton recoil spectrometer has an efficiency comparable to the Compton magnetic spectrometer (0.1% per incident photon) but considerably better resolution. A detailed description of this Compton recoil spectrometer can be found in references [19] and [20].

In the Compton recoil spectrometer a well collimated beam of gamma rays impinges on a detector (scintillator crystal or solid state), and the backscatter quanta are detected by a second detector. A coincidence circuit is used to select and count Compton events of definite backscatter angle. By selecting only Compton events backscattered at an angle near 180°, a single peak for monoenergetic gamma rays is obtained. Selection of suitable events is done by accepting only those pulses from the second detector which are in coincidence with the back-scattered quantum detected in the first detector. This spectrometer is characterized by its efficiency, its response function, and its energy resolution. The total intrinsic efficiency is approximately 10^{-3} at maximum and decreases toward higher and lower energies. This is due to absorption losses - especially for backscattered quanta at lower energies - and to the decreasing Compton cross-section at higher energies.

2.3. Gamma scanning

Gamma scanning has been used by a number of investigators. Various fission products have been used or proposed for measuring burn-up. Frequently, Ce-Pr-144, with a half-life of 284 d and 221.90 keV gamma line is used for burn-up determinations, because the self-absorption in the fuel
element is lower at the higher energies. Furthermore, cerium has no tendency to migrate to the surface in the high temperature fuels.

Cs-Ba-137 with a half life of 30 yr is very advantageous for use in burn-up analysis because of its long half life and approximately equal yields for $^{235}\text{U}$ and $^{239}\text{Pu}$ fission. For normal fuel element utilization times of up to three years, the time factor can be estimated with good accuracy without information on reactor operation. Because of the quality of fission yields for the mass number 137, the determination of $^{137}\text{Cs}$ leads directly to the sum of the $^{235}\text{U}$ and $^{239}\text{Pu}$ fissions in the fuel containing only these fissionable isotopes. For practical cooling times of the order of weeks, the 661 keV line is very difficult to measure due to the neighbouring much stronger gamma lines from other fission products. Also, there is the possible influence of the migration of $^{137}\text{Cs}$ in the fuel.

Westinghouse have used gamma scanning to determine burn-up in a Yankee type reactor [21]. They have summarized their results as follows: "Within certain limitations gamma scanning measurements can be used to determine relative burn-up distribution, local peaking factors, the maximum-to-average burn-up ratio, and gross core asymmetries". In the Yankee type core, a simple one-to-one correlation between gamma activity and burn-up does not exist.

One of the difficulties encountered was the effect of the impurities in the stainless steel tubing from which the fuel rods were fabricated. It has been found that the gamma emission which accompanies the decay was approximately 17% of the total gamma activity measured along the length of the fuel rod. This does not present a problem if all the scanned fuel rods and assemblies have been fabricated from tubing drawn from the same heat of steel, or from heats which contain approximately the same composition of impurities; however, if the cladding material is obtained from several heats, each having a different composition of impurities, a significant bias can be introduced into the data, leading to erroneous conclusion. Cobalt-59 was found to be the troublesome impurity in the clad.

A second difficulty, which caused an increase in the uncertainty of the gamma scan data obtained during the programme, was the rather long time interval from the beginning to the end of the gamma scanning measurements. Approximately 18 months elapsed between the gamma scanning measurements of the initial and final fuel rods considered in the project. The fission-product decay which occurred during this 18 month period, was grossly, but adequately, accounted for by normalizing all fuel rod scans to a frequently scanned "standard" rod which was retained for this purpose throughout the entire programme.

Using the burn-up and gamma activity of a standard irradiated assembly as a reference, and assuming a one-to-one correlation between burn-up and gamma activity, it was determined that the gross gamma scans made in the assembly centre-holes predicted the assembly average burn-up to within approximately 8% on the average. The gamma scans underpredicted burn-up by as much as 17% for low burn-up assemblies, and overpredicted burn-up by as much as 18% for high burn-up assemblies. These discrepancies, which include gross errors caused by the non-uniform cobalt distribution, can be reduced with the use of a burn-up:gamma correlation. For Yankee Core I, application of the burn-up:gamma correlation to the assembly average
gamma activities reduced the average discrepancy from 8% to 4%, and narrowed the range of the discrepancies from -12 to +18%, down to -5 to +10%. If the gamma scans were corrected for cobalt non-uniformity, the difference between the burn-up and the gamma distribution would be somewhat smaller than the values given above. Considering the fact that the assembly average gamma activities were inferred from volume weighted pellet burn-ups measured in the central and corner subassemblies, the agreement between the burn-up and gamma activity distributions is quite good.

The use of gamma scans overpredicts the maximum point-to-average burn-up ratio by 14% if the gamma activity is taken to be directly proportional to burn-up. Again, the use of the burn-up:gamma correlation given previously sharply reduced the observed discrepancy. Gamma scanning measurements of fuel rods do not reliably predict gross burn-up asymmetries in the xy-plane unless the measurements are free from the bias which can be introduced by non-uniformities in the fuel-rod cladding.

Hanford [22] has constructed an underwater facility for scanning irradiated fuel. In this facility the fuel element can be moved linearly and vertically in front of a small-aperture collimator. The scintillation detector is well shielded and connected to the collimator. The gamma-ray energy band used for a fuel element scan is centred at 0.70 MeV with a window width of 0.31 MeV. Destructive analysis for comparison with the non-destructive assay are made by chemically analysing for $^{137}$Cs. Good correlation has been obtained with the destructive-analysis results. Sources of error in making measurements are caused by: fuel warping; changes in fuel-to-collimator geometry; repositioning of the specimen after a background count; drift in the electronic circuits.

Many other investigators, including Diggle and Blockadder [23], Cornish [24], Hart and coworkers [25], and Fudge and coworkers [26], have used gamma scanning to study burn-up. The work of Fudge and coworkers is especially interesting because they are making activity contour plots of individual fuel elements by use of highly collimated detectors.

Owen [27] has used the gamma activity of two specific fission products for measuring burn-up. A short half-life fission product gives a measure of the power distribution in the fuel for the period immediately before discharge, while a long half-life product gives a measure of the power integrated over the whole irradiation period. During irradiation of fuels in a reactor, the short half-life fission products build up to a state of secular equilibrium or saturation after a few half-lives. The power reactor changes their concentration characteristic of the new power level after a few half-lives. A build-up of long half-life fission products is approximately linear for about one half-life. Owen has chosen two fission products which have the following properties: characteristic gamma radiation which can be readily discriminated from the general background and high percentage yields from the main fissile isotopes.

The fuel elements are scanned using a sodium iodide scintillator connected to a photomultiplier tube. The desired pulses are counted using a multichannel analyser. To protect the detecting system from the high flux the specimens are placed behind a lead shield having a small collimating
slit. To minimize errors due to self shielding, the fuel element is rotated while being counted.

The short half-life product chosen for use in its analysis was Ba-La-140. Barium-140 is formed by the decay of two very short half-life fission products. The $^{140}$Ba has a half-life of 12.8 d decaying to $^{140}$La with a half-life of 40 h. The gamma ray emitted in the decay has an energy of 1.6 MeV. There is very little if any interference from other fission products. The long half-life product chosen was Ce-Pr-144. Cerium-144 has a half-life of 285 d decaying to the 17 min $^{144}$Pr. The gamma ray emitted has an energy of 2.2 MeV. There is little interference from other fission products. Table IV shows the fission yields of $^{140}$Ba and $^{144}$Ce.

This technique can be used either to give a relative measure of burn-up or to make absolute measurements. In the latter case the equipment must be calibrated using known sources of the appropriate fission products or by radiochemical analysis of fuel specimens which have been analysed using the equipment. The accuracy of the method is determined by the accuracy of calibration.

2.4. Activation analysis

To evaluate the feasibility of fission product activation analysis for $^{235}$U burn-up determinations in fuel, an extensive examination of all the stable fission products and their stable daughters was made [28]. The $^{103}$Rh isotope was chosen because of its high thermal-neutron activation cross-section (140 b) its short half-life (44 sec) and because of the relatively high fission yield (3%) of its parent, $^{103}$Ru, which has a 39.7 d half-life. A detailed analysis of this technique and calculations can be found in Ref. [28].

This technique has the following limitations: (1) a waiting time of about six months after shut-down is required so that essentially all $^{103}$Ru has decayed to $^{103}$Rh; (2) only the analysis of highly-enriched fuel plates will produce any meaningful data because of the extremely high standard counting deviation for low enrichment fuel plates; (3) conventional counting techniques are inefficient for counting the 556-keV gammas. For 90% enriched fuel plates, the resulting statistical error is significant. A 10.7% standard deviation in counting was calculated for the case of 90% enrichment and 2% burn-up using a solid state detector and $2\pi$ geometry. Scintillation detectors are not practical in this case because of poor resolution and high gamma background from the irradiated fuel plate under test.

Nevertheless, depending on the accuracy desired, the neutron source available and the possibility of circumventing some of the counting difficulties, this technique may very well prove to be of practical significance in determining fuel burn-up of relatively highly enriched fuel plates.

2.5. Prompt and delayed neutrons

In the fission process both prompt and delayed neutrons are emitted. The measurement of prompt and delayed fission neutrons had been proposed as a method for measuring burn-up. The basic principle of this method is the measurement of the fast fission neutron emitted as a result of thermal
neutron irradiation of the irradiated fuel elements. Such a measurement is sensitive only to the presence of unknown materials. Although the method will measure directly the total fissionable material in the specimen, one cannot directly determine the $^{235}\text{U}$ and plutonium content without other knowledge.

A refinement of the above technique would be to measure both the total fission neutrons and the delayed neutrons. The fast neutron yields from plutonium-239 and uranium-235 will be markedly different in these two measurements. The number of fast neutrons emitted per thermal fission are 2.43 and 2.89 for $^{235}\text{U}$ and $^{239}\text{Pu}$ respectively. The number of delayed neutrons per fission are 0.158 and 0.0061 for $^{235}\text{U}$ and $^{239}\text{Pu}$, respectively.

### 2.6. Capture gamma rays

When a thermal neutron is captured by a nucleus, the resultant compound nucleus is left in an excited state with an energy equal to the excited state from which decay occurs promptly with the emission of capture gamma rays. The spectra resulting from the capture gamma-ray spectra are complex, containing both low and high energy gamma rays. In the case of fissionable materials the prompt gamma spectrum consists of two parts: the prompt gamma rays emitted in the fission process and the capture gamma rays emitted in the neutron-gamma reaction. The prompt gamma spectra, resulting from the fission reaction in $^{235}\text{U}$ and $^{239}\text{Pu}$ are essentially identical. However, the capture gamma-ray spectra from $^{235}\text{U}$ and $^{239}\text{Pu}$ in the neutron-gamma reaction are different.

The basic principle of the method is to irradiate the fuel elements with thermal neutrons and detect the capture gamma-ray lines characteristic of $^{235}\text{U}$ and $^{239}\text{Pu}$. One of the problems in this technique is that the capture gamma-ray intensity from the fuel element must not be much weaker than the fission product activity of the fuel element. The sodium iodide scintillators cannot resolve the individual capture gamma ray lines, but use of solid state detectors, which have greater resolution, would be needed in this study.
2.7. Neutron transmission

The attenuation of neutrons in spent fuel elements occurs due to attenuation by $^{235}\text{U}$, $^{239}\text{Pu}$ and the fission products. However, the main effect is due to the combined effects of $^{235}\text{U}$ and $^{239}\text{Pu}$. Therefore, measurement of the thermal-neutron transmission through the spent fuel element can provide a measurement of the fissionable material present. Practical thermal-neutron transmission measurements should be possible using an external neutron beam from a reactor, or thermalized neutrons from a neutron generator or one of the stronger isotopic sources. In this method it is necessary to know the integrated neutron dose or burn-up to which the specimen has been subjected. In many reactors, neutron flux varies along the axis. In addition, local variations due to self-shielding and interference from other irradiation specimen, lead to a variation in the burn-up along the axis of the specimen.

2.8. Stable isotopes

There are a number of problems in using radioactive isotopes for burn-up analysis. One of the major problems is the influence of reactor operation and cooling time on the test results. The use of stable isotopes for burn-up analysis has been proposed. In the use of stable isotopes a detailed irradiation history is of secondary importance and cooling time is unimportant. Also there is more precise nuclear data for the stable fission product nuclei. Stable isotopes are being used in conjunction with mass spectrometry for destructive burn-up analysis.

There are many factors which dictate the choice of a stable isotope for use as a measure of burn-up. The main considerations are concerned with the nuclear properties of the isotope and it is essential that the number of atoms of the nuclide formed in the specimen is directly related to the number of fissions that have occurred during irradiation.

The requirements of a fission product nuclide for use as a measure of burn-up in a specimen can be summarized as follows. It must be found on or near the peaks of the fission-product yield curve. This minimizes error due to the fission yield variations with neutron energy and fissile nuclide. It must be capable of determination by isotopic dilution analysis and mass spectrometry. It must have a low capture cross-section for neutrons for itself and its precursors. Also, it must not be produced by a secondary process during irradiation. Its volatility must be similar to that of the matrix. Its volatile precursors, if any, must be short lived and remain in the specimen. For destructive analysis it must be easily separated on a carrier-free basis from radioactive species. It must not be present in significant amounts in the unirradiated matrix or picked up in subsequent chemical processing.

Two elements with isotopes which meet most of the above requirements are molybdenum and neodymium. Neodymium is unlikely to undergo any migration under irradiation, and the precursors that do exist are of short half-life and of low neutron capture cross-section.

Although there are about 30 stable fission products which have known fission yields, the possible choice of isotope(s) is limited to molybdenum
and neodymium. The neutron cross-sections of most of the molybdenum isotopes are significantly lower than those in neodymium.

The isotopes of molybdenum, mass numbers 97, 98, and 100 formed in nuclear fuel by fission, have the advantage of high fission yields, low cross-section for neutron capture reaction, no false precursors, and both molybdenum-98 and molybdenum-100 are primary fission products.

The determination of the amount of a given stable isotope might be determined by such nuclear techniques as activation analysis, or capture gamma-ray analysis. However, much experimental work must be done before these methods would become feasible or useful for the non-destructive determination of burn-up.

2.9. Slowing down spectrometer

Chandramoleshwar and coworkers [29] have used the slowing down-time spectrometer for assay of $^{239}\text{Pu}$. In the energy range from 0.01 to 1.0 eV, the fission cross-section of $^{235}\text{U}$ and $^{239}\text{Pu}$ are similar, except at 0.3 eV, where $^{239}\text{Pu}$ has a resonance whose value is about twenty times that of $^{235}\text{U}$. This difference can be used to determine the $^{239}\text{Pu}$ content of irradiated fuel if the relative fission rates in $^{239}\text{Pu}$ and $^{235}\text{U}$ are measured as a function of neutron energy.

Chandramoleshwar [29] used a Van de Graff accelerator as a source of pulsed neutrons. Short bursts of fast neutrons were injected periodically into thick lead. Neutrons of various energies, having a small energy spread about a mean energy, can be obtained as a function of time from each burst. The experimental results indicate that it is possible to detect plutonium in an irradiated specimen. However, the system has not been optimized as yet, so that the sensitivity and resolution of the method is not known as yet.

2.10. Comparative studies

Hart, Lounsbury and McKay [3] have made a comparative study of six methods of determining burn-up in uranium dioxide fuel. The six methods compared were: (1) uranium-235 depletion; (2) cobalt monitoring; (3) caesium-137 production; (4) plutonium production; (5) calorimetry; (6) calculation from reactor power and standard flux maps. It has been found that all of the methods agree to within their analytical accuracy with the exception of the cesium-137 method, which gives results that are, on the average, 15% low. The uranium-235 depletion method has the largest probable error. The methods are briefly summarized below:

1. The uranium-235 depletion method is the least ambiguous of the methods described. However, the mass spectrometer used in the initial study was rather inaccurate at low burn-ups. Mass spectrometry is also the most time-consuming method. Use of a different mass spectrometer can extend the limit of accurate measurements to considerably lower burn-ups with considerably less effort.

2. The cobalt monitor method involves estimation of the effective cross-section and self-shielding. Both of these quantities are a function of neutron spectrum and involve an estimation of surface-to-average flux. However,
the ease and accuracy of measurement make it useful, and the application of dilute alloy wires and epithermal index monitors could help to overcome some of the uncertainties in the measurement.

(3) The caesium-137 should be unambiguous but caesium migration in the hot zones of the fuel elements and conflict in the half-life values make the results uncertain. Similar complications in cerium-137 determinations have been reported by many others. Precautions must be taken to avoid caesium adsorption on glassware when making wet-chemistry analysis.

(4) The plutonium method involves an estimation of the ratio of the total/thermal/(plus 1/v) captures in uranium-238 for each specimen. Since this ratio varies with neutron spectrum the method is only applicable where the ratio is well known.

(5) The calorimetric method must be combined with another method for estimating flux distribution among the specimens in the fuel charge before it can be considered reliable. Corrections must also be made for departure from adiabatic conditions.

(6) When calculating burn-up from the reactor power, the vertical flux distribution must be adequately known before accurate results can be expected.

3. CONCLUSION

Various non-destructive methods for determination of fuel burn-up have been briefly reviewed. There is perhaps no best method applicable for all burn-up specimens and determinations. There are however several methods capable of giving good accuracy on appropriate specimens if proper experimental technique is followed. Instruments such as the Compton magnetic spectrometer which have high resolution are too inefficient to be used for burn-up studies on a production line basis. The use of stable isotopes for burn-up analysis seems promising, but much experimental work must be done to adequately evaluate this method.

REFERENCES

McGONNAGLE


THE NON-DESTRUCTIVE EXAMINATION OF IRRADIATED NUCLEAR FUEL FOR BURN-UP BY GAMMA SPECTROMETRY WITH MECHANICAL SCANNING

A.J. FUDGE, E. FOSTER AND L. MURPHY
ATOMIC ENERGY RESEARCH ESTABLISHMENT, HARWELL, BERKS., UNITED KINGDOM

Abstract — Résumé — Аннотация — Resumen

THE NON-DESTRUCTIVE EXAMINATION OF IRRADIATED NUCLEAR FUEL FOR BURN-UP BY GAMMA SPECTROMETRY WITH MECHANICAL SCANNING. Gamma-ray spectrometry with mechanical scanning of irradiated nuclear fuel has been used to determine the absolute burn-up, non-destructively, and the distribution and total burn-up for a fuel assembly. In addition it gives valuable information on the state of the fuel, on the movement of fuel within the cladding, and on detailed neutron flux distribution within the reactor.

Fully automatic equipment has been designed and operated to enable the distribution of specific radioactive fission product nuclides to be obtained for both rod and plate type fuel assemblies. Physical resolution and gamma attenuation is achieved by variable collimation, filtration, and distance. Electronic detection and resolution is obtained using sodium iodide crystals and solid state detectors followed by pulse height analysis. Profile recording of the intensity is carried out by analogue and digital measurement, and an X-Y-Z plotter.

Mechanical scanning of the fuel element is carried out at either a continuous uniform speed or in a series of equal increments equal to the collimator slit width. A high degree of reproducibility can be achieved for both systems and corresponding recording equipment by electronic pulse circuitry control. Two-dimensional scanning is used for plate specimens. Automatic control is maintained from a programmed input and ensures continuous operation.

The precision and accuracy of this technique of burn-up determination is discussed in relationship to fissile material distribution and the calibration technique employed. Examples are included of the different types of information that have been obtained using this equipment and their interpretation is also commented on.

MESURE NON DESTRUCTIVE, PAR SPECTROMETRIE GAMMA ET EXPLORATION MECANIQUE, DU TAUX DE COMBUSTION DES COMBUSTIBLES NUCLEAIRES IRRADIES. La spectrométrie gamma associée à l'exploration mécanique a été utilisée pour la détermination absolue et non destructive du taux de combustion atteint par les combustibles irradiés ainsi que de la distribution et de la valeur totale de ce taux pour un assemblage d'éléments combustibles. Elle fournit également des renseignements intéressants sur l'état du combustible, le mouvement du combustible à l'intérieur de la gaine et la distribution exacte du flux de neutrons dans le réacteur.

Des appareils entièrement automatiques ont été mis au point afin de permettre le calcul de la distribution des produits de fission radioactifs dans des assemblages combustibles en barres et en plaques. La résolution physique et l'atténuation du rayonnement gamma sont obtenues à l'aide d'un collimateur réglable et en faisant varier le filtrage et la distance. La détection et la résolution électroniques sont réalisées au moyen de cristaux d'iode de sodium et de détecteurs à semi-conducteurs, associés à un analyseur d'amplitude. La courbe d'intensité est établie au moyen de mesures par ordinateurs analogiques et numériques et d'un traceur de courbes X-Y-Z.

L'exploration mécanique de l'élément de combustible est effectuée, soit de manière continue, à une vitesse uniforme, soit par bandes successives correspondant à la largeur de la fente du collimateur. Un contrôle électronique du circuit à impulsions assure une bonne reproductibilité avec les deux systèmes et leurs enregistreurs. L'exploration à deux dimensions est appliquée pour les éléments en plaques. Un contrôle automatique par programmation assure la marche continue des opérations.

Le mémoire analyse la précision et l'exactitude de cette méthode de mesure du taux de combustion en fonction de la distribution des produits fissiles et de la technique d'étalonnage utilisée. Il donne des exemples des différents types de renseignements qui ont été obtenus à l'aide des appareils décrits et il étudie la manière dont il convient de les interpréter.
ПРОВЕРКА ОБЛУЧЕННОГО ЯДЕРНОГО ТОПЛИВА НА ВЫГОРАНИЕ БЕЗ РАЗРУШЕ-
НИЯ ОБРАЗЦА С ПОМОЩЬЮ ГАММА-СПЕКТРОМЕТРИИ С МЕХАНИЧЕСКИМ СКЕНИР-
ВАНИЕМ. Гамма-спектрометрию с механическим скеннированием облученного ядерного
топлива использовали для определения абсолютного выгорания без разрушения образца и для
определения распределения и полного выгорания для топливной сборки. Это дает ценную
информацию о состоянии топлива, движении топлива внутри оболочки и подробном распреде-
лении нейтронного потока в реакторе.

Было сконструировано полностью автоматическое оборудование для того, чтобы обеспе-
чить распределение конкретных осколочных радиоизотопов, которые могут быть получены
как для стержневых, так и пластинчатых топливных элементов топливных сборок. Физическое
разрешение и гамма-затухание достигаются с помощью разнообразной коллимации, фильтра-
ции и расстояния. Электронное обнаружение и разрешение достигаются путем использования
кристаллов нодистого натрия и твердых детекторов, за которыми следует аналита-
затор импульсов. Запись интенсивности в разрезе осуществляется с помощью аналогового
и цифрового измерений и графической записи X-Y-Z.

Механическое скеннирование топливных элементов осуществляется либо при постоянной
однаковой скорости, либо в целой серии равных инкриментов, приравненных к ширине щели
коллиматора. Для обоих систем может быть получена высокая степень воспроизводимости,
а соответствующее записывающее оборудование работает с помощью управления электронной
импульсной схемы. Для пластинчатых образцов используется двухразмерное скеннирование.
Автоматический контроль осуществляется за счет программных сигналов и обеспечивает
непрерывность работы.

Точность и четкость этого метода определения выгорания обсуждается в связи с рас-
пределением делящихся материалов и калибровкой используемого метода. Приводятся приме-
ры различных видов информации, полученной благодаря использованию этого оборудования,
а также комментируются их интерпретации.

DETERMINACION NO DESTRUCTIVA DEL GRADO DE COMBUSTION POR ESPECTROMETRIA GAMMA CON
EXPLORACION MECANICA. Los autores han empleado la espectrometría gamma con exploración mecánica
para determinar en forma no destructiva el grado absoluto de combustión de los elementos irradiados y la
distribución y el grado total de combustión de un montaje de combustible. Este método proporciona, además,
valiosa información sobre el estado del combustible, sobre sus movimientos dentro del revestimiento y sobre
la distribución detallada del flujo neutrónico en el reactor.

Para poder obtener la distribución de los núcleos radiactivos productos de fisión en montajes de combus-
tible tanto en barra como en placa, los autores han ideado un equipo totalmente mecánico. La resolución
física y la atenuación de los rayos gamma se logran variando la colimación, la filtración y la distancia. La
detección y resolución electrónicas se obtienen con cristales de yoduro de sodio y detectores de estado sólido,
y mediante el análisis de amplitud de impulsos. Con mediciones numéricas y analógicas se traza el perfil de
la intensidad y se prepara un gráfico X-Y-Z.

La exploración mecánica del elemento combustible se efectúa a una velocidad uniforme continua o con
una serie de incrementos iguales a la anchura de la redija del colimador. Mediante el control de impulsos
electrónicos se puede lograr un alto grado de reproductividad para ambos sistemas y para el correspondiente
equipo de registro. Cuando se trata de muestras de placas se emplea la exploración bidimensional. La pro-
gramación mantiene el control automático y asegura el funcionamiento continuo.

Los autores analizan la precisión y la exactitud de esta técnica en relación con la distribución de los
materiales fisionables y con la técnica de calibración empleada. Dan algunos ejemplos de los diferentes
tipos de información obtenidos con el equipo descrito y formulan algunas observaciones sobre su interpre-
tación.

INTRODUCTION

The determination of burn-up, or the total number of fission events,
that have occurred during an irradiation of a small nuclear fuel test speci-
men, can be carried out by a number of different, but fairly well-established
procedures. The choice of the method used depends on such factors as the
accuracy required, the chemical nature of the specimen, and irradiation and decay times.

These methods fall into four main classes:

(a) Isotopic analysis of the fissile and fertile nuclides before and after irradiation \([1, 2, 3]\);
(b) Radiochemical analysis for a radioactive fission product nuclide produced in a known yield at a known uniform rate \([4, 5]\);
(c) Determination of a long-lived or stable fission product nuclide produced in known yield by conventional chemical analysis \([9, 10]\), or by isotopic-dilution mass-spectrometric analysis \([6, 7, 8, 11]\); and
(d) Measurement of heat output and external flux monitoring for certain specialized irradiations \([1, 16]\).

Unless the whole of the specimen is taken for analysis, the results obtained by the first three methods would relate only to a small fraction of the specimen. The high degree of accuracy of which the first three methods are capable is of little use if applied to only a small volume of a nuclear fuel element. Before an accurate assessment of variation of burn-up within an irradiated fuel specimen can be obtained, a prohibitively large number of samples must be analysed chemically. Methods of class (d) have a very limited application and, although they are non-destructive and capable of good precision, they are difficult to calibrate for the determination of absolute burn-up.

A need existed, therefore, for a non-destructive technique for measuring the distribution of burn-up within nuclear fuel specimens. The selected technique should have adequate precision to enable the magnitude of the burn-up variations measured to be checked by subsequent chemical analysis. The chemical result could then be used for absolute calibration of the non-destructive procedure.

The initial approach involved the measurement of the radioactivity of specific radio-nuclides found in single pellets or pieces of fuel element by gamma-ray spectrometry. The radioactivity was reduced to acceptable levels for the sodium iodide detector by rough collimation, attenuation with lead absorbers, and distance. It was soon apparent that this approach was severely limited by the specimen size and by the difficulty of absolute calibration. Consequently, the technique was developed further to enable the gamma-ray activity of selected volumes of irradiated material to be observed individually. To give complete coverage of larger specimens, including those in the form of plates, improved collimation and ability to scan were incorporated into the equipment. Recently, improvements in the electronic resolution of the detectors have been achieved and this has further enhanced the usefulness of the apparatus. Observations on the gamma spectra of nuclear fuels irradiated and cooled for varying periods have been very informative in selecting the best fission product nuclide for use as a fission monitor.
EQUIPMENT FOR GAMMA SCANNING

The mechanical equipment has been described already [12], and a more detailed description of the scanning control is to be published separately [13]. Only the main features of interest will be described here.

In order to measure the gamma-rays from a definite volume of fuel and to reduce the contribution from adjacent material, high density shielding is placed as close to the specimen as possible. Tungsten was selected in preference to most other shielding materials because of its higher density and ease of machining. The scanning mechanism has been designed to allow accurate positioning of the fuel specimen in relation to the slit to be achieved. Samples for subsequent chemical analysis for calibration purposes can then be taken from a well-defined position. The synchronous recording of count rate with both continuous and incremental scanning is very important for the highest precision and accuracy. This has been achieved by electronic control of pulsed stepping motors, a variable frequency pulse generator and limit scalers. Completely automatic operation is also possible using this system.

DETECTION, MEASUREMENT AND RECORDING OF THE RADIOACTIVITY

Gross gamma-ray counting or even a simple form of energy discrimination, as used by many workers in this field, is of little use for burn-up determinations. The highest possible resolution of the gamma-rays must be achieved to make the technique of maximum usefulness. The main detection system used, so far, has been a thallium-activated sodium iodide crystal and photomultiplier. The chief limiting features of this method are the insufficient resolution of the gamma-rays, pulse pile-up, and spectrum distortion at high activity levels.

Two other detector systems are under trial at the present time. The first system, which has already shown promising results, is a lithium-drifted germanium diode operated at liquid nitrogen temperatures. The other is a Compton spectrometer consisting of a lithium-drifted silicon diode and a number of sodium iodide crystals. Both these systems are less efficient than the sodium iodide detector, although this is an advantage when scanning specimens with high gamma emission rates since less attenuation of the activity is required.

After suitable amplification of the pulses from the detector, it is necessary to carry out pulse height analysis. A multichannel analyser has definite advantages for spectral recognition, but these instruments are limited by a high dead time, and the production of too much data for scanning purposes. High stability single channel analysers having low fixed dead times are therefore used. These can be set to count only the photopeak response of a particular radionuclide, and they give an output that can be recorded in a number of different ways.

Two systems of recording the photopeak count rate from the single channel analyser can be used. For preliminary work on the distribution of burn-up, an analogue trace is obtained using a linear ratemeter and a recorder driven synchronously with the scanning rate. An analogue display
using colour to represent radiation intensity has been developed. This en-
ables a rapid visual assessment of the fission density to be made, and it is
particularly useful for plate specimens and for radial cross-sections of fuel
rods. For detailed measurements of burn-up within a specimen and for
determination of the integrated burn-up of the same specimen, the analysen
output is fed into a scaler and subsequently into a readout system printing
on paper tape or punched cards for computer analysis.

CHOICE OF RADIO-NUCLEIDE AS FISSION MONITOR

The radioactive fission product nuclide chosen as fission monitor for
a non-destructive procedure must have most of the properties required for
radiochemical analysis. In addition it must be possible to measure it in
the presence of the gamma-spectrum of mixed fission products emitted by
an irradiated fuel element composed of dense material.

The nuclide must fulfil the following conditions:

(a) Adequate sensitivity is required, therefore it must have a high
gamma-ray abundance and fission yield. Fission product nuclides produced
in high yield are also less dependent on changes in incident neutron energy
and of the nuclide undergoing fission.

(b) It must have a low neutron absorption cross-section, and both it
and its precursors must have a low volatility. This ensures that the fission
product concentration as measured by the activity can be related directly
to the burn-up throughout the specimen.

(c) The gamma-rays have to penetrate the dense fuel element hence
a high gamma-energy is required. This also reduces corrections that have
to be made in some cases for self absorption. It also reduces errors caused
by unsymmetrical fission across a fuel rod.

(d) Variations in neutron intensities within the specimen or within the
core of the reactor will cause the fission rate to vary during the irradiation.
The nuclide chosen therefore must have a long half life, since a short-lived
fission product would not adequately record relative flux changes that occur
early in a long irradiation. For short irradiations, or where it is known
that the relative flux throughout the core or the specimen remains reasonably
constant, nuclides of shorter half life could be used without too great an
error.

For most burn-up determinations the above factors limit the choice
to the following fission product nuclides: $^{95}$Zr-$^{95}$Nb, $^{144}$Ce-$^{144}$Pr and $^{137}$Cs-$^{137}$Ba.

Zirconium-95 — niobium-95

For most of the irradiations and decay periods encountered in fuel ele-
ment test irradiations, this parent-daughter pair of fission product nuclides
gives rise to the greatest abundance of gamma-rays. These are readily observed with a sodium iodide crystal as a single photopeak at 0.75 MeV and this suffers little from interference from any other gamma-emitting fission product. They can be resolved by a lithium-drifted germanium diode into a $^{95}$Zr photopeak at 0.724 MeV and a partially resolved 0.756 MeV peak of $^{95}$Zr and 0.764 MeV peak of $^{95}$Nb. The fission yield is high and relatively constant for all types of fission. The volatility of these nuclides and their precursors is very low, as is their neutron absorption cross-section. Unfortunately, the half lives of both radionuclides are comparatively short (65 d for $^{95}$Zr and 35 d for $^{95}$Nb). In order to calculate burn-up from an activity measurement using this pair of nuclides, it is necessary to know the contribution of the $^{95}$Zr alone. This cannot be obtained using a sodium iodide crystal and in this case it must be calculated from a knowledge of the irradiation history. This increases the uncertainty of the measurement (the better resolution obtained with the lithium-drifted germanium diode should overcome this difficulty). Despite these limitations it is the only practical monitor for burn-up determination for many irradiation conditions.

_Cerium-144 – praseodymium-144_

The parent nuclide has a half life of 285 d and gamma-energy of 0.134 MeV, while the daughter half life is 17 min and its energies are 0.695, 1.48 and 2.18 MeV. The gamma-abundance is low, however, so that the total activity at the detector must be very high to obtain a useful count rate at a particular energy. This leads to large dead time corrections or pulse pile-up problems when using a sodium iodide crystal. With a germanium diode the scanning rate must be reduced very considerably. There is a significant difference in the fission yields for $^{144}$Ce for $^{235}$U and $^{239}$Pu fissions. Interference can also occur between the 2.18-MeV photopeak of this pair and the 2.10-MeV photopeak from $^{106}$Ru-$^{106}$Rh in $^{239}$Pu fission, since the fission yield of this latter pair is enhanced in this case.

_Caesium-137 – barium-137_

The half life of caesium-137 is 30.4 ± 0.4 yr [14] and the thermal fission yield for $^{235}$U and $^{239}$Pu is 6.15% and 6.63% respectively [15]. This system is therefore almost ideal as a fission monitor for long irradiations. It is not possible with a sodium iodide detector to resolve completely the caesium-137 – barium-137 photopeak at 0.66 MeV from the zirconium-95 – niobium-95 photopeak at 0.75 MeV. It can only be used as a burn-up monitor on long-irradiated and long-decayed fuel specimens or where a lithium-drifted germanium diode detector is available to give the required resolution. There are two other factors that affect the usefulness of this pair of nuclides. These are the high volatility of caesium compounds above 600°C and the high self absorption of the gamma-rays of energy 0.66 MeV in the dense fuel material.

The particular radionuclide selected as a monitor will depend therefore on the type of fuel, the lengths of the irradiation and decay periods, and the temperature of the fuel during the irradiation.
EVALUATION OF BURN-UP

The concentration of a specific fission product nuclide in any volume of an irradiated nuclear fuel can be used, within limits, as a measure of the total number of fission events that have occurred in that volume during irradiation. To relate the activity of a radionuclide to burn-up, or even fission density, it is necessary to know very precisely the homogeneity of the fissile material and its concentration throughout the fuel element. For many systems this is not a source of error and homogeneity of a high degree can be accepted for compounds and metals of single fissile species. Where mixed fissile materials are irradiated in alloys and cermets the homogeneity must be determined before irradiation, preferably by a non-destructive technique. Inhomogeneity could easily be the biggest error in the accuracy of a burn-up measurement carried out subsequently. Where changes occur in fuel thickness, as for example between fuel specimens irradiated simultaneously in the same reactor, allowance can be made for the change in self absorption of the gamma-rays using the equation

$$\frac{A}{A_0} = \frac{1}{\mu l} (1 - e^{-\mu l})$$

where

- $A$ is the observed activity of gamma-rays of this particular energy
- $A_0$ is the unattenuated activity of these gamma-rays
- $\mu$ is the absorption coefficient for gamma-rays in material of the particular composition
- $l$ is the thickness of the specimen.

This assumes that the measurement is being made at right angles to a plane face of a right cylinder.

The absolute calibration of the efficiency of the detecting and measuring system for gamma-rays is not strictly necessary, or desirable. Subsequent chemical analysis of an accurately known volume of the scanned fuel for burn-up by methods such as fissile element depletion and stable fission product analysis, can be related to the count rate of a selected fission product radionuclide. Burn-up in other parts of the specimen can then be obtained from the observed count rates by simple proportion, after they have been corrected for the dead time of the system. This method of calibration reduces uncertainties caused by irradiation and decay times, decay scheme and fission yield. The reproducibility of the technique is controlled only by the stability of the electronic equipment, which must be checked frequently against standardized sources. The precision of the measurement of the activity is usually 5%, but 2% can be achieved by careful attention to detail. Care must be taken to obtain an accurate measure of dead time of the equipment by electronic means and it is advisable to record total count rate as well as the analyser channel output.
RESULTS

A large number of irradiated fuel assemblies in the form of pelleted and powder compacts, and metals, alloys and cermets as rods and plates, have been examined by scanning and gamma-spectrometry. The information obtained has shown fuel movement, dimensional changes, fission product migration, and the presence of unsuspected material in the irradiation assemblies, as well as providing burn-up values. The fission density measurements have been used to relate physical changes to definite causes, and to obtain more realistic neutron flux profiles for different reactor systems.

One example is included here of the over-all accuracy of this technique for burn-up determination. An irradiation assembly, consisting of eight cans of pellets of enriched uranium dioxide of various diameters and irradiated simultaneously for 72 d in a test reactor, was scanned and subsequently chemically analysed. Counts from a 0.05 in. x 0.05 in. collimator aperture for the $^{95}$Zr-$^{95}$Nb photopeak around 0.75 MeV were taken along the centre line of the pellets. A typical plot is shown in Fig. 1. As the pellets were of differing diameters, the count rates were corrected for self absorption as well as dead time. Sections of some of the pellets, 0.05 in. wide, from known positions were then sliced out and analysed for burn-up by radiocchemical [4, 5] and mass-spectrometric procedures [1, 2, 3, 16]. The mean value obtained for one of the pellets was used to calibrate the system and the burn-up for the various pellets was expressed relative to this one. These results were then compared with the results on seven other pellets sectioned and analysed chemically. The results obtained by counting show good agreement with the chemical results and in all but one case are within the one standard deviation quoted for the mean of the chemical results. The values obtained are shown in Table I.

CONCLUSIONS

The measurement of the concentration of selected radioactive fission product nuclides by gamma-spectrometry and mechanical scanning has enabled much useful information on the distribution of fission density in nuclear fuel elements and test specimens to be obtained. The data has been converted into an absolute measurement of burn-up by suitable calibration. The over-all accuracy of the burn-up measurement has been increased by using methods based on stable fission product nuclides and fissile element depletion for the calibration analysis on samples taken from a well-defined position in the specimen. Uncertainties in half lives and decay schemes are eliminated in this way. The nuclides used for gamma-scanning are selected on the basis of irradiation and decay times, fuel type, the temperature during irradiation, and gamma-energy. Compared with chemical methods, an over-all accuracy of 10% has been achieved on the example reported, and this is typical of all other systems at this level of burn-up.

This technique is capable of measuring the burn-up or total number of fission events that have occurred during the irradiation of fissile material in a nuclear reactor. It will only be possible to calculate the depletion of
FIG. 1. Incremental scan of oxide fuel assembly
The fissile material from this data if the capture-to-fission value \( a \) and the breeding ratio are well known and constant throughout the system. Information on these two aspects is still under investigation.

**TABLE I**

**BURN-UP DETERMINATION: VALUES OBTAINED BY COUNTING AND CHEMICAL ANALYSIS**

<table>
<thead>
<tr>
<th>Fuel can No.</th>
<th>Pellet No.</th>
<th>Count rate (corrected)</th>
<th>Burn-up in at.-%</th>
<th>Mean burn-up as measured chemically</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>1</td>
<td>3.54</td>
<td>0.85</td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>3.24</td>
<td>0.87</td>
<td></td>
<td></td>
</tr>
<tr>
<td>3</td>
<td>3.09</td>
<td>0.83</td>
<td></td>
<td></td>
</tr>
<tr>
<td>4</td>
<td>3.08</td>
<td>0.83</td>
<td></td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>3.06</td>
<td>0.82</td>
<td></td>
<td></td>
</tr>
<tr>
<td>6</td>
<td>3.05</td>
<td>0.82</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Fuel can No.</th>
<th>Pellet No.</th>
<th>Count rate (corrected)</th>
<th>Burn-up in at.-%</th>
<th>Mean burn-up as measured chemically</th>
</tr>
</thead>
<tbody>
<tr>
<td>2</td>
<td>1</td>
<td>5.26</td>
<td>1.41</td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>5.16</td>
<td>1.39</td>
<td></td>
<td></td>
</tr>
<tr>
<td>3</td>
<td>5.12</td>
<td>1.38</td>
<td></td>
<td>1.49 ± 0.11</td>
</tr>
<tr>
<td>4</td>
<td>5.15</td>
<td>1.38</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Fuel can No.</th>
<th>Pellet No.</th>
<th>Count rate (corrected)</th>
<th>Burn-up in at.-%</th>
<th>Mean burn-up as measured chemically</th>
</tr>
</thead>
<tbody>
<tr>
<td>3</td>
<td>1</td>
<td>5.54</td>
<td>1.49</td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>5.43</td>
<td>1.46</td>
<td></td>
<td>1.48 ± 0.11</td>
</tr>
<tr>
<td>3</td>
<td>5.46</td>
<td>1.47</td>
<td></td>
<td></td>
</tr>
<tr>
<td>4</td>
<td>5.47</td>
<td>1.47</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Fuel can No.</th>
<th>Pellet No.</th>
<th>Count rate (corrected)</th>
<th>Burn-up in at.-%</th>
<th>Mean burn-up as measured chemically</th>
</tr>
</thead>
<tbody>
<tr>
<td>4</td>
<td>1</td>
<td>5.95</td>
<td>1.60</td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>5.86</td>
<td>1.57</td>
<td></td>
<td>1.63 ± 0.08</td>
</tr>
<tr>
<td>3</td>
<td>5.86</td>
<td>1.57</td>
<td></td>
<td></td>
</tr>
<tr>
<td>4</td>
<td>5.84</td>
<td>1.57</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Fuel can No.</th>
<th>Pellet No.</th>
<th>Count rate (corrected)</th>
<th>Burn-up in at.-%</th>
<th>Mean burn-up as measured chemically</th>
</tr>
</thead>
<tbody>
<tr>
<td>5</td>
<td>1</td>
<td>5.88</td>
<td>1.58</td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>5.76</td>
<td>1.55</td>
<td></td>
<td>1.65 ± 0.08</td>
</tr>
<tr>
<td>3</td>
<td>5.73</td>
<td>1.54</td>
<td></td>
<td></td>
</tr>
<tr>
<td>4</td>
<td>5.86</td>
<td>1.57</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Fuel can No.</td>
<td>Pellet No.</td>
<td>Count rate (corrected)</td>
<td>Burn-up in at.-% as measured chemically</td>
<td>Mean burn-up as measured chemically</td>
</tr>
<tr>
<td>-------------</td>
<td>-----------</td>
<td>------------------------</td>
<td>----------------------------------------</td>
<td>-----------------------------------</td>
</tr>
<tr>
<td>6</td>
<td>1</td>
<td>5.46</td>
<td>1.47</td>
<td></td>
</tr>
<tr>
<td></td>
<td>2</td>
<td>5.51</td>
<td>1.48</td>
<td></td>
</tr>
<tr>
<td></td>
<td>3</td>
<td>5.49</td>
<td>1.47</td>
<td></td>
</tr>
<tr>
<td></td>
<td>4</td>
<td>5.50</td>
<td>1.48</td>
<td></td>
</tr>
<tr>
<td></td>
<td>5</td>
<td>5.61</td>
<td>1.51</td>
<td></td>
</tr>
<tr>
<td>7</td>
<td>1</td>
<td>5.17</td>
<td>1.39</td>
<td></td>
</tr>
<tr>
<td></td>
<td>2</td>
<td>5.13</td>
<td>1.38</td>
<td></td>
</tr>
<tr>
<td></td>
<td>3</td>
<td>5.13</td>
<td>1.38</td>
<td></td>
</tr>
<tr>
<td></td>
<td>4</td>
<td>5.26</td>
<td>1.41</td>
<td></td>
</tr>
<tr>
<td></td>
<td>5</td>
<td>5.39</td>
<td>1.45</td>
<td></td>
</tr>
<tr>
<td>8</td>
<td>1</td>
<td>3.17</td>
<td>0.85</td>
<td></td>
</tr>
<tr>
<td></td>
<td>2</td>
<td>3.00</td>
<td>0.81</td>
<td></td>
</tr>
<tr>
<td></td>
<td>3</td>
<td>2.92</td>
<td>0.78</td>
<td></td>
</tr>
<tr>
<td></td>
<td>4</td>
<td>2.82</td>
<td>0.76</td>
<td></td>
</tr>
<tr>
<td></td>
<td>5</td>
<td>2.81</td>
<td>0.75</td>
<td></td>
</tr>
<tr>
<td></td>
<td>6</td>
<td>2.87</td>
<td>0.77</td>
<td></td>
</tr>
</tbody>
</table>

**REFERENCES**

DISCUSSION

N. C. RASMUSSEN: Why is it so important that the sample be homogeneous?

A. J. FUDGE: No burn-up determination is possible unless the concentration or the total number of atoms of fissionable material before irradiation is known. If some other means of obtaining this exists, then integrated burn-up can be obtained. If it is not known, then the signal obtained will vary with the fissile concentration.
SYMPOSIUM ON NUCLEAR MATERIALS MANAGEMENT

HELD AT VIENNA, 30 AUGUST - 3 SEPTEMBER 1965

CHAIRMEN OF SESSIONS

Session II  D. JANISCH  United Kingdom Atomic Energy Authority, Risley
Session III  J. SORNEIN  Commissariat à l'énergie atomique, Paris
Session IV  K. NAKAJIMA  Atomic Fuel Corporation of Japan, Tokai-Mura
Session V  W. J. WRIGHT  Australian Atomic Energy Commission, Sutherland
Session VI  A. KAMAEV  Institute of Physics and Power, Moscow
Session VII  E. MALASEK  Czechoslovak Atomic Energy Commission, Prague
Session VIII  W. McGONNAGLE  Illinois Institute of Technology, Chicago
Session IX  M. J. HIGATSBERGER  Reactor Centre, Seibersdorf

SECRETARIAT OF THE SYMPOSIUM

Scientific Secretaries:  C.L.A. BUECHLER Division of Safeguards and Inspection
Y. MOTODA Division of Safeguards and Inspection
Administrative Secretary:  P. GHELARDONI Division of Scientific and Technical Information
Editor:  A. GUILLON Division of Scientific and Technical Information
Records Officer:  D. MITCHELL Division of Languages
# LIST OF PARTICIPANTS

<table>
<thead>
<tr>
<th>Name</th>
<th>Institution</th>
<th>Nominating State or Organization</th>
</tr>
</thead>
<tbody>
<tr>
<td>Agu, B. N. C.</td>
<td>Federal Radiation Protection Service, University of Ibadan</td>
<td>Nigeria</td>
</tr>
<tr>
<td>Andersson, L. H.</td>
<td>Research Institute of National Defence, Sundbyberg</td>
<td>Sweden</td>
</tr>
<tr>
<td>Artaud, J.</td>
<td>Commissariat à l'énergie atomique, Grenoble</td>
<td>France</td>
</tr>
<tr>
<td>Barendregt, T. J.</td>
<td>EUROCHEMIC, Mol, Belgium</td>
<td>OECD/ENEA</td>
</tr>
<tr>
<td>Bennett, C. A.</td>
<td>Battelle Northwest Laboratories, Richland, Wash.</td>
<td>United States of America</td>
</tr>
<tr>
<td>Billy, G.</td>
<td>Commissariat à l'énergie atomique, Paris</td>
<td>France</td>
</tr>
<tr>
<td>Bisby, H.</td>
<td>Atomic Energy Research Establishment, UKAEA, Harwell, Berks</td>
<td>United Kingdom</td>
</tr>
<tr>
<td>Blechscheidt, M.</td>
<td>Physikalisch-Technische Bundesanstalt, Braunschweig</td>
<td>Federal Republic of Germany</td>
</tr>
<tr>
<td>Bonnieux, J. M.</td>
<td>Commissariat à l'énergie atomique, Paris</td>
<td>France</td>
</tr>
<tr>
<td>Byrne, J. T.</td>
<td>Dow Chemical Company, Golden, Colo.</td>
<td>United States of America</td>
</tr>
<tr>
<td>Cardwell, R. G., Jr.</td>
<td>Oak Ridge National Laboratory, Oak Ridge, Tenn.</td>
<td>United States of America</td>
</tr>
<tr>
<td>Celeri, J.</td>
<td>Electricité de France, SEG, Paris</td>
<td>France</td>
</tr>
<tr>
<td>Cerrolaza, J. A.</td>
<td>Junta de Energía Nuclear, Madrid</td>
<td>Spain</td>
</tr>
<tr>
<td>Chenouard, J.</td>
<td>Commissariat à l'énergie atomique, Saclay</td>
<td>France</td>
</tr>
<tr>
<td>Colsmann, P.</td>
<td>Brookhaven National Laboratory, Upton, N. Y.</td>
<td>United States of America</td>
</tr>
<tr>
<td>Name</td>
<td>Institution</td>
<td>Nominating State or Organization</td>
</tr>
<tr>
<td>-------------------</td>
<td>------------------------------------------------------------------------------</td>
<td>----------------------------------</td>
</tr>
<tr>
<td>Donato, M.</td>
<td>EURATOM, Brussels, Belgium</td>
<td>EURATOM</td>
</tr>
<tr>
<td>Drujon, C.</td>
<td>Commissariat à l'énergie atomique, Paris</td>
<td>France</td>
</tr>
<tr>
<td>Elliott, F.</td>
<td>Windscale Works, UKAEA, Sellafield, Cumberland</td>
<td>United Kingdom</td>
</tr>
<tr>
<td>Elston, K. E.</td>
<td>Hydro-Electric Power Commission of Ontario, Toronto</td>
<td>Canada</td>
</tr>
<tr>
<td>Erbacher, W.</td>
<td>Oesterreichische Verbundgesellschaft, Vienna</td>
<td>Austria</td>
</tr>
<tr>
<td>Fletcher, G. W.</td>
<td>Atomic Energy of Canada Ltd., Ottawa</td>
<td>Canada</td>
</tr>
<tr>
<td>Forcella, A.</td>
<td>Allis-Chalmers Manufacturing Company, Bethesda, Md.</td>
<td>United States of America</td>
</tr>
<tr>
<td>Frank, P.</td>
<td>NEWAG, Maria-Enzersdorf, N. Ö.</td>
<td>Austria</td>
</tr>
<tr>
<td>Franzen, F.</td>
<td>Institut für Reaktorsicherheit d. TÜV. e. V.</td>
<td>Federal Republic of Germany</td>
</tr>
<tr>
<td>Frenzel, W.</td>
<td>EUROCHEMIC, Mol, Belgium</td>
<td>OECD/ENEA</td>
</tr>
<tr>
<td>Fruchard, Y.</td>
<td>Commissariat à l'énergie atomique, Marcoule</td>
<td>France</td>
</tr>
<tr>
<td>Fudge, A. I.</td>
<td>Atomic Energy Research Establishment, UKAEA, Harwell, Berks</td>
<td>United Kingdom</td>
</tr>
<tr>
<td>Gilles, A.</td>
<td>Commissariat à l'énergie atomique, Fontenay-aux-Roses</td>
<td>France</td>
</tr>
<tr>
<td>Hazelton, L. D.</td>
<td>Dow Chemical Company, Golden, Colo.</td>
<td>United States of America</td>
</tr>
<tr>
<td>Hein, H. J.</td>
<td>Gesellschaft für Wiederaufarbeitung von Kernbrennstoffen m. b. H., Frankfurt</td>
<td>Federal Republic of Germany</td>
</tr>
<tr>
<td>Higatsberger, M. J.</td>
<td>Reactor Centre, Seibersdorf</td>
<td>Austria</td>
</tr>
<tr>
<td>Highfill, J. P.</td>
<td>New Brunswick Laboratory, New Brunswick, N. J.</td>
<td>United States of America</td>
</tr>
<tr>
<td>Name</td>
<td>Institution</td>
<td>Nominating State or Organization</td>
</tr>
<tr>
<td>-----------------</td>
<td>------------------------------------------------------------------------</td>
<td>----------------------------------</td>
</tr>
<tr>
<td>Hildenbrand, G.</td>
<td>Siemens-Schuckertwerke A.G., Erlangen</td>
<td>Federal Republic of Germany</td>
</tr>
<tr>
<td>Hiramatsu, I.</td>
<td>Japan Atomic Energy Research Institute, Tokai-mura</td>
<td>Japan</td>
</tr>
<tr>
<td>Hood, St.C.C.</td>
<td>United Kingdom Atomic Energy Authority, London</td>
<td>United Kingdom</td>
</tr>
<tr>
<td>Hydén, L.</td>
<td>A.B. Atomenergi, Stockholm</td>
<td>Sweden</td>
</tr>
<tr>
<td>Jantisch, D.</td>
<td>United Kingdom Atomic Energy Authority, Production Group, Risley, Warrington, Lancs.</td>
<td>United Kingdom</td>
</tr>
<tr>
<td>Jasmy, G.R.</td>
<td>Oak Ridge Y-12 Plant, Oak Ridge, Tenn.</td>
<td>United States of America</td>
</tr>
<tr>
<td>Jennekens, J.</td>
<td>Atomic Energy Control Board, Ottawa</td>
<td>Canada</td>
</tr>
<tr>
<td>Jessnitzer, K.</td>
<td>Leybold Hochvakuuum Anlagen G.m.b.H., Cologne</td>
<td>Federal Republic of Germany</td>
</tr>
<tr>
<td>Jolly, D.C.</td>
<td>OECD Dragon Project, AEE, Winfrith, Dorset, UK</td>
<td>OECD/ENEA</td>
</tr>
<tr>
<td>Kamaev, A.</td>
<td>Institute of Physics and Power, Moscow</td>
<td>Union of Soviet Socialist Republics</td>
</tr>
<tr>
<td>Kauffman, G.F.</td>
<td>Goodyear Atomic Corporation, Piketon, Ohio</td>
<td>United States of America</td>
</tr>
<tr>
<td>Ketzinel, Z.</td>
<td>Israel Atomic Energy Commission, Tel-Aviv</td>
<td>Israel</td>
</tr>
<tr>
<td>Kiestling, R.F.</td>
<td>Technischer Ueberwachungs-Verein Bayern e.V., Munich</td>
<td>Federal Republic of Germany</td>
</tr>
<tr>
<td>Kinderman, E.M.</td>
<td>Stanford Research Institute, Menlo Park, Calif.</td>
<td>United States of America</td>
</tr>
<tr>
<td>Klinken, A.B.</td>
<td>OECD/ENEA, Paris, France</td>
<td>OECD/ENEA</td>
</tr>
<tr>
<td>Knudsen, P.</td>
<td>Danish Atomic Energy Commission, Roskilde</td>
<td>Denmark</td>
</tr>
<tr>
<td>Koning, H.</td>
<td>Institut für Theoretische Physik, University of Innsbruck</td>
<td>Austria</td>
</tr>
<tr>
<td>Name</td>
<td>Institution</td>
<td>Nominating State or Organization</td>
</tr>
<tr>
<td>--------------------</td>
<td>------------------------------------------------------------------------------</td>
<td>----------------------------------</td>
</tr>
<tr>
<td>Kops, S.</td>
<td>United States Atomic Energy Commission, Argonne, Ill.</td>
<td>United States of America</td>
</tr>
<tr>
<td>Kotschi, H.</td>
<td>Technischer Uberwachungs-Verein Bayern e.V., Munich</td>
<td>Federal Republic of Germany</td>
</tr>
<tr>
<td>Kuznetsov, F. M.</td>
<td>Institute of Physics and Power, Moscow</td>
<td>Union of Soviet Socialist Republics</td>
</tr>
<tr>
<td>Lang, F.</td>
<td>Bundeskanzleramt, Vienna</td>
<td>Austria</td>
</tr>
<tr>
<td>Laurent, H.</td>
<td>Centro Comune di Ricerche, Ispra, Varese, Italy</td>
<td>EURATOM</td>
</tr>
<tr>
<td>Laurent, R.</td>
<td>Centro Comune di Ricerche, Ispra, Varese, Italy</td>
<td>EURATOM</td>
</tr>
<tr>
<td>Ledich, A.</td>
<td>Bundesministerium für Lebensmitteluntersuchung, Vienna</td>
<td>Austria</td>
</tr>
<tr>
<td>Lesur, P.</td>
<td>Commissariat à l'énergie atomique, Fontenay-aux-Roses</td>
<td>France</td>
</tr>
<tr>
<td>Lieser, K. H.</td>
<td>Technische Hochschule Darmstadt</td>
<td>Federal Republic of Germany</td>
</tr>
<tr>
<td>Lint, J. M. , Van</td>
<td>(See under &quot;Van Lint&quot;)</td>
<td></td>
</tr>
<tr>
<td>Lockett, E.</td>
<td>Atomic Energy Establishment, UKAEA, Winfrith, Dorset</td>
<td>United Kingdom</td>
</tr>
<tr>
<td>Longo, G.</td>
<td>EURATOM, Brussels, Belgium</td>
<td>EURATOM</td>
</tr>
<tr>
<td>Lorrain, C.</td>
<td>Commissariat à l'énergie atomique, Le Bouchet</td>
<td>France</td>
</tr>
<tr>
<td>Malasek, E.</td>
<td>Czechoslovak Atomic Energy Commission, Prague</td>
<td>Czechoslovakia</td>
</tr>
<tr>
<td>McCluen, W. D.</td>
<td>Oak Ridge Gaseous Diffusion Plant, Oak Ridge, Tenn.</td>
<td>United States of America</td>
</tr>
<tr>
<td>McGinley, F.</td>
<td>United States Atomic Energy Commission, Grand Junction, Colo.</td>
<td>United States of America</td>
</tr>
<tr>
<td>McGonnagle, W. J.</td>
<td>Illinois Institute of Technology, Chicago, Ill.</td>
<td>United States of America</td>
</tr>
<tr>
<td>Merz, E.</td>
<td>Kernforschungsanlage Jülich</td>
<td>Federal Republic of Germany</td>
</tr>
<tr>
<td>Name</td>
<td>Institution</td>
<td>Nominating State or Organization</td>
</tr>
<tr>
<td>--------------</td>
<td>------------------------------------------------------------------------------</td>
<td>----------------------------------</td>
</tr>
<tr>
<td>Metz, C.F.</td>
<td>Los Alamos Scientific Laboratory, Los Alamos, N. Mex.</td>
<td>United States of America</td>
</tr>
<tr>
<td>Milner, G.</td>
<td>Atomic Energy Research Establishment, UKAEA, Harwell, Berks</td>
<td>United Kingdom</td>
</tr>
<tr>
<td>Moeken, H. H. Ph.</td>
<td>EUROCHEMIC, Mol, Belgium</td>
<td>OECD/ENEA</td>
</tr>
<tr>
<td>Möller, E.</td>
<td>Technischer Ueberwachungs-Verein Norddeutschland e.V., Hamburg-Stellingen</td>
<td>Federal Republic of Germany</td>
</tr>
<tr>
<td>Montojo, Rosa</td>
<td>Junta de Energia Nuclear, Madrid</td>
<td>Spain</td>
</tr>
<tr>
<td>Mostert, P.</td>
<td>Gemeenschappelijke Kernenergie Centrale Nederland, Arnhem</td>
<td>Netherlands</td>
</tr>
<tr>
<td>Nadal, J.</td>
<td>Société industrielle de combustible nucléaire, Annecy</td>
<td>France</td>
</tr>
<tr>
<td>Nakajima, K.</td>
<td>Atomic Fuel Corporation of Japan, Tokai-mura</td>
<td>Japan</td>
</tr>
<tr>
<td>Neven, Z.</td>
<td>Helsingør Skibsbaerftog Maskinbyggeri A/S Elsinore</td>
<td>Denmark</td>
</tr>
<tr>
<td>Oszuszyk, F.</td>
<td>Oesterreichische Elektrizitätswirtschafts A.G., Vienna</td>
<td>Austria</td>
</tr>
<tr>
<td>Pictet, J.M.</td>
<td>Eidgenössisches Institut für Reaktorforschung, Würenlingen</td>
<td>Switzerland</td>
</tr>
<tr>
<td>Pilgenröther, A.</td>
<td>Nuklear-Chemie- und Metallurgie G. m. b. H., Hanau</td>
<td>Federal Republic of Germany</td>
</tr>
<tr>
<td>Pinochet, J.</td>
<td>Commissariat à l'énergie atomique, Saclay</td>
<td>France</td>
</tr>
<tr>
<td>Prawitz, J.</td>
<td>Research Institute of National Defence, Stockholm</td>
<td>Sweden</td>
</tr>
<tr>
<td>Prem, L.</td>
<td>NEWAG, Maria-Enzersdorf, N. U.</td>
<td>Austria</td>
</tr>
<tr>
<td>Putz, F.</td>
<td>Oesterreichische Studiengesellschaft für Atomenergie G. m. b. H., Vienna</td>
<td>Austria</td>
</tr>
<tr>
<td>Rasmussen, N.C.</td>
<td>Massachusetts Institute of Technology, Cambridge, Mass.</td>
<td>United States of America</td>
</tr>
<tr>
<td>Regnaud, F.</td>
<td>Commissariat à l'énergie atomique, Fontenay-aux-Roses</td>
<td>France</td>
</tr>
<tr>
<td>Name</td>
<td>Institution</td>
<td>Nominating State or Organization</td>
</tr>
<tr>
<td>------------</td>
<td>------------------------------------------------------------------------------</td>
<td>----------------------------------</td>
</tr>
<tr>
<td>Riedel, E.</td>
<td>Kernkraftwerk Obrigheim, Stuttgart</td>
<td>Federal Republic of Germany</td>
</tr>
<tr>
<td>Ronteix, F.</td>
<td>Commissariat à l'énergie atomique, Fontenay-aux-Roses</td>
<td>France</td>
</tr>
<tr>
<td>Ross, A.</td>
<td>United Kingdom Atomic Energy Authority, Risley, Warrington, Lancs</td>
<td>United Kingdom</td>
</tr>
<tr>
<td>Salvi, P.</td>
<td>Ministero dell'Industria e del Commercio, Rome</td>
<td>Italy</td>
</tr>
<tr>
<td>Santarelli, E.</td>
<td>Comitato Nazionale per l'Energia Nucleare, Rome</td>
<td>Italy</td>
</tr>
<tr>
<td>Schimits, J.</td>
<td>Centre d'étude de l'énergie nucléaire, Mol</td>
<td>Belgium</td>
</tr>
<tr>
<td>Schmidt, O.</td>
<td>Technischer Ueberwachungs-Verein Baden e.V., Mannheim</td>
<td>Federal Republic of Germany</td>
</tr>
<tr>
<td>Schmitt, B.F.</td>
<td>Bundesanstalt für Materialprüfung, Berlin</td>
<td>Federal Republic of Germany</td>
</tr>
<tr>
<td>Schmitt, M.</td>
<td>EURATOM, Brussels, Belgium</td>
<td>EURATOM</td>
</tr>
<tr>
<td>Schüller, W.</td>
<td>EUROCHEMIC, Mol, Belgium</td>
<td>OECD/ ENEA</td>
</tr>
<tr>
<td>Shelley, W.J.</td>
<td>Mallinckrodt Chemical Works, St. Charles, Miss.</td>
<td>United States of America</td>
</tr>
<tr>
<td>Sornein, J.</td>
<td>Commissariat à l'énergie atomique, Fontenay-aux-Roses</td>
<td>France</td>
</tr>
<tr>
<td>Tarrice, R.R.</td>
<td>Stanford Research Institute, Menlo Park, Calif.</td>
<td>United States of America</td>
</tr>
<tr>
<td>Tingey, F.H.</td>
<td>National Reactor Testing Station, Phillips Petroleum Co., Idaho Falls, Idaho</td>
<td>United States of America</td>
</tr>
<tr>
<td>Valetino, M.F.</td>
<td>Combustion Engineering, Inc., Windsor, Conn.</td>
<td>United States of America</td>
</tr>
<tr>
<td>Van Lint, J.M.</td>
<td>BelgoNucléaire, Brussels</td>
<td>Belgium</td>
</tr>
<tr>
<td>Vertès, P.</td>
<td>Société de raffinage d'uranium, Courbevoie</td>
<td>France</td>
</tr>
<tr>
<td>Vetter, H.</td>
<td>Technischer Ueberwachungs-Verein Baden e.V., Mannheim</td>
<td>Federal Republic of Germany</td>
</tr>
<tr>
<td>Name</td>
<td>Institution</td>
<td>Nominating State or Organization</td>
</tr>
<tr>
<td>--------------</td>
<td>------------------------------------------------------------------------------</td>
<td>----------------------------------</td>
</tr>
<tr>
<td>Wanlin, J.</td>
<td>Saint-Gobain Techniques Nouvelles, Courbevoie</td>
<td>France</td>
</tr>
<tr>
<td>Weinzierl, P.</td>
<td>Oesterreichische Studiengesellschaft für Atomenergie G. m. b. H., Vienna</td>
<td>Austria</td>
</tr>
<tr>
<td>Welch, G.A.</td>
<td>Dounreay Experimental Reactor Establishment, UKAEA, Thurso, Caithness</td>
<td>United Kingdom</td>
</tr>
<tr>
<td>Whitham, G.K.</td>
<td>Argonne National Laboratory, Idaho Falls, Idaho</td>
<td>United States of America</td>
</tr>
<tr>
<td>Wright, W.J.</td>
<td>Australian Atomic Energy Commission, Sutherland, N. S. W.</td>
<td>Australia</td>
</tr>
<tr>
<td>Zühlke, P.</td>
<td>Gesellschaft zur Wiederaufarbeitung von Kernbrennstoffen m. b. H., Frankfurt</td>
<td>Federal Republic of Germany</td>
</tr>
</tbody>
</table>
AUTHOR INDEX

Arabic numerals underlined refer to the first page of a paper by the author concerned.
Further Arabic numerals denote comments and questions in discussions.
Literature references are not indexed.

Al-Khafaji, T. : 809
Artaud, J. : 353, 373, 656
Bennett, C. A. : 373, 465, 480, 746, 796
Billy, G. : 315, 323
Bir, R. : 707
Bisby, H. : 433
Bod, R. : 353
Bokelund, H. : 527
Brown, D. L. : 375
Buck, P. : 749
Buechler, C. L. A. : 49, 688, 847
Burtscher, A. : 817
Byrne, J. T. : 447, 462, 489, 656, 676, 688, 704, 816
Cardwell, R. G., Jr. : 65, 86, 235, 257, 611
Ceja, S. N. : 463
Cerrolaza, J. A. : 413
Chenouard, J. : 257, 707
Colsmann, P. : 677, 688, 797
Cordin, R. A. : 141, 151
Davidson, A. S. : 325
Downing, J. J. : 323, 462, 612
Dubovskv, B. G. : 515
Elliott, F. : 325
Feldman, M. J. : 123
Fletcher, G. W. : 237, 257
Forcella, A. A. : 153
Foster, E. : 867
Frenzel, W. : 205
Fruchard, Y. : 311, 481, 489
Fudge, A. J. : 689, 847, 867, 878
George, D. E. : 30, 175, 202, 323, 411, 568, 612, 613, 625, 689
Gilles, A. : 167, 175
Giraud, R. : 571
Hazelton, L. D. : 89, 121
Hick, H. : 817
Higatsberger, M. J. : 292, 480, 626, 746, 817, 828, 847
Highfill, J. P. : 639, 656, 689
Hiramatsu, I. : 213
Hocking, D. R. : 229
Hoq, St. C. C. : 597, 611, 625
Janisch, D. : 151, 202
Jasny, G. R. : 189, 201, 311, 568
Jenkinson, W. G. : 237
Jennekens, J. : 373
Kamaev, A. V. : 515
Kauffman, G. F. : 719
Kazi, A. H. : 514, 749, 773
Kinderman, E. M. : 31, 275, 292
Kops, S. : 43, 49, 611
Kuehn, M. N. : 547
Kuznetsov, F. M. : 515
Lago, A. : 413
Langridge, R. W. : 375
Lesur, P. : 571
Lewis, B. S. : 293
Lint, J. M., van: see Van Lint
Lockett, E. : 257
Lorrain, C. : 177, 353
Lucas, Monique : 707
Machida, C. : 213
Mayman, S. A. : 829
McCluen, W. D. : 341
McDowell, S. C. T. : 585
McGinley, F. E. : 375, 391
McGonnagle, W. J. : 851
Metc, C. F. : 657, 676, 688, 705
Mills, J. S. : 275
Milner, G. W. C. : 691, 704
Mooek, H. H. Ph. : 527
Montejo, Rosa M. : 413
Motoda, Y. : 462
Murphy, L. : 867
Nadal, J. : 51
Nakajima, K. : 259, 373, 626
O'Leary, W. J. : 153
AUTHOR INDEX

Phillips, G.: 691
Pinochet, J.: 51
Powell, R.: 325
Putz, F.: 809
Raber, M.: 499
Rasmussen, N.C.: 828, 829, 847, 878
Regnaud, F.: 629, 705
Reid, H.B.: 237
Rodden, C.J.: 639
Ronteix, F.: 19, 30, 203, 257, 323, 612, 796
Rosztoczy, Z.R.: 775
Rumpold, K.: 817
Saito, N.: 259
Santarelli, E.: 3
Schmets, J.: 30, 463, 489, 514, 626
Schüller, W.: 205
Shelley, W.J.: 393, 411, 547, 557
Shields, W.R.: 411, 480, 737, 746
Sofer, G.A.: 749
Sornein, J.: 30, 49, 175, 201, 236, 391, 557, 568, 625
Sovka, J.A.: 829
Spalding, T.R.: 123
Swinburn, K.A.: 325
Tabor, C.D.: 719
Tarrice, R.R.: 31
Thiriet, L.: 571
Tingey, F.H.: 121, 202, 293, 311, 491, 537
Tomonto, J.R.: 749
Valerino, M.F.: 775, 796
Van Lint, J.M.: 559, 568
Vertès, P.: 411
Vladyдов, G.M.: 515
Wanlin, J.: 571
Waterbury, G.R.: 657
Weinzierl, P.: 809, 816, 817, 847
Welch, G.A.: 202, 462, 489, 848
Whitham, G.K.: 123
Wright, W.J.: 86, 121, 229, 235, 257, 463, 772
Ziegler, W.A.: 393
Orders for Agency publications can be placed with your bookseller or any of our sales agents listed below:

**ARGENTINA**  
Comisión Nacional de Energía Atómica  
Avenida del Libertador  
General San Martin 8250  
Buenos Aires - Suc. 29

**AUSTRALIA**  
Hunter Publications,  
23 McKillop Street  
Melbourne, C.J

**AUSTRIA**  
Georg Fromme & Co.  
Spengergasse 39  
A-1050, Vienna V

**BELGIUM**  
Office international de librairie  
30, avenue Marnix  
Brussels 5

**BRAZIL**  
Livraria Kosmos Editora  
Rua do Rosario, 135-137.  
Rio de Janeiro  
Agencia Exponte Oscar M. Silva  
Rua Xavier de Toledo, 140-1º Andar  
(Caixa Postal No. 5,614)  
São Paulo

**BYELORUSSIAN SOVIET SOCIALIST REPUBLIC**  
See under USSR

**CANADA**  
The Queen's Printer  
Ottawa, Ontario

**CHINA (Taiwan)**  
Books and Scientific Supplies  
Service, Ltd.,  
P.O. Box 83  
Taipei

**CZECHOSLOVAK SOVIET SOCIALIST REPUBLIC**  
S.N.T.L.  
Spolenska 51  
Nové Mesto  
Prague 1

**DENMARK**  
Ejnar Munksgaard Ltd.  
6 Nørregade  
Copenhagen K

**FINLAND**  
Akateeminen Kirjakauppa  
Keskuskatu 2  
Helsinki

**FRANCE**  
Office international de documentation et librairie  
48, rue Gay-Lussac  
Paris 5e

**GERMANY, Federal Republic of**  
R. Oldenbourg -  
Rosenheimer Strasse 145  
8 Munich 8

**HUNGARY**  
Kultura'  
Hungarian Trading Co. for Books and Newspapers  
P.O.B. 149  
Budapest 62

**ISRAEL**  
Heiliger and Co.  
3 Nathan Strauss Street  
Jerusalem

**ITALY**  
Agenzia Editoriale Internazionale  
Organizzazioni Universali (A.E.I.O.U.)  
Via Meravigli 16  
Milan

**JAPAN**  
Maruzen Company Ltd.  
6, Tori Nichome  
Nihonbashi  
(P.O. Box 605)  
Tokyo Central

**MEXICO**  
Libreria Internacional  
Av. Sonora 206  
Mexico 11, D.F.

**NETHERLANDS**  
N.V. Martinus Nijhoff  
Lange Voorhout 9  
The Hague

**NEW ZEALAND**  
Whitcombe & Tombs, Ltd.  
G.P.O. Box 1894  
Wellington, C.J
NORWAY
Johan Grundt Tanum
Karl Johans gate 43
Oslo

PAKISTAN
Karachi Education Society
Haroon Chambers
South Napier Road
(P.O. Box No. 4866)
Karachi 2

POLAND
Ośrodek Rozpowszechniania
Wydawnictw Naukowych
Polska Akademia Nauk
Pałac Kultury i Nauki
Warsaw

ROMANIA
Cartimex
Rue A. Briand 14-18
Bucarest

SOUTH AFRICA
Van Schaik’s Bookstore (Pty) Ltd.
Libri Building
Church Street
(P.O. Box 724)
Pretoria

SPAIN
Libraría Bosch
Ronda de la Universidad 11
Barcelona

SWEDEN
C.E. Fritzes Kungl. Hovbokhandel
Fredsgatan 2
Stockholm 16

SWITZERLAND
Librairie Payot
Rue Grenus 6
1211 Geneva 11

TURKEY
Librairie Hachette
469, Istiklal Caddesi
Beyoğlu, Istanbul

UKRAINIAN SOVIET SOCIALIST REPUBLIC
See under USSR

UNION OF SOVIET SOCIALIST REPUBLICS

UNITED KINGDOM OF GREAT BRITAIN AND NORTHERN IRELAND
Her Majesty’s Stationery Office
P.O. Box 569
London, S.E.1

UNITED STATES OF AMERICA
National Agency for International Publications, Inc.
317 East 34th Street
New York, N.Y. 10016

VENEZUELA
Sr. Braulio Gabriel Chacares
Gobernador a Candilito 37
Santa Rosalia
(Apartado Postal 8092)
Camistas D.F.

YUGOSLAVIA
Jugoslovenska Knjiga
Temžije 27
Belgrade

IAEA publications can also be purchased retail at the United Nations Bookshop at United Nations Headquarters, New York, at the news-stand at the Agency’s Headquarters, Vienna, and at most conferences, symposia and seminars organized by the Agency.

In order to facilitate the distribution of its publications, the Agency is prepared to accept payment in UNESCO coupons or in local currencies.

Orders and inquiries from countries where sales agents have not yet been appointed may be sent to:

Distribution and Sales Group, International Atomic Energy Agency,
Kämtner Ring 11, A-1010, Vienna I, Austria