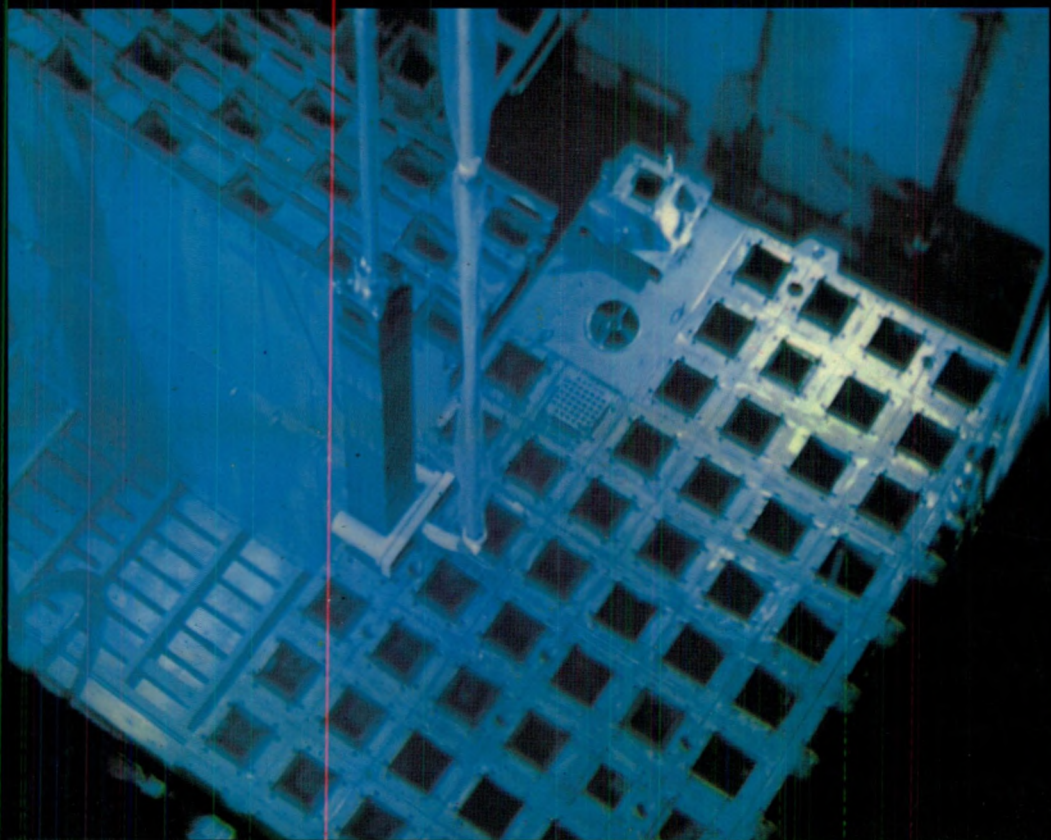


# NUCLEAR VOL.2 SAFEGUARDS TECHNOLOGY 1986

PROCEEDINGS OF A SYMPOSIUM  
VIENNA, 10 - 14 NOVEMBER 1986



INTERNATIONAL ATOMIC ENERGY AGENCY, VIENNA, 1987

**NUCLEAR SAFEGUARDS TECHNOLOGY 1986**  
**VOLUME 2**

PROCEEDINGS SERIES

# NUCLEAR SAFEGUARDS TECHNOLOGY 1986

PROCEEDINGS OF AN INTERNATIONAL SYMPOSIUM  
ON NUCLEAR MATERIAL SAFEGUARDS  
ORGANIZED BY THE  
INTERNATIONAL ATOMIC ENERGY AGENCY  
AND HELD IN VIENNA, 10-14 NOVEMBER 1986

*In two volumes*

## VOLUME 2

INTERNATIONAL ATOMIC ENERGY AGENCY  
VIENNA, 1987

**NUCLEAR SAFEGUARDS TECHNOLOGY 1986**  
**IAEA, VIENNA, 1987**  
**STI/PUB/722**  
**ISBN-92-0-070287-2**



## FOREWORD

This publication presents the results of the sixth in a series of international symposia on nuclear material safeguards. The symposia are sponsored by the IAEA at four-year intervals in order to foster a broad exchange of information related to technologies needed in international safeguards. The Symposium was held in Vienna during the week of 10-14 November 1986 and, as indicated by the list of participants at the end of Volume 2, attracted close to three hundred participants from some thirty Member States and four international organizations.

From a first glance at these Proceedings it might seem that there has been little progress since the last symposium, in 1982. A closer examination, however, should prove that this impression is not correct. It is true that some papers can be described as updated versions of earlier work presented elsewhere. The policy of the Symposium organizers specifically permits this. Moreover, many participants do not routinely receive the proceedings of professional society meetings at which safeguards relevant work is usually presented. However, there was still much that was new.

Development efforts related to safeguards for reprocessing plants constituted over twenty per cent of the programme. For example, in Paper IAEA-SM-293/24 the authors suggest that under certain admittedly optimistic (but in the authors' opinions not unrealistic) assumptions the uncertainty in a 12-month material balance for a 4 t/d reprocessing plant might be as low as 0.1% (standard deviation). Other papers present results of over four years of field testing of near real time material accountancy at a plant in Japan (IAEA-SM-293/34), and results for a lesser period of time at a plant in Scotland (IAEA-SM-293/22).

Papers reporting work on destructive and non-destructive measurement procedures or equipment constituted another thirty per cent of the programme, more if measurements in reprocessing and poster presentations are included. In honour of the tenth anniversary of the founding of the Safeguards Analytical Laboratory, two sessions were devoted to a review of destructive analytical measurement procedures.

Some subjects received only minor attention during the Symposium. The statistical theory of random sampling, safeguards for uranium enrichment plants, material accountancy systems and several other topics appear only incidentally in the programme, but primarily because there are few remaining problems, not because there is little remaining interest.

## **EDITORIAL NOTE**

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**CONTAINMENT-SURVEILLANCE  
TECHNOLOGY**

**(Session 9)**

**Chairman**

**Yu.I. KONNOV**

**Union of Soviet Socialist Republics**



# DEVELOPMENT OF CONTAINMENT AND SURVEILLANCE SYSTEMS AT THE JAPAN ATOMIC ENERGY RESEARCH INSTITUTE (JAERI)

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## Abstract

DEVELOPMENT OF CONTAINMENT AND SURVEILLANCE SYSTEMS AT THE JAPAN ATOMIC ENERGY RESEARCH INSTITUTE (JAERI).

Three C/S systems have been developed for international safeguards use: (1) The compact closed circuit TV (CCTV) surveillance system, COSMOS, is designed to replace the currently used Twin Minolta system. COSMOS is able to record more than 30 000 still pictures in three months, during which it need only operate by battery. A unique one-shot video tape recorder (VTR) was developed for COSMOS, based on the 8 mm VTR technique. A prototype of COSMOS was completed by the end of October 1986, and demonstrated the performance expected. (2) The electronic verifier of the COBRA seal is designed to read and digitize a COBRA seal pattern in situ. Comparisons between the digitized patterns are made by calculating correlation coefficients of these patterns, one to be verified and one taken as reference when the seal is initially installed. The experimental results obtained with the prototype verifier have demonstrated that it provides simple, accurate quantitative measures for COBRA seal verification. (3) The fast critical assembly (FCA) portal and penetration monitor is a comprehensive C/S system developed for the JAERI fast critical facility. The monitor, which is a multi-sensor system to satisfy tamper safe requirements, consists of a metal detector, a CCTV surveillance system, motion detectors, and a control/recording unit. The monitor was completed in 1985. A field trial of the entire system was started in September 1986 under field conditions using regularly scheduled inspectors to carry out inspections to obtain performance data and to gain international credibility for the system.

## 1. INTRODUCTION

The facilities under IAEA safeguards have been increasing in number while the IAEA has been facing budgetary limitations. Newly built facilities are becoming more complicated and automated, and accessibility to nuclear material in them for inspection activities is becoming more difficult. Under these circumstances, containment and surveillance (C/S) measures become increasingly important.

Various kinds of proven techniques are now available for C/S equipment, whereas some existing safeguards devices have become outdated. The development of an in situ verifiable seal to replace the metal cap seal (types E and X), and a video surveillance system to replace the Twin Minolta, are badly needed.

With these circumstances in mind, the development of the compact closed circuit TV (CCTV) surveillance system, COSMOS, and the electronic verifier of the COBRA seal, was started in order to assist IAEA safeguards. Development of these devices is based on experience acquired in developing the fast critical assembly (FCA) portal and penetration monitor, which was initiated in 1979.

After several modifications, and following joint field tests by the IAEA and the State inspection authorities, the portal and penetration monitor was completed in 1985 and the field operation test began in September 1986 for twelve months.

This report describes the design features and performance data of (1) COSMOS, (2) the electronic verifier of the COBRA seal, and (3) the FCA portal and penetration monitor.

## 2. COMPACT CCTV SURVEILLANCE SYSTEM

### 2.1. Design criteria

The Compact Surveillance Monitoring System (COSMOS) has been designed to replace the Twin Minolta, now the standard IAEA surveillance device.

The Twin Minolta, a simple but very reliable surveillance device, has the important features of compactness and being battery driven, valuable from the viewpoint of minimum intrusion into facility operation. Despite these advantages it has too small a capacity for recording pictures - 7000 pictures is the maximum as long as 8 mm film cameras are used. Furthermore, the manufacturer of the 8 mm camera, which is used in the Twin Minolta, has stopped production because of reduced demand for it.

The CCTV technique has been highly developed, and reliable devices are available for a surveillance system. Therefore, COSMOS was designed as a CCTV system, whereby more information such as time, date, etc., not obtainable in the Twin Minolta, can be easily annotated on the tape.

The following are the basic requirements for COSMOS as a replacement for Twin Minolta:

- Compact and portable configuration
- Battery driven for three months
- Picture storage capacity of 30 000
- High reliability
- High quality of pictures
- Tamper resistance
- Reasonable cost.

The picture storage capacity of 30 000 is needed where a standard procedure of a surveillance device is such that pictures can be recorded at five-minute intervals for three months.

To be battery driven for three months is a very severe requirement with a system composed of CCTV devices, even if the power consumption is very low. The power consumption of a small CCD (solid state image sensor) camera, used in robots, and of an 8 mm VTR is as low as 3.6 and 6 W, respectively. The energy required to continuously operate a 10 W device for three months is 21 600 W·h (= 10 W x 24 h/d x 90 d). On the other hand, stored energy in a 12 V 50 A·h car battery is 600 W·h. Comparing these figures, it may be concluded that the continuous driving of CCTV devices for three months by a reasonable size battery is not feasible. Therefore, to reduce the power consumption the system should be switched off during non-recording and be powered only for the recording period. But problems exist when a normal VTR is used with a frequent switch-on and -off mode. Frequent switch-on and -off of a VTR will shorten the lifetime of those mechanical parts which have to move at high speed. Another problem is energy waste even with the switch-on and -off mode operation. Since it takes a few seconds for a recording head to reach the constant rotating speed and for recording to be initiated, energy is wasted during this idling period.

Taking into consideration these factors, it was considered necessary to develop a new type of 8 mm VTR suitable for COSMOS and based on commercially available techniques.

A further factor should be borne in mind - sufficient time to supply the instrument when a standard inspection device is designed. Should such instruments be safeguards specific not many will be required, which means that the price will be high and a manufacturer may not be interested in producing it. To avoid this situation, safeguards standard instruments should be assembled from commercially available equipment or be designed for general purpose or public use.

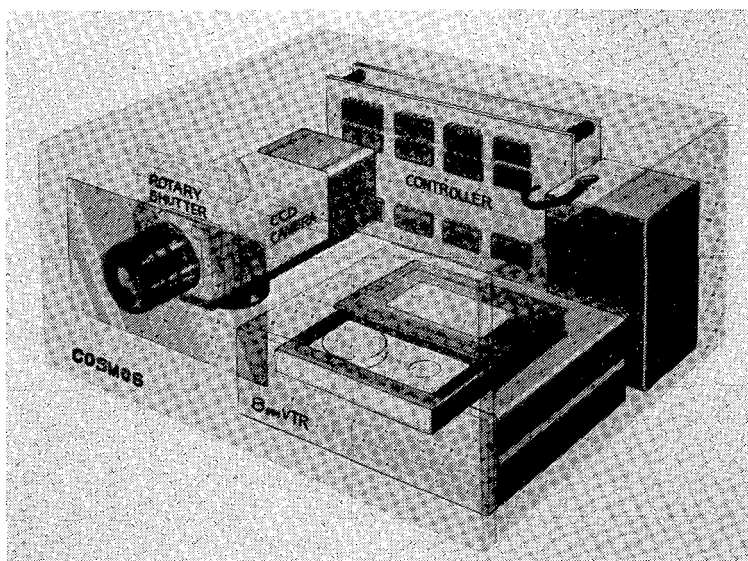


FIG. 1. Conceptual drawing of COSMOS.

As already explained, the COSMOS design is for general applications. It was originally designed as the surveillance instrument in safeguards, but was also designed for general applications as an observation tool. For example, COSMOS can be used for long term observation of wildlife or natural phenomena where the power supply relies on batteries.

If an increased demand for COSMOS from outside the safeguards community can be expected, its price will be reduced and procurement and maintenance will be easier. Furthermore, continuous production and manufacturer's improvements can be expected.

## 2.2. Description of COSMOS

A conceptual drawing of a completed COSMOS system is shown in Fig. 1. It consists of:

- (1) A CCD camera with a rotary shutter
- (2) One or two one-shot 8 mm VTRs
- (3) A system controller
- (4) Battery pack
- (5) Tamper safe housing case.

A set-up unit and miniature video monitor are used for the initial system installation.



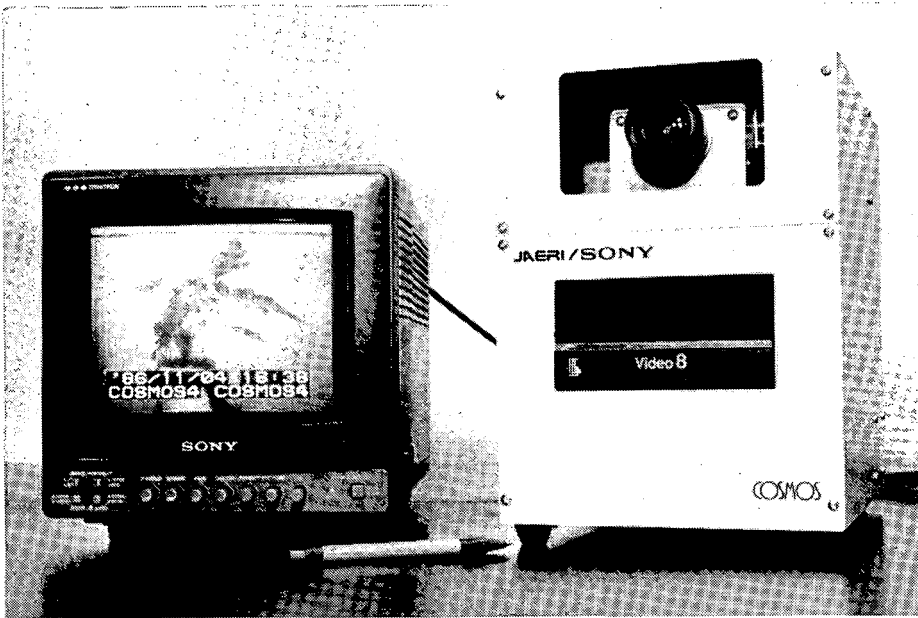


FIG. 2. Prototype unit of COSMOS.

In the newly developed 8 mm VTR, a video recording head rotates with a speed  $1/8$  that of a normal 8 mm VTR; the tape speed is also slower. During non-recording intervals every component except the system controller is switched off. By the random time interval base, the controller starts up the whole system, then the rotary shutter is opened for  $1/30$  s. The video recording head of the VTR makes only three turns. One still video image on the CCD camera is slowly read out and recorded on a tape during these three turns. Then the system is switched off within 2 s after the start. Time, date and other information are annotated on a video tape.

The estimated energy consumption of the field use model is 200 W·h for 30 000 picture recordings for three months when an IC (integrated circuit) is used for the system control.

A COSMOS prototype was completed in October 1986 (see Fig. 2). Its expected performance, namely very good picture quality and very low energy consumption (250 W·h), was demonstrated. The performance test will be continued until the end of March 1987 to obtain data on reliability, energy consumption, failure rate, etc. After modifications based on the test results of the prototype, demonstration units will be built by October 1987.

### 3. ELECTRONIC VERIFIER OF COBRA SEAL

#### 3.1. COBRA seal

A few tens of thousands of metal cap seals (type E and X seals) are in routine IAEA use. These seals are removed during an inspection visit and returned to IAEA Headquarters for verification. This process results in a delay in verification and resolution of anomalies. An in situ verifiable seal eliminates this delay and provides timely information to enable the operator and the inspectors to resolve anomalies during an inspection visit. The in situ verification also enables repeated use of the same seal without its being removed for verification, thus providing a considerable saving in effort compared with that required for conventional sealing, sampling, replacement and subsequent verification at IAEA Headquarters.

The COBRA seal was developed at the Sandia National Laboratories as the in situ verifiable sealing system [1]. It consists of a fibre optic loop with a seal body and a seal pattern recorder/verifier. The seal pattern is photographed by the seal recorder/verifier. Visual verification by photograph overlay comparison is effective and accurate under normal circumstances. However, when the number of seals in question is large, or the quality of two photographs varies, as is often the case in film processing, to avoid ambiguity and to help the inspector make an objective judgement, the development of a simpler and quantitative means for in situ verification is needed.

Taking into consideration the appropriate features of the COBRA seal as a replacement for the type E seal, and the necessity of easier approaches for in situ verification, the development of the electronic verifier of the COBRA seal has been initiated at JAERI. The possibility also exists for the electronic verifier to be applied as a joint or common seal between the IAEA and the State inspection authorities.

#### 3.2. Development of COBRA seal verifier

The design of an electronic verifier is based on the following considerations:

- In situ verification of the COBRA seal
- To be simple and easily quantitative
- Portable equipment
- Low cost.

The prototype electronic verifier, shown in Fig. 3, consists of the following:

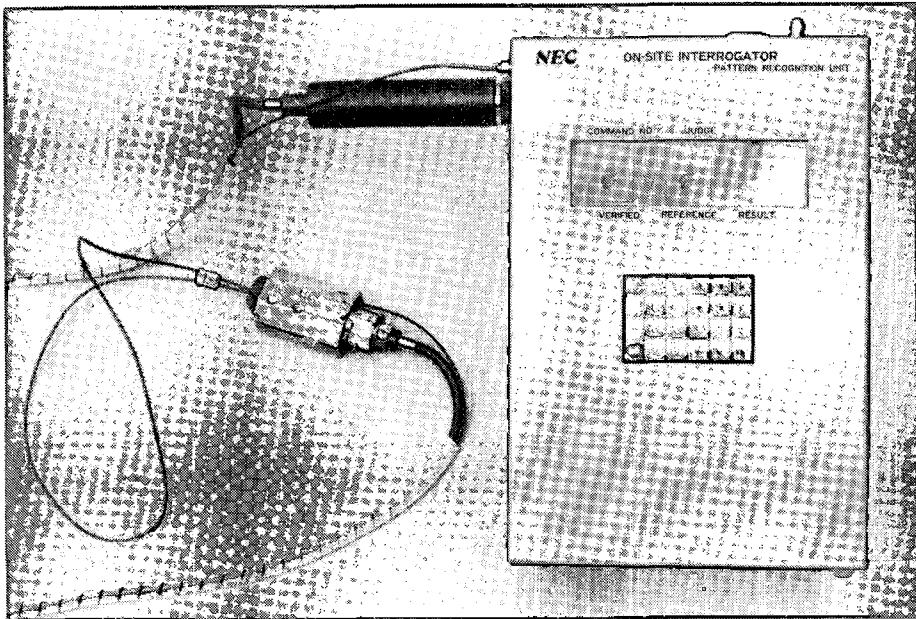


FIG. 3. Prototype of electronic verifier for COBRA seal.

- (1) Probe, to view the seal pattern: The seal body is inserted into a viewer receptacle where one end of the seal loop is illuminated and the light traverses uncut fibres to the other end of the loop; then the unique pattern is produced by the uncut fibres.
- (2) Pattern detection part, to generate the image of a seal pattern by the CCD camera, the video signal of which is sampled and digitized.
- (3) Memories, to store the digitized patterns temporarily.
- (4) Auxiliary memory, to store the digitized reference patterns in the compact floppy disk with a capacity of 200 patterns for later use.
- (5) Processor and comparator, to calculate the correlation coefficients between the digitized patterns.
- (6) Display, to display the correlation coefficients.

### 3.3. Experimental results obtained with the prototype verifier

Comparisons have been made of seal patterns for 35 seals of various lengths ranging from 10 cm to 1 m. The results of the measurements are:

- (1) Uncertainty of the electronic data processing: Each seal pattern was compared ten times with its reference while

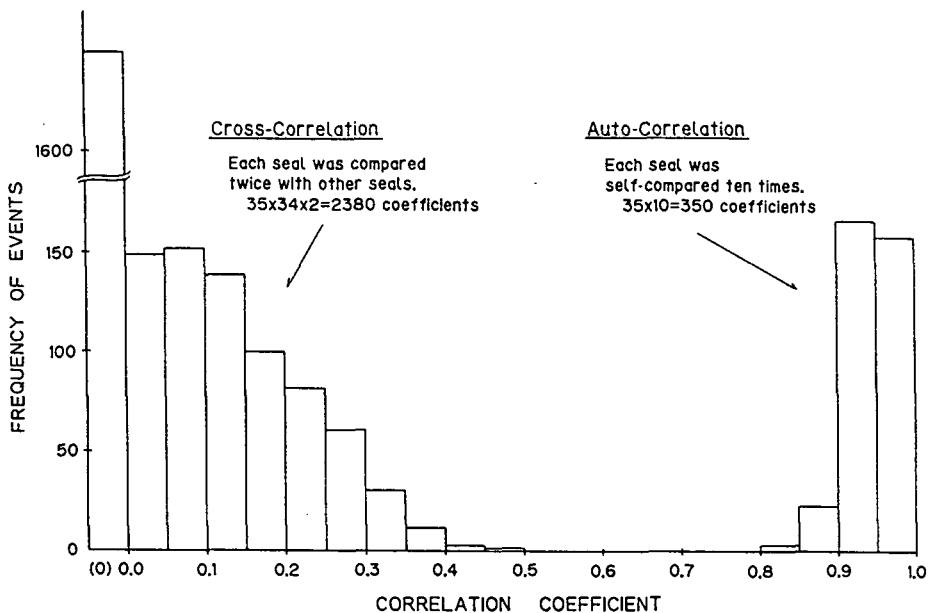


FIG. 4. Distribution of correlation coefficients of pairs of patterns for 35 COBRA seals.

the seal was kept inserted in the receptacle. The average correlation coefficient was  $0.95 \pm 0.02$  for the 350 data.

- (2) Reproducibility of image positioning on a CCD camera:  
The position of the image focused on the CCD sensor will be changed slightly owing to mechanical looseness at the seal receptacle. Each seal was repeatedly inserted into, and ejected from, the receptacle and the correlation coefficients were measured. The change of correlation was less than 4% for 350 measurements. The effect of looseness of other parts of the optics system was found to be about 4%. Combining these uncertainties, the total of 6% uncertainty in the correlation coefficients is caused by looseness of the optical connection.
- (3) Correlation coefficients measured. A histogram of correlation coefficients obtained by repeated comparisons of the same seal and of different seals is shown in Fig. 4. The repeated comparisons of the same seal (auto-correlation) are highly correlated while the patterns of the different seals (cross-correlation) show little correlation. The difference in correlation coefficients between auto- and cross-correlation is large enough for inspectors to decide to accept or to reject the seal.

### 3.4. Summary

Experimental results have confirmed that the electronic verifier is sufficiently promising and provides a simple but unambiguous quantitative means of verifying the COBRA seal in situ. Plotting of digitized patterns is available if necessary, together with the correlation coefficient, and this provides a means of visual comparison when it is desirable for redundancy in verification.

The modified model is being redesigned to improve the reproducibility of an image on the image sensor and to reduce the size and the weight of the verifier housing.

## 4. PORTAL AND PENETRATION MONITORS FOR THE FAST CRITICAL ASSEMBLY (FCA)

### 4.1. The FCA from the safeguards viewpoint

Because of experimental requirements the fast critical assembly of JAERI possesses a large inventory of fissile material in relatively pure form. Moreover, the fact that this nuclear material is clean, virtually unirradiated, accessible and easily handled poses unique safeguards problems. Consequently, the FCA is considered to be among the most sensitive of facilities from the safeguards viewpoint; thus, inspection activities are quite frequent. Manpower and radiation exposure problems, accompanied by frequent NDA based inspections, are a burden for both the inspectorates and the facility operator. In the hope of alleviating these burdens, the development of C/S measures for the FCA was initiated.

### 4.2. Concept of C/S application to the FCA

Part of the nuclear material is stored in birdcages, which are placed in the fuel storage vaults. The remainder is loaded into the zero power reactor for experiments. Nuclear material in the birdcages is secured by inspectorate seals, but for material in the reactor it is difficult to apply seals because of the reactor structure and the experimental procedures.

The main inspection effort is inventory verification by NDA of non-sealed material in the reactor.

The reactor building provides an ideal containment measure because of its explosion-proof, airtight structure and the limited number of penetrations, most of which are seldom

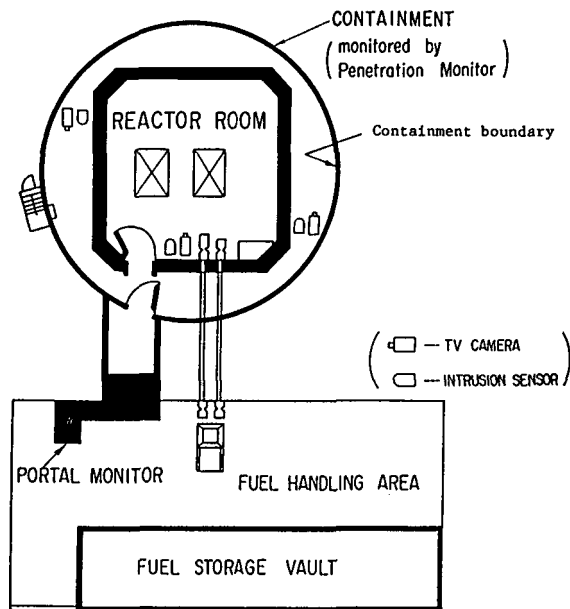


FIG. 5. Concept of C/S system for FCA.

opened. Only the personal doorway is frequently used. Therefore, the combination of monitoring by penetration monitor of containment boundaries and all their penetrations except for the doorway, and monitoring by portal monitor, provides complete coverage of all realistic diversion routes. The assurance that the integrity of the containment itself remains unimpaired is provided by reviewing the operation record of the monitor.

The concept of the FCA C/S system is shown in Fig. 5.

#### 4.3. Description of FCA portal and penetration monitors

Detailed descriptions of the FCA C/S system are given in Refs [2-4] and include the portal monitor and the penetration monitor as shown in Fig. 5.

##### Portal monitor

The function of the portal monitor is to detect undeclared removal of nuclear material from the reactor building through the doorway. The major characteristics are:

- (1) A walk-through metal detector, to detect metal nuclear material. FCA nuclear materials are in the form of

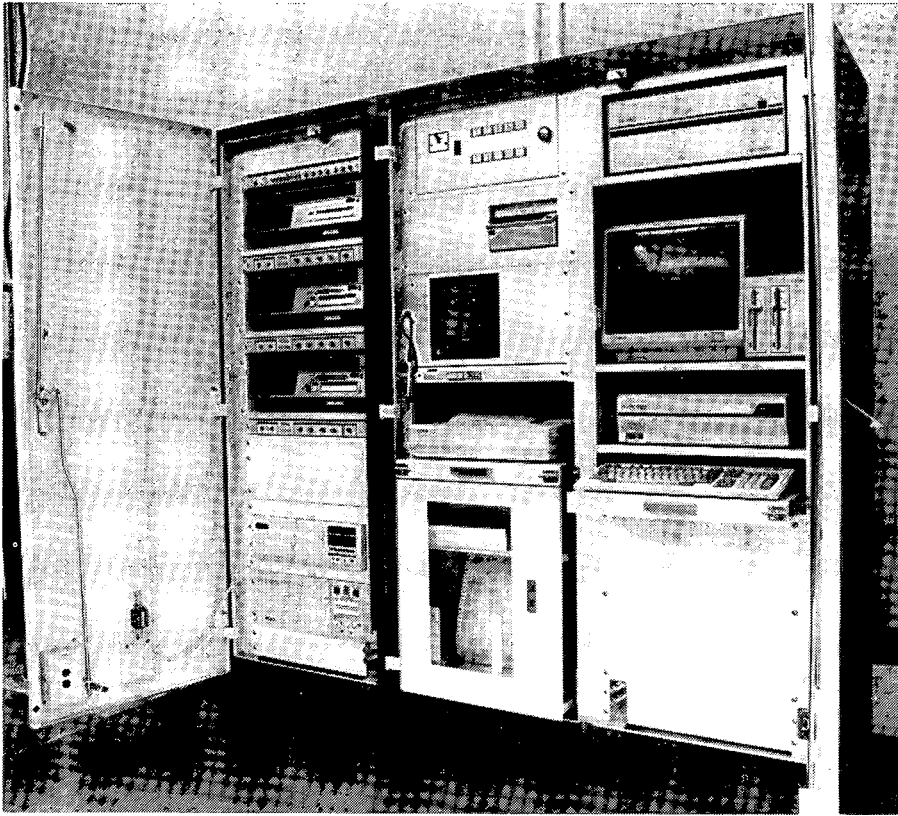


FIG. 6. Control/recording unit of FCA portal monitor.

variously sized metal plates. The metal detector has a special coil arrangement to detect a single metal coupon of 2 in x 2 in x 1/16 in<sup>1</sup> regardless of its orientation relative to the passage direction.

- (2) Video surveillance, to support the unattended operation.
- (3) Tamper indication, to indicate tamper attempts with the system.

The control/recording unit of the portal monitor is shown in Fig. 6.

#### Penetration monitor

The penetration monitor is designed for surveillance of diversion routes through containment boundaries, and of

<sup>1</sup> 1 in = 2.54 cm.



safeguards related activities which involve the resolution of metal nuclear material for bypassing the portal monitor (the detection of metal solution by a metal detector is rarely possible).

A number of TV cameras and infrared motion detectors are distributed in the reactor building to provide complete coverage of all realistic diversion routes.

#### 4.4. Operation of the portal and penetration monitors

The portal monitor and the penetration monitor are individually controlled by their own computers and specific inspector interactive software. Every event on the monitor is recorded in the bubble cassette memory and is printed out at a fixed time of day. The data in the bubble memory are retained during an inspection period. Video recording is initiated by motion detection or anomaly detection, and by the random time interval.

Inspectors are asked to follow instructions displayed on the screen to review the data obtained during the previous inspection period and to restart the system for the next inspection. Video tape review is also part of the data review.

Should anomalies be recorded, inspectors will analyse the data and review the recorded tape. When they conclude that the recorded anomaly is not false, appropriate response procedures will be carried out.

#### 4.5. Present status of the portal and penetration monitors

The FCA C/S system development was basically completed in 1985. Since 1981 several functional and performance tests have been carried out jointly by the IAEA, the State authorities and JAERI, the final ones being done in February 1986.

Several times the system was modified, based on comments made during these tests and is now fully functional.

The field trial operation by the IAEA and the State authorities began in September 1986 with assessment by both inspectorate parties and the facility operator to see whether the system should be routinely implemented. Inspection, training, performance monitoring, testing and maintenance procedures required in routine use will also be tested during the trial operation.

A significant relief of the work-loads of both the inspectors and the operator, which arise from the NDA

inspection, is expected once the FCA C/S system is put into routine use for international safeguards.

### ACKNOWLEDGEMENT

The authors gratefully acknowledge the significant contributions made by Sony Corp. for the development of COSMOS, and of NEC Ltd for the development of the electronic verifier of the COBRA seal. Thanks are gratefully extended to P. Vodrazka, A. Matolcsy, K. Taylor and L. Watkins of the IAEA for valuable discussions during the FCA C/S system development. This work has been performed within the framework of the Japan Support Programme for Agency Safeguards (JASPAS).

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# REAL TIME ITEM ACCOUNTING SYSTEM FOR SPENT FUEL AT RECEIVING AND STORAGE AREAS

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## Abstract

### REAL TIME ITEM ACCOUNTING SYSTEM FOR SPENT FUEL AT RECEIVING AND STORAGE AREAS.

In 1983 a real time item accounting system (RIAS) for spent fuel at the receiving and storage areas was implemented as a new JASPAS (Japan Support Programme for Agency Safeguards) programme at the Tokai Reprocessing Plant. This is an effective measurement system for safeguards implementation by the continuous monitoring of movement of fuel assemblies from the receiving to shipping areas and the instantaneous reproduction of transfer paths of any spent fuels and inventories, etc. This equipment was installed at the end of October 1985, and a field test is being carried out to ensure that the device is reliable for safeguards.

## 1. INTRODUCTION

A survey and evaluation of various detectors and a conceptual design were done in June 1982 and March 1983, respectively. The specifications of the system were determined after the conceptual design was reviewed in December 1983. On this basis the real time item accounting system (RIAS) for spent fuel at the receiving and storage areas was proposed. This system uses a crane position detector and existing signals.

The system was installed in two stages. First, crane position detectors were installed at the fuel storage pool (FSP), the data collection and management module (DCMM) and the data input unit. Second, crane position detectors were installed at the cask unloading pool (CUP), the fuel transfer pool (FTP), and the input equipment of the mechanical processing cell (MPC)<sup>1</sup>.

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<sup>1</sup> This is a Furukawa Electric Co. system.

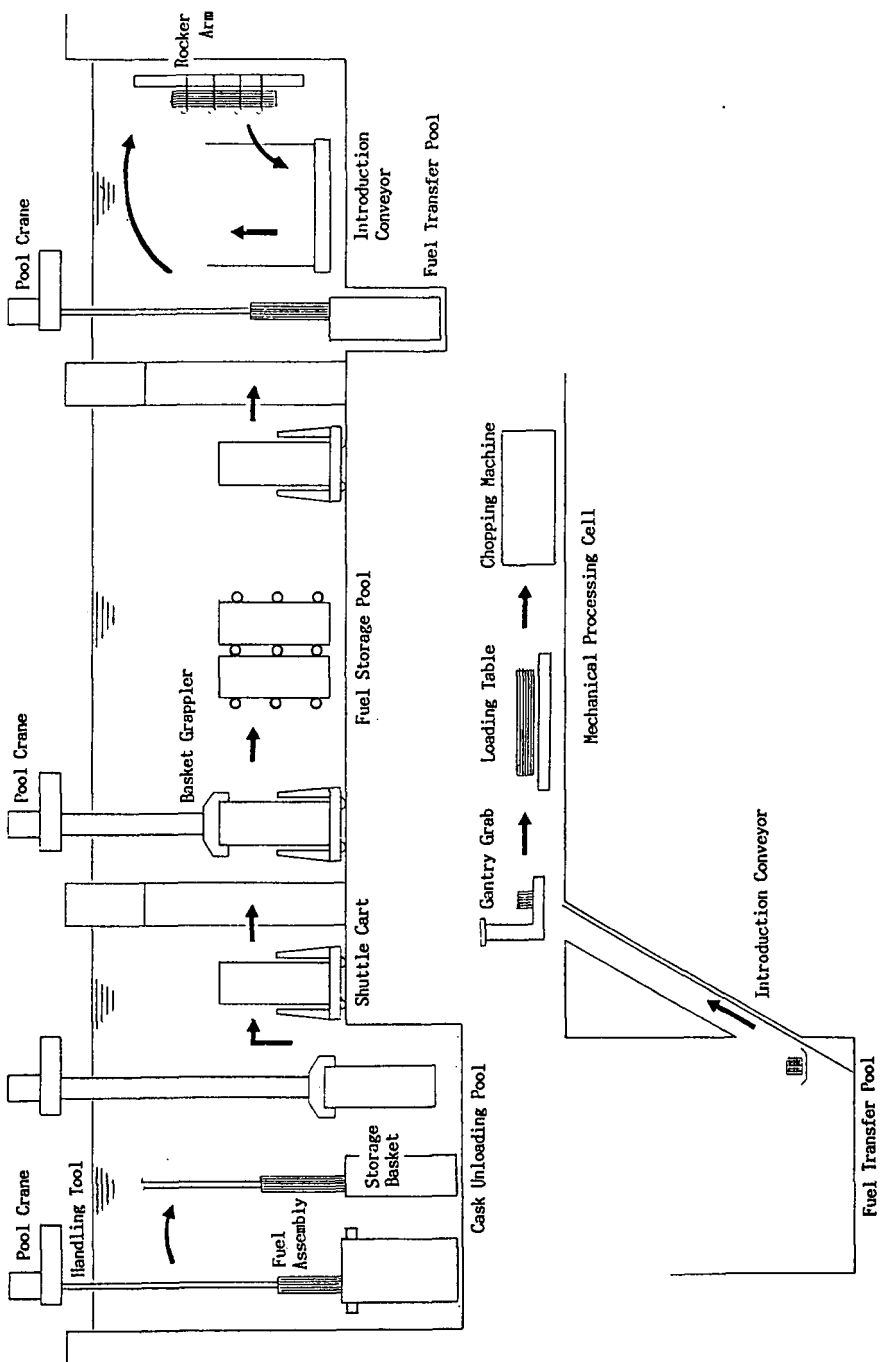


FIG. 1. Flow diagram of fuel assemblies.

Spent fuel in the fuel storage basket is transferred by a pool crane. Thus, by this system, a crane transfer path is traced by combining the crane position detector with the existing position signals. The system can monitor all transfer of fuels which are removed from the casks and transferred to the MPC, and can control the data of physical inventory and location of fuels in real time. Moreover, it stores information concerning all past fuel transfer so that, if required, it can reproduce the inventory status and fuel transfer path in any period on the display or the printer.

## 2. FUEL HANDLING AND TRANSFER OPERATIONS

The fuel assembly flow diagram is given in Fig. 1. Fuel handling and transfer operations in this area are as follows.

### 2.1. Cask unloading pool (CUP)

Fuel is removed from the cask by using the pool crane and fuel handling tool, then inserted into the storage basket, which is then placed by the pool crane on the shuttle cart which runs between the CUP and the fuel storage pool (FSP), and placed in the FSP.

### 2.2. Fuel storage pool (FSP)

After the basket has been transferred to the FSP it is lifted from the shuttle cart by the pool crane and placed in a storage rack. The basket containing the fuel for reprocessing is placed by the pool crane on the shuttle cart which runs between the FSP and the fuel transfer pool (FTP), and put into the FTP.

### 2.3. Fuel transfer pool (FTP)

After the basket has been transferred to the FTP a fuel assembly is removed from the basket by the fuel handling tool and placed on the rocker arm, which tilts from the vertical position to the horizontal one and places a fuel assembly on the conveyor, which then carries the assembly through a tunnel into the mechanical processing cell (MPC).

### 2.4. Mechanical processing cell (MPC)

After a fuel assembly on the conveyor has been carried to the MPC, it is transferred by the gantry grab to the loading table and inserted into the chopping machine by the pusher. A fuel assembly is chopped into equal parts.

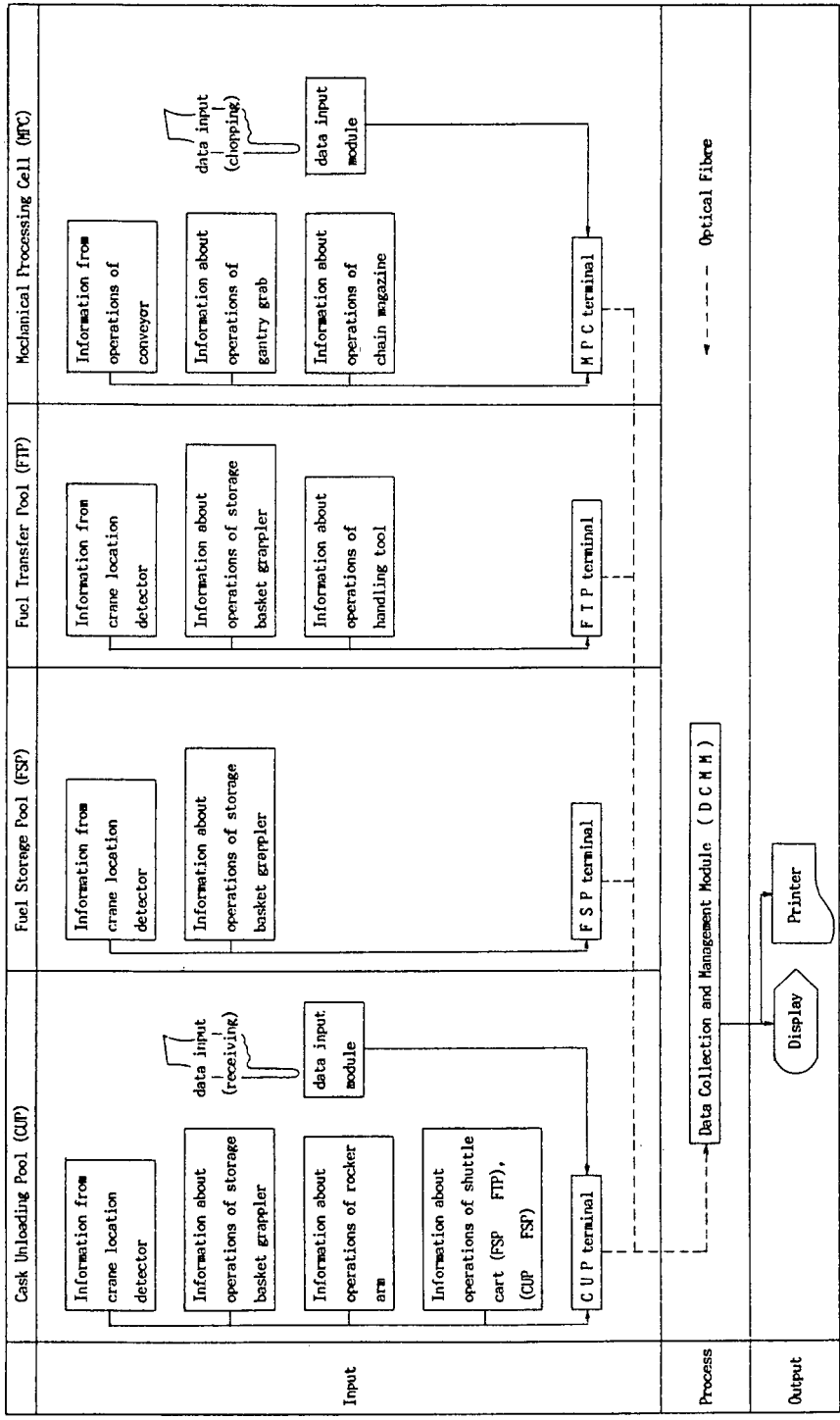


FIG. 2. Block diagram of the RIAS.



### 3. COMPONENTS

A system block diagram is shown in Fig. 2. Each monitoring sub-area is equipped with a monitoring terminal to collect the data on the fuel operation that is carried out in each area and transmitted to the data collection and management module (DCMM).

However, signals of each shuttle cart and rocker arm will be connected with the CUP monitoring terminal. The data from the input module will be transmitted to the CUP and MPC monitoring terminals.

#### 3.1. Data collection and management module (DCMM)

The data collection and management module processes the data from each monitoring terminal by a processing program. As a 576 kbyte microcomputer is used, it is compact and its functions are equivalent to a minicomputer. It has a higher availability and reliability because of a backup battery and memory. Any trouble or interruption in the transmission path and monitoring terminal is detected by a response to the questions/answers from each monitoring terminal at a specified interval. All information about receiving/shipping and transfer of fuels is stored in the memory and simultaneously written into the floppy disk.

#### 3.2. Monitoring terminals

The monitoring terminals are installed in four monitoring areas, and information on the fuel transfer is transmitted to the DCMM by an established form. The interface input signal from other equipment and details of monitoring terminals are as follows:

- (i) Monitoring terminal of CUP
  - CUP crane loading status signal (loading, unloading)
  - Status signal of the shuttle cart between the CUP and FSP (arrival, travelling)
  - Status signal of the shuttle cart between the FSP and FTP (arrival and travelling)
  - Status signal of the FTP rocker arm (up, down, etc.)
- (ii) FSP monitoring terminal
  - FSP crane loading status signal (loading, unloading)
- (iii) FTP monitoring terminal
  - FTP crane loading status signal (loading, unloading)
  - Handling tool status signal (loading (fuel), unloading)
- (iv) MPC monitoring terminal
  - Conveyor status signal (fuel transfer between FTP and MPC)

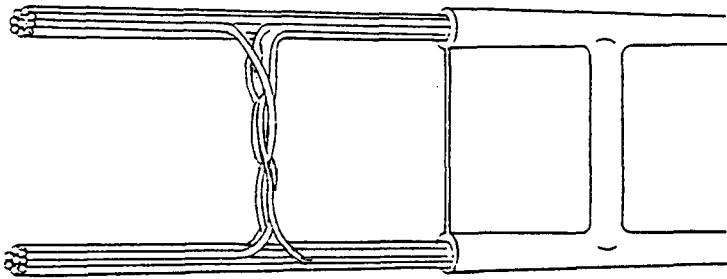


FIG. 3. Address cable.

- Gantry grab status signal (fuel transfer from conveyor to loading table)
- Loading table cart status signal (fuel transfer from loading table to chopping machine)
- Chain magazine status signal (open/close, last cut)

### 3.3. Crane position detector

The crane position detector detects travel/traverse positions and transmits them to the DCMM. The following description concerns the position detector using an inductive radio address cable, which is shown in Fig. 3.

The position detection (PD) is classified into two types, an absolute position detection (APD, position detection pitch: 10 cm), with an inductive radio system and an address cable, composed of multiple cross-parallel cables with assorted cross-distance and, second, a high resolution position detection system (HRPD, position detection pitch: 1 cm) with signals received from two cross-parallel cables.

In the PD system the oscillator signal generated at the vehicle station is transmitted to the address cable by an antenna. The phases of electrical signals produced by electromagnetic induction in each pair of cables are compared to ascertain the absolute address. For convenience of description, as shown in Fig. 4, two cross-parallel cables are used as examples. Two signal phases induced in one cross-pair and one parallel pair are compared at the receiver, and if two signals are co-phase then the vehicle position is determined as address "0", and if two signals are in-phase then as address "1". In practical use the address cable is composed of  $N + 1$  pairs needed to get  $2^N$  addresses. And the basic principle of the HRPD system is that the APD resolution is divided into 1 cm pitch by phase signals received from two cross-parallel cables,

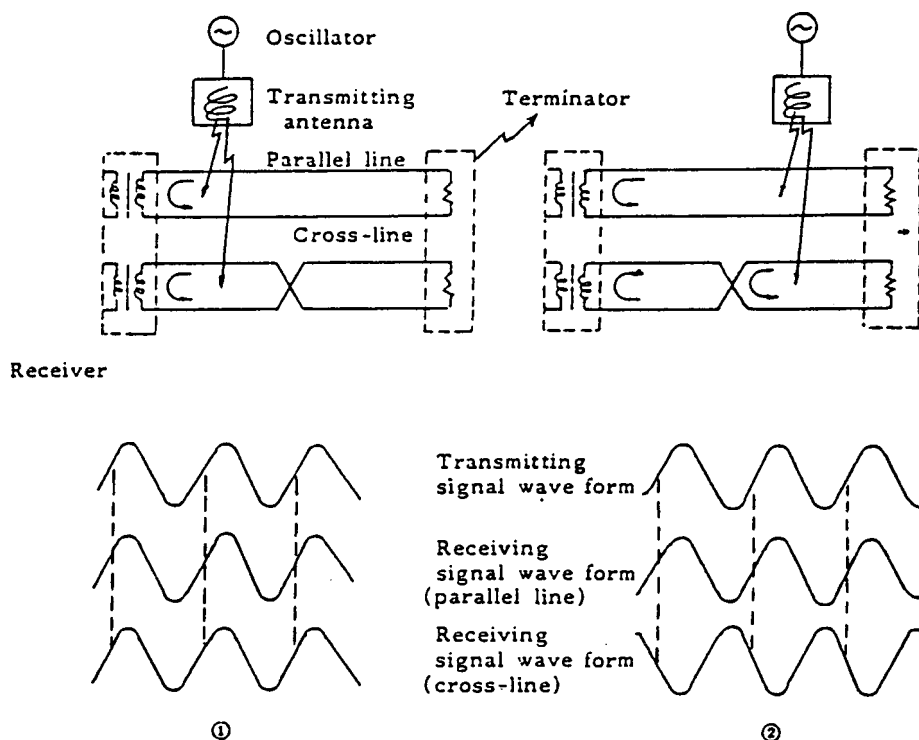


FIG. 4. Phases within the address cable.

and the unique data processing is combined with APD to establish a practical PD system.

This system has many advantages, for example no contact is needed at position detection, the absolute position can be detected and high reliability is achieved even in a bad environment, such as dust, oil, steam, etc. Also, it has a high resistance to external noise and signal fluctuations. This is due to the phase detection technique and unique address code, the "Random Address Code", shown in Fig. 5, which reduces cross-talk between pairs and eliminates variation of phase constant.

### 3.4. Data input module

RIAS is provided with three data input modules which need to be totally tamperproof. One is used to input data such as fuel identification number, kind of fuel, receiving date and time, etc., when spent fuels are received. The second inputs data such as fuel identification number, kind of fuel and chopping date and time when a fuel assembly was chopped. The

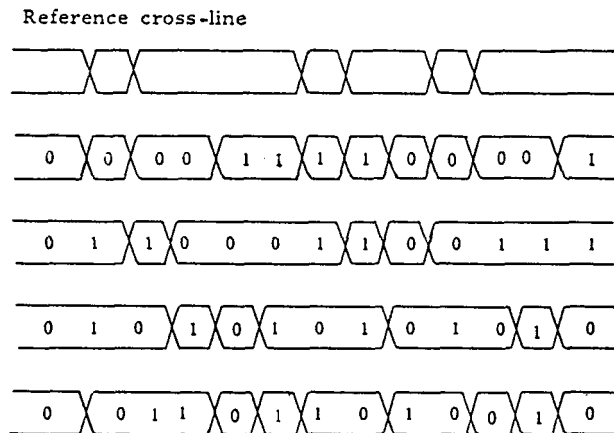


FIG. 5. Random address code.

third renews the stored information when the system is under maintenance. In addition, this module may display and print the inventory and fuel transfer information. The operator should first input his registered password.

#### 4. FUNCTIONS

RIAS has the following functions.

##### 4.1. Real time monitoring of fuel transfer (transfer monitoring function)

The transfer of fuel removed from the cask and inserted into the storage basket at the CUP is continuously traced and monitored in real time, during the process from storage in the pool to chopping by the MPC. In other words, the movement is monitored of each pool crane and basket grapppler of the CUP, FSP and FTP, shuttle carts between the pools, and the rocker arm.

##### 4.2. Real time management of fuel inventory (inventory management function)

The inventory information in the monitoring area is managed in real time by this function, which is operated by information obtained by the above transfer monitoring function and the manual input data on fuel receiving and fuel chopping processes. The commands designated by the DCMM keyboard operation actuate the CRT display and printer to output four

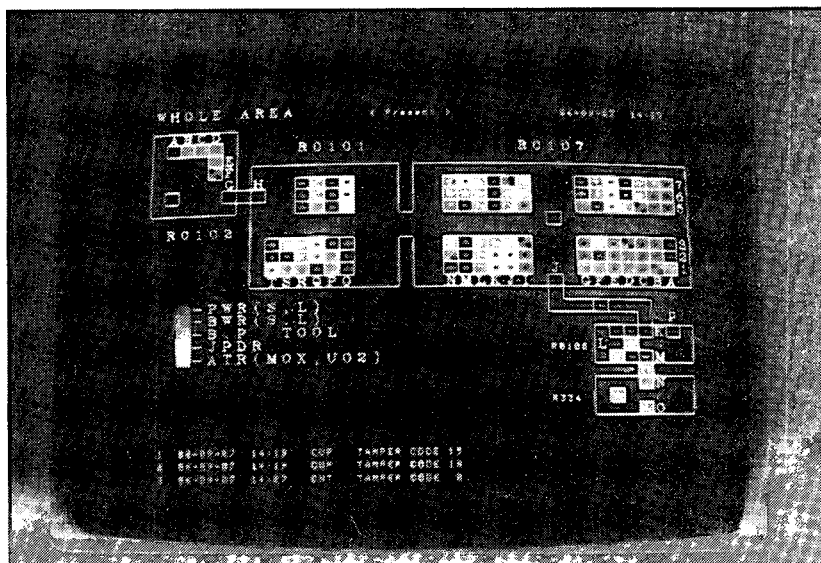


FIG. 6. Graphic display for the whole monitoring area.

items (graphic data display, if necessary) for an easier check of the inventory status. This is done as follows:

- (i) Presence of fuel in the monitoring area. Graphic display is given to make it easy to check the presence of stored fuel, fuel identification number, and the type of fuel for the whole monitoring area or sub-area. A graphic display for the whole area is shown in Fig. 6.
- (ii) Indication of the fuel volume in the monitoring area. The overall volume of actual storage or volume of each fuel shall be displayed for the whole monitoring area or each sub-area.
- (iii) Display of fuel information. Input data of the stored fuels are displayed.
- (iv) Preparation of daily report. A daily report is prepared in a specified format.

#### 4.3. Reproduction of fuel transfer operation (reproduction function)

All data available at each operation, such as fuel receiving, shipping or transfer, are stored as historical data. This function reproduces past fuel operation or inventory status on to the CRT display unit or printer in

accordance with the historical data. The reproduction contents are of the following four types:

- (i) Transfer path (starting point to stopping point), date and time of a specified fuel
- (ii) All fuel operations in a specified time
- (iii) Inventory status at a specified time
- (iv) Type, position and time of anomalous operation generated in a specified period.

#### 4.4. Detection of anomalous operation for spent fuel in connection with diversion (anomalous operation detection function)

The transfer monitoring function and the inventory management function are both used to detect anomalous operation in connection with diversion. This enables timely issuance of the report. Apart from an anomalous operation, reversed transfer can be carried out when unavoidable circumstances arise in the process. Detectable anomalous operations are:

- (i) After the fuel is inserted into the CUP basket, it may be transferred to another position without using manual data input.
- (ii) The basket may be placed in any position other than in the specified ones such as in storage racks or at the stop position of the shuttle cart at each pool.
- (iii) The fuel or basket may be transferred to a position where another fuel or basket is stored.

#### 4.5. Detection of abnormality of system components (tamper detection function)

Unreasonable human interference with system components, and component troubles can be detected and a timely report issued. Some of the detectable items are:

- (i) Open/close operation of the doors of the DCMM, the monitoring terminal unit and the data input unit
- (ii) Transmission failure between the DCMM and one of the monitoring terminals
- (iii) Suspension of the DCMM power supply
- (iv) Failure of a DCMM disk
- (v) Abnormality of crane position detectors.

## 5. CONCLUSIONS

Containment and surveillance is carried out by tracing the movement of the entire fuel continuously at the receiving

and storage areas. The transfer monitoring function and inventory management can work well in real time, and detect in a timely manner tamper or anomalous operations which may cause diversion of spent fuels. Owing to manual input of the data at the receiving/shipping stages, data checking is important in order to control system reliability, and the operators should be checked by the use of a password. We conclude that this system may be applied as an inspection device and be supported by continuous efforts to make it even more reliable.

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## **ВОЗМОЖНЫЕ ПРОЦЕДУРЫ ГАРАНТИЙ ДЛЯ ЭНЕРГЕТИЧЕСКИХ РЕАКТОРОВ НА БЫСТРЫХ НЕЙТРОНАХ С ИНТЕГРАЛЬНОЙ КОМПОНОВКОЙ**

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### **Abstract—Аннотация**

#### **POSSIBLE SAFEGUARDS PROCEDURES FOR POOL TYPE FAST POWER REACTORS.**

Recommendations are being formulated under the USSR's programme of scientific and technical support for Agency safeguards on carrying out Agency safeguards procedures on BN series pool type fast power reactors. By examining the design features of this type of reactor and taking into account the technical criteria of safeguards and the experience and practice of Agency safeguards procedures at other facilities, the paper analyses the main elements in developing an approach to the application of safeguards: (a) the structure of a nuclear material accounting and control system for this type of reactor; (b) possible nuclear material diversion paths at power plants with BN type reactors; (c) the organization of nuclear material flow control during receipt at and dispatch to nuclear power plants and also during loading into or unloading from an inaccessible area (reactor vessel and store containing liquid sodium coolant) by means of operator independent control systems; (d) verification of the nuclear material inventory at the nuclear power plant; (e) use of methods and devices for implementing containment and surveillance measures. On the basis of this analysis, the directions of scientific research for the development of recommendations concerning Agency safeguards procedures are defined with reference to the example of the USSR designed BN series pool type liquid metal cooled reactor. Additional ways of increasing the effectiveness of Agency safeguards procedures for this type of facility are discussed.

#### **ВОЗМОЖНЫЕ ПРОЦЕДУРЫ ГАРАНТИЙ ДЛЯ ЭНЕРГЕТИЧЕСКИХ РЕАКТОРОВ НА БЫСТРЫХ НЕЙТРОНАХ С ИНТЕГРАЛЬНОЙ КОМПОНОВКОЙ.**

В рамках Программы СССР по научно-технической поддержке гарантий МАГАТЭ проводится разработка рекомендаций по осуществлению контрольных процедур МАГАТЭ для энергетического реактора на быстрых нейтронах с интегральной компоновкой типа БН. На основе рассмотрения конструктивных особенностей реактора такого типа, а также с учетом технических критериев гарантий, опыта и практики осуществления контрольных процедур МАГАТЭ на других установках, в докладе проводится анализ таких важных элементов разработки подхода к применению гарантий: (a) структура системы учета и контроля ядерного материала для рассматриваемого типа реактора; (b) возможные пути переключения ядерного материала на АЭС с реактором типа БН; (c) организация контроля потока ядерного материала во время его получения и отправления на/из АЭС,

а также при загрузке/выгрузке такого материала из труднодоступной зоны (корпуса реактора и хранилищ со средой жидкого натрия) с использованием независимых от оператора средств контроля; (d) проверка инвентарного количества ядерного материала на АЭС; (e) использование методов и приборов для осуществления средств сохранения и наблюдения. На основе проведенного анализа определяются направления научно-исследовательских работ, связанных с разработкой рекомендаций по процедурам гарантий МАГАТЭ на примере реактора советской конструкции типа БН с интегральной схемой компоновки и жидко-металлическим теплоносителем. Обсуждаются дальнейшие пути повышения эффективности контрольных процедур МАГАТЭ для рассматриваемых установок.

## ВВЕДЕНИЕ

Большинство государств с развитой ядерной энергетикой предполагает осуществлять дальнейшее ее развитие с использованием реакторов на быстрых нейтронах.

Несмотря на общее замедление развития ядерной энергетики в мире, ряд государств уже до 2000 г. планирует независимо или на региональной основе продемонстрировать в коммерческом масштабе (10 ГВт (эл.) со всеми предприятиями замкнутого топливного цикла) технологию использования быстрых реакторов.

Из разрабатываемых концепций реакторов на быстрых нейтронах наиболее развитой является концепция реактора с жидкометаллическим теплоносителем. Как правило, большинство таких реакторов имеют интегральную компоновку [1]. В Советском Союзе уже работает такой реактор БН-600, строятся и проектируются более мощные реакторы БН-800 и БН-1600.

Необходимость разработки процедур гарантий для реакторов на быстрых нейтронах связана с развитием системы гарантий МАГАТЭ.

С точки зрения применения процедур гарантий основное отличие топливного цикла реакторов на быстрых нейтронах от цикла с легководными реакторами заключается в том, что, начиная с этапа изготовления топлива, используемый ядерный материал (плутоний или высокообогащенный уран) представляет намного большую потенциальную опасность с точки зрения переключения его на немирное использование. Изотопный состав плутония в отработавшем топливе реакторов на быстрых нейтронах более подходит с точки зрения его использования в ядерных взрывных устройствах по сравнению с плутонием, наработанным в легководных реакторах. Постановка в ближайшем будущем энергетических реакторов на быстрых нейтронах под гарантии МАГАТЭ вызывает необходимость разработки процедур контроля ядерного материала на таких установках.

Цель настоящей работы заключается в анализе современного состояния данной проблемы и в определении направлений научно-исследовательских работ при разработке рекомендаций по процедурам гарантий для быстрых реакторов на примере реактора советской конструкции типа БН-600.

Более подробно результаты проведенного анализа изложены в работе [2].

## 1. АНАЛИЗ ТЕХНИЧЕСКИХ ХАРАКТЕРИСТИК АЭС С РЕАКТОРАМИ НА БЫСТРЫХ НЕЙТРОНАХ, ВАЖНЫХ С ТОЧКИ ЗРЕНИЯ ПРИМЕНЕНИЯ ГАРАНТИЙ МАГАТЭ

Рассмотрим конструкционные особенности реактора на быстрых нейтронах на примере реактора с интегральной компоновкой типа БН-600 (табл. I). Общая схема реактора показана на рис. 1. Важной особенностью является то обстоятельство, что в герметически закрытом корпусе реактора под слоем жидкого натрия кроме активной зоны и зоны воспроизводства размещается весь первый контур с теплообменниками и насосами. Таким образом, доступ к топливным сборкам активной и экранной зон может быть осуществлен только с помощью штатных механизмов перегрузки.

Активная зона состоит из ТВС, регулирующих и компенсирующих сборок, использующих стержневые топливные элементы. Для выравнивания тепловыделения по радиусу активной зоны используются ТВС нескольких (в настоящее время двух) обогащений — ТВС зоны большого обогащения (ЗБО) и ТВС зоны малого обогащения (ЗМО) (см. табл. I).

Разница в обогащении зон составляет в среднем 20–50%. Каждая топливная сборка состоит из активной части и торцевых экранов из обедненного урана. По периметру активная зона окружена экранными сборками (зоной воспроизводства), содержащими двуокись обедненного урана.

Можно сформулировать основные причины, которые усложняют контроль МАГАТЭ за ядерным материалом на АЭС с реактором на быстрых нейтронах (например, в сравнении с легководными реакторами):

- наличие большого инвентарного количества специальных ядерных материалов;
- наличие примерно одного значимого количества (ЗК) ядерного материала в каждой ТВС активной зоны (со свежим или отработавшим топливом) и в одной–двух отработавших сборках воспроизводства — экранных сборках (ЭС);
- практическое отсутствие возможности визуальной проверки и идентификации большей части ядерного материала, находящегося под слоем жидкого натрия, либо в атмосфере натриевых паров (активная и экранная зоны, внутриреакторное хранилище, промежуточные хранилища — так называемая ”труднодоступная зона”, отсек отмывки);
- отсутствие прямого доступа к ТВС с отработавшим топливом, хранящимся в герметически закрытых контейнерах (пеналах);
- наличие в активной зоне реактора ТВС, имеющих существенно разное обогащение, а также различного вида оборот (активной и экранной зон, регулирующих, поглощающих и др.).

Вместе с тем существуют некоторые особенности реакторов с интегральной компоновкой, способствующие осуществлению гарантий:

- доступ к ядерным материалам, находящимся в корпусе реактора, возможен только с помощью стационарных перегрузочных устройств;

ТАБЛИЦА 1. НЕКОТОРЫЕ ХАРАКТЕРИСТИКИ АЭС С РЕАКТОРОМ ТИПА БН-600 [2]

Характеристика реактора		
Год пуска		1980
Мощность, МВт:		
тепловая		1470
электрическая		600
Конструкция		Баковая
Активная зона:		
вид топлива		UO <sub>2</sub>
высота, м		0,75
эквивалентный диаметр, м		2,06
число ТВС:		
ЗБО		154
ЗМО		215
масса основного делящегося элемента, кг		1880 ( <sup>235</sup> U)
Зона воспроизводства:		
вид топлива (UO <sub>2</sub> )		Обедненное по <sup>235</sup> U
число ТВС		379
Характеристики ТВС:	Активной зоны	Экранных сборок
масса, кг	105	115
длина, мм	3500	3500
размер под ключ, мм	96	96
число твэлов в ТВС, шт	127	37
вид топлива	UO <sub>2</sub>	UO <sub>2</sub>
Обогащение топлива по <sup>235</sup> U:		
в активной зоне, %	21; 33	0,4
в торцевых экранах (обедненный уран), %	0,4	0,4
Максимальный тепловой поток на (оболочке твэла, ккал/ (м <sup>2</sup> · 4)	214 · 10 <sup>6</sup>	0,82 · 10 <sup>6</sup>
Максимальное выгорание тяжелых атомов, %:		
ЗМО	7	
ЗБО	10	
Длительность работы на номинальной мощности (сут) :		
ЗМО	300	900-2400
ЗБО	450	

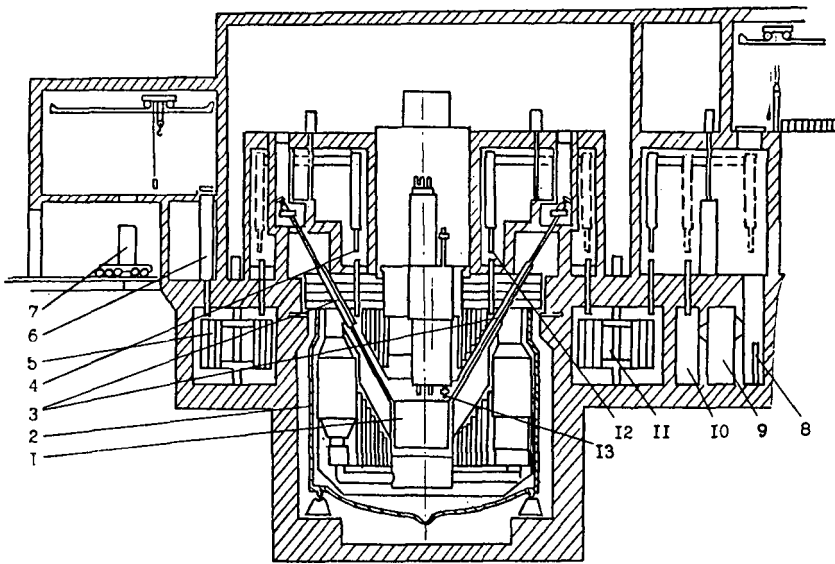


РИС. 1. Схема перегрузки ядерного топлива в реакторе типа БН: 1 – внутриреакторное хранилище, активная и экранная зоны; 2 – корпус реактора; 3 – элеваторы загрузки сборок в реакторе и выгрузки из него; 4 – перегрузочное устройство для свежих сборок (передаточный бокс); 5 – БСС (промежуточное хранилище); 6 – устройство для загрузки в БСС; 7 – транспортировочный контейнер (чехол) со свежими сборками; 8 – отсек загрузки ТВС в герметичные контейнеры; 9 – отсек наблюдения; 10 – отсек отмывки ТВС от натрия; 11 – БОС (промежуточное хранилище); 12 – перегрузочное устройство для отработавших сборок (передаточный бокс); 13 – внутриреакторное перегрузочное устройство (перегрузочная машина).

- извлечение твэлов или их частей требует разрушения ТВС, имеющих сварную конструкцию т. е. требует размещения специального оборудования в герметичном отсеке;
- ТВС с отработавшим топливом обладает высоким уровнем радиоактивного излучения и высоким остаточным тепловыделением, что требует для их транспортировки специальных защитных контейнеров;
- среда жидкого натрия, в которой находятся сборки, затрудняет доступ к ним и требует специальных процедур очистки для обращения с ТВС.

## 2. ПОДХОД К ПРИМЕНЕНИЮ ГАРАНТИЙ НА РЕАКТОРАХ НА БЫСТРЫХ НЕЙТРОНАХ

При разработке подхода и процедур гарантий учитывался опыт МАГАТЭ по разработке аналогичных подходов для легководных реакторов и реакторов, перегружаемых на мощности. Кроме этого, в рамках программ научно-технической поддержки развития гарантий МАГАТЭ разрабатываются рекомендации по проце-

дурам учета и контроля ядерного материала и по процедурам гарантий для отдельных типов реакторов на быстрых нейтронах. В последнее время МАГАТЭ накопило некоторый опыт применения гарантий на небольших исследовательских и опытно-промышленных реакторах. Однако предварительный анализ показывает, что при разработке подхода к применению гарантий для крупных коммерческих реакторов на быстрых нейтронах невозможны простое перенесение и экстраполяция существующих процедур контроля.

Подход к применению гарантий для реакторов на быстрых нейтронах с жидко-металлическим теплоносителем основывается на поштучной системе учета топливных и экранных сборок, дополняемой мерами сохранения и наблюдения.

Обычно свежие топливные сборки тщательно проверяются инспекторами МАГАТЭ на заводе по изготовлению топлива и их отправляют на АЭС опломбированными. Непосредственный поштучный подсчет, идентификация и измерение методами неразрушающего анализа (НРА) свежих топливных сборок могут проводиться в определенных местах в пределах доступной части зоны обращения со сборками на реакторе. Для труднодоступной зоны процедуры гарантий должны включать проверку потока ядерного материала на входе и выходе совместно с применением соответствующих мер по сохранению и наблюдению. В этом случае также может применяться проверка эксплуатационных записей оператора установки о расположении и использовании ядерного материала (например, записей об операциях по загрузке—выгрузке, а также картограмм загрузки активной зоны реактора и хранилищ).

Перед отправкой облученные топливные и экранные сборки должны проверяться путем визуального осмотра и (или) с помощью методов НРА, применяемых на выборочной основе; затем эти сборки помещаются в транспортный контейнер, который пломбируется. В соответствии с временными критериями для проведения проверки МАГАТЭ на таких установках (один раз в четыре недели), инспекции свежего топлива (если такое имеется на реакторе) должны проводиться чаще, чем облученного топлива. Однако некоторые инспекции для проверки инвентарного количества свежего топлива частично могут совмещаться с проверками запланированных отправок облученного топлива из реактора.

Проанализируем основные элементы подхода к применению гарантий на энергетических реакторах на быстрых нейтронах с жидкометаллическим теплоносителем, которые позволяют определить систему мер гарантий и процедуры контроля для установок такого типа.

### 3. ВОЗМОЖНОСТИ ПЕРЕКЛЮЧЕНИЯ ЯДЕРНОГО МАТЕРИАЛА НА АЭС С РЕАКТОРАМИ ТИПА БН

Анализ возможных путей переключения ядерного материала на АЭС требуется для разработки подхода по применению гарантий МАГАТЭ, который, как правило, включает рассмотрение двух экстремальных стратегий:

- единовременного переключения ЗК;
- постепенного переключения ядерного материала, общее количество которого примерно равно (или более) ЗК (например, посредством переключения небольших количеств ядерного материала из твэлов, целиком нескольких твэлов или нескольких сборок экранной зоны).

Для целей рассмотрения возможных переключений ядерного материала на энергетическом реакторе на быстрых нейтронах удобно подразделить зоны обращения с ядерным материалом на две категории:

1. "Труднодоступная зона" — зона, где ядерный материал находится под слоем жидкого натрия (реактор; БОС, БСС; перегрузочное устройство).
2. "Доступная зона" — зона, к которой можно отнести хранилище свежих сборок, водный бассейн выдержки хранилища отработавшего топлива, помещения для транспортировки топлива.

В качестве первой категории переключения ("единовременное") можно рассмотреть следующие возможности:

- переключение одной или нескольких ТВС со свежим топливом из доступной зоны. Данное переключение может быть проведено без использования стационарного подъемно-транспортного оборудования, так как масса основной сборки (около 100 кг) позволяет обращение с ней вручную или с помощью небольшого подъемного устройства. Оценки показывают, что время переключения одной сборки составляет 1–4 ч. Для сокращения переключения ТВС может быть использована их подмена макетами сборок, которые могут имитировать геометрические размеры, массу и идентификационные номера штатных сборок;
- переключение ТВС со свежим топливом из труднодоступной зоны. Данное переключение может быть произведено только с помощью стационарных механизмов перегрузки: элеваторов загрузки-выгрузки, внутриреакторного перегрузочного устройства, устройства для загрузки сборок в БСС. Кроме того, учитывая, что сборки в этом случае будут покрыты слоем натрия, необходим специальный контейнер для их перемещения из здания реактора. Данное переключение может быть проведено с подменой сборок макетами или без нее. Время переключения составляет 12–48 ч;
- переключение отработавшей ТВС активной зоны или нескольких отработавших сборок зоны воспроизводства. Данное переключение может быть произведено также только с использованием стационарных механизмов перегрузки и специального контейнера для транспортировки. В конструкции контейнера должны быть учтены не только наличие натрия на поверхности сборки в случае ее извлечения из труднодоступной зоны, но и высокий уровень радиоактивного излучения сборки и остаточное тепловыделение. Время переключения может составить (в зависимости от вида сборок и их количества) от 12 ч до 5–7 сут. Такое переключение может быть проведено с заменой сборок на макеты (или без нее). В макетах могут быть предусмотрены источники нейтронов и  $\gamma$ -излучения, а также источники тепла (однако изготовление таких макетов очень трудное и дорогостоящее мероприятие).

В качестве примера второй категории переключения ("постепенное") можно рассматривать следующие возможности:

- переключение топливных элементов или их частей. Как указывалось выше, ТВС реактора типа БН являются, как правило, неразборными, следовательно, для осуществления данного переключения необходимы специальное оборудование и большие трудозатраты. Кроме того, в случае переключения отработавших топливных элементов высокая радиоактивность и остаточное тепловыделение представляют существенный барьер для обращения с ними. Следует также отметить, что для того, чтобы переключение не было зафиксировано средствами НРА (исходя из оценок точности измерения), из ТВС может быть извлечено не более 1–2% общей массы специального ядерного материала, т.е. два–три топливных элемента. Учитывая, что в одном элементе находится примерно 50 г плутония, для переключения одного ЗК требуется извлечение около 160 топливных элементов, т.е. разрушение и последующее восстановление 50–80 ТВС. Время, необходимое для осуществления подобного переключения, оценивается примерно в 10–20 недель;
- переключение ТВС зоны воспроизводства с незначительным временем облучения из труднодоступной зоны. Для такого переключения требуется использование стационарных механизмов перегрузки и специальных контейнеров, тем не менее такое переключение можно осуществить во время перегрузки зоны воспроизводства. Следует учитывать, что риск обнаружения переключенной сборки с низким содержанием плутония пропорционален содержанию плутония, т.е. сборка должна содержать не более 100–200 г плутония (пределы ошибок приборов при качественном неразрушающем анализе). Следовательно, необходимо будет переключить несколько десятков такихборок для получения одного ЗК, что сравнимо с общим количеством этихборок на реакторе. Время такого переключения может составить 2–3 недели.
- наработка незаявленного плутония. Незаявленный естественный уран может быть облучен в зоне воспроизводства и других зонах внутри корпуса реактора. В этом случае исходный материал может быть введен в активную зону или зону воспроизводства с помощью обычных механизмов перегрузки.

#### 4. ОРГАНИЗАЦИЯ ОСУЩЕСТВЛЕНИЯ УЧЕТА И КОНТРОЛЯ ЯДЕРНОГО МАТЕРИАЛА НА АЭС С РЕАКТОРАМИ НА БЫСТРЫХ НЕЙТРОНАХ

При выборе числа ЗБМ и определении их границ необходимо учитывать, что на АЭС с реакторами на быстрых нейтронах с жидкометаллическим теплоносителем существуют три различные зоны обращения с ядерным топливом:

- хранилище свежего топлива (со свежим топливом в воздушной среде);
- промежуточные хранилища свежего и отработавшего топлива – БОС и БСС, активная и экранная зоны реактора, внутриреакторное хранилище (с топливом, находящимся под слоем натрия);



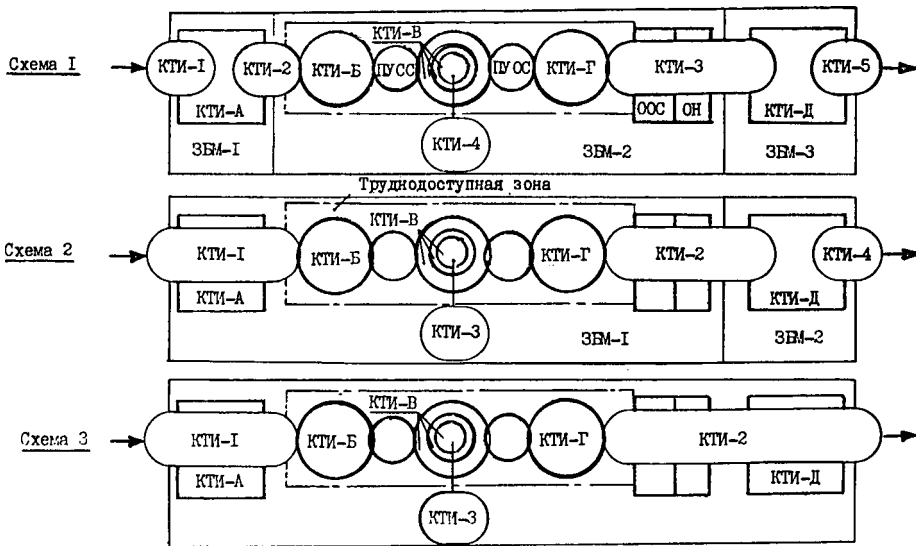


РИС. 2. Возможные схемы организации учета и контроля ядерного материала на АЭС с энергетическим реактором типа БН.

— приреакторное временное хранилище (с отработавшим топливом, помещенным в герметичные пеналы в специальные контейнеры для хранения, содержащиеся в водных бассейнах или в атмосфере инертного газа).

В отличие от легководных реакторов на реакторах типа БН отсутствует техническая возможность непосредственного подсчета и идентификации топливных сборок после их поступления в труднодоступную зону, поэтому необходимо организовать контроль ядерного материала в этой зоне для целей гарантий с помощью мер по сохранению и наблюдению, а также методов НРА. Контроль отработавшего топлива в приреакторном временном хранилище может быть организован аналогично процедурам, описанным в работе [3] для бассейнов выдержки долговременных хранилищ отработавшего топлива.

На исследовательских реакторах и прототипах энергетических реакторов число свежих и отработавших ТВС вне корпуса реактора может быть значительным, в этом случае такие хранилища топливных сборок могут быть выделены в отдельные ЗБМ (рис. 2, схема 1). При замкнутом топливном цикле реактора на быстрых нейтронах стремятся к сокращению времени нахождения ядерного материала вне реактора. В этой связи можно ожидать, что количество свежих сборок и время их хранения перед загрузкой в реактор будут сведены к минимуму — вплоть до загрузки в реактор ТВС из транспортных контейнеров, прибывших с завода по изготовлению топлива.

Время хранения отработавшего топлива также будет уменьшено до минимальной величины, связанной с необходимой выдержкой и охлаждением, допускающими безопасную транспортировку топлива на завод по химической переработке в специаль-

ном защитном контейнере. Такие мероприятия позволят снизить в два–три раза инвентарное количество ядерного материала, находящегося в реакторе, и в этом случае достаточно будет иметь на АЭС одну ЗБМ (рис. 2, схема 3). На переходном этапе развития топливного цикла с реакторами на быстрых нейтронах будут существовать приреакторные хранилища со значительным количеством топлива. В этом случае может быть выбрана схема 2 (см. рис. 2).

Технические средства контроля МАГАТЭ, используемые на АЭС с реактором на быстрых нейтронах, условно подразделяют на средства идентификации ТВС, приборы для осуществления мер сохранения и наблюдения, а также приборы НРА ядерного материала, содержащегося в топливных и экранных сборках. Для идентификации сборок реактора на быстрых нейтронах, находящихся вне труднодоступной зоны, могут быть использованы обычные методы, включающие визуальный осмотр и применение оптических систем, например, подводный телескоп и телекамера.

Для идентификации отработавших ТВС, хранящихся не в герметических пена-лах в водном бассейне, можно использовать, как и в случае легководных реакторов, приборы, регистрирующие излучение Черенкова — Вавилова.

В настоящее время не существует методов непосредственной идентификации сборок, хранящихся под слоем жидкого натрия. Можно, однако, отметить некоторые перспективные разработки, предназначенные для этой цели.

Прибор с использованием принципа сонара (USV) для наблюдения объектов в натриевой среде, позволяющий получить изображение торца сборки с высоким разрешением (нескольких десятых долей миллиметра) [4].

Ультразвуковой метод (FAID) идентификации сборок, имеющих в верхней части специальные устройства — метки, представляющие собой керамическую матрицу с вкраплениями металла в защитном чехле [5]. Считывание кода устройства-метки может осуществляться с помощью специального ультразвукового прибора (излучателя-приемника) и записывается на магнитную ленту для расшифровки на ЭВМ).

Эффективным качественным методом определения принадлежности сборки к тому или иному классу может служить регистрация их нейтронного и  $\gamma$ -излучения [6].

Основными количественными методами НРА ядерного материала, которые могут использоваться на реакторах на быстрых нейтронах с жидкометаллическим теплоносителем, являются  $\gamma$ -спектрометрия высокого разрешения, а также пассивный и активный нейтронные методы. Для оперативного сбора, обработки и хранения информации как для их поштучного учета, так и для контроля количества содержания ядерного материала в топливных и экранных сборках на АЭС с реакторами на быстрых нейтронах может использоваться ЭВМ. Так, например, для исследовательского реактора на быстрых нейтронах БОР-60 разработана система на базе ЭВМ [7], которая позволяет вести непрерывный учет количества и местонахождения всех ТВС и ЭС на АЭС, учет содержания ядерных материалов (урана и плутония) в каждой сборке. Важно, что с помощью такой системы можно оперативно получать информацию о расположении сборок на АЭС (картограммы активной и экранной зон, хранилищ), а также о количестве ядерного материала (списки инвентарного количества).

## 5. ПРОЦЕДУРЫ ПРИМЕНЕНИЯ ГАРАНТИЙ МАГАТЭ НА РЕАКТОРАХ НА БЫСТРЫХ НЕЙТРОНАХ

Конкретные процедуры, связанные с проверкой учетной и эксплуатационной документации на использование и хранение ядерного материала на АЭС с реакторами на быстрых нейтронах, во многом аналогичны таким процедурам для реакторов на тепловых нейтронах.

Материально-балансовые учетные документы должны подвергаться проверке с целью убедиться, что содержащиеся в них данные правильны и взаимно согласуются, а также чтобы подтвердить, что имеющаяся в них информация совпадает и соответствует информации, содержащейся в отчетах, представленных в МАГАТЭ государством. Эксплуатационные учетные документы проверяются точно таким же образом, как и материально-балансовые учетные документы и используются для определения нахождения топливных сборок на установке. Для свежих топливных сборок предусматривается, что перед отгрузкой на реактор типа БН с установки по изготовлению смешанного окисного топлива они должны подвергаться проверке МАГАТЭ методом НРА и транспортные контейнеры опечатываться. По прибытии транспортных контейнеров печати на них проверяются инспектором, а ТВС идентифицируются (по серийным номерам) и подсчитываются в момент, когда сборки перегружаются из транспортных контейнеров в контейнеры зоны хранения свежего топлива при длительном хранении на АЭС. Каждое отдельное гнездо или контейнер для хранения сборок свежего топлива и сборок зоны воспроизводства может опечатываться инспектором МАГАТЭ, а зона хранения подлежит оптическому наблюдению. Механизмы для перегрузки топлива в активную зону и из нее могут опечатываться.

После того, как свежие топливные и экранные сборки помещаются загрузочной машиной в среду с натриевым теплоносителем в целях перемещения во временные хранилища или для введения собственно в реактор, процедура проверки становится весьма сложной. Фактически оказывается невозможным непосредственно проверить инвентарное количество в активной зоне реактора. Поэтому необходимо, чтобы во время начальной загрузки активной зоны и зоны воспроизводства присутствовали инспекторы МАГАТЭ в целях проверки начального количества. После загрузки топлива в реактор и временные хранилища устройства для перегрузки топлива могут опечатываться. Затем присутствие инспекторов требуется во время последующих загрузок и разгрузок активной зоны и зоны воспроизводства. В промежутках между перегрузками активной зоны реактора применяют оптическое наблюдение (кино- и телекамерами) и опечатывание. В дополнение к этим мерам в будущем могут использоваться другие меры наблюдения и идентификации, осуществляемые в специально выбранных для этого ключевых местах.

К таким мерам можно отнести, например, использование приборов для контроля и идентификации сборок, загружаемых и выгружаемых из труднодоступной зоны. В таких приборах могут быть использованы методы НРА на основе регистрации нейтронного и  $\gamma$ -потоков топливных и экранных сборок. В частности, такие приборы могли бы регистрировать не только число прошедших через них сборок, но и

их класс (например, свежие ТВС активной зоны, отработавшие ТВС активной и экранной зон, неклассифицируемые сборки<sup>1</sup> [6].

Информация от таких приборах могла бы либо накапливаться в их блоках памяти, либо собираться и обрабатываться в едином блоке автоматизированной системы слежения за прохождением топлива на реакторах типа БН.

Для контроля и перемещения топливных и экранных сборок с помощью перегрузочных механизмов могут использоваться следующие устройства – мониторы. Для независимой регистрации мощности реактора необходимо разработать прибор (монитор) на основе регистрации ионизирующего излучения реактора [8].

Можно ожидать, что вышеперечисленные дополнительные меры позволят осуществлять надежный контроль МАГАТЭ за ядерным материалом в труднодоступной зоне на реакторе типа БН, обнаруживать возможное незаявленное облучение ядерного материала в реакторе, способствовать удовлетворению критериев своевременности обнаружения для описанных выше возможных способов переключения.

После выгрузки отработавших сборок из хранилища с натриевым теплоносителем и отмывки сборки выборочно могут подвергаться идентификации инспекторами МАГАТЭ с помощью оптических приборов в отсеке для наблюдения (см. рис. 1). В водных бассейнах сборки могут храниться либо отдельно на стеллажах, либо по нескольку штук в специальных контейнерах (чехлах). В случае хранения отработавших сборок в закрытой емкости в атмосфере инертного газа они могут контролироваться аналогичными приборами (мониторами), как и в случае емкостей под слоем натрия.

Контрольные процедуры МАГАТЭ для отработавших сборок, хранящихся в водных бассейнах выдержки, аналогичны процедурам для долговременных хранилищ отработавшего топлива с водными бассейнами. Заключительным этапом контроля МАГАТЭ на АЭС с реакторами на быстрых нейтронах является проверка отправки отработавших сборок на завод по химической переработке топлива. Большое стратегическое значение ядерного материала в отработавших сборках обуславливает присутствие инспектора МАГАТЭ во время загрузки таких сборок в транспортный контейнер и опечатывания контейнера. В дополнение к этому на АЭС может быть организовано оптическое наблюдение с помощью приборов МАГАТЭ за зоной для отправки ТВС и ЭС и работой погрузочно-транспортного оборудования.

При разработке процедур гарантий для реактора на быстрых нейтронах важно учесть критерий своевременности обнаружения возможного переключения, в особенности для свежего ядерного топлива. Такой учет обуславливает необходимость проверки топлива один раз в три недели. Однако общая интенсивность инспекционной проверки свежего и отработавшего топлива на реакторе может быть снижена с учетом графиков получения и отправки свежих и отработавших сборок на АЭС с реактором на быстрых нейтронах, их загрузки в активной зоне, а также с учетом рационального планирования проведения контрольных процедур МАГАТЭ.

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<sup>1</sup> К неклассифицируемым сборкам можно, например, отнести свежие сборки экранной зоны, контрольные и регулирующие сборки без ядерного материала и т. д.

## ЗАКЛЮЧЕНИЕ

Система мер гарантий для энергетических реакторов на быстрых нейтронах должна состоять из следующих основных элементов:

- системы документальных данных, отражающих число и местонахождение сборок, количество и состав ядерного материала в них и информацию о порядке его хранения, перемещения и использования. Хранение и обработка данных могут быть осуществлены с использованием ЭВМ на АЭС и необходимого набора программ (как, например, в случае реактора БОР-60);
- специальных стендов, оборудованных приборами НРА для проведения оценок и измерений содержания ядерного материала в свежих ТВС перед их загрузкой и в отработавших топливных и экранных сборках после их выгрузки из труднодоступной зоны (т. е. емкостей реактора и промежуточных хранилищ с натриевым теплоносителем). Такие измерения могут быть организованы на основе плана случайной выборки в соответствии с критериями гарантий, и они должны быть согласованы с технологическими операциями по перегрузке топлива на установке. Методы НРА могут включать  $\gamma$ -спектрометрические и нейтронные измерения;
- автоматизированных систем с использованием приборов для идентификации и подсчета сборок перед их загрузкой и выгрузкой из труднодоступной зоны. Детекторы этих систем должны быть установлены на пути перемещения сборок (штатные транспортные каналы, перегрузочные механизмы и т. д.). Важно также обеспечить регистрацию сборок в реальном масштабе времени, либо с минимальной задержкой в процессе их перемещения и перегрузки. При необходимости можно также организовать идентификацию и подсчет сборок по четырем классам: свежие ТВС, отработавшие ТВС, отработавшие ЭС и прочие сборки (например, свежие ЭС, кассеты для регулирования и т. д.);
- дополнительной системы, состоящей из устройства и приборов сохранения и наблюдения, разработанной и спроектированной для конкретной конструкции на быстрых нейтронах с интегральной компоновкой. Приборы такой системы должны позволять осуществление контроля за перемещением ядерного материала на АЭС вне труднодоступной зоны (хранилища свежего и отработавшего топлива), а также обеспечивать контроль за нахождением такого материала в пределах конструкционно выбранных границ (оболочки) для обращения с ним. В качестве таких устройств и приборов могут быть использованы оптические системы наблюдения (кино- и телеаппаратура), печатающие системы, следящие системы—мониторы;
- системы независимой регистрации мощности реактора на протяжении всего времени его работы. Такая система должна состоять из датчика и регистрирующего устройства.

Необходимо обеспечить, чтобы сбор информации от контрольных систем, используемых для целей гарантий МАГАТЭ на реакторах на быстрых нейтронах, осуществлялся бы персоналом МАГАТЭ во время инспекционных проверок на АЭС.

Представляет также интерес изучение возможности сбора и анализа такой информации автоматически, независимо от оператора установки. Высокая стратегическая ценность топлива с точки зрения его переключения и возможность нахождения большого количества такого топлива на АЭС с прототипными образцами реакторов на быстрых нейтронах могут потребовать трудозатрат на осуществление контрольных процедур МАГАТЭ на таких установках, близких к максимальным.

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## THE LASER SURVEILLANCE SYSTEM FOR SPENT FUEL POOLS

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### Abstract

#### THE LASER SURVEILLANCE SYSTEM FOR SPENT FUEL POOLS.

The Laser Surveillance System (LASSY) is a safeguards device primarily designed for use at spent fuel pools. Two beams of blue light scan an underwater plane just above the assemblies in the storage racks. When an assembly or tool penetrates this plane of light the change of reflectivity at the corresponding angle position is detected and the system computer calculates the position of the disturbance in the pool. Each of the two optical emitter detector assemblies ('eyes') turns on its own axis and takes measurements at 1150 points or angle positions. These readings are compared to a continuously updated background. If deviations meeting a number of criteria occur the corresponding X/Y co-ordinates of the disturbance in the pool are calculated, as well as a rough measure of its size and shape. These shape-figures are used to interpret an obstacle such as a certain type of assembly or tool. Scans with assembly or tool movements and their interpretation are stored and documented for retrieval by the inspector. LASSY has been successfully tested under field conditions during a two-week period in a fuel storage pool of the Paks Nuclear Power Plant, Hungary. A total of 114 fuel assemblies were monitored and registered with LASSY. During this test LASSY was exposed to various changes in environmental conditions such as change in water transparency and mechanical vibrations. In some cases the measured signals were not interpreted correctly because the fuel assemblies were guided manually into the grid position. A large amount of experimental data concerning environmental conditions, signals of assemblies and time characteristics of fuel transfer was collected. These data will be used to further develop the system to improve detection probability and interpretation of detected incidents.

### INTRODUCTION

New challenges for Safeguards are arising from the introduction of fuel assemblies designed for dismantling and reconstruction in LWR storage pools, and the growing number of long term storage facilities. Along with this there is a growing need for counting assemblies being loaded into, or unloaded from, a shipment cask.

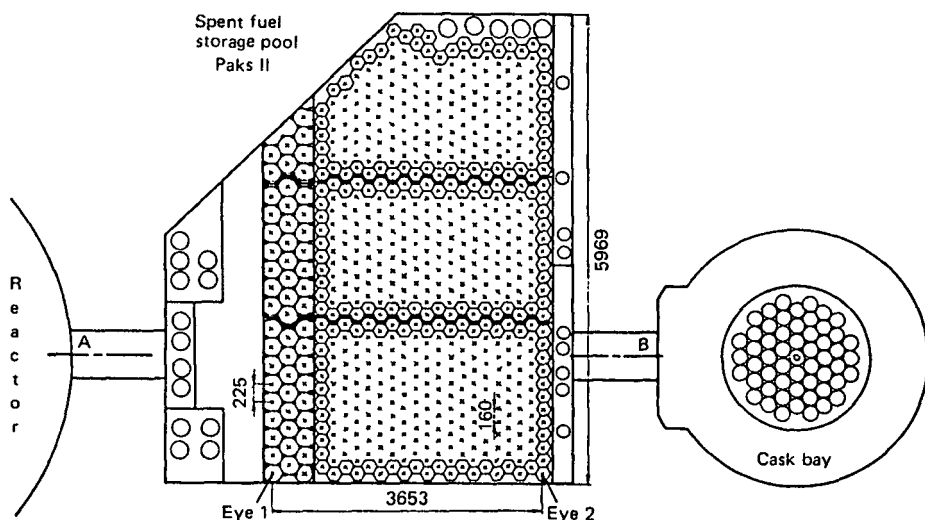


FIG. 1. Layout of the pool at Paks.

A clear distinction between positions where nothing happened since the last inventory on an inspection visit and locations where action was detected, would be an effective means to direct inspector activities.

The laser surveillance system (LASSY), which was designed for this purpose, is a novel safeguards system developed under IAEA research contract No. 3458/RB at the Atominstitut, Vienna.

Using experience gained with a first laboratory prototype system, a second-generation LASSY was assembled under IAEA research contract No. 4273/RB at the Atominstitut together with Mr. Thomsen, IAEA consultant.

This second-generation LASSY was essentially an upgraded version of the first complete prototype with important improvements in detection probability and scanning speed.

The successful field test of the second-generation LASSY at Paks NPP, Hungary, proves that assemblies which were either loaded or unloaded from a cask, or transferred within the spent fuel pool, can be efficiently detected and located.

## FIELD TEST RESULTS

From 23 June to 11 July, 1986, an extensive field test of the second-generation prototype Laser Surveillance System was performed at the spent fuel storage pool of Block-2 at the Paks



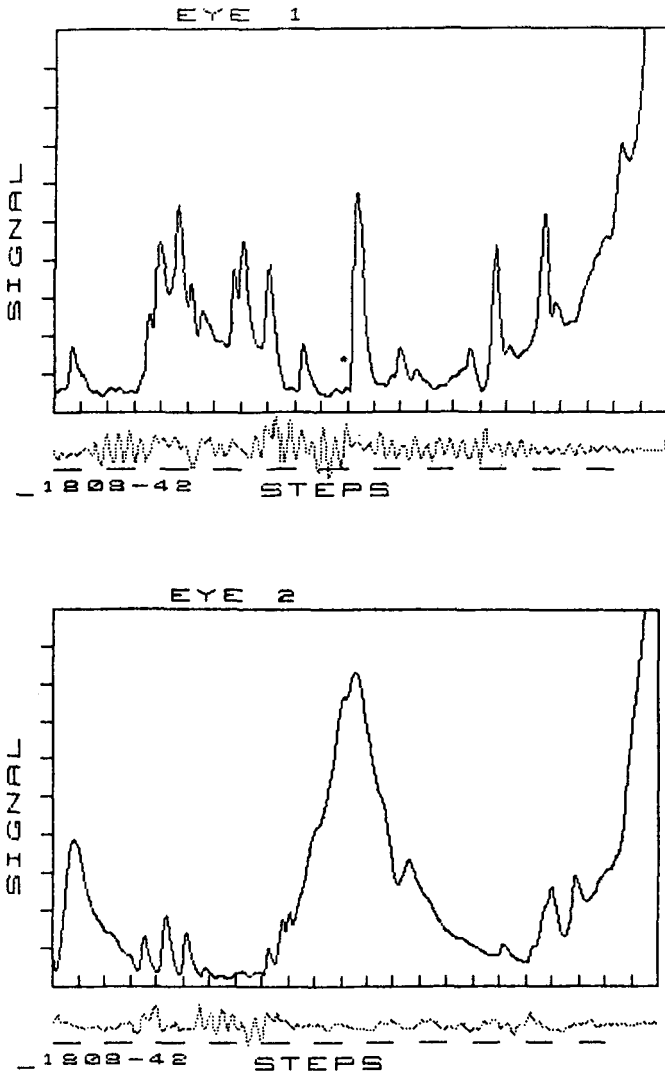


FIG. 2. Background in undisturbed pool with good water transparency.

Nuclear Power Station in Hungary. The transfer of 114 assemblies into the pool was monitored plus a number of preliminary actions with a dummy element. Much experimental data concerning the demands on a future regular-use LASSY were collected.

The two optical emitter detector assemblies (eyes) of LASSY were installed at two adjacent corners of the grid in the

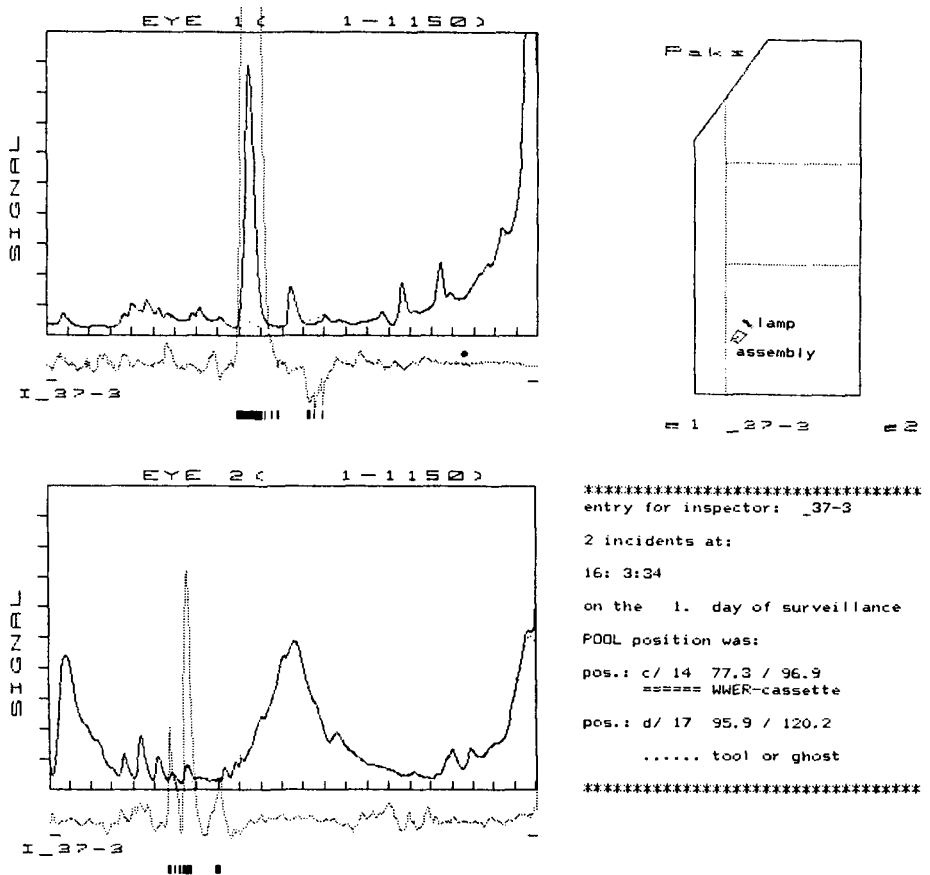


FIG. 3. Incidents caused by an assembly and a lamp at the same time.

pool (Fig. 1). For this purpose two supporting devices were prepared by Paks operators. The bottom of the support for eye 1 was shaped like a hermetic container for leaking fuel assemblies since this eye was to be placed into a corner position of the grid designed for such containers. The mount for eye 2 had the same hexagonal dimension as WWER fuel elements. The heads of the supporting devices were the same as on fuel assemblies. After fixing the eyes on to their supports they were manually guided into the designated grid positions.

Before and during the first scans a number of measurements on environment conditions were performed.

The 'fingerprint' of the pool, i.e. the reflected signals from the walls of the undisturbed pond, is shown in Fig. 2. It is very structured owing to several pipes along the pool walls

of the pool. Some of the peaks in the scans originate from the corners and from surface structures of the pool liner.

Under good environment conditions at the beginning of the test period (see also Fig. 4) LASSY was able to detect a simulated single rod.

From 1 July till 10 July the transfer of 114 fuel assemblies into Block-2 was monitored. The assemblies were loaded into a transport container, TKG, at Block-1 using the fuel handling machine. In this container, which is designed to house 48 assemblies, six assemblies at a time were transferred to Block-2. There the assemblies were moved by crane and guided manually from the container to their grid position.

The scan "-37-3" in Fig. 3 is a typical example of an assembly being manually directed into its position. To enable this and to light up the grid, slim 24 V operated lamps were used. They were also detected by LASSY and identified as 'tool or ghost'.

Using the fuel handling machine it takes more than a minute to pick up an assembly or lower it into a grid position. With the manually directed crane the same operation could be performed in less than 30 seconds, much faster than expected. Nevertheless, LASSY was able to 'see' almost all assemblies.

Typical times needed for element transfer are as follows:

(a) Operating by refuelling machine:

- lowering of the mast	40-45 s
- turning and locking	40-45 s
- picking up an assembly	30-25 s
- removing an assembly from the pond and placing it into the container	4-5 min
(all operations)	on average

(b) Operating the crane:

- aiming and locking an assembly	25-70 s
- picking up an assembly	25-30 s
- transfer through the pool	50-200 s
- placing an assembly into a socket in the pool	80-170 s
- removing an assembly from the container and placing it into a socket in the pool	4-5 min
(all operations)	on average

The major objective of the LASSY field test at a real power plant was to find out what relevant environment conditions such a system would find there, and if it was possible to cope with them. It turned out that the conditions at Paks NPP were much more adverse to successful LASSY performance than at Ispra or at the non-nuclear pool next to

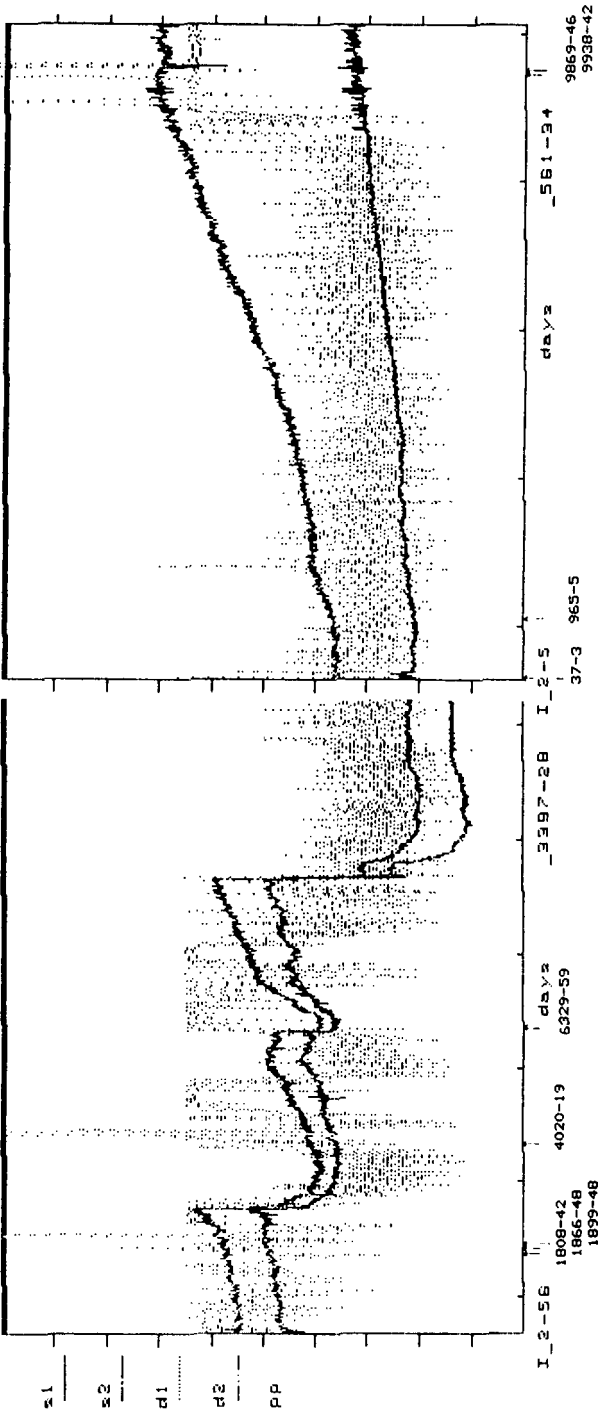


FIG. 4. Integral signals, transparency changes.

the IAEA headquarters. Nevertheless the present LASSY prototype was able to detect assemblies and to determine the grid position where they were placed.

Problems met can be summarized either as 'noise' or as those specific to the installation. All can be cured to some extent by upgrading the used software but to overcome them completely hardware changes in the system will be necessary.

Six contributions to the noise in the detected signals could be identified:

(a) Voltage fluctuations of the power line. Only after inserting a line stabilizer was it possible to use the lock-in amplifiers at the necessary high gain (100  $\mu$ V fuel scale).

(b) Noise as the result of pick-up in the electrical cables was detected with the laser beam blocked.

(c) During video recordings, when an excessive amount of light was focused into the pool, gain fluctuations of the overloaded avalanche-photo-diodes were observed.

(d) While the system was operating over longer periods short and long term changes in water transparency were registered. Drastic variations were induced when the water level in the pool was raised or lowered about 8 m during preparations of the fuel transfer (Fig. 4). As soon as the water level stabilized the transparency in the pool increased slightly.

When the big transport cask was placed into the cask bay strong water torrents were induced, and the level rose about one meter. During the fuel transfer the short term noise, most probably due to stirred-up particles, increased.

The maximum value for the attenuation length of the water for the 488 nm laser was about 9 m. The minimum value was about 3.5 m (Ispra: 5 m for 442 nm, Donaupark 7 m for 488 nm).

(e) Noise, which was similar to particle or current induced short term variations of the detected signals, originated from instabilities of the mechanical position of the eyes. As the stainless steel liners of the pool were polished with an irregular pattern even small changes in the eye position caused large deviations in the reflected signal. Such changes were sometimes induced when a crane was moving and a slight tremor could be felt all over the pool.

(f) With an assembly present the response from the wall of the pool was sometimes blurred when the distance between assembly and the eye was about one meter or less. Multiple reflections between eye, assembly and the wall of the pool resulted in additional deviations in the scan.

Apart from the unstable mechanical fixation of the eyes, problems arose from the exact location where they were installed. Both eyes were scanning at the same level, about 80 cm above the grid. In combination with the special way the fuel was handled during the test surveillance period, this resulted in problems with interpretation of the measured signals. Fuel assemblies were guided manually into their grid position. This took between 30 s and 3 min during which time the assembly was swaying around its designated position. In the noisy pool two successive scans were needed to establish an incident, and when the fuel element changed its place between the scans no response and no strange shape were recorded. These difficulties were further accentuated by not only the assembly but also up to three lamps were waving around at the level of the beams.

An important shortcoming of the present prototype system was the slow effective scanning rate. Although the scanning forward and data sampling only takes about 15 s the time needed to identify an incident was around two minutes. The system controller is not prepared for multi-tasking and can only perform sections of the program one after the other. Evaluation routines that involve much data transfer are also quite time consuming on the HP 9826 computer.

## CONCLUSIONS

Despite adverse environment conditions the second generation Laser Surveillance System Prototype was able to detect and locate 120 assembly movements at a real spent fuel storage pool.

It was no problem for the system to cope with changes in the reflected signals by a factor of 2 during 30 minutes as long as the attenuation produced no substantial short term noise. Big variations in the signal from scan to scan can only be partly handled by upgraded software, and therefore additional features for the LASSY hardware are necessary.

- The impact of all described sources of noise on LASSY could be minimized in a next generation if not only the amplitude of the reflected signal is measured but also its phase. Going over to higher chopping frequencies would permit the dependency of the phase on the light path length to be detected. Each eye would give complete information on angle position and distance of an obstacle. Redundancy gained with two eyes would enhance the detection and differentiation abilities of LASSY considerably.

- Measures to ensure successful operation of the system's components will have to be taken in the form of power line stabilization, battery back-up and shielding of the electrical cables between the eyes and the electronics.

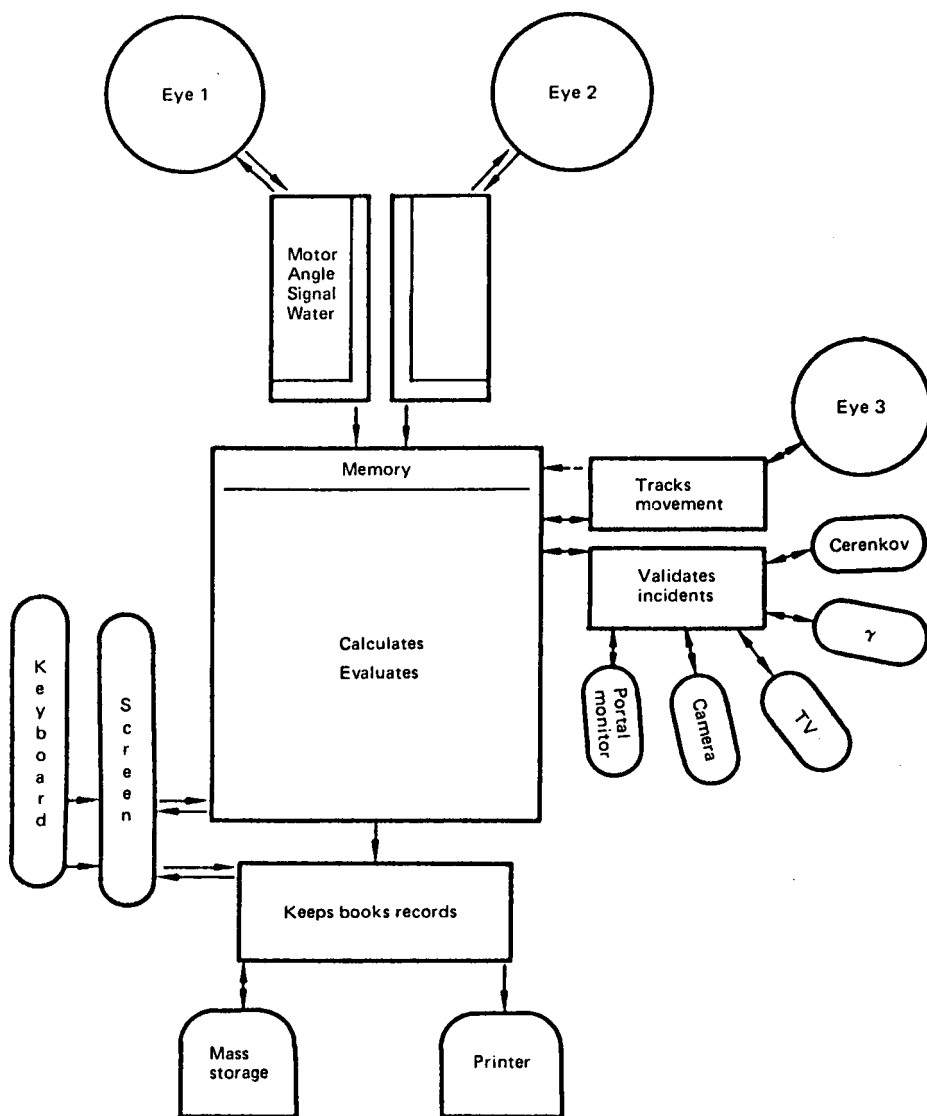


FIG. 5. Modular LASSY.

- The time characteristics of fuel handling at Paks indicate that the time needed for identifying an incident should not be longer than 20 s. Under adverse environment conditions this goal can only be reached with a more powerful computer concept. Either a multiple task configuration of the system controller or, even better, a design incorporating several CPUs is needed. Such a modular LASSY (Fig. 5) should have processors for each eye (or additional 'sense'). The main processor is only used for scan evaluation.

- Much attention should be paid to the mechanical setup of the eyes in order to place them at a well-defined and stable location. At the same time the eyes should scan only the 'plane of the highest interest'. For spent fuel storage pools this would mean as close as possible to the heads of the assemblies in the rack.

- In accordance with a modular hardware concept software would also be structured and flexible to adapt LASSY to a wide range of surveillance tasks and environments.

## OUTLOOK

The field test of the laser surveillance system (LASSY) at Paks NPP proved the applicability of this system under field conditions. Some improvements on hard and software remain to be done in the future. Taking into account LASSY's unique features (ability to monitor relevant movements, also underwater) and the results of this field test, further development of the system and systems studies on LASSY implementation would be very useful for safeguards purposes. This was distinctly pointed out by the Advisory Group Meeting on Safeguards Surveillance Techniques at the IAEA, 14-18 July, 1986.

At the moment system studies on LASSY and its possible applications are being undertaken in the United Kingdom and Australia. The development of the next generation Laser Surveillance System will be done at the Euratom's Joint Research Centre, Ispra. Depending on the outcome of the studies the next prototype will be designed to meet additional requirements of certain tasks.

## ACKNOWLEDGEMENT

This work was supported by the IAEA under the Research Contract 4279/RB. We express our gratitude to the representatives of the Hungarian National Atomic Energy Commission and the staff of PAKS Nuclear Power Station for their valuable cooperation and assistance in organizing and performing the LASSY field test.



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## EXTENSION OF THE USE OF BOILING WATER REACTOR (BWR) FUEL ASSEMBLY SEALING TECHNIQUES TO OTHER SAFEGUARDS APPLICATIONS

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### Abstract

#### EXTENSION OF THE USE OF BOILING WATER REACTOR (BWR) FUEL ASSEMBLY SEALING TECHNIQUES TO OTHER SAFEGUARDS APPLICATIONS.

The sealing of light water reactor (LWR) fuel assemblies has been under consideration by safeguards authorities and research laboratories for about a decade. More recently, decisive improvements have been made on fuel assembly underwater ultrasonic seals and the equipment used to handle and verify them. A long term demonstration has been conducted at the reactor facility of Kahl, Federal Republic of Germany, and successfully completed in 1986. On the basis of their experiences during that demonstration, the Joint Research Establishment, Ispra, Commission of the European Communities, and the Sandia National Laboratories, Albuquerque, United States of America, are jointly considering the extension of the BWR fuel assembly sealing systems (BFASS) technology to other possible areas of application. These are the MOX BWR and the PWR fuel assemblies (normal and MOX), transport and/or storage containers of various kinds, fuel assembly racks and stacks. Referring to existing techniques, the difficulty of application to other feasible solutions is evaluated. Among these areas, the sealing of storage flasks seems to pose few additional problems, an application being already on the way. On the other hand, the sealing of PWR fuel assemblies is shown to raise several questions regarding the exact constitution of an assembly, the operator's handling practices and the various possible safeguards strategies. In general, knowledge of the handling practices is an essential point from which to start the feasibility of a sealing system. As long as the applications contemplated do not envisage a more severe environment than that of the BFASS, the main technical points are essentially mechanical and can be resolved in a joint effort involving manufacturers, developers, operators and safeguards inspectors.

## 1. INTRODUCTION

In safeguards, ultrasonic techniques have been under investigation in several laboratories for more than a decade. Basically, these techniques offered good possibilities of underwater verification of items containing fissile materials, such as fuel assemblies, containers or storage racks — a typical containment and surveillance (C/S) problem.

In particular, on the basis of a solution first proposed by the Joint Research Establishment (JRC), Ispra [1], two systems using ultrasonic underwater verifiable seals for safeguarding BWR fuel assemblies have been developed: one by the Sandia National Laboratories (SNL), Albuquerque and the other one by JRC. These BWR fuel assembly sealing systems (BFASS) have both been field tested by the respective teams in the BWR reactor of Kahl, Federal Republic of Germany (FRG) for several years, in the framework of collaboration with an FRG/IAEA support programme (Task D3) which is now completed [2].

The results obtained during the demonstration campaigns at Kahl [3, 4] have clearly shown the feasibility of the technique. It is possible to place a seal on a new fuel assembly, either at the manufacturer's plant, or in storage. It is possible to re-verify that seal after a stay of several years in the reactor core, once the fuel assembly is stored in a rack of the water storage. It is also possible to install a seal on a spent fuel assembly never previously sealed. It is possible to extract a seal from its assembly when it is to be repaired, or on its way for disassembling operations. The handiness of the tools for placing, extracting, and verifying the seals has been demonstrated, along with the flexibility of the portable ultrasonics-electronics instruments which allow, with the present versions, a fuel assembly to be easily identified on the spot, because an automatic statement is delivered to the inspector within a few minutes. Furthermore, the seals have been approved by the licensing authorities and, after several years, their use and handling have not caused operational difficulties or damage to the fuel assemblies.

Owing to these results, the application of ultrasonic seals has been considered from a more general viewpoint, in order to assess their suitability for other C/S requirements. The following areas were reviewed, taking into account their significance to the safeguards approach and the corresponding estimated technical difficulties. They are:

- (a) BWR fuel assemblies
- (b) BWR MOX fuel assemblies
- (c) PWR fuel assemblies
- (d) PWR MOX fuel assemblies
- (e) Transport and storage flasks (bottles), casks or containers for spent fuel
- (f) Spent fuel storage stacks or racks.

Our main interest is to compare the requirements in each area with the features available on the current BFASSs we have developed and demonstrated. In some

areas, the problems raised and the expected effort for a solution seem to be easier than in the original study. For instance, although these techniques have been developed for underwater remote verification they were shown to be suitable also for dry condition identification, and can thus apply to the tamperproof closure of fresh fuel containers, or other surface transport spent fuel containers.

## 2. BRIEF DESCRIPTION OF THE PRESENT TECHNIQUE

The sealing technique for BWR fuel assemblies is based on the concept of 'locking' a fuel assembly (typically: a square array of 36, 49 or 64 rods, about  $0.15 \times 0.15 \times 4.5$  m in size, mounted between a lower and an upper tie-plate) by means of a seal which must be broken to allow the disassembly of the fuel bundle. The seal bears a fingerprint obtained by a distribution of random marks which confers on it an *identity*. It also provides an *integrity check* indicating whether or not it has been broken. Both identity and integrity can be verified with appropriate extendable tools operating under water and fitted on the seal during the verification operation. These tools incorporate ultrasonic transducers emitting ultrasounds and also conveying the reflected echoes to portable instruments. Different transducers may be used for identification and for integrity verification. A seal has the shape of a small cylinder clamped vertically on top of a fuel assembly. It is about 15 mm o.d. and 40 mm high, and weighs about 30 g.

The system developed by SNL-Albuquerque is called FAID (fuel assembly identification device). The system developed by JRC is called VAK-III, after the name of the Kahl reactor and the version number. These two systems have been described elsewhere [5, 6] and their progress reported several times, in particular on the occasion of the INMM and ESARDA meetings [7-9]. Nevertheless, their main characteristics and features are given in Table I. It can be seen how the present solutions fulfil certain essential criteria (*basic functions*) and meet operational requirements (*operating-handling functions*) as they were defined in the Task D3 Working Group [2].

## 3. BWR FUEL ASSEMBLIES (area (a))

It is felt that the problem of sealing BWR fuel assemblies has now been solved by the D3 study, and that the basic safeguards requirement of keeping a fuel assembly (or bundle) under control as long as it exists has been fulfilled (Fig. 1).

TABLE I. MAIN CHARACTERISTICS OF THE VAK-III AND FAID SYSTEMS

**Basic functions**

1. *Locking*: Seal attached by a one-way clamping device. Must be extracted to allow fuel assembly dismounting.

- |          |   |
|----------|---|
| FAID:    | Needs a gripping nut to be preinstalled at fabric. plant  |
| VAK-III: | Needs an extended tie rod end to be welded on one tie rod |

2. *Identity*: Unique signature obtained by random marking process.

- |          |  |
|----------|--|
| FAID:    | External irregular annular weld on top of seal                                     |
| VAK-III: | Internal cavities randomly distributed and filled by an incomplete brazing process |

3. *Integrity*: Broken or intact state of the seal evidenced by ultrasounds, a fracture link being broken when seal is pulled off.

**Handling and operating functions**

4. *Installation*: Seals can be clamped by hand on fresh fuel assemblies or placed with an underwater positioning tool on spent fuel.

- |          |   |
|----------|---|
| FAID:    | Can be installed directly on Exxon fuel assemblies  |
| VAK-III: | Can be installed directly on KWU fuel assemblies; and with an intermediate bushing on Exxon fuel assemblies |

5. *Extraction*: A special underwater breaking tool is used.

- |          |   |
|----------|---|
| FAID:    | Fracture link breaks at 1500 N traction force   |
| VAK-III: | Fracture link breaks at 1300 N, then the tie rod extension is broken at 2600 N traction force |

6. *Identification*: Identification tool fitted on the seal.

- |          |  |
|----------|--|
| FAID:    | Underwater long tool with 2 steady 10 MHz focused transducers (plus one flat central trans. for integr.) |
| VAK-III: | Underwater or air extendable tool with one 10 MHz revolving focused transducer and driving motor         |

7. *Integrity check*: Fracture link interrogated with:

- |          |   |
|----------|---|
| FAID:    | A central transducer on same tool as above          |
| VAK-III: | A central transducer on independent extendable tool |

8. *Readout instrument*: A portable instrument allows process and deliver data to be acquired on the spot (at present one seal can be identified every 7 to 10 min). It embodies ultrasonics and electronics and issues data on an incorporated printing machine.

- |          |   |
|----------|---|
| FAID:    | SPAR developed by SNL. Uses conversational step program and bubble cassettes  |
| VAK-III: | Compact box (VAK 45 or 18) incorporating commercially available SONIC instrument and SHARP PC 1500. It uses conventional cassettes and low cost printer. It processes identity signals and checks integ. on CRT |

**Costs**

- |   |
|---|
| A portable instrument costs about US \$6500 to US \$8000                |
| A seal costs about US \$150 in small series and US \$50 in large series |
| A set of handling tools and transducers costs US \$5000                 |

NB: integ. = integrity (check); trans. = transducer.

### 3.1. Manufacturers

Two different fuel manufacturers were involved in that study, because the Kahl reactor uses Exxon Nuclear and KWU<sup>1</sup> assemblies. Their staff were open and collaborative, and accepted a slight modification of the assemblies' upper parts to allow the seals to be clamped on. Such modifications were defined by common agreement and designed so that no thermal, mechanical or physical effect could be induced by the clamping element, or by the eventually attached seal. They were made for experimental reasons, but it can be envisaged that they could become permanent features of the fuel assembly production. We do not consider specific restrictions, because all the BWR fuel manufacturers use very similar assembly designs.

### 3.2. Users

As mentioned before, the seals were well accepted by the users as soon as the licensing authorities had approved them. It is probable that the main disturbance we caused to the operators was in carrying out our demonstrations. But, on a routine basis, the seals by themselves do not cause inconvenience and do not interfere with the normal handling, channelling and dechannelling operations. It is obvious that, during inspections, according to the number of assemblies to be verified, a certain 'intrusion' is necessary, but it is limited to using the moving bridge above the racks, since no fuel assembly handling is required to inspect the seals.

### 3.3. Reconstitution and consolidation

The pending question of the possible use of *reconstitution* and *consolidation* practices in plants is important because these operations represent an interruption of the information a seal can give about a fuel assembly. When a fuel assembly is totally or partially dismantled, it ceases to exist as a unique identifiable item, and if it was previously sealed its seal has to be broken. As shown during the Kahl demonstration, extracting one seal with a simple tool takes only a few minutes and would cause very limited inconvenience to the operator involved in a disassembly process or a reconstitution. According to recent surveys [10, 11] and experimental demonstrations [12] conducted in the USA, the reconstitution or the consolidation of one BWR fuel assembly is estimated to take between 5 and 10 hours. This minimizes the related importance of seal removal or installing operations. In fact, we understand that, at least in certain techniques, the individual rods are accumulated in special canisters, having the capacity of two consolidated assemblies. These canisters might well be closed and safeguarded by seals similar to the BFASS ones (see area (f) — Section 8). During the discontinuity of information, the safeguards scheme might be to rely on an inspector, or optical surveillance to be triggered for the circumstance.

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<sup>1</sup> KWU = Kraftwerk Union Aktiengesellschaft, FRG.

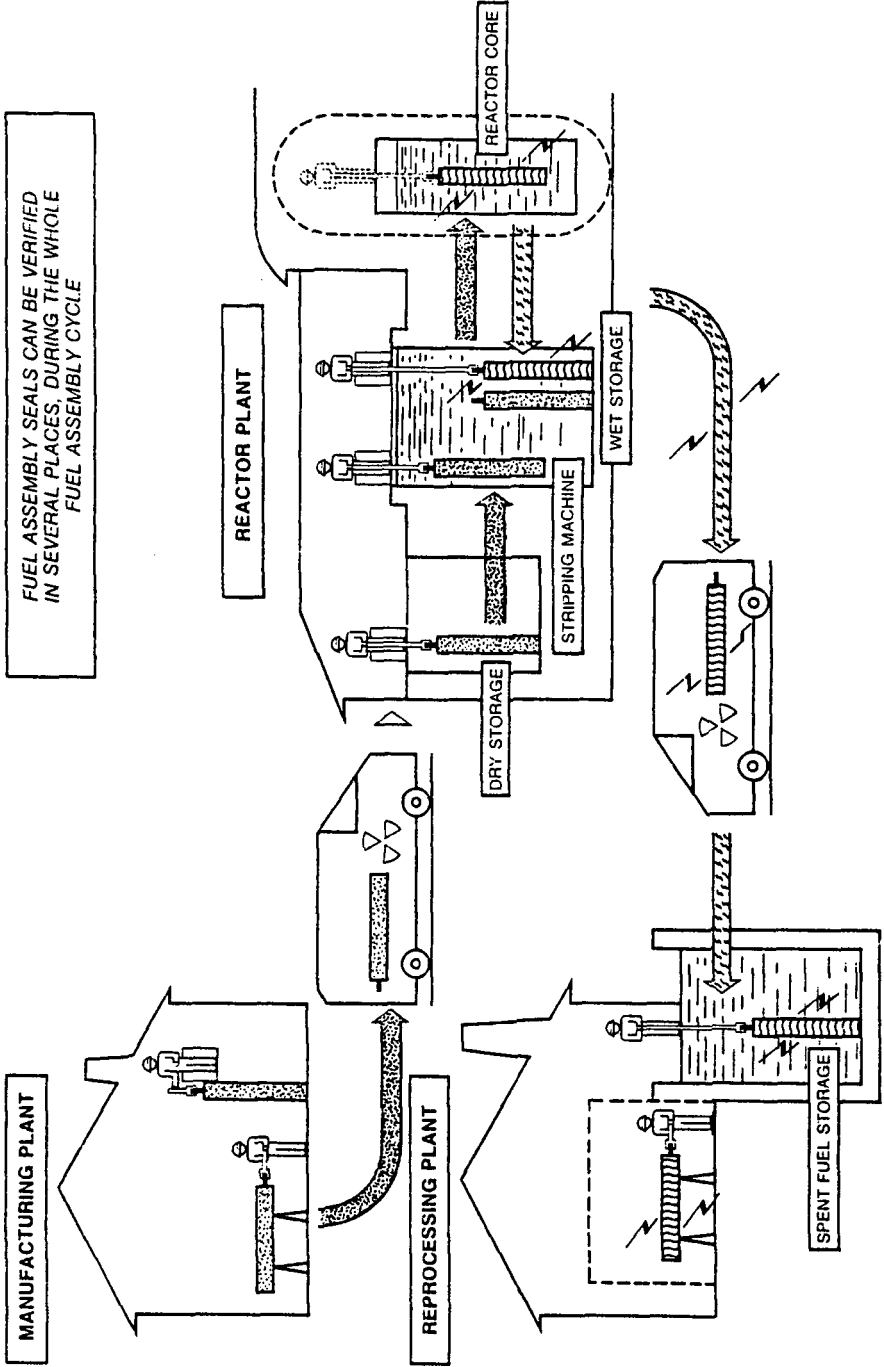


FIG. 1. The fuel assembly seal cycle.



But we do not know how frequent such exercises would be and it is not our purpose to discuss this matter here. As for the reconstitution, we must add that a fuel assembly, since it has three or four possible 'sealing' sites, can easily be resealed after it has been reconstituted (or repaired).

#### **4. BWR MOX FUEL ASSEMBLIES (area (b))**

Because of their plutonium content, MOX fuel assemblies are of concern to the safeguards authorities, but they have the same structure as normal fuel assemblies.

##### **4.1. Safeguards schemes**

It can be imagined that such fuel assemblies would be sealed at the manufacturer's plant, and that the seals would be taken away, at the end of the assemblies' life — for instance, at the reprocessing plant. As for normal fuel, no particular problem is foreseen for the installation and initial verification, in a normal environment (dry conditions), either at the fuel assembly or at the storage vault of the manufacturer. This operation can also be carried out at the moment fresh fuel assemblies are stored at the reactor. Or, according to various possible safeguards schemes, it can be decided to seal the MOX assemblies only during their 'fresh' life, in order to provide transportation guarantees. If 'whole life' sealing is preferred, 'de-sealing' operations can take place either at the reactor's disassembly station or at the reprocessing plant.

##### **4.2. User's practices**

Concerning the user's practices, we see no particular differences in the handling procedures between normal and MOX fuel bundles. Consolidation is envisaged and fuel repair is still also possible. If we refer to the figures reported by a Federal German manufacturer such as KWU [13], the medium in-core defective rate of the fuel rods is 0.003%. Actually, this refers to the normal UO<sub>2</sub> fuel elements but it still indicates that on a 1300 MW(e) BWR reactor using a three-year renewable load of 900 assemblies with  $9 \times 9$  rods, only two assemblies will be repaired every three years for rod defect!

##### **4.3. Suitability**

We consider that, in the absence of very particular specifications, the underwater ultrasonic sealing techniques are suitable for the application to BWR MOX fuel assemblies, and that the technology has been sufficiently demonstrated and is now ready to be studied for industrial production. Depending on eventual requirements from the safeguards authorities, an evaluation programme could be started with a

limited batch of MOX fuel assemblies to be sealed and followed during a complete cycle, or during the shipment of fresh fuel.

## 5. PWR FUEL ASSEMBLIES (area (c))

The concept of applying ultrasonic sealing techniques to PWR fuel assemblies is not new. From the very beginning of these studies the whole LWR fuel production was considered as the field of application for the safeguards approach. But effort was put first on the smaller and easier to handle BWR fuel assemblies which were also those used in the Kahl reactor. The safeguards requirements are the same for both types, but preventing the normal disassembly of a PWR fuel bundle is more complicated, even if we again have the problems of 'locking' a fuel assembly.

### 5.1. Description

A PWR fuel assembly is typically a square array of 196, 225, 256 or 289 rods about  $0.20 \times 0.20 \times 4.20$  m in size, mounted between a lower and an upper fitting. According to a survey from Oak Ridge National Laboratory (ORNL), more than ten different PWR fuel assembly configurations exist in the USA [14], but the differences between them are related more to special mechanical features than to the general concept, which is the same for almost all types.

Essentially a PWR fuel assembly is characterized by the existence of *guide tubes* serving to guide the *control rods* descending into the fuel assembly. These can be 5, 16, 20 or 24 according to the array or design type. They are distributed with a regular pattern into the fuel rod array and occupy similar geometrical positions (Fig. 2). The lower end of these tubes is blind (thimble tubes) so as to have no water flow through them. They are fastened on the lower plate (nozzle) by means of screws, nuts, welds or other devices. They are kept apart by several spacer grids with which they constitute the assembly's skeleton. The spacer grids are welded on the guide tubes but hold the fuel rods by friction so that these do not apply their weight directly on the bottom plate and there is a gap between the lower rod ends and the upper surface of the bottom plate. Similarly, there is a gap of about 20 to 30 mm between the upper rod ends and the lower surface of the top plate. During the assembly life, some rods may slide to a small extent, the maximum being when they come into contact with the top or the bottom plate. The guide tubes are made of Zircaloy-4 with an i.d. of about 13 mm. A central instrument tube of the size of the guide tubes is often installed in the middle of the array.

### 5.2. Disassembly and cutting operations

It is our understanding that when the configuration is said to have the 'disassembly capability', the top or bottom plate can be withdrawn after certain fastening

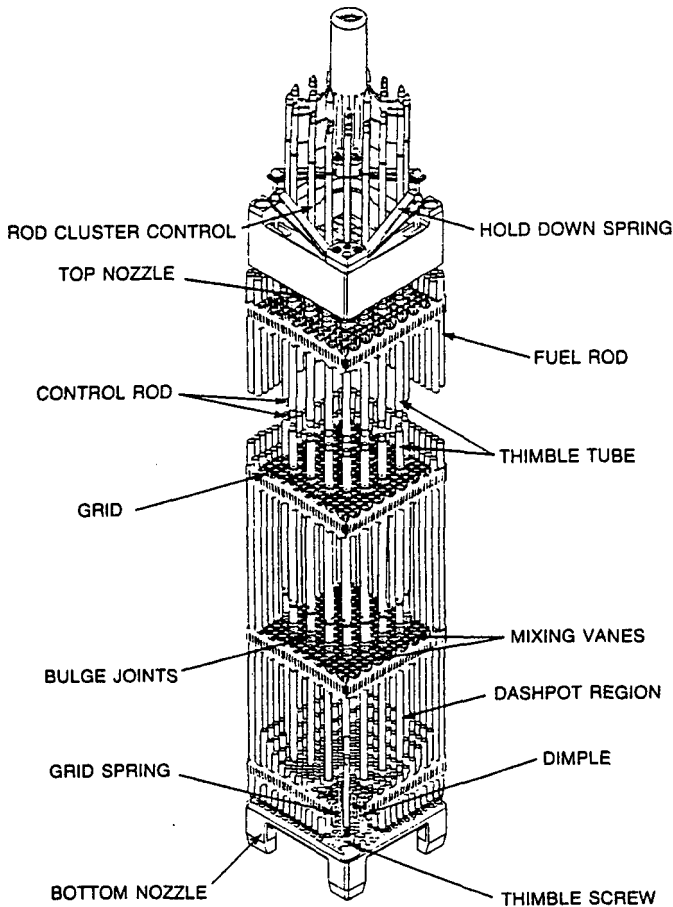


FIG. 2. Schematic view of a typical PWR fuel assembly.

devices have been unscrewed, rotated or otherwise unlocked, so that some or all rods can be removed from the structure. Nevertheless, in the absence of quick disconnections, it seems that cutting the guide tubes — or machining the guide tube welds — are faster methods of disassembly, at least for the consolidation operation [15].

Johnson [15] reports a 15–20 min time to cut (we assume) 16 guide tubes and one instrument tube, which seems a very fast operation.

### 5.3. Sealing capability

From the sealing viewpoint it is interesting to keep in mind the existence of guide tubes which act as connecting elements between the top and the bottom of what we can call a 'container'. This container is very vulnerable in the zones where it can

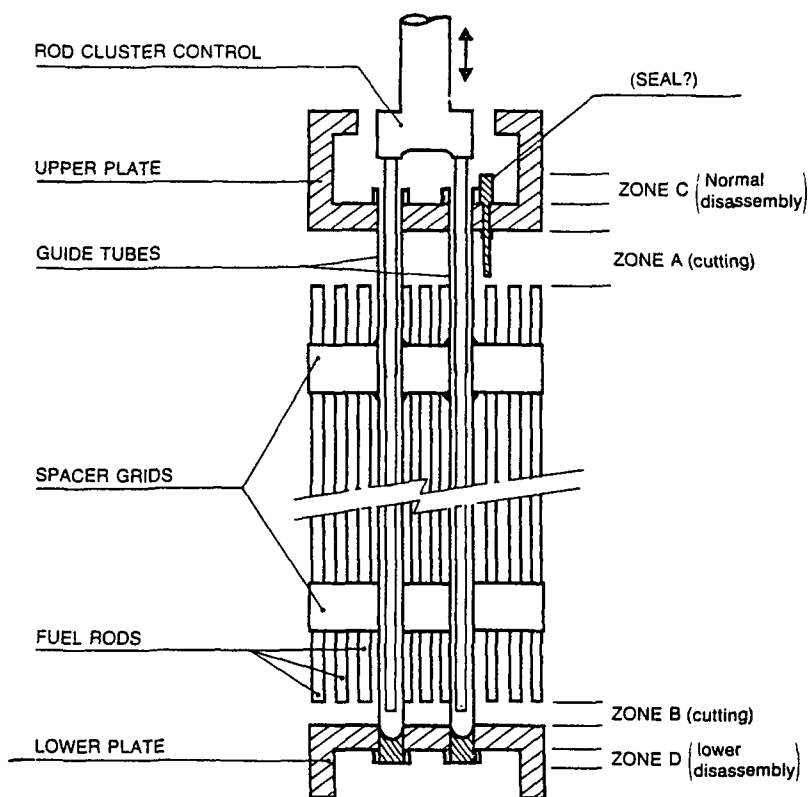


FIG. 3. The four 'vulnerable' zones of a typical fuel assembly (order of importance: C, A, B, D).

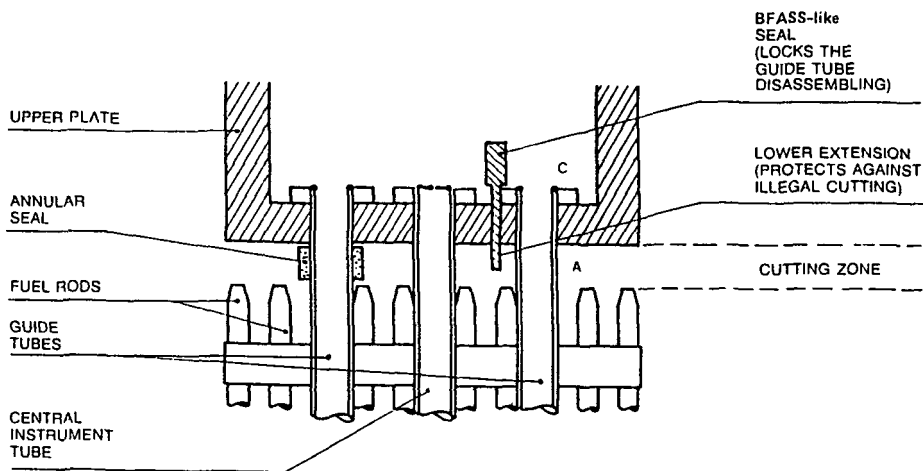


FIG. 4. Two possible solutions for safeguarding Zone A (cutting) and Zone C (normal disassembly) of a PWR fuel assembly.

be cut easily, i.e. the two gaps mentioned before. The second-level vulnerability is related to the actual disassembly capability. We presume that the most convenient PWR fuel assembly concepts provide normal upper plate removability, which our seal should impede. For disassembly from the lower side, another locking or sealing device should be designed.

We can contemplate four zones of interest with decreasing safeguards importance while looking for a seal location (Fig. 3). In zone C, where it is logical to place a seal, the guide tube end protrusion varies with the fuel type. The space available between the plate and the *rod cluster control* (spider) at its lowest position is probably large enough, even if not clearly understandable from the available drawings. We feel that a seal protruding about 30 mm would not create problems. Zone A can be protected, for instance, by an annular seal or by a downward extension of the seal in zone C (Fig. 4). Zones B and D must be reached from the upper part of the assembly, probably by using a transducer descending into one guide tube and interrogating a modified end plug from inside. Such an end plug might well be derived from a FAID, with its upper marks facing the inside of the tube.

#### 5.4. Feasibility

The existing BFASS technique would apply quite well to a sealing of zones A and C (Fig. 3), which are located in the upper side of the PWR assembly. Should a lower protection be recognized as a necessary safeguards measure, this would imply a special development study of at least two years. In such studies the use of the central instrument tube should be taken into account, provided that more information about the way it is used be available.

### 6. PWR MOX ASSEMBLIES (area (d))

We can draw a parallel between the BWR and PWR situations. Also in the PWR case, since the fuel assemblies from the same manufacturer are supposed to have exactly the same configuration whether they are MOX or not, the possible difference in sealing procedures would come only from the safeguards strategy.

#### 6.1. Safeguards approach

If the safeguards approach concentrates on the beginning of the MOX assemblies' life, as is supposedly the case for BWR fuel assemblies, fresh fuel would be sealed at the production plant and verified prior to loading into the reactor. In such a case, control of the integrity and identity of fresh assemblies during transportation should be afforded by the proposed sealing system. First, the vulnerable zones A and C described previously would have to be protected.

## 6.2. Handling assessment

These questions are expected to be answered provided that technical surveys will be available on the PWR fuel assembly 'whole life' handling. It is absolutely necessary to have exact knowledge of the MOX fuel cycle (or cycles), from fabrication to reprocessing, or from fabrication to reconstitution, or from fabrication to consolidation, and also from consolidation to reprocessing or from consolidation to definitive storage, etc. For example, depending on the existence of a consolidation installation (a 12–15 m high rig) in a reactor facility, it can be known whether a determined population of assemblies is going to undergo cutting or normal repair disassembly.

## 7. TRANSPORT AND STORAGE FLASKS, CASKS AND SPENT FUEL CONTAINERS (area (e))

For these applications, the safeguards requirements are quite clear. Provided that all types of container are closed with bolted lids, it is required that at least one of the bolts be replaced by a special bolt with the characteristics of a seal, i.e. a verifiable unique identity and an integrity feature indicating whether or not this bolt has been unscrewed.

### 7.1. Flasks

Among other possible applications, one has already been indicated by British Nuclear Fuels plc (BNFL), who raised the problem of sealing spent fuel transport flasks, also called multi-element bottles (MEB), and started developing a technique in collaboration with AECL<sup>2</sup> for the hardware, and with SNL-Albuquerque for the electronic instrumentation. The idea was to apply the technique which is under development for safeguarding CANDU spent fuel storage stacks, and uses a cap seal with an AECL random coil (ARC) as unique identity marking which is verified by the Sandia seal pattern reader (SPAR) instrument, in a modified version [16, 17].

### 7.2. Suitability of a BFASS (MARK-II)

In discussing the problem above, it appeared in 1984 that, owing to its small size and good temperature and irradiation resistance, the fuel assembly seal could be a valid candidate for the specific application of the flasks, and JRC soon started a

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<sup>2</sup> AECL = Atomic Energy of Canada Limited.

study on the applicability of its VAK-III sealing technique to the spent fuel containers [18]. In August 1986, a pre-series of new 'bolt-seals', incorporating a VAK-III seal into an MEB bolt, had been manufactured, dry tested and identified at Ispra. They have been sent to Sellafield where they have been installed on a dummy lid in the test pond. A first on-site verification campaign is planned for the end of 1986. Apart from the fact that this new seal (called MARK-II) has the same shape as the normal bolts and can thus be handled with conventional tools, it also has the property of being verifiable in dry conditions (container in a hall) and under water (storage pond). One of its two possible versions is also able to be transformed in an irreversible way into a seal, after the container closure.

### 7.3. Operators' viewpoints

From the reactor operators' point of view the closing of the containers in their plant raises no particular problem. Using the MARK-II system, they could receive sealing heads already identified at the safeguards office and place them at the moment of shipping without the presence of an inspector. If special leak tests have to be made, one can screw in or torque the sealable bolt. Then its head can be replaced by a sealing head which prevents any loosening. The seal would have to be read after such operation either at the shipping place or at the destination plant.

For the pond operators, there is no special difficulty, except that the use of the skip-handler (moving bridge) is required to perform verifications of the containers in the pond. Also, the provision of electric AC power is needed.

### 7.4. Costs

A sealing bolt costs about US \$100 if produced in small series. The instrumentation (excluding the underwater TV system ) is estimated as for the VAK-III system, at about US \$6500, and the verification tools about US \$2500. The operator generally has his own underwater TV system.

### 7.5. Casks

The same technique can be proposed for sealing the large transport containers (casks) in which the flasks (MEB) are transported. It is a matter of screwing in one special sealing bolt which cannot be loosened without breaking its integrity feature. As the bolts for closing the casks are larger than those described above, there is room enough to incorporate in them a seal system. As mentioned, such sealing bolts are verifiable in water or in air, vertically or horizontally (for instance, in a railway carriage).

## 7.6. Other containers and canisters

Similarly, several kinds of containers are used to transport or store fuel material in a tamperproof manner. We can list:

- shipping containers (dry)
- failed fuel rod canisters (wet)
- consolidation rod canisters (wet or dry).

The first example is used to transport pairs of fresh fuel assemblies. The last two are used for storing failed or consolidated fuel assemblies. They are generally closed tamperproof with four bolts or nuts [12]. One of these could be replaced easily by an actual seal such as the MARK-II sealing bolt which has been designed for staying 10 to 15 years in water and provides an identification, which a normal fastener does not. A sealing bolt is used by the operator like a normal bolt and requires only a minimum screwing-in torque to become effective.

## 8. SPENT FUEL STORAGE STACKS OR RACKS (area (f))

In addition to the transport-storage flasks (see area (e) — Section 7) and containers, *fixed* storage structures must also be safeguarded. This is particularly true for the CANDU 600 spent fuel stacks which enclose several hundred spent fuel bundles. But other requirements could also arise for the safeguarding of vertical racks. The principle is to lock such structures with at least one underwater ultrasonic seal which is broken and replaced by another one each time the structure must be opened.

### 8.1. Present technique

Started about ten years ago, the development of the CANDU sealing system has many common points with the BFASS development. Its original shape and concept were studied in collaboration with JRC [19] and derived from the very first fuel assembly seals whose identity was given by internal inclusions. The electronics instrument used is derived from the seal pattern reader developed by SNL-Albuquerque for the FAID verification. Today the CANDU cap seal keeps the original structure. Its identity pattern is given by an AECL random coil placed in an open cavity (see Section 7.1). The ARC serves also as an integrity element. This seal is currently being evaluated at the Canadian Gentilly-2 CANDU reactor where it appears to be easily read with automated equipment.

### 8.2. Alternative techniques

Since the present technique has been reported to comply with the IAEA criteria of seal acceptance [20], it might be that other solutions are not required at all.



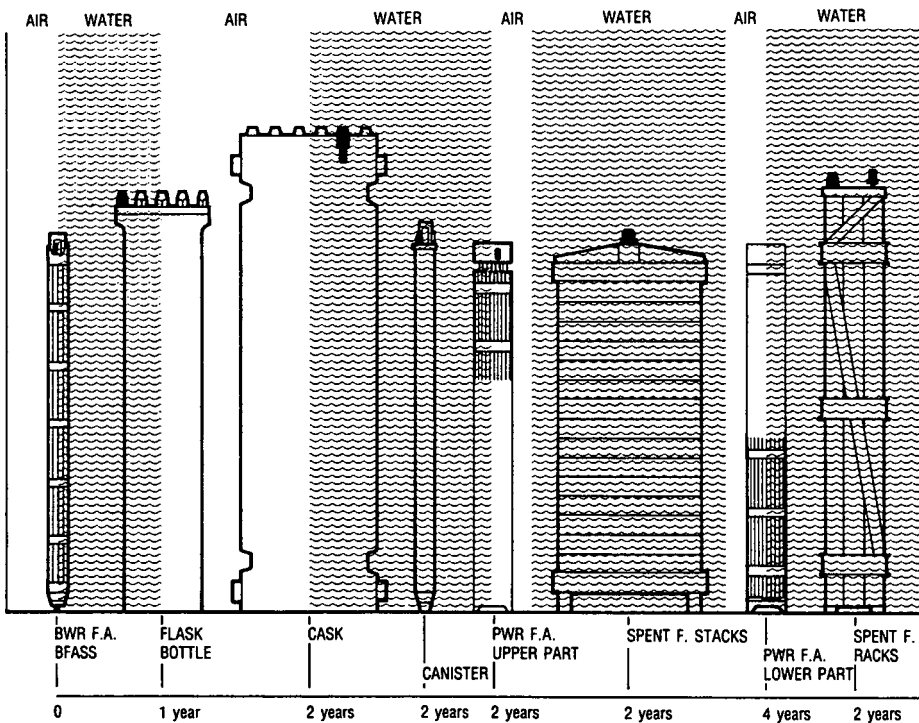


FIG. 5. Estimated time to reach feasible solutions based on the BFASS technology.

Nevertheless, taking into account the progress made with the BFASS, in particular concerning the stability of the signature pattern, it is possible to propose an alternative solution for sealing spent fuel stacks. This technique is directly derived from the VAK-III seal and uses internal cavities randomly distributed in the cap seal body so that no flushing is required to perform ultrasonic measurements. The main progress with respect to the first CANDU seals consists of a marking with large random defects in place of inclusions, and a reading by a scanning transducer in place of a steady one. The handling tools would be the same as in Table I with the exception of the extraction tool (item 5) and the installation tool (item 4) which are not necessary, since normal tools at the plant are used.

Such techniques could apply to any vertical rack locked by a central bar and a cap seal, but the sealing bolts, as described in Section 7, might also be used. It is mainly a question of the mechanical requirements of the locking system and of the possibility of allowing a broken extension to drop into the rack bottom.

Either a SPAR instrument or VAK-45 instruments could be used for the seal verification.

The present costs are shown in Table I. The cost of a cap seal is estimated to be about US \$200 in small series.

## 9. CONCLUSIONS

In recent years, SNL and JRC have devoted considerable effort to demonstrating the BFASS to a feasible level. We now jointly review the application of such systems to other possible safeguards domains which are the sealing of BWR MOX fuel assemblies, PWR fuel assemblies, and containers of different kinds used for assembly transportation and/or storage (Fig. 5). We have considered the practical aspects and shown that we are very near to a feasible solution for BWR MOX assemblies, provided that the safeguards strategy and the fuel cycle are well determined. We are also close to a field solution concerning the sealing of flasks or similar containers. On the other hand, the sealing of PWR fuel assemblies has been shown to be conceptually possible but still necessitates a better knowledge of the actual disassembly capability of the bundles and of the operator's normal practices.

It is suggested that two themes be studied in common by our two laboratories. The first of these is the practical sealing of a limited population of BWR fuel assemblies, for instance from the fabrication plant to the reactor. The second would be the conceptual design of sealing PWR fuel assemblies, considering as a minimum the first level of tamper vulnerability of such structures.

## ACKNOWLEDGEMENT

The authors wish to thank Neil L. Harms from Battelle Pacific Northwest Laboratories for the procurement of useful information about the handling and the constitution of BWR and PWR fuel assemblies.

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## THE MODULAR INTEGRATED VIDEO SYSTEM (MIVS)

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### Abstract

#### THE MODULAR INTEGRATED VIDEO SYSTEM (MIVS).

The Modular Integrated Video System (MIVS) is intended to provide a simple, highly reliable closed circuit television (CCTV) system capable of replacing the IAEA Twin Minolta film camera systems in those safeguarded facilities where mains power is readily available, and in situations where it is desired to have the CCTV camera separated from the CCTV recording console. The paper describes the MIVS and the programme plan which is at present being followed for the development, testing and implementation of the system.

### 1. INTRODUCTION

The International Atomic Energy Agency (IAEA) currently uses containment and surveillance (C/S) as measures complementary to nuclear material accountancy. The principal surveillance measure is optical imagery, achieved through the use of film cameras and, to a limited degree, closed circuit television (CCTV). The IAEA currently has deployed approximately 250 Twin Minolta film camera systems (two XL-401 cameras) to provide most of its optical surveillance requirements.

The Minolta Company, as well as other film camera manufacturers, has discontinued, or soon will discontinue, production of these cameras, and is concentrating on the video market. Because of this, the IAEA plans to replace the Twin Minolta film camera systems with CCTV systems within the next five years.

Upon a request from the IAEA the United States Program for Technical Assistance to IAEA Safeguards (POTAS) has established Task E.67 - Simple, Highly Reliable CCTV System - and assigned it to the Sandia National Laboratories (Sandia).

Under the sponsorship of the US Department of Energy's Office of Safeguards and Security, Sandia has an ongoing project to carry out research, development, testing and evaluation on a wide variety of C/S technologies. As an extension of this project, Sandia was able to respond to the IAEA's request and to commence development of the Modular Integrated Video System (MIVS). This is being designed to replace the Twin Minolta film camera system in (1) those safeguarded facilities where mains power is readily available, and (2) in situations where it is desired to have the CCTV camera separated from the CCTV recording console.

This paper describes the MIVS and the programme plan which is at present being followed for the development, testing and implementation of the system.

## 2. SYSTEM DESIGN GOALS

The MIVS design goals are high reliability, simple inspector interface, ease of maintenance, high tamper resistance and minimum facility impact.

The reliability goal is to provide a system whose reliability is equal to, or better than, the Twin Minolta film camera system. The film capacity of the Minolta camera limits the number of scenes to about 7000. If the system is used to provide unattended surveillance for 90 days, the time interval between scenes must be about 20 minutes. The MIVS video tape recorder (VTR) has a capacity to record about 26 000 scenes, allowing the interval between scenes to be reduced by a factor of four. Such a reduction would be desirable in many safeguards applications. This means, however, that the MIVS would be required to record four times as many scenes. To achieve the desired reliability the MIVS will have two VTRs which would be replaced at regular intervals.

The simple inspector interface goal will be met by the use of a self-instructive, interactive display which guides the operator through all system operations. An auto-start feature will be incorporated, as well as automated record keeping and a small printer which provides a hard copy of pertinent system set-up, diagnostic and environmental exposure data.

Ease of maintenance will be provided by self-monitoring features and modular construction. First-level maintenance, consisting of module replacement, can be performed by inspectors.

Video authentication will be provided to ensure that recorded scenes are from the system's camera. In addition, tamper resistant/indicating housings will be used. These features will provide a high level of tamper resistance.

As regards minimum facility impact, the MIVS consists of small unobtrusive housings, has very low power consumption and is simple to install.

### 3. SYSTEM DESCRIPTION

The MIVS consists of a recording control unit (RCU) in a wall mounted housing, a camera module (CM) and a cable to transmit power and video between the RCU and the CM. A separate portable review station provides a means of reviewing the video tape and will verify that all scenes were recorded.

The RCU, shown in Fig. 1, contains two recorder modules, a control module and a power supply module. The three modules can be easily replaced, with no special tools required. The recorder module consists of a commercial video recorder, with a recording capacity in excess of 26 000 scenes. The recordings are annotated with time, date and event information. The control module [1] contains a liquid crystal display (LCD), a panel of four 'soft-keys' (push-buttons), a small printer and the control electronics. The system's control circuits are microprocessor based. The 8-line by 40-column LCD and four 'soft-keys' operate in an interactive mode (similar to an automatic bank teller) to provide information concerning system status and instructions and choices available for set-up and review of event and diagnostic data. The displayed information is human engineered in a sequential form, making the set-up procedure simple to follow. The printer operates automatically to provide a hard copy of the set-up, environmental and surveillance summary data. Critical maintenance requirements are also printed when necessary.

The power supply module contains an AC/DC power supply and a battery. The AC/DC power supply operates from a 100 to 240 V AC power source at a frequency from 45 to 60 Hz. The battery provides power for system operation during losses of AC power for a minimum of three hours with the system recording at five-minute intervals.

The camera module consists of a solid state video camera and a video authentication circuit board, contained in a tamper resistant housing. The video signal transmitted from the camera module to the RCU is periodically authenticated.

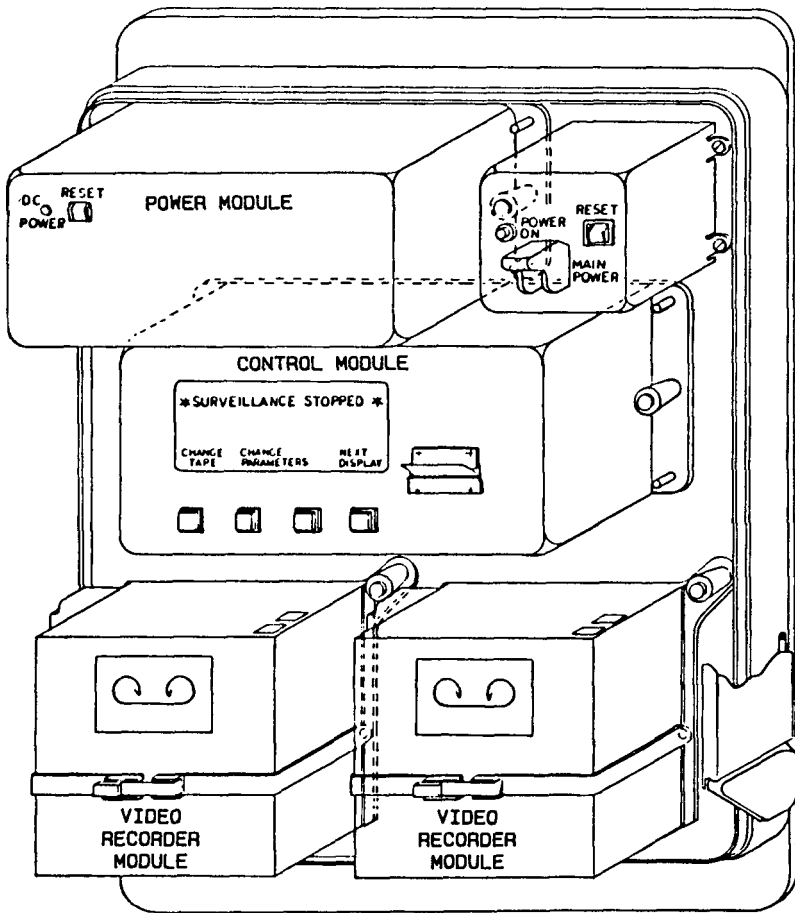


FIG. 1. Artist's concept of the Modular Integrated Video System (MIVS).

Failure to authenticate the video signal results in an event scene being recorded and a tamper event being stored. This authentication is performed at random times and on random pixels in the video.

The review station consists of a recorder module, a review control module, a video monitor and a power supply mounted in a small suitcase type enclosure. The recorder module is the same as that used in the RCU. The review control module is similar to the control module used in the RCU but has different software. Three modes of review are available: normal playback, playback with scene verification, and search and pause at a specified time and date on the video tape.



The MIVS is designed to operate in an indoor environment in ambient temperatures from 5 to 45° Celsius, and relative humidity varying from 10 to 90%. The system, when packaged in its shipping containers, withstands shock and vibration environments found in normal air, truck and rail shipping without operational damage.

Field maintenance consists only of replacing one or more of the four basic modules. No special tools are required for this level of maintenance. It is expected that the recorder module will be replaced routinely about once every 18 months, or after approximately 150 000 scene recordings. Module repair will be performed at Agency Headquarters.

The system reliability will be equal to or better than that of the Twin Minolta film camera system, regardless of the cause of failure. Diagnostic aids are built in to assist the inspector in identifying faulty modules. Module replacement is simple, minimizing the mean time to repair.

The MIVS design incorporates the latest control, display, recording and video camera technologies. Being microprocessor based, it is being designed to use, with minor software modifications, new portable video tape recorders as they become available. Thus, the system will not become obsolete when the recorder chosen for initial use is no longer available.

The MIVS, in production quantities, is expected to cost approximately US\$12 000. The review station will cost about US\$10 000.

#### 4. PROGRAMME PLAN

From the inception of the MIVS, the necessity for a well structured programme plan was recognized. The 1982 IAEA C/S Advisory Group developed a Plan of Activities Leading to Routine Use of C/S Devices. The basic elements of this plan have been applied to the MIVS programme activities, with emphasis on the responsibilities of the IAEA and Sandia throughout the project.

The principal elements of the MIVS programme plan [2], summarized in Fig. 2, are described below.

(1) Identification of need. Before initiating a C/S device development effort it is necessary to determine what safeguards functions the device will perform for the facility(ies) in which it will be used - in essence, the need for the device.

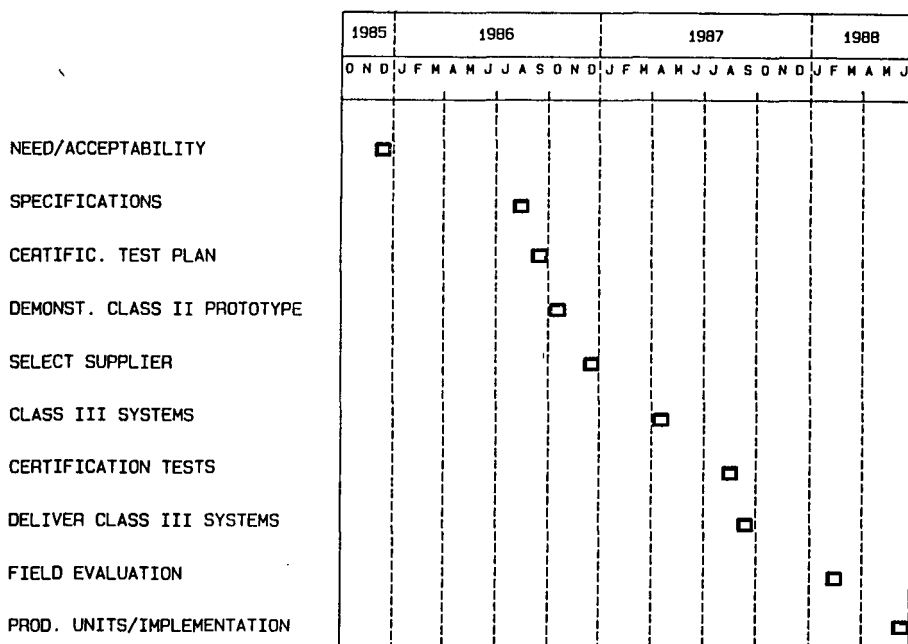


FIG. 2. MIVS programme plan schedule.

In the case of the MIVS, this was very straightforward. The function of the Minolta systems in a wide variety of safeguards approaches is well established. The MIVS is intended to replace this system, thus clearly establishing the need.

(2) Assessment of State and facility operator acceptance. As in (1), the assessment of State and facility operator acceptance was also very straightforward. The current widespread use of optical surveillance provides a high degree of assurance that the MIVS will be accepted by State and facility operators. Despite the apparent simplicity of this step of the programme plan for the MIVS, its importance cannot be overlooked. It is most important in cases where a new C/S device is proposed to serve a new safeguards function. Here the safeguards needs must be clearly defined, and the State and facility operator acceptance thoroughly investigated.

(3) Formulation and approval of system specifications. The need for an agreed-upon set of performance specifications early in a development effort is considered fundamental. The pursuit of such an effort without specifications introduces a high

level of risk that the device may require repetitive development phases that may extend the development period unreasonably (to years), as well as delay the fulfillment of a well established need.

In the case of the MIVS, work on the specifications began some four months before official submission to the IAEA for comments and formal acceptance. Sandia and IAEA personnel had many discussions, resulting in a comprehensive specification to define the functional characteristics of the MIVS and the associated review station, including system reliability requirements.

The resulting specification was submitted to the IAEA by the US Government and approved by the IAEA in August 1986. This process provided a clear understanding of the system requirements, which should result in a timely implementation in the field, provided the system requirements are met.

(4) Formulation and approval of performance and reliability certification test programme. Following IAEA approval of the specifications, Sandia and IAEA personnel are preparing a detailed certification test programme plan, which describes all tests to be performed on the class III field evaluation test units before delivery to the IAEA. The successful completion of these tests will ensure, to the maximum extent possible, that the systems meet IAEA requirements. The certification test programme plan will also be submitted to the IAEA for approval.

(5) Class II prototype development, test and demonstration. The MIVS prototype system development is in progress and was expected to be completed in October 1986. Following tests to verify correct performance of all system elements, the prototype will be demonstrated to the IAEA. This demonstration was planned for November 1986, to be conducted for approximately two weeks to permit all interested IAEA personnel to witness the system operation and to make comments. Based on the previous approval of the specifications, only minor system modifications are expected to be necessary. The agreed-upon modifications will be carried out at Sandia shortly after the demonstration.

(6) Selection of a commercial supplier and manufacture of class III field evaluation units. To expedite implementation of the MIVS, the class III system will be produced by a commercial supplier, selected on a competitive bid basis. Sandia will monitor the production process, perform the acceptance tests, and ensure that adequate design documentation is available at the completion of class III system production.

The use of a commercial supplier at this phase of the project will confirm that the MIVS can be produced effectively and, if desired, provide a ready source for procurement of class IV production units. The proven design documentation will be available for use by any other commercial supplier that may be selected. Initial production is expected to be completed in April 1987.

(7) Performance and reliability certification tests. The certification tests on the class III field evaluation units will commence in May 1987 and will be performed according to the agreed-upon certification test programme plan. These tests will require five months, the time dictated by the period necessary to provide sufficient data to verify statistically the system reliability. The reliability tests will be conducted at IAEA Headquarters by Sandia and IAEA personnel. The environmental and transportation shock/vibration tests will be conducted at Sandia in parallel with the reliability tests. The safety certification test will be performed by an independent safety testing organization.

Upon successful completion of the certification tests, the IAEA and Sandia will prepare a detailed report on all the test results. Six class III field evaluation units and two review stations will be supplied to the IAEA for field evaluation. The test programme is expected to form the basis for rapid entry into the field evaluation phase. Since the six class III systems will be part of the test programme, very little, if any, laboratory tests at IAEA Headquarters will be necessary.

During the certification test period, Sandia and IAEA training and technical services personnel will prepare the initial inspector training and maintenance programmes. These will be completed when the systems and review stations are delivered to the IAEA, the expected date being September 1987.

(8) Field evaluation. Field evaluation of the MIVS is expected to commence in October 1987. Before then the IAEA will obtain State and facility operator permission to install four or five systems in operational environments for evaluation. The field evaluations are expected to last five to six months. Given successful results, the IAEA will place the class III systems in the "limited routine field use" category.

(9) Production order. During or after the field evaluation phase the IAEA is expected to be in a sound position to order a quantity of class IV units. The selected supplier, who may be the same one who produced the class III units, should be capable of producing approximately ten systems per month, with

first deliveries by June 1988. Sandia will assist the Agency, as required, in performing the necessary acceptance testing on the production units.

(10) Implementation. Based on the class III system field experience, full implementation of the MIVS is expected to commence shortly after delivery of the first class IV systems. All training and maintenance procedures/manuals will be completed. At that time, the system would be placed in the "routine safeguards use" category.

## 5. SUMMARY

The programme objective of reaching the implementation stage in a 30-month time frame is ambitious. However, correct execution of the programme plan, with well identified goals and responsibilities, is expected to result in attaining the 30-month objective. A key element in such an achievement is successful system performance during the certification test phase.

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# MONITORING AND LOGGING IN SAFEGUARDS APPLICATIONS

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## Abstract

### MONITORING AND LOGGING IN SAFEGUARDS APPLICATIONS.

In plants which have been designed and built over the last few years, increasing attention has been paid to questions of security and health physics. This tendency, together with advances in measurement technology, nuclear material handling techniques and computerization, has led to increased automation of processes within the nuclear fuel cycle. From the aspect of safeguards, the material being controlled has become increasingly inaccessible. As a result, the Euratom Safeguards Directorate has been designing monitoring and logging systems for safeguards and has now built up a considerable amount of practical experience. In general terms these systems are designed with a sensor to respond to all 'events of interest' — e.g. movement of a crane, passage of a fuel element — and a logging mechanism which records the event in a tamperproof way. If necessary, the system can be connected to a video system which is triggered by an event to record what is happening in the field of view. It is felt that these systems are an important new element for safeguards strategies, helping to solve problems which other safeguards measures cannot do. The essential characteristics of these systems are given, together with some design considerations and practical examples.

## 1. INTRODUCTION

The original technical measures used in safeguards for containment and surveillance (C/S) — camera units and cap seals — have served and continue to serve a useful function. Up to now no satisfactory alternative has been available for the twin-camera unit which has been extensively applied in installations under Euratom and IAEA safeguards. However, the camera is no longer the best solution, and frequently its application requires more than this simple technique can provide. Cameras are most useful in situations where no movement is expected between inspections, and this can be confirmed by the camera. This is rarely, if ever, met in practice and the inspectors are left with the almost impossible task of determining whether or not observed movements could signify an attempted diversion. Thus, one must conclude that the camera, while performing an immensely useful function as a safeguards technical 'work-horse' for a number of years, has only been a stop-gap measure until a better solution was

found. This move away from reliance on cameras has been hastened by the fact that such units are now becoming obsolete.

The cap seal has been extensively used and has proved very effective. However, the inability to verify in situ its relatively crude design and the fact that it can only be used once have led to several interesting new developments in sealing techniques.

Several changes have occurred that indicate the direction safeguards should take in applying C/S measures. More stringent safety and security requirements, in particular the need to reduce radiation exposure of personnel, are now recognized so that installations are now being designed in which nuclear material subject to safeguards control is made less accessible than previously. This means that straightforward optical surveillance devices no longer have a view of the material under control, thus highlighting the limitations of traditional camera surveillance.

Other changes are recent developments in video technology, detectors (crane sensors, motion detectors, gamma and neutron detectors, etc.), tamperproof recording/interrogation systems, triggering systems and electronic, in situ verifiable seals. These can be combined in such a way that the safeguards measure is applied much nearer to the material being controlled and is therefore much more satisfactory as a safeguards tool. An event of interest is defined - the passage of a drum through an exit, any movement during periods where no movement is permitted, the passage of an irradiated fuel element at a key point in a storage pond, etc. - and a detector is designed to recognize the event. A logging system which can be interrogated periodically by the inspector records the event in a tamperproof way, and the recognition of an event of interest can trigger a video camera for a prescribed period to record the event in question or, if the camera operates quasi-continuously, special reviewing techniques can be used to concentrate on certain events within the period in question.

## 2. DATA LOGGING SYSTEM

Traditional C/S measures must therefore be supplemented, using for instance material and motion sensitive sensors. These sensor-recording systems, termed monitoring and logging systems, must be tamper resistant and operate in unattended mode, thereby replacing the inspector over periods of time. Sensors may be optical (video cameras), on/off type (electronic seals, or threshold detectors such as video motion detectors, intrusion detectors, etc.), analogue type (count rate, item



counters, temperature, liquid level, etc.) and/or complex measurement devices (doorway monitors, neutron coincidence counters or, for example, LASSY, the underwater laser surveillance system for storage ponds).

Depending on the application, a suitable choice of sensors must be made and the relevant information must be stored in a protected way. Thus, it should be possible to design a system which provides independent verification for safeguards relevant to areas where changes of safeguards interest occur in the absence of inspectors.

A particular problem is the time consuming review of optical surveillance records, especially if safeguards relevant events seldom happen. With the help of suitably chosen sensors, special events may be logged or marked on the video tape (modern surveillance recorders have alarm recording features permitting rapid access to these alarm points). Alternatively, depending on the sensor, the recorder may only be operated during 'alarm' situations. However, in many cases the scenes prior to the alarm (particularly if a video motion sensor is used) will also be of interest for the review. Also, a check on whether the sensor detects all relevant events can only be done if the whole period is covered by the video recordings.

Although, as described above, such systems are being applied more and more, similar systems have already been applied in some cases for some years.

### 3. EXAMPLES OF APPLICATION IN EURATOM SAFEGUARDS

The first unattended device was a compact pebble sampling device for the fuel pebble fabrication plant (1975) for the Thorium High Temperature Reactor [1]. The device and sampling tube are mounted and sealed on to the pebble storage (shipment) barrel. The pebbles are counted (up to 1000) and recorded by an electromechanical counter and up to 10 pebbles (preselectable between 1 and 1000) are diverted into the sampling tube. With 10 such devices at least two weeks' production is counted and sampled in the absence of inspectors. After the NDA measurement the inspector returns the pebbles to the appropriate barrels, seals them and prepares the barrels for the next production period [1].

Two functionally similar pebble sampling devices are being constructed for the loading station of the pebble bed reactor in order to verify that only pebbles of the known type have been transferred into the inaccessible core. These devices are

very complex and incorporate a measure of redundancy to meet the high safety and reliability constraints required for the reactor operation, ensuring the ability to feed pebbles under all circumstances.

Until these sampling devices become operational a gamma monitor has been set up in a sealed box underneath the position of the pebble drum attached to the loading station. A gamma measurement is made every two minutes during the one and a half days (approximately) which it takes to unload the barrel; the printed result shows the barrel being positioned and unloaded and the barrel loading operations through the typical radiation-time pattern. The monitor consists of a NaI detector, and a SAM-2 (with interface) operated by an HP41CX. The area is subject to camera unit surveillance. The system has operated reliably for over a year. In future this system will be used to characterize the contents of spent fuel drums being shipped. See also Ref. [2].

During the design and construction of the fast reactor SNR 300 in collaboration with Euratom the inaccessible inventory instrumentation system (IIIS) was developed (starting in 1973) as part of the safeguards system. This system<sup>1</sup> is a microprocessor controlled monitor which combines the information of two (redundant) position indicators of the fuel handling devices with one neutron and gamma measurement system for each device, and is independent of the operator's instrumentation. The measurement system is designed so that the different types of fresh and irradiated assembly can be distinguished. The sensor information is collected automatically and evaluated, and the inventories of the different inaccessible areas are continuously updated. The reliability is assured by a redundancy of sensors and memory devices; in addition, a system is being installed by which Euratom is informed periodically in a tamperproof way of any component breakdown so as to provide timely repair. The IIIS has been cold tested. The reactor plant has not yet started operation.

A more recent development is the level monitoring system for a Pu nitrate store. In this case the operator's instrumentation is used (the capacitance sensors with the signal conversion units). The data collection and evaluation system is a straightforward computing task. The output data will be printed and be available on a mass data store.

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<sup>1</sup> Joint Programme on the Technical Development and Further Improvement of IAEA Safeguards between the Government of the Federal Republic of Germany and the IAEA.

The software is designed for two functions:

- To monitor continuously the status of each tank, e.g. constant volume, increasing/decreasing volume or homogenization procedure. For any status change, an output is prepared.
- To evaluate at any time the liquid volume in each tank on the basis of the calibration function of the tank.

The output of the monitoring system combined with the verification measurements of the input batches and with concentration measurements will allow the Pu content of the store and of the transfers to the process to be verified.

#### 4. DESIGN CONCEPT FOR MONITORS

The design of monitoring devices as unique, complex instruments is no longer considered for future developments, because the costs are very high, and because operation and maintenance experience must be developed each time from scratch; therefore, the reliability of the system will on average not be satisfactory or will even be unknown initially.

Therefore, we support a modular design with the following main components:

- (a) Computer-controller (monitor console);
- (b) Input/output devices (monitor console);
- (c) Interface unit for communication between the controller and the sensor units (monitor console);
- (d) Power supply with partial autonomy (monitor console);
- (e) Sensors (sensor unit);
- (f) Sensor control unit for communication between the interface unit and the sensor (sensor unit).

Components (c), (d) and (f) can be standardized; for (a) and (b), certain standards may be defined but they can be chosen from a wide range of commercial components depending on the amount of software and data flow rate (anything between a programmable pocket calculator and a personal computer with the related peripherals may be chosen). The sensors (e) and (f) are the only components which may be specific for monitoring applications.

The authenticity of the collected information can be ensured through encrypted communications, or through protected communication lines. For low rate data transmission we choose encryption of standard length words with a type of DES code.

For high rate data a partial encryption or a protected optical fibre cable would be available as a choice.

The overall reliability of the monitoring system is a function of the reliability of the components. Owing to the modularity of the system only the sensors, and that part of the controller software which contains the logic combinations of the different sensor signals, differ between any two monitoring systems. Suitable redundancy, in particular by diversification of the sensors, is the basic requirement to arrive at the required high reliability.

Components (a) to (d) are commercially available. The sensor control unit is commercially available integrated with the VACOSS seal and as a separate unit to be combined with any on/off sensor. The control unit for a generalized sensor (for an analogue sensor or complex sensors) has been specified and is expected to be available during 1987.

The sensors are the main problem in designing a monitoring device for a specific application. It is our experience that they require the main development effort, particularly the video system. To record optical surveillance information a set of cameras and redundant recording systems are normally used. A typical video sensor may be a video motion detector or a more sophisticated image processing device indicating the existence of a possible safeguards relevant situation. This information (threshold signal) may be combined logically with the information of other sensors and documented (printout, mass storage) and recorded with the video information as an alarm (feature of modern time lapse recorders). Today's recording systems also allow remote operation through an external controller.

## 5. IMPLEMENTATION PROBLEMS

Normally, the design of monitoring systems should mostly go in parallel with plant design and construction. Sometimes this requires consultations with the safeguards authorities many years before the device in question becomes operational. The IIIS is an outstanding example.

The use of standardized equipment, as mentioned before, will allow the main characteristics and the reliability to be assessed in advance. Only the sensor problem (choice, design, installation) would remain as the specific R & D problem if the monitor uses sensors installed in the operator's equipment.

Tamperproof data transmission is also a problem in practice. Installed cables are in most cases no longer verifiable. However, the credibility of monitors depends ultimately on the authenticity of the data. The proposed data encryption, together with the monitoring procedure (interrogation of sensors by the monitor) and with the specific characteristics of the sensor control units, allows substantial security against occasional line deficiencies, which are, of course, detected within one interrogation cycle.

## 6. OPERATIONAL CONSIDERATIONS

Last but not least, the monitoring system must be designed in an inspector friendly way. The inspector will interrogate the system periodically and obtain an output in a format adapted to his needs with date and time of events (including the description of events) and statistics on the monitor performance. It is also desirable to provide the safeguards authorities at regular intervals with the condition of the monitor in order to allow timely repair of faulty components. This would considerably raise the reliability beyond that which can be obtained by redundancy alone.

## 7. CONCLUSION

Monitoring and logging systems represent an important development in safeguards technology and provide an opportunity to apply effective safeguards in a wide range of circumstances.

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## **О ВОЗМОЖНОСТИ ПОЛУЧЕНИЯ КОЛИЧЕСТВЕННЫХ ЗАКЛЮЧЕНИЙ ДЛЯ ЦЕЛЕЙ ГАРАНТИЙ С ПОМОЩЬЮ ТЕХНИЧЕСКИХ МЕР СОХРАНЕНИЯ/НАБЛЮДЕНИЯ**

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### **Abstract—Аннотация**

#### **THE POSSIBILITY OF USING TECHNICAL CONTAINMENT/SURVEILLANCE MEASURES IN ORDER TO REACH QUANTITATIVE CONCLUSIONS FOR SAFEGUARDS PURPOSES.**

The report sets out the results obtained using experimental (prototype) versions of an irradiated assembly counter (SOS-1) designed to provide an independent, automatic record of irradiated fuel assemblies loaded under water into a transport container for dispatch from the spent fuel store of a power plant with WWER-440 type light water reactors, and the possibility is examined of using the counter to reach quantitative conclusions on the dispatch of assemblies from such a plant.

#### **О ВОЗМОЖНОСТИ ПОЛУЧЕНИЯ КОЛИЧЕСТВЕННЫХ ЗАКЛЮЧЕНИЙ ДЛЯ ЦЕЛЕЙ ГАРАНТИЙ С ПОМОЩЬЮ ТЕХНИЧЕСКИХ МЕР СОХРАНЕНИЯ/НАБЛЮДЕНИЯ.**

В докладе излагаются результаты разработки опытных (предсерийных) образцов счетчика облученных сборок (SOS-1), предназначенного для независимой автоматизированной регистрации облученных тепловыделяющих сборок (ТВС), загруженных под водой в транспортный чехол для отправки из хранилища атомной электростанции (АЭС) с легководными реакторами типа ВВЭР-440, и рассматривается возможность получения с его помощью количественных заключений об отправке сборок со станции.

На легководных АЭС облученные ТВС после выгрузки из реактора помещаются в бассейн выдержки. По истечении 2—3 лет облученные ТВС могут быть отправлены со станции. Для этой цели ТВС необходимо переместить под водой хранилища в защитный чехол, который затем извлекается наружу и транспортируется. Учитывая, что все операции по перемещению ТВС в защитный чехол осуществляются непосредственно под водой, то современные технические средства наблюдения для целей международных гарантий, например, телевизионные системы и фильмовые системы, как правило могут предоставить лишь информацию об использовании оборудования,

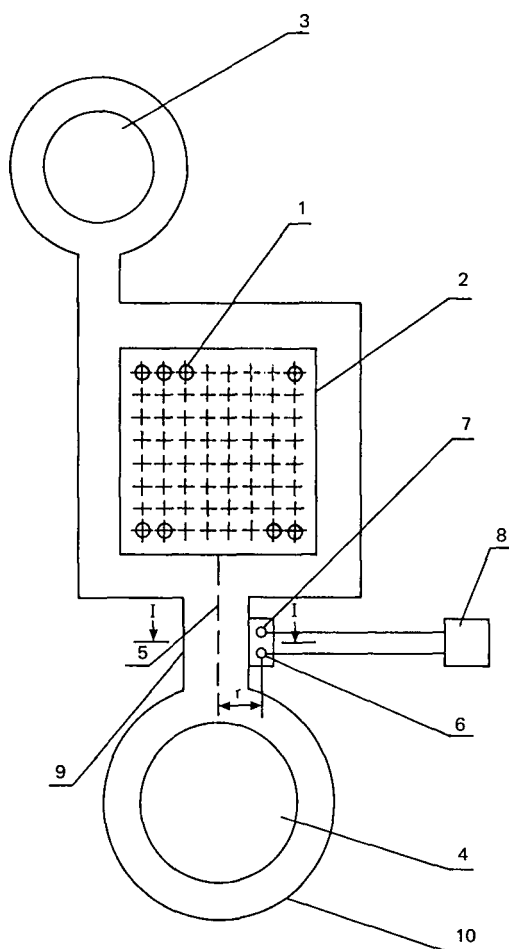


Рис. 1. Схема размещения счетчика облученных ТВС в бассейне выдержки: 1 – ТВС; 2 – стеллаж; 3 – реактор; 4 – транспортный контейнер; 5 – штатный маршрут перемещения ТВС; 6 – блок детектирования нейтронного излучения; 7 – блок детектирования гамма-излучения; 8 – блок обработки информации; 9 – транспортный коридор; 10 – транспортный отсек бассейна выдержки.

предназначенного для транспортировки сборок (краны, контейнеры и т.п.). Представляется, что такая информация не может обеспечить в полной мере решение проблемы, связанной с получением МАГАТЭ независимым способом надежной и достоверной количественной информации, подтверждающей сообщения оператора о количестве ТВС, отправленных со станции. Повысить эффективность гарантий на этом участке топливного цикла могло бы применение технических средств, регистрирующих автоматически независимо от оператора АЭС количество облученных ТВС,



перемещаемых под водой в транспортный чехол. Одно из возможных решений этой проблемы применительно к АЭС с реакторами типа ВВЭР-440 было предложено в работах [1–3]. Для предварительной оценки этого способа регистрации был изготовлен и опробован в течение короткого промежутка времени в условиях АЭС прототип счетчика отработавших ТВС [2].

Первоначальные результаты, полученные по этой разработке, показали принципиальную возможность создания такого прибора-счетчика. Однако эти результаты носили лишь оценочный характер, так как были получены на макете, не предназначенном для длительных испытаний.

Для специалистов МАГАТЭ в 1984 г. в СССР была проведена демонстрация этого макета на Нововоронежской АЭС (НВ АЭС). Учитывая последующие рекомендации МАГАТЭ о необходимости разработки опытных образцов приборов, предназначенных для долговременных испытаний в реальных условиях АЭС, такая работа недавно завершена в рамках Программы СССР научно-технической поддержки гарантий МАГАТЭ. Создание этих опытных образцов потребовало новых конструктивных и схематехнических решений.

Разработанные опытные образцы предназначены для установки на АЭС с реакторами типа ВВЭР-440, на которых имеется транспортный коридор между бассейном выдержки и отсеком для транспортного контейнера (рис. 1). Облученные ТВС размещаются на стеллажах, располагаемых в отсеке хранения бассейна выдержки реактора. Транспортный контейнер расположен в транспортном отсеке бассейна выдержки. Для отправки сборки с АЭС необходимо переместить сборку из бассейна выдержки в транспортный отсек и загрузить в чехол, что осуществляется по штатному маршруту, обозначенному на рис. 1 пунктирной линией.

Для установления перемещения ТВС счетчик регистрирует как нейтронное, так и гамма-излучения непосредственно под водой. Регистрация факта прохождения ТВС происходит только в случае обнаружения обоих видов излучения. Это значительно уменьшает вероятность незафиксированного перемещения ТВС с использованием преднамеренных действий, так как экранировка или имитация одновременно двух излучений затруднены.

Предложенный радиационный способ регистрации отработавших ТВС по двум видам излучения (нейтронному и гамма) позволяет определить направление перемещения ТВС и дает возможность автоматизировать непосредственно в условиях АЭС счет числа отработавшихборок, отправляемых со станции. Счетчик содержит три блока детектирования нейтронного излучения, блок детектирования гамма-излучения, а также блок накопления и обработки информации, который может быть отнесен от блоков детектирования на расстояние не менее 25 м. Расположение блоков счетчика показано на рис. 1 и рис. 2.

При таком расположении аппаратуры отработавшие ТВС перемещаются по штатному маршруту из отсека выдержки мимо блоков детектирования нейтронного и гамма-излучений в транспортный отсек хранилища.

Эти блоки детектирования расположены на расстоянии около 0,5 м от перемещаемых ТВС. Все блоки детектирования при помощи соединительных кабелей под-

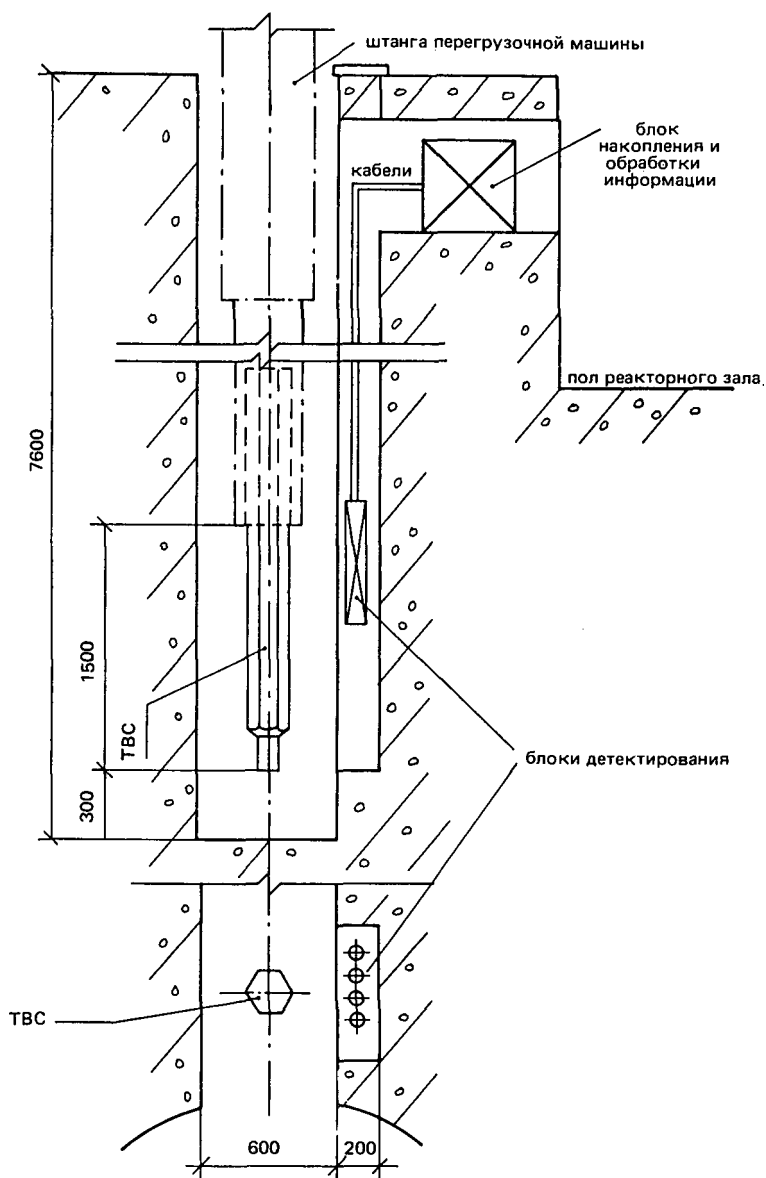


РИС. 2. Схема расположения прибора-счетчика числа ТВС на станции (разрез I-I рис. 1).

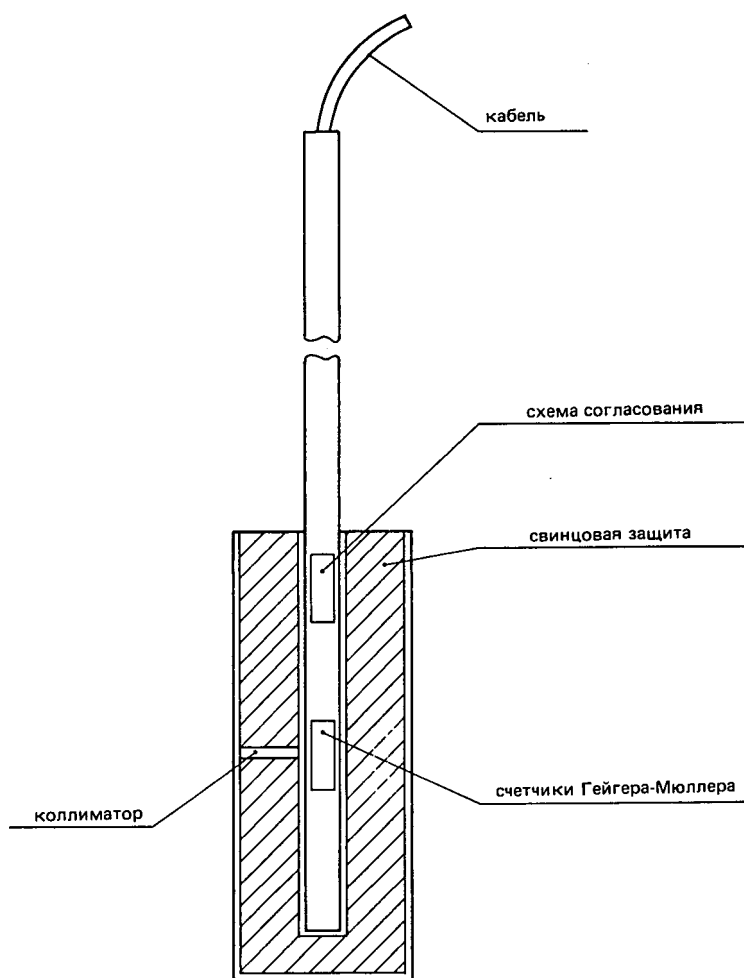


РИС. 3. Блок детектирования гамма-излучения.

ключены к входу блока накопления и обработки информации. При прохождении ТВС в непосредственной близости от блоков детектирования на их выходах формируются сигналы в виде импульсных последовательностей, поступающих на вход блока накопления и обработки информации, который выдает информацию о количестве перемещенных ТВС в соответствии с заданными алгоритмами идентификации, как в прямом направлении, так и в обратном.

Блок детектирования гамма-излучения представляет собой цилиндрический защитный корпус, подвешиваемый на тросах в сухой нише транспортного коридора или непосредственно в воде бассейна выдержки (рис.3). Корпус изготовлен из

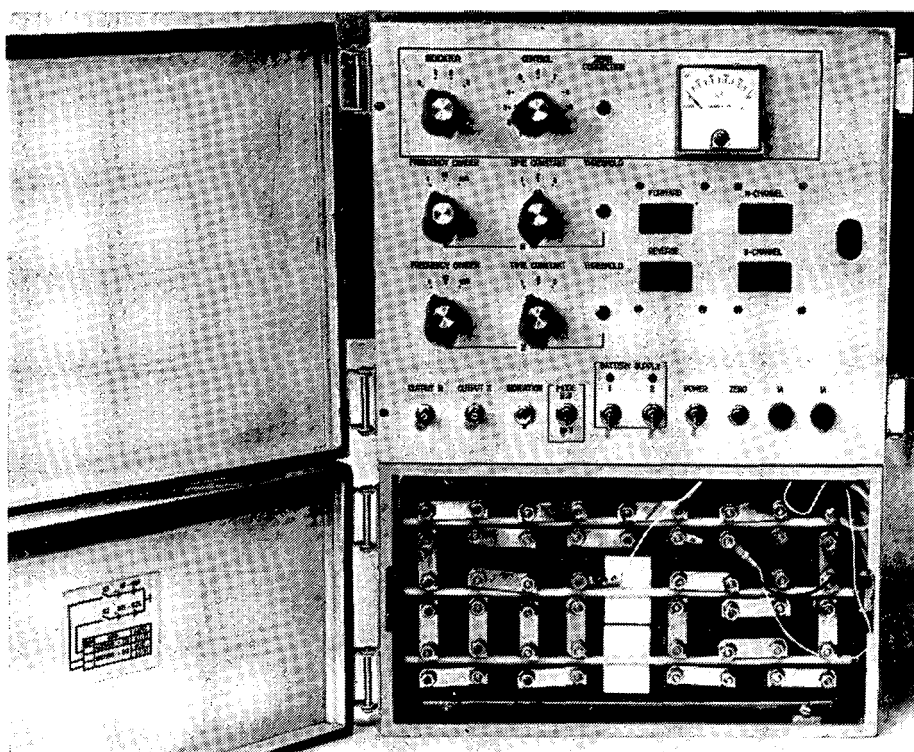


РИС. 4. Фото передней панели блока накопления и обработки информации.

свинца, помещенного в кожух из нержавеющей стали. Внутри защитного корпуса введена стальная труба с тонкостенным стеклом, с размещенными в нем двумя газоразрядными счетчиками Гейгера-Мюллера, напротив которого расположено коллимационное отверстие. Крепление счетчиков в цанговых гнездах обеспечивает их удобную замену в процессе эксплуатации.

Защитный корпус устанавливается в бассейн выдержки или нишу один раз при монтаже прибора. В процессе эксплуатации счетчика нет необходимости в поднятии защитного корпуса на поверхность бассейна, т.к. для замены счетчиков на поверхность бассейна вытаскивается только легкая тонкостенная труба диаметром 40 мм.

Блок детектирования имеет габаритные размеры 400×185×155 мм, а суммарный вес гамма-детекторов со схемой согласования не превышает 2 кг.

Конструкция блока детектирования нейтронного излучения выполнена аналогично. Внутри защитного корпуса размещено три коронных счетчика тепловых нейтронов.

Габариты блока 1690×185×155 мм, однако суммарный вес всех нейтронных детекторов со схемой согласования не превышает 4 кг.

Блок накопления и обработки информации (рис. 4) выполнен в виде настенного шкафа, состоящего из двух отсеков, закрытых откидывающимися панелями. В верхнем отсеке расположена кассета с печатными платами и блок с трансформаторами. На переднюю панель вынесены переключатели, цифровые табло и другие органы управления.

В нижнем отсеке расположены 25 герметических никель-кадмиевых аккумуляторов емкостью 11 А·ч. Разъемы для внешнего подключения кабелей вынесены на нижнюю поверхность блока.

Для предотвращения доступа внутрь блока передняя панель нижнего отсека имеет приспособление для опломбирования, а верхнего — крышку с замком.

Габариты блока 410×510×338 мм. Вес до 30 кг.

В состав блока накопления и обработки информации входит зарядное устройство, блок низковольтного питания, блок усилителей, блок питания высоковольтный, блок алгоритмов, блок интенсивметров, блок индикации.

Зарядное устройство предназначено для автоматической зарядки аккумуляторов в процессе эксплуатации.

Блок низковольтного питания обеспечивает получение стабилизированных напряжений 5, 9 и 12 В.

Блок усилителей обеспечивает усиление и дискриминацию по уровню импульсов напряжения, поступающих из блоков детектирования.

Блок интенсивметра содержит два идентичных канала. На вход первого поступают импульсы из нейтронного канала, а на вход второго из гамма-канала. После преобразования сигналы с выходов каналов в виде перепадов напряжения поступают на выход блока алгоритмов.

Блок алгоритмов предназначен для обработки сигналов, поступающих с блока интенсивметра в соответствии с заданными алгоритмами идентификации ТВС.

Блок индикации содержит две группы семисегментных индикаторов по три индикатора в каждой группе, на которых отображается информация о количестве идентифицированных ТВС.

При этом прибор обеспечивает регистрацию на отдельных цифровых табло количества перемещаемых ТВС в прямом (штатном) и обратном направлениях при совместной регистрации двух видов излучений (нейтронного и гамма), а также обеспечивает регистрацию количества перемещаемых объектов, испускающих только один вид излучения — нейтронное или гамма, без указания направления перемещения.

Таким образом, счетчик облученных тепловыделяющих сборок позволяет:

- осуществить идентификацию ТВС и определить направление их перемещения на основе регистрации нейтронного и гамма-излучений продуктов деления сборок;
- осуществить счет отработавших ТВС при их перемещении со скоростью до 0,25 мм/с в бассейне выдержки реакторов типа ВВЭР-440;
- сохранить работоспособность блоков детектирования при температуре воды от +45 до +65° С;

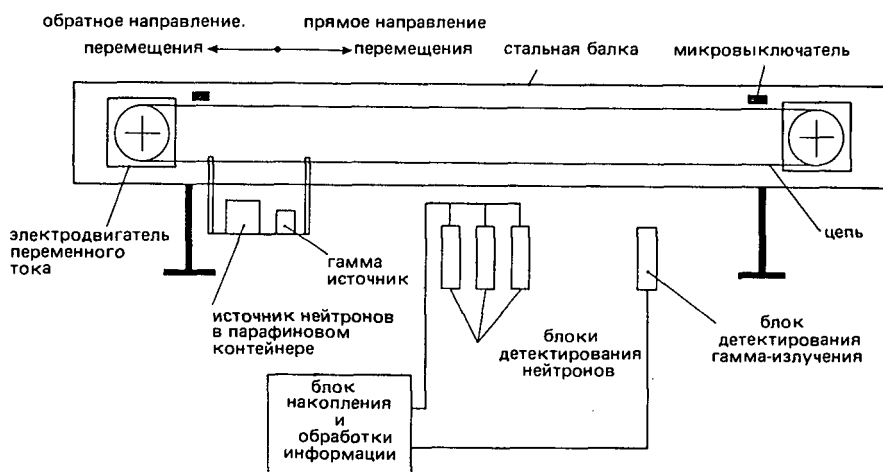


РИС. 5. Установка для испытаний счетчика.

- сохранить работоспособность при работе от промышленной сети переменного тока напряжением  $220 \pm \frac{10}{15}\%$  частотой  $(50 \pm 1)$  Гц. При отключении от сети счетчик автоматически переключается на батарейное питание;
- сохранить работоспособность при вибрации с частотой 25 Гц и амплитудой не более 0,1 мм;
- эксплуатировать его на АЭС в течение 6 лет при минимальном обслуживании.

Для проведения лабораторных испытаний счетчика облученных ТВС была разработана и изготовлена специальная установка, показанная на рис. 5. Установка представляет из себя сканирующее устройство с кареткой. На каретке устанавливаются источники гамма-излучения и источники нейтронов, которые могут автоматически перемещаться с определенной скоростью мимо блоков детектирования счетчика, располагаемых на определенном расстоянии от сканирующего устройства.

При испытаниях имитация физических полей отработавших ТВС производилась с использованием плутоний-берилиевых источников быстрых нейтронов с выходом нейтронов  $5 \times 10^5 \text{ с}^{-1}$ , окруженных парафиновой защитой и источников гамма-излучения на основе радионуклидов америция-241 с активностью  $1,3 \times 10^{10} \text{ Бк}$  (35 Ки) и цезия-137 с активностью  $1,23 \times 10^8 \text{ Бк}$  (3,3 мКи).

Вероятность безошибочного подсчета имитаторов ТВС при их перемещении в прямом и обратном направлении мимо блоков детектирования счетчиков подсчитывалась по формуле:

$$P_{\text{прям. (обр.)}} = \left( 1 - \frac{N - n_{\text{прям. (обр.)}}}{N} \right) \times 100\%,$$

где  $N$  — число перемещенных ТВС в прямом и обратном направлениях;  $n_{\text{прям. (обр.)}}$  — показания счетчика о перемещениях в прямом и обратном направлениях.

При многократных перемещениях ТВС не было зафиксировано просчетов или ложных срабатываний прибора-счетчика, т. е. вероятность безошибочного подсчета ТВС при лабораторных испытаниях была равна 100%.

В процессе лабораторных испытаний проверялась также работоспособность прибора при его питании от автономных аккумуляторов. Для этого при регистрации перемещений ТВС в произвольные моменты времени прибор отключался от промышленной сети. Сбоев показаний и просчетов также не обнаружено. Максимальная гарантированная длительность работы счетчика на автономном питании — 48 часов. По теоретической оценке прибор может сохранять работоспособность до 72 час.

В процессе лабораторных испытаний проводилась проверка прибора на устойчивость к механическим воздействиям, к воздействию повышенных и пониженных температур.

Результаты испытаний подтвердили соответствие прибора установленным техническим требованиям.

Лабораторные испытания опытных предсерийных образцов счетчика показали, что проделанные конструкционные переработки макетного образца и введение ряда других новых технических решений позволили значительно повысить надежность работы прибора и экспериментально подтвердили основные технические характеристики SOS-1.

Результаты лабораторных испытаний свидетельствуют о целесообразности проведения длительных полномасштабных испытаний образцов SOS-1 в реальных условиях АЭС.

В случае разработки соответствующих модификаций представляется возможным использование подобного счетчика для различных ядерных установок, например, крупномасштабных долговременных хранилищ и АЭС других типов.

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## POSTER PRESENTATIONS

IAEA-SM-293/41P

### DEVELOPMENT OF AN ULTRASONIC TAMPER INDICATOR

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The inspectorate requires a suitable surveillance measure to be applied to a container for nuclear material or for an inspector's equipment so as to ensure the container's integrity during his absence. The most effective surveillance can be obtained by monitoring the entire container surface, and by releasing an alarm should it be tampered with. From this aspect, we have developed a tamper indicator which can be applied to a box type container, and uses the ultrasonic technique. This has been developed during the last two years.

The indicator consists of two basic units — a monitoring and a verification unit (Fig. 1). The monitoring unit has an omnidirectional transducer and a microcomputer. In the verification unit a keyboard (ten-key), an LCD display and a printer are installed. The monitoring unit is operated by the keyboard of the verification unit to which it is connected. Each unit can be driven by dry batteries.

The monitoring unit is installed in a container together with the material(s) to be surveyed. When the container is closed and the containment condition is established, the transducer begins to transmit ultrasonic waves towards the inner walls of the container and also to receive waves reflected by these walls within a very short time of transmission. If the container shape is not changed, the reflected waves always appear with almost identical waveforms (Fig. 2). However, if the container shape has been changed by, for example, its lid being opened, an obvious difference should be observed between the two waves (Fig. 3). After receiving the reflected waves, the microcomputer converts them to 256 point digital data according to the reception sequence, and these digital data are stored in the microcomputer memory, and subsequently analysed by comparing

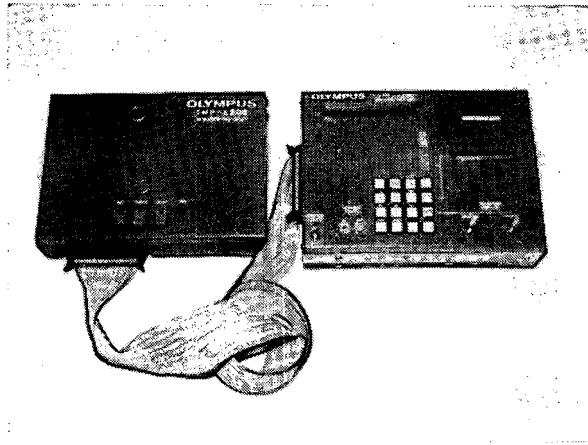


FIG. 1. Monitoring unit (left); and verification unit (right).

them with the two data set, produced by the preceding waves. This is repeated periodically at a predetermined interval, which can be selected by the verification unit.

Preliminary experiments showed that a difference in the energy of the ultrasonic waves can be expressed by the mean value of the differences between the two waves. Also it was confirmed that the two waveforms, corresponding to a normal case and an abnormal case, can be distinguished from each other if we compare the magnitudes of two numerical values defined by the following equation:

$$X = (1/256) \times \Sigma [F^n(I) - G^{n+1}(I)]$$

$$Z = \sqrt{(1/256) \times \Sigma [F^n(I) - G^{n+1}(I) - X]^2}$$

where  $F^n(I)$  is the wave corresponding to a normal case; and  
 $G^{n+1}(I)$  is the wave corresponding to an abnormal case.

When the two resulting reflected waves have a similar waveform [ $F^n(I) \cong G^{n+1}(I)$ ], both the mean value  $X$  and the variance value  $Z$  shall be small. But if they have different waveforms, these two values shall increase, depending on the degree of the difference between the two waveforms. Whenever calculated, these values are compared with a reference threshold value that is preset by an inspector using the verification unit. If these values are larger than their respective threshold values, they are recorded in the memory of the monitoring unit together with the date and time of occurrence. The recorded values can be

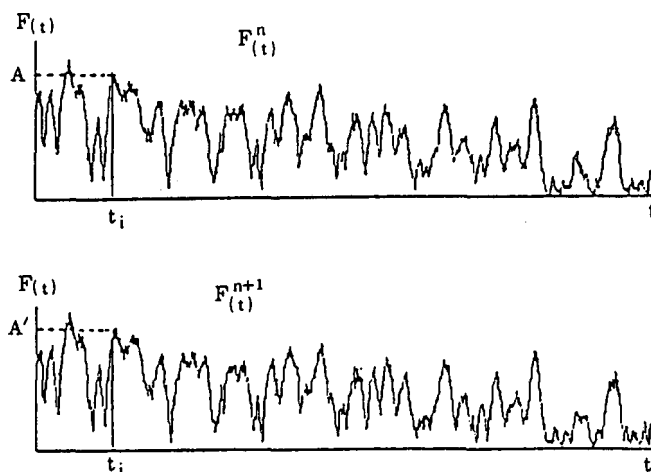


FIG. 2. Reflected waves of the normal case.

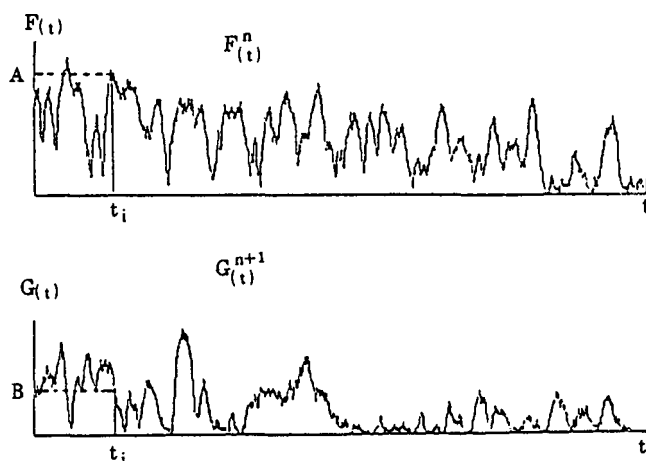


FIG. 3. Reflected waves of the normal and the abnormal case.

read out by an inspector using the verification unit by connecting it to the monitoring unit.

The equipment described above was tested in early 1985. The results show that the equipment is able to detect a hole with a diameter greater than 4 cm anywhere on the box. Three false alarms were observed during continuous operation (three months). The equipment can run for more than three months without the batteries being changed, and it can be used as effective surveillance equipment applicable to a box type container used for nuclear material or inspector's equipment.

IAEA-SM-293/137P

**A TELEVISION LINK WITH HIGH  
TAMPER RESISTANCE****B. RICHTER, G. STEIN**Kernforschungsanlage Jülich GmbH,  
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Within the framework of the programme of the Federal Republic of Germany in support of the IAEA, the development of a TV link with high tamper resistance is under way. In a preceding paper [1] the principle of authentication with the objective of securing data transmission is discussed. This poster presents the technical realization of the authentication method for transmitting video information.

One of the basic safeguards measures is optical surveillance with film camera systems. Within the coming years film cameras will be replaced by closed circuit television (CCTV) systems, which offer certain advantages compared with film cameras. CCTV systems can in principle be operated in compact systems — camera and recorder in one unit ('compact CCTV') — or in systems with camera and recorder in separate housings. Both units are connected to each other via cable or optical fibre. In this case the transmission of video information from the camera to the recorder has to be secured. Investigations on the technical measures to be applied resulted in excluding preventive measures, but focusing on indicating features. A cryptographic approach in connection with standard transmission lines has been selected as the basic application in CCTV systems.

The authentication system comprises two units, transmitter and receiver units. They are connected to each other via a standard coaxial cable or an optical fibre. Both video information and special data are transmitted via this cable. The video information corresponds to the customary worldwide standards (Comité consultatif international des Radiocommunications (CCIR) or the Electrical Industry Association (EIA)), so that the image information can be constantly displayed on a standard monitor on the receiver side. Securing data are normally not displayed on a standard monitor. However, in principle this information exchange is accessible to every opponent. The transmission path is also not under surveillance.

Attached to the video camera is a transmitter unit to generate an authentication signal that is built into the video signal.

The plain text of the signal may be composed of the following elements:

- time
- (x, y) co-ordination pair, generated by a random number generator
- time integral, formed in the video image via line segment on the (x, y) position, and
- external physical parameter such as pressure, temperature, moisture, etc., in the camera.

This plain text is enciphered in the transmitter unit by a suitable procedure (e.g., DES) and transmitted to the receiver unit.

The receiver unit forms the same time integral to the corresponding image on the (x, y) position and compares this with the deciphered information of the transmitter unit. If the plain text does not correspond to the calculated information (time integral) plus time with the deciphered information in the receiver unit within a given tolerance range, a deception attack is taking place. The receiver unit can then report the deception attack on a control instrument.

For proof of time authenticity, the authentication signal contains the time (transmitter unit). This time is a fundamental inherent constituent of the security process. If the time is dispensed with, the procedure can be breached by the simplest means.

## REFERENCE

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IAEA-SM-293/39P

**CERENKOV RADIATION VIEWING DEVICE –  
DEVELOPMENT AND EXPERIMENTAL RESULTS\***

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Spent fuels in a storage pool produce Cerenkov radiation ranging from ultra-violet to infrared. Observation of this radiation is believed to be very useful for identifying spent fuels. However, some trials revealed a difficulty due to weakness of the Cerenkov radiation itself. A conventional image intensifier (NVD) could not be applied without turning off the facility lights, e.g. the mercury, halogen and fluorescent lamps on the ceiling and in the pool. Because the NVD spectral sensitivity covers the spectral region of these lamps it cannot detect faint Cerenkov radiation under such a very intense ambient light. To switch off all facility lights would be dangerous and unacceptable for facilities. Another trial used NVD together with an optical filter UG11 to eliminate the influence of ambient light. But it was still difficult to observe the Cerenkov radiation, especially under mercury lamps because the filter could not reduce the ambient light completely. To solve this problem a new device, UV-11, has been developed since 1983. It has spectral sensitivity only in the ultraviolet region where only a negligible contribution comes from ambient light, particularly from mercury lamps. Accordingly, it becomes possible to observe the Cerenkov radiation image without switching off the facility lights.

Figure 1 shows a schematic diagram of the new UV-11 device. A large aperture reflection type lens with  $f/1.55$  opening can collect enough photons to activate the subsequent UV-11, which consists of a Cs-Te UV sensitive photocathode, microchannel plate and phosphor screen.

The optical UV-11 gain is more than 10 000. This device is shown in Fig. 2. Figure 3 shows the relative sensitivity of UV-11 and NVD + UG11. Also, the relative intensities, of Cerenkov radiation, the mercury lamp and the halogen lamp are shown. UV-11 has a reasonable sensitivity only in the ultraviolet region. Accordingly, it appears that this device can observe the UV region of Cerenkov radiation

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\* Government support through the Japanese Science and Technical Agency.

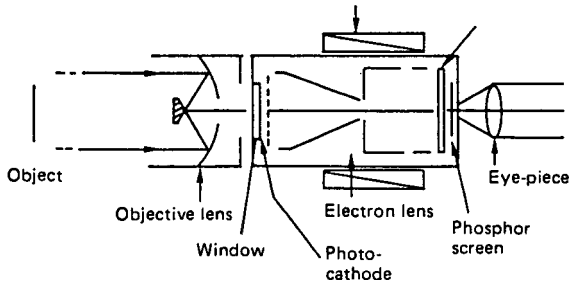


FIG. 1. Schematic diagram of UV-11.

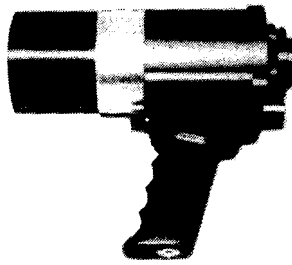


FIG. 2. The UV-11.

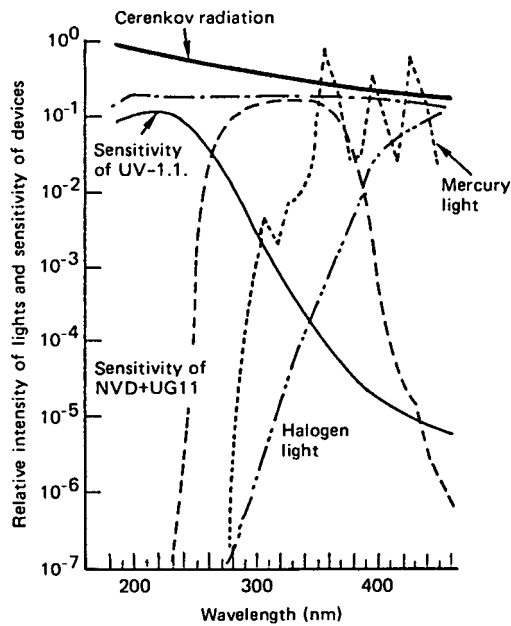


FIG. 3. Spectrum distributions of various lights and sensitivities of the UV-11, and the NVD + UG11.

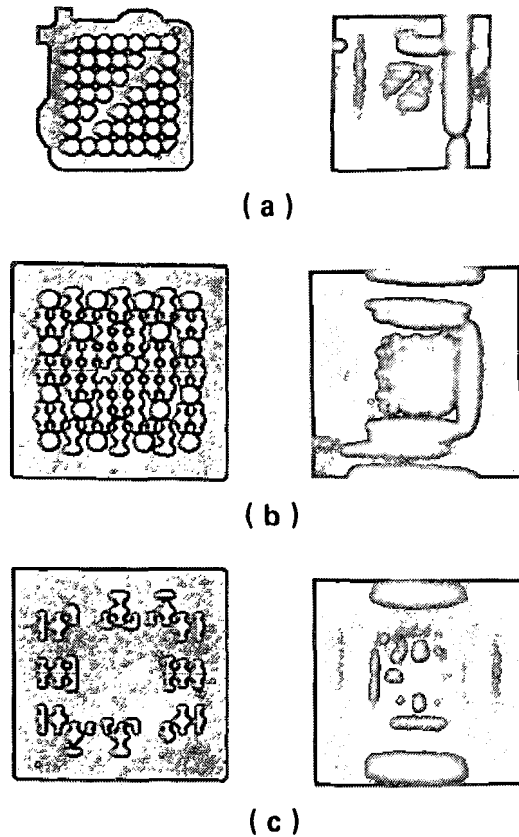


FIG. 4. Experimental result of UV-11.

(a) BWR type  $8 \times 8$  spent fuel;

(b) PWR type  $14 \times 14$  spent fuel without burnable poison;

(c) PWR type  $14 \times 14$  spent fuel with burnable poison.

Pictures taken by using the UV-11 under normal facility lighting conditions.

in the presence of normal facility light. On the other hand, the sensitivity of NVD + UG11 overlaps with the spectra of the mercury and halogen lamps and it becomes difficult to see the Cerenkov radiation in their presence.

Figure 4 shows Cerenkov radiation images taken by this device under full facility illumination. The left of the figure shows the predicted image pattern of Cerenkov radiation at the top of the spent fuel, and the right side shows experimental results. It seems very clear from Fig. 4 that the device is very useful for observing Cerenkov radiation in the presence of normal facility lights.



IAEA-SM-293/91P

# MEASUREMENT OF CERENKOV RADIATION FROM USED CANDU FUEL

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Further advances have been made in the measurement of Cerenkov glow from CANDU fuel stored in water bays for safeguards attribute verification. Previous studies [1] involving the use of a Cerenkov viewing device (CVD) in CANDU fuel bays demonstrated the ability to detect the weak Cerenkov glow from two-year cooled fuel. This work was done in the presence of artificial lighting filtered to remove ultraviolet light. At the Douglas Point Nuclear Generating Station, fuel bundles cooled from one to eleven years were examined. Each bundle was measured for its Cerenkov light intensity in complete darkness using a 1P28 photomultiplier tube and a light collimator. The individual fuel bundles were then measured with two advanced CVDs under ultraviolet filtered mercury vapour lights.

The intensity of Cerenkov light from used fuel bundles was calculated using the computer codes LATREP [2] and CANIGEN [3] and equations derived by Cerenkov [4]. The results are shown in Fig. 1 as a series of curves. The experimental

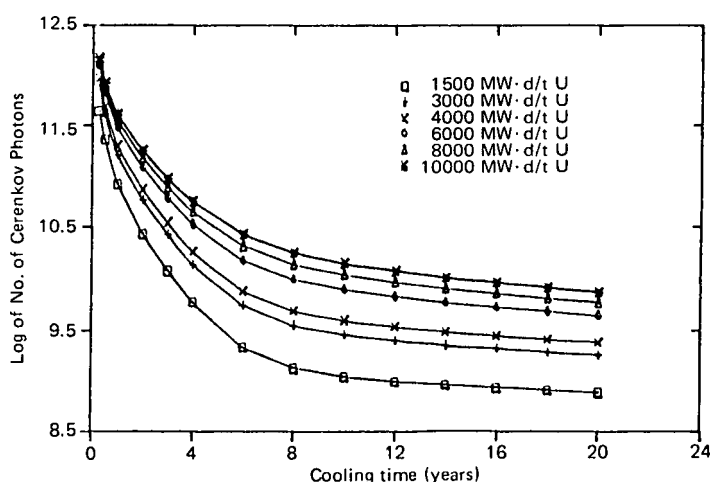


FIG. 1. Cerenkov light intensity from individual bundles as a function of cooling time and burnup.

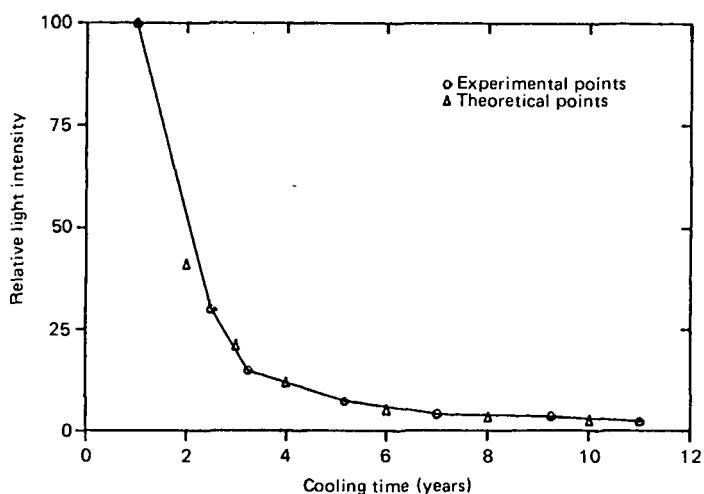


FIG. 2. Plot of Cerenkov light intensity from used Douglas Point fuel bundles as a function of cooling time.



FIG. 3. Quartz lens Cerenkov viewing device.

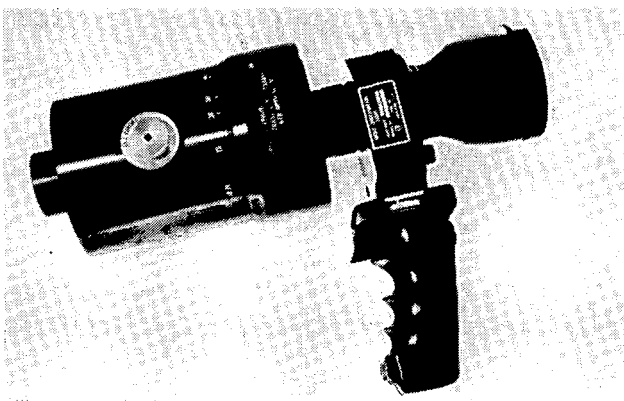


FIG. 4. Mirror lens Cerenkov viewing device.

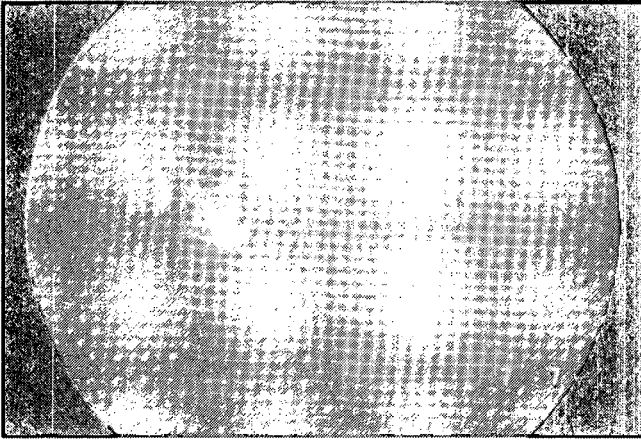


FIG. 5. Photograph of unused fuel bundle using the Cerenkov viewing device.

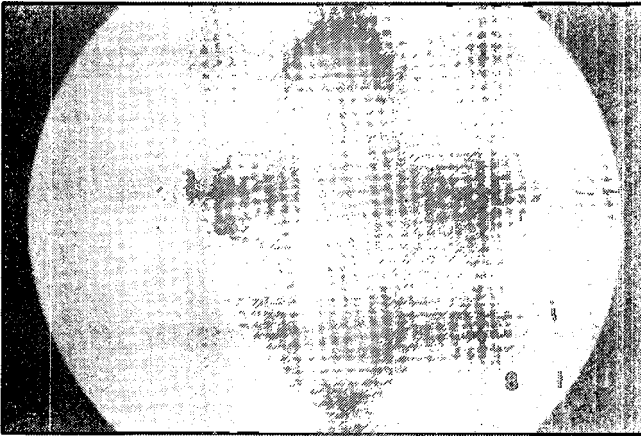


FIG. 6. Photograph of a five-year cooled used fuel bundle using the Cerenkov viewing device.

data obtained from the measurements on Douglas Point fuel bundles cooled from one to eleven years were plotted with the theoretical data for a bundle with similar burnup ( $8000 \text{ MW}\cdot\text{d/t U}$ ). The results are given in Fig. 2 and show excellent agreement.

Two CVDs were used to measure the Cerenkov glow from the Douglas Point fuel bundles (Figs 3 and 4). Both units contain second-generation image intensifiers with approximately the same optical gain. The quantum efficiency of the photocathodes of these units at 300 nm is about 19% compared with 8% for the older CVD

used in previous studies. The CVD with the catadioptric mirror lens, which has a maximum lens opening of  $f/1.4$  (Fig. 4), was found to transmit seven times more light than the quartz lens CVD (Fig. 3), which has a maximum lens opening of only  $f/4.3$ . The two devices were both capable of detecting Cerenkov glow from fuel cooled for five years. Figure 5 shows an unused fuel bundle with no detectable Cerenkov glow. Figure 6 shows a five-year cooled fuel bundle with the characteristic Cerenkov glow. Longer cooled fuel was not detected visually using the CVDs; however, the devices could detect Cerenkov glow from eleven-year cooled fuel by using time exposure photography.

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IAEA-SM-293/7P

## CHAINE COMPLETE D'AUTOMATES POUR L'ANALYSE PAR SPECTROMETRIE DE MASSE DES SOLUTIONS DE COMBUSTIBLES IRRADIES

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## INTRODUCTION

La détermination des concentrations d'uranium et de plutonium dans les solutions de combustibles irradiés est généralement effectuée par dilution isotopique en spectrométrie de masse.

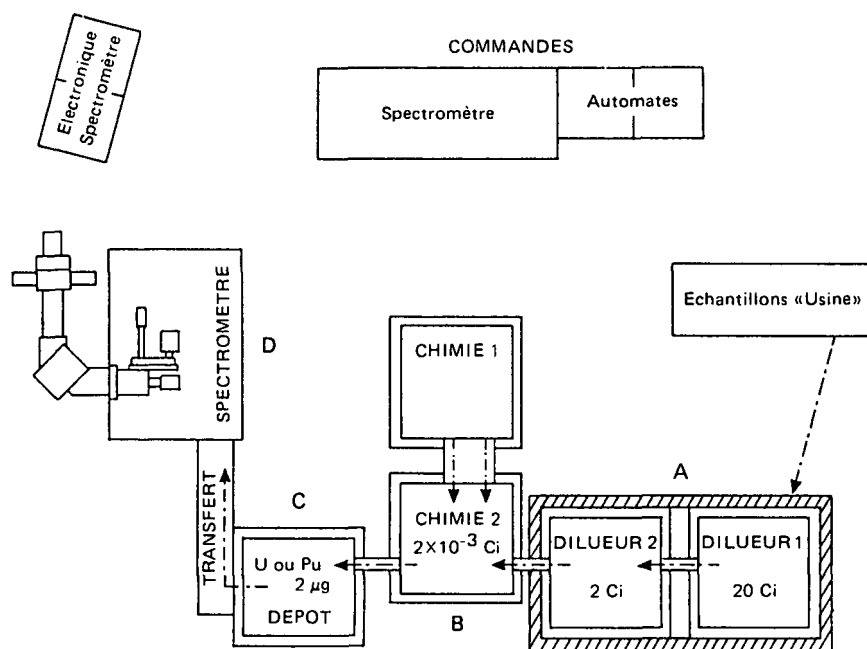


FIG. 1. Schéma de l'installation.

Un traitement chimique préalable permet l'élimination des produits de fission et la séparation des fractions «U» et «Pu» purifiées. La grande activité et la concentration élevée des solutions initiales imposent de procéder auparavant à une dilution d'un facteur connu avec précision.

Pour diminuer l'exposition du personnel aux radiations, ainsi que pour améliorer la reproductibilité des opérations, il a été décidé d'entreprendre l'étude et la construction d'un ensemble d'automates contrôlés par microprocesseurs (fig. 1) capables d'accomplir les fonctions décrites ci-après.

## MODULE A: DILUTION ET AJOUT DE TRACEURS ISOTOPIQUES

Ce module, qui recevra les échantillons de solutions de combustible irradié, fonctionnera en enceinte blindée. Les flacons d'échantillons de haute activité comportent des bouchons de caoutchouc. Tous les prélèvements, les transferts et les ajouts de solutions se font par percement à l'aide d'aiguilles et de seringues. On espère atteindre une reproductibilité de quelques  $10^{-4}$  dans la connaissance du facteur de dilution en opérant par pesées.

## MODULE B: SEPARATION CHIMIQUE

On a ici mécanisé les opérations classiques de séparation sur colonnes de résine échangeuse d'ions. Les activités introduites sont suffisamment faibles pour un fonctionnement en boîte à gants non blindée.

## MODULE C: DEPOTS SUR SUPPORTS D'ECHANTILLONS

Les dépôts sur filament central ou latéral sont effectués goutte à goutte à l'aide d'une seringue originale dont l'aiguille, changée à chaque dépôt, est un tube d'acier inoxydable coupé en place à l'exacte longueur voulue. L'appareil est adapté à l'utilisation des supports de filaments en verre utilisés dans les spectromètres VG ou CAMECA; il pourrait être modifié pour l'utilisation d'autres supports.

## MODULE D: SPECTROMETRE DE MASSE

Un prototype de spectromètre de masse automatique a été fabriqué par la Société CAMECA selon nos spécifications. L'appareil peut introduire, analyser, puis évacuer en continu un nombre quelconque de filaments porteurs d'éléments variés, sous le seul contrôle d'un ordinateur.

## CONTROLES

Les modules A, B, et C sont pilotés par des microprocesseurs MOTOLA 6802; un quatrième microprocesseur de même type règle le fonctionnement simultané de ces modules. L'ensemble sera supervisé par le calculateur DEC PDP 11 du spectromètre de masse.

Tous les mouvements dans les modules sont réalisés par des moteurs à courant continu et des réducteurs PORTESCAP. Les positions sont repérées par des détecteurs électro-optiques.

Pour une maintenance plus facile, chaque moteur est relié au microprocesseur par un seul type de carte d'interface, qui reçoit les informations numériques du calculateur, les convertit en un courant de sens approprié, et renvoie en retour les informations de position.

Les prototypes des éléments A, B, et D sont en cours de test.

L'élément C est en cours de commercialisation. Un exemplaire fonctionne à l'usine Cogéma de La Hague, où il a déjà réalisé plus de 10 000 dépôts.

IAEA-SM-293/20P

# A $^{233}\text{U}/^{235}\text{U}/^{238}\text{U}$ 1/1/1 ISOTOPIC REFERENCE MATERIAL

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An isotopic reference material with isotope ratios  $^{233}\text{U}/^{235}\text{U}/^{238}\text{U} = 1/1/1$  has been prepared from highly enriched isotopes with abundances  $^{233}\text{U}/\text{U} = 0.9947$  -  $^{235}\text{U}/\text{U} = 0.9994$  -  $^{238}\text{U}/\text{U} = 0.99997$ . It is intended to be used for calibrating isotope dilution measurements and verifying the dependence of mass discrimination upon mass.

The starting materials were purified by ion exchange and peroxide precipitation. In the first step, an anion exchange resin Dowex 1  $\times$  4 (100–200 mesh;  $\text{H}^+$  form) in 7M nitric acid was used to eliminate transuranium elements and most cations. In the second step a cation exchange resin Dowex 50  $\times$  8 (100–200 mesh; acid form) in a medium of 90% tetrahydrofuran — 10% 6M nitric acid was employed. The ion exchange purification was followed by a peroxide precipitation with  $\text{H}_2\text{O}_2$  at  $\text{pH} = 2$ .

The enriched isotope oxides were calcined simultaneously in three quartz tubes for 16 h at  $900^\circ\text{C}$  in a furnace with a temperature uniformity of  $\pm 2.5^\circ\text{C}$ . The calcination and weighing operations were performed in a glove box with a controlled humidity.

An extensive evaluation of the total uncertainty of the isotope ratios in the prepared mixture was performed, identifying correctly the different uncertainty contributions and their propagation.

The certified results are:

$$^{233}\text{U}/^{238}\text{U} = 1.00001 \pm 0.00030$$

$$^{234}\text{U}/^{238}\text{U} = 0.00205 \pm 0.00001$$

$$^{235}\text{U}/^{238}\text{U} = 1.00015 \pm 0.00020$$

$$^{236}\text{U}/^{238}\text{U} = 0.00025 \pm 0.00001$$

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\* Visiting scientist from Western Australian Institute of Technology, Bentley, Australia.

This corresponds to the following isotopic composition:

	Isotopic atom %	Isotopic mass %	Uncertainties
$^{233}\text{U}/\text{U}$	33.3064	32.9756	$\pm 0.0059$
$^{234}\text{U}/\text{U}$	0.0683	0.0679	$\pm 0.0003$
$^{235}\text{U}/\text{U}$	33.3110	33.2639	$\pm 0.0040$
$^{236}\text{U}/\text{U}$	0.0082	0.0082	$\pm 0.0003$
$^{238}\text{U}/\text{U}$	33.3061	33.6844	$\pm 0.0049$

It is intended to transform this reference material into a certified European Community Nuclear Reference Material, the number of which will be EC-NRM 199.

IAEA-SM-293/148P

## MEASUREMENTS OF URANIUM ISOTOPIC RATIOS BY TRANSPORTABLE QUADRUPOLE MASS SPECTROMETERS

*Three years of experience*

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The exact determination of the enrichment factor in the different phases of the nuclear material cycle is an essential parameter in safeguards. Taking samples and sending them to the analytical laboratories of Direction du contrôle de sécurité (DCS) suffers two major drawbacks – transportation problems and delay of results from the laboratories concerned.

To avoid these drawbacks, DCS is using transportable quadrupole mass spectrometry for in-field control of fissile materials with the required accuracy.

The results obtained during three years on  $\text{UF}_6$  isotopic ratio measurements allow us to confirm the reliability of the technique, even under the severe conditions to which the instrument is exposed.



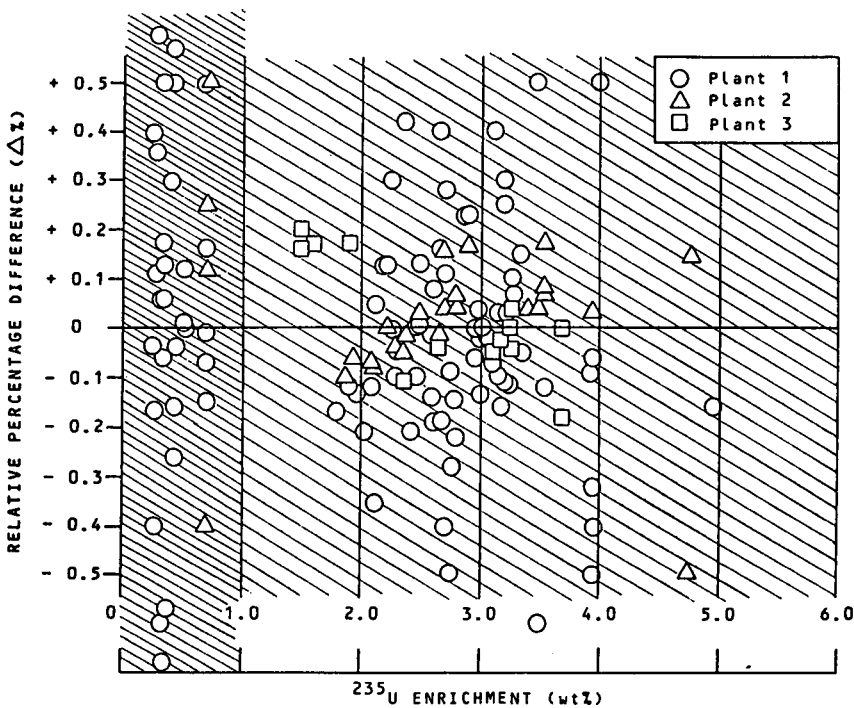


FIG. 1. Typical differences between declared and measured values in three different enrichment installations over the last three years.

In  $\text{UF}_6$  enrichment measurements Fig. 1 shows typical differences between plant declarations and transportable mass spectrometry measurements in three different installations during recent years. It can be concluded that, for the enrichments of 'product materials', a discrepancy of 0.5% is already significant. For enrichments lower than 1%, this discrepancy increases, probably due to the inhomogeneity of the depleted material.

To avoid transportation of the  $\text{UF}_6$  reference material used during analyses, a limited amount is permanently stored in the plants. For the product materials measurements, differences of 100% between the reference enrichment values and the sample values affect the accuracy of the results within a factor of 0.2% — these values are still acceptable for safeguards purposes.

The estimated analysis costs with a transportable mass spectrometer have remarkably confirmed our estimates. For about 60 samples a year the analysis costs are equivalent to those performed in a laboratory.

Contamination problems remained within the expected limits. No contamination occurred which could not be eliminated by normal maintenance. Two

modifications are planned in order to minimize the volume of the machine and the sample consumption: one is the flow controller which would replace the actual expansion tanks, giving flow stability in the source; the other is the miniaturization of the computer.

Positive experience with the gas ( $\text{UF}_6$ ) quadrupole is being continued with the in situ utilization of a thermo-ionic quadrupole used for measuring solid U and Pu samples. Recent provisional results show an accuracy of better than 0.5%.

For the two machines at present running, transportation necessitates a small truck with an elevator system. The  $\text{UF}_6$  instrument require half a day for installation and one night to reach working conditions ( $10^{-7}$ – $10^{-8}$  torr)<sup>1</sup>. The thermo-ionic one can be operated within a few hours. The installation of both instruments could easily be done by one technician.

IAEA-SM-293/40P

#### DEVELOPMENT OF AUTOMATED ANALYTICAL SYSTEM FOR INPUT SOLUTION SAMPLES BY THE ISOTOPE DILUTION METHOD

*System design and component test*

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An automated analytical system for input solution samples has been designed to increase analytical capability as well as to improve timeliness of measurement at a future large reprocessing plant. A component of the system was prepared and tested.

Analytical procedure of the system is based on isotope dilution mass spectrometry coupled with anion exchange separation. After receiving six vials containing input solution samples at the pneumatic tube terminal station in a hot cell the system begins operation. Six small amounts of the samples are first taken on a weight basis and diluted with nitric acid. Two aliquots of each diluted sample are transferred into two

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<sup>1</sup> 1 torr = 133.3 Pa.

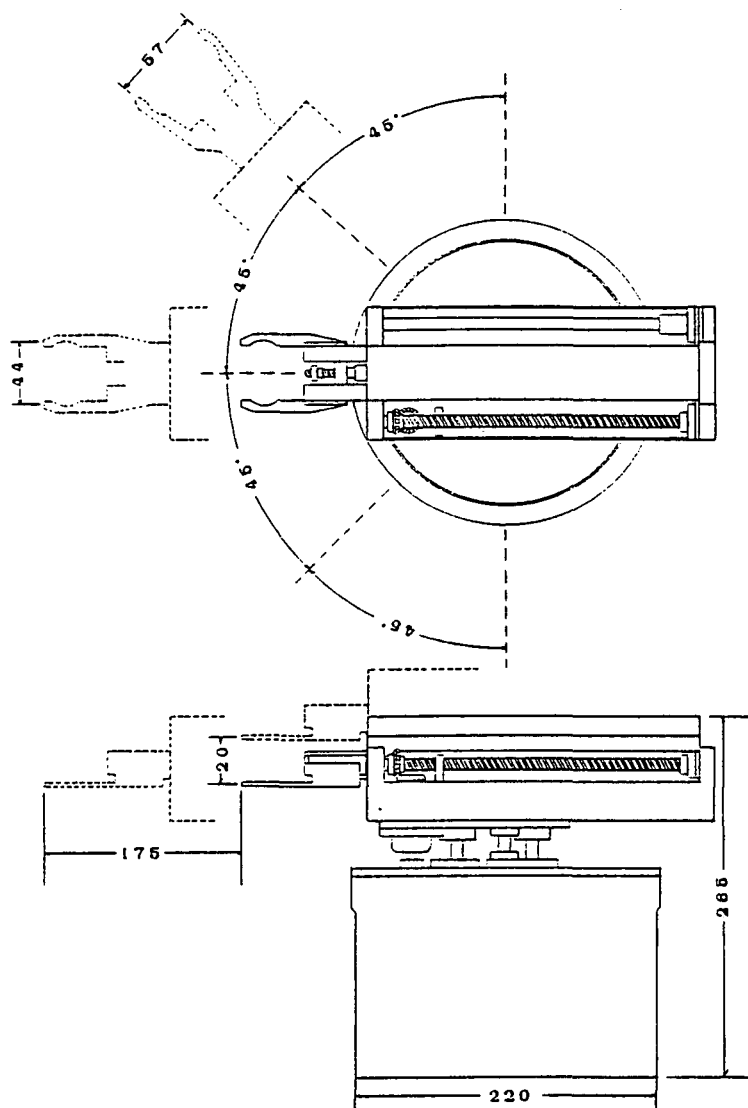


FIG. 1. Two-fingered hand robot (units: mm).

small beakers, one of which contains mixed Pu-U isotope. Then the Pu valency in the solution is adjusted to the tetravalent state by redox procedure, and the uranium and plutonium are separated from each other by the anion exchange technique. A small volume of each fraction separated is automatically evaporated to dryness on a filament by using a loading device, and the magazine with filaments is manually installed in a mass spectrometer ion source housing. In addition, another small fraction of plutonium is deposited on a stainless steel dish to prepare a sample for alpha spectrometry of plutonium. Concentration and isotopic composition of each element in the sample are obtained by a calculation based on change of isotopic ratios and density of sample solution.

The sample preparation system, which basically consists of hand robots with two fingers or a syringe, and turntables, can prepare 1200 samples a year (200 working days). Taking into account the difficulties of maintenance by remote manipulation, each system component has been designed to have a simple and modular structure. Much attention is also paid to the standardization of parts used, and the radiation and corrosion resistance of materials. Throughout the whole process, samples are identified by reading bar-codes on the walls of beakers, and sample delivery is confirmed by measuring weight change at some key points. The sample preparation process of the system can be divided into five operation units, each of which is run by a programmable controller. A host computer controls not only mechanical and electrical operation through five programmable controllers, but also collects and processes data from the instruments and prints out analytical results.

Based on the system design described, a two-fingered hand robot shown in Fig. 1, which is one of the most important component devices of the system, has been prepared and tested for basic motions such as vertical, expansion and contraction, swing and gripping movements. As a result, it has been found that each movement stops at the specified positions within a precision of  $\pm 2$  mm.

**SAFEGUARDS FOR  
REACTORS OR  
LONG TERM STORAGE FACILITIES  
(Session 10)**

**Chairman**

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Sweden



## A MODIFIED INSPECTION PROCEDURE FOR LIGHT WATER REACTOR SAFEGUARDS

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### Abstract

#### A MODIFIED INSPECTION PROCEDURE FOR LIGHT WATER REACTOR SAFEGUARDS.

Safeguards for light water reactors are based on annual inventory verifications, supplemented by surveillance cameras to verify that there have been no unreported changes in the inventory. Interim inspections at three-month intervals are primarily to recover camera records. A modification of the surveillance equipment and procedures has been suggested whereby surveillance records could be sent by the operator to the IAEA at scheduled times. If the records indicated no spent-fuel shipments, the three-month interval between inspections could be extended. Tamper resistance would be an important requirement; design approaches are described.

### THE CONCEPT

The present International Atomic Energy Agency (IAEA) approach for safeguarding Light Water Reactors (LWR) has developed over many years and hundreds of inspections, and the problems of safeguarding other, more sensitive, types of facilities have drawn more attention in recent years. However, while LWRs demand only about twenty percent of the Agency's inspection resources, that portion is expected to grow as new reactors come on line. In any case, it would be desirable to allocate a smaller fraction to LWRs in order to make more inspection effort available for facilities that are of greater concern [1].

Safeguards strategies for various types of facilities are based on quantitative detection goals, which are a function of the material and its form. Nuclear material in LWRs is in the form of unirradiated U-235 in fresh fuel and plutonium in the spent fuel and reactor core, for which the defined significant quantities are 75 kg and 8 kg respectively. (Safeguards for LWRs using mixed oxide fuel are not considered in this paper). The detection goal is based on the times estimated to convert those materials to forms directly usable in an explosive, which are specified as on the order of one year for fresh fuel and one to three months for spent fuel. The LWR strategy is

intended to detect a potential diversion at those threshold levels. The general approach is:

1. Item accountancy of individual fuel assemblies:
  - a. periodic physical-inventory verification
  - b. containment and surveillance to indicate any flows while the inspector is not present.
2. Measures to assure the integrity of fuel assemblies and to detect substitutions.

The purpose of this paper is to examine ways by which that approach could be implemented with further use of instrumentation, thereby reducing inspection effort.

An improved inspection procedure is sought for the purpose of saving inspector resources, not to achieve better timeliness or more complete verification, which we assume to be adequate under the present approach. Only trivial gains could be made if some, but not all, inspector activities were eliminated. For any substantial saving we must eliminate some of the three-month interim inspections without compromising the timeliness goal. It is necessary that we justify the elimination of any of the present interim inspection activities that require the on-site presence of the inspector, and second, that we find some way of performing the remaining essential operations without inspector presence.

If the fuel assemblies are accounted for as discrete, integral items, it is not necessary as part of the reactor safeguards to measure the nuclear material content. Therefore, LWR safeguards consist of counting, identifying, and to some degree verifying the integrity of the fresh fuel, at least annually and the spent fuel at least every three months. The irradiated core, which also contains plutonium, cannot be directly verified except at refueling, when the reactor is opened. That is done nominally at 12 to 18 month intervals, for different reactors, and it may be longer. At those times the core is verified by physical-inventory taking. Other inspections are made at three-month intervals, with the fresh fuel inventoried at each fourth inspection, in order to meet the respective timeliness goals for the plutonium and the unirradiated uranium.

It is necessary to make a distinction between interim inspection activities required to meet the inspection goal and those that are performed at the same time for other reasons. Presently, an inspector visit is necessary every three months to reload film cameras. If the inspector is there anyway, physical-inventory verification, at least to the extent of item counting, can be done without adding substantially to the inspector man-day total.

The question arises, what information is essential to the three-month timeliness goal, assuming that the information need



not be made available for any other reason? We start with the premise that the timeliness goal is met if certain, but not necessarily all, of the present verification functions are carried out every three months. Following the reasoning of the current approach we conclude that the physical-inventory verification during interim inspections is not essential, in the absence of any surveillance information to the contrary. Further, we assume that an interim inspection need not be made for the purpose of auditing the operator's records. The State/operator is required to report any inventory changes, and so the Agency can keep its own book inventory between annual inspections. In any case, a principal reason for examining the records at the interim inspection is to bring the inspector up to date on the current inventory, in preparation for the physical-inventory verification.

It is apparently not necessary that everything be verified, since the spent fuel in the core is not verified more often than at refueling, every 12 to 18 months. We assume that surveillance information that assures that there were no inventory changes is sufficient justification for omitting the three-month inspection. It follows that what is necessary if an inspection is to be omitted is the acquisition of surveillance information adequate to assure that there were no spent-fuel shipments in or out of the facility since the last physical-inventory taking. We make one final judgment: that a spent-fuel movement in or out of the facility is presumed to require a cask, or a large cask-like object.

A modified system to reduce inspection effort would include the following functions:

1. Surveillance means that can provide adequate assurance that a cask-like object at any exit point would be detected.
2. Means by which the surveillance information can be acquired by the Agency without an inspector visit, early enough to make an inspection at the reactor within three months of the indicated cask presence.

Surveillance cameras, whether film or TV, are a means presently available for cask detection. The problem is that the information must be collected on-site by the inspector. An alternative would be to equip the surveillance TV with a secure, detachable recorder and arrange for the State or facility operator to send it to the Agency on a regular schedule. The facility operator would replace the recorder with another, previously furnished by the Agency. The schedule would be arranged so that the information would reach the Agency in time for review and any follow-up inspection actions in accordance with the timeliness goal.

Characteristics of the system that we consider to be essential are that it must be fail-safe; that is, any failure,

as well as any indication of tampering, would lead to a timely on-site inspection; and second, that no circumstances, including tampering or failures of any sort, would lead to inspections more frequently than every three months, as with present interim inspections.

How much inspection effort might be saved by the suggested procedure depends on how often spent fuel might be shipped from the storage pool at the reactor. On the basis of published information and information from private sources we estimate that in 1988 more than 75 percent of all interim inspections at LWRs, more than 250 inspections, could be avoided. The estimate covers all safeguarded LWRs in non-nuclear-weapon States outside of CMEA countries. Of the 17 States, 10 plan no shipments. The remaining 7 countries ship some fuel, but infrequently enough that there would be no shipment during most inspection intervals.

#### SYSTEM DESCRIPTION

Consider a system based on detachable recorders, as shown in Figure 1. The TV camera(s) would be positioned to provide clear imagery of the zone(s) through which spent-fuel casks could enter or exit the facility. Frame intervals would have to be set short enough to ensure that a cask could not pass

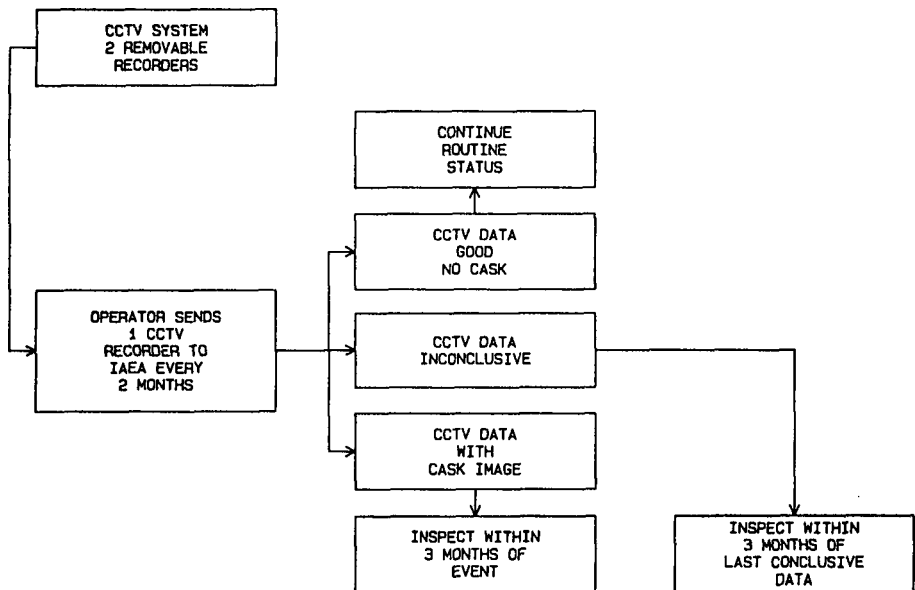


FIG. 1. Functional block diagram of suggested system. CCTV = closed circuit television.

completely through the zone(s) between successive frames. The TV system would be equipped with two recorders, with the camera output recorded in each recorder in parallel. The recorders would be designed so that either could be removed without affecting the operation of the other. Each recorder would have a recording capacity in excess of four months. The entire system, including the separated recorders, the open interface, and any associated signal lines would be designed such that any indications of tampering would appear on the TV record or a data storage medium in the recorders.

On scheduled dates, once every two months, the operator would remove one of the recorders and send it to the Agency, replacing it with another previously supplied. Allowing an estimated 15 days for transit and for review by the Agency, in the extreme case 15 days would remain within the three-month timeliness-goal limitation for the Agency to make an inspection at the reactor. An inspection would be made if (a) the recorder were not received within the 15-day limit, (b) there were an indication that a cask, or cask-like object, was at any time present in the facility entry/exit area, (c) an indication of loss of surveillance coverage, such that a cask might not have been detected, including equipment failure or inadequate lighting, and (d) any indication of tampering. The inspection would be scheduled for some date within three months of the date of the anomalous event or loss of coverage.

Thus, following an annual inspection which would be the same as now, interim inspections could be deferred until three months after the date when there was no longer assurance that the spent-fuel inventory had not changed. The consequence of any equipment failure, false detection, or tampering, would be that the next interim inspection could not be avoided. The expected frequency of interim inspections, especially at reactors with little spent-fuel shipping activity, should be substantially reduced.

The system would have to be fail-safe in order to provide positive assurance during periods when no casks were detected. The price for being fail-safe is an inspection each time there is a malfunction, tampering, or a false or ambiguous detection. If there is at least one such occurrence in each three-month period, or an actual cask presence, there will be no saving of inspection effort, and hence nothing to be gained. It is therefore very important that false alarms and malfunctions be held to the minimum possible, and that cask detections be conclusive.

#### SYSTEM DESIGN CONSIDERATIONS

The design of the video system described in this paper is in a preliminary conceptual stage. The benefits that may be

derived from the suggested procedure are highly dependent on the effectiveness of an operator-interfaced video system and its tamper indicating features.

Conceptually, two Video Recorder Modules are installed in a System Console which contains a Master Controller; the System Video Camera Module with its internal video authentication circuit is connected to the System Console. The Recorder Module (RM) contains an 8-mm video recorder, its internal control circuitry, and a small battery for power backup and circuit operation. The battery is charged by the System Console when installed therein, or kept in a special charger during storage. The video system with two RMs is installed at a facility and a third RM is left at the facility. This means that at all times the system must have a high degree of tamper indication: (1) when both RMs are installed, and (2) when one RM has been removed, exposing the interface between the RM and the System Console. In addition, the bare RM must also have high tamper indicating features while at the facility and during the transportation to and from the IAEA. Existing technology offers the potential of accomplishing this objective.

The RM housing is made of a material that provides evidence of any attempt to penetrate its exterior surfaces. The material used for this housing is also sufficiently rugged to survive the expected handling environments. The RM housing is designed to accommodate an IAEA seal on the normal recorder access point which must be removed before the housing lid can be opened. Opening the lid is detected by a lid switch and a special light sensing-circuit; failure to deactivate the light sensor or the lid switch is recorded by circuitry in the RM. Only IAEA personnel will have the proper code to open the lid and remove the video tape without deactivating the detectors. The RM has no external electrical connector. All signals to and from the RM are transmitted over optical links. The RM contains a small battery for operation of its electronics which is charged through a special transformer designed into the rear of the housing. The housing is protected against the introduction of moisture, has a humidity sensor to register exposure to excessive moisture, and contains a video authentication circuit for direct communication with the Camera Module (CM) through the System Console. All authentication of the video takes place inside the RM. Since the Video System requires two recorders, each RM is capable of independently communicating with the CM to establish authentication of the video it is recording.

All data and control signals passing into the RM are encrypted. Reverse communication from the RM to the master controller is sent in the clear - no encryption.

There will be a need to protect the video data during its shipment from the facility to the point of review. This is accomplished by providing the RM with a unique code stored in a

volatile memory. Opening the RM any time will destroy the code. As long as the code is present, it is periodically recorded on the video during the shipment to the point of review. Time and date information are also recorded in the video during shipment. Examination of this information will verify that the video data has not been tampered with during shipment.

Each RM has a unique internal and external identification, with an external bar code which is read when it is inserted into the System Console. The bar code is then transmitted to the RM by the encrypted data link where it is compared with the internal code. If the codes agree, then the recorder can be accessed to start recording the video information.

The operation of the video system recordings starts with the installation of the RM in the System Console. The Master Controller will read the bar code and transmit it to the RM after it is in place. If all indications are proper, then the Master Controller will activate an electric lock to secure the RM in place.

The recordings now take place until it is time to remove the RM. At that time, a facility operator inserts a request key, after which the Master Controller initiates a final recording sequence on the recorder. After the final recording has taken place, the electric lock is opened and the facility operator informed that the RM can be removed. The replacement RM can then be installed into the System Console and is automatically activated, as described above.

## SUMMARY

The suggested modified inspection procedure appears to warrant further development. It follows the general inspection strategy now used by the Agency. It offers the prospect of a substantial reduction in the number of interim inspections, while providing for inspections consistent with timeliness goals whenever a possible change in inventory is indicated. As a step toward implementation, a hardware design concept is envisioned that provides the essential tamper indication by means of special containment, encryption, and recorded information.

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## **ИСПЫТАНИЕ И ДЕМОНСТРАЦИЯ ТЕХНИЧЕСКИХ СРЕДСТВ ГАРАНТИЙ НА СОВЕТСКИХ ЯДЕРНЫХ УСТАНОВКАХ**

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### **Abstract—Аннотация**

#### **TESTING AND DEMONSTRATION OF SAFEGUARDS EQUIPMENT AT USSR NUCLEAR FACILITIES.**

As part of its programme of scientific and technical support for IAEA safeguards, the USSR has since 1979 regularly organized courses for IAEA inspectors, who are given the opportunity to acquaint themselves with nuclear power plant design and to gain personal experience of carrying out the main inspection procedures on specially selected fuel assemblies. Particular attention is paid to acquiring practical skills in the use of non-destructive analysis equipment and methods on the fuel contained in reactor fuel assemblies. These courses have been held for some years at a WWER-440 reactor, but in September 1985 the first courses at a fast reactor were held on the BOR-60. The paper examines the experience with the use and

testing of the non-destructive fuel analysis methods and equipment usually employed in IAEA inspections gained during the courses for inspectors at the WWER-440 reactor and, in particular, at the BOR-60 fast reactor.

#### ИСПЫТАНИЕ И ДЕМОНСТРАЦИЯ ТЕХНИЧЕСКИХ СРЕДСТВ ГАРАНТИЙ НА СОВЕТСКИХ ЯДЕРНЫХ УСТАНОВКАХ.

В рамках Программы СССР научно-технической поддержки гарантий МАГАТЭ, начиная с 1979 года в Советском Союзе регулярно организуются курсы инспекторов МАГАТЭ, во время которых слушателям представляется возможность знакомиться с конструкцией ядерных энергетических установок и практически участвовать в проведении основных инспекционных процедур на выделенной для этой цели совокупности ТВС. Особое внимание при этом уделяется приобретению практических навыков в использовании методов и аппаратуры неразрушающего контроля топлива в ТВС реактора. Такие курсы проводились в течение нескольких лет на реакторе ВВЭР-440, а в сентябре 1985 года они были впервые организованы на установке с реактором на быстрых нейтронах BOR-60. В докладе рассмотрен опыт использования и испытания аппаратуры и методов неразрушающего контроля топлива, обычно применяемых в инспекционной деятельности МАГАТЭ, в процессе проведения курсов для инспекторов на реакторе ВВЭР-440 и, особенно, для установки с реактором на быстрых нейтронах BOR-60.

Программа научно-технической поддержки МАГАТЭ охватывает ряд проблем, решение которых позволит существенно повысить эффективность гарантий. Важнейшее значение в рамках этой Программы придается вопросу подготовки квалифицированных кадров для работы в инспекционной деятельности МАГАТЭ. До 1985 года вклад СССР в подготовку инспекционных кадров МАГАТЭ состоял в организации и проведении учебных курсов на базе водо-водяных реакторов (ВВЭР-440, ВВЭР-1000, ИР-8), а также опытной установки по переработке отработавшего топлива [1,2]. Учебные курсы предназначались для начинающих инспекторов и для персонала национальных систем учета и контроля ядерных материалов.

В 1985 году состоялись курсы повышенной сложности по применению гарантий к АЭС с реакторами на быстрых нейтронах на базе BOR-60 с ознакомительной поездкой на Белоярскую АЭС. В дальнейшем планируется регулярное проведение таких курсов.

Программа курсов охватывает практически все виды инспекционной деятельности на реакторах при проведении обычной инспекции. Особое внимание уделяется вопросу приобретения инспекторами навыков выполнения практических измерений количества ядерных материалов с помощью неразрушающих методов контроля. Это один из важнейших элементов инспекции, поскольку только независимые от оператора количественные измерения могут с требуемой достоверностью обеспечить уверенность в отсутствии переключения значимых количеств ядерных материалов.

В развитии методов неразрушающего контроля топлива и соответствующей электронной аппаратуры в последнее десятилетие достигнут столь существенный прогресс, что создание эффективной системы гарантий для большинства установок, на которых используется, перерабатывается или хранится специальный ядерный материал, стало реальной возможностью.



Решены, однако, еще не все проблемы. Применение в ходе развития атомной энергетики необычных топливных композиций приводит, и, по-видимому, еще некоторое время будет приводить к получению ненадежных, а иногда и просто неверных результатов измерений.

Прогресс в развитии методов и аппаратуры неразрушающего контроля ядерных материалов стимулировался в немалой степени потребностью атомной промышленности в средствах технологического контроля. В связи с этим уделялось недостаточно внимания таким специфическим для системы гарантий проблемам, как измерения количества ядерных материалов в условиях возможной фальсификации количественных характеристик и топливной композиции, или просто в условиях недостаточных априорных знаний.

Понимание этого обстоятельства необходимо при организации учебных курсов, особенно — повышенной сложности, и, конечно, при планировании новых методических и аппаратурных разработок для применения в системе гарантий МАГАТЭ.

Учебные курсы инспекторов МАГАТЭ в СССР проводились с учетом стройной и хорошо организованной системы подготовки кадров, принятой в МАГАТЭ; при этом принимались меры к созданию условий, максимально приближенных к осуществлению инспекции на реальной установке. Для этого выделялась намеренно ограниченная совокупность ТВС, содержащихся в зоне баланса материалов установки, на базе которой организовывались курсы, при этом существенно упрощались учет ядерных материалов и проверка отчетных документов, что уместно для начальных курсов.

Для практического применения неразрушающих методов оборудовались участки неразрушающего контроля ядерных материалов, на которых выполнялись следующие упражнения:

- измерение длины активной части ТВС с помощью прибора НМ-4, регистрирующего собственное гамма-излучение топлива;
- измерение обогащения урана в различных типах ТВС сцинтилляционным детектором и многоканальным анализатором РМСА;
- измерение количества ядерных материалов в ТВС пассивным детектором нейтронов (HLNC) и счетчиком нейтронных совпадений НЕС-100;
- измерение количества ядерных материалов в ТВС активным колодезным детектором (UNCL) со счетчиком нейтронных совпадений НЕС-100;
- измерение количества ядерных материалов в отработавших ТВС и измерение изотопного состава конечной топливной композиции с помощью полупроводникового детектора гамма-излучения высокого разрешения из сверх-чистого германия и многоканального анализатора импульсов SILENA CICERO 8K или портативного многоканального анализатора (РМСА).

Эта совокупность измерений применялась в течение нескольких лет на курсах для начинающих инспекторов, проводимых на базе ВВЭР-440, что свидетельствует об определенной стабилизации комплекта используемых приборов и методик. За этот период произошла замена прибора SAM-II на анализатор РМСА, работающий в комплекте со стабилизированным сцинтилляционным детектором, при измере-

ниях обогащения уранового топлива. Микрокалькуляторы HP-97, работавшие ранее в комплекте со счетчиком нейтронных совпадений НЕС-100, были заменены на более эффективный и удобный микрокомпьютер EPSON (HX-20). Многоканальные анализаторы SILENA 1K и 4K заменены на более мощный анализатор SILENA CICERO 8K, оснащенный встроенным микропроцессором, обеспечивающим анализ результатов измерений [3].

Опыт проведения практических занятий на курсах показал, что перечисленная аппаратура в сочетании с микрокомпьютером HX-20 обеспечивает возможность достаточно достоверной проверки наличия ядерных материалов на реакторной установке. В условиях реактора ВВЭР-440 эта аппаратура позволяла оперативно получать надежные данные по глубине выгорания, распределению по длине ТВС и относительному содержанию урана-235, которые были подтверждены результатами разрушающих измерений, выполненных на некоторых ТВС в Радиовом институте им. В.Г. Хлопина совместно с сотрудниками НВ АЭС.

Можно отметить также достаточно высокую надежность аппаратуры неразрушающего контроля топлива. Отказы наблюдались только у анализатора SILENA CICERO. Этот прибор имеет и еще один недостаток — громоздкие размеры и вес. Поэтому желательна замена его на более портативный анализатор РМСА при условии, что последний будет иметь не менее совершенный усилительный тракт и не менее мощное программное обеспечение.

К достоинствам аппаратуры, используемой в настоящее время для инспекций и в процессе обучения, несомненно относится ее насыщенность микропроцессорной техникой и обеспеченность стандартными интерфейсами для соединения с внешними компьютерами и периферийными устройствами. Это позволяет надеяться на ее достаточную стабильность (долговременное использование однотипных приборов) при одновременной возможности постоянного совершенствования за счет улучшения программного обеспечения.

АЭС с реакторами на быстрых нейтронах и исследовательские реакторы такого типа представляют собой особую проблему с точки зрения применения гарантий МАГАТЭ. Не имеется еще достаточного опыта применения гарантий к таким установкам, а опыт накопленный на установках с реакторами на тепловых нейтронах зачастую непригоден из-за ряда существенных особенностей быстрых реакторов.

К этим особенностям, в первую очередь, относятся:

- практическая недоступность для непосредственного измерения ядерных материалов в ТВС активной зоны и воспроизводящих экранов, за исключением момента сразу после доставки свежих ТВС на установку;
- использование топлива с высоким обогащением по урану-235 с высокой долей плутония, т.е. топлива, представляющего собой высокую стратегическую ценность;
- высокая интенсивность процессов преобразования ядерных материалов, обуславливающая накопление вторичного горючего, имеющего также достаточную стратегическую ценность;
- жесткий спектр нейтронов в активной зоне, позволяющий применять различные способы маскировки изъятия ядерных материалов.

Процессы интенсивного накопления вторичного горючего создают также определенные проблемы при ведении учетной документации и при подготовке отчетов в стандартных форматах МАГАТЭ.

Курсы инструкторов МАГАТЭ по применению гарантий на АЭС с реакторами на быстрых нейтронах на базе БОР-60 были первым мероприятием такого рода в Советском Союзе.

При подготовке к практическим занятиям и их проведении был выявлен ряд проблем, обусловленных конструктивными особенностями ТВС активной зоны реактора БОР-60. К таким особенностям относятся:

- гомогенная топливная композиция активной части ТВС, состоящая из смеси диоксидов плутония и урана высокого обогащения (от 36 до 75%);
- верхний и нижний торцевые экраны в ТВС активной зоны изготовлены из спеченных таблеток диоксида естественного урана, что обуславливает возможность проникновения небольших количеств мелкой фракции топлива активной зоны в пространство между внутренней поверхностью оболочки твэла и таблетками торцевых экранов;
- В ТВС активной зоны (тип "О") использовано оборотное топливо с довольно высоким уровнем собственной радиоактивности.

По-видимому, разработчики методик измерения и программного обеспечения использованной аппаратуры с подобными свойствами топлива ранее не встречались.

Приборы, предназначенные для использования в практических занятиях, были доставлены заблаговременно, что позволило тщательно подготовить рабочие места для измерений и выполнить ряд контрольных экспериментов. По материалам этих экспериментов и по практическим занятиям можно сделать следующие замечания:

Измерение длины активной части ТВС выполнялось прибором НМ-4, обычно используемым в инспекционной деятельности МАГАТЭ. При измерении ТВС типа "А" со спеченным урановым двуокисным топливом (90% обогащение) случайная погрешность не превышала 2 мм на длине активной зоны, что составляет менее 0,5% и является вполне удовлетворительным результатом.

Для ТВС со смешанным уран-плутониевым топливом ("тип О") потребовалось установить биологическую защиту из свинца и борированного полиэтилена, чтобы снизить дозу облучения на рабочем месте до допустимого уровня, и изготовить дополнительный коллиматор для прибора НМ-4.

Но в этих условиях длина активной зоны была определена с погрешностью 20—30 мм. Систематическая погрешность в сторону увеличения длины активной зоны возникла из-за упомянутого ранее проникновения мелких фракций топлива активной зоны в пристеночное пространство экранов. В этом случае формальное применение принятой процедуры измерений приводит к завышению результатов.

Измерение обогащения урана в ТВС типа "А" и ТВС зоны воспроизводства выполнялось с помощью портативного многоканального анализатора РМСА со сцинтилляционным гамма-детектором, а для градуировки прибора использовались ТВС с обогащениями 23%, 36% и 90%.

Эти измерения дают вполне удовлетворительные результаты, если обеспечена идентичность геометрии эксперимента.

Изотопный состав топлива в ТВС типа "О", контролировался с помощью детектора высокого разрешения из сверхчистого германия и многоканального анализатора SILENA CICERO 8K.

Работоспособность прибора проверена с помощью набора стандартных образцов. Определялись отклонения измеренного процентного содержания изотопов плутония от паспортного значения. Стандартные образцы представляли собой контейнеры диаметром 44 мм, заполненные диоксидом плутония и смесью диоксидов плутония и смесью диоксидов урана и плутония. Использовалось топливо двух типов (1-го и 2-го) с различающимся изотопным составом и содержанием плутония-239 в диапазоне 63–75%.

Для основных изотопов плутония получены удовлетворительные результаты при временах измерения, не превышающих 30 мин.

Измерения изотопного состава в ТВС со смешанным уран-плутониевым топливом, содержащим уран высокого обогащения, не были удачными. Программа, встроенная в анализатор SILENA CICERO 8K, не предусматривала разделения комплекса интерферирующих линий плутония-239 (203,5 кэВ), плутония-241 (208 кэВ) и урана-235 (205,3 кэВ и 202,1 кэВ). Этот недостаток может быть устранен путем надлежащей корректировки программного обеспечения приборов.

Количество урана-235 и урана-238 в ТВС типа "А" измерялось прибором, состоящим из нейтронного детектора, америций-литиевого нейтронного источника, счетчика совпадений НЕС-100 и микрокалькулятора НХ-20. Градуировка приборов выполнялась с помощью разборных экспериментальных чехлов ТВС, содержащих твэлы с обогащением по урану-235 23% и 36%. Из-за высокой концентрации урана-235 градуировочные кривые имели нелинейный характер с насыщением, появляющимся по мере добавления твэлов в чехол. Прибор успешно использовался во время практических занятий. К его недостаткам следует отнести слабую помехоустойчивость к электромагнитным наводкам, часто возникающим при включении силового оборудования, расположенного в соседних помещениях. Для него следует предусматривать надежное заземление и включить в состав программного обеспечения специальный блок статистического анализа на выбросы. Все это повысит надежность и уменьшит время измерений.

Измерение массы плутония на единице длины ТВС типа "О" выполнялось прибором PWCC, состоящим из 4-х секционного блока детектирования нейтронов, счетчика совпадений НЕС-100 и микрокомпьютера НР-97. Установка градуировалась по ТВС, содержащим примерно одинаковые массы плутония, что не позволило определить градуировочную кривую в широком диапазоне масс плутония. Относительные измерения количества плутония в ТВС были выполнены успешно в предварительных экспериментах и на практических занятиях. Абсолютные измерения выполнить не удалось из-за неудачи при определении изотопного состава топливной композиции, знание которого необходимо для правильной интерпретации нейтронных измерений.

Оценивая результаты, отметим, что в настоящее время МАГАТЭ располагает комплектом измерительной аппаратуры и соответствующими методиками, обеспечивающими контроль количества ядерных материалов на АЭС с реакторами на быстрых нейтронах с приемлемой точностью и хорошим уровнем достоверности. Применяемая электронная аппаратура и блоки детектирования нейтронного и гамма-излучения отвечают современным требованиям, в большинстве своем достаточно компактны, удобны в обращении и надежны.

Отмеченные недостатки не принципиальны и устранение их возможно как посредством приспособления процедур измерения к новым условиям, так и совершенствования методического и программного обеспечения.

Существенно то, что они проявились при измерении топливных композиций, с которыми ранее специалисты МАГАТЭ не встречались. Из этого следует, что в настоящее время еще трудно рассчитывать на создание универсальной методики и соответствующей процедуры измерений, пригодных для контроля любых топливных композиций. Поэтому для обеспечения контроля топлива реакторов на быстрых нейтронах требуется разработка специализированных методик для существующей аппаратуры, учитывающих специфические особенности топлива быстрых реакторов. По крайней мере, это надо делать в тех случаях, когда появляются существенные отличия топливных композиций от обычных.

Программа СССР научно-технической поддержки гарантий МАГАТЭ предусматривает участие специалистов различных научных учреждений Советского Союза в работах по совершенствованию системы гарантий, в т.ч. и в работах, относящихся к применению на АЭС и других установках с реакторами на быстрых нейтронах.

Поскольку реакторы на быстрых нейтронах являются наиболее перспективными для развития атомной энергетики, разработка эффективной системы гарантий для них становится актуальной, и особое значение приобретает вопрос о подготовке квалифицированных кадров для проведения инспекций на таких реакторах.

Основными задачами в этом направлении являются:

- разработка предложений по усовершенствованию вопросника МАГАТЭ о конструкции установки;
- анализ и испытание существующих методов неразрушающего контроля топлива применительно к быстрым реакторам, разработка новых методов и процедур;
- разработка учебно-методических материалов для использования при проведении учебно-тренировочных курсов инспекторов МАГАТЭ.

В Советском Союзе работы в этом направлении будут развиваться в основном опираясь на экспериментальную и техническую базу, представляемую реактором БОР-60, с участием ведущих научно-исследовательских учреждений СССР и, возможно, зарубежных исследовательских организаций.

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## **ДИСТАНЦИОННОЕ МОНИТОРИРОВАНИЕ ЭНЕРГОВЫРАБОТКИ РЕАКТОРА ПО ЕГО НЕЙТРИННОМУ ИЗЛУЧЕНИЮ КАК ВОЗМОЖНОЕ СРЕДСТВО КОНТРОЛЯ ЯДЕРНЫХ МАТЕРИАЛОВ НА АЭС**

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### **Abstract—Аннотация**

**REMOTE MONITORING OF A REACTOR'S POWER OUTPUT BY ITS NEUTRINO RADIATION AS A POSSIBLE MEANS OF SAFEGUARDING NUCLEAR MATERIALS AT POWER PLANTS.**

A fundamentally new approach to the problem of remote monitoring for safeguards purposes of reactor power output and plutonium buildup over the fuel campaign is put forward. It is based on recording the neutrino radiation released during the fission chain reaction. The proposal hinges on the fundamental neutrino research carried out by the I.V. Kurchatov Institute of Atomic Energy at the Rovno nuclear power station. The neutrino detectors used are briefly described. It is noted that the provision of specialized research as applied to the goals of the IAEA is opportune, as is the development of a strategy for using the neutrino method as a safeguards tool.

**ДИСТАНЦИОННОЕ МОНИТОРИРОВАНИЕ ЭНЕРГОВЫРАБОТКИ РЕАКТОРА ПО ЕГО НЕЙТРИННОМУ ИЗЛУЧЕНИЮ КАК ВОЗМОЖНОЕ СРЕДСТВО КОНТРОЛЯ ЯДЕРНЫХ МАТЕРИАЛОВ НА АЭС.**

Для целей гарантий предлагается принципиально новый подход к проблеме дистанционного определения энерговыработки реактора и контроля за накоплением плутония в ходе кампании, основанный на регистрации нейтринного излучения, возникающего при протекании цепной реакции деления. Предложение опирается на опыт фундаментальных нейтринных исследований, ведущихся Институтом атомной энергии им. Курчатова на Ровенской АЭС. Кратко описаны используемые детекторы нейтрино. Отмечается своевременность постановки специальных исследований применительно к целям МАГАТЭ, а также разработки стратегии использования нейтринного метода технического средства гарантий.

Для целей международных гарантий операторы АЭС сообщают МАГАТЭ расчетные данные по выгоранию урана-235 и производству плутония за определенные периоды эксплуатации реакторов [1]. Точность таких расчетов может быть различна. Кроме того, без независимой экспериментальной проверки этих данных они могут иметь лишь ограниченное значение для МАГАТЭ, а возможная экспериментальная

оценка концентрации плутония и обогащения урана в отработавшем топливе на стадии его переработки отрицательно сказывается на своевременности обнаружения. Для своевременной проверки данных оператора МАГАТЭ совместно с государствами в течение ряда лет разрабатывает технические средства, позволяющие осуществлять мониторинг энерговыработки энергетических реакторов [2, 3]. Упомянутые приборы являются довольно компактными, однако они имеют ряд недостатков, например, принципиально возможно вмешательство оператора в процессе измерений, окончательные результаты получаются косвенным путем, в отдельных случаях необходимо использование лабораторных услуг и др. В этой связи предлагается рассмотреть принципиально новый подход к проблеме независимого определения энерговыработки реактора, основанный на регистрации нейтринного излучения.

1. Нейтринное излучение ядерного реактора неразрывно связано с процессом деления, является его неотъемлемой стороной и проявлением: каждый акт деления дает определенный вклад в общую энерговыработку, увеличивает массу осколков деления и порождает нейтрино. Нейтрино проникают без поглощения через любые материалы и могут быть зарегистрированы с помощью детекторов, расположенных за биологической защитой или, возможно, за пределами здания реактора.

Из сказанного выше ясно, что дистанционное мониторинг нейтринного потока дает в определенном масштабе число ядер, испытавших деление, энерговыработку реактора и выгорание ядерного горючего.

Нейтринное излучение реактора содержит также информацию о текущем составе ядерного горючего в активной зоне. Это связано с существенными различиями в энергетических спектрах нейтрино, генерируемых различными делящимися изотопами: урана-235 и -238, плутония-239 и -241. Таким образом, спектральный анализ нейтринного излучения реактора может создать основу определения динамики выгорания и накопления делящихся изотопов в ходе кампании и, в первую очередь, накопления плутония, представляющего наибольший интерес.

2. Сечение взаимодействия  $\tilde{\nu}_e$  с веществом в  $10^{19} - 10^{20}$  раз меньше, чем у других частиц, например, у нейтронов или гамма-квантов. Связанные с этим трудности привели к тому, что в течение длительного времени (1953—1979 гг.) только известная группа исследователей в США, во главе с Ф. Райнесом, регистрировала реакторные нейтрино.

Восьмидесятые годы характеризуются быстрым прогрессом в методах регистрации нейтрино. С 1982 г. работы Института атомной энергии им. И.В. Курчатова в этой области ведутся в специализированной нейтринной лаборатории на Ровенской АЭС [4] (рис. 1), где используются два различных детектора  $\tilde{\nu}_e$ . Еще две группы, располагающие двумя идентичными детекторами, публикуют результаты, полученные на реакторах в Швейцарии [5] и во Франции [6].

Вопрос о возможном использовании нейтринного излучения в практических целях был впервые поставлен в 1975—1976 гг. [7, 8] в связи с подготовкой к работам на Ровенской АЭС. В 1983 г. осуществлен демонстрационный эксперимент [9], в котором было измерено энерговыделение реактора ВВЭР-440 по его нейтринному излучению.



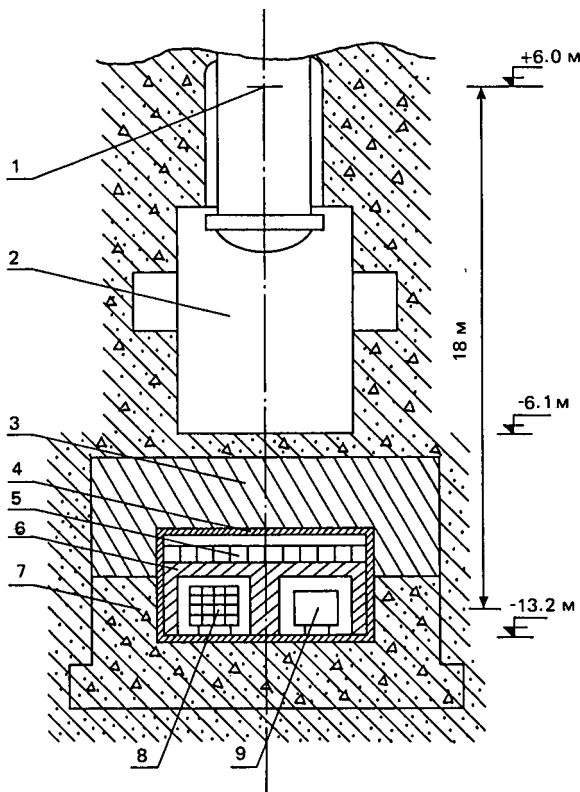


РИС. 1. Разрез нейтринной лаборатории на Ровенской АЭС: 1 – центр активной зоны реактора; 2 – технологическое помещение; 3 – тяжелый бетон; 4 – железо; 5 – баки активной защиты; 6 – полиэтилен; 7 – бетон; 8 – детектор интегрального типа; 9 – сцинтилляционный спектрометр.

Следует отметить, что нейтринные эксперименты, ведущиеся на реакторах, направлены на выяснение фундаментальных свойств нейтрино. Работы в области практического использования  $\bar{\nu}_e$  [7–9] являются пока побочным продуктом фундаментальных исследований и имеют основной целью привлечь внимание к потенциальным возможностям нейтринного метода.

3. Регистрация осуществляется по продуктам реакции



в которой ядерно-свободный протон под действием  $\bar{\nu}_e$  превращается в нейтрон и по-

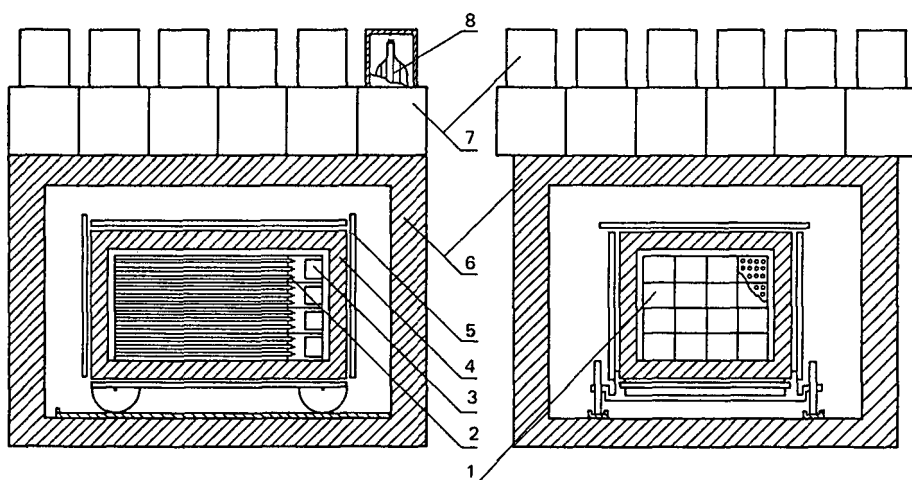


РИС. 2. Детектор интегрального типа: 1 – детектор; 2 – нейтронные счетчики; 3 – предусилитель; 4 – борированный полиэтилен; 5 – пласти активной защиты; 6 – полиэтилен; 7 – баки активной защиты; 8 – ФЭУ.

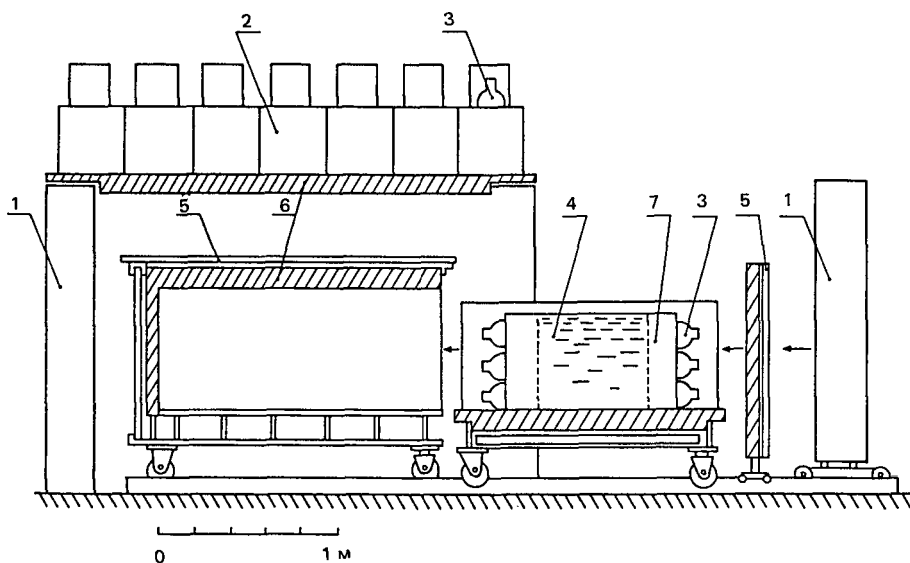


РИС. 3. Сцинтилляционный спектрометр: 1 – полиэтилен; 2 – баки активной защиты; 3 – ФЭУ; 4 – жидкий сцинтиллятор; 5 – пласти активной защиты; 6 – борированный полиэтилен; 7 – световоды.

зитрон. Существенная особенность этой реакции состоит в том, что кинетическая энергия возникающего позитрона  $T_e$  связана с энергией нейтрино  $E_\nu$  соотношением  $T_e = E_\nu - 1,8 \text{ МэВ}$ .

Таким образом, полное число зарегистрированных в детекторе событий дает интенсивность нейтринного излучения реактора, а измерение энергетического спектра позитронов позволяет восстановить спектр этого излучения.

В лаборатории действуют две экспериментальные установки. В одной из них (рис. 2) мишенью для нейтрино и замедлителем образующихся нейтронов служит полиэтилен. Замедлившиеся нейтроны регистрируются в газовых счетчиках, заполненных гелием-3. Этот детектор позволяет измерять поток  $\tilde{\nu}$ , но не дает информации об их энергетическом спектре.

Другая установка — сцинтилляционный спектрометр  $\tilde{\nu}_e$  (рис. 3). Мишенью для  $\tilde{\nu}_e$  и замедлителем нейтронов, а также детектором позитронов и нейтронов служит жидкий органический сцинтиллятор с добавками гадолиния. Энергия позитрона определяется по величине сцинтилляционной вспышки.

Полезная масса мишени (полиэтилена в первом и жидкого сцинтиллятора — во втором случае) составляет приблизительно 190 кг. Детекторы расположены на расстоянии 18 м от центра активной зоны. Скорость счета нейтринных событий — 300–500 в сутки. Реально за кампанию реактора ВВЭР-440 от перегрузки до перегрузки регистрируется  $(3-5) \times 10^4$  нейтринных событий. Более подробное описание детекторов можно найти в работах [10, 11].

4. Связь между числом зарегистрированных прибором за определенное время событий  $N_\nu$  и числом делений  $N_f$ , имевших место в реакторе за это же время, дается соотношениями:

$$N_\nu = \gamma \frac{N_f N_H}{R^2}; \quad \gamma = \frac{\epsilon \bar{\sigma}_f}{4\pi}, \quad (2)$$

где  $N_H$  — число атомов водорода в мишени,  $1/R^2$  — геометрический фактор,  $\epsilon$  — эффективность регистрации, реально составляющая 30–50%,  $\bar{\sigma}_f \cong 6 \times 10^{-43} \text{ см}^2/\text{деление}$  — среднее по изотопному составу горючего в зоне сечение взаимодействия.  $\bar{\nu}_e$  с протоном:  $\bar{\sigma}_f = \sum \alpha_i \sigma_f^i$ , где  $\alpha_i$  — вклад данного делящегося изотопа в общее число делений. Характерный для водо-водяных реакторов с топливом низкого обогащения состав зоны:

изотоп	$^{235}\text{U}$	$^{238}\text{U}$	$^{239}\text{Pu}$	$^{241}\text{Pu}$
вклад	60%	7%	27%	6%

В ходе кампании вклад урана-238 остается практически постоянным. Из-за выгорания урана-235 и образования изотопов плутония в ходе кампании величина  $\bar{\sigma}_f$ , входящая в основное соотношение (2), меняется приблизительно на 4%, что необходимо учитывать при достаточно точных измерениях.

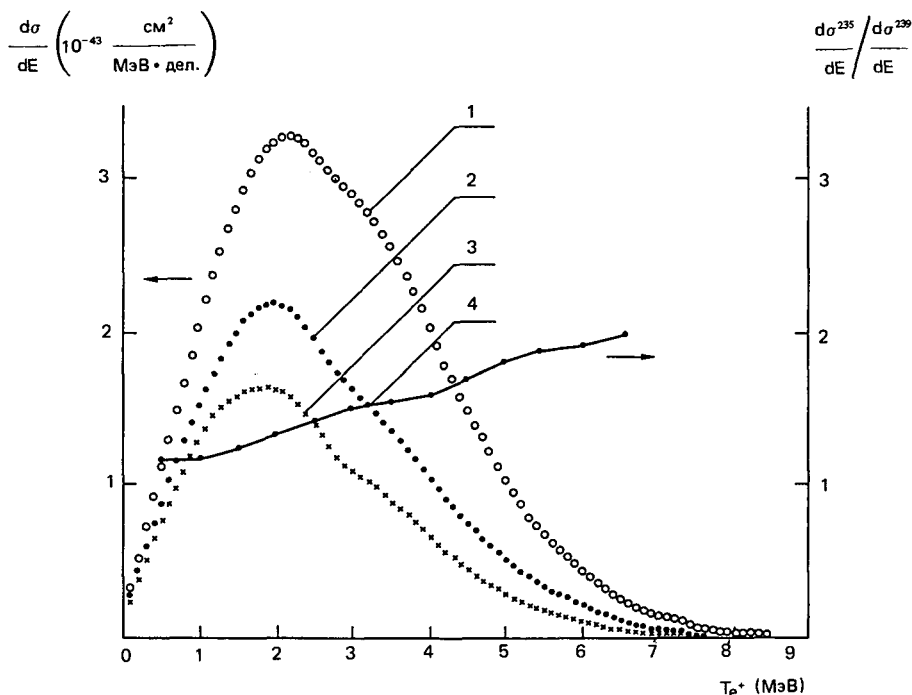


РИС. 4. Дифференциальные расчетные спектры позитронов  $d\sigma/dE$  реакции обратного бета-распада: 1 – от  $^{238}\text{U}$ , 2 – от  $^{235}\text{U}$ , 3 – от  $^{239}\text{Pu}$ ; 4 – отношение спектров позитронов

$$\frac{d\sigma^{235}}{dE} \bigg/ \frac{d\sigma^{239}}{dE}.$$

По измеренному числу нейтринных событий  $N_\nu$  из (2) можно найти число делений  $N_f$  и далее, практически без потери точности, энерговыработки  $W$  и массу выгоревшего горючего  $M$ .

Накопление плутония, как указано выше, слабо влияет на интегральную величину  $N_\nu$ . По этой причине интегральные измерения потока  $\nu_e$  не могут быть прямо использованы для определения количества плутония. Нейтринная спектроскопия открывает здесь гораздо большие возможности. На рис. 4 представлены энергетические спектры позитронов, возникающих в реакции (1) под действием урана-235 и -238, плутония-239. Видно, что с ростом энергии различие в спектрах урана-238 и плутония-239 растет, так что при больших энергиях  $\tilde{\nu}_e$  от урана-235 дает в 1,6–2,0 раз больше событий, чем от плутония-239.

5. На данной стадии целесообразно подчеркнуть особенности рассматриваемого подхода, отличающие его от других, более традиционных методов:

(а) нейтринный метод контроля по самой природе дистанционный. Он не требует контакта с активной зоной. Показания нейтринных приборов не зависят от возможных ошибок оператора. Действие этих приборов не мешает работе оператора;

(б) измеряемая интенсивность нейтринного излучения прямо связана с основной величиной, характеризующей работу реактора, а именно с числом ядер испытывших деление в его активной зоне. Для реакторов типа ВВЭР нейтринный метод может иметь такую же точность, как и применяемый в настоящее время тепловой. Для реакторов других типов нейтринный метод может оказаться более точным. В любом случае, однако остаются особенности, отмеченные в (а);

(с) уникальны перспективы определения накопления плутония непосредственно в ходе его производства. Развитие этого направления позволит создать принципиально новые средства определения полного потока расщепляющихся материалов в долгосрочные хранилища;

(д) градуировка нейтринных детекторов и спектрометров может в принципе производиться на одном — эталонном реакторе. Это открывает возможность стандартизации приборов контроля.

Одним из главных итогов последнего периода состоит в том, что достигнут существенный прогресс в методах измерения потока и спектра нейтрино. На Ровенской АЭС создана и функционирует специализированная нейтринная лаборатория, где могут при необходимости проводиться соответствующие исследования и испытания.

Предлагаемый подход не противопоставляется существующим методам покассетного анализа и контроля. Нейтринный подход мы рассматриваем как принципиально новый, дополняющий имеющиеся методы.

Представляется целесообразным провести целенаправленные исследования применительно к проблеме гарантий МАГАТЭ. Еще предстоит доказать экспериментально возможность измерения энергovyработки по нейтринному излучению с погрешностью не большей 1—1,5%. Предстоит более полно, чем это сделано сейчас, исследовать реальные возможности нейтринного спектрального анализа. Нуждается в уточнении и константное обеспечение этого направления.

Из изложенного ясно, что нейтринные детекторы относятся к классу транспортных, но не портативных детекторов. В связи с этим целесообразно определить хотя бы в предварительном порядке желательное место размещения таких приборов на действующих энергетических реакторах. Что касается проектируемых реакторов, было бы целесообразно заранее предусмотреть стандартное положение таких приборов, что способствовало бы в будущем унификации системы нейтринного контроля.

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## SAFEGUARDS CONCEPT FOR THE THTR-300 THORIUM HIGH TEMPERATURE REACTOR

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### Abstract

#### SAFEGUARDS CONCEPT FOR THE THTR-300 THORIUM HIGH TEMPERATURE REACTOR.

The nuclear power plant in Hamm, Federal Republic of Germany is a plant with a high temperature reactor. The fuel elements are spheres with a diameter of 60 mm, an enrichment of 93%, a  $^{235}\text{U}$  content of 0.96 g and a thorium content of 10.2 g. The facility is divided into two material balance areas (MBA 1: fresh fuel and spent fuel storage; MBA 2: loading facility, reactor core and discharge facility) in order to increase the transparency of the fuel element flow, and because a direct physical inventory of the core is not possible. The inventory of the core is determined by the quantity received and the quantity withdrawn, which are to be reported to Euratom. The quantity received in the core will be verified by the inspectors using a special sampling machine. The quantity withdrawn from the core is counted by independent counters at the exit of the core. At the exit of the spent fuel storage a monitoring and logging system verifies all the drums leaving the storage. The route of the spent fuel drums is continuously observed by video systems until they are finally packed into shipping containers which are to be sealed by the operator using electronic seals.

### 1. BRIEF DESCRIPTION OF THE POWER PLANT AND THE FUEL ELEMENTS

The nuclear power plant in Hamm, Federal Republic of Germany, has a high temperature reactor, with a net electrical power of 300 MW(e). The fuel elements are spheres with a 60 mm diameter, an enrichment of 93%, a  $^{235}\text{U}$  content of 0.96 g and a thorium content of 10.2 g. The total weight ranges from 200.8 to 208.0 g. The fresh fuel element storage has a maximum capacity of 168 barrels each containing approximately 1000 elements, so that the maximum inventory amounts to about 150 e. The equilibrium reactor core consists of 674 500 fuel elements (about 234 e on average). The spent fuel storage has a maximum capacity of 243 drums each containing about 2100 spent fuel elements (about 122 e).

### 2. THE AIM OF THE SAFEGUARDS CONCEPT FOR THE THTR-300

The aim of the concept described in this paper is to show which safeguards measures are applied to meet the following

inspection goal - the detection of diversion of fresh fuel elements within one month; and of spent fuel elements within three months.

This goal is subject to the condition of minimizing the inspection effort by the use of automatic safeguards measures.

When sampling techniques are applied then the basis for determining sample size is as described in the following sections.

## 2.1. Attribute testing

(1) Hypergeometric function

(2) Operating characteristic: 
$$\beta = \sum_{x=0}^c \frac{\binom{M}{x} \binom{N-M}{n-x}}{\binom{N}{n}}$$

(3) N = Total number of items in a finite group

(4) C = 0

(5) M = Number of discrepancies

(6) n = Number of items examined by the inspector

(7) x = Observed number of discrepancies

(8)  $\beta \leq 0.1$  for M = 26 000 fresh fuel elements = 25 kg  $^{235}\text{U}$

M = 34 000 spent fuel elements = 8 kg  $^{233}\text{U}$

(9) Null hypothesis  $H_0$ : M = 0

(10) Alternative hypothesis  $H_1$ : M > 0

(11)  $H_0$  will be accepted if x = C

(12)  $H_1$  will be accepted if x > C

## 2.2. Variable testing

Destructive analysis (DA) and variable testing are not foreseen in this concept. Consequently, a variable sampling plan is not necessary.



### 3. MBA STRUCTURE OF THE FACILITY

The facility is divided into two material balance areas - MBA 1: fresh fuel storage, and spent fuel storage; and MBA 2: loading facility, reactor core and discharge facility - in order to increase the transparency of the fuel element flow and because a direct physical inventory of the core is not possible. The inventory of the core is determined by the quantity received and the quantity withdrawn, which are to be reported to Euratom.

### 4. INTERMITTENT ROUTINE INSPECTION ACTIVITIES (see Fig. 1)

These inspections will take place once a month and last about two days if only fresh fuel elements are verified, and four days if both fresh and spent fuel elements are verified.

#### 4.1. Examination of records

An examination of the records consists of examining the book inventory using facility data (e.g. for the purpose of updating); examining the records; and reconciling reports with records.

#### 4.2. Verification of the fresh fuel storage (MBA 1)

The assumption is that appropriate sealing is applied in the shipper's facility on all fresh fuel barrels received. Verification consists of:

- Counting the barrels;
- Identification and seal checking of the barrels:
  - 100% of the newly arrived barrels
  - the remainder on a sampling plan basis;
- Application, removal and replacement of seals on barrels (replacement on a sampling plan basis); and
- NDA measurements of unsealed barrels on a sampling plan basis.

#### 4.3. Verification of the reactor core (MBA 2)

Verification of the reactor core, MBA 2, consists of counting the elements which go into the core by the sampling machine counters (see Section 7.1) at the entry to the core; carrying out NDA measurements of the elements sampled by the sampling machine; and counting the elements which leave the core by the independent counters at the core exit (see Section 7.2).

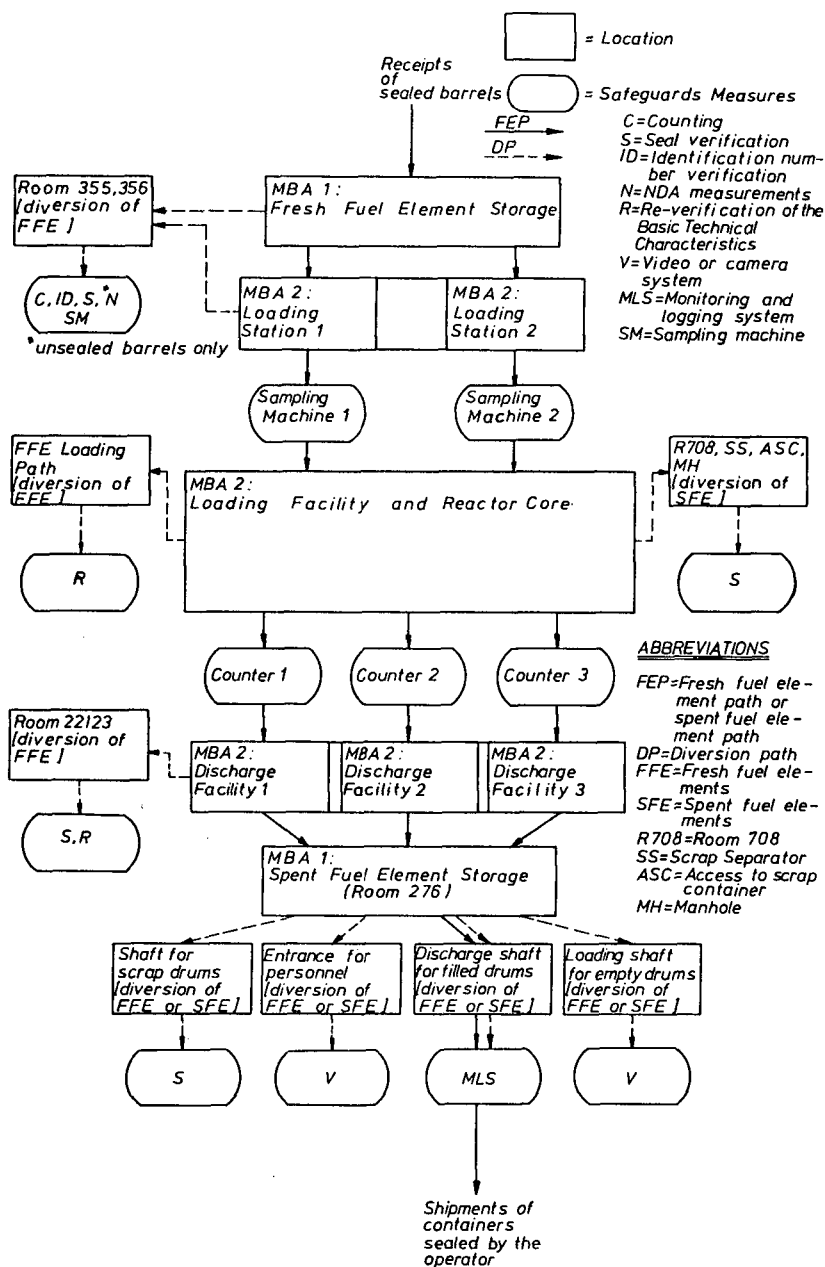


FIG. 1. Fuel element path and diversion path.

#### 4.4. Verification of the spent fuel storage (MBA 1) during the run-in phase

During the run-in phase (about three years) graphite, absorber and fuel elements are stored in the spent fuel storage. Based on the independent counters at the exit of the core the total number of newly arrived elements (absorber, graphite and spent fuel elements) is known. But, in order to know how many of them are spent fuel elements, additional information and verifications are necessary. The operator has to report to Euratom the number of newly arrived spent fuel elements (shipment from MBA 1 to MBA 2). Consequently, the inspectors have to verify the quantity received by the following measures:

- Counting, identification and NDA measurements of spent fuel drums which go to the spent fuel storage (identification and NDA measurements (see Section 7.6) will be carried out on a sampling plan basis every three months).
- Automatic NDA measurements of all drums which leave the spent fuel storage (see Section 7.5).

Based on the quantity received and the quantity withdrawn the inventory of the spent fuel storage can always be determined because the continuity of knowledge is maintained by C/S measures. A direct inventory verification is also possible but very time consuming (it can take 11 days).

#### 4.5. Verification of the spent fuel storage (MBA 1) when the equilibrium reactor core is reached

When the equilibrium reactor core is reached then the elements withdrawn from the core can only be fuel elements. Consequently, it is no longer necessary to carry out NDA measurements and identification verifications of the drums which went to the spent fuel storage, because all possible exits of the store are under safeguards control. The number of fuel elements which go to the spent fuel store is determined by the counters at the exit of the core. The drums leaving the spent fuel storage can be determined by the monitoring and logging system at the exit of the spent fuel storage. Consequently, the inventory of the spent fuel storage is always known.

The routes of spent fuel drums are continuously observed by video systems until they are finally packed into shipping containers which will be sealed by the operator using electronic seals. These electronic seals, e.g. VACOSS seals, have the property that any application or reopening is monitored in a tamperproof way.

#### 4.6. Other activities

Other activities include:

- Application, checking, removal and replacement of seals on safeguards equipment, scrap separators, access to scrap containers, manhole, and to rooms 708, 22/23 and 276;
- Servicing the sampling machine at the entry to the core (see Section 7.1);
- Servicing the counters at the exit of the core (see Section 7.2);
- Servicing the video system at the shaft of the spent fuel storage for empty drums (see Section 7.3);
- Servicing the camera unit at the personnel access to the spent fuel storage (see Section 7.4);
- Servicing the monitoring and logging system at the exit of the spent fuel storage (see Sections 7.5 and 7.6).

#### 5. PHYSICAL INVENTORY VERIFICATION (PIV)

The PIV will take place once a year and last about four days. The inspection activities will be exactly the same as described in Section 4, but 100% identification and seal checking of the fresh fuel element barrels in the fresh fuel storage will be carried out.

#### 6. ADDITIONAL ROUTINE INSPECTION ACTIVITY (see Fig. 1)

A further routine inspection activity is the reverification of the basic technical characteristics (BTC) of the fresh fuel element loading path because the use of C/S techniques alone would not cover all possible strategies to divert fresh fuel elements. This BTC reverification will be carried out on a random basis about five times per year. Such inspections can be accomplished in less than half a day.

#### 7. SAFEGUARDS HARDWARE

##### 7.1. Sampling machine (see Fig. 2)

The purpose of the sampling machine is to verify the quantity received in the reactor core by counting, and by sampling randomly selected pebbles which are measured by NDA. The sampling machine is located behind the loading station in room 302. One sampling machine consists of the function block with doser and switch; the sample container; counters 1, 2, 3;

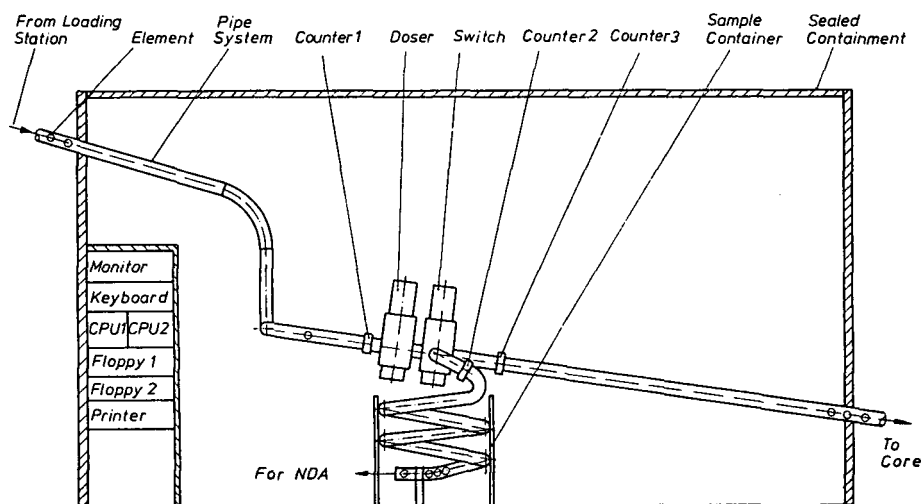


FIG. 2. Sampling machine.

two microcomputers (redundancy) - a CPU1 with real time clock and a CPU2 with real time clock; and a monitor, keyboard, floppy disk drive I, floppy disk drive II, memory 1 (program: EPROM; data: CMOS-RAM), memory 2 (program: EPROM; data: CMOS-RAM), multiplexer with data buffer, a printer, and interfaces for input and output signals.

The function block is a cast steel block containing the doser and the switch unit. These are standard components used in other function blocks in the reactor.

The total capacity of the sample container is 66 elements.

The whole system including the microcomputer and accessories is stored in a sealed containment.

The system operates as follows:

The inspectors either manually enter 66 random numbers between 1 and 22 000 or these are automatically generated by the computer. The CPU counts the signals from counter 1 and compares them with the random numbers 1...66. If a random number and the accumulated signals from counter 1 are equal the doser is blocked; the switch is turned to a 45° position; the doser releases the pebble; the pebble is counted by counter 2 and falls into the sample container; counter 2 gives a signal to the CPU; the switch is turned to its vertical position; random number, date and time are printed.

If a random number and the accumulated signals from counter 1 are not equal the pebble passes counter 3 and goes to the core.

During each inspection the 'balance period', 'balance report', 'error diagnosis' and the date and time when the inspectors have interrupted the program are printed.

During the PIV the total number of pebbles which went to the core within the material balance period is printed.

#### 7.2. Counters at the exit of the core

The purpose of the counters is to count the elements which leave the core.

#### 7.3. Video system at the shaft for empty drums in the spent fuel storage

The purpose of this video system is to record when a drum leaves the spent fuel storage via the shaft. This is not foreseen in the BTC.

#### 7.4. Camera unit at the personnel entrance to the spent fuel storage

The camera is placed outside the spent fuel storage with a view of the personnel entrance. The purpose of the camera unit is to record if a drum leaves the spent fuel storage via the personnel entrance. This is not foreseen in the BTC.

#### 7.5. Monitoring and logging system at the exit of the spent fuel storage during the run-in phase (see Fig. 3)

The system consists of video and NDA units. The video unit records when a drum leaves the storage; based on these records the inspectors can determine the number of drums which have left the storage during a certain period.

The NDA unit automatically carries out NDA measurements of all drums leaving the storage to ensure that a drum contains graphite or absorber elements (as declared by the operator) and no fresh fuel elements. Also it prints the date, time and the measurement results when a drum leaves the storage; the printed date and time must harmonize with the video records.

#### 7.6. Monitoring and logging system at the exit of the spent fuel storage when the equilibrium reactor core is reached (see Fig. 3)

The system consists of video and NDA units. The purpose of the video unit is to record when a drum leaves the storage;

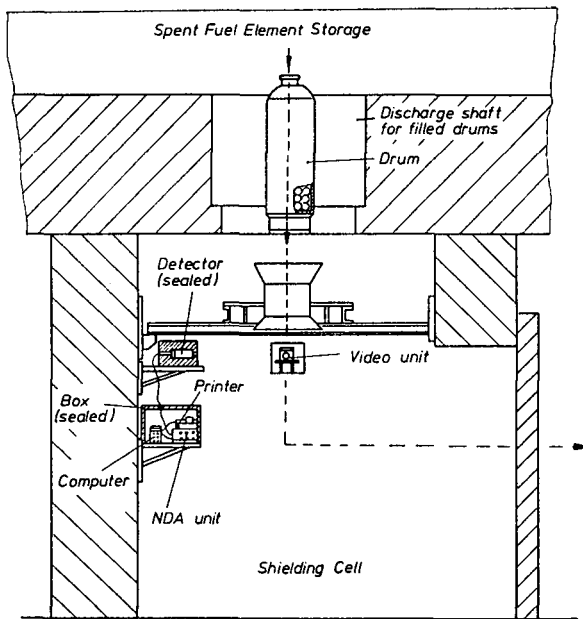


FIG. 3. Monitoring and logging system.

based on these records the inspectors can determine the number of drums which have left the storage during a certain period; to ensure that the drum which was measured by the NDA unit really goes into the shipping container; and to observe the shipping container until it is finally closed and sealed by the operator using electronic seals.

The NDA unit automatically carries out NDA measurements of all drums leaving the storage to ensure that a drum packed into a shipping container contains spent fuel elements. Also it prints the date, time and the measurement results when a drum leaves the storage; the printed date and time must harmonize with the video records.

## 8. FINAL REMARKS

Safeguarding a high temperature reactor such as the THTR-300 is more complicated than, for example, safeguarding light water reactors because of the complex fuel element route involved and because a direct physical inventory of the core is not possible. The above concept demonstrates that it is

possible to safeguard the THTR with a reasonable inspection effort when special safeguards hardware and inspection activities such as described in Section 6 are applied.

#### ACKNOWLEDGEMENTS

This paper is based mainly on the Joint Programme<sup>1</sup>. The author acknowledges the contributions of R. Baeumer (VEV, Dortmund), M.J. Canty (KFA, Jülich), S. Mika (HRB, Mannheim) and S. Nicolai (KFA, Jülich) for their extensive work in the development of the THTR-300 safeguards approach.

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<sup>1</sup> Joint Programme on the Technical Development and Further Improvement of IAEA Safeguards between the Government of the Federal Republic of Germany and the IAEA.



## SOME SAFEGUARDS IMPLICATIONS OF INACCESSIBLE INVENTORIES

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### Abstract

#### SOME SAFEGUARDS IMPLICATIONS OF INACCESSIBLE INVENTORIES.

In the medium term future a considerable increase is expected in the quantities of nuclear material which will have to be regarded as being inaccessible for safeguards verification purposes. Three classes of inaccessible inventory are identified, based upon the length of time during which the nuclear material resides within the inaccessible area and the ease with which the inventory can be reduced to near zero. The authors conclude that credible safeguards will require effective monitoring of the flow of safeguards material into and out of the inaccessible inventories, combined with an effective containment and surveillance regime. There does not appear to be a requirement for novel technologies to achieve such a regime, but high equipment reliability will be essential. An effective regime might also benefit from closer routine co-operation between the facility operator and the inspectorates with respect to the functioning of surveillance devices.

### 1. INTRODUCTION

In the medium term future there is expected to be a considerable increase in the quantities of nuclear material within safeguarded facilities which, for all practical safeguards purposes, will have to be regarded as being inaccessible to the Inspectorates. The inaccessibility may have safeguards implications. The basis of IAEA safeguards is "the use of (nuclear) materials accountancy as a safeguards measure of fundamental importance, with containment and surveillance as important complementary measures"[1]. The process of materials accountancy entails the periodic closing of material balances by checking the physical inventories and comparing them with the 'book' inventory derived from the previous inventory corrected for declared receipts and shipments. An essential safeguards element of this procedure is the independent verification of the materials accountancy data in a timely manner. This is achieved

by physical inventory verification and by flow monitoring techniques at strategic points using batch or item counting and the use of non-destructive assay (NDA) and chemical analytical methods. The particular techniques employed and their frequency of application depend upon the type and form of the material and upon the operational procedures at each facility. Where nuclear materials are inaccessible for the purpose of independent verification it may be necessary to adopt alternative safeguards techniques, or relax the inspection goals (particularly the timeliness goal), or modify the safeguards philosophy on the needs for independent verification. In this paper the authors discuss the implications of the impact of inaccessible nuclear material upon the use of nuclear materials accountancy, in particular with respect to physical inventory verification, and upon the role and requirements for containment and surveillance.

The presence of nuclear material in nominally inaccessible inventories is not a new feature in the Agency's range of experience. For example, the inventory of fuel in the core of LWRs is not accessible for verification more frequently than the frequency of refuelling, which may be at intervals of between 12 and 18 months [2], although spent fuel in the storage ponds can be and is verified at the inspection guideline frequency of every 3 months. In CANDU, gas cooled reactors and fast reactors, the core inventory is not accessible for verification purposes because of the design and mode of operation. There are also examples where spent fuel assemblies are stored in ponds in such a way that, whilst in principle they could be accessed, there are operational complexities and hazards which make it undesirable and costly to do so. Looking to the future it is clear that there will be a considerable increase in the number and types of facility where nuclear materials under safeguards will have to be regarded as inaccessible for the purposes of physical inventory taking and verification; for example, in long term storage facilities.

## 2. A TAXONOMY OF INVENTORIES

There is no absolute definition of the term "inaccessible"; it is a matter of degree, depending upon the availability of resources and the determination and resourcefulness of the individual(s) seeking access. For the purposes of this paper the term is used to describe the state or location of nuclear material where the location, identity and/or integrity of items or the location, identity and quantity of material cannot be independently verified with present technology without excessive intrusion into routine operations at the facility or involving undesirable costs and risks. Using this definition it is possible to classify inaccessible inventories into three types:

- Type A, where the residence time of the material within the inventory is relatively short, generally less than 2-3 days, and where the inventory can conveniently be reduced to near zero at chosen intervals;
- Type B, where material passing through the inventory region has a residence time of several months or a few years before it is again accessible for safeguards purposes;
- Type C, where material is placed in the inventory for long periods with no expectations of a change in its accessibility status for, perhaps, several tens of years.

We now identify the relevant features of the three classes of inaccessible inventory and assess the significant differences between them from a safeguards point of view.

### 3. SAFEGUARDS FEATURES

Type A is typified by the in-process inventories in bulk handling facilities as in the chemical separation stages of a reprocessing plant or in certain process stages in nitrate to oxide conversion plants. The size of the inventory can and does fluctuate during routine operations. Process control procedures are designed to maintain the inventories within design limits in order to achieve product quality, to meet safety standards and for efficient operation. In the solvent extraction contactors of a reprocessing plant the in-process hold-up is constrained by the plant geometry (pulsed columns or mixer-settlers) and the need to keep the aqueous and organic phases within flow and concentration limits in order to achieve effective separation of the feed materials. The nuclear materials inventory can be shown to approximate to a simple mathematical model, the inputs to which are plant and process data readily available to the inspectors [3,4]. This mathematical simulation of the in-process inventory has relevance in the technique of near-real-time materials accountancy (NRTMA) where mass balances are closed at frequent intervals (daily, weekly, for example) as a form of process monitoring. NRTMA permits mass balances to be closed without the need to have plant shutdown with a washout in order to measure the inventory; an estimate of the inaccessible inventory can be derived from accessible information. The use of a mathematical model is still under test and its value as a safeguards tool will be closely bound to the success of near-real-time accountancy itself. The analogous inventories in a nitrate to oxide conversion plant would primarily be the drying and calcining furnaces. The inventories of transport casks are also consistent with the definition of Type A inventories.

Because of the ability to reduce the immeasurable inventories to a minimum (e.g. during plant shutdown) Type A inventories are notably different from the inaccessible inventories of Types B & C. The Inspectorates are involved both through existing safeguards activities and through Member States Support programmes with the issues and methods of treating Type A inaccessible inventories and we choose not to consider them further in this paper.

Type B is typified by the core and in-sodium inventory of a fast reactor with fuel assemblies being transferred into the inaccessible inventory where they may reside for typically between two and six years before being discharged for further processing. Depending upon the particular design of the reactor, during the period of inaccessibility the items are either moved remotely under the cover of the reactor vessel (i.e. within the containment structure in the case of 'pool' type designs), or they are transferred from location to location within a closed containment vessel (i.e. above the reactor working face as in some 'loop' type designs). In the 'loop' design fuel assemblies could be regarded as being accessible for NDA purposes whilst in a transfer container but unique identification might be more difficult to achieve. However, in both types of reactor designs, fuel assemblies will be under sodium for most of their lifetime at the reactor complex and effectively inaccessible to the safeguards inspectorate for location, identification and materials verification purposes for periods which exceed the timeliness detection guidelines: there could be no possibility of their removal to a location where they could be verified without an extended cooling period.

Further examples of Type B inaccessible inventories exist. As referred to above, most reactor cores come within this category but with shorter residence times than is the case in the fast reactor. The short/medium term storage of thermal reactor fuel assemblies within a single container whilst awaiting reprocessing simplifies handling and transport requirements but at the same time makes the assemblies inaccessible for safeguards verification purposes unless it is credible to either withdraw individual assemblies from the container or make some attribute measurement of the sealed containers. Other methods of short/medium term storage of thermal reactor fuel assemblies involved either the stacking of individual assemblies or the stacking of trays or baskets of assemblies within the storage pond. It could be argued that such items are more accessible than fast reactor core assemblies: in the sense of the above definition they could be located and moved so as to be accessible for verification purposes. However, the shuffling of items and racks would be intrusive into operations, is time consuming and costly and would give rise to additional personnel radiation exposure and involve additional risks of damage to stored items.

Type C is typified by some of the schemes proposed for the long term storage of thermal reactor fuels assemblies where reprocessing is not being considered as a foreseeable option. There are two basic types of long term storage designs; wet storage and dry storage [2]. Whilst there are a range of design variations within wet storage design, from the safeguards point of view they differ little from the range of spent fuel storage ponds considered as Type B except that the inventories may continue to increase over many years. In the case of dry storage, spent fuel assemblies are contained in storage flasks with no intention for them to be physically accessible during their stored lifetime. It is proposed that the storage flasks will be placed in a covered enclosure with no intention that they should be moved again until the expiry of their long term storage period. They are therefore, inaccessible for safeguards purposes.

Between Types B & C it may be possible to identify an alternative method of classification which emphasises the common features rather than the differences. For example, it would appear from a safeguards point of view that no distinction needs to be made in the safeguards treatment of fuel assemblies placed in a container at the reactor pond, whether the assemblies are intended to be shipped to a reprocessing plant or to a long term dry storage facility. In both cases it is necessary to know the contents of the multi-element bottle or cask prior to shipment, in order to maintain the book inventory of the reactor storage pond. It can be argued that verification of the multi-element bottle inventory could be achieved at the reprocessing plant immediately prior to reprocessing, but in the case of the design being developed by the Federal Republic of Germany for long term dry storage, where there are no facilities for opening a storage cask, verification would have to be done at the reactor pond prior to closure of the cask. This would require the deployment of additional resources at the reactor spent fuel pond unless fuel assembly shipments can be acceptably determined from inventory differences deduced at successive interim inventory inspections. Any proposals to increase the inspection effort at the reactor irradiated fuel pond will need to take into account the actual routine inspection effort (ARIE) negotiated between the IAEA and the State.

In the case of shipment to long term storage the continued identity of individual fuel assemblies becomes of less importance. The cask should be treated as a new item of known nuclear material content and the safeguards requirement is to provide assurance of the continuing existence and integrity of that new item. In the case of the multi-element bottle the assurance required is that 'what comes out is equal to what went in'.

It is clear from the examination of the features of inaccessible inventories that independent verification of these inventories in a timely manner cannot be achieved. The safeguards approaches will have to be based on materials flow monitoring with containment and surveillance techniques providing continuity of accountancy information. We now consider what this implies in terms of item counting and containment and surveillance.

#### 4. SAFEGUARDS TECHNIQUES

The core and in-sodium inventory of a fast reactor will be used as an example of Type B inaccessible inventory. In the pool and loop designs presently constructed or published there is only a single access port for transferring fuel assemblies into or out of the in-sodium locations (store and core areas). In view of this simplification of the fuel routes, safeguarding of the inaccessible inventory might be expected to be based on the simple principles of accounting for items entering and leaving the inventory through the designed route and providing assurance that no other (undeclared) route has been used. There is a need to account for fresh assemblies entering the in-sodium inventory in order to maintain a book account for the fresh fuel inventory; similarly irradiated assemblies need to be accounted for since they are components of the materials flow from the reactor. Thus the safeguards approach could consist primarily of a surveillance regime with NDA on the access port to the in-sodium inventory for item counting purposes. Surveillance of the reactor containment structure may be required in order to ensure that alternative fuel routes are not introduced following the design verification inspections. Although a fast reactor may be shutdown for the core/breeder refuelling process, assemblies are likely to be transferred through the access port to the in-sodium stores on a more continuous basis so there is no possibility of closing that access port with safeguards seals during the major part of the year. Continuous surveillance is clearly required.

Monitoring the flow of items into and out of the inaccessible inventory permits the construction of a running book item inventory of that region. The safeguards value of a book inventory under routine operations is not clear since it cannot be compared in a timely way with the physical inventory; in fact, not until the reactor is decommissioned. If there is a total shut-down of the reactor with all fuel assemblies removed, due to some major fault conditions, then an estimate of the number and types of items to be removed from the inventory is clearly of accountancy value. In order to maintain effective monitoring of the flow of items through the access port there must be a very

low (zero) probability of failure of the surveillance and NDA monitor(s) and no ambiguity with respect to the types of items being monitored and to their direction of flow, since many items will contain more than one significant quantity of nuclear material. This places higher demands upon the reliability of instrumentation than has been demanded or achieved in the past. Alternatively it may require higher levels of redundancy in the equipment used for surveillance than has been the case in the past. Re-verification of the design information will probably require little if any additional demands above that required at other plants. Seals may have a role to play in this function.

Irradiated material being shipped from a reactor storage pond to a reprocessing plant in (say) multi-element bottles may well be regarded as inaccessible for periods in excess of the timeliness criterion. It would, therefore, seem to be equally important that items entering such an inventory are accounted for at the time of loading, with continuity of knowledge provided by seals on the transit containers. An NDA attribute test of sealed containers would appear to have no value. Verification of seals on the bottle lids would provide assurance of no tampering between the reactor and the receipt pond/head-end of the reprocessing plant. The principal significant difference between such an inventory and the fast reactor inventory is that the unloaded multi-element bottle can be verified as being empty much earlier than can the reactor core. The safeguards requirements are closely analogous to those of the thermal reactor core where verification is achievable but not within the timeliness criterion. The safeguards regime must rely upon containment and surveillance to provide a high level of assurance of the continuity of knowledge of the contents of the inventory.

Within Type C the scheme proposed by the Federal Republic of Germany for the long term dry storage of fuel assemblies make use of a variation of the multi-element bottles. Several irradiated fuel assemblies will be stored in each sealed container and the containers will be spaced in rows on a concrete floor. Once the containers have been so located there is no intention to move them for, perhaps, tens of years. The inventory of the storage will increase with time until all the container storage locations have been filled; thereafter the inventory should remain constant. No facilities are to be provided at the storage area to enable the containers to be opened in order that the contents can be verified. The contents should be verified in some way at the reactor prior to sealing for shipment to the storage area. The containers can then be treated as new items for the purposes of accountancy at the storage and the inventory verification can consist of items accountancy with integrity checks. In principle some attribute NDA might be possible on the new items, but the

purpose of such measurements is not clear. If pursued, some decay-time corrections will be required over the extended storage period. The integrity checks will entail inspection of any seals applied to the containers and this should provide sufficient assurance of the continued integrity of each new item.

The Agency appear to have a policy of reverifying parts of inventories which have been placed under seal for several years, partly to ensure that the containment has not been breached and that the seal has not been by-passed and partly to check the efficacy of the seal itself. Optical surveillance of the storage area should be able to provide additional assurance that no significant tampering has taken place and to identify the receipt and location of new items. This may avoid the need for partial re-verification of material under seal. In the case of dry storage there is again a need for the maintenance of a book inventory and a demand for a highly reliable surveillance system.

In most designs for the long term wet storage the fuel assemblies are transferred at the storage facility from shipping casks to long term storage racks or baskets and are accessible to some extent for inventory verification purposes [5] and are not relevant to the theme of this paper. Exceptions arise where baskets are stacked in such a way that some fuel is not readily accessible for inventory verification purposes. The use of seals on stacks of baskets whose contents have been verified, the use of surveillance techniques for monitoring flows into the storage pond and to detect any undeclared movements, and the use of underwater TV to enable visual access to the lower baskets for verification purposes should be capable of providing sufficient assurance of the physical inventory. The safeguards options will to some extent be facility specific, but again there is an emphasis on the need for a highly reliable system of containment and surveillance.

## 5. CLOSING REMARKS

We have identified examples of inaccessible inventories in each of three Types and conclude that most of them present no safeguards issues which require novel solutions. Within Type B, the core inventory of fast reactors and CANDU type reactors must be regarded as totally inaccessible but effective safeguards can be achieved by a combination of flow monitoring using installed NDA equipment for attribute measurements and containment and surveillance measures. Maintenance of a book inventory is only of value in this case in the event of a non-routine total clearance of the inaccessible inventory and at final decommissioning. Also in Type B is the short/medium term storage



and transport of irradiated fuel in multi-element bottles which can be regarded as a variant of the long term dry storage containers of Type C. For the long term storage it is necessary to verify the contents of the containers prior to their final closure and sealing and for timeliness reasons it may be necessary to do the same for multi-element bottles, although verification of their contents can be achieved later when the fuel assemblies are unloaded for reprocessing. In both cases, the containers should be treated as new items once they have been sealed; they can then be treated as accessible inventories to which standard safeguards techniques can be applied, namely item accounting and containment and surveillance.

Four key factors can now be seen to emerge in order to achieve adequate safeguards for inaccessible inventories:

- the need for effective monitoring of the flow of fuel assemblies
- the need for effective monitoring of shipments of fuel assemblies
- the need for highly reliable containment and surveillance regimes;
- the need for periodic re-verification of design information, particularly with respect to the containment structure of the reactor and of the long term storage facilities.

The containment and surveillance requirement must also imply that the measures are effective as well as reliable; effective in the sense that any undeclared but safeguards significant activities can be detected. Improved reliability of a surveillance system is normally considered to be achieved by improving the reliability of individual devices together with the introduction of additional devices to provide redundancy whilst avoiding common-mode failures. The reliability of a surveillance system might be further improved if some method can be devised for providing the facility operator with an indication that Agency devices have developed a malfunction. The operator could then advise the Agency in good time to effect a repair before malfunctions occur in other devices. This idea is contrary to the established principle that an operator should not be aware of a loss of continuity of surveillance, but it is often argued that an operator could easily detect the sounds of an operating camera or tape recorder, if he so wished, in order to check the operational status of the device.

De Montmollin [6] has suggested that the facility operator could contribute to a reduction in the frequency of inspections

by replacing data-tape packages and forwarding them to the Agency on a regular scheduled basis. Advising the Agency of equipment malfunctions would be an extension of this concept of greater cooperation between operator and the Agency. It is not suggested that all types of malfunction could or should be indicated to an operator, but many could. Recent containment and surveillance Advisory Group meetings have indicated the feasibility of using facility operator's CCTV systems for Agency purposes [7], since it would now appear to be possible to electronically authenticate the signals from the camera. If there is some method for authenticating that the view seen by the camera is the view agreed by the inspectors to be covered by the camera, then it might be possible for the Agency to make use of the operator's surveillance cameras in addition to their own devices. This could ensure early detection and correction of malfunctions.

This concept of greater operator involvement in surveillance functions may not be quite as simple to implement as is suggested in this paper but the idea is worthy of closer study. Clearly it can function most effectively in a safeguards regime developed in a co-operative rather than a conflict environment. There is an added responsibility placed upon the operators in return for the reduction in the likelihood of a loss of surveillance. The outcome in the event of an operator being unable to (or failing to) observe and report a possible loss of continuity of surveillance needs to be considered. The possibility of such an event must be reduced to near zero.

There will be increasing demands upon the Agency's resources in terms of equipment and manpower for surveillance reviewing procedures. In the long term storage facilities there should not be a significant number of flask movements to be detected and it can be expected that the application of motion detection techniques either as a trigger for the surveillance devices or as a reviewing aid could help to reduce reviewing requirements. The use of more devices for increased system reliability should not produce proportionate increases in reviewing requirements. It should not be necessary to review all the data collected by all the devices unless there arises the need to resolve an anomaly.

The requirements for effective monitoring of the flow of fuel assemblies through the reactor and of shipments from the reactor imply that there will be a greater demand for inspection resources at fast reactors and at some thermal reactor irradiated fuel ponds where fuel assemblies are being placed into inaccessible inventories. The technological problems are not likely to be too difficult to solve; the CANDU fuel bundle counter [5], the NDA instrumentation on the Kalkar IIIS scheme [8] and the Swedish development for their long term wet storage facility [9] indicate possible technical solutions to the problem

of monitoring material flows. Verification and interpretation of the data will still require inspector effort.

The resources required for re-verification of design information should not be very great, being an additional function to routine inspections.

The authors conclude that there exist relatively standard methods to achieve effective safeguards at the various types of inaccessible inventories which will be subjected to Agency safeguards in the future. A high reliance will have to be placed upon effective containment and surveillance techniques using devices which have a low failure rate and with sufficient redundancy to reduce the probability of a loss of surveillance to an extremely small value. There will be a need to monitor item flows and installed NDA devices should be capable of contributing in such cases. "The most desirable combination of these measures is the one that permits the safeguards objectives to be achieved at acceptable costs and with minimum intrusion into routine plant operations"[2].

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# SAFEGUARDING AN AWAY-FROM-REACTOR STORAGE FACILITY (WET TYPE)

## *Concept and initial experience*

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### Abstract

#### SAFEGUARDING AN AWAY-FROM-REACTOR STORAGE FACILITY (WET TYPE): CONCEPT AND INITIAL EXPERIENCE.

In December 1985, the Zwischenlager für Abgebrannte Brennstäben (ZAB), a storage facility for WWER type spent fuel assemblies, went into operation in the German Democratic Republic (GDR). It is one of the first away-from-reactor storages to come under IAEA safeguards and is therefore of particular interest. The spent fuel is stored under water in open stainless steel baskets, which are also used as inserts in the transport container. After a description of the relevant safeguards design features, the development of the safeguards approach is discussed. Starting with an analytical investigation by the System Studies section of the IAEA's Department of Safeguards, practical experience and close co-operation between various sections of the Department of Safeguards, the SSAC (State's system of accounting for and control of nuclear material) and the operator, a safeguards approach was elaborated that promises to ensure the attainment of IAEA inspection goals with reasonable effort. Its main element is the ease of direct verification of spent fuel assemblies in their final storage position. The particular criteria for inspection goal attainment at this facility and the ways to meet them are discussed in detail. The GDR's offer to include this facility in its support programme for IAEA safeguards, e.g. as training ground for newly recruited inspectors, is highlighted.

### 1. INTRODUCTION

Contrary to previous expectations the reprocessing of spent fuel elements from nuclear power plants has not yet become general practice. Up to now, final decisions on reprocessing or direct disposal of spent fuel elements have not been made in any country. In view of this situation, the construction of interim storages became a necessity, since spent fuel ponds at nuclear power plants have usually a rather limited capacity. This also applies to the German Democratic Republic (GDR) where the storage ponds of the five operational nuclear reactors are already employed to capacity, and those of another six reactors under construction are only designed for about three years each of reactor operation. In designing the interim storage facility, the

option of wet storage in open baskets was chosen. From the safeguards viewpoint this offers the great advantage of maintaining the accessibility of the individual spent fuel assemblies to verification.

## 2. DESCRIPTION OF THE ZAB<sup>1</sup> STORAGE FACILITY FOR SPENT FUEL ASSEMBLIES

The ZAB<sup>1</sup> facility for storage of spent fuel assemblies was established at the site of the Bruno Leuschner Nuclear Power Plant (about 20 km north-east of the town, Greifswald) and put into operation in December 1985. It is thus one of the first away-from-reactor storages to come under IAEA safeguards, and represents a prototype of other facilities at the design stage, or under construction in various CMEA<sup>2</sup> countries. The spent fuel assemblies from the four WWER-440 units of the Greifswald nuclear power plant, as well as those from the WWER-70 of the Rheinsberg nuclear power plant, will be stored there for about ten years. In the future, the spent fuel assemblies of the WWER-440 units 5 to 8 of the Greifswald nuclear power plant, at present under construction, will also be stored there.

The ZAB facility is a separate building covering 3300 m<sup>2</sup>. Figure 1 shows the plan of it. It has rail and road connections with both the reactors at the Greifswald site and with the public communication network. The facility consists of a reloading area, a storage area, and several support areas of no relevance to safeguards. The *reloading area* includes:

- the rail corridor;
- the receiving section (the hatch between rail corridor and the receiving section is the only transfer path for spent fuel assemblies between the storage area and the outer world);
- the reloading section;
- the decontamination shaft;
- the decontamination room; and
- the repair section, a shaft for storing special instruments, and a room for technological inspections.

The *storage area* comprises three operational ponds and a reserve pond, all plated with austenitic material. For reasons of residual heat removal and radiation protection, the spent fuel assemblies are stored under water in stainless steel baskets containing 30 fuel assemblies each. Figure 2 shows a diagram of a fuel basket. These baskets serve both as inserts of the shipping container and are also used for the final transport, when the spent fuel is re-exported to the USSR.

The storage ponds are 21.8 m long and 8.3 m wide, and are loaded on either side of a transport passage with two rows each of baskets.

<sup>1</sup> ZAB = Zwischenlager für Abgebrannte Brennstäben.

<sup>2</sup> CMEA = Council for Mutual Economic Assistance.

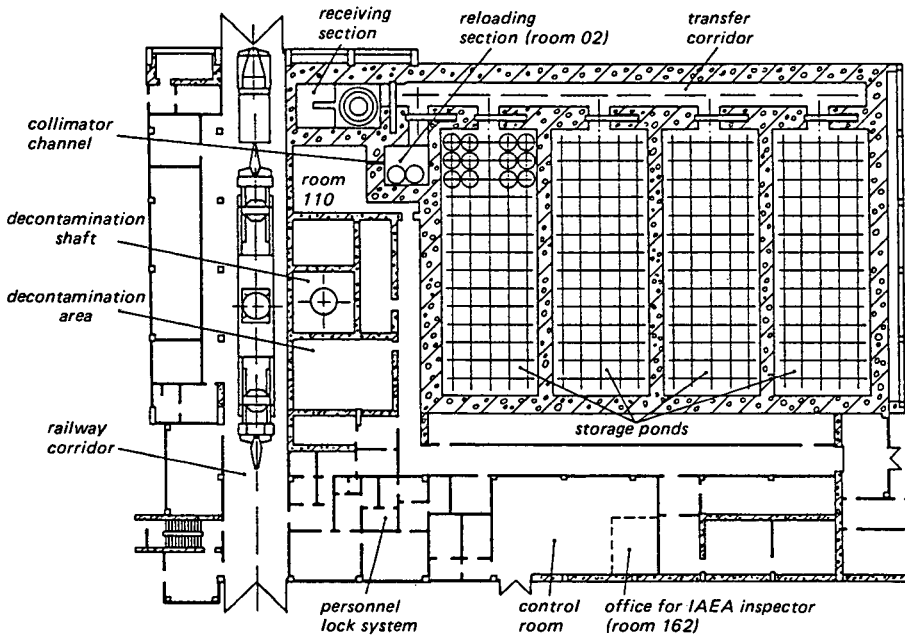


FIG. 1. Plan of the ZAB facility.

The storage capacity is designed for 52 baskets per pond, corresponding to 4680 spent fuel assemblies. The reserve pond accommodates a further 1560 spent fuel assemblies. Though not planned for the time being, it is technically possible to increase the storage capacity, both by constructing further storage ponds and by a higher density of packing. Two-layer storage, however, is not possible, as the ponds are only 7.20 m deep. The storage ponds are connected with the receiving section by a transfer corridor provided with water locks. For technical safety and radiation protection the ponds and corridors are covered with steel plates with slits along which the fuel baskets are transported by the gripper bars of the crane. For safety, the crane is interlocked in such a way that, during transport, the fuel baskets can be lifted only 0.5 m above the floor of the pond or the transfer corridor.

Normally, the fuel assemblies are placed in the reactor spent fuel pond in their final position within the fuel basket. This also applies to damaged fuel assemblies that are placed in hermetically sealed cans in the fuel baskets. If necessary, however, fuel assemblies or cans can also be reloaded into other fuel baskets in the reloading section of the ZAB facility. The reloading section has, in addition, a measuring position with a collimator, enabling measurements to be done at individual fuel assemblies from an adjacent shielded room. Some important physical and technical data of the spent fuel assemblies to be stored are summarized in Table I. The fuel assemblies are integral welded units which also retain their integrity during storage. They have a unique mark, hard to alter, enabling them to be identified in the storage position.

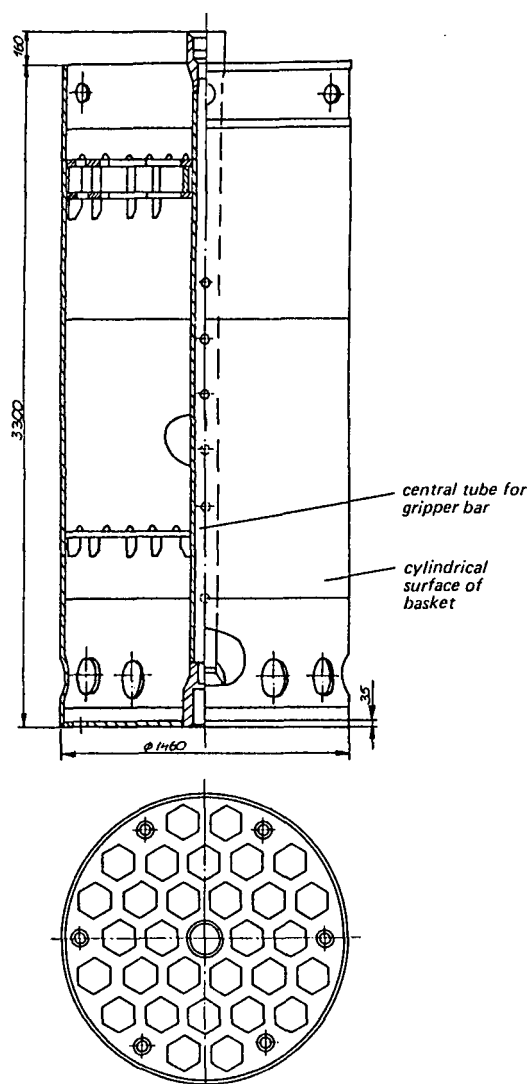


FIG. 2. Diagram of fuel basket for transporting and storing fuel assemblies.

Depending on the burnup and type of fuel assembly the plutonium content varies between 650 g and 1200 g, so that 6 to 10 fuel assemblies equal one significant quantity [1].

### 3. TECHNOLOGICAL PROCESS OF STORING SPENT FUEL ASSEMBLIES

Figure 3 is a sketch of receipt, handling and shipment of fuel assemblies. Spent fuel assemblies are transported to and from the spent fuel storage facility exclusively



TABLE I. PHYSICAL-TECHNICAL DESCRIPTION OF SPENT FUEL ASSEMBLIES

FA <sup>a</sup> type	K 1.6	R 1.6	K 2.4	R 2.4	R 3.6	O 4.2	K.2
Active length of FA (mm)	2320	2420	2320	2420	2420	2500	2400
Fuel rod dia. (mm)	9.1	9.1	9.1	9.1	9.1	10.2	10.2
Number of fuel rods per FA	126	126	126	126	126	90	90
U total (approx.) (g)	110 000	116 000	111 500	115 000	114 500	114 000	109 000
<sup>235</sup> U (approx.) (g)	750	900	750	800	1300	900	900
Average burnup (approx.) (MW·d/t U)	12 000	11 000	26 000	26 000	32 000	16 000	14 000
Pu total (approx.) (g)	700	700	1050	1100	1200	700	650
Maximum burnup (approx.) (MW·d/t U)	17 000	12 000	29 000	29 000	39 000	18 000	18 000

<sup>a</sup> FA = fuel assembly.

in shielding containers mounted on railway trucks. There are three types of shielding containers, but for each of them the same type of fuel basket is used as a transport insert for up to 30 fuel assemblies.

From the railway corridor the container is lifted by the 125 Mp<sup>3</sup> crane through the hatch and positioned inside the receiving section in the place where the cover is positioned. Figure 4 shows a shipping cask over the reloading area. The cover of the container, which in this position is still above the water surface, is loosened from a working platform. Thereafter, the container is lowered down to the reloading position, the cover is removed, and the fuel basket is moved by a gripper bar and the 15 Mp crane via the transfer corridor to one of the storage ponds, and put in the final storage position. The gripper bar can only be moved in the slits of the cover plates of the storage pond, which are uncovered for this operation. The operations in the receiving section are monitored by an underwater TV camera.

<sup>3</sup> Mp = megapond (1 p =  $9.807 \times 10^{-3}$  N).

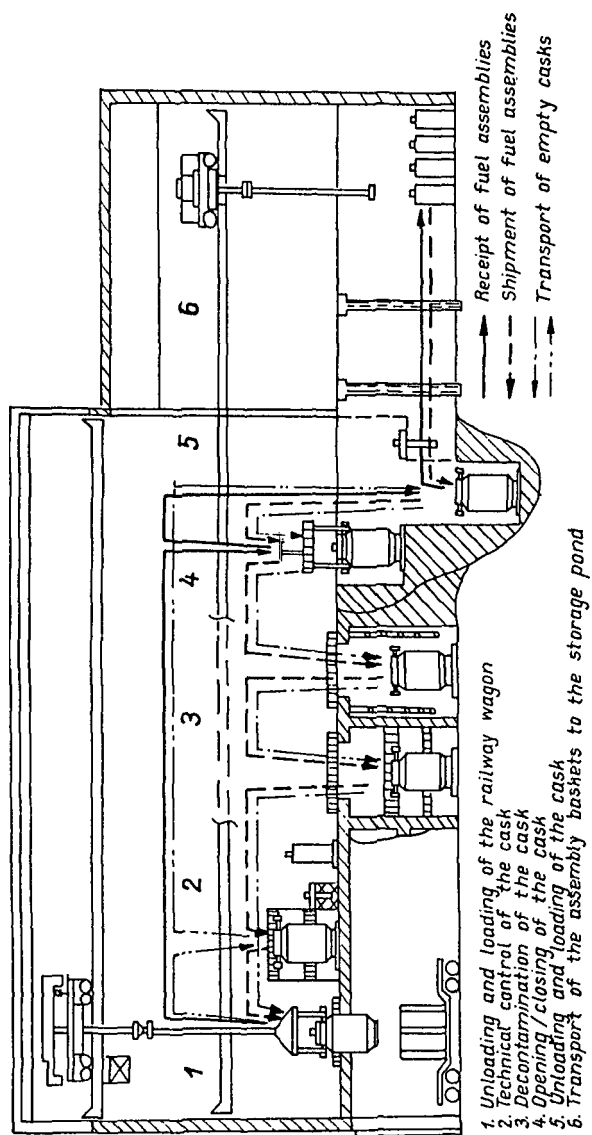
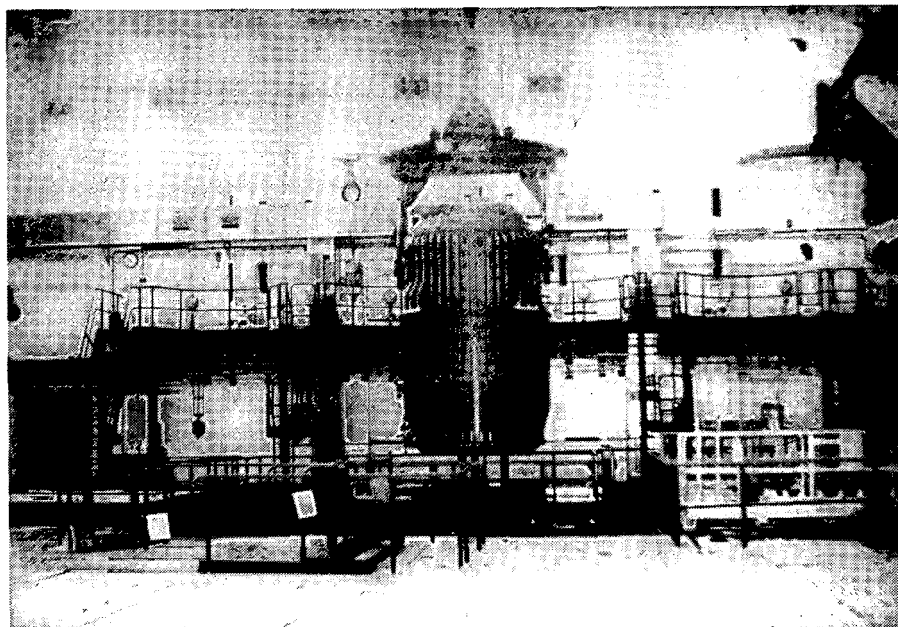


FIG. 3. Sketch of receipt, handling and shipment of fuel assemblies.



*FIG. 4. Shipping cask over the reloading area.*

The empty container is closed, decontaminated (if necessary, decontamination is repeated in the decontamination cell) and placed again through the hatch on the railway truck in the railway corridor. About 12 hours are required for the whole operation, whereas the fuel basket reaches its storage position within about five hours. The anticipated shipment of spent fuel assemblies out of the storage facility will need much longer (about four days), which is especially necessary for attaining a stationary thermal balance (ca. 88 hours), and for a final technological control.

The transport frequency of fuel assemblies depends on the number of reactor refuellings annually. With 90–120 fuel assemblies discharged per WWER-440 unit, the corresponding three or four fuel baskets must be expected for interim storage per reactor unit refuelling.

#### 4. GENERAL CONSIDERATION OF THE SAFEGUARDS APPROACH

In recent years the IAEA has paid great attention to the issue of safeguarding separate storage facilities for spent fuel assemblies. Various studies, advisory groups and other international meetings have dealt with this matter. The facility presented

in this paper served under the name AFRS NORD as the basis of a model for the design information for an away-from-reactor storage facility for spent fuel assemblies, presented by the GDR at an IAEA Consultants Meeting in December 1984 [2], and also as an example in an IAEA analytical study [3]. The design data provided to the IAEA for the ZAB facility largely correspond to the model mentioned. The analytical study was the starting point for elaborating the actual safeguards approach. Basically, two inspection regimes are discussed.

- (1) The IAEA inspector is present whenever spent fuel assemblies are shipped to the storage facility. He verifies the receipt and seals after termination of the storing operation, the storage position, all transfer routes and intermediate stations, as well as the transport container. Because of the relatively frequent shipment of transport containers this inspection regime is rather unrealistic and is therefore no longer considered.
- (2) The IAEA inspector is not present when spent fuel assemblies are shipped to, and stored in, the facility. During the next inspection he verifies the fuel assemblies received in the meantime and seals the storage position. Transfer routes and possible intermediate stations are not sealed.

The safeguards approach is then evaluated in the analytical study on the basis of the Safeguards Effectiveness Assessment Methodology (SEAM). This method, still in the development stage, compares the inspection activities with a systematically derived list of diversion paths and concealment methods, and analyses any indication of possible diversions (anomalies) detectable during the inspection activities. The designed safeguards approach evaluation is based on the detection probability for individual diversion paths. The detection probability is rated in six steps from zero to very high, taking into account three technical complexity levels of different diversion paths, from easy to very complicated.

There are 96 diversion paths, described by time, location, type of nuclear material and concealment method, facing 60 detectable anomalies if the following inspection activities are carried out:

- Examination of accounting records;
- Verification measures (item counting, identification, NDA measurement);
- Sealing of the storage pond cover after verification;
- Surveillance measures (film or TV cameras), including an additional underwater camera in the reloading section;
- Presence of an IAEA inspector during the receipt of hermetically sealed cans containing leaking fuel assemblies, which were transported from the shipping facility in a shielding container under IAEA seals, and which have been verified by an IAEA inspector before hermetic sealing.

According to the IAEA study a high (70–90%), or very high (90–100%), detection probability for almost all diversion paths considered will be achieved if these preconditions are fulfilled.

## 5. PRESENT STATUS OF THE SAFEGUARDS APPROACH

The example of the ZAB facility shows again that a realistic, reasonably achievable safeguards approach can only be brought about stepwise by taking into account practical experience. In this context the safeguards approach presented in Section 4 was subject to various modifications and refinements. Apart from practical experience, close co-operation between various units of the IAEA's Department of Safeguards, the State System of Nuclear Material Control of the GDR, and the operator, was required. Fully in keeping with the final declaration of the third NPT Review Conference, all parties considered the implementation of effective safeguards as the common objective to enable the IAEA to create and maintain confidence between States by the verified assurance that States use nuclear material exclusively for the declared peaceful purposes. In this sense the national regulatory body of the GDR is just as interested as the operator to take an active part in implementing technically credible safeguards through the IAEA, and to receive positive conclusions from the IAEA, arising from its inspection activities. Besides the designated IAEA inspectors from the responsible operations section, staff members of other divisions of the Department of Safeguards have shared in elaborating the current safeguards approach. Experience gained during several visits to the spent fuel storage facility was a valuable contributing factor.

As a yardstick for technically credible safeguards, one can consider the minimum uniform criteria for attaining the inspection goals. During the visit of representatives of the Safeguards Effectiveness Evaluation Section to the storage facility in May 1986, a relevant set of criteria for this facility was compiled and discussed in detail.

To the spent fuel storage facility the set of requirements for 'facilities other than reactors' applies. To attain the inspection goal, all the following inspection activities must be performed:

- (1) Examination of accounting records and their comparison with the reports to the IAEA have to be performed to the end of the last material balance period;
- (2) The accountancy verification goal is to be set to 1 SQ (significant quantity, see Ref. [4]);
- (3) Spent fuel assemblies received from other facilities within the same State have to be verified by non-destructive analysis in the attribute mode (e.g. with the Cerenkov viewing device). Identification of the serial number will suffice for integral, welded fuel assemblies if, as in this case, they have not been produced in the same country.
- (4) For direct-use nuclear material (including Pu in spent fuel assemblies) two complete physical inventory verifications a year are necessary. The second physical inventory verification can be replaced by an interim inventory verification, or omitted in the case of successful application of containment and surveillance measures throughout the year.

(5) Inventory verifications must meet the following conditions:

- The quantity for which no operator's data are available must not exceed 0.3 SQ;
- The non-verified quantity must be less than 1 SQ;
- At least the methods indicated in (3) must be applied (i.e. item counting and identification for welded fuel assemblies or qualitative NDA measurement).

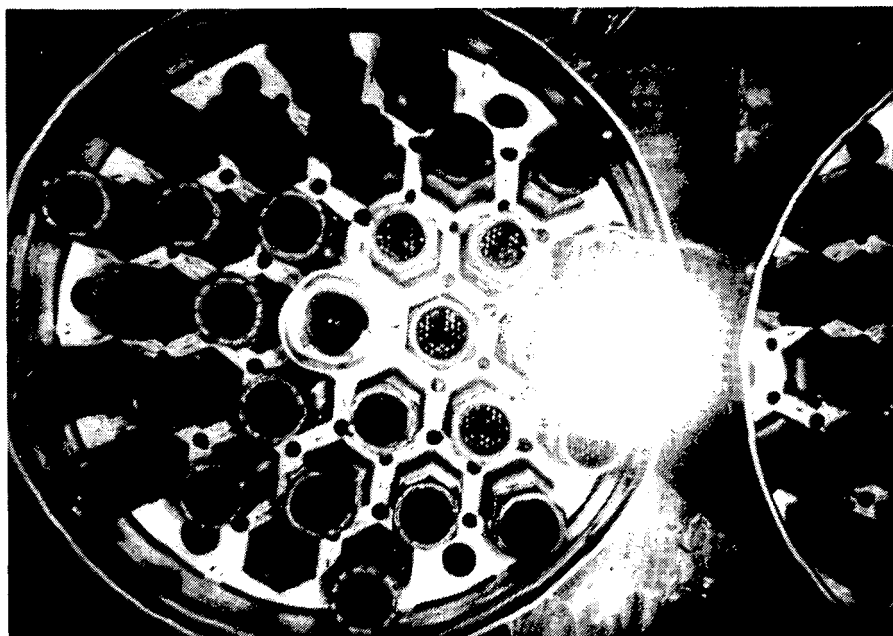
If containment or surveillance according to (4) is applied instead of material verification, the prescribed verification measures must be taken before, or at the latest, three months after, the attachment of the seal or the beginning of the surveillance period. Seals are to be replaced on a random basis, and material sealed for more than a year is also to be reverified on a random basis. In the case of successful surveillance, item counting is regarded as adequate reverification.

- (6) If other facilities in the same State handle more than 0.3 SQ of the same type of nuclear material, at least one simultaneous inspection per year of all these facilities is required in order to exclude mutual borrowing of material, unless the application of containment and surveillance measures excludes borrowing.
- (7) To attain the timeliness goal (detection time) of three months for spent fuel assemblies, the prescribed verification measures must be repeated for at least half the time intervals within this time, or the examination of the seals, or the review of surveillance films, must be finished within this time. In addition, discrepancies or anomalies exceeding 1 SQ must be resolved within these three months.
- (8) The receipt of spent fuel assemblies must be confirmed to the IAEA by the receiver within three months on an average.

Meeting all these somewhat complicated conditions is possible. It requires neither great IAEA inspection effort, nor extraordinary endeavours of the national regulatory body or the operator, as the following considerations indicate:

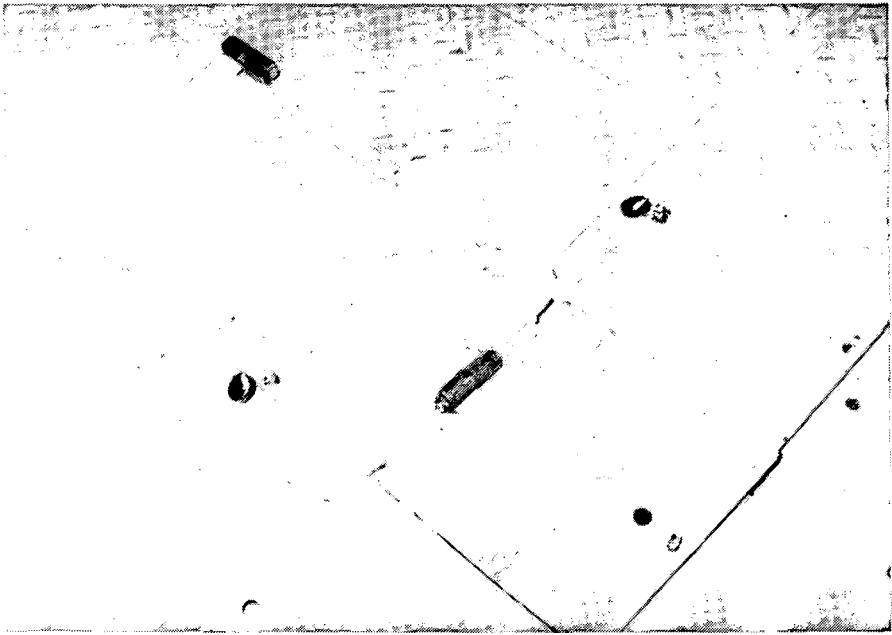
- (a) Correct keeping of the accounting records and timely reporting are a commitment that goes without saying.
- (b) The vertical storage of the spent fuel assemblies in open fuel baskets allows item counting, identification and NDA measurement (with the Cerenkov viewing device) of all fuel assemblies in their storage position.

Figure 5 shows fuel baskets in storage position through the slit in the storage pond cover. One can distinguish not only the individual fuel elements, but also individual fuel rods within the fuel assemblies surrounded by the bluish light of the Cerenkov radiation. Also, this should not be changed at more compact storage. Incidentally, verification is also possible under daylight conditions in the storage hall, because in the case of selective lifting of an individual cover, the storage pond still remains sufficiently dark to apply the Cerenkov viewing device.



*FIG. 5. View of fuel baskets with spent fuel assemblies in the storage position through the slit of the pond cover.*

- (c) Sealing, discussed earlier, of all storage positions, which would prevent a view into the storage pond and thus the inventory taking by the operator, was modified in favour of sealing by sectors the cover plates of the filled storage positions. Figure 6 shows IAEA seals applied to the transport slits. This makes occupied and verified storage positions inaccessible to the operator, but enables him to take inventories, and allows the inspector to perform complete or random verification without breaking the seals, thus avoiding the unnecessary effort of replacing a large number of seals.
- (d) Further considerations of the actual situation show also that the envisaged and extensively discussed camera surveillance does not seem to be essential. For timely detection at least four inspections a year must be made, and verification of the inventory and of inventory changes is possible relatively easily by direct verification supported by the adequate application of seals. Only the movement of the transport containers would be left to camera surveillance. However, camera surveillance alone provides only very limited information on the quantity and the direction of nuclear material movements. In addition, the lighting conditions are expected to vary widely because of daylight in the storage hall. This would considerably aggravate review of the surveillance films. Figure 7 shows the covered storage area with a gripper bar in the transport slit of the transfer corridor.



*FIG. 6. IAEA seals attached to the transport slits of the pond cover.*

- (e) Under the conditions as they exist in the GDR control over borrowing spent fuel assemblies from other facilities that handle the same type of material is easy, even without surveillance measures, since nearly all these facilities are located at one site, the Greifswald nuclear power plant, and thus simultaneous inspections are easy to carry out, and in the past have taken place almost automatically. The only separate site with comparable fuel assemblies, the Rheinsberg NPP, is situated between Berlin, the usual place of arrival and departure of the inspectors, and the ZAB facility, so that it can be verified almost at the same time en route.
- (f) The acknowledgement of receipts for international transfers of spent fuel assemblies does not belong solely to the sphere of influence of the national regulatory body or of the IAEA. If, however, the receiver fulfils the usual reporting obligations, this condition should also be always met.

## 6. FINAL REMARKS

The safeguards approach presented makes one hope that, under normal conditions and with appropriate planning of inspections, it will always be possible to attain inspection goals.



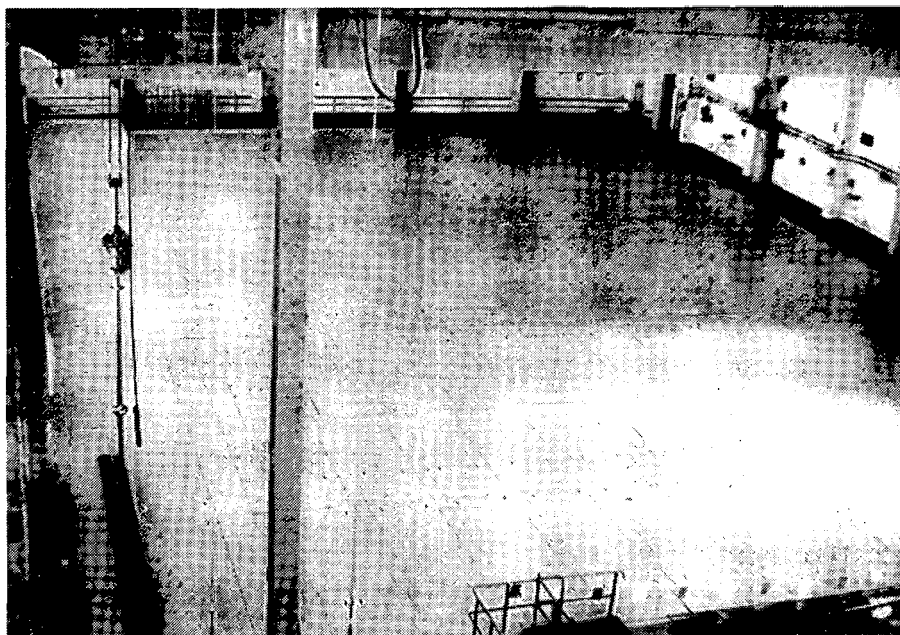


FIG. 7. View of the covered storage area in daylight with a gripper bar for fuel baskets in the transport slit of the transfer corridor.

Beyond the current safeguards approach and the inspection goal criteria the following questions may become of safeguards interest and could, as far as the operator is concerned, be accommodated in the inspection practice:

- (a) Verification of damaged fuel assemblies before they are hermetically encased, and subsequent attachment of seals, either in the reactor spent fuel pond or in the reloading section of the ZAB facility;
- (b) Special measurements of selected fuel assemblies (e.g. NDA of variables in attribute mode) in the reloading section of the ZAB facility;
- (c) Camera surveillance of the whole storage hall of the ZAB facility to prove that the fuel assemblies in their storage position did not become accessible by lifting the whole steel construction of the cover of the storage ponds.

Because of considerable general interest in nuclear material control at storage facilities for spent fuel assemblies, the GDR has included the ZAB facility in its support activities for IAEA safeguards. Thus, a course for trainees from developing countries was conducted in October 1986 at which simple verification methods, accompanied by an accounting exercise, were demonstrated at this interim storage facility for spent fuel assemblies.

It is planned to include verification and bookkeeping exercises at the ZAB facility in the eighth Comprehensive Inspection Exercise in February 1987 as part of the ICAS Module 12<sup>4</sup>, the final inspection exercise in training newly recruited IAEA inspectors.

Since, unlike a nuclear power plant, the interim storage facility is not immediately involved in production, the training activities and experiments serving the development of control methods can be carried out without interfering too much with the routine facility operation. In this respect, preliminary discussions have already taken place with the IAEA; however, specification of tasks and the creation of required technical preconditions have not yet started.

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<sup>4</sup> ICAS = Introductory Course on Agency Safeguards. Module 12: Comprehensive Inspection Exercise.

# LONG TERM STORAGE AND FINAL DISPOSAL CONDITIONING OF SPENT FUEL

## *Facility characteristics and safeguards aspects*

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### **Abstract**

#### LONG TERM STORAGE AND FINAL DISPOSAL CONDITIONING OF SPENT FUEL: FACILITY CHARACTERISTICS AND SAFEGUARDS ASPECTS.

The back end of the fuel cycle concept of the Federal Republic of Germany now includes, as a complement to reprocessing, the development and demonstration of final disposal techniques for spent fuel. In this way, together with intermediate dry storage, the Deutsche Gesellschaft für Wiederaufarbeitung von Kernbrennstoffen (DWK) has created two new facility types. These facilities require new safeguards concepts. Whereas, regardless of court decisions pending with respect to licensing, intermediate dry storage facilities are ready for operation (Gorleben), under construction (Ahaus) or at the design stage (Wackersdorf), a pilot conditioning plant for the final disposal of spent fuel is still at a conceptual stage. However, the licensing procedure pursuant to the Atomic Energy Act was opened in 1986. The paper describes the two types of facility in the fuel cycle and discusses basic aspects of the envisaged safeguards concepts with their facility specific features.

### **1. INTRODUCTION**

The long term storage of spent fuel is becoming more and more important to the nuclear fuel cycle. In the Federal Republic of Germany a decision has been reached in favour of dry storage. The first facility, located at Gorleben, is ready for operation, and a second facility is now under construction at Ahaus. Furthermore, a similar facility will be integrated into the Federal German industrial scale reprocessing plant at Wackersdorf, currently at the design stage.

The purpose of "away from reactor stores" is to shift the burden of long term storage away from spent fuel ponds in reactors and reprocessing plants to dedicated interim storage facilities. The "interim" character of these facilities is reflected in the limited licensing period of 40 years.

After completing extensive comparative studies, both of reprocessing and of direct final disposal, in January 1985 the Federal Government came out in favour of establishing a reprocessing facility and at the same time of further developing direct final disposal until ready for licence application /1/.

The task of the conditioning plant designed by the DWK in this connection is to package radioactive residues, which cannot be used in reprocessing either for technical or for economic reasons, ready for intermediate and final disposal.

## 2. TECHNICAL DESCRIPTION

### 2.1. Dry Storage Facility

All storage facilities already commissioned, currently under construction or at the design stage have a similar layout. The following description refers to the Gorleben Dry Storage Facility for spent fuel /2/.

The storage facility consists of a single hall measuring 182 m by 38 m and 20 m in height. The store is designed for a capacity of 420 casks or a maximum of 1500 t of uranium before burnup. The hall, shown in Fig. 1, is split into two functional areas:

- reception area, where the transport/storage casks are lifted from the transport vehicle after prescribed checks and safety procedures. In this part of the facility a maintenance area is provided for cask preparation prior to storage (or shipment) as well as for repairs in the event of leakage or sensor failure.
- storage area, where the casks are positioned by means of a 140 t crane for interim storage prior to reprocessing or further treatment.

The two areas are separated by a concrete shielding wall approximately 8 m in height with a sliding steel

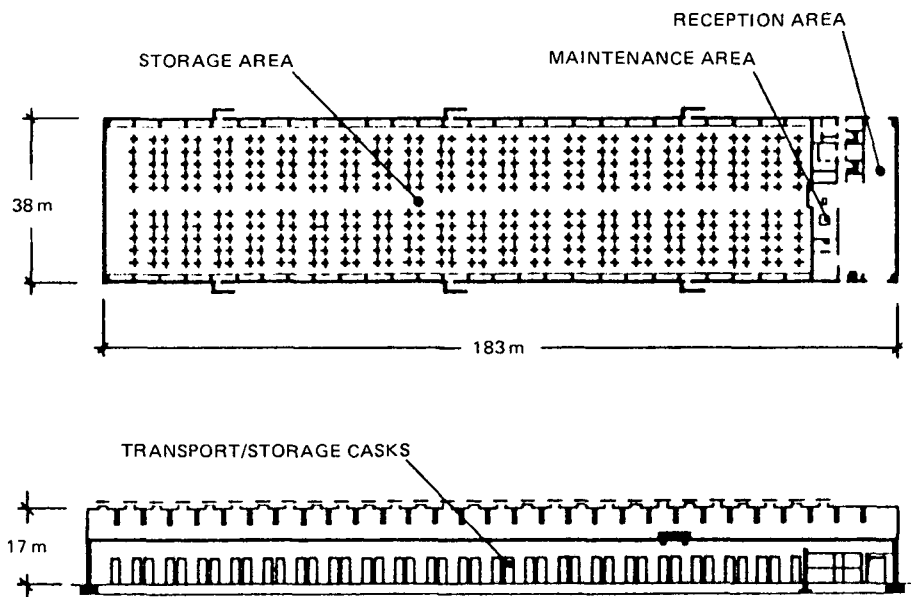


FIG. 1. Floor plan and longitudinal section of an interim dry storage facility.

door. The entrance (with two sliding doors for incoming and outgoing casks on heavy duty trucks, as well as for personnel access) is situated in the reception area.

The reception area consists of two floors with the following layout:

- ground floor: personnel entrance with health physics facilities and electrical distribution
- first floor: electrical distribution, air conditioning and ventilation room, cask maintenance room with measurement devices and auxiliary equipment storage area.

A travelling crane is mounted over the entire length of the hall and allows casks and equipment to be transported to any desired position. In the cask storage area the casks are placed upright on the base plate of the hall.

Vents are incorporated into the outer walls of the building and the roof structure allowing natural convection and ensuring passive removal of the radioactive decay heat.

### Loading, Transport and Storage Procedures

The following is a brief summary of the procedures involved upon arrival of a cask at the storage facility:

- unloading the transport vehicle: removal of protective cover and shock absorbers; tipping the cask into an upright position, depositing the cask in the maintenance position
- insertion of pressure sensor into the secondary lid
- leak test of the metal seal of the secondary lid as well as of the pressure sensor
- pressurization of the control volume between the primary and secondary lid to 6 bar with inert gas
- leak test of the secondary lid
- bolting on of a protective plate, fastening of connector and cable for the pressure monitoring system
- transport via overhead crane to the storage position in the storage area and connection to the pressure monitoring system

In the event of an alarm from the pressure monitoring system, indicating a leak, the cask is brought to the maintenance area. There the cask is investigated and repaired, if necessary, by welding on a protective lid.

Shipment of casks away from the storage facility involves the following activities:

- uncoupling from the pressure monitoring system and removal to the maintenance area
- disconnection of the monitoring system cable and removal of the protective plate
- lifting and mounting of the cask onto the transport vehicle, installation of shock absorbers and protective cover.

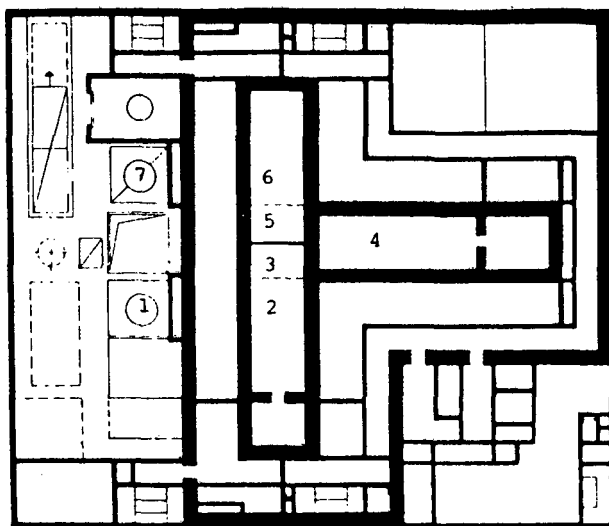


FIG. 2. Final disposal of spent fuel: Floor plan of the pilot conditioning plant. 1. Receipt of spent fuel in transport/storage casks. 2. Unloading cell. 3. Buffer store I. 4. Dismantling cell. 5. Buffer store II. 6. Loading cell. 7. Dispatch of final disposal casks.

## 2.2. Conditioning Plant

The planned facility described here is a pilot plant enabling all necessary operations in the field of fuel element and waste conditioning to be demonstrated on a representative scale. Furthermore, it is also envisaged that the procedures should be further developed and applied on an industrial scale.

The constructional and process engineering plans for the multipurpose conditioning plant are based on a maximum throughput of 35 t of heavy metal per year. However, the later operating programme assumes a lower throughput. The decisive factor in the order of magnitude, is the transferability of the technology to increased throughputs and the trial of a wide range of activities such as:

- the treatment and packaging of spent fuel elements and radioactive wastes in a form suitable for final disposal
- loading transport and storage casks with dismantled or chopped fuel elements in suitable packaging.

Further aspects include the production of final storage packages for other types of waste (i.e. not fuel elements), the reloading of waste packages not suitable for final disposal, as well as maintenance work on transport, storage and final disposal containers.

In the field of fuel element conditioning, the pilot plant will primarily treat those fuel elements not currently envisaged for reprocessing, e.g.

- LWR fuel elements of reprocessed uranium, certain mixed oxide fuels or with a very high burnup
- fuel elements from the high-temperature reactor
- core elements and special fuel elements from the breeder reactor.

The conditioning plant consists of a process building and several smaller auxiliary facilities.

The process building (see Fig. 2) serves the following functions:

- receipt of the incoming loaded or empty casks, dispatch of the outgoing casks and preparation of the casks for loading and unloading
- preconditioning, i.e. for LWR fuel elements: identification, dismantling, loading the fuel rods into bins, compaction of the structural components, loading the pressed pieces into baskets
- final conditioning, i.e. loading, sealing and examining the final disposal canisters.

There are also various other process steps connected with the conversion of the delivered radioactive waste into a form suitable for final disposal.

In the following, two examples will be selected from the broad range of operations at the multipurpose conditioning plant connected with international nuclear material safeguards.



(i) Packaging LWR fuel elements

- The transport cask is transferred into the facility, opened, unloaded, prepared for dispatch and transferred out again.
- The fuel elements removed from the transport cask are identified and placed in a buffer store.
- Control elements (PWR) or channels (BWR) are detached.
- Fuel elements are dismantled by separating the tie plates and withdrawing the fuel rods from their spacers in series.
- Structural components of the fuel elements are compacted.
- The fuel rods are loaded into bins and the compacted structural components into baskets.
- The baskets and bins are placed into final disposal canisters, the primary lid is put in position and bolted on.
- The secondary lid is put in position and welded, the protective lid is screwed in, the whole package is examined, transferred out of the facility and transported away.

(ii) Handling casks in the conditioning of high-temperature reactor fuel elements is largely identical to the treatment of LWR fuel elements described above.

However, upon delivery the HTR fuel elements are in cans.

- The can is removed from the transport cask, placed into a relocation device and opened.
- The fuel pebbles are pneumatically transported individually via a pipe into a final disposal canister.
- The empty can is then replaced into the transport cask and transferred out.

Further handling of the containers is carried out as described in the example of LWR fuel elements.

### 3. SAFEGUARDS ASPECTS

#### 3.1. General Remarks

Owing to the fact that the facility attachment for the first Federal German dry storage facility is still under negotiation and a safeguards system for the conditioning facility has not yet been designed, the present paper describes international safeguards aspects rather than complete approaches.

#### 3.2. Interim Dry Storage Facility

##### 3.2.1. Nuclear Material Accounting

It is envisaged that one material balance area will be established for the entire storage facility. The storage facility operator's accounting records are based entirely on shipper's (e.g. reactor) data as to the nuclear material contents of the transport/storage cask. An appropriate rebatching of shipped spent fuel assemblies (either at the reactor site or upon receipt at the store), in which a single cask is designated as one batch will facilitate accounting procedures.

Radioactive decay of nuclear material during the storage period will not be considered in accounting. Physical inventory taking is restricted to the counting and identification of casks with their associated contents.

##### 3.2.2. Inspection and Verification Activities

The most stringent design principle of the dry storage facility under consideration affecting accountancy verification is the inaccessibility of the nuclear material. Transport casks sent to the dry store cannot be reopened; this would make no sense operationally and thus no facilities are available for doing so. Therefore, in the entire facility neither direct verification of spent fuel assemblies (e.g. by serial number identification) nor non-destructive assay are applicable. Any safeguards approach will, necessarily, rely extensively upon containment/surveillance measures which enable unambiguous identification of the transport casks themselves and ensure continuity of knowledge about their contents. Consequently, the input verification of spent fuel entering the storage facility has to be done at the shipping facility. This situation is also unique with regard to the extent of the resulting inspection activities.

### Sealing of Casks and Optical Surveillance:

It is assumed that transport casks arrive sealed at the interim store in every case. A detailed discussion of the various possibilities of sealing, especially of the active participation of the reactor operator in sealing the casks, can be found in /3/.

It is proposed that the electronic seal system VACOSS /4/ be used in such a way as to guarantee the integrity of all spent fuel shipments. If the absence of inspectors during the loading phase or alternatively during arrival at the store is considered, it will become necessary to correlate the sealing procedure or seal detachment with some optical and/or electronic surveillance.

All transport/storage cask movements to and from the storage area must take place via the reception area of the interim store (Fig. 1). A permanently installed surveillance device (film or video camera) will suffice for verifying declared receipts or shipments of casks. Empty transport/storage casks are also intended to be stored temporarily next to filled ones in the storage area. Since there is no apparent difference between filled and empty casks (the empty ones are, of course, not subject to safeguards), conventional surveillance systems are not capable of making such a discrimination. Additional measures are therefore necessary.

Seals attached to the secondary lids of transport/storage casks during transportation are either of the VACOSS type or wire seals (if inspectors were present during loading). Seal detachment is required in cask preparation for long term storage. This will take place after removal of the protective cover and shock absorbers, either prior to lifting from the transport vehicle or after positioning in the maintenance area. In order to maintain continuity of knowledge after detachment of the secondary lid seal, the cask can be sealed to any appropriate equipment in the maintenance area. When storage preparation is completed, a seal attached to the protective plate on the top of the cask can provide assurance of integrity as well as of identity of the cask and its associated inventory. This seal can remain in position during the entire storage period. As for shipments of transport/storage casks from the interim store, the whole procedure will take place in reverse.

### Physical Inventory Verification:

Safeguards verification of physical inventory (and inventory changes) will be exclusively based on item counting and identification. To facilitate item identification, clearly readable serial numbers will be placed on the transport/storage casks. Moreover, the casks will be positioned in such a way that the serial numbers can easily be read from the corresponding corridors (see Fig. 1). As the radiation level within the storage hall is about 30 mrem/h<sup>1</sup>, visual item identification should take place according to a well-defined plan, perhaps on the basis of statistical sampling. An alternative to direct observation would be remote visual identification with the aid of a video camera attached to the overhead crane cabin via a telescoping arm.

### 3.3. Conditioning Plant

#### 3.3.1. Nuclear Material Accounting

The design and operation of the planned facility would suggest that two material balance areas should be established for safeguarding fissionable materials. One balance area comprises the storage area for nuclear fuels in incoming and outgoing transport or storage casks. A second MBA is formed by the process area in which the incoming fuel is mechanically processed until in a form ready for final disposal.

From the operational point of view, there is no need to determine the fissionable material by measuring and consequently no measuring devices are present. This means that the entire material accountancy must be based on data from the shipper, i.e. from a reactor plant or a fuel element interim storage facility. The incoming items which each form one accountancy batch are destroyed in the course of the process and converted into new batches each corresponding to the contents of one final disposal canister.

The accountancy procedure is very similar for HTR spherical fuel elements. In this case, the can forms the incoming batch unit, whereas the final disposal canister filled with approx. 8,500 pebbles - corresponding to approx. 4 cans - forms the outgoing accountancy unit.

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<sup>1</sup> 1 rem = 10<sup>-2</sup> Sv.

### 3.3.2. Inspection and Verification Activities

All loading and unloading processes and handling of the fuel take place by remote control in process cells but can nevertheless be observed at all important handling positions through windows in the cell walls.

Thus, for example, LWR and FBR elements can be identified in the familiar way by reading the serial numbers and optically examined with respect to their integrity. The storage positions of the individual fuel elements will probably only be visible during the loading and unloading process and concealed for the remaining time.

The destruction of a fuel element item by detaching the end section and withdrawing the pins thus also terminates the identifiability of the fuel in the further course of the process. Containment/surveillance measures thus necessarily replace material accountancy as the essential safeguards. Their application can be compared to the procedure in the headend area of reprocessing plants. Once again the basic principles are that the limits to be defined as the containment should be as close as possible to the material to be safeguarded, which is almost ideally applicable to the walls of the process cells. In spite of the necessary openings in the containment (ventilation, utilities, maintenance and repairs), there is no need for extensive C/S instrumentation if the strategic value of the material to be processed is taken into sufficient consideration. Since the diversion of even the slightest amounts of fissionable material would always require large volumes (due to the necessary shielding), or in the case of minimum quantities an extremely large number of diversion processes would have to take place before significant quantities had been collected, the very presence of inspectors during the handling of fuel represents an important component in the safeguards concept.

The material flow is channelled by structural features at key measurement points so that the optical monitoring (e.g. by TV systems) of the fuel input into the process cell and also the loading of conditioned fuel into the casks for further transport, particularly during the absence of inspectors, does not present any basic difficulties.

The plant operates on a seasonal basis corresponding to the demands made on it. A large number of operating periods serve to condition waste without fuel. During

this period the process MBA is not necessarily safeguarded as long as it can be assumed that the fuel from one operating period has been completely processed (i.e. conditioned) and that the physical inventory coincides with the end of the operating period. In this case inspection activities are restricted to verifying the empty state of the process area and if necessary to inventory taking in the cask storage area.

#### 4. COMPARATIVE EVALUATION

Neither type of plant - interim dry storage or conditioning facilities - represents any basically new problems with respect to safeguards.

Owing to the physical nature of the material and techniques applied, the nuclear material under safeguards is inaccessible at every stage of the operation. It is either stored inside shielded casks or handled in hot cells.

Dry working conditions and technical precautions render even criticality assay unnecessary. Therefore no measurement data are available and no operator equipment is installed for use together with the inspectorates.

Thus, both facilities have in common that neither the incoming nor the outgoing uranium and plutonium is determined by measuring techniques.

Neither is a shipper-receiver difference determinable with respect to quantities of fissionable material nor the evaluation factors for the balance: the material unaccounted for (MUF). The resulting necessity of using containment/surveillance methods therefore requires non-facility-specific safeguards concepts for both types of plant.

Rebatching is meaningful and should be applied in both types of facility. Rebatching during long term storage is rather a formal and temporary procedure in defining all fuel assemblies inside a single cask as one batch. In contrast, spent fuel in the conditioning facility loses its identity at the moment of disassembly. Since the final disposal canisters contain rods stemming from several fuel assemblies a new item for accountancy purposes has to be established once and for all.

It should also be noted that this new item can no longer be reverified at any time with respect to the inventory.

This is a special situation in the safeguards sector which must be taken into consideration in final disposal following the conditioning step /5/.

## 5. CONCLUDING REMARKS

Long term dry storage for special fuel is ready for operation and only court orders have prevented it from coming into use. A high priority is therefore attached to working out the safeguards concept and establishing the particular safeguards provisions and the facility attachments. Since the major objective for the conditioning plant is first of all an examination of licensability under the Atomic Energy Act and a final construction decision has not yet been made, an opportunity is thus presented of examining the safeguards aspects very meticulously and of including them in the plans at an early stage. The earliest possible commissioning date is envisaged for the late nineties and thus enables the introduction of the most modern C/S technology, such as the communication of electronic seals with TV systems, image processing and similar procedures. All this is included in the German programme in support of the IAEA /6/.

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## **КОНСТРУКЦИОННЫЕ И ТЕХНОЛОГИЧЕСКИЕ ОСОБЕННОСТИ ДОЛГОВРЕМЕННЫХ ХРАНИЛИЩ ОТРАБОТАВШЕГО ТОПЛИВА БАССЕЙНОВОГО ТИПА**

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### **Abstract—Аннотация**

#### **DESIGN AND ENGINEERING FEATURES OF POOL TYPE LONG TERM SPENT FUEL STORAGE FACILITIES.**

The paper gives the essential data on the purpose and main features of the USSR design for a pool type long term spent fuel storage facility to take and store the fuel from WWER-440 type power reactors. Matters relating to the layout of the store and the use of the main items of equipment are discussed. The operating regimes of the store are examined along with the main transport and process operations on the spent fuel. Special attention is given to those structural and process design elements which were included specifically to facilitate the application of the IAEA safeguards. Recommendations are made for increasing the efficiency of IAEA safeguards application to this type of storage facility.

#### **КОНСТРУКЦИОННЫЕ И ТЕХНОЛОГИЧЕСКИЕ ОСОБЕННОСТИ ДОЛГОВРЕМЕННЫХ ХРАНИЛИЩ ОТРАБОТАВШЕГО ТОПЛИВА БАССЕЙНОВОГО ТИПА.**

В докладе приводятся необходимые данные о назначении и основных характеристиках советского проекта долговременного хранилища отработавшего топлива (ДХОТ) с бассейновым способом хранения, который используется для приема и хранения топлива с АЭС с реакторами типа ВВЭР-440. Обсуждаются вопросы компоновки хранилища и использования основного оборудования. Рассматриваются режимы работы ДХОТ и основные транспортно-технологические операции с отработавшим топливом. Обращается особое внимание на те элементы конструкции и технологического процесса, которые были специально внедрены в проект для облегчения осуществления гарантий МАГАТЭ на ДХОТ. Даны рекомендации по повышению эффективности применения гарантий МАГАТЭ для рассматриваемого типа хранилищ.

В настоящее время государства с развитой ядерной энергетикой ориентируются на длительные сроки хранения отработавшего топлива АЭС до 10–20 лет [1].

Проблема длительного хранения топлива решается путем строительства крупномасштабных хранилищ для долговременного хранения отработавших тепло-выделяющих сборок (ОТВС). ДХОТ становится одним из новых элементов ядер-

ного топливного цикла и на него в полной мере будут распространяться гарантии МАГАТЭ.

Появляющийся опыт в области разработки ДХОТ в ряде стран свидетельствует о возможности использования различных способов хранения (в специальных контейнерах, в горных массивах, в водных бассейнах и др.). В зависимости от выбора способа хранения принимаются принципиально различные конструкционные решения для ДХОТ, которые в свою очередь оказывают непосредственное влияние на технологические операции с ядерным материалом и обращение с ним.

Советский проект ДХОТ по ряду конструктивно-технологических особенностей отличается от других подобных установок [2], на которых применяются гарантии МАГАТЭ к облученному топливу. Различия в способах, сроках хранения отработавшего топлива, технологии получения и отправки такого топлива определяют различия в подходе по применению на них гарантий МАГАТЭ.

Обсуждаемый тип долговременного хранилища предназначен для длительного хранения под водой ОТВС от реакторов ВВЭР-440. Емкость хранилища рассчитана на прием всего отработавшего топлива из четырех реакторов АЭС в течение 10 лет, что соответствует приблизительно 5000 ОТВС.

Перевозка ОТВС и их хранение осуществляется в штатных транспортных чехлах. Перевозка чехлов со сборками из реакторных блоков АЭС в долговременное хранилище осуществляется с помощью контейнера после трехлетней выдержки их в приреакторных бассейнах.

Емкость чехла составляет 30 герметичных или 18 дефектных ОТВС. Дефектные ОТВС размещаются в чехле в индивидуальных герметичных пеналах. ОТВС реактора ВВЭР-440 содержит приблизительно 1200 г плутония [3]. Таким образом, чехол с герметичными ОТВС включает в себя четыре значимых количества ядерного материала прямого использования по плутонию.

В чехле ОТВС располагаются в вертикальном положении по треугольной решетке.

Ежегодный вывоз из каждого реактора составляет 120 ОТВС (одна перегрузка реактора), что соответствует емкости четырех чехлов. Таким образом, в ДХОТ поступает ежегодно 16 чехлов.

В хранилище ОТВС располагаются в отсеках хранения в чехлах под защитным слоем воды толщиной около 2,8 метров.

ДХОТ представляет собой отдельно стоящее здание, состоящее из следующих основных отделений (рис. 1, 2):

- отделение перегрузки контейнеров;
- отделение хранения ОТВС;
- отделение технологического обеспечения ХОТ.

Отделение перегрузки контейнеров включает в себя (рис. 2):

- транспортный коридор (помещение 101);
- зал отделения перегрузки (помещение 202);
- отсек приема контейнеров (помещение 01);
- шахты стендов дезактивации;

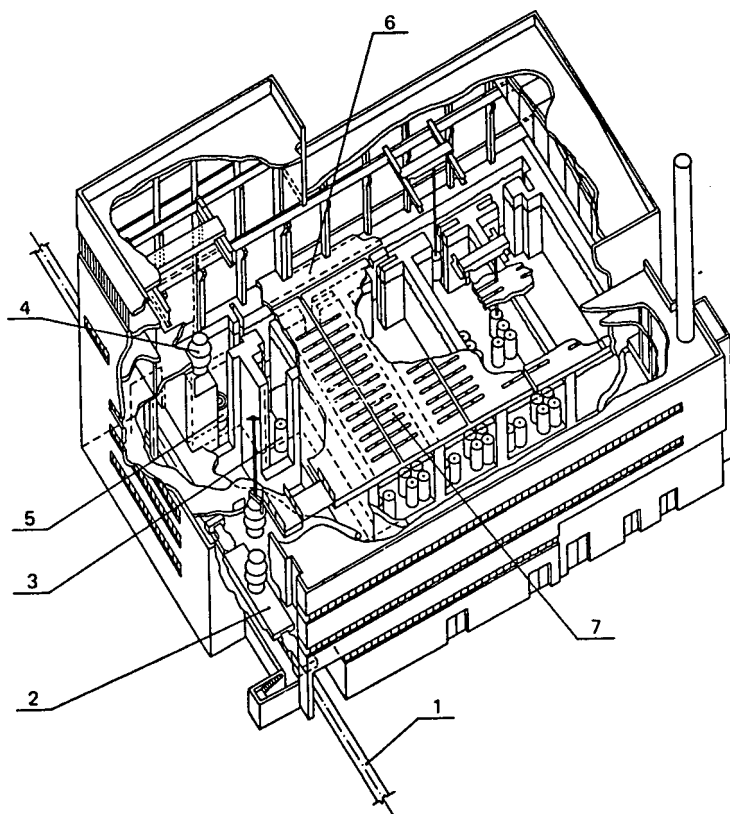


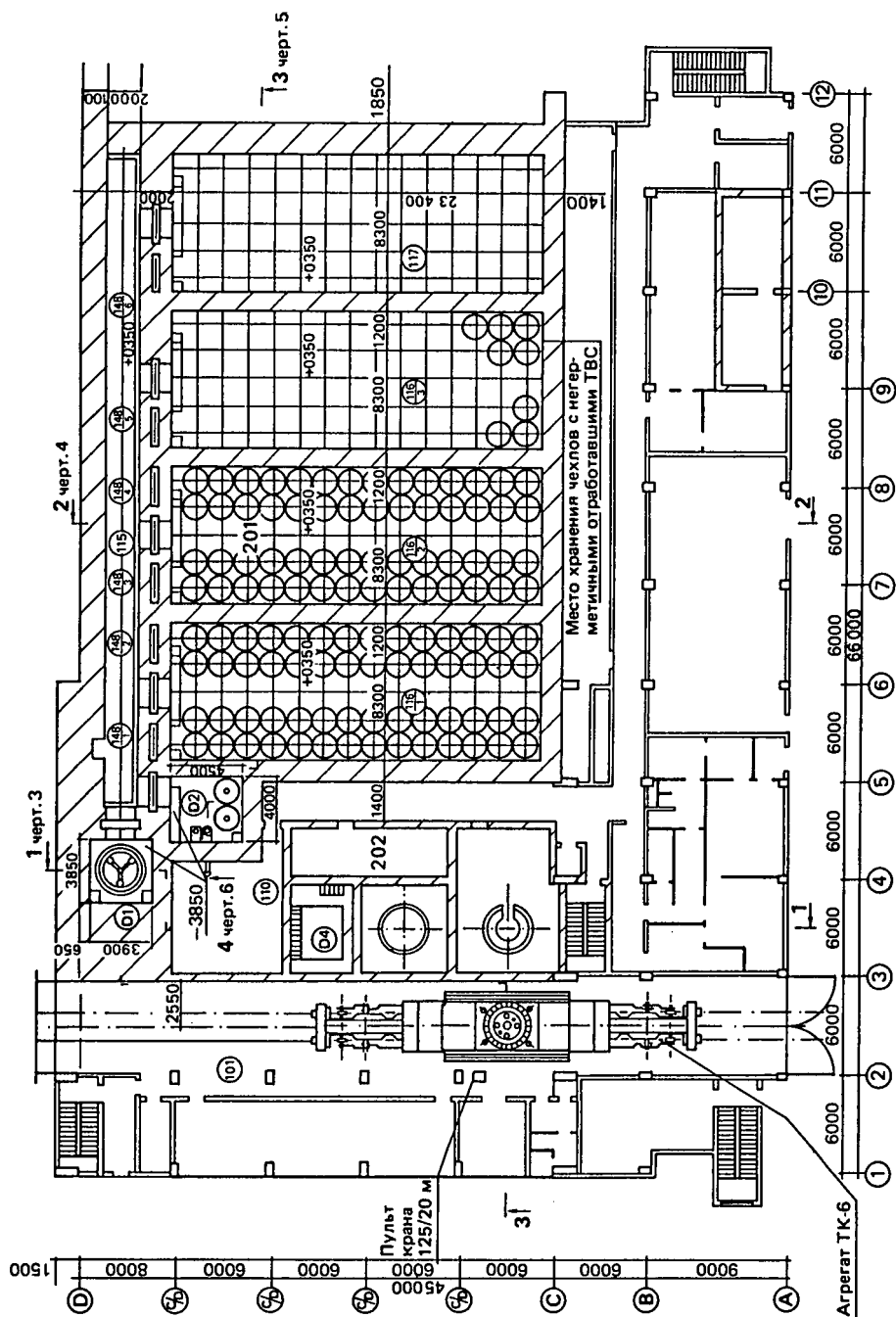
РИС. 1. Общий вид долговременного хранилища отработавшего топлива: 1 – транспортный коридор; 2 – агрегат ТК-6; 3 – зал отделения перегрузки; 4 – отсек приема контейнеров; 5 – стенд технологического контроля; 6 – передаточный коридор; 7 – отделение хранения ОТВС.

– шахту хранения инструмента (помещение 04).

Для выполнения технологических операций отделение перегрузки контейнеров оснащено следующим оборудованием (рис. 3):

- краном специальным 125/20 т;
- траверсом для контейнера;
- штангой с подсветкой;
- площадкой передвижной;
- гайковертом;
- стендом технологического контроля;
- внутристанционным контейнером;
- захватами для сборок и пеналов;
- другим оборудованием и приспособлениями.

Отделение хранения ОТВС включает в себя:

РИС. 2 ДХОТ. План на отм.  $\pm 0,000$ .

- отсеки хранения отработавших ТВС, 3 рабочих и 1 резервное помещения 116/1—3 и 117);
- передаточный коридор (помещение 115);
- отсек перегрузки сборок (помещение 02);
- зал отделения хранения (помещение 201).

Для выполнения технологических операций отделение хранения ОТВС оснащено следующим оборудованием (рис.3):

- краном специальным 16 т;
- штангой с подсветкой;
- гидрозатворами;
- копирами;
- чехлами для ОТВС.

Отделение технологического обеспечения расположено в обстройке здания и состоит из:

- спецхимводочистки и систем охлаждения воды;
- системы вентиляции;
- водоснабжения и канализации;
- других систем поддержания и обеспечения технологических условий работы хранилища.

Внутристанционный контейнер, загруженный чехлом с ОТВС в реакторном блоке АЭС, перевозится в хранилище транспортным средством.

Краном 125/20 т и траверсой контейнер снимается с транспортного средства, поднимается в зал перегрузки и спускается в воду отсека приема на отметку + 3,60 м.

Производится раскрепление крышки контейнера.

Контейнер с раскрепленной крышкой переставляется с отметки + 3,60 м на отметку — 3,85 м отсека приема.

Штангой с подсветкой на крюке 20 т кран 125/20 т снимается с контейнера крышка и устанавливается на отметку + 3,60 м. Краном 16 т и штангой с подсветкой из контейнера извлекается чехол с отработавшим ТВС и по передаточному коридору под водой переносится в загружаемый отсек, где устанавливается на место хранения.

В разгруженный контейнер краном 16 т устанавливается порожний чехол, краном 125/20 т на контейнер устанавливается крышка. Контейнер краном 125/30 т поднимается с отметки — 3,85 м и на отметку + 3,60 м, где производится закрепление крышки на контейнере.

Краном 125/20 т контейнер снимается с отметки + 3,60 м и поднимается из отсека приема в зал перегрузки. При подъеме производится обмывка контейнера водой. Отмытый контейнер подвергается радиационному контролю. При необходимости контейнер подается в шахты дезактивации для дополнительной отмывки.

Контейнер краном 125/20 т переносится к люку в транспортный коридор.

Открывается крышка люка. Контейнер опускается в транспортный коридор, устанавливается на транспортное средство и отправляется в реакторный блок АЭС за очередным чехлом с отработавшими ТВС.

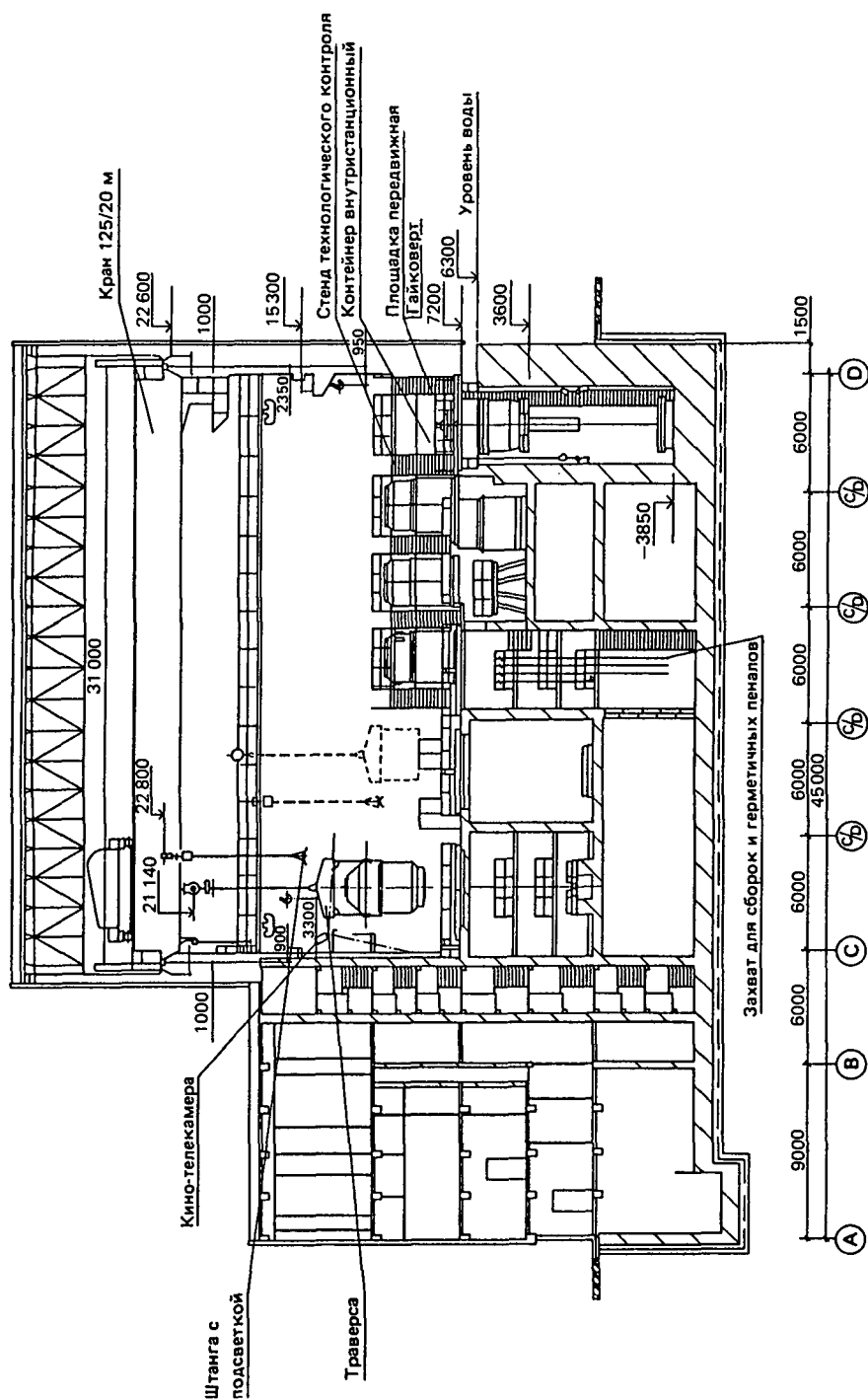


РИС. 3 ДХОТ. Разрез 1 - 1.

Чехлы располагаются на дне отсеков с определенным шагом. Шаг расстановки определяется расположением транспортных щелей перекрытия отсеков и размером (длиной) откидных крышек на щелях.

Каждый отсек рассчитан на размещение в нем 56 чехлов. Каждое место чехла в отсеке на перекрытии имеет нумерацию с 1 по 56.

В процессе хранения ОТВС проведение каких-либо транспортно-технологических операций с ними в хранилище не требуется.

За время нахождения чехлов с ОТВС в хранилище (~10 лет) могут быть осуществлены 2 перестановки чехлов из отсеков хранения в резервный отсек для проведения профилактического ремонта и очистки отсеков (один раз в 5 лет каждый отсек).

Вывоз отработавших ТВС из хранилища производится транспортным контейнером ТК-6.

Транспортный контейнер с порожним чехлом подается в транспортный коридор хранилища, откуда он подается в зал перегрузки и устанавливается на стенд технологического контроля, где производится входной контроль.

Проверенный контейнер устанавливается в отсек приема на отметку + 3,60 м, где производится раскрепление крышки и ее снятие с контейнера.

Из контейнера извлекается порожний оборотный чехол. Контейнер без чехла с отметки + 3,60 м переставляется на отметку — 3,85 м.

Чехол с ОТВС, намеченный к отправке, краном 16 т транспортируется с места его хранения и загружается в контейнер. На контейнер устанавливается крышка.

В случае невозможности установки чехла в контейнер (повреждение чехла, деформация и т. д.) производится перегрузка ОТВС из дефектного чехла в исправный. Эта операция производится в отсеке перегрузки сборок.

Контейнер поднимается на отметку + 3,60 м, где гайковертом производится закрепление крышки, после чего контейнер извлекается из приемного отсека, обмывается и устанавливается на стенд технологического контроля.

На технологическом стенде контейнер, загруженный чехлом с отработавшими ТВС, выдерживается до выхода температуры и давления внутри его на стационарный уровень.

Подготовленный к отправке контейнер краном 125/20 т. и траверсой снимается с технологического стенда и через открытый люк устанавливается на агрегат ТК-6, находящийся в транспортном коридоре и вывозится из хранилища.

В соответствии с назначением и принятым технологическим процессом работа хранилища предусматривается в следующих технологических режимах: прием отработавшего топлива; хранение отработавшего топлива; отправка отработавшего топлива.

Хранилище рассчитано на одновременный прием и технологическое обслуживание только одного загруженного контейнера.

Хранение отработавшего топлива является технологическим процессом, независимым от режима приема и отправки, и обеспечивается соответствующим размещением чехлов с отработавшими ТВС в отсеках хранилища и поддержанием в них и

других помещениях хранилища необходимых технологических режимов и параметров.

Технологические системы обеспечения хранения отработавшего топлива работают без присутствия персонала в хранилище. Основные контрольные параметры – превышение предельных уровней отсеков, температура воды и радиационная обстановка в хранилище, передаются на пульт ДХОТ.

На технологическом стенде контроля параметров может накапливаться до четырех загруженных контейнеров.

В соответствии с технологическим процессом и техническими характеристиками транспортно-технологического оборудования, время, затрачиваемое на прием, загрузку и отправку контейнера при двухсменной работе составляет 16 часов. С учетом выхода контейнера на стационарный режим, общее время пребывания контейнера в хранилище составляет 72 часа, а прием и отправка загруженных контейнеров будет осуществляться с интервалом 30 часов.

Для отправки в течение года 16 загруженных контейнеров потребуется примерно 18 суток и 16 суток для их приема.

Исходя из вышеизложенного, работа персонала по обслуживанию хранилища носит периодический характер и в общей сложности составляет примерно 34 суток в год.

В проекте ДХОТ для АЭС с реакторами типа ВВЭР-440 можно выделить следующие особенности конструкции, облегчающие применение гарантий:

- (а) хранилище представляет собой замкнутое сооружение, ограниченное трудно-разрушаемыми физическими барьерами;
- (б) хранилище имеет один вход–выход через транспортный проем в отделении перегрузки, изъятие ядерного материала через другие проемы (двери, люки) – невозможное;
- (с) штатное транспортное оборудование спроектировано таким образом, что чехол со сборками нельзя удалить из бассейна хранения;
- (д) возможен доступ инспекторов к местам хранения отработавшего топлива в течение всего времени хранения, благодаря тому, что:
  - хранение ОТВС в бассейнах ДХОТ осуществляется в один слой;
  - ОТВС хранятся в чехлах, конструкция которых не мешает оптическому наблюдению верхней части сборок и их идентификации без выемки из чехла;
  - ОТВС имеет неразборную конструкцию и трудноизменяемый заводской номер, выгравированный на верхней ее части;
  - щелевая конструкция перекрытия отсеков в зоне хранения, а также возможность вращения чехлов вокруг вертикальной оси позволяют идентифицировать сборки без их извлечения из чехлов;
  - проект предусматривает обеспечение прозрачности воды, достаточной для подсчета и идентификации ОТВС;
- (е) в отсеке перегрузки предусмотрена возможность извлечения отдельных ОТВС из чехла и проведения измерений с помощью методов неразрушающего анализа, для чего предусматривается установка коллиматора, блоков детектиро-



вания, устройства для вертикального перемещения ОТВС, а также аппаратуры для определения наличия ядерного материала по высоте ОТВС;

(f) имеется возможность опечатывания крышек щелевого перекрытия в секциях отсеков хранения, гидрозатворах, люках над транспортным коридором;

(g) имеется возможность использования средств наблюдения (кино- и телекамер), обеспечивающих обзор вероятных маршрутов транспортировки ОТВС, смонтированы специальные телекамеры, производящие периодический осмотр помещения хранения;

(h) предусмотрена возможность нормального и аварийного освещения на участках залов хранения, наблюдаемых с помощью кино- и телекамер;

(i) предусмотрено применение телевизионной системы с подсветкой для идентификации и подсчета ОТВС в чехлах под слоем воды;

(j) обеспечено наличие картограмм и маркировки расположения чехлов с ОТВС в бассейнах хранилища и сборок в чехлах.

В качестве рекомендаций по дальнейшему повышению эффективности применения гарантий на ДХОТ можно предложить:

- автоматизированную систему учета ядерного материала с применением персональных компьютеров и представлением результатов на магнитной ленте в требуемой МАГАТЭ форме;

- применение нового поколения опечатывающих устройств, позволяющих их многократное использование;

- установку следящих мониторов, позволяющих регистрировать перемещение и осуществлять подсчет чехлов ОТВС по транспортному каналу между отсеками перегрузки и бассейнами выдержки;

- дистанционный сбор информации, поступающей от следящих мониторов и опечатывающих устройств с помощью автоматической системы контроля на базе ЭВМ.

Анализ советского проекта ДХОТ показал, что предлагаемая конструкция удовлетворяет требованиям, способствующим облегчению применения гарантий на ДХОТ.

Осуществление контроля в соответствии с процедурами и с использованием технических средств может позволить МАГАТЭ обеспечить надежный контроль ядерного материала для этого типа хранилищ.

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## **FUTURE IAEA SAFEGUARDS INFORMATION PROCESSING**

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### **Abstract**

#### **FUTURE IAEA SAFEGUARDS INFORMATION PROCESSING.**

The IAEA Safeguards Information System (ISIS) has been continuously expanded and enhanced since it was put into production in 1981; several new components have been introduced to meet the growing needs of safeguards. At present, ISIS is the principal information resource for the Department of Safeguards. Changes in safeguards operations requirements, the expansion of scope and flow of information, and new data processing technologies require continuous development and further exploitation of information technologies for safeguards. The paper describes the present ISIS — its main functions, the flow of information, the ISIS architecture and the ISIS subsystems. Justifications for a review of the current system are provided; areas of necessary revision required for further integration of the major ISIS subsystems are defined. New data processing technology, especially microcomputers, and its impact on future ISIS development are considered; a future approach of information processing during inspections is proposed.

### **1. INTRODUCTION**

The current IAEA Safeguards Information System (ISIS) has been in operation since January 1981 and has therefore expended about half of the life span of ten years commonly accepted for large corporate information systems. Its architecture was conceived and designed in the late 1970s to cope with anticipated growth in the amount of safeguards information to be processed and stored. Reports on this system have been given previously [1-5].

At present, ISIS is the principal information resource for the Department of Safeguards. Changes in safeguards operations requirements, the expansion of scope and flow of information, and new data processing technology require continuous development and further exploitation of information technology for safeguards. An information system meeting the current and foreseeable needs to the greatest extent possible will assume a central role in the department and will therefore influence significantly the effectiveness and efficiency of the department's operation.

## 2. CURRENT SYSTEM

### 2.1. Information flow

Information collected by the Agency is the product of the implementation of agreements with Member States and the result of procedures within the Agency itself. In general, the Member States supply accounting and facility design information. Information from the inspectorate, analytical laboratories, evaluation and management are supplied by the Agency. The basic users of the Agency safeguards information are the Member States, the Board of Governors and the Agency. Of these users, the Agency utilizes the largest portion of information, specifically to carry out its safeguards function. In accordance with official agreements, the Member States are regularly provided with information which includes the Semi-Annual Statement of Consolidated Book Inventories and Transit Accounts, results of the inspection activities, the Safeguards Implementation Report (SIR) and the IAEA Annual Report.

Figure 1 illustrates the general information flow and shows the main data sources and destinations. The amount of information handled and its development over the last five years are illustrated in Fig. 2 by the volumes of incoming and outgoing reports.

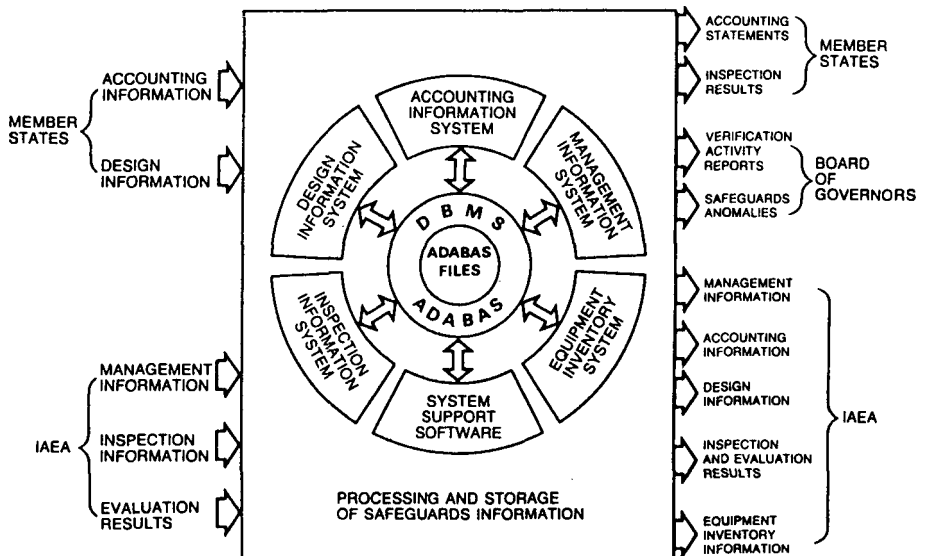


FIG. 1. General information flow. DBMS: Database Management System.

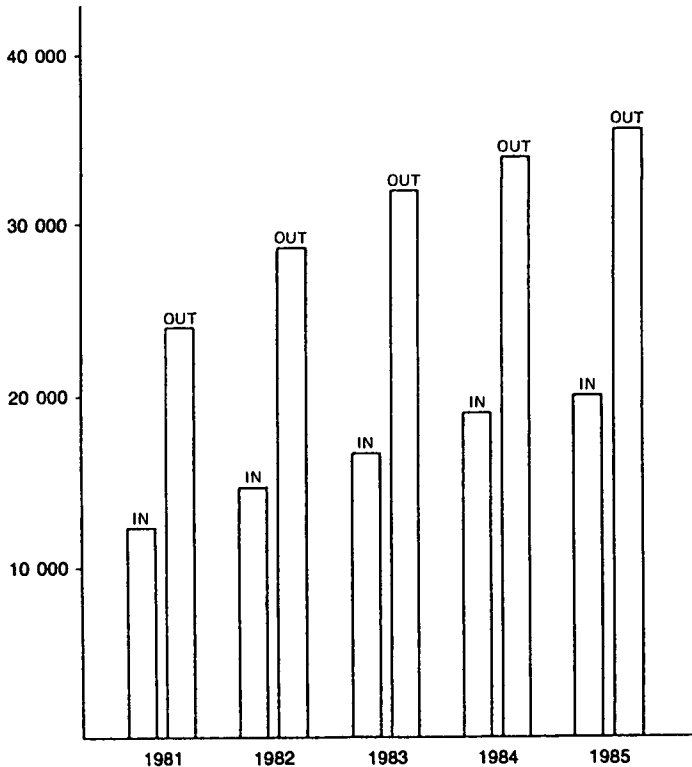


FIG. 2. Numbers of incoming and outgoing reports.

## 2.2. Main functions

ISIS comprises three main functions:

- Storage and maintenance of safeguards data;
- Processing of safeguards data;
- Evaluation of safeguards information.

The first function deals with the maintenance of the safeguards database, including data entry, data loading and quality control, as well as with protecting and ensuring the integrity of information contained in the database.

The second function deals with the requirements of safeguards data processing, e.g. data reduction, information retrieval, database queries, information comparison and merging of data from different sources according to the needs of the department.

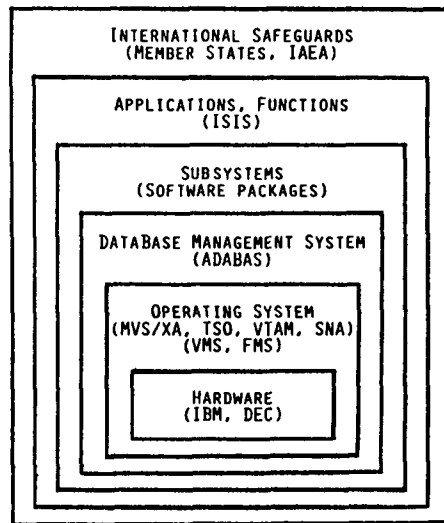


FIG. 3. ISIS architecture.

The third function includes the evaluation of information and the production of results that are either supplied directly to management or used by other departmental units as a basis for furnishing safeguards reports to Agency management or to Member States.

### 2.3. System architecture

The system architecture of ISIS has a hierarchical structure which is common for computerized information systems (Fig. 3). Each higher level governs its lower level by sending requests which take different forms as they proceed down the hierarchy to the lowest level – computer hardware. These requests are gradually changed from requesting some services (expressed in natural language), through queries and application programs (written in high level programming languages) to instructions, micro-instructions, etc., at the computer level. Lower levels send the messages to their higher levels in forms that change from electronic signals to the final reports, documents or answers on the screen.

The highest level of the architecture represents the overall mandate of the ISIS, which is to support the Department of Safeguards, and thereby the IAEA, in carrying out its statutory obligations in the international safeguards area as



expressed in official documents: the IAEA Statute (Article III.A.5), the NPT (Non-Proliferation Treaty) (Article III.1), INFCIRC/66/Rev.2 (para. 2) and INFCIRC/153 (Part II, paras 28-30). A wide variety of functions and services are expressed in bilateral agreements between the Agency and the Member States.

The next level corresponds to the main data processing functions of ISIS mentioned above: services and reports delivered to the Member States and the Agency. This entails storage, maintenance and processing of safeguards information as well as its evaluation.

The subsystems and database management levels in the ISIS architecture are very complex and their interconnections, mutual relations and internal structure do not form a simple hierarchy. Each level can be divided into six parts corresponding to the main areas of the Agency's safeguards activities as shown in Fig. 1. Additionally, there are software components which are common for all six subsystems. On the higher, subsystem level, these common parts are programming languages (e.g. Natural, PL/I, Fortran, macro language) and special software packages (e.g. ISPF, LIBRARIAN, DATA MANAGER, TEL-A-GGRAPH, SAS, MINITAB, DATATRIEVE). On the lower level, a commercial database management system (DBMS), ADABAS, manages data files and all access to the data.

Each subsystem has two components - software packages which define the processing functions performed by a particular subsystem, and database files which contain the subsystem related data. The software packages of all subsystems comprise the ISIS production library; the growth of this library during the last five years is shown in Fig. 4. Similarly, the ISIS production database growth during the last five years is depicted in Fig. 5.

The ISIS production library contains more than 1500 in-house developed software modules - 44% of the total belong to the Accounting Data subsystem, 30% to the Inspection Data subsystem, 14% to the System Support subsystem, and 10% to the Management Information subsystem; the remaining 2% represent software of the Design Information and Equipment Inventory subsystems.

The distribution of data in the ISIS production database, which holds over 4 million records (equivalent to 1 Gbyte), is substantially different. The largest portion belongs to the Accounting Data subsystem, with about 80% of the whole database. The data related to the Inspection Data subsystem comprise 10%, and the other 10% correspond to the remaining four subsystems.

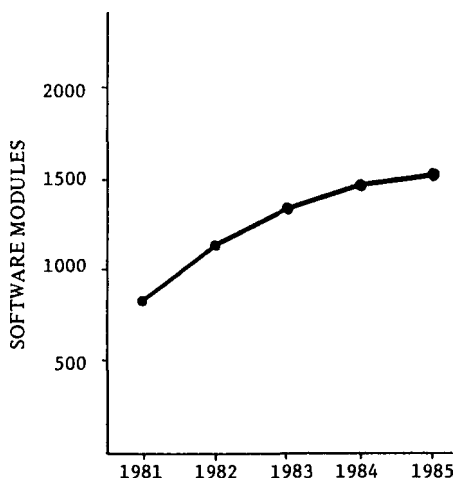


FIG. 4. Size of ISIS production library.

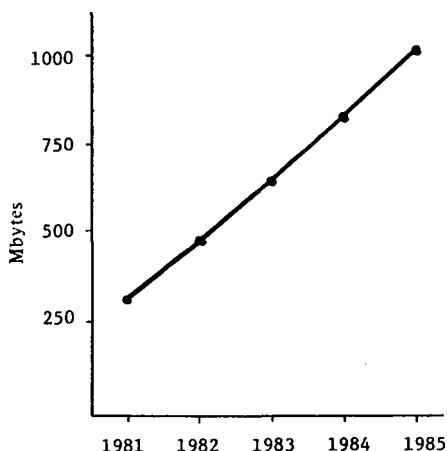


FIG. 5. Size of ISIS production database.

The operating system software for the IBM 3083 consists of the MVS/XA operating system, VTAM/NCP and SNA for teleprocessing, DFP to interface the cache-memory of the IBM 3880, and TSO/ISPF to support software development and to handle on-line transaction processing screens. The corresponding system software on the VAX 11/750 consists of the VMS operating system and FMS (Form Management System) to develop and handle data entry/data display screens. In addition, there are operating systems for microcomputers and personal computers (DOS for IBM and P/OS for DEC).

The ISIS hardware comprises an IBM 3083 with 16 Mbyte of main storage, a VAX 11/750 with 2 Mbyte of main storage and several microcomputers (DEC and IBM). The two main computers are coupled; more than 80 terminals are connected to them. Some of the microcomputers are linked to the mainframes and can be used as terminals. The IBM disk storage consists of two different models (2 IBM 3380 units and 14 IBM 3350 units) with a total capacity of approximately 10 Gbyte; the VAX disk capacity (2 RM80s) is approximately 250 Mbyte. The five tape units are mainly used for the input of accounting data reported by the Member States and for system maintenance tasks. To support applications in the field, several microcomputers have been installed in the Field Offices and at the Agency's Headquarters. In addition, they are used as data communication devices between Headquarters and the Field Offices in Canada and Japan through a public mailbox service.

### 3. FUTURE SYSTEM

Especially over the past several years international safeguards have developed and progressed. Approaches and procedures that were not clearly defined when the current ISIS was designed have now reached a degree of maturity that, if integrated into ISIS, will improve considerably the effectiveness of international safeguards and, in a time of zero growth budget constraints, will enable the Agency to make use of gains in operating efficiency as well as to cope with the continuously growing volume of information.

To improve the responsiveness of ISIS with respect to the needs of the Department of Safeguards, it is necessary to change the orientation of ISIS from a data collection and storage oriented system to a system geared to produce immediately usable results for safeguards. During this change of direction, new information processing techniques with improved cost effectiveness will be introduced to operate adequately under the present financial constraints.

Therefore, the transition has to be accomplished from a data collection and storage oriented system, with a limited degree of integration into the current safeguards and management process, to a result and product oriented system fully integrated into the safeguards and management information processing and decision support processes.

To promote system integration, it is envisioned that ISIS be reorganized into three major subsystems supporting nuclear material accountancy, inspection and management applications.

The accounting system area was reasonably well established during the early design of the current ISIS although some new applications have been added as they have become necessary. These will need some attention for optimization, further integration and consolidation.

Only recently has computer based support for inspection activities been implemented. This subsystem requires further definition of safeguards results which will lead into a review and consolidation of computerized inspection reporting. Computer support has become necessary for inspection activities in the field, and different techniques of on-line and off-line data transfer will be implemented.

The management information subsystem, covering several different areas (budget, personnel, training, schedules, designations, equipment), is at the stage where some fundamental steps are still to be taken - namely, to determine users and their needs, to define information flow, to define products and sources, etc. A master plan for the implementation of the department's management information system is under development.

A corresponding growth in information processing software is required to respond to these general requests and to more specific requests from the Department of Safeguards. This software covers a range from further analysis and reduction of data at Headquarters to software required to assist the safeguards inspector's activities at the facility with no direct support from Headquarters.

One more source of performance improvement is enhancement, redesign or replacement of some software parts which constitute bottlenecks for the performance of particular subsystems.

The system work-load increased during 1984-1986 at an average rate of 15% per year. If a continued average work-load increase of 15% per year is assumed, the IBM 3083 will be saturated in its present configuration at the beginning of 1989; by that time, upgrading of the hardware configuration will be necessary.

Major influences on the development of any computerized information system are new data processing methods and computer technologies, including advances in mainframe processors, microcomputers, local area networks and new on-line storage technologies. Microcomputers or personal computers will influence the future ISIS at all levels of the architecture. Several examples will illustrate this:

- The use of microcomputers at facilities during inspection.
- Direct data transmission via secure telecommunication links for communicating safeguards information from field locations to Vienna.
- During 1985, Member States, in agreement with the Agency, initiated the use of microcomputer diskettes as a medium for the transmission of nuclear material accounting data.
- Within the Headquarters operation itself, the use of microcomputers for office automation is anticipated.
- For software development, the use of microcomputers for design support and documentation promises a considerable reduction in work and improvement in quality for new applications.

#### 4. INFORMATION PROCESSING IN THE FIELD

Microcomputers will be necessary for inspectors at facilities where large volumes of data have to be recorded and evaluated. Thus, the scope of safeguards information processing will continue to expand to provide the necessary support in this area. As this develops, the availability of safeguards data on microcomputers will be utilized for inspection reporting purposes. Therefore, the integration of field support systems with the Agency's Headquarters system is essential.

The information flow loop will be completed by providing the inspector with a diskette of data to be used at the facility. This is an area of great potential where the microcomputer becomes the focal point for the collection of operator and inspection data as well as for on-site evaluation. Obviously, at facilities under continuous inspection the benefits of using a microcomputer would be most noted.

The potential exists for the connection of a microcomputer to the facility's computer system to perform direct auditing of the operator's records. Additional areas of application worth considering include use of the microcomputer to aid in comparison of records and reports, inventory verification and generation of sampling plans.

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## PARTICULAR PROBLEM SOLUTIONS WITHIN THE EURATOM INFORMATION SYSTEM

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### Abstract

PARTICULAR PROBLEM SOLUTIONS WITHIN THE EURATOM INFORMATION SYSTEM.

In recent years information systems have multiplied rapidly, reflecting the wide ranging developments in the technologies upon which they depend. However, it is often difficult to assess the effectiveness of information systems because the criteria upon which such a judgement may be based have not been defined. The paper proposes that three general considerations should be observed, namely that information must be handled efficiently, economically and with due regard to security. Since these considerations obviously present themselves differently according to circumstances, the paper examines the form they took in the particular case of the Euratom Safeguards Directorate. It might be interesting to consider whether this approach could be used as a guide to evaluating the contribution of information systems in the field of safeguards.

### 1. INTRODUCTION

The activity of nuclear safeguards exists to provide information. This information is of course of a highly specialized nature concerning the use to which nuclear material is put but, in more general terms, it may be considered that the exponential growth in the amount of information available is one of the most characteristic developments of the present day.

This development has been made possible by the emergence of what are sometimes called the 'new technologies' of informatics and telecommunications. The practice of safeguards could not have taken the course which it has in the past decade without being inextricably meshed into the growth of these technologies.

For this reason it is perhaps not easy to step back and to consider what are the essential requirements of an information system. The exercise is nevertheless necessary in order to assess whether safeguards are being conducted in such a way as to make optimum use of the means available.

This paper argues that there are three general criteria which an information system must meet, namely that information should be handled efficiently, economically and with due regard to security. The exact nature of these criteria obviously varies according to the circumstances under which the system is designed and developed, so the intention here is to consider the way in which each of these presented itself in a particular environment, namely the Euratom Safeguards Directorate, and the response which was made.

The first criterion is the efficiency of resource utilization, which is highlighted with the transfer of applications from a large general system to a dedicated machine. This necessitated a review of the applications design and of the data input element (resulting in a gradual move from manual to machine readable input).

Secondly, to deploy resources economically, users were integrated fully into the development cycle, fourth-generation tools were widely propagated and a database strategy was adopted which permitted the gradual introduction of a range of newly developed systems.

Data security, the final essential requirement, demanded a combination of different solutions. Proprietary software products were installed according to requirements. The corresponding hardware element involved use of independent personal computers to maintain the integrity of the database even when remote data transmission methods were employed.

## 2. EFFICIENCY

During the period that the Safeguards Directorate has been responsible for handling reports submitted by operators, the organization of this work has naturally undergone numerous transformations, some of which have already been discussed elsewhere [1]. Until 1981, the Directorate's informatics needs were served by a large central general data processing facility operated by the Informatics Directorate of the Commission of the European Communities, a situation which often required safeguards needs to be subordinated to the working patterns of the central installation. This sometimes caused, for example, considerable delays in reporting. A requirement to intensify and diversify the Safeguards Directorate's informatics activities led to the installation of a dedicated machine on which further systems could be developed under its own responsibility. The transfer to this machine of the existing system for nuclear material accountancy immediately required the solution of a specific problem.

This problem arose from the quantity of data which had to be handled by the accountancy system, combined with the system's necessarily cyclical nature. In terms of data quantities, it should be noted that about 670 facilities at present come under Euratom safeguards, submitting a total of about 330 000 entry lines annually. (In terms of quantities of material, the figures are approximately 105 t plutonium, 13 t highly enriched uranium, 20 000 t low enriched uranium, 70 000 t natural uranium, 60 000 t depleted uranium, 700 t thorium and 400 t heavy water.)

The system handling these data was naturally designed so as to reflect the submission of monthly reports and the requirement for monthly statistics. In the previous environment (where the system accounted for only about 5% of the processing requirement), this regular 'peak' in the work-load merged with the independent peaks of other applications to produce a relatively constant utilization of resources.



However, when this application was run alone on a dedicated computer, the total work-load was initially very unevenly spread throughout the month, which implied (since the capacity of the machine was necessarily determined by the maximum load) that the resources were under-utilized for much of the time. In overall terms, there was sufficient capacity for the development of new systems designed to rationalize safeguards activities, but in practice the work-load needed to be more evenly spread out before the available margin could be effectively used. Consequently, it was essential to 'smooth out' the peaks by means of a series of measures designed to improve the efficiency of machine utilization.

### **2.1. Buffer file**

Input data were previously entered as an ongoing process over several weeks but became integrated into the accountancy system only after the monthly update suite. The requirement for up to date statistical summaries caused numerous procedures to be run immediately after that integration, thereby aggravating the monthly processing bottleneck caused by the update suite. It was possible with some program modifications to make the new data now available to the listing procedures from the moment of data entry in a 'buffer file', which permitted them to be spread over a longer period, alleviating the bottleneck.

### **2.2. Data input**

A sustained attempt was made to reduce to a minimum the errors in data input. This is a critical consideration because the correction of input errors at an advanced stage of processing is a time consuming and therefore expensive operation. Installation operators were therefore encouraged as far as possible to submit their reports in computer readable form, that is on magnetic tapes or floppy disks, as this eliminates one possible source of error which exists where data are entered by keyboard operators. With the operators' co-operation, it has been possible to ensure that about 64% of the yearly 330 000 entry lines are now in this form, and it is hoped to raise this figure still further in the future.

### **2.3. Indexed file**

As an intermediate stage to transferring the accountancy data to an ADABAS database, an indexed sequential version of this file was created, giving the alternatives of sequential or indexed processing. The latter is a more efficient access mechanism where a relatively low proportion of records is to be retrieved, such as in situations where procedures are analysing only a subset of the data (e.g. records relating to a single material balance area).

## **2.4. Table searching**

It is a common feature of Euratom's data processing for certain key data to be held in tables which are retained in main memory for comparison with values read from input files. These may be regarded as playing a similar role to the IAEA's authority files. In some cases, these tables were searched sequentially to find equal values, the average number of comparisons being equal to half the number of entries in the table. This number may be so high that it is worth while sorting the table beforehand in order to employ the binary search, for which the number of searches required is the binary logarithm of the number of entries in the table.

## **2.5. File handling**

The running times of certain suites of programs were reduced in some cases where intermediate files were produced between programs, by amending the second program to write its output directly on to the area of disk occupied by the intermediate input file. This obviously reduced head movement on the disk units, which in typical cases accounts for over 95% of the running time of the programs.

Obviously, this measure has an impact upon recovery strategy, in that a program which fails to terminate normally (e.g. because of a system failure) cannot simply be rerun, but the suite must be restarted at an earlier point so as to recreate all intermediate files which have been so handled. However, the reliability of the Directorate's configuration is such that this overhead was found to be negligible.

## **2.6. Benefits**

The effect of the measures implemented was to reduce and, even more important, to redistribute the work-load on the computer. During 1985 the monthly CPU utilization diminished by about 25% from 363 to 274 giga-instructions, although the quantity of input data did not fall but actually rose (from an average of about 26 500 input records in 1985 to 27 750 in 1986). More significantly, however, the proportion of the work-load which was accounted for by the monthly update cycle (a few days' very intensive use of resources) fell from 36% to 20%. Therefore, what had been a very pronounced monthly 'peak' in machine utilization was now smoothed out to make for a more even distribution of work.

# **3. ECONOMY**

The measures described above were carried out concurrently with the task of equipping the Euratom Safeguards Directorate's inspectors with the informatics infrastructure necessary for modern safeguards work. In this, the Directorate was

faced with what has been termed the 'software crisis' of the 1980s [2], which may be described as follows.

Whereas almost all items of hardware are continually dropping in price (at least in real terms), the cost of software is rising because it is largely a function of salaries and related personnel overheads. The cost of proprietary software such as operating systems is shared among many customers, but application software, being developed 'in-house' for the sole use of the developer, is relatively far more expensive. In addition, the current climate of economic restraint makes it impossible for publicly funded bodies to recruit qualified computer staff at the rate necessary to undertake large scale development projects. The Euratom Safeguards Directorate's response to this situation is outlined below.

### **3.1. User involvement**

The user is integrated into all stages of the development process. His involvement at an early stage of software production diminishes the risk of inaccurate specification, which often lies at the root of unsatisfactory systems and resource wastage.

Development priorities are identified at regular meetings of a working party, in which all divisions of the Euratom Safeguards Directorate are represented. All staff are encouraged to become familiar with computer technology, with terminals and microcomputers being widely available (at present 34 terminals and 17 microcomputers for 200 staff). Thus computer literacy is not confined to the relatively small informatics service but is spread as widely as possible through the Directorate. This process is stimulated by regular training programmes in which members of the informatics service present both general software products and locally developed utilities to all interested staff. The result of this approach is that many computer procedures developed by individual users for their own purposes are now in general usage.

### **3.2. Fourth-generation software**

Rapid applications development is facilitated by the use of fourth-generation software tools. Among these may be mentioned the report writer, Easytrieve; the screen template generator, IFG/FHS; the database query language, Natural; and the data dictionary, Predict. Such tools, being closer to normal speech patterns than earlier computer languages, make it easier to formulate problems and hence solutions relatively rapidly. A ratio of 4:1 between time expended on the same problem using third-generation and fourth-generation techniques has been suggested in one study [3]. The self-documenting nature of the latter also reduces another considerable overhead of development.

### 3.3. Database approach

The commercially available database management system ADABAS was introduced through an evolutionary approach because the resources were not available for a revolutionary conversion exercise. Some applications were transferred to ADABAS gradually as manpower permitted; in other cases, data continued to be maintained by existing programs and copied regularly to ADABAS for on-line query, thus offering the advantage of rapid access with minimal outlay. New developments are made by principally using ADABAS.

### 3.4. Applications development

Within the strategy described above, a range of applications has been made available at the Euratom Safeguards Directorate within a relatively short period. Constraints of space prohibit a full catalogue here, but the following areas of activity may be mentioned briefly.

#### 3.4.1. *Inspection planning*

Details of each inspection planned, including names of inspectors participating and also details of instruments needed, are recorded by the co-ordinators of the inspection divisions. This permits each inspector to receive a personal inspection programme (either on-line or in printed form) and also enables the sector responsible for technical support to assess the requirement for each category of instrument and so to plan the itinerary of individual items.

#### 3.4.2. *Activity programme control*

Information is received from each installation, on an annual basis, of the principal activities relating to safeguards (physical inventory taking, etc.) which are planned for the coming year. This is integrated to give a general picture of requirements which is used by the Directorate in turn in planning its inspection programme.

#### 3.4.3. *Destructive analysis*

A system was developed to assist in the monitoring of samples taken for destructive analysis. This permits the follow-up of the sample from the date of taking, through its return to Luxembourg and submission to the laboratory, to receipt of the results and recording of chemical and isotopic details.

#### 3.4.4. *Seals*

This system has already been examined in some detail elsewhere [4] and so need not be discussed further here.

### 3.4.5. *Personnel*

The basic structure and a number of modules of the Management Information System have been implemented for routine use.

### 3.4.6. *Nuclear Material Statistical Analysis System (NUMSAS)*

NUMSAS, which was originally developed by the Joint Research Centre at Ispra, has been implemented in a modified form at the Euratom Safeguards Directorate [5]. Its purpose is to evaluate whether differences found in material balances as reported by the operator can be explained solely by inherent and unavoidable uncertainties in the operator's measurement system.

### 3.4.7. *Informatics hardware*

The informatics service has naturally been among the first to rationalize its own stock control, recording technical, administrative and contractual data related to each item of computer hardware.

## 4. SECURITY

In recent years, cases of unauthorized intrusions into computer systems have often attracted widespread publicity, which has helped to create a more general recognition of the problem of data security. An important consequence of this growing public awareness is that unauthorized access even to non-sensitive data may damage the credibility of the organization concerned. The Euratom Safeguards Directorate, being charged with the maintenance of data supplied by operators, has a responsibility therefore to ensure a degree of security at least equal to that prevailing where these data originate.

Data security may be implemented on the levels of either hardware or software, and any satisfactory strategy must allow for adequate provisions on both of these levels.

### 4.1. Software

The software mechanism implemented at the Euratom Safeguards Directorate has already been discussed elsewhere [6], so it is sufficient to note here in summary form that the following levels of security are implemented:

- (a) **Volume level** — restricting utilities which circumvent user identification protection to selected terminals in secure locations.
- (b) **File level** — through double password protection (user identification and file) on sensitive data.

- (c) **Application level** — based upon program libraries requiring a specific authorization profile with negative default.
- (d) **User level protection** — shielding of certain data elements even where other elements on the same file are accessible.

#### 4.2. Hardware

With respect to hardware security precautions, traditional methods were concerned with supervision (and physical locking at appropriate times) of data preparation, central processing and output handling areas. Subsequently, the mushrooming of local terminal networks increased the difficulty of supervising the physical location of all connected equipment, and more recently the introduction of remote terminals (using standard telephone lines while the Integrated Standard Data Network is awaited [7]) has brought with it a range of new problems.

These concern not so much the data actually being transferred (which are in any case no less safe than when they were entrusted to the mail service and may, if a higher level of security is required, be protected by a ciphering system). The vital consideration is that the privacy of the information already stored in the database should not be compromised. The requirement was to reconcile this with the need to provide a means of data transmission from remote sites to the Safeguards Directorate.

The strategy devised to meet this requirement was based upon the use of an Olivetti M24 microcomputer as a receiving point for remote data exchange. This decision takes advantage of the now almost universal availability of microcomputers in industry and business. In the five years which have elapsed since the appearance of the IBM PC, the emergence of an effective standard based upon compatibility with this machine has made possible the growth of a vast selection of software, including communications packages, and made the transfer of data a more practicable possibility.

The connection for data exchange may be made at any of several levels of sophistication. A simple communication may be established between two compatible systems using modems with the same speed (baud rate), but such a method has been found unsatisfactory in practice owing to the unreliability of dial-up lines and the absence of protocols for sequence checking and error handling. Such integrity checks have been defined in the X25 standard established by the Consultative Committee on International Telegraphy and Telephony (CCITT) to conform with the lowest three levels of the International Organization for Standardization (ISO) seven-layer reference model for open system interconnection [8] and implemented in the packet switching services currently offered by most national telecommunications bodies (e.g. PSS, DATEX-P, Transpac, Luxpac). Asynchronous data terminal equipment (such as a microcomputer) may be connected into the essentially synchronous world of packet switching by means of a PAD (Packet Assembler and Disassembler) in which the individual characters are buffered (defined in CCITT standards X28, X29 and X3). Therefore, this implies connection to a host computer on which the PAD is maintained.

Data transmitted from a remote site to a PC in Luxembourg may subsequently be captured on the Directorate's central database. This is achieved off-line by physically transferring the diskette on which they have been stored to a similar PC linked to the Directorate's mainframe in synchronous mode (i.e. emulating a local terminal). A standard file transfer utility can then be executed. The policy of assigning these two operations to two separate PCs is deliberate, since the manual intervention which it requires acts as a contact breaker in the chain of transmission. This ensures that the unauthorized transfer of data by an external agent is completely prohibited by the hardware configuration, whatever the efficacy of the software mechanism currently installed. The computer on which the data are maintained, together with the associated terminal network, remains within protected premises and without any outside connections, so that the level of data security is not compromised.

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# DEVELOPMENT OF A FACILITY RECORD AND REPORT MODEL SYSTEM AND A COMPUTERIZED BOOK AUDIT PROCEDURE

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## Abstract

DEVELOPMENT OF A FACILITY RECORD AND REPORT MODEL SYSTEM AND A COMPUTERIZED BOOK AUDIT PROCEDURE.

Computerization of the book audit procedure will not only help increase the credibility of safeguards applied to a nuclear facility but also decrease the manpower needed to carry out the inspection activity at the facility. A precondition of this computerization is the availability of a computerized record and report system at the facility level. Such a system will also improve the quality of data to be reported to the safeguards authority and reduce the manpower required to maintain the nuclear materials accountancy system at the facility. A facility record and report model system (FARMS) was developed and demonstrated as a standardized model materials accountancy information system. On the basis of this system, a book audit procedure was designed and computerized.

## 1. INTRODUCTION

In many countries, the introduction of computers has been progressing on various levels in the field of nuclear activities, including materials accounting and control. In order to develop an effective and efficient safeguards procedure, it is very important to cope with the recent trend of computerization.

While a measurement instrument with built-in microcomputer has been widely used and a procedure to collect measured data with the aid of a computer in the field has been investigated, a book audit is still a manual procedure to be carried out by an inspector. It is considered, however, that the establishment of a computer aided book audit procedure will help increase the credibility of safeguards applied to a nuclear facility because a thorough checking of records and reports is thereby made possible. Besides, it will decrease the manpower needed to carry out the inspection activities at the plant.

A precondition of the introduction of a computer into the audit activity is the availability of a computerized standard record and report system at the facility level. This requirement, however, will be easily satisfied if the facility already has a computer system.

Reasons for computerization may differ from facility to facility. Establishing a facility's information system, however, will improve the quality of the data to be used for the plant operator's and other purposes, and reduce the manpower required to maintain the operator's nuclear materials accountancy system. Standardization of the facility's system, on the other hand, could provide well defined information to both the operator and the inspector, thus making an inspection easier and at the same time reducing the amount of intrusion into plant operation.

The objectives of the study described here are to investigate the potential standardization of an accountancy information system and to study the feasibility of establishing a computerized book audit procedure. A facility record and report model system (FARMS) was developed and demonstrated as a standardized model materials accountancy information system [1]; this is described in Section 2. On the basis of this system, a book audit procedure was designed and computerized; details are given in Section 3.

## 2. FACILITY RECORD AND REPORT MODEL SYSTEM

A computer installed in a facility could be used for many jobs, ranging from the accounting, control and physical protection of nuclear materials, including process monitoring and control, to the management of personnel and financial property. In order to do these jobs, computer programs corresponding to each job have been developed in many facilities independently. One of them is an information system for nuclear materials accountancy. Since its primary purpose is to support the safe and economical operation of a nuclear facility, it has to satisfy many requirements, which differ from facility to facility. As a result, some standardization is needed if the computerization of a book audit procedure should be implemented on the basis of this information system, although other requirements from the State's system of accounting for and control of nuclear material (SSAC) could be fulfilled without any substantial work on the system.

An information system for the accounting and control of nuclear materials could be established so as to cope with three stages of operation: planning, implementation and reporting.

At the planning stage, a plant operation schedule will be worked out as well as a nuclear materials procurement programme. Planning data will be used in the operator's financial accounting, personnel management and safety considerations and for the confirmation of compliance with the national laws and regulations. It will also be requested that some of the data be submitted to the SSAC authority for inspection planning.

At the implementation stage, confirmation of materials receipts, shipments, transfers and inventories will be made by checking an item identification or by measuring an amount of nuclear materials. The result will be recorded by plant staff and be authorized by a responsible person of the plant staff as the established figures. Then the records will be kept in the system for a considerable time, e.g. ten years, for book audits by the authority or for reporting and management purposes at the facility.

At the reporting stage, a data processing operation on the data stored in the system will be carried out and a report will be produced in a given form for the SSAC authority or plant management.

FARMS was designed to handle the last two stages, i.e. recording of events, including authorization, and reporting.

A major function of a materials accountancy information system, as viewed from the spatial aspect, is to provide an inventory listing at an arbitrary time for a properly defined area of the plant. In the case of a facility's information system, the area is a unit area which is normally constructed around an inventory key measurement point with additional flow measurement points between the areas, and the official material balance area would be a combination of such areas. If flow data are given at the flow measurement points together with the nuclear loss and production data, the amount of materials in the unit area can be easily updated and accounted for, giving a book value of inventory at that area. When a physical inventory is taken, the book data can be compared with the corresponding physical value, which provides a material balance.

FARMS was so standardized as to fulfil the requirements mentioned above. It was also intended to make FARMS a standard system in terms of the portability of the software, by preparing computer programs in a common language and by making them operable on a minicomputer, the introduction of which is not expensive and can provide a capability for automating the book audit procedure at any type of facility.

The system has the following functions: input preparation and editing, including authorization of the records stored in the database; quality control; database management; data processing and reporting; and system backup and security.

The characteristics of the system are summarized as follows:

- (1) Input data can be entered into the system in an interactive manner with the computer, and a minimum amount of data is required for input, reducing manpower for data handling and decreasing the possibility of clerical error.
- (2) The quality control procedure which was provided for checking the consistency of the accountancy data will improve the quality of the records.
- (3) The record authorization mechanism will further improve the quality of records and at the same time prevent one from deleting records mistakenly; this mechanism can also make it possible to handle the records before authorization.
- (4) The system can update the book inventory at a time when inventory change data are given to the system, and it can produce reports at any time on request.

- (5) The present version of the system was provided for an item handling facility. It would, however, not only provide a basis for future expansion to a bulk handling facility but would also be useful for safe, reliable, smooth and efficient plant operation as well as for verification purposes, if applied.
- (6) It is possible to use an inexpensive computer system for the implementation of this record and report system, and the computer itself, of course, can be utilized for purposes other than materials accountancy. For handling the database, a simple management system was developed. Use of a commercially available database system, however, is possible and a modification of the system for such use would be easy.

FARMS was demonstrated and used as a training tool during the regional training course on the SSAC held in Tokai and Tokyo in March 1985 [1].

### 3. CONSTRUCTION OF A COMPUTERIZED BOOK AUDIT PROCEDURE

The book audit is one of the fundamental inspection activities at a plant. From the viewpoint of diversion path analysis, the book audit is one of the most important safeguards verification activities because the falsification of records and reports is the easiest and cheapest way of concealing a diversion. Therefore, a thorough checking of records and reports is required.

The book audit, however, is time consuming, complicated work. While records at the facility promptly reflect the movement of nuclear materials, reports are associated with a considerable time delay in showing the changes and distribution of inventories. The timing of inspection, on the other hand, does not necessarily correspond to the occurrence of inventory changes. Moreover, records and reports are susceptible to mistakes, making it necessary to correct the data from time to time.

Such reasons make it desirable to establish a computerized book audit procedure.

In order to attain an inspection goal, the following are required with regard to records and reports:

- (a) The book audit should be carried out over a period from the last physical inventory verification (PIV) before the current year to the last PIV during that year.
- (b) Records should be examined and compared with reports. If there are no reports, it should be confirmed that there are no events to report.
- (c) The audit procedure should have a capability of detecting inconsistencies among records and between records and reports.
- (d) Book inventory values should be determined at the time of inventory verifications and compared with those determined on the basis of reports.

To realize these requirements, a computerized book audit procedure should have the following functions:

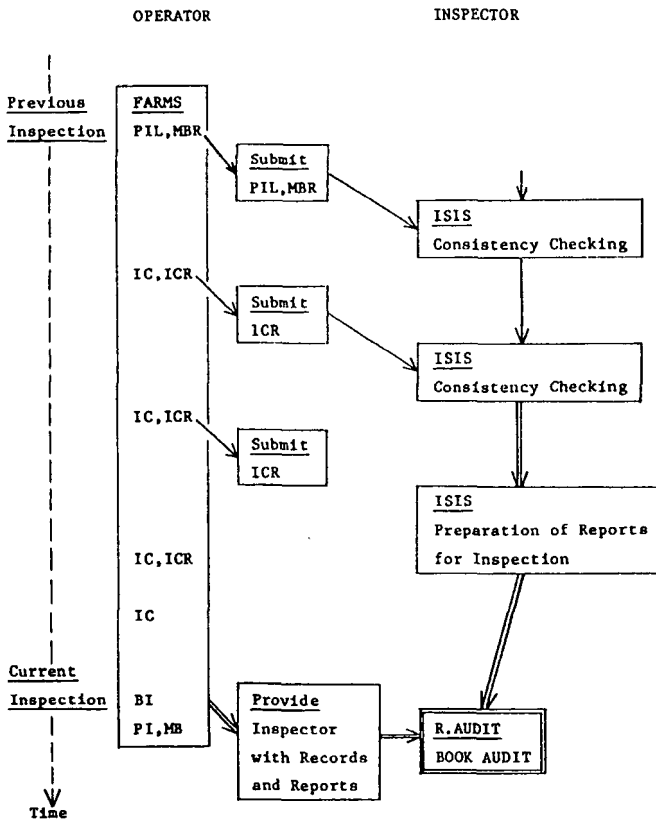


FIG. 1. The book audit system. IC, BI, PI and MB are records of inventory changes, book inventories, physical inventories and material balances; R: report; PIL: physical inventory listing.

- (i) A facility's records on receipts, shipments, book inventories, physical inventories and material balances, including corrections and adjustments, should be read into the audit system, and consistencies among batch data and other accounting records should be checked.
- (ii) Accountancy reports kept by the plant should be read in and compared with the records as well as with reports brought by an inspector from the International Atomic Energy Agency to the plant.
- (iii) A list of batch data on the receipts, shipments and inventories which are to be inspected should be generated.
- (iv) The book inventory should be updated on the basis of the records.
- (v) It should be confirmed that there are no changes in the records which were audited in the previous routine inspection.
- (vi) In cases where corrections to the batch data are made by the operator during inspection, it should be confirmed with the aid of the computer that such corrections have been accurately performed.

A model record audit system (R.AUDIT) was designed so as to fulfil the requirements mentioned above. It can simulate the book audit procedure at an item facility such as a light water reactor or a research reactor. The basic concept of the system is shown in Fig. 1.

It is assumed that an inspector who performs a book audit as an inspection activity at the facility brings to the facility a portable personal computer together with the accountancy reports which have been stored on floppy disk at the Agency. These reports are those provided by the plant operator with the aid of FARMS and then sent to the Agency through the national authority. At the plant, the inspector enters the accountancy records from FARMS into the portable computer by means of floppy disk or communication line, i.e. indirectly or directly. Then R.AUDIT automatically executes a book audit procedure by comparing records and reports, checking consistency within the records, updating a book inventory to the time of inspection and providing a list of inventories for each inventory key measurement point (KMP).

R.AUDIT was developed as a demonstration system using a commercially available relational database system, dBASE III. In R.AUDIT, the first processing step is to read a file, of ASCII format, in which the accountancy reports are provided by a conversion of the format from the Agency's ADABAS file, and to transfer its contents into the system's database file. The second step is to incorporate the operator's records into the system's file via an ASCII type file. Then, a series of processing operations are carried out; a thorough comparison between these two files is performed; a consistency check between the physical inventory listing (PIL) and the book inventory is carried out by calculating the book inventory based on the previous PIL and inventory changes during the period between the two PILs; a consistency check of the material balance report (MBR) is made by summarizing inventories and inventory changes and comparing them with the corresponding MBR entries; and in case there are some events which have not yet been reported to the Agency, consistency within such entries as well as between them is examined. Finally, a file is provided for the inspector, who takes it back to the Agency for future checking.

#### 4. DISCUSSION

The R.AUDIT system needs an additional program at the Agency for providing the inspector with the reports on floppy disk in ASCII format, and for comparing the facility's record with the report which will be sent later by the plant operator.

The interface between the facility's information system and the inspector's book audit system was provided in two ways, as mentioned earlier. The case of direct access, however, would create problems of data security and authentication. Therefore, indirect data transmission would be recommended. If these problems could be solved, then the use of the facility's computer by the inspector would be possible, providing a most efficient audit procedure at the plant.

One of the problems of the audit system is how to handle the plant specific data, for example transfers of nuclear materials within the material balance area. The plant is not obliged to report such data to the Agency, but they are essential to calculate a book inventory at each inventory KMP. The Agency has the right of access to such information as part of verification activities. Therefore, the audit system should have a capability of handling these data. The operator, however, might not want the data as well as the detailed records to be taken to the Agency. In that case, it is possible to modify the audit system so as to confine the data processing within the plant, keeping the data secure.

Another problem is how to maintain consistency of report entry numbers for records and reports in relation to the correction procedure. The method used in this study is to apply the same principle to both the records and the reports and to assign a specific 'report' number for the records which consist only of plant specific data. Such standardization of the facility's data preparation procedure is necessary to solve the problem.

The authors are trying to extend the computerized book audit procedure to bulk handling facilities. Two types of modification are needed: the first is concerned with the fact that the unit area for the facility's smallest material balance is not necessarily constructed around an inventory KMP. In some facilities, the unit area consists of several inventory KMPs, making it impossible to directly relate the book inventory to the physical ones because a physical inventory is given for each inventory KMP. The second modification requirement is due to the necessity to determine the amount of nuclear materials by measurements. Thus, standardization of measurement records as well as of quality control data is needed for both the facility's and the inspector's systems. These modifications are feasible, but not simple.

The impact of the implementation of a computerized book audit procedure on the facility's current system will depend on the degree of standardization of the system. Additional work might be necessary to improve the system. In such cases, the extended FARMS would be useful.

Owing to the adoption of a commercial database system in R.AUDIT, it will be possible to expand the system in the future to include other inspection data without major development efforts.

## 5. CONCLUSION

In view of the recent trend of office automation with the aid of computers, it is feasible to computerize the book audit procedure, thus making the audit less time consuming and more thorough and cost effective. Also, such computerization becomes most effective if the operator's accountancy information system is standardized and is fully utilized.

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# **NDA MEASUREMENT TECHNIQUES**

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# IN-FIELD CALIBRATION OF NEUTRON CORRELATION COUNTERS VIA CALORIMETRY AND HIGH COUNT RATE GAMMA RAY ISOTOPIC ABUNDANCE MEASUREMENTS

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## Abstract

IN-FIELD CALIBRATION OF NEUTRON CORRELATION COUNTERS VIA CALORIMETRY AND HIGH COUNT RATE GAMMA RAY ISOTOPIC ABUNDANCE MEASUREMENTS.

The use of transportable calorimetric assay equipment to perform an in-field calibration of a neutron correlation counter is described. During 1984-1986 this combination of instrumentation has been used to measure the plutonium content of a variety of oxide, salt and metal samples. An increase in sample throughput has been achieved by using this combination of measurements.

## INTRODUCTION

The use of transportable calorimeters and gamma ray isotopics systems for in-field assay of plutonium for safeguards purposes is well established [1,2,3]. Since 1980, the Safeguards Research and Development section at Mound has performed calorimetric assay (calorimetry and gamma ray spectroscopy) measurements at other U. S. laboratories and plants for the U. S. Department of Energy. The purpose of these measurements is to provide an independent verification of the measurement systems used by the site. Performing these measurements in the field (as opposed to shipping the plutonium to Mound) has definite advantages in addition to the accuracy and precision inherent in calorimetric assay. Some of the more pronounced advantages are improved material control of the plutonium, ready access to the material for repeat measurements, and ready access to process personnel. This ability to interact with people who are familiar with the history of the sample has been especially advantageous for

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\* Mound is operated by Monsanto Research Corporation for the United States Department of Energy under Contract No. DE-AC04-76DP00053.

developing measurement strategies for heterogeneous and other irregular samples. The primary challenge with these in-field calorimetric assay measurements has been measuring enough samples that a defensible conclusion concerning the sample category can be reached. The length of the exercise (typically two weeks), the time required for each calorimeter measurement (typically 4 to 6 h ), and the availability of the instruments and samples (typically during daytime shift on weekdays) limit the number of samples that can be measured. The pressure to produce measurements must also be balanced against the need for measurement control and quality assurance activities that help to validate the accountability measurements. This limit on the number of samples for which calorimetric assay measurements can be performed could prevent the achievement of the goal of the measurement exercise: the assessment of the quality of the measurement system employed by the site for the material category of interest.

Strategies for increasing throughput for calorimetric assay usually involve implementation of a servo control mode for the calorimeter's operation, sample preconditioning, optimizing the calorimeter's sample chamber for the sample size, and careful attention to sample packaging. These strategies require control over some of the parameters of the sample. In this program of in-field measurements, there is not sufficient control of these parameters to achieve a significant reduction in the time required for a calorimeter measurement. Consequently, the possibility of improving the throughput for a measurement exercise by including a second type of measurement was investigated. In this paper, the combined use of calorimetric assay with neutron correlation counting is reported. By combining the accuracy, precision, and matrix insensitivity of calorimetry with the speed of neutron correlation counting, a synergistic union was achieved.

#### APPROACHES TO THE PROBLEM

There are several approaches one could take in implementing this combination. In-field calibration of the neutron counter by calorimetric assay is the focus of this report. Calorimetric assay measurements are obtained for a set of samples from the material category of interest. These samples are selected to span the range of plutonium content within that material category. Neutron correlation measurements are also performed for these samples, and a calibration curve of neutron counter response as a function of plutonium content is constructed. Then, the remaining samples from the category of interest are assayed by the neutron correlation counter.

TABLE I. SUMMARY OF NEUTRON CORRELATION MEASUREMENTS

Category	Plutonium content (kg)	Number of samples	
		Neutron <sup>a</sup>	Cal. assay
Product oxide	0.1-0.9	26	6
Electrorefining salt residue	0.3-1.2	21	13
Low fired oxide	0.5-1.6	35	7
Direct oxide reduction salt	0.3-0.7	15	5
Direct oxide reduction metal	0.2-0.6	19	10

<sup>a</sup> Includes calibration data.

Since the neutron data can be obtained in nominally 10 min, the number of samples assayed during an in-field exercise is no longer limited by the time required for the calorimeter measurement, but by the number of samples which can be transported to the neutron counter.

## EXPERIENCE

During the last three years, this combination has been used in five in-field measurement exercises to measure the plutonium content of a variety of sample types. These categories included pure product oxide, electrorefining salt residues, low fired oxide, salt residues from direct oxide reduction, and metal buttons from direct oxide reduction. The plutonium content of these samples ranged from 0.1 to 1.6 kg. This information is summarized in Table I.

The neutron correlation data were obtained with a high level neutron coincidence counter (IRT Corporation Model Number HLNCC-100). The neutron data were acquired until the statistical uncertainty in the real coincidence rate was approximately 1%. This required 5 to 15 min of data acquisition, depending on the material type. For the calibration samples, replicate measurements were obtained. A calibration curve of the real coincidence rate as a function of plutonium content measured by calorimetric assay was constructed. A calibration curve of the form:

$$CR = a*PU^2 + b*PU$$

where CR = real coincidence rate  
PU = plutonium content

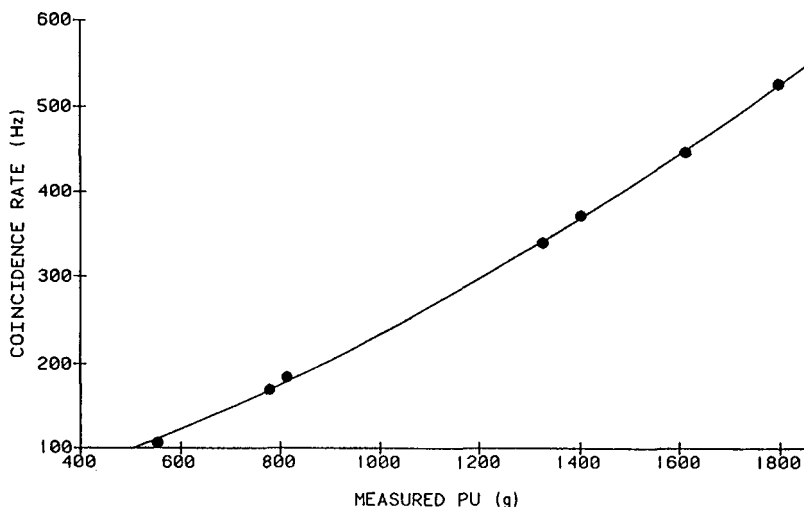


FIG. 1. Neutron correlation counter calibration for low fired oxide samples. The real coincidence rate measured with the neutron correlation counter is plotted as a function of the plutonium content measured by calorimetric assay. The uncertainties in the data points are approximately the size of the circles. The calibration function is a quadratic that passes through the origin.

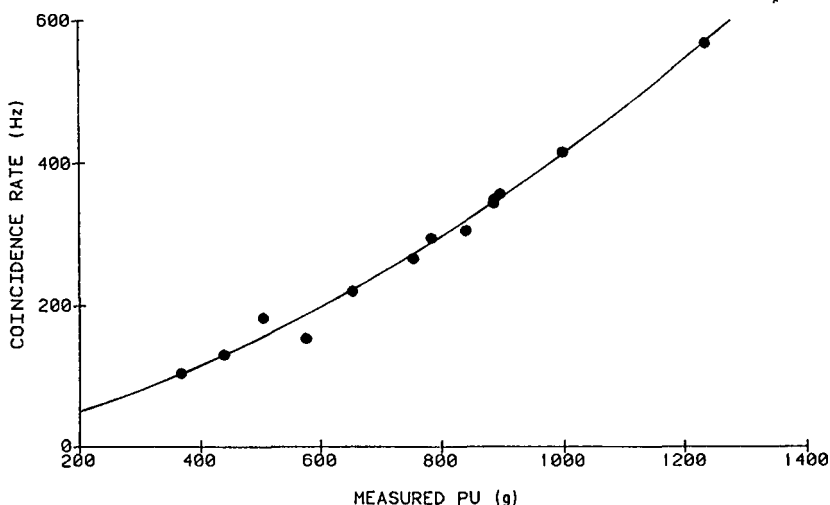


FIG. 2. Neutron correlation counter calibration for electrorefining salt residue samples. The real coincidence rate measured with the neutron correlation counter is plotted as a function of the plutonium content measured by calorimetric assay. The uncertainties in the data points are approximately the size of the circles. The calibration function is a quadratic that passes through the origin.

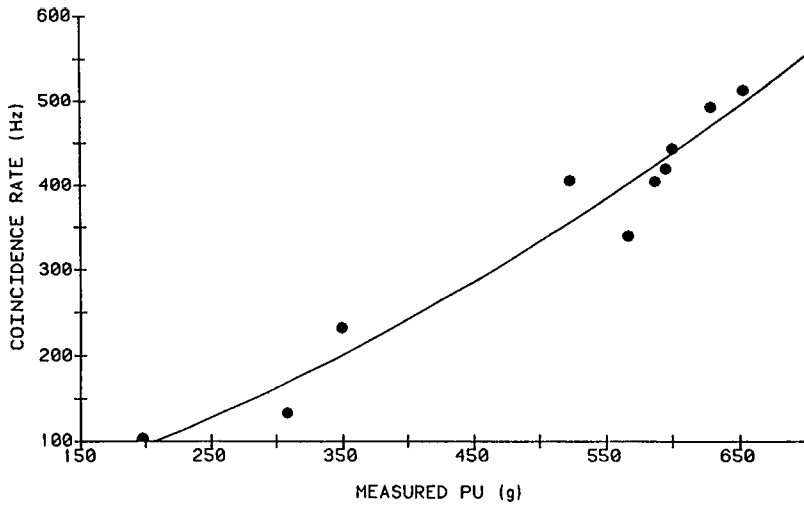


FIG. 3. Neutron correlation counter calibration for metal samples from direct oxide reduction. The real coincidence rate measured with the neutron correlation counter is plotted as a function of the plutonium content measured by calorimetric assay. The uncertainties in the data points are approximately the size of the circles. The calibration function is a quadratic that passes through the origin.

was constructed. Calibration curves for the oxide, salt, and metal categories are shown in Figures 1, 2, and 3, respectively.

In Tables II, III, and IV, the plutonium content determined from the calibration curve is compared to the plutonium content determined by calorimetric assay. The samples in these tables form the calibration sets for these categories. Table V is an example of the determination of the plutonium content for the samples outside the calibration set. The samples listed in Table V are low fired oxides. The assay values from the neutron correlation counting are compared to the operator's declared values for the plutonium content.

The calorimeter data were obtained with one of Mound's transportable calorimeters. These calorimeters are operated in the heater replacement mode and employ the prediction of equilibrium algorithm.

The gamma ray data were obtained with one of Mound's transportable gamma ray assay systems. A significant improvement in the data acquisition capability of this system is discussed in the next section.

TABLE II. HLNCC<sup>a</sup> CALIBRATION DATA FOR LOW FIRED OXIDE SAMPLES

Sample ID	Coin. rate	Plutonium (g)		Diff. (%)
		HLNCC assay	Cal. assay	
1	106.88	531.3	552.0	3.74
2	340.53	1319.0	1325.9	0.52
3	372.51	1407.8	1401.1	-0.48
4	170.56	778.6	776.1	-0.33
5	527.82	1801.1	1797.5	-0.20
6	185.14	830.8	813.7	-2.11
7	447.32	1604.3	1611.8	0.46

Notes: Coincidence rate = average of three measurements.

Calibration curve: coincidence rate =  $7.238654E-05PU^2 + 0.162683PU$ .

Difference (%) =  $[(\text{calorimetry} - \text{HLNCC})/\text{calorimetry}] \times 100$ .

<sup>a</sup> High level neutron coincidence counter.

Seventy-five samples were measured using the neutron correlation counter calibrated by calorimetric assay. Forty-one samples were measured by calorimetric assay. The number of neutron measurements was limited by the number of samples in a category and the time required to transport samples. The time required to make the measurement was not a limiting factor.

#### HIGH COUNT RATE GAMMA RAY MEASUREMENTS

The interpretation of both the neutron counter data and the calorimetry data involves information on the isotopic composition of the sample. Previously, the throughput for the isotopics measurement system had to match the throughput of the calorimeter. Gamma ray measurements performed at low count rates and with long (4 h) data acquisition times resulted in an uncertainty in the effective specific power of less than 1%. When this is combined with the uncertainty in the power measurement, an uncertainty in the plutonium content of less than 1% was obtained. With the increase in the number of gamma ray measurements required to support calorimetry and neutron counting, an improvement in the gamma ray throughput



TABLE III. HLNCC<sup>a</sup> CALIBRATION DATA FOR ELECTROREFINING SALT RESIDUE SAMPLES

Sample ID	Coin. rate	Plutonium (g)		Diff. (%)
		HLNCC assay	Cal. assay	
1	105.04	374.5	366.1	-2.30
2	131.64	445.4	436.9	-1.95
3	154.15	501.4	574.6	12.73
4	182.36	567.3	503.6	-12.65
5	220.29	649.8	651.4	0.24
6	266.08	742.0	753.2	1.48
7	294.25	795.4	783.5	-1.52
8	305.58	816.3	840.4	2.86
9	343.37	883.5	854.6	-3.39
10	349.74	894.5	886.2	-0.93
11	356.92	906.8	895.7	-1.24
12	416.32	1004.6	997.7	-0.70
13	569.67	1231.4	1232.6	0.10

Notes: Coincidence rate = average of three measurements.

Calibration curve: coincidence rate =  $2.125583E-04\text{PU}^2 + 0.200853\text{PU}$ .

Difference (%) =  $[(\text{calorimetry} - \text{HLNCC})/\text{calorimetry}] \times 100$ .

<sup>a</sup> High level neutron coincidence counter.

was required to maintain the uncertainty in the calorimetric assay measurement. The in-field gamma ray assay system was equipped with commercially available electronics appropriate for acquisition of gamma ray spectral data at a high count rate (approx. 50 kHz). The preamplifier for the detector and the analogue to digital converter (ADC) were optimized for high count rates. The detector's preamplifier was tuned to enable low resolution performance at energy dissipation rates of 4000 MeV/s. A multichannel analyzer (MCA) with a 450 MHz Wilkinson type ADC was used. Figure 4 depicts the throughput (count rate measured at MCA) as a function of the input (count rate at the detector) for a sample measured with the high count rate electronics. This representation of the pulse processing efficiency is depicted for three different amplifier shaping time constants: 1, 3, and 6  $\mu\text{s}$ . Also shown is the ideal case in which the count rate at the multichannel analyzer matches the count rate at the detector.

TABLE IV. HLNCC<sup>a</sup> CALIBRATION DATA FOR METAL SAMPLES FROM DIRECT OXIDE REDUCTION

Sample ID	Coin. rate	Plutonium (g)		Diff. (%)
		HLNCC assay	Cal. assay	
1	103.00	211.2	197	-7.20
2	132.70	257.0	308	16.55
3	232.70	389.9	349	-11.43
4	404.50	569.2	522	-9.04
5	339.00	505.3	566	10.73
6	403.10	567.9	586	3.10
7	418.20	581.9	594	2.03
8	442.80	604.4	599	-0.90
9	491.50	647.3	628	-3.07
10	511.50	664.3	652	-1.89

Notes: Coincidence rate = average of three measurements.

Calibration curve: coincidence rate =  $6.228755E-04PU^2 + 0.356159PU$ .

Difference (%) = [(calorimetry - HLNCC)/calorimetry] × 100.

<sup>a</sup> High level neutron coincidence counter.

It is important to note that the count rate at the multichannel analyzer is not a monotonically increasing function of the count rate at the detector. In Figure 5, the resolution (full width, half maximum at 129 keV) of the gamma ray system is plotted as a function of the input count rate. Figure 6 depicts the uncertainty in the effective specific power measured with this system as a function of data acquisition time (real time). These data were acquired with a count rate at the detector of 45 kHz. Note that an uncertainty of 1% is obtained in less than 5 min.

## SUMMARY

There are several benefits to in-field calibration of neutron correlation counters by calorimetric assay. The throughput for the measurement exercise is greater than that achieved with calorimetric assay alone. The neutron counter is calibrated with the same material and packaging as are

TABLE V. HLNCC<sup>a</sup> ASSAY DATA FOR LOW FIRED OXIDE SAMPLES

Sample ID	Coin. rate	Plutonium (g)		Diff. (%)
		HLNCC assay	Declared	
1	403.98	1492.3	1449	2.90
2	367.13	1393.1	1367	1.88
3	387.79	1449.2	1418	2.15
4	190.69	850.4	805	5.34
5	443.45	1594.5	1515	4.99
6	429.41	1558.6	1514	2.86
7	410.03	1508.2	1457	3.40
8	409.76	1507.5	1507	0.04
9	421.47	1538.1	1491	3.06
10	412.93	1515.8	1523	-0.47
11	412.75	1515.4	1500	1.01
12	404.37	1493.4	1450	2.90
13	439.45	1584.3	1553	1.98
14	382.61	1435.3	1444	-0.61
15	372.20	1407.0	1418	-0.78
16	440.38	1586.7	1568	1.18
17	376.05	1417.5	1430	-0.88
18	435.28	1573.7	1547	1.70
19	423.61	1543.6	1495	3.15
20	435.46	1574.2	1583	-0.56
21	434.34	1571.3	1527	2.82
22	368.00	1395.5	1372	1.69
23	488.26	1706.1	1689	1.00
24	350.20	1346.2	1309	2.77
25	430.10	1560.4	1529	2.01
26	409.51	1506.9	1547	-2.66
27	463.37	1644.7	1648	-0.20
28	492.23	1715.8	1667	2.84
				1.62 Av.
				1.86 SD

Notes: Coincidence rate = average of three measurements.

Calibration curve: coincidence rate =  $7.238654E-05PU^2 + 0.162683PU$ .

Difference (%) =  $[(\text{calorimetry} - \text{HLNCC})/\text{calorimetry}] \times 100$ .

<sup>a</sup> High level neutron coincidence counter.

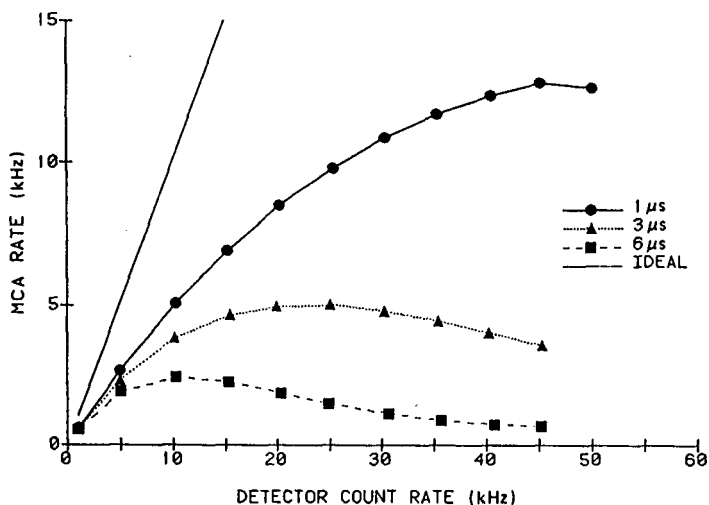


FIG. 4. Pulse processing efficiency for high count rate gamma ray assay system. The count rate recorded at the MCA is depicted as a function of the count rate at the detector for three values of the amplifier shaper time setting: 1, 3, and 6  $\mu$ s.

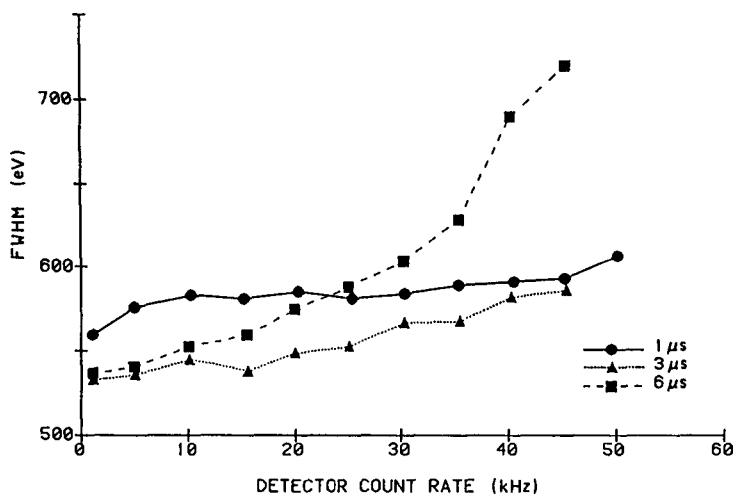


FIG. 5. Effect of count rate on resolution for high count rate gamma ray assay system. The resolution (FWHM at 129 keV) is plotted as a function of the count rate at the detector for three values of the amplifier shaper time setting: 1, 3, and 6  $\mu$ s.

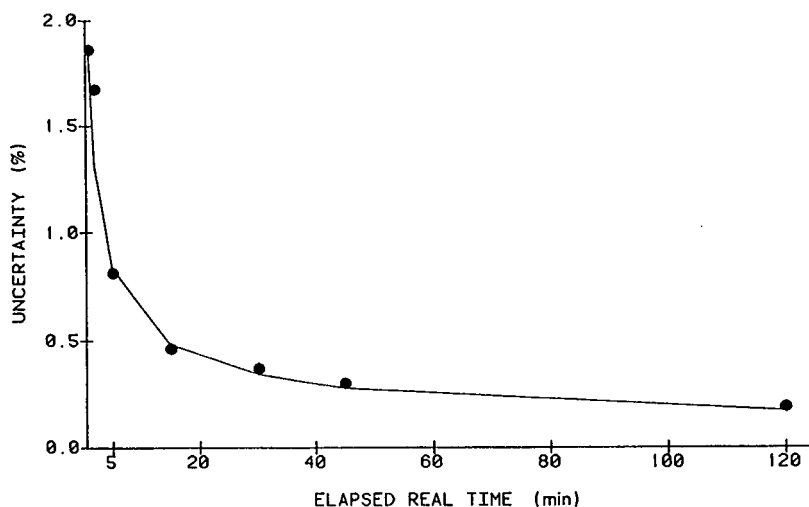


FIG. 6. Uncertainty in effective specific power as a function of duration of data acquisition. These data were acquired at a detector count rate of 45 kHz. The uncertainty is less than 1% for acquisition times of less than 5 min.

experienced during the neutron measurements. The reliability of the neutron correlation counter data is better than that obtained with calibration curves determined in the laboratory.

#### ACKNOWLEDGEMENTS

The author wishes to thank the following individuals who have helped with the acquisition of the data reported here: K. L. Breakall, J. G. Fleissner, R. L. Mayer, E. F. Novak, W. W. Rodenburg, and M. R. Spivey.

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## USE OF RADIOMETRIC CALORIMETERS IN A GLOVEBOX ENVIRONMENT

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### Abstract

#### USE OF RADIOMETRIC CALORIMETERS IN A GLOVEBOX ENVIRONMENT.

The use of radiometric calorimeters in a glovebox environment requires that special consideration be given to instrument design, measurement methodology, and measurement control. The measurement system described in the paper consists of five radiometric calorimeters installed in production facility glovebox lines at Mound, a United States Department of Energy contractor operated facility.

### 1. INTRODUCTION

Radiometric calorimeters are instruments designed to measure the rate of thermal power production for items placed within their measurement chambers. Calorimeters are routinely used as assay instruments for plutonium and other radioactive materials. Items placed within the measurement chamber of a calorimeter are separated from a constant temperature environment by a thermal resistance element. Thermal equilibrium is attained when the rate of thermal energy transfer to the environment is equal to that generated in the measurement chamber. Under these conditions, the temperature difference across the thermal resistance element is proportional to the thermal power produced in the measurement chamber. Items that generate a known amount of heat are used to calibrate the instrument, thus providing a means of relating the temperature difference to rate of power production. When coupled with the rate of heat generation per unit mass of a material, calorimetry can provide a precise nondestructive estimate of the amount of that material present in a container.

Radioactive material is frequently processed in a glovebox environment to provide an inert, low moisture atmosphere, to contain the material, and to protect workers from contamination.

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\* Mound is operated by Monsanto Research Corporation for the United States Department of Energy under Contract No. DE-AC04-76DP00053.

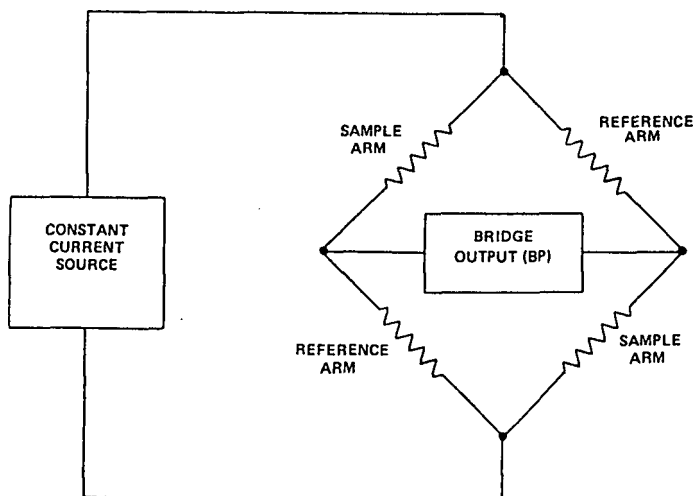


FIG. 1. Calorimeter circuit with Wheatstone Bridge.

It is, therefore, often desirable to install calorimeters in glovebox lines so that accountability, material control, and process monitoring data can be obtained expediently without violating the glovebox environment. Such a system has recently been designed and fabricated at Mound and is currently in operation. Subsequent paragraphs describe the design, operation, and measurement control considerations applied to this system which are unique to the use of calorimeters in a glovebox environment.

## 2. DESIGN CONSIDERATIONS

Design considerations for glovebox installation of radiometric calorimeters include isothermal environment design, number of penetrations into the glovebox floor in a limited area, height of the glovebox floor from the room floor, ease of installation and removal, ease of sample loading and unloading, and the need for isolation of the sample chamber from fluctuations in glovebox environmental conditions. Added major design considerations which are not unique to glovebox operation are criticality requirements and measurement precision and accuracy.

### 2.1. Calorimeter Design

There are three basic types of calorimeter designs commonly used at Mound. These all employ a Wheatstone Bridge (see



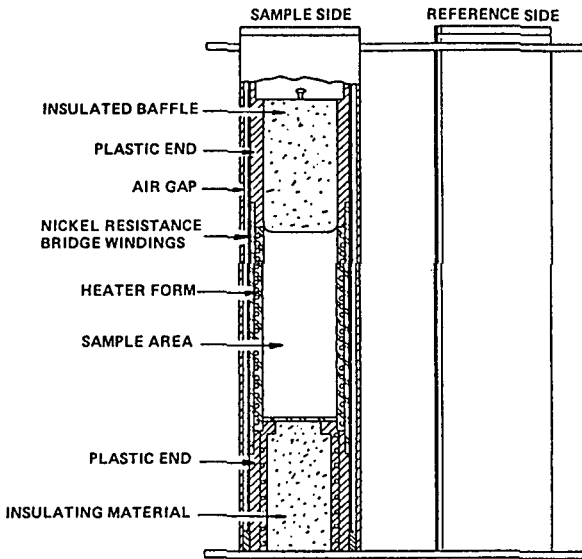


FIG. 2. Cross-section of a full twin bridge calorimeter.

Figure 1) with two arms of the bridge used as sample chamber sensing elements, and the other arms as reference elements which are maintained at the temperature of the isothermal environment. The three calorimeter designs each employ different physical configurations of the Wheatstone Bridge, as shown in Figures 2, 3, and 4. The full twin bridge design provides the best precision, accuracy, and long term stability. It is larger, however, since two identical chambers are placed side by side, and it requires two glovebox penetrations per calorimeter. The over/under bridge design is significantly smaller than the full twin, provides almost as good accuracy and precision, and requires only one glovebox penetration per calorimeter. It is, however, the tallest of the three designs, thus requiring the glovebox floor to be at a greater height above the room floor. The gradient bridge design is the smallest, and also requires only one glovebox penetration per calorimeter. The precision and accuracy of the gradient bridge design, however, are the poorest of the three designs.

The gradient bridge design was chosen for use in this system, since space requirements were minimized, and prior experience indicated that the desired precision of 0.15% in the range 0.5 W to 5 W could be met without difficulty.

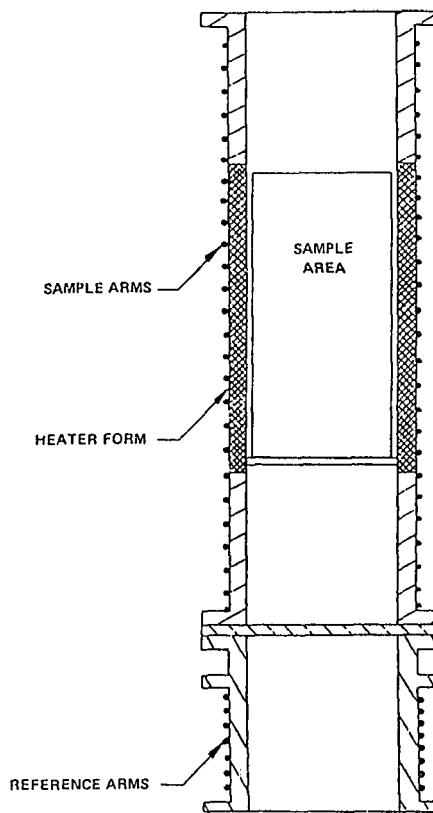


FIG. 3. Cross-section of an over/under bridge calorimeter.

## 2.2. Bath Design

Isothermal water baths are used to provide the isothermal environment for all calorimeters currently in use at Mound. When water is used as a heat transfer medium, temperature of the isothermal environment can be maintained to within  $0.001^{\circ}\text{C}$ . Two water bath designs were considered for the box line calorimeter installation: environmental and recirculatory. Environmental water baths are large tanks of water with electrical heaters immersed in the water, stirrers to circulate the water, and either evaporative cooling or a gas based coolant. These baths provide the greatest isothermal stability and must be used in applications with full twin bridge calorimeters. The disadvantages of the environmental water bath are size (the baths are typically 1.5 m wide, 2 m long, and 1 m high), quantity of water required (typically over 800 liters), and difficulties in performing maintenance on baths installed under

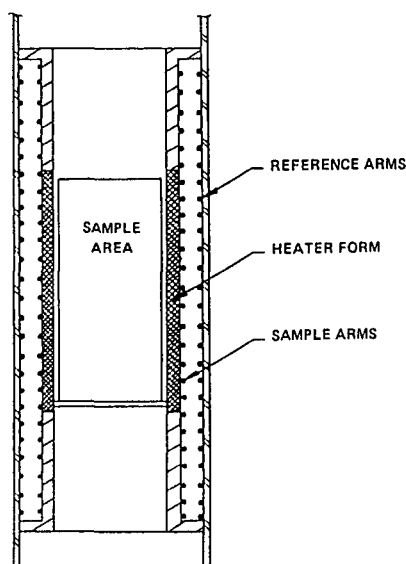


FIG. 4. Cross-section of a gradient bridge calorimeter.

glovebox lines. Recirculatory water baths are small tanks of water in which the temperature of the water is maintained by electrical heating and gas based cooling. Water is pumped continuously from the small isothermal tank through a jacket which contains the calorimeter and then returned to the isothermal tank. These baths provide only slightly poorer isothermal stability than the environmental baths. The advantages of this bath design are small size, small quantity of water required, and ease of maintenance. Although criticality was not a consideration for this glovebox installation, the recirculatory bath design, coupled with the gradient bridge calorimeter design, provides criticality limits which are about the same as for a high level neutron coincidence counter. The recirculatory water bath design was chosen for this installation because of its small size and ease of maintenance.

### 2.3. Glovebox Design

Several features were included in the glovebox design to facilitate the use of calorimeters. The floor of the glovebox was fabricated from 2.5 cm thick stainless steel to support the weight of the calorimeters, as well as to provide a heat sink to insulate the calorimeters from small fluctuations in the temperature of the glovebox. Stainless steel covers with O-ring seals were bolted to the floor to cover the openings under which the calorimeters were to be installed. The floor of the

glovebox was located about 1.3 m above the room floor to provide sufficient space beneath the glovebox floor in which to place the calorimeters. The calorimeters were installed by first bolting the calorimeter/water jacket assemblies to the underside of the floor, and then removing the stainless steel covers, thus maintaining the integrity of the box line during calorimeter installation. If the need to remove the calorimeters arises, they can also be removed without violating the glovebox seal by first bolting the stainless steel covers back in place, and then bagging and removing the calorimeters. O-ring seals and hold-down clamps were provided for the lids of the calorimeters to prevent fluctuations in glovebox line pressure from affecting measurement results. An overhead electrical hoist was supplied on a manually operated X-Y transport system. The X-Y transport system was designed so that the hoist will center itself over the calorimeters as tension is placed on the hoist cable.

### 3. MEASUREMENT CONSIDERATIONS

The primary measurement considerations were to minimize measurement times while maintaining the required precision of 0.15% or better in the range of 0.5 W to 5 W. Three options were available to accomplish these goals: heater replacement operation, servo-controlled operation, and prediction of equilibrium. The heater replacement mode of calorimeter operation provides the best precision of the three methods. The heater replacement method is based on a three-step procedure to determine the power generated by an item. First, an equilibration baseline, which is the bridge potential with the calorimeter empty and no heat being input to the calorimeter, is determined. Next, after the item has been placed into the calorimeter and permitted to reach thermal equilibrium, a second bridge potential value is determined. The item is then removed, and an electrical heater is used to input roughly the same amount of heat to the calorimeter as the item. The calorimeter is permitted to reach thermal equilibrium, and a third bridge potential value is determined. The wattage of the measured item may then be determined based on these three values, the power output by the electrical heater, and the sensitivity or change in bridge potential induced by a known amount of heat input to the system.

Prediction of equilibrium follows the same procedure as the heater replacement mode; however, equilibrium values are estimated mathematically using a prediction routine. Since thermal equilibrium is never attained with this method of operation, measurement times are reduced significantly, usually

on the order of 50%. Some loss of precision occurs, however, when prediction of equilibrium is used.

The servo-controlled method of operation is the fastest method available. An electrical heater continuously supplies heat to the calorimeter, with the output of this heater varied to maintain a constant bridge potential value. A thermal equilibrium value for the power supplied by the heater is first determined. The item to be assayed is then loaded, and the power supplied by the electrical heater decreases so that the total power (electrical plus item) supplied to the calorimeter remains constant. The power of the sample is, then, the difference between the electrical power supplied to the calorimeter with the sample chamber empty and with the item loaded. The precision of the servo-controlled method is the poorest of the three methods of operation.

The servo-controlled method of operation was chosen for this calorimeter measurement laboratory. Items can be assayed using this method within a time frame of about 3.5 h, making it possible to measure up to two items per calorimeter (optimally) in an 8 h workday. Even though the precision for this method is the poorest of the three operating methods considered, the design requirement of 0.15% precision was met without difficulty.

The primary differences between operating a calorimeter inside a glovebox and outside a glovebox are measurement time and controlled atmospheric conditions. Measurement times are reduced when measuring inside a glovebox as a result of less stringent packaging requirements. Items measured outside a glovebox normally require secondary, or in some cases, tertiary containment. These additional levels of containment retard the flow of heat from the sample to the environment, and thus increase the time required to reach thermal equilibrium. For glovebox operations, however, the glovebox acts as the secondary containment, thus reducing assay time requirements. Improved control of atmospheric conditions inside a glovebox results in an improvement in measurement precision, as the magnitude of the random fluctuations of the bridge potential is reduced.

#### 4. MEASUREMENT CONTROL

The primary measurement control is based on weekly measurement of plutonium-238 heat standards, calibrated to an uncertainty of 0.02%. Upon completion of a heat standard measurement, the results of the latest five measurements are

compared to the design specifications for precision and accuracy. In addition, the error for the latest measurement is compared with warning and action limits. If warning limits are exceeded, an additional measurement of the standard is performed, and all tests are repeated. If precision and accuracy limits are not met, action limits are exceeded, or if warning limits are exceeded for two consecutive measurements, the calorimeter is removed from service. The calorimeter may be returned to service once appropriate repairs have been made, and a series of five standard measurements has been completed indicating that the instrument is again operating properly.

Since standard measurements are performed only on a weekly basis, diagnostic indicators are also used to provide an early warning of control loss. These indicators are baseline measurements, box line temperature, and temperature of the isothermal water bath. If action limits are exceeded for tests on the baseline measurements, a series of standards is measured to assess the condition of the affected calorimeter. If action limits for box line or bath temperature are exceeded, measurement results are rejected, and the measurement is repeated. Measurement results are carefully reviewed by operators and their management before being released if warning limits are exceeded for any of the three diagnostic indicators.

## 5. USE IN INSPECTIONS

Calorimeters installed in a glovebox line are also useful to inspectors during audit verification measurements. The inspector can first authenticate the operation of the calorimeter by measurement of a certified heat standard or by passing standard material maintained by the inspector into the glovebox line for measurement. Following authentication, on-site measurement of materials requiring glovebox containment can be performed.

## 6. CONCLUSIONS

Installation of radiometric calorimeters in a glovebox environment is both practical and useful. Although some special considerations need to be given to instrument and glovebox design, measurement methodology, and measurement control, these considerations are not prohibitive to the installation and operation of calorimeters in glovebox lines. Some operating benefits, such as shortened measurement times and better precision, have been observed. The benefits of in-situ measurement of material contained in glovebox lines for

accountability, materials control, inspections, and process monitoring make these installations desirable for many practical applications.

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# **CONTROLE DE LA MASSE D'UN ECHANTILLON PLUTONIFERE PAR COINCIDENCES NEUTRONIQUES COMPTE TENU DES FISSIONS INDUITES**

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## **Abstract—Résumé**

### **DETERMINATION OF THE MASS OF PLUTONIUM SAMPLES BY NEUTRON COINCIDENCE COUNTING WITH ALLOWANCE FOR INDUCED FISSIONS.**

If the composition of a plutonium sample is known, it appears to be possible to check its mass with a neutron counting system, by comparing the measurements with coincidence calculations. After a short description of the measuring device used, the calculation methods are presented and their results are compared with the measured values for known sources. When induced fissions can be disregarded, the calculations are clearly excellent. When this is not the case — and this is the most important situation in practice — the effect of induced fission multiplication for a single spontaneous fission must first be evaluated. Comparison of the results then finally calculated with measurements seems very encouraging, even though the results are not as good as in the previous case. It seems that it would be appropriate to continue the study, with improvement of the means of measurement as a next step.

### **CONTROLE DE LA MASSE D'UN ECHANTILLON PLUTONIFERE PAR COINCIDENCES NEUTRONIQUES COMPTE TENU DES FISSIONS INDUITES.**

Quand la composition d'un échantillon plutonifère est connue, il paraît possible de contrôler la masse de plutonium à l'aide d'un système de comptage neutronique, par comparaison des mesures aux calculs de coïncidences. Après une description sommaire du dispositif physique utilisé, on présente les méthodes de calcul mises au point dont on compare les résultats aux valeurs mesurées pour des sources connues. Lorsque les fissions induites peuvent être négligées, les calculs sont manifestement excellents. Dans le cas contraire, le plus important en pratique, on doit d'abord évaluer l'effet de multiplication des fissions induites sur une fission spontanée. La comparaison aux mesures des résultats finalement calculés, bien que moins bonne que dans le cas précédent, paraît très encourageante. Il convient, semble-t-il, de prolonger l'étude dans le sens d'une amélioration des moyens de mesure.

## 1. INTRODUCTION

Un moyen de contrôler la masse d'un échantillon plutonifère de composition isotopique connue est basé sur l'utilisation d'un dispositif particulier de comptage de neutrons dont l'électronique établit la distribution du nombre de particules détectées dans une fenêtre temporelle: cette distribution mesurée est comparée à celle que l'on obtient par le calcul grâce à une modélisation du système physique [1].

Ce mémoire a pour objet essentiel la présentation simplifiée de la modélisation du système (utilisant largement les propriétés des fonctions génératrices de probabilités) et de résultats obtenus sur des échantillons réels.

On sait qu'un échantillon plutonifère est en général le siège de fissions spontanées, de réactions ( $\alpha, n$ ) et de fissions induites [2]. Ces dernières, négligeables dans le cas de très petites sources, peuvent être importantes pour les masses élevées. On distinguera dans notre exposé plusieurs parties:

- la description succincte du fonctionnement du dispositif de comptage;
- la modélisation sans participation des fissions induites;
- la prise en compte des fissions induites.

Dans les deux dernières parties, les valeurs calculées sont comparées aux résultats des mesures.

## 2. DESCRIPTIF DU DISPOSITIF EXPERIMENTAL

Une chambre en polyéthylène, dans laquelle est placé l'échantillon à contrôler, renferme 18 compteurs à  $^3\text{He}$  fonctionnant en collection de courant. Les impulsions logiques issues des amplificateurs-discriminateurs sont traitées dans un système électronique à registre à décalage (SCRD<sup>1</sup>) générant des fenêtres de comptage de largeur  $\tau$  selon deux modes possibles (méthode de Böhnel):

— Mode à autodéclenchement: toute impulsion neutronique déclenche l'ouverture d'une fenêtre avec un retard  $\theta$  nul ou très faible (inférieur à  $8 \mu\text{s}$ ).

— Mode à déclenchement aléatoire: l'ouverture des fenêtres est aléatoire par rapport aux instants de détection des neutrons. Ceci est obtenu en donnant au retard une valeur importante [1] ( $\theta \approx 1 \text{ ms}$ ).

Associé au SCRD, un ensemble d'échelles (deux groupes de neuf) fournit la distribution des neutrons comptés selon chaque mode de déclenchement.

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<sup>1</sup> SCRD: appareillage fabriqué et commercialisé par la société Novelec établie à Meylan (France).

### 3. MODELISATION DU SYSTEME DE DETECTION SANS FISSIONS INDUITES

#### 3.1. Mode à déclenchement aléatoire

On suppose ici que la source n'est constituée que d'un isotope spontanément fissile.

Pour une fission spontanée quelconque, la fonction génératrice<sup>2</sup> du nombre de neutrons émis est de la forme:

$$H(s) = a_0 + a_1 s^1 + a_2 s^2 + \dots + a_8 s^8 \quad (1)$$

où  $a_0, a_1, \dots$  sont des données de physique nucléaire.

Pour toute fenêtre de comptage de largeur  $\tau$ , la probabilité de détection  $p_\tau(t)$  d'un neutron, émis à l'instant  $t_0$  de l'intervalle d'émission  $T$  correspondant à cette fenêtre, s'écrit:

$$p_\tau(t) = \int_{(t)} \epsilon \alpha e^{-\alpha(t-t_0)} dt$$

avec

$\epsilon$ : efficacité de détection

$\alpha$ : inverse du temps de vie des neutrons.

D'autre part, la fonction génératrice du nombre de neutrons comptés dans une fenêtre pour une fission spontanée se produisant dans  $T$  a pour expression:

$$\frac{1}{T} \int_{(T)} H[(1-p) + ps] dt \quad (2)$$

où  $p$  est mis pour  $p_\tau(t)$ .

Si l'on considère maintenant l'éventualité de  $k$  fissions spontanées pendant le temps  $T$ , la fonction génératrice du nombre de neutrons comptés dans une fenêtre est donnée par:

$$\frac{(\lambda T)^k}{k!} e^{-\lambda T} \left\{ \frac{1}{T} \int_{(T)} H[(1-p) + ps] dt \right\}^k \quad (3)$$

$\lambda$  étant le taux de fissions spontanées.

<sup>2</sup> Dans tous nos calculs présentés ici on se limite au degré 8 (en général suffisant dans la pratique).

Avec  $k$  variant de 0 à l'infini, la fonction génératrice finale est:

$$\exp \left( \lambda \left\{ \int_{(T)} H[(1-p) + ps] dt - T \right\} \right) \quad (4)$$

expression que l'on peut mettre sous la forme d'un polynôme en  $s$ , soit:

$$A_0 + A_1 s + A_2 s^2 + \dots + A_8 s^8 \quad (5)$$

Les coefficients  $A_0, A_1, \dots, A_8$  sont les probabilités de compter 0, 1, ..., 8 neutrons dans une fenêtre de largeur  $\tau$  déclenchée aléatoirement.

### 3.2. Réaction spontanée équivalente: définition

Les sources et échantillons mesurés sont souvent en fait constitués de plusieurs isotopes spontanément fissiles et sont le siège de réactions ( $\alpha, n$ ). On peut prendre en compte tous ces isotopes  $j$  émetteurs de neutrons en définissant une réaction spontanée équivalente à l'ensemble de toutes les réactions différentes, de taux  $\lambda_j$ . La réaction équivalente est caractérisée par les formules suivantes,  $\lambda$  étant son taux:

$$\lambda = \sum_j \lambda_j \quad (6)$$

et

$$\left\{ \begin{array}{l} a_0 = \left( \sum_j a_{0j} \lambda_j \right) / \lambda \\ a_8 = \left( \sum_j a_{8j} \lambda_j \right) / \lambda \end{array} \right. \quad (7)$$

$a_{ij}$  étant la probabilité que l'isotope  $j$  émette spontanément  $i$  neutrons.

Dans le cas de réactions ( $\alpha, n$ ) on a:

$$\left\{ \begin{array}{l} a_{0j} = a_{2j} = \dots = a_{8j} = 0 \\ a_{1j} = 1 \end{array} \right. \quad (8)$$

### 3.3. Mode à autodéclenchement

Le déclenchement des fenêtres est provoqué par les impulsions neutroniques elles-mêmes (avec un retard  $\theta$  nul ou faible, fixé par l'opérateur).

#### 3.3.1. Cas d'un retard au déclenchement $\theta$ nul

Dès lors qu'une fenêtre est prise en considération, on admet qu'elle existe et qu'elle a été ouverte par un neutron détecté.

A) Ceci implique qu'une réaction spontanée équivalente s'est produite et que cette réaction a émis au moins un neutron. Les probabilités d'émission de 0, 1, ..., 8 neutrons par cette réaction sont alors:

$$\begin{cases} a'_0 = 0 \\ a'_1 = \frac{a_1}{a_1 + a_2 + \dots + a_8} \\ a'_8 = \frac{a_8}{a_1 + a_2 + \dots + a_8} \end{cases} \quad (9)$$

La fonction génératrice du nombre de neutrons détectés dans la fenêtre considérée est alors donnée par l'expression:

$$\frac{1}{T} \int_0^T H'[(1-p) + ps] dt \quad (10)$$

que l'on met sous la forme:

$$f_0 + f_1 s + f_2 s^2 + \dots + f_8 s^8$$

avec:

$$H'(s) = a'_0 + a'_1 s + a'_2 s^2 + \dots + a'_8 s^8$$

B) Ceci implique aussi qu'un neutron a certainement été détecté dans la fenêtre; les probabilités pour qu'un seul des neutrons émis, ou 2, ..., ou 8 soient comptés dans la fenêtre sont données par:  $f'_0, f'_1, \dots, f'_7$  (le neutron responsable de l'ouverture de la fenêtre n'étant pas compté dedans); on a:

$$\begin{cases} f'_0 = \frac{f_1}{f_0 + f_1 + \dots + f_8} \\ f'_7 = \frac{f_8}{f_0 + f_1 + \dots + f_8} \end{cases} \quad (11)$$

TABLEAU I. SOURCE DE CALIFORNIUM 252

Valeurs de m, nombre aléatoire de neutrons comptés dans une fenêtre	Probabilités p (m)					
	Pour le mode 1 (à déclenchement aléatoire)			Pour le mode 2 (à autodéclenchement)		
	Calculs	Mesures	Ecart (%)	Calculs	Mesures	Ecart (%)
0	0,4289	0,4280	0,2	0,4061	0,419	1,0
1	0,3438	0,3435	0,09	0,3475	0,3470	0,1
2	0,1564	0,1570	0,4	0,1665	0,1683	1,1
3	$5,238 \times 10^{-2}$	$5,293 \times 10^{-2}$	1,0	$5,826 \times 10^{-2}$	$6,010 \times 10^{-2}$	3,1
4	$1,431 \times 10^{-2}$	$1,408 \times 10^{-2}$	1,6	$1,653 \times 10^{-2}$	$1,713 \times 10^{-2}$	3,5
5	$3,359 \times 10^{-3}$	$3,494 \times 10^{-3}$	3,9	$4,017 \times 10^{-3}$	$4,335 \times 10^{-3}$	7,3
6	$7,003 \times 10^{-4}$	$8,316 \times 10^{-4}$	16	$8,639 \times 10^{-4}$	$9,365 \times 10^{-4}$	7,8
7	$1,325 \times 10^{-4}$	$1,373 \times 10^{-4}$	3,5	$1,682 \times 10^{-4}$	$2,039 \times 10^{-4}$	18
$\geq 8$	$2,312 \times 10^{-5}$	$3,433 \times 10^{-5}$	33	$3,013 \times 10^{-5}$	$3,589 \times 10^{-5}$	16
Moyenne	0,8931	0,8963	0,4	0,9481	0,9614	1,4
Ecart-type	9,9948	0,9980	0,3	1,0225	1,0321	0,9

Note: Paramètres de comptage:  $\epsilon = 8\%$  $\tau = 74,6 \times 10^{-6}$  s $\lambda_f = 4,015 \times 10^4$  fissions par seconde

T = 400 s.

Il est évident par ailleurs que la réaction certaine est accompagnée de  $k$  autres réactions possibles ( $k$  variant de 0 à l'infini). La fonction génératrice résultante du nombre de neutrons comptés dans la fenêtre considérée est alors obtenue en effectuant le produit polynômial de la fonction génératrice associée aux réactions possibles (équation 5) avec la fonction génératrice liée à la réaction certaine, soit:

$$(A_0 + A_1s + A_2s^2 + \dots + A_8s^8) \times (f'_0 + f'_1s + \dots + f'_7s^7)$$

### 3.3.2. Cas d'un retard au déclenchement $\theta$ non nul

Dans le cas où le retard  $\theta$ , avec lequel les impulsions neutroniques ouvrent les fenêtres de comptage, n'est pas nul, l'intervalle d'émission  $T$  est réduit d'un temps  $\theta$ , du côté de la borne finie, en ce qui concerne seulement la réaction certaine.

### 3.3. Résultats (tableaux I et II)

Les résultats donnés dans ces tableaux paraissent tout à fait satisfaisants. Il faut noter que, dans le cas de la source ( $\alpha, n$ ), les probabilités mesurées et calculées sont identiques pour les deux modes de déclenchement, ce qui était prévisible.

## 4. PRISE EN COMPTE DES FISSIONS INDUITES

### 4.1. Distribution du nombre de neutrons de fuite issus d'une fission spontanée

#### 4.1.1. Position du problème

On suppose que la source n'est constituée que de deux isotopes connus, fissiles, l'un spontanément, l'autre par fissions induites (par exemple  $^{239}\text{Pu}$ ). On se donne aussi la probabilité  $f$  qu'un neutron émis induise une fission.

#### 4.1.2. Hypothèses de base

Le puits de comptage utilisé ayant du cadmium sur sa surface interne, le retour des neutrons thermiques vers l'échantillon est très improbable, seules des fissions rapides existent au niveau de l'échantillon à contrôler. On peut donc considérer que la fission spontanée et les fissions induites associées ont lieu au même instant: super-fission spontanée.

Les réactions de capture étant supposées négligeables, le neutron n'a que deux possibilités: créer une fission induite ou fuir, donc être détectable, avec la probabilité  $(1 - f)$ .

TABLEAU II. SOURCE D'AMERICIUM-BORE

Valeurs de m, nombre aléatoire de neutrons comptés dans une fenêtre	Probabilités p (m)					
	Pour le mode 1 (à déclenchement aléatoire)			Pour le mode 2 (à autodéclenchement)		
	Calculs	Mesures	Ecart (%)	Calculs	Mesures	Ecart (%)
0	0,1251	0,1264	1,0	0,1251	0,1253	0,2
1	0,2601	0,2596	0,2	0,2601	0,2578	0,9
2	0,2703	0,2680	0,9	0,2703	0,2699	0,1
3	0,1873	0,1867	0,3	0,1873	0,1865	0,4
4	$9,730 \times 10^{-2}$	$9,704 \times 10^{-2}$	0,3	$9,730 \times 10^{-2}$	$9,797 \times 10^{-2}$	0,7
5	$4,044 \times 10^{-2}$	$4,169 \times 10^{-2}$	3,0	$4,044 \times 10^{-2}$	$4,170 \times 10^{-2}$	3,0
6	$1,401 \times 10^{-2}$	$1,440 \times 10^{-2}$	2,7	$1,401 \times 10^{-2}$	$1,477 \times 10^{-2}$	5,1
7	$4,160 \times 10^{-3}$	$4,543 \times 10^{-3}$	8,4	$4,160 \times 10^{-3}$	$4,475 \times 10^{-3}$	7,0
$\geq 8$	$1,081 \times 10^{-3}$	$1,518 \times 10^{-3}$	29	$1,081 \times 10^{-3}$	$1,532 \times 10^{-3}$	29
Moyenne	2,0779	2,0828	0,2	2,0779	2,0897	0,6
Ecart-type	1,4402	1,4516	0,8	1,4402	1,4519	0,8

Note: Paramètres de comptage:  $\epsilon = 8\%$  $\tau = 74,6 \times 10^{-6} \text{ s}$  $\lambda_a = 3,51 \times 10^5 \text{ réactions par seconde}$  $T = 400 \text{ s}$



#### 4.1.3. *Génération successive de neutrons*

Considérons une fission spontanée, la fonction génératrice du nombre de neutrons émis, dits «de première génération», est donnée par l'équation (1). La fonction génératrice du nombre de neutrons capturés pour cette première génération est donc, en remplaçant  $s$  par  $((1-f) + fs)$  dans l'équation (1):

$$C_1(s) = H((1-f) + fs)$$

Celle relative aux neutrons fuyards est au contraire donnée par:

$$F_1(s) = H(f + (1-f)s)$$

Considérant la fonction génératrice  $I(s)$  du nombre de neutrons émis par fission induite, analogue à l'équation (1), la fonction génératrice du nombre de neutrons émis, dits de «deuxième génération», est donnée par  $C_1(I(s))$ .

Ces neutrons de deuxième génération vont fuir à leur tour ou être capturés et ainsi de suite. Finalement, l'ensemble des neutrons détectables parce que fuyards est caractérisé par la fonction génératrice-produit, soit, si l'on se limite à  $n$  générations:

$$F_1(s) \times F_2(s) \times F_3(s) \times \dots \times F_n(s)$$

On peut en général donner à  $n$  une valeur largement inférieure à 20 dans les calculs pratiques.

#### 4.1.4. *Résultats (tableaux III et IV)*

Ces tableaux présentent des résultats de calcul obtenus pour différentes valeurs de  $f$ , probabilité de fission induite par neutron émis. On observe que le nombre moyen de neutrons de fuite calculé, par superfission, s'identifie parfaitement à celui donné par la formule (13) présentée ci-après, bien connue [3-5].

### 4.2. **Distribution du nombre de neutrons comptés dans une fenêtre compte tenu des fissions induites**

#### 4.2.1. *Position du problème*

Un échantillon à vérifier, de masse et de composition isotopique censées connues, peut être le siège de fissions induites non négligeables. Nous avons déjà défini une «réaction spontanée équivalente» (sous 3.2): si l'on parvient à définir une «fission induite équivalente» et à évaluer la probabilité  $f$  pour qu'un neutron émis provoque une telle fission, on sera ramené au cas étudié sous 4.1 et une évaluation de la distribution du nombre de neutrons comptés dans une fenêtre sera possible.

TABLEAU III. CALCUL DES PROBABILITES D'EMISSION NEUTRONIQUE PAR SUPERFISSION DU  $^{240}\text{Pu}$  POUR UNE PROBABILITE DE FISSION INDUITE DU  $^{239}\text{Pu}$  ( $f = 0,100$ )<sup>a</sup>

i	Données utilisées dans le calcul		Résultats		
	Spectre de fission induite du $^{239}\text{Pu}$	Spectre de fission spontanée du $^{240}\text{Pu}$	2 générations	5 générations	20 générations
0	0,00	0,0677	0,0746	0,0688	0,0686
1	0,09	0,2120	0,2119	0,1961	0,1957
2	0,13	0,3479	0,2919	0,2722	0,2716
3	0,56	0,2639	0,2042	0,1964	0,1962
4	0,11	0,0924	0,0964	0,1023	0,1024
5	0,06	0,0149	0,0538	0,0647	0,0650
6	0,05	0,0011	0,0331	0,0427	0,0430
7	0	0,0001	0,0176	0,0249	0,0251
8	0	0	0,0089	0,0144	0,0146
≥ 9	0	0	0,0076	0,0175	0,0178
Moyenne	3,07	2,1509	2,5301	2,7857	2,7934
$C_v^2$		1,8616	3,4660	4,5242	4,5577
$C_v^3$		0,8080	3,7917	6,2203	6,3040

<sup>a</sup> Evolution en fonction du nombre de générations associées à la fission induite.

#### 4.2.2. Fission induite équivalente

Désignant par  $\sigma$  la section efficace microscopique d'un isotope pour une certaine réaction, si une cible de N atomes de cet isotope est plongée dans un flux  $\varphi$  supposé uniforme, le taux de réactions est donné par:  $\varphi\sigma N$ . Dans le cas de notre source supposée constituée de plusieurs isotopes fissiles j, le taux des fissions induites pour chacun d'eux est donné par  $\varphi\sigma_j N_j$ , soit au facteur  $\varphi$  près:  $\lambda_j = \sigma_j N_j$ .

Pour chaque isotope,  $\sigma_j$  et la distribution du nombre de neutrons émis par fission sont connues,  $N_j$  est censé connu, et l'on peut donc définir une «fission induite équivalente» en utilisant les formules (6) et (7) indiquées sous 3.2.

TABLEAU IV. CALCULS DES MOMENTS D'ORDRE 1, 2 ET 3 ASSOCIES A UNE SUPERFISSION DU  $^{240}\text{Pu}$  POUR DIFFERENTES VALEURS DE  $f$  (cas du  $^{239}\text{Pu}$ )

$f$	$C_\nu^1 = \bar{\nu}_{\text{res}}$	$C_\nu^2 = \left( \frac{\nu(\nu-1)}{2} \right)_{\text{res}}$	$C_\nu^3 = \left( \frac{\nu(\nu-1)(\nu-2)}{6} \right)_{\text{res}}$
0	2,1509	1,8616	0,8080
0,002	2,1599	1,8992	0,8746
0,004	2,1689	1,9371	0,9420
0,006	2,1781	1,9754	1,0101
0,008	2,1874	2,0141	1,0791
0,010	2,1968	2,0532	1,1489
0,012	2,2064	2,0927	1,2196
0,016	2,2258	2,1730	1,3640
0,018	2,2357	2,2139	1,4377
0,020	2,2458	2,2552	1,5126
0,040	2,3539	2,6985	2,3350
0,060	2,4784	3,2108	3,3392
0,100	2,7934	4,5577	6,3040
0,150	3,3881	7,4145	14,0930
0,200	4,4349	13,6134	36,2196

Note: Le calcul a été effectué en prenant 20 générations successives de neutrons. Ces résultats sont à comparer avec ceux donnés dans la référence [3].

#### 4.2.3. Détermination de la probabilité $f$ pour qu'un neutron émis induise une fission

Plutôt que de faire une étude théorique physique, longue et difficile, de l'ensemble du système (source et détecteur), on détermine  $f$  pour l'échantillon plutonifère considéré en utilisant le nombre moyen  $m$  de neutrons réellement comptés dans une fenêtre, en mode aléatoire.

TABLEAU V. ECHANTILLON METALLIQUE DE 3,2 g  $^{240}\text{Pu}$  EQUIVALENT (f mesuré = 0,001)

Valeurs de m, nombre aléatoire de neutrons comptés dans une fenêtre	Probabilités p (m)			
	Pour le mode 1 (à déclenchement aléatoire)		Pour le mode 2 (à autodéclenchement)	
	Calculs	Mesures	Calculs	Mesures
				Ecart (%)
0	0,9726	0,9730	0,9304	0,9255
1	0,0259	0,0255	0,0658	0,0690
2	0,0014	0,0014	0,0035	0,0051
3	0,0001	0,0001	0,0002	0,0003
4	0	0	0,0001	0,0001
5	0	0	0	0
6	0	0	0	0
7	0	0	0	0
$\geq 8$	0	0	0	0
				0,5 4,9 46,1 50,0 — — — — —
Moyenne	0,0291	0,0290	0,0736	0,080
Ecart-type	0,1778	0,176	0,2768	0,294
				8,7 6,2

Note: Paramètres de comptage:  $\epsilon = 12\%$  $\tau = 75,8 \times 10^{-6} \text{ s}$  $\theta = 2,4 \times 10^{-6} \text{ s}$  $T = 400 \text{ s}$

TABLEAU VI. ECHANTILLON METALLIQUE DE 108,8 g  $^{240}\text{Pu}$  EQUIVALENT (f mesuré = 0,127)

Valeurs de m, nombre aléatoire de neutrons comptés dans une fenêtre	Probabilités p (m)					
	Pour le mode 1 (à déclenchement aléatoire)			Pour le mode 2 (à autodéclenchement)		
	Calculs	Mesures	Ecart (%)	Calculs	Mesures	Ecart (%)
0	0,2723	0,2839	4,2	0,2472	0,2361	4,7
1	0,3216	0,3175	1,3	0,3146	0,3000	4,9
2	0,2193	0,2106	4,1	0,2281	0,2237	2,0
3	0,1124	0,1085	3,6	0,1231	0,1287	4,5
4	0,0479	0,0481	0,4	0,0549	0,0633	15,3
5	0,0179	0,0196	9,3	0,0213	0,0285	33,8
6	0,0060	0,0075	25,7	0,0074	0,0120	62,1
7	0,0019	0,0028	45,7	0,0024	0,0048	99,1
$\geq 8$	0,0007	0,0015	120,8	0,0010	0,0030	204,4
Moyenne	1,4336	1,4312	0,2	1,5358	1,655	7,7
Ecart-type	1,3146	1,3610	3,5	1,3572	1,476	8,8

Note: Paramètres de comptage:  $\epsilon = 12\%$   
 $\tau = 75,8 \times 10^{-6} \text{ s}$   
 $\theta = 2,4 \times 10^{-6} \text{ s}$   
 $T = 400 \text{ s}.$

On sait en effet que:

$$m = \lambda_{sp} \cdot \bar{\nu}_{res} \cdot \tau \cdot \epsilon \quad (12)$$

$$\bar{\nu}_{res} = \bar{\nu}_{sp} \cdot \frac{1-f}{1-f \cdot \bar{\nu}_{ind}} \quad (13)$$

où

$\lambda_{sp}$  = taux de réactions spontanées équivalentes

$\bar{\nu}_{sp}$  = nombre moyen de neutrons émis par réaction spontanée

$\bar{\nu}_{ind}$  = nombre moyen de neutrons émis par fission induite

$\bar{\nu}_{res}$  = nombre moyen de neutrons «de fuite» par réaction spontanée équivalente, compte tenu de la cascade de fissions induites.

C'est là un système de deux équations à deux inconnues:  $\bar{\nu}_{res}$  et  $f$ .

#### 4.2.4. Résultats (tableaux V et VI)

Deux échantillons métalliques ont été testés, l'un très petit, siège de peu de fissions induites, l'autre plus important. On remarque une bonne cohérence entre les moments d'ordre 1 et 2 associés aux probabilités de détection mesurées et calculées, pour les deux modes de déclenchement considérés.

## 5. CONCLUSION

En ce qui concerne la formulation sans fissions induites, appliquée à de petites sources ( $^{252}\text{Cf}$ , Am-B), les résultats sont manifestement excellents. Quant à la prise en compte des fissions induites dans le cas de sources importantes et volumineuses, les résultats calculés présentent par rapport aux valeurs mesurées des écarts plus importants. Ceci est probablement dû au fait que les hypothèses de base prises pour notre calcul sont trop simplistes, à savoir:

- efficacité uniforme;
- réactions de capture négligées;
- fissions induites supposées rapides donc instantanées, alors qu'il y a probablement des fissions thermiques;
- temps mort électronique supposé nul.

Quoi qu'il en soit, les résultats paraissent encourageants; il convient de prolonger cette étude, sans doute dans le sens d'une amélioration des moyens de mesure.

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# FURTHER DEVELOPMENTS ON PLUTONIUM MEASUREMENTS BY PASSIVE NEUTRON ASSAY

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## Abstract

### FURTHER DEVELOPMENTS ON PLUTONIUM MEASUREMENTS BY PASSIVE NEUTRON ASSAY.

Numerical procedures were developed for determination of the source neutron multiplicity distribution caused by a fission chain and for dead time correction of neutron pulse trains. The induced fission chain is started by a spontaneous fission event and is followed by a computer, generation after generation, yielding the correct neutron multiplicity distribution of the leakage neutrons for a point source model. The Monte Carlo computer code for the dead time correction uses an updating dead time counting process and assumes that the pulses are randomly distributed inside an observation interval.

## 1. INTRODUCTION

The Euratom Time Correlation Analyser has been tested for many years for the non-destructive assay of plutonium [1]. It has been used to correct for the non-linearity of the neutron coincidence rate caused by waste matrices [2, 3] or to correct for neutron multiplication of large Pu quantities [4].

The instrumental basis of the Euratom Time Correlation Analyser is the opening of a gate of length  $g$  either sequentially (pulse fluctuation analysis) or by each pulse in the pulse train (pulse to pulse analysis) and thereafter the counting of the number of pulses,  $k$ , which appear within the gate length. This leads to count rate distributions  $R_{k,g}$ .

The first problem to become apparent with the extension of measurements up to 1 kg Pu was the effect of detector dead time on the  $R_{k,g}$  distributions. An empirical correction has been used by simulating the effect of the dead time on a true  $R_{k,g}$  distribution using Monte Carlo techniques. For count rates between 5 and 75 kHz the empirical corrections were evaluated as a function of three parameters:

- (i) Ratio of single events/total events
- (ii) Ratio of coincident neutrons/coincident events
- (iii) Total count rate.

To correct the measured  $R_{k,g}$  distributions, interpolation between the derived correction points was performed by fitting polynomials to the deduced behaviour as a function of these three parameters. In order to calculate the Pu content from the measured  $R_{k,g}$  distributions, the interpretation model utilizes among other things the neutron multiplicity distributions  $P_j$  and their modifications due to neutron multiplication. These distributions were obtained from point source Monte Carlo calculations with the approximation that the sample consists of  $^{239}\text{Pu}$ ,  $^{240}\text{Pu}$  and an  $(\alpha, n)$  source, and that the probability of provoking an induced fission has the unique value  $p$  [5, 6].

In this paper, new, convenient methods are presented for determining both the source neutron multiplicity distribution and the dead time correction.

## 2. NEUTRON MULTIPLICITY DISTRIBUTION

The mathematical model developed for the calculation of the neutron multiplicity distribution of a point source subdivides the induced fission chain into a well defined sequence of neutron generations. After a sequence of  $\ell$  generations, the physical state of the induced fission chain is described by the fission neutron state matrix  $\vec{S}(\ell)$ . The element  $S_{n,\nu}(\ell)$  is defined as the probability that the system consists of: firstly,  $\nu = 0, 1, 2, \dots$  leakage neutrons, which already leaked out from the point source and cannot induce further fissions; and secondly,  $n = 0, 1, 2, \dots$  fission neutrons produced in the previous generation which still have the chance either to leak out from the point source or to induce further fissions.

After this, the system will change to an intermediate fission state. The fraction of fission neutrons leaking out increases the number of leakage neutrons, and the rest induce further fissions. This intermediate state is described by the fission state matrix  $\vec{F}(\ell)$ . The element  $F_{i,\nu^*}(\ell)$  is defined as the probability of counting  $\nu^* = 0, 1, 2, \dots$  leakage neutrons and  $i = 0, 1, 2, \dots$  fissions. The accumulated leakage neutrons and the neutrons generated by the last fissions belong to the next neutron generation described by the state matrix  $\vec{S}(\ell + 1)$ . In this way the state matrix can be obtained by a recurrence formula.

Let  $B_{k,n}$  be the probability that  $k$  of the  $n$  fission neutrons leak out and  $n - k$  induce fissions; then

$$B_{k,n} = \binom{n}{k} (1-p)^k p^{n-k}$$

where  $p$  is the probability that a neutron induces a fission.

Let  $P_{n,i}$  be the probability that  $n$  neutrons are generated by  $i$  induced fissions, each belonging to the same generation; then

$$P_{n,i} = \sum_{m=0}^n P_{m,j} P_{n-m,i-j} \quad 0 < j < i$$

The initial values are:  $P_{n,0} = 0$  if  $n > 0$ ,  $P_{0,0} = 1$ .  $P_{n,1}$  is the neutron multiplicity distribution for induced fission.

The state matrices are calculated by summation of the singular transition probabilities weighted with the matrix element of the previous state:

$$F_{i,\nu^*}(\ell) = \sum_{n=i}^{i+\nu^*} B_{n-i,n} S_{n,\nu^*-(n-i)}(\ell)$$

$$S_{n,\nu}(\ell+1) = \sum_{i=0}^{\infty} P_{n,i} F_{i,\nu}(\ell)$$

At the beginning the neutron state matrix  $\bar{S}(1)$  is described by the spontaneous fission neutron multiplicity distribution  $P_n$ :

$$S_{n,0}(1) = P_n \quad \text{and} \quad S_{n,\nu}(1) = 0 \quad \text{if} \quad 0 < \nu$$

where  $P_n$  is the probability that  $n$  neutrons are generated by a spontaneous fission event.

After a number of recurrent steps, each of the matrix elements  $S_{n,\nu}(\dots)$  for  $0 < n$  becomes negligible because the probability for further fission in this branch becomes negligible. The first line of the state matrix  $\bar{S}(\dots)$ , namely  $S_{0,\nu}(\dots)$ , is the neutron multiplicity distribution.

The number of neutron generations required depends strongly on the numerical value of  $p$ . The recurrence procedure is terminated if the sum

$$\sum_{\nu=0}^{\infty} S_{0,\nu}(\dots)$$

has reached a value very close to 1.

For the execution of the calculations, at least a normal personal computer is necessary. A complete Fortran program is presented in the Appendix.

### 3. DEAD TIME CORRECTION

In Fig. 1 some  $R_{k,g}$  distributions are plotted as a function of  $k$ . For simplicity these distributions are normalized to one gate.  $R_{k,g}$  is then the probability of finding  $k$  pulses in the time interval of length  $g$ . The experiments with and without dead time were simulated by the Monte Carlo technique [7]. A Pu metal point source of 815 g  $^{240}\text{Pu}$  with an induced fission probability of 12% was analysed. The overall detection probability was set to 17% and the die-away

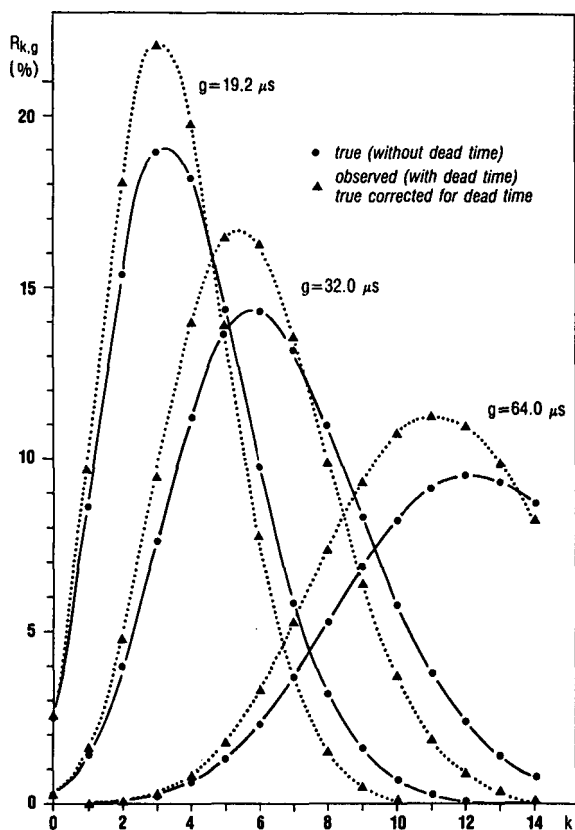


FIG. 1.  $R_{k,g}$  distributions from pulse to pulse analysis.

time to  $45 \mu\text{s}$ . This led to a total count rate of 200 000 counts/s. The neutron detector head consisted of six groups of proportional counters functioning as six equivalent separate channels, each having a dead time of  $3.3 \mu\text{s}$ . For the neutron detection, an updating dead time counter was selected. Only those neutrons contribute to the pulse train which have a longer period from the previous neutron of the same channel than the dead time.

The multiplet distributions of Fig. 1 demonstrate a strong dead time effect. The true distributions are shifted to the left and accumulated at lower  $k$ -values, producing the observed distributions.

To obtain the observed  $R_{k,g}^*$  distribution from the true  $R_{k,g}$  distribution, each of the  $R_{k,g}$  elements is to be substituted by a distribution  $Z_{k^*,k,g}$  which describes the probabilities that from  $k$  only  $k^*$  pulses will be observed:

$$R_{k^*,g}^* = \sum_{k=k^*}^{\infty} Z_{k^*,k,g} R_{k,g}$$

By inversion of the matrix  $\bar{Z}$ , the true  $R_{k,g}$  distribution is determined from the observed one.

Neutrons appearing during the dead time immediately before the observation interval contribute to the dead time effect. The matrix  $\bar{Z}$  is slightly modified by this effect. Limiting the loss to a maximum of one pulse,

$$(1 - PA_{k,g})Z_{k^*,k,g} + PA_{k,g}Z_{k^*+1,k,g} \Rightarrow Z_{k^*,k}$$

where  $P$  is the probability of finding any neutron before the observation interval and  $A_{k,g}$  is the probability of losing one pulse owing to such a neutron.

For the determination of  $\bar{Z}$  and  $\bar{A}$ , a Monte Carlo computer code has been developed. The Fortran program is presented in the Appendix. The calculations were performed for an updating dead time counting process and assume that the pulses are randomly distributed inside the gate.

It was found that it is not necessary to take into consideration a neutron correlation effect. In a practical case, the detector system is subdivided into channels with 2-3% detection efficiency per channel. Any neutron correlation inside a channel is sufficiently weak that a random distribution of pulses for dead time correction can be used. In Fig. 1 the true distributions corrected for dead time fit perfectly the observed distributions. Any differences are smaller than the plotted symbols.

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## Appendix

```

C-----
C---- NEUTRON MULTIPLICITY DISTRIBUTION -----
C-----
      SUBROUTINE NEMUDI(DP,DPSF,DPIF,DPFC,DPR,NGN)
      IMPLICIT REAL*8 (D)
      DIMENSION DPSF(12),DPIF(12),DPFC(108),
      *          DV1(108),DV2(108),DV3(108)
      VIRTUAL   DMP(36,12),DMB(36,36),
      *          DMS(36,108),DMF(12,108)
C-----
C      "DP" PROBABILITY TO PROVOKE INDUCED FISSION
C      "DPSF(N)" NEUTRON MULTIPL. OF SPONT. FISS. ISOTOPE
C      "DPIF(N)" NEUTRON MULTIPL. OF INDUC. FISS. ISOTOPE
C      "DPFC(L)" NEUTRON MULTIPL. AT THE END OF THE CHAIN
C      "DPR" SUM OF "DPFC(L)" OVER L /NEARLY TO 1.0
C      "NGN" NUMBER OF GENERATION FOLLOWED
C-----
      1 LMAX=108
      NMAX=36
      IMAX=12
      DMIN=1.0D-14
      DSF=0.0D+00
      DIF=0.0D+00
      DO 11 N=1,12
      DSF=DSF+DPSF(N)
      11 DIF=DIF+DPIF(N)
      DO 12 N=1,12
      DPSF(N)=DPSF(N)/DSF
      12 DPIF(N)=DPIF(N)/DIF
C-----
      2 DO 21 I=1,IMAX
      DO 21 N=1,NMAX
      21 DMP(N,I)=0.0D+00
      DMP(1,1)=1.0D+00
      DO 22 N=1,12
      22 DMP(N,2)=DPIF(N)
      I=2
      I1=1
      29 I1=I1+1
      I2=I1-1
      DO 23 J=1,2
      I=I+1
      IF (I.GT.IMAX) GO TO 3
      I2=I2+1
      DO 24 N=1,NMAX
      24 DV1(N)=DMP(N,I1)
      DO 25 N=1,NMAX
      25 DV2(N)=DMP(N,I2)
      DO 26 N1=1,NMAX
      N2X=NMAX-N1+1
      DO 26 N2=1,N2X
      N=N1+N2-1
      26 DMP(N,I)=DMP(N,I)+DV1(N1)*DV2(N2)
      23 CONTINUE
      GO TO 29
C-----
      3 DV1(1)=1.0D+00
      DV2(1)=1.0D+00
      DV3(1)=1.0D+00

```

```

DO 31 LD=2,NMAX
DV1(LD)=DV1(LD-1)*(1.0D+00-DP)
DV2(LD)=DV2(LD-1)*DP
31 DV3(LD)=0.0D+00
DO 32 N=1,NMAX
DO 32 LD=1,NMAX
32 DMB(LD,N)=0.0D+00
DMB(1,1)=1.0D+00
DO 33 N=2,NMAX
N2=N
DMB(1,N)=DV2(N2)
DOLD=1.0D+00
DO 33 LD=2,N
DNEW=DV3(LD)
DV3(LD)=DOLD+DV3(LD)
DOLD=DNEW
N2=N2-1
33 DMB(LD,N)=DV3(LD)*DV1(LD)*DV2(N2)
C-----
4 DO 49 L=1,LMAX
DO 49 N=1,NMAX
49 DMS(N,L)=0.0D+00
DO 48 N=1,12
48 DMS(N,1)=DPSF(N)
NGN=0
40 NGN=NGN+1
TYPE *, ' NGN',NGN
DO 41 L=1,LMAX
DO 41 I=1,IMAX
41 DMF(I,L)=0.0D+00
DO 42 L=1,LMAX
DO 43 N=1,NMAX
43 DV1(N)=DMS(N,L)
DO 44 N=2,NMAX
IF (DV1(N).LE.DMIN) GO TO 44
LDX=MAX0((N-IMAX+1),1)
DO 45 LD=LDX,N
45 DV2(LD)=DMB(LD,N)
DO 46 LD=LDX,N
LL=L+LD-1
IF (LL.GT.LMAX) GO TO 44
D=DV1(N)*DV2(LD)
IF (D.LE.DMIN) GO TO 46
II=N-LD+1
DMF(II,LL)=DMF(II,LL)+D
46 CONTINUE
44 CONTINUE
42 DMF(1,L)=DMF(1,L)+DV1(1)
C-----
5 COUNT=0.0
DO 51 L=1,LMAX
DO 51 N=1,NMAX
51 DMS(N,L)=0.0D+00
DO 52 L=1,LMAX
DO 53 I=1,IMAX
53 DV1(I)=DMF(I,L)
DO 54 I=2,IMAX
IF (DV1(I).LE.DMIN) GO TO 54
DO 55 N=1,NMAX
55 DV2(N)=DMP(N,I)
DO 56 N=1,NMAX
D=DV1(I)*DV2(N)

```

```

      IF (D.LE.DMIN) GO TO 56
      DMS(N,L)=DMS(N,L)+D
      COUNT=COUNT+1.0
56  CONTINUE
54  CONTINUE
52  DMS(1,L)=DMS(1,L)+DV1(1)
      IF (COUNT.NE.0.0) GO TO 40
C-----
      6 DPR=0.0D+00
      DO 61 L=1,LMAX
      DPFC(L)=DMS(1,L)
61  DPR=DPR+DPFC(L)
C-----
      RETURN
      END
C-----
      DP 0.12000
DPSF 0.06126 0.23316 0.34344 0.24849 0.09354 0.01819
      0.00183 0.00009 0.00000 0.00000 0.00000 0.00000
DPIF 0.00710 0.06383 0.21212 0.33258 0.26103 0.10161
      0.01976 0.00188 0.00009 0.00000 0.00000 0.00000
           1         2         3         4         5         6
DPFC 0.06146 0.20730 0.27534 0.19269 0.09300 0.04984
      0.03391 0.02324 0.01595 0.01143 0.00839 0.00622
      0.00468 0.00356 0.00274 0.00212 0.00165 0.00129
      0.00102 0.00081 0.00064 0.00051 0.00041 0.00033
      0.00027 0.00022 0.00018 0.00014 0.00012 0.00009
      0.00008 0.00006 0.00005 0.00004 0.00004 0.00003
      0.00002 0.00002 0.00002 0.00001 0.00001 0.00001
      0.00001 0.00001 0.00001 0.00000 0.00000
DPR 0.9999999984992      NGN      27
C-----

```

```

C-----
C--- DEAD TIME CORRECTION ---
C-----
      SUBROUTINE DETICO
      COMMON      TE,LE,NZ,KZ,NH,VPH(16),VTH(16),STA,
*                AZ(22,16),ZZ(22,22,16)
      DIMENSION  PH(16),PC(16),TH(16),LH(16),
*                PT(22),QT(22),KD(22,16)
C-----
C GATE LENGTH="TE"*L L=1,2,...LE"      "TE"[MICRO-SEC]
C      "NZ" MAXIMAL NUMBER OF TRUE-PULSES
C      "KZ" MAXIMAL NUMBER OF LOSS-PULSES !!!
C      "NH" NUMBER OF THE DETECTOR-CHANNELS
C "VPH(I)" DETECT. EFF. OF CHANNEL I      /I=1,2,...NH
C "VTH(I)" DEAD TIME OF CHANNEL I      [MICRO-SEC]
C      "STA" IS THE NUMBER OF HISTORIES
C "AZ(N,L),ZZ(K,N,L)" RESULT N:TRUE K:LOSS!!! L:GATE
C      RAFU: UNIFORMLY DISTR. RANDOM NUMB. 0.<RAFU<1.
C-----
      20 SCH=0.0
      DO 201 IC=1,NH
201  SCH=SCH+VPH(IC)
      SP=0.0
      DO 202 IC=1,NH
      P=VPH(IC)/SCH

```



```

      PC(IC)=P
      SP=SP+P
202  PH(IC)=SP
      PH(NH)=1.0
      DO 203 L=1,16
      DO 203 N=1,22
      AZ(N,L)=0.0
      DO 203 K=1,22
203  ZZ(K,N,L)=0.0
      STAI=0
209  STAI=STAI+1.0
      TYPE *, ' STA',STAI
      21  DO 211 L=1,LE
      DO 211 N=1,NZ
211  KD(N,L)=1
      DO 212 IC=1,NH
      TH(IC)=0.0
212  LH(IC)=NZ+1
      ST=0.0
      M=NZ
      PT(NZ)=0.0
      QT(NZ)=TE
      22  R=RAFU
      ST=ST+(1.0-ST)*(1.0-R**(1.0/M))
      R=RAFU
      IC=0
221  IC=IC+1
      IF (R.GT.PH(IC)) GO TO 221
      TT=TH(IC)
      NN=LH(IC)
      TH(IC)=ST
      LH(IC)=M
      T=VTH(IC)
      23  DO 231 N=M,NN-1
      VV=(ST-PT(N))*QT(N)
      RR=AMAX1(0.005,VV)
      LL=T/RR
      IF (LL.EQ.0) GO TO 231
      LL=MIN0(LL,LE)
      DO 232 L=1,LL
232  AZ(N,L)=AZ(N,L)+PC(IC)*(T-L*VV)/T
231  CONTINUE
      24  IF (TT.EQ.0.0) GO TO 229
      V=ST-TT
      DO 241 N=NN,NZ
      VV=V*QT(N)
      RR=AMAX1(0.005,VV)
      LL=T/RR
      IF (LL.EQ.0) GO TO 241
      LL=MIN0(LL,LE)
      DO 242 L=1,LL
242  KD(N,L)=KD(N,L)+1
241  CONTINUE
229  M=M-1
      PT(M)=ST
      QT(M)=TE/(1.0-ST)
      IF (M.NE.0) GO TO 22
249  DO 248 L=1,LE
      DO 248 N=1,NZ
      K=KD(N,L)
      IF (K.GT.KZ+1) GO TO 248
      ZZ(K,N,L)=ZZ(K,N,L)+1.0

```

```

248 CONTINUE
    IF (STAI.LT,STA) GO TO 209
29  DO 291 L=1,LE
    DO 291 N=1,NZ
    AZ(N,L)=AZ(N,L)/STA
    DO 291 K=1,KZ+1
291  ZZ(K,N,L)=ZZ(K,N,L)/STA
C-----
      RETURN
      END
-----
TE-LE-NZ-KZ-NH 6.4000      16      22      21      6
      VPH 0.1700 0.1700 0.1700 0.1700 0.1700 0.1700..
      VTH 3.3000 3.3000 3.3000 3.3000 3.3000 3.3000..
GL 32.00  N/K      0      1      2      3      4
AZ 0.0086 1 ZZ 1.0000 0.0000 0.0000 0.0000 0.0000..
AZ 0.0170 2 ZZ 0.9672 0.0328 0.0000 0.0000 0.0000..
AZ 0.0254 3 ZZ 0.9043 0.0941 0.0015 0.0000 0.0000..
AZ 0.0338 4 ZZ 0.8160 0.1751 0.0088 0.0001 0.0000..
AZ 0.0422 5 ZZ 0.7110 0.2603 0.0277 0.0010 0.0000..
AZ 0.0500 6 ZZ 0.5971 0.3373 0.0608 0.0046 0.0001..
AZ 0.0580 7 ZZ 0.4817 0.3921 0.1116 0.0139 0.0007..
AZ 0.0659 8 ZZ 0.3744 0.4193 0.1698 0.0332 0.0032..
AZ 0.0740 9 ZZ 0.2793 0.4155 0.2301 0.0647 0.0096..
AZ 0.0816 10 ZZ 0.2009 0.3832 0.2825 0.1076 0.0228..
AZ 0.0893 11 ZZ 0.1380 0.3328 0.3155 0.1592 0.0458..
AZ 0.0972 12 ZZ 0.0906 0.2715 0.3259 0.2106 0.0795..
AZ 0.1045 13 ZZ 0.0565 0.2086 0.3111 0.2530 0.1233..
AZ 0.1122 14 ZZ 0.0345 0.1496 0.2783 0.2771 0.1709..
AZ 0.1195 15 ZZ 0.0199 0.1025 0.2296 0.2812 0.2144..
AZ 0.1267 16 ZZ 0.0110 0.0668 0.1761 0.2638 0.2450..
AZ 0.1335 17 ZZ 0.0057 0.0410 0.1284 0.2266 0.2564..
AZ 0.1407 18 ZZ 0.0028 0.0239 0.0875 0.1824 0.2451..
AZ 0.1475 19 ZZ 0.0013 0.0131 0.0552 0.1362 0.2178..
AZ 0.1541 20 ZZ 0.0006 0.0067 0.0328 0.0952 0.1772..
AZ 0.1610 21 ZZ 0.0003 0.0033 0.0183 0.0616 0.1348..
AZ 0.1678 22 ZZ 0.0001 0.0013 0.0095 0.0372 0.0944..
-----

```

# A LIQUID SCINTILLATOR NEUTRON COINCIDENCE COUNTER (LSNCC) FOR PLUTONIUM ASSAY\*

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## Abstract

### A LIQUID SCINTILLATOR NEUTRON COINCIDENCE COUNTER (LSNCC) FOR PLUTONIUM ASSAY.

Neutron coincidence counting (NCC) is a widely used technique for the assay of fissionable or spontaneously fissioning isotopes. The instrument presently in general use by the IAEA is the high level neutron coincidence counter (HLNCC), a  $^3\text{He}$  based polyethylene moderated well counter. Although this detector provides accurate results for well defined samples with low neutron multiplication, the detector lacks the ability to provide satisfactory results for samples of an unknown composition. The basic limitation of the detector results from its low neutron detection efficiency (17.5% for HLNCC-II). In order to obtain information concerning sample neutron multiplication, a high efficiency liquid scintillation detector has been constructed for plutonium assay. One of the major attributes of liquid scintillator neutron coincidence counting is that the technique is based on absolute nuclear data, as opposed to calibration standards. Preliminary results for  $\text{PuO}_2$  samples of up to 200 g have indicated that the technique provides measurements accurate to within a few per cent within this mass range. These results are presented and discussed. Progress in dead-time corrections and neutron multiplication studies is also outlined.

## 1. INTRODUCTION

The concept of neutron coincidence counting (NCC) has been applied with considerable success to the measurement of plutonium, especially in the waste product from reprocessing plants. The technique makes use of the multiplicity of the prompt neutron emission accompanying the spontaneous fission of  $^{240}\text{Pu}$ . These neutrons are emitted within  $10^{-16}\text{s}$  of spontaneous fission, and this time correlation may be exploited to enable the selective detection of spontaneous fission events within a large neutron background. This fission rate information can then be related to the mass of  $^{240}\text{Pu}$ , and with additional isotopic information, a full plutonium assay may be obtained.

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\* Research performed under contract for the Foreign Affairs Department, Australia.

Many safeguards instruments have been based on the principle of NCC. The most notable of these, the high level neutron coincidence counter (HLNCC), has achieved a precision as high as 1% [1]. However, this precision is dependent on the provision of standards with similar properties to the samples being assayed. Furthermore, the accuracy of the HLNCC, as with all neutron coincidence counters, is limited by neutron multiplication effects. Existing procedures for treating neutron multiplication generally require a large number of standards, and are only suitable for samples of well defined composition [2,3].

To make a significant improvement in neutron coincidence counters for plutonium assay, attention is turning towards high efficiency neutron detection systems [4]. This trend has been accompanied by the development of a more theoretical analysis of time correlated neutron detection [5]. In keeping with this direction of NCC, we have developed a technique that is based on a high efficiency liquid scintillation detector [6,7]. This type of detector has been found to offer distinct advantages over the more conventional detector.

## 2. PROTOTYPE LIQUID SCINTILLATOR NEUTRON COINCIDENCE COUNTER (LSNCC)

A prototype liquid scintillation detector for safeguards purposes has been designed and constructed at the A.A.E.C. A cross section of the LSNCC is shown in Figure 1. The detector contains 70 litres of NE323, a tri-methyl benzene based liquid scintillator, containing 0.5% loading of gadolinium by weight. The development of the technique followed from previous measurements of the average number of prompt neutrons ( $\bar{\nu}_p$ ) emitted in fission [8].

### 2.1. Neutron Detection Properties of the LSNCC

Neutron detection within the scintillator is viewed by six photomultiplier tubes, arranged in two groups of three. A coincidence signal between the two groups is required for a true event. This condition is imposed to eliminate the small double pulsing probability associated with the photomultiplier tubes. The time distribution for neutron capture within the scintillator is shown in Figure 2. The average neutron lifetime is 12.3  $\mu$ s, with 99.8% of the neutrons being detected within 60  $\mu$ s.

The energy dependence of the scintillator has been determined by a computer calculation [9], and is shown in Figure 3. Assuming a

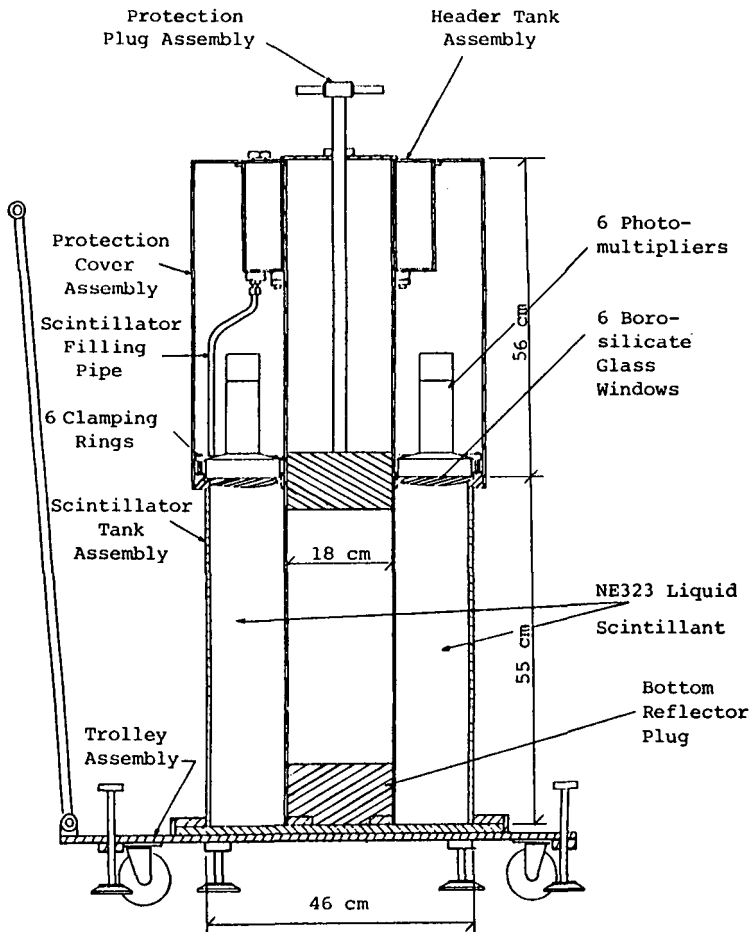


FIG. 1. Cross section of prototype liquid scintillator neutron coincidence counter (LSNCC).

Maxwellian distribution of the form  $\sqrt{E} \exp(-E/T_M)$  with  $T_M = 1.42$  MeV for the spontaneous fission of  $^{252}\text{Cf}$ , the neutron absorption for the reference spectrum was calculated to be 62.4%. Although the actual fission neutron spectrum for  $^{252}\text{Cf}$  shows small departures from the Maxwellian shape [10], the departures are sufficiently small to be neglected in the present application.

Neutron absorption within the scintillator is almost entirely neutron capture within the gadolinium loading. The prompt gamma rays emitted in neutron capture (7.9 MeV total energy) cause scintillations within the

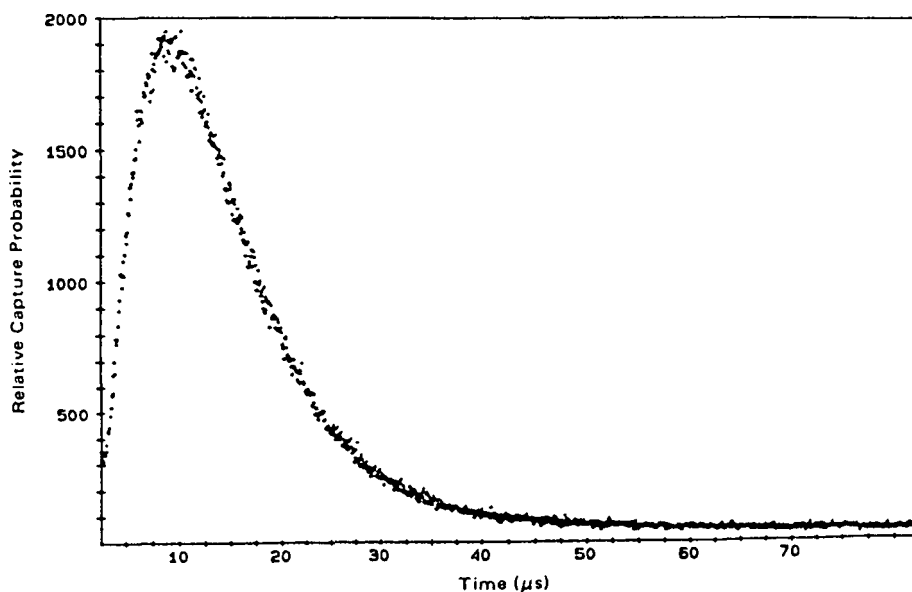


FIG. 2. Neutron lifetime distribution for prototype LSNCC.

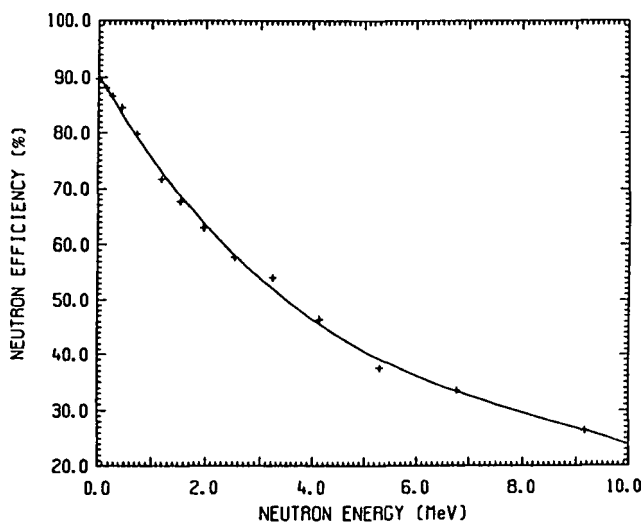


FIG. 3. Calculated neutron efficiency of prototype LSNCC as a function of neutron energy.

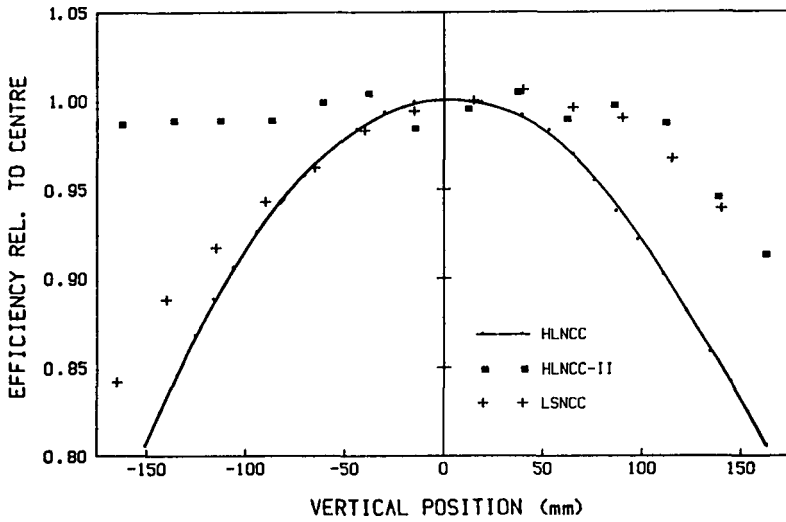


FIG. 4. Comparison of neutron efficiency profile for prototype LSNCC (polyethylene end plugs) with HLNCC and HLNCC-II.

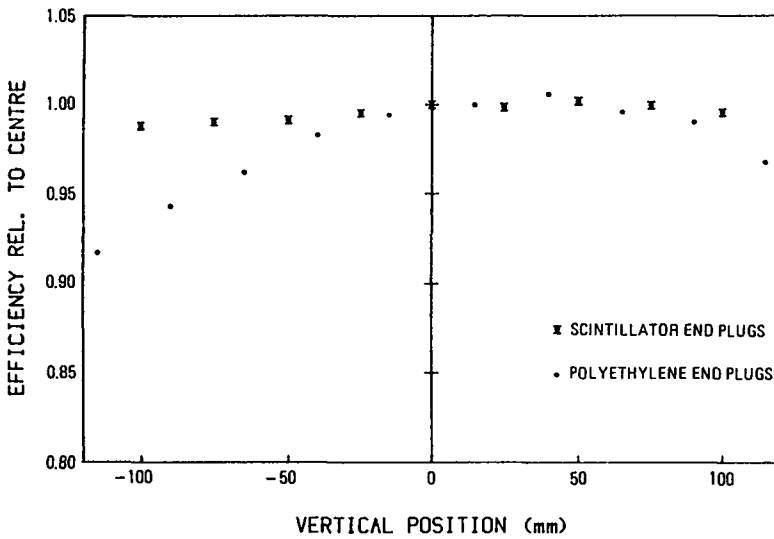


FIG. 5. Neutron efficiency profile for prototype LSNCC: scintillator and polyethylene end plugs.

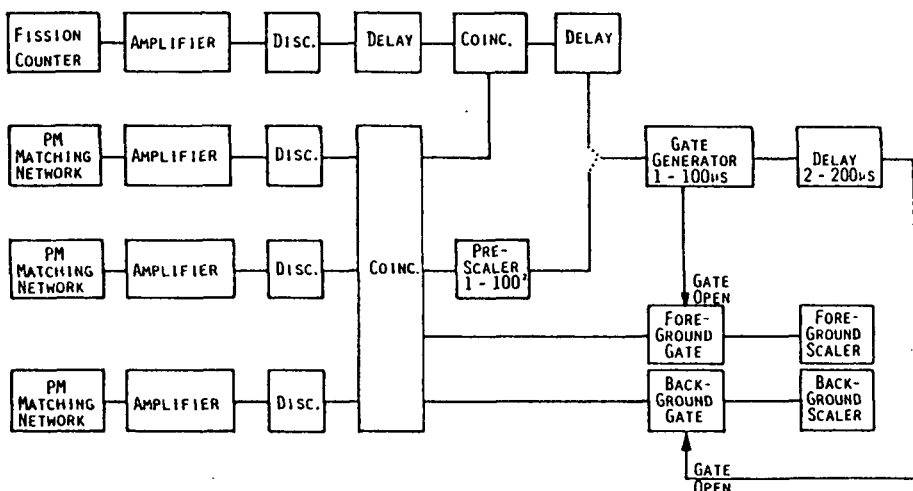


FIG. 6. Block diagram of scintillator electronics.

scintillator, which can be observed with high efficiency by the photomultiplier tubes. In practice, the experimental neutron efficiency that is used for NCC measurements is reduced because of the need to discriminate against the large gamma ray background, which is sample dependent. This efficiency variation is obtained by adjustment of the lower level discriminators on the photomultiplier tubes. A neutron efficiency of 45% has been used for the assay of plutonium samples ranging from 16 to 200 g.

One of the important characteristics of a neutron coincidence counter is the variation of the neutron efficiency within the sample cavity. The original design of the detector included two 10 cm polyethylene end plugs to flatten the axial response of the detector. Figure 4 shows the normalised neutron efficiency of the detector over the sample cavity. For comparative purposes, similar curves for the HLNCC and the HLNCC-II are also shown. As illustrated, the efficiency profile of the LSNCC is marginally better than that of the HLNCC, but is greatly inferior to the profile for the new HLNCC-II.

In order to improve the efficiency profile of the LSNCC, alternative plug designs were considered. It was found that the use of scintillator plugs produced the desired response. The plugs consisted of 17.5 mm diameter stainless steel cans containing the liquid scintillator NE323. The optimum size of the plugs was found to be 15 cm for the base and 13 cm for the top. Figure 5 compares the profiles obtained with scintillator



and polyethylene end plugs. The efficiency obtained with the scintillator plugs is flat to within 1.5% over a distance of 22.5 cm. The flat counting zones for the HLNCC and the HLNCC-II are 11.0 cm and 30.5 cm respectively [11]. Appropriate changes to the design of the scintillator are expected to increase the flat counting zone, and these changes will be implemented in the next prototype. There was no detectable neutron efficiency variation in the horizontal direction.

## 2.2. LSNCC Electronics

The electronics for the LSNCC are shown in Figure 6. The scalers on the output of the liquid scintillator can be gated on for a preset period between 1 and 100  $\mu$ s. The gate trigger may be either a fission signal from a fission chamber located at the centre of the cavity, or a randomly selected (prescale 1 to  $10^4$ ) scintillator signal. A second gate to count background events is initiated a preset delay (1-100  $\mu$ s) after the initial gate. The appropriate scaler (32 maximum for the foreground) corresponding to the number of events counted during the foreground gate is incremented by one at the completion of the counting cycle. Similarly, the appropriate background scaler (maximum of 16) is incremented by one.

In the manner described above, the electronics provide a very simple method of analysing the time correlation associated with a series of events. To calibrate the system, the coincidence between a fission chamber event and a scintillator event is used to preferentially detect the spontaneous fission neutrons. Since the number of neutrons emitted in this event is accurately known ( $\bar{\nu}_p(^{252}\text{Cf})=3.757$ ) [12], the absolute neutron efficiency of the detector may be accurately measured in a short time (typically 10 minutes results in 0.1% accuracy).

It should be noted that the prescaler setting on the random trigger line (1 to  $10^4$ ) is chosen so that the time between consecutive triggers is much greater than the total count cycle (typically 160  $\mu$ s). In this way the trigger is truly random, and the electronics operate in the same manner as the conventional shift register, with the exception that all the information is not used. We are at present designing alternative "shift register" electronics for the LSNCC.

## 3. FISSION RATE CALCULATION

The method of fission rate determination using the LSNCC involves a series of measurements and calculations; this procedure is outlined below.

(i) Initially, a measurement of the detector efficiency for  $^{240}\text{Pu}$  spontaneous fission neutrons is required. This is obtained by use of a  $^{252}\text{Cf}$  fission chamber and correcting the neutron efficiency to account for the difference in the two fission neutron spectra. This difference has been calculated as 2.14% [9], for a  $^{240}\text{Pu}$  spectrum described by a Maxwellian distribution of the form  $\sqrt{E} \exp(-E/T_M)$ , with a temperature  $T_M = 1.27$  MeV. Unfortunately, measurements of the  $^{240}\text{Pu}$  spontaneous fission neutron spectrum are limited to those of Bonner (1961) [13] and Alexandrova et al. (1974) [14]. The two results presented in these references assign temperatures of 1.16 and 1.27 MeV to the Maxwellian distribution respectively. In order to clarify this discrepancy, we are performing another measurement of the  $^{240}\text{Pu}$  spontaneous fission neutron spectrum.

(ii) By referring to standard nuclear data [15], it is possible to calculate the distribution of neutron events correlated with a randomly selected spontaneous fission neutron (i.e. random trigger). Firstly, the neutron distribution associated with spontaneous fission ( $P_v$ ) must be renormalised to the experimental neutron efficiency  $\epsilon_n$ . This new distribution,  $D_i$ , is given by

$$D_i = \epsilon_n^i \sum_{j=0}^{N-i} \frac{(i+j)!}{i! j!} (1-\epsilon_n)^j P_{i+j} \quad (1)$$

where  $i=0,1,2,\dots,N$ .

The next step involves the generation of a distribution that describes the probability of obtaining  $i$  correlated neutrons in a gate of length  $\tau$ . This distribution,  $M_i$ , must be calculated from the neutron capture distribution  $f(t)$  shown in Figure 2. The  $M_i$  are given by

$$M_i = \sum_{j=1}^{j_{\max}} D_{i+j} C(i,j) \int_0^{\infty} f(t) \sigma(t)^i [1 - \sigma(t)]^{j-1} dt \quad (2)$$

where

$$C(i,j) = \frac{(i+j)!}{i! (j-1)!}$$

and

$$\sigma(s) = \int_s^{s+\tau} f(t) dt$$

(iii) Spontaneous fission is accompanied by a multiplicity of prompt fission gamma rays ( $\sim 6.8$  MeV total energy) which, because of the short time-scale involved, are detected as a single event. Since this emission is correlated with the neutrons, a count cycle that is triggered by such a pulse will contribute to the correlated count. The observed distribution of neutron events in this gate would correspond to the fission triggered distribution  $D_i$ .

(iv) For a fission source of rate  $f$ , the distribution of events detected in a randomly triggered gate  $G_i$  of length  $\tau$  is

$$G_i = B_i + \frac{\tau \bar{D}}{\bar{B}} \sum_{j=0}^i [M_i B_{i-j} - B_i] + \frac{\tau f \epsilon_\gamma}{\bar{B}} \sum_{j=0}^i [D_i B_{i-j} - B_i] \quad (3)$$

where  $\bar{B}$  represents the average number of events detected in the background gate (distribution  $B_i$ ), and  $\bar{D}$  is the average of the fission triggered distribution  $D_i$ , given by

$$\bar{D} = \sum_{i=1}^{i_{\max}} i D_i$$

From (3) it is a simple process to obtain a number of estimates of the fission rate from a single measurement. The number of equations that provide realistic solutions is limited by the neutron efficiency and the ratio of the spontaneous fission emission to the total background.

## 4. CORRECTIONS FOR THE LSNCC

### 4.1. The Prompt Gamma Efficiency $\epsilon_\gamma$

For small samples, it is possible to use a  $^{240}\text{Pu}$  spontaneous fission chamber to provide an estimate of the efficiency of the LSNCC for the spontaneous prompt fission gamma rays. Unfortunately, in larger samples there will be a sample dependent attenuation of the gamma rays. In theory, it is possible to determine this efficiency for the particular sample by manipulation of equation (3). Two simultaneous measurements are performed, one counting cycle has a gate length ( $\tau$ ) of  $10 \mu\text{s}$ , while the other has a gate length of  $30 \mu\text{s}$ . Since the fission rate  $f$  may be eliminated from the two simultaneous equations, the variable  $\epsilon_\gamma$  may be determined. The statistical significance of this determination is highly dependent on the detector and sample characteristics, and the general applicability of the method is under investigation.

## 4.2. Dead-Time Corrections

Because of the gamma ray sensitivity of the LSNCC, high background count rates are obtained, and hence dead-time effects become very significant for large samples. In general, because NCC techniques involve a time analysis of events, the dead-time corrections are very difficult to calculate analytically. For the present, a general procedure for correcting for dead-time has been proposed. Before the necessary analytical calculations are performed, a thorough investigation of dead-time is being undertaken by use of a Monte Carlo computer program. By simulating the detection of events in the LSNCC, as well as the counting system, the program provides a complete quantitative description of dead-time effects. This analysis is in progress.

## 4.3. Neutron Multiplication Studies

A study of neutron multiplication in the LSNCC has extended the theoretical developments of the procedure outlined by Bohnel [16]. Unfortunately this procedure, as applied to the LSNCC, involves the solution of a series of intractable algebraic expressions. It is intended to pursue this line of investigation using an algebraic manipulation program called REDUCE.

## 5. PLUTONIUM ASSAY USING THE LSNCC

Fission rate measurements have been performed using a number of  $\text{PuO}_2$  samples. The maximum amount of plutonium measured was 200g, and the composition of the samples is summarised in Table I. Table II

TABLE I. ISOTOPIC COMPOSITION  
OF  $\text{PuO}_2$  SAMPLES

Isotope	Isotopic composition
$^{239}\text{Pu}$	91.6 wt%
$^{240}\text{Pu}$	7.76 wt%
$^{241}\text{Pu}$	0.63 wt%
$^{242}\text{Pu}$	< 400 ppm

TABLE II. RESULTS OF FISSION RATE MEASUREMENTS OF PuO<sub>2</sub> SAMPLES

PuO <sub>2</sub> sample	Mass of Pu (g)	Fission rate estimates					Average estimate	Calculated fission rate	Ratio
		f <sub>0</sub>	f <sub>1</sub>	f <sub>2</sub>	f <sub>3</sub>	f <sub>4</sub>	$\bar{f}$	f <sub>c</sub>	$\bar{f}/f_c$
1	14.08	567	577	550	551	587	564	539.8	1.044
2	24.05	962	981	924	956	1033	960	922.0	1.042
2+1	38.13	1557	1588	1528	1480	1673	1552	1462	1.062
3	62.56	2452	2508	2378	2388	2634	2458	2398	1.025
3+1	76.64	2950	2983	2870	3007	3032	2962	2938	1.008
3+2	86.61	3364	3327	3326	3357	3693	3408	3320	1.027
4	98.88	4186	514	4116	3753	4029	4054	3836	1.057
3+2+1	100.7	3868	3928	3739	3894	3970	3880	3860	1.005
4+1	113.0	4443	9141	4322	4603	4540	4504	4332	1.040
4+2 <sup>a</sup>	123.4	5036	8157	4835	4692	4985	4928	4730	1.042
4+2+1 <sup>a</sup>	137.0	5408	9153	5457	5391	5629	5450	5253	1.038
4+3 <sup>a</sup>	161.4	6321	6210	6212	6555	6046	6324	5189	1.022
4+3+2+1 <sup>a</sup>	200.0	7869	7364	7966	7550	7808	7810	7668	1.019

<sup>a</sup> Indicates that the measurement was performed at an efficiency for <sup>252</sup>Cf spontaneous fission neutrons of 0.463. The efficiency for all other measurements was 0.490.

shows fission rate results obtained from the first four equations in (3), as well as an average estimate (f) which is based on the average number of events in the foreground and background gates. It should be noted that there are a few anomalous fission rate estimates shown in Table II. These anomalies are due to the fact that the foreground and background probability distributions almost cross over at this point, resulting in the subtraction of almost identical probabilities. The gate length for the measurements was 30  $\mu$ s and the neutron and prompt gamma efficiencies of the detector were 49% and 72% respectively. Also shown in Table II are fission rates calculated from the stated isotopic composition and the spontaneous fission half-life of  $^{240}\text{Pu}$ . The value of  $T_{1/2}$  was taken as  $1.15 \times 10^{11}$  years [17].

The accuracy of the fission rate measurement is not only dependent on counting statistics, but on the accuracies of the prompt gamma and neutron efficiency determinations, as well as the dead-time of the detection system. In general, the results shown in Table II indicate an overestimate of the fission rate of approximately 3%. Since the accuracy of the fission rate measurement is of the order of 1.5%, this overestimate suggests a small systematic error. However, the calculated fission rates ( $f_c$ ) shown in Table II are based on an assumed value for  $T_{1/2}(^{240}\text{Pu})$ , which has a quoted accuracy of 3%. There is therefore a need for a considerable improvement in the precision of the spontaneous fission half-life. We are at present performing a new measurement of  $T_{1/2}$  using the neutron coincidence counting method. In order to perform the experiment, we have purchased a high purity  $^{240}\text{Pu}$  spontaneous fission source ( $\sim 8$  mg Pu) from ORNL.

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# DETERMINATION OF PLUTONIUM ABUNDANCES, CONCENTRATIONS AND ISOTOPIC RATIOS BY X-RAY AND GAMMA RAY SPECTROMETRY ASSAY TECHNIQUES\*

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## Abstract

DETERMINATION OF PLUTONIUM ABUNDANCES, CONCENTRATIONS AND ISOTOPIC RATIOS BY X-RAY AND GAMMA RAY SPECTROMETRY ASSAY TECHNIQUES.

Two X-ray and gamma ray systems were recently installed at-line in glove boxes and will measure Pu solution concentrations from 5 to 105 g/L. These NDA systems, developed and refined over the past decade, are now used domestically and internationally for nuclear material process monitoring and accountability needs. In off- and at-line installations, they can measure solution concentrations to an accuracy of 0.2%. The K X-ray fluorescence analysis systems use a transmission source to correct for solution density. The gamma ray systems use peaks from 59 to 208 keV to determine solution concentrations and isotopic ratios. A Pu check source monitors system stability. These two NDA techniques can be combined to form a new NDA measurement methodology. With the instrument located outside a glove box, both Pu isotopic ratios and absolute Pu abundances of a sample located inside a glove box can be measured. The new technique works with either single- or dual-source excitation, the former for a detector 6–20 cm away with no geometric corrections needed, the latter with geometric corrections or source movement if the sample cannot be measured at the calibration distance.

## 1. INTRODUCTION

The Safeguards Technology Program (STP) at the Lawrence Livermore National Laboratory (LLNL) delivered its first off-line plutonium solution assay system to the Savannah River Plant (SRP) in 1974, the first at-line Pu solution isotopic ratio analysis system in 1979 to the Tokai-mura plant in Japan [1] and the first off-line K X-ray fluorescence analysis (K-XRFA) system to the SRP in 1981 [2]. This paper describes two state of the art gamma and X-ray systems that were recently installed for accountability needs at the SRP for at-line analyses of Pu solution concentrations and Pu isotopic ratios.

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\* Work supported by the United States Department of Energy Office of Safeguards and Security and by E.I. Du Pont de Nemours and Company, Inc., and performed under the auspices of the US DOE by the Lawrence Livermore National Laboratory under contract W-7405-Eng-48.

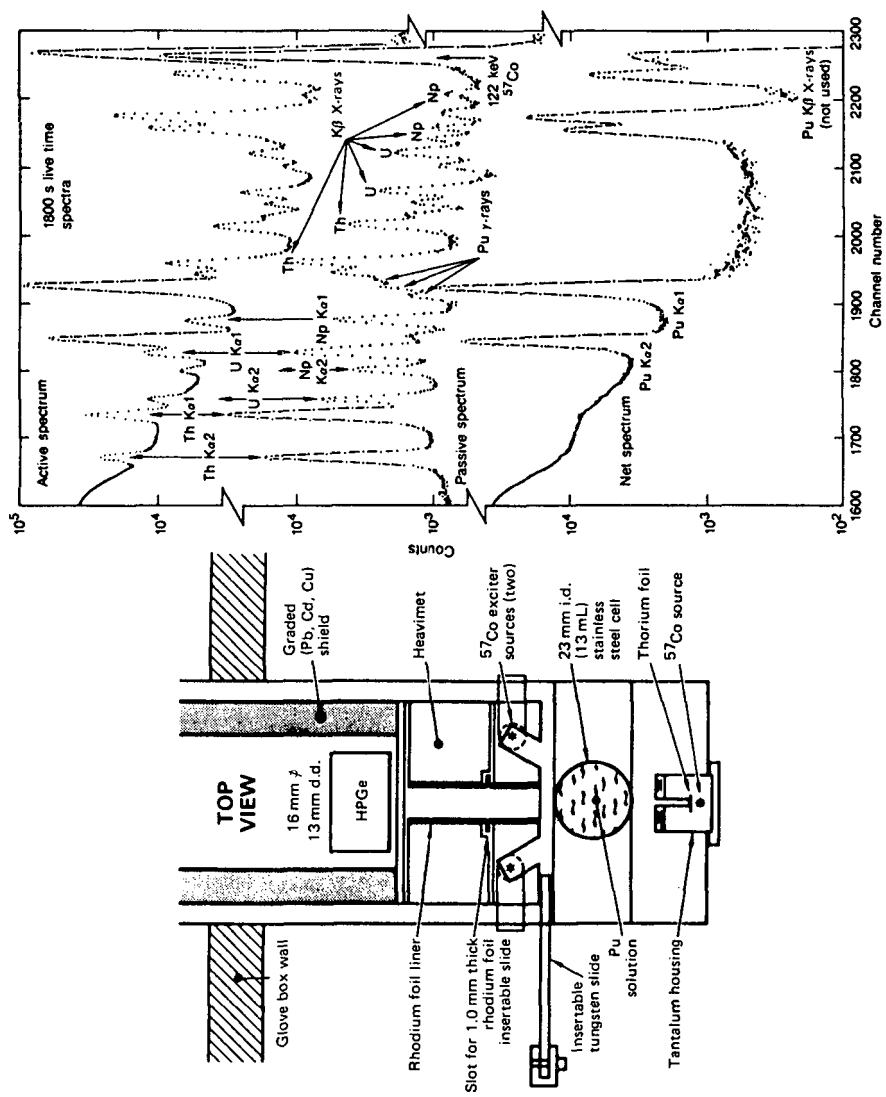


FIG. 1. X-ray fluorescence analysis system and three spectra of a 50 g Pu/L solution obtained from its use (d.d.: depletion depth).

In addition, we report on a new NDA concept that combines the two techniques to form a field portable NDA methodology that measures Pu abundance and isotopic ratios and solution concentration if the volume is known, from outside a glove box in a way that is nearly independent of geometry. The principle and restrictions of this new methodology are discussed.

## 2. X-RAY FLUORESCENCE SOLUTION CONCENTRATION MEASUREMENTS

The left side of Fig. 1 shows a top view of our X-ray fluorescence analysis system. The detector is shielded from glove box activity and from two 0.74 GBq (20 mCi)  $^{57}\text{Co}$  exciter sources. In the active mode the 122 keV gamma rays cause Pu X-ray fluorescence and produce a spectrum as shown on the right side of Fig. 1. In the passive mode, the tungsten slide closes and eclipses the exciting radiation so that solution radioactivity can be measured. A slide with a 1 mm thick rhodium foil is inserted automatically if the solution is sensed to contain excessive amounts of the 59.5 keV americium gamma ray. Both the rhodium and tungsten slides are operated under computer control. A third  $^{57}\text{Co}$  source housed in tantalum and located behind 0.4 mm thick thorium foil provides a highly collimated beam of 122 keV photons and thorium X-rays that go through the solution. Their intensities from the passive spectrum (Fig. 1, middle) are used to correct the measured actinide  $\text{K}\alpha_1$  X-ray intensity for solution matrix effects and for self-absorption. The (active minus passive) data yield a net X-ray spectrum (Fig. 1, bottom). Solution concentrations are proportional to the measured  $\text{K}\alpha_1$  X-ray intensity, which can be converted to grams of plutonium per litre after system calibration. Results of the K-XRFA technique are discussed in the next section.

## 3. GAMMA RAY SOLUTION AND ISOTOPIC COMPOSITION MEASUREMENT SYSTEM

Plutonium solution concentrations and isotopic ratios can also be measured using gamma ray spectrometry. Figure 2 shows a side view of the system. Solution is drawn into a 3 cm diameter by 1 cm thick stainless steel cell that is part of an assembly installed at-line in the glove box. Large/small collimators in a Ta slide are used for low/high concentrations to collimate gamma ray activity. There is also a Pu source vacuum deposited on stainless steel and welded into the slide. It is used to monitor system performance periodically. All of the spectra accumulated are referenced to the intensity of the 88 keV gamma ray from a  $^{109}\text{Cd}$  source attached to the cell

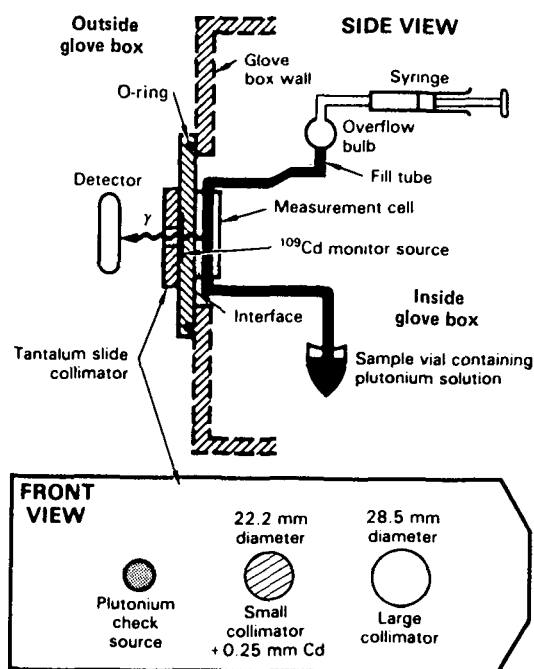


FIG. 2. Sketch of gamma ray spectrometry system.

TABLE I. PRECISION OF Pu ISOTOPIC ABUNDANCE MEASUREMENTS

Isotope	Abundance (%)	Precision (%)	Code error (%)
Pu-238	0.0271	1.0	1.4
Pu-239	93.36	0.03	0.04
Pu-240	6.05	0.47	0.55
Pu-241	0.522	0.20	0.21
Am-241	0.054	0.34	0.30

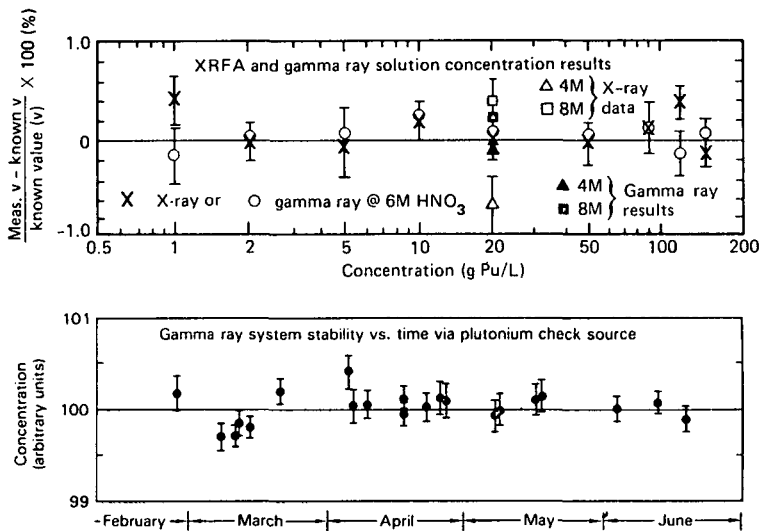


FIG. 3. Comparison of atomic K-XRFA and nuclear gamma ray spectrometry techniques for measurement of Pu solution concentrations.

window. This intensity is used to monitor system geometry, and to correct for actual counting time and for any detector efficiency changes.

Both the solution concentration and Pu isotopic ratios are determined from the intensities of peaks in the rather complex 94–104 keV region and from the 59, 129, 148, 152 and 208 keV peaks using software codes developed by the STP [3]. The system is calibrated using one or more known concentrations. Solution density is determined by a densitometer installed in the same glove box so that small self-absorption corrections can be made.

The gamma ray spectrometry technique also has the ability to measure the Pu isotopic composition. Table I lists the precisions of isotopic abundance measurements obtained using the large collimator for 20 runs of 20 g Pu/L solution, with each run accumulated for 66 min.

The results of measuring solution concentrations from 1 to 150 g Pu/L using both the atomic X-ray fluorescence and nuclear gamma ray methods are shown in the top half of Fig. 3.

Both these NDA methods are accurate to 0.2% over the central, 5-105 g/L range, and this represents the state of the art for their use at- or off-line for nuclear material accountability measurements. The range of the atomic and nuclear techniques can be extended by a factor of three to five to lower or higher concentrations; however, careful calibration is required beyond either end to ensure 0.2% accuracy to such limits. The bottom half of Fig. 3 shows that the gamma ray system remained stable to within 0.1% over five months as monitored by the plutonium check source.

#### 4. COMBINED X-RAY AND GAMMA RAY NDA SYSTEM

##### 4.1. Introduction

We have developed a new measurement concept. It combines the advantages of both the K-XRFA and gamma ray methods, and may offer international inspectors a portable measurement system that can measure Pu isotopic ratios and abundances from outside a glove box in a manner that is nearly independent of the sample-detector geometry. The arrangement is shown in Fig. 4. An HPGe detector is located in front of a glove box that contains a Pu solution sample cell, which preferably is located at or near the inside edge of the glove box and at a known distance. First, we consider only one X-ray fluorescence

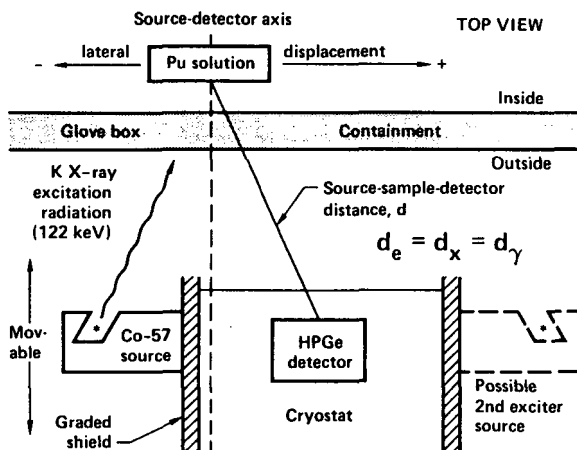


FIG. 4. Top-view geometry for single-source active/passive X-ray + gamma ray methodology. See text for details if two sources are used.

exciter source. In this case the sample must be positioned midway between the axes of the detector and source. Then, the excitation distance  $d_e$  will equal the X- or gamma ray distances. X-ray production in the sample and gamma ray intensities from the sample vary as the inverse square of the distance, while X-ray intensity from the sample varies as the inverse fourth power of the distance.

As shown in Eq. (1), gamma ray counts will be proportional to the sample activity, detector efficiency, gamma ray branching ratio and counting time divided by the distance squared and a self-absorption correction term  $a_\gamma$ :

$$I_\gamma \propto A_\gamma \epsilon_\gamma b_\gamma t (d_\gamma^2 a_\gamma)^{-1} \quad (1)$$

Therefore, the Pu abundance in the sample will be related to the sample activity by a calibration constant, as shown in Eq. (2), which represents the passive gamma ray spectrum from the sample:

$$\text{Pu abundance} = k_\gamma A_\gamma = k_\gamma a_\gamma d_\gamma^2 I_\gamma (\epsilon_\gamma b_\gamma t)^{-1} \quad (2)$$

We can write a similar equation that relates the Pu abundance to the X-ray fluorescence analysis intensity:

$$\text{Pu abundance} = k_x A_x = k_x d_e^2 d_x^2 a_e a_x I_x (\epsilon_x f t)^{-1} \quad (3)$$

where  $a_e$  and  $a_x$  are the absorption correction factors for the excitation radiation entering the sample and for the X-radiation leaving the sample, and  $f$  is the K X-ray fluorescence yield.

Using Eqs (2) and (3), we can express Pu abundance as the square of Eq. (2) divided by Eq. (3). This yields

$$\text{Pu abundance} = K (\text{geometry term}) (\text{absorption term}) I_\gamma^2 / I_x t \quad (4)$$

where  $K$  is a single calibration constant that includes the detector efficiency, gamma ray branching and X-ray fluorescence yield; the geometry term is

$$d_\gamma^2 d_\gamma^2 / d_e^2 d_x^2$$

and the absorption term is

$$a_\gamma^2 / a_e a_x$$

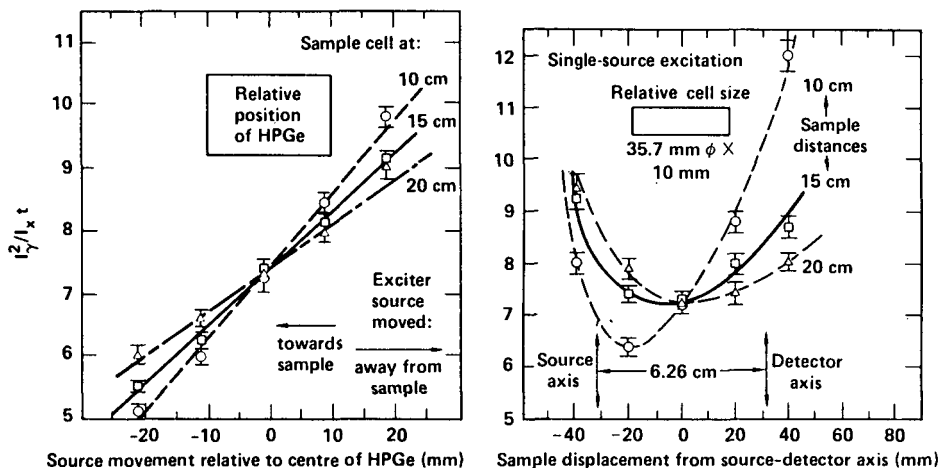


FIG. 5. Variations in intensity ratio (proportional to abundance) with (left) axial movements of the source towards or away from the sample, and with (right) lateral displacements of the sample off the midline axis towards either the detector or source axis.

Note that for a single-source excitation,  $d_e = d_x = d_y$ ; thus, the geometry term becomes unity. The last term in Eq. (4) must be experimentally measured: that is,  $I_{\gamma}^2$  is determined from the passive gamma ray spectrum and is divided by the X-ray fluorescence intensity, obtained from the net (active minus passive) spectrum, times the active count time.

We have performed several proof of principle experiments using uranium solutions spiked with  $^{153}\text{Gd}$ . Uranyl nitrate solutions are easier to handle (no glove box), and have very nearly the same density and fluorescence efficiency as plutonium nitrate; and the emitted  $^{153}\text{Gd}$  97.4 and 103.2 keV gamma rays will simulate the gamma and X-rays of Pu samples in this energy region.

A concentrated solution of uranyl nitrate was made to which a 1% by volume solution of  $^{153}\text{Gd}$  activity was added. From the master solution, various dilute concentrations down to 5 g U/L were gravimetrically prepared. The 30 d difference in half-life between  $^{153}\text{Gd}$  and  $^{57}\text{Co}$  leads to a differential correction of only 1% after 3 d, hence experiments carried out within one week required no half-life corrections. To simulate field conditions, a portable HPGe planar detector, 25.4 mm in diameter and 20 mm deep, was used with a 5 d Dewar; the detector had a FWHM of 550 eV at 122 keV with 3  $\mu\text{s}$  Gaussian shaping; and a computer based, 4096-channel analyser was used.



A code, GRPANL [4], was used to fit the X- and gamma ray peaks. In all measurements, the ratios of  $K\alpha_2$  to  $K\alpha_1$  and of the weaker to the stronger gamma ray were monitored. After satisfactory fits were obtained, the weaker X-ray (gamma ray) peak was multiplied by 1.61 (1.38) to normalize the weaker peak intensity to the stronger. Then, the mean X- and gamma ray intensities were calculated and became the  $I_Y$  and  $I_X$  used to obtain the intensity ratio required by Eq. (4).

#### 4.2. Single-source excitation

When only one source is used, its location must be optimized relative to the detector. The left side of Fig. 5 shows the intensity ratio term measured at three distances versus the distance that the source has been moved towards or away from the sample, which remained on the midline axis. The intensity ratio is independent of sample distance when the source is located next to the mass centre of the detector.

Next, the intensity ratio was measured for lateral displacements of the sample off a midline axis that is half-way between the source and detector (right side of Fig. 5). As the sample is moved towards the detector axis, X-ray production and intensity decrease, thus increasing the intensity ratio at all distances. However, when the sample is moved towards the source axis, X-ray production increases (dramatically at close sample distances), and causes the broad minima in the ratios at intermediate distances and deeper minima at closer distances.

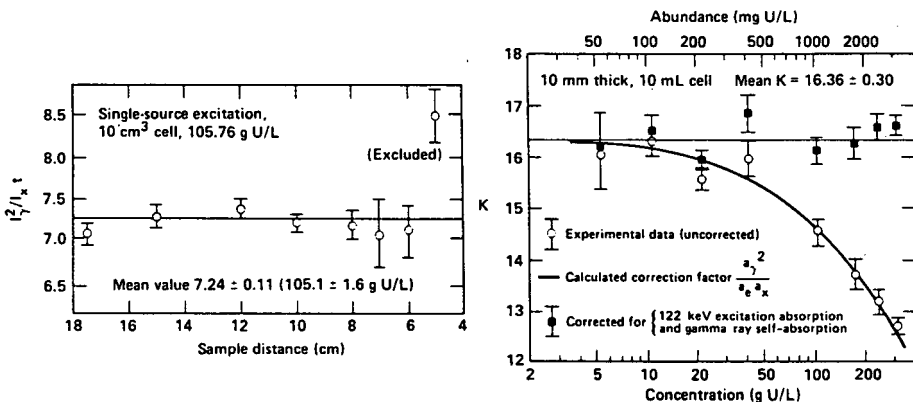


FIG. 6. Left: the intensity ratio stays constant if the sample remains on the midline axis. Right: measurements of different concentrated solutions uncorrected/corrected (circles/squares) for 122 keV source absorption and X- and gamma ray sample self-absorption.

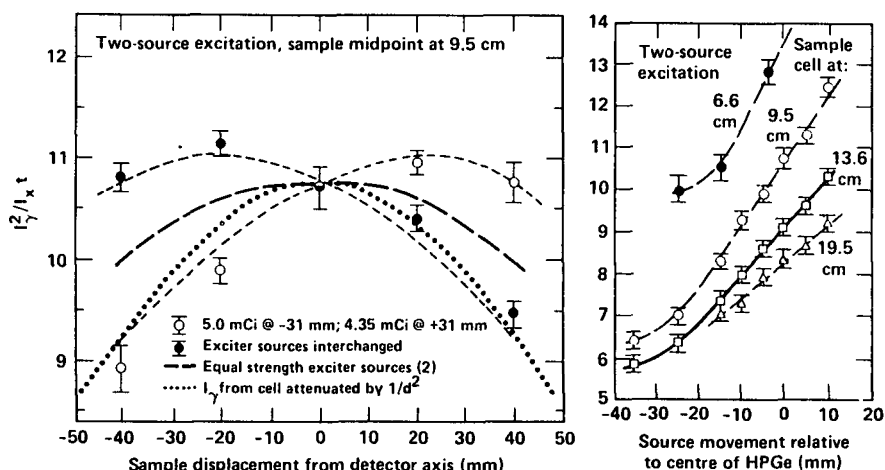


FIG. 7. Variation in measured intensity ratio with (left) lateral displacement of sample off-axis and with (right) source movement from detector centre with two sources.

For distances between 15 and 20 cm, an off-axis sample misalignment of 1 cm or less will lead to an error of 5% or less in the intensity ratio (abundance). The 6.26 cm between the detector and source axes used in our experiments can be reduced to 3.8 cm by using a smaller diameter cryostat and less source shielding. This would flatten out each parabolic curve in Fig. 5 and lessen the need for exact on-axis sample alignment.

If the sample and midline source detector axes are collinear and the source is located on the same line as the mass centre of the detector, then Eq. (4) states that the actinide abundance or solution concentration should remain constant for any sample distance. The data on the left side of Fig. 6 show that a constant intensity ratio with an error of 1.5% is obtained and indeed is independent of the distance. Eventually, at some close distance (in this case 5 cm), the source radiation exciting the sample begins to be partially eclipsed by the shielded detector assembly (Fig. 4) and the intensity ratio increases.

A series of constant volume (10.0 mL) spiked solutions were measured. The concentrations ranged from 5 to 310 g U/L and are shown on the right side of Fig. 6. The data represented by circles are uncorrected for absorption effects of source radiation entering the sample or for gamma and X-rays leaving the sample. But the correction amounts are calculated

easily from geometric and mass absorption considerations (solid line). The data represented by squares are obtained when calculated corrections are applied to the uncorrected data. The mean value shown with its 2% uncertainty is the K of Eq. (4) and is used to convert a measured and corrected ratio to solution concentration.

#### 4.3. Double-source excitation

If two sources are used (Fig. 4), the sample and detector axes become collinear. However, the excitation distance  $d_e$  no longer equals the gamma ray and X-ray distances, hence the geometry term no longer equals unity. Any sample lateral displacements off-axis do not produce the dramatic variations seen with only one source. The left side of Fig. 7 shows the variation in measured intensity recorded with two sources of unequal strength for a sample at 9.5 cm. Interchanging the sources reverses the asymmetry. For equally intense sources, the measured intensity ratio data remain nearly constant, i.e. they stay within 5% of the on-axis ratio at distances up to 3 cm. At greater distances the heavy dashed curve becomes even flatter, and thus less sensitive to off-axis sample misalignments.

The right side of Fig. 7 shows the changes in the measured intensity ratio at four sample distances when the two sources are moved relative to the detector centre. The curves

TABLE II. CALCULATED AND MEASURED  $I_\gamma^2/I_x$  VERSUS SAMPLE DISTANCE

Sample distance $d_x$ (mm)	Measured $I_\gamma^2/I_x t$ , $v^*$	Geometry factor $g^{**}$	Calculated $I_\gamma^2/I_x t$ , $gv$	Measured $I_\gamma^2/I_x t$ at a source displacement of:
95	10.7	0.848	9.07	$9.22 \pm 0.19$ @ -11 mm
115	~9.8	~0.922	~9.04	$9.14 \pm 0.22$ @ -6 mm
135	9.1	1.000	9.10	$9.27 \pm 0.26$ @ 0 mm
165	~8.7	~1.044	~9.08	$8.86 \pm 0.18$ @ +4 mm
195	8.3	1.095	9.09	$9.06 \pm 0.21$ @ +10 mm
Mean values:			~9.08	$9.11 \pm 0.16$

\* Read/interpolated from data in Fig. 7 for a source movement of zero.

\*\* Geometric correction  $g = (d_\gamma^2/d_e^2)(d_\gamma^2/d_e^2(135))$ ;  $d_e^2 = [d_\gamma^2 + (62.6)^2]$ .

cannot cross as they did when a single source was used (Fig. 5). Only if both sources were placed in front of the detector would exciter distance equal the gamma or X-ray distance, and then only for that distance. Thus, there is no source position that makes the measurement insensitive to sample distance. So, we have two choices: either to leave both sources fixed at the detector's mass centre line and apply simple calculated geometric corrections, or to move the two sources (towards or away from the sample) by an amount precalibrated for sample distance (like focusing of a camera).

To demonstrate both options, a set of measurements was made for five sample distances from 9.5 to 19.5 cm. Table II summarizes these data. Columns 4 and 5 agree, as they should. The error on the mean value is 2%.

## 5. SUMMARY

The NDA techniques of atomic X-ray fluorescence analysis and nuclear gamma ray spectrometry are both well developed. If a suitable transmission source is used with the former and solution densities are supplied for the latter, then both can be used to make accountability measurements. We have demonstrated that for at-line installations accuracies of 0.2% can be obtained for concentration ranges from below 1 to above 150 g/L, if they are operated under well designed quality control programmes. Similar accuracies could be obtained in off-line systems. Gamma ray spectrometry also measures the relative Pu isotopic abundances. Since the two NDA techniques are fundamentally different, they can serve as basically independent measurement methods; and if installed side by side, each can authenticate the other and reduce tampering probability.

We have developed a potentially new NDA methodology by combining X-ray and gamma ray spectrometry. Our experiments show that it may be possible to make in-field measurements for Pu isotopic ratios and concentrations of known solution volumes using only simple and portable measurement equipment. Although this methodology is not designed to offer accountability accuracies, it can make confirmatory measurements, since its accuracy will be from 2 to 5%. If one exciter source is used, then a measurement can be made almost independent of distance, but the sample axis must be aligned to 1 cm or better with the midline axis between the source and detector. If two sources are used, any unknown must be measured at the same distance as the calibration standard unless a geometric correction is applied to the data or the sources are moved by a precalibrated amount relative to the detector. Additional experiments are

planned with Pu to verify that this new NDA methodology can measure Pu abundances from outside a glove box almost independently of geometry. This new method may benefit the international inspection community.

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## FIELD EXPERIENCE WITH A PORTABLE K-EDGE DENSITOMETER

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### Abstract

#### FIELD EXPERIENCE WITH A PORTABLE K-EDGE DENSITOMETER.

The portable K-edge densitometer developed by the Los Alamos National Laboratory was used by Euratom in a number of in-field applications. The objective of these field trials was to check the instrument's performance, its ease of use and its compatibility with plant operational procedures. The instrument was used by inspectors to measure Pu product solutions of reprocessing plants with Pu concentrations ranging from about 10 to about 370 g/L. In total about 100 samples were measured. On average the differences between measured and declared values were of the order of 1-2% for concentrations larger than 100 g/L. In separate experiments the stability and reproducibility of the instrument were checked. Another aspect looked at was the effect of changing hardware components and sources (different portable multichannel analyser, different Ge detector, use of new as opposed to old sources) on the value of the calibration constant and on the performance of the instrument. An overall evaluation of the measurement campaigns indicates that the calibration constant is increasing with time. This effect should be further examined. Some other practical recommendations and feedback obtained from operators are given.

### 1. INTRODUCTION

The portable K-edge densitometer developed by the Los Alamos National Laboratory [1] was demonstrated to a team of Euratom-Luxembourg, Euratom-Ispira and IAEA staff at the European Institute for Transuranium Elements (hereinafter referred to as the TUI) at Karlsruhe in June 1985. Subsequently, the instrument was used several times during field tests to measure plutonium nitrate end product solutions from reprocessing plants. In addition, some specific laboratory tests were carried out.

The portable K-edge densitometer is of considerable interest to both the safeguards authorities and the operators because it permits the inspectors to carry out in situ measurements on plutonium nitrate solutions with the following advantages:

- Measurement results are made available without the usual long delays due to transport and chemical analysis.
- There is a reduction in the number of samples to be packed and shipped, with a corresponding saving in operators' efforts and assistance and cost reduction for safeguards authorities.
- Requirements are reduced for long term storage of reference samples affected by radiolysis and self-concentration.

A disadvantage at present is that the portable K-edge is less accurate than the existing X-ray based K-edge instruments or chemical analysis methods.

## 2. DESCRIPTION AND ASSEMBLY OF EQUIPMENT

A detailed description of the instrument was presented at the ESARDA Symposium in Venice [1]. Both the Davidson multichannel analyser (MCA) and the Ge detector (model Ortec GLP 16195/10 S, planar, 10 mm  $\times$  200 mm<sup>2</sup> with a resolution of 505 eV at 122 keV) provided for the demonstration failed and were replaced with another Davidson MCA and another Ge detector (model Ortec GLP/F-43, planar, 13 mm  $\times$  800 mm<sup>2</sup> with a resolution of 650 eV at 122 keV).

The three sources, <sup>75</sup>Se (activity 1.924 GBq (52 mCi)), <sup>57</sup>Co (0.962 GBq (26 mCi)) and <sup>109</sup>Cd (3.7 MBq (100  $\mu$ Ci)) were delivered directly from Los Alamos. The Se and Co sources were mounted into the source holder. During the mounting it was noticed that the diameter of the sources was about 0.2 mm larger than the diameter of the hole in the source holder. The workshop of the TUI enlarged the hole so that the sources could be fitted in position.

The following dose rates were measured for the sources:

(a) Direct contact

<sup>109</sup> Cd:	0.05 mSv/h	(5 mrem/h)
<sup>75</sup> Se:	20 mSv/h	(2000 mrem/h)
<sup>57</sup> Co:	4 mSv/h	(400 mrem/h)

(b) at 30 cm

<sup>75</sup> Se:	1 mSv/h	(100 mrem/h)
<sup>57</sup> Co:	0.2 mSv/h	(20 mrem/h)

The mounting of the sources, including health physics checks, took about 30 min.

The unpacking and assembly of the whole equipment, including the transfer of the sample vials and the sample holder in the glove box, took about 2 h. To this time the time required for cooling the Ge detector had to be added before measurements could be started.

In a later field trial, the Davidson MCA and Ge detector were replaced with the original equipment delivered to the TUI to check for effects due to a change of components of the instruments which may require replacement. Such changes may be required under field conditions if one component fails or does not work with the performance expected.



TABLE I. CHARACTERIZATION OF REFERENCE SAMPLES USED AT THE TUI

Sample No.	Concentration (g/L)
S1 Pu nitrate	$200.36 \pm 0.34$ (Pu)
S2 Pu nitrate	$151.05 \pm 0.35$ (Pu)
S3 Pu nitrate	$100.25 \pm 0.28$ (Pu)
S4 Pu nitrate	$50.50 \pm 0.15$ (Pu)
S5 Pu nitrate	$30.32 \pm 0.23$ (Pu)
S6 U nitrate	$220.64 \pm 0.36$ (U)
S11 (Pu + U) nitrate	$180.40 \pm 0.43$ (Pu) $10.47 \pm 0.10$ (U)
S12 (Pu + U) nitrate	$180.18 \pm 0.28$ (Pu) $5.52 \pm 0.44$ (U)
S13 (Pu + U) nitrate	$180.34 \pm 0.44$ (Pu) $2.38 \pm 0.56$ (U)
S31 Pu nitrate	$100.42 \pm 0.22$ (Pu)

### 3. MEASUREMENT RESULTS

#### 3.1. Demonstration at the TUI

##### 3.1.1. Characterization of reference samples

Ten reference samples were provided and characterized using the X-ray densitometer installed at the TUI [2]; six samples were pure plutonium nitrate samples, three samples had uranium additions varying from about 2 to 10 g/L and one sample was a pure uranium nitrate solution. The sample data are summarized in Table I.

##### 3.1.2. Calibration constant $K$

The calibration was done using the procedure recommended in the draft operation manual provided by Los Alamos. Sample S2 (151 g Pu/L) was used as calibration standard throughout. Three calibration runs were made during the demonstration:

- (a)  $4 \times 300$  s measurement time, straight-through measurement with empty vial, calibration run with sample S2;  
result:  $K = 0.00647 \pm 6 \times 10^{-5}$  (i.e.  $\pm 0.9\%$ );

- (b)  $4 \times 3000$  s measurement time, empty vial, sample S2;  
result:  $K = 0.00637 \pm 4.5 \times 10^{-5}$  (i.e.  $\pm 0.7\%$ );
- (c)  $4 \times 300$  s measurement time, vial with 3M  $\text{HNO}_3$ , sample S2;  
result:  $K = 0.00644 \pm 7 \times 10^{-5}$  (i.e.  $\pm 1.1\%$ ).

The standard deviations given above are the counting statistics errors for the relevant measurements and include the standard deviation in the concentration value of the reference sample taken, i.e. 0.2% in the above case. The results show that the standard deviation for the calibration constant derived from a  $4 \times 300$  s measurement is typically of the order of 1%.

### 3.1.3. Measurement results

The measurement results obtained at the TUI are presented in Table II. They were obtained with the calibration constant  $K = 0.00644$  (based on calibration with an acid sample as under (c) above). The table also includes the repeat measurements carried out on some samples. The measurement time used for all the samples was  $2 \times 300$  s.

The results indicate that the reproducibility of the data is consistent to within the counting statistics error given in column 6 of Table II. A check on the variation in the transmission ratio  $r = I_M^{121} I_o^{122} / I_M^{122} I_o^{121}$  for sample S2 over the three measurement days gives a value of  $\pm 0.9\%$ , consistent with the counting statistics.

Furthermore, the difference between the declared and measured values ( $\Delta\rho$ ) is more or less consistent with the counting statistics error (columns 5 and 6 of Table II). There appears to be, however, a bias in that for low concentrations the measured values are always systematically lower than the declared values.

The results show further that, in agreement with calculations, small uranium concentrations do not affect the results to within the error of the measurement (the correction is estimated to be 0.29 g/L for sample S11 with about 10 g U/L) [3]. From the difference between samples S0 and S6 in Table II, a correction of about 0.02 per g U/L can be derived.

### 3.2. Further field test results

In a reprocessing plant the instrument was used to measure the concentration of seven Pu output samples. The calibration was carried out with a sample having a high Pu concentration ( $\geq 300$  g/L, sample 1 in Table III) and a sample with 3M  $\text{HNO}_3$  for the background measurement. The calibration constant was determined to be  $K = 0.00665$ .

The measurement results are presented in Table III, where the ratio of the measured to the declared value is given for comparison. The lowest concentrations measured were of the order of 10–30 g Pu/L.

All measurement results were obtained using the standard  $2 \times 300$  s runs per sample. As can be seen from Table III, the difference between declared and measured values is consistent with the expected standard deviation of the measurement. The uncertainties in the declared values were not known.

It was observed that the gas bubbles released in high concentration Pu samples affected the assay results. When the samples were swivelled before each measurement to degas, the measurement results were consistent.

A series of other measurements showed that the best consistency was obtained if the straight-through measurement was taken with an acid sample having the same molarity as the sample to be assayed.

During another field application in the same installation, three Pu output samples were measured. The calibration constant derived from four repeat measurements on one sample gave  $K = 0.00676 (\pm 0.9\%)$ .

The ratio of the measured to the declared value for a sample having a very high Pu concentration was 0.999. The standard deviation for the measured value was 0.4% compared with an expected standard deviation of 0.7% (six repeat measurements of  $2 \times 300$  s each). The same ratio for a sample having a very low Pu concentration was 0.937. The counting statistics error for the measured value was 18%.

### 3.3. Some more specific results

#### 3.3.1. Stability of instrument

A series of measurements were carried out during various applications to check the stability of the K-edge densitometer. The following parameters were examined, both for an acid sample and for a Pu sample with about 150 g Pu/L:

- Net cadmium count rate  $P_o(\text{Cd})$  for acid sample and  $P_M(\text{Cd})$  for plutonium sample,
- Net cobalt count rate  $P_o(\text{Co})$  for acid sample and  $P_M(\text{Co})$  for plutonium sample,
- Net selenium count rate  $P_o(\text{Se})$  for acid sample and  $P_M(\text{Se})$  for plutonium sample,
- The ratios  $I_o(\text{Se})$ ,  $I_o(\text{Co})$ ,  $I_M(\text{Se})$  and  $I_M(\text{Co})$ , where  $I$  is the background corrected ratio of the source count rate (Se or Co) to the relevant cadmium count rate. The index 'o' refers to an acid sample, and 'M' refers to a plutonium sample.

A representative set of data, obtained from  $6 \times 300$  s runs for each parameter, is given in Table IV. For each parameter the observed standard deviation  $S$  and the standard deviation due to the counting statistics,  $\sigma$ , are given for comparison.

As can be seen from the data in Table IV, the observed standard deviations in the cadmium measurements vary between 0.5 and 0.8%. The expected counting

TABLE II. MEASUREMENT RESULTS AT THE TUI  
( $K = 0.00644$ , acid sample for straight-through measurement)

Sample	Concentration declared (g/L)	Concentration measured (g/L)	$\Delta\rho$ (g/L)	$\Delta\rho$ (%)	$\sigma$ (%)	$\Delta\rho$ (%) <sup>a</sup>
S5	30.32 $\pm$ 0.23	29.2 $\pm$ 1.8 28.65 $\pm$ 1.8 26.19 $\pm$ 1.8	-1.12 -1.67 -4.13	-3.7 -5.5 -13.3	6.2 6.2 6.2	+4.1 +2.3 -5.6
S4	50.50 $\pm$ 0.15	48.07 $\pm$ 1.8 49.17 $\pm$ 1.8	-2.43 -1.33	-4.8 -2.6	3.7 3.7	-0.8 +1.3
S3	100.25 $\pm$ 0.28	99.62 $\pm$ 2.2 98.43 $\pm$ 2.1 98.52 $\pm$ 2.1	-0.65 -1.82 -1.73	-0.6 -1.8 -1.7	2.2 2.2 2.2	+0.4 -0.7 -0.6
S31	100.42 $\pm$ 0.22 (8M HNO <sub>3</sub> )	98.08 $\pm$ 2.1	-2.34	-2.3	2.1	-1.2
S2	151.05 $\pm$ 0.35	150.2 $\pm$ 2.6	-0.85	-0.6	1.7	-0.5
S12	180.18 $\pm$ 0.28 ( $\pm$ 5.52 U)	179.9 $\pm$ 2.9 180.1 $\pm$ 2.9	+0.10 -0.08	+0.2 0.0	1.6 1.6	-0.4 -0.3
S13	180.34 $\pm$ 0.44 ( $\pm$ 2.38 U)	181.5 $\pm$ 2.9	+1.60	+0.6	1.6	+0.4

S11	180.40 ± 0.43 (± 10.47 U)	180.5 ± 2.9 180.7 ± 2.9	+0.10 +0.30	0.0 +0.2	1.6 1.6	-0.2 0
S1	200.36 ± 0.34	202.7 ± 3.1	+2.34	+1.2	1.5	+0.8
		200.6 ± 3.1	+0.24	+0.1	1.5	-0.3
		200.9 ± 3.1	+0.54	+0.3	1.5	-0.1
		201 ± 3.1	+0.64	+0.3	1.5	0
		201.5 ± 3.1	+1.14	+0.6	1.5	+0.2
S0	Pure acid, 3M	-2.08 ± 1.7				
S6	220 U	-6.23 ± 3.0				

<sup>a</sup>  $\Delta\rho$  (%) obtained from fitted parameters  $K = 0.00656$  and  $\ln t_0 = -0.019$  (see Section 4).

TABLE III. MEASUREMENT RESULTS FOR Pu OUTPUT SAMPLES IN A CONCENTRATION RANGE OF ABOUT 10–350 g Pu/L ( $K = 0.00665$ )

Sample	Ratio of measured to declared value	Expected standard deviation for measured value (%)	Difference between declared and measured value (%)
1	0.998	0.9	−0.2
2	1.010	1.0	+1.0
3	0.998	1.3	−0.2
	0.995	1.3	−0.5
	0.994	1.3	−0.6
4	1.029	1.4	+2.9
	1.023	1.4	+2.3
5 <sup>a</sup>	1.021	3.0	+2.1
	1.005	2.9	+0.5
	1.004	3.0	+0.4
6	1.091	5.7	+9.1
	1.139	5.7	+13.9
7	0.944	18	−5.6
	0.737	18	−26.3
	1.006	18	+0.6

<sup>a</sup> This sample was used in a different series of measurements, where the calibration constant was 0.00675 (from sample 1 using an empty vial for background measurements).

statistics error is 0.1%. Similarly, the cobalt and selenium measurements show a variation of 0.3–0.7% at a counting statistics error level of 0.2%. The variations in the cadmium, cobalt and selenium measurements are propagated in the respective  $I_0$  and  $I_M$  values, which are used to determine the concentration values.

Measurements carried out in the laboratory with different Davidson MCAs and a  $^{57}\text{Co}$  source ( $8 \times 3000$  s) confirmed that the stability of the instrument varies between about 0.5 and 0.7%.

### 3.3.2. Reproducibility

Some samples were measured repeatedly over two or three days to check the reproducibility. Each day a new calibration run was carried out using an acid sample. The measurements confirmed that the results were reproducible to within the counting statistics error, which, depending on the age of the source, varied for samples between 100 and 200 g/L from about 2% (fresh sources) to about 4% (sources seven months old) for a measurement time of  $2 \times 300$  s.

TABLE IV. EXPECTED ( $\sigma$ ) AND OBSERVED (S) STANDARD DEVIATIONS (%) OF VARIOUS PARAMETERS FOR  $6 \times 300$  s ACID AND Pu SAMPLE MEASUREMENT

	Cd peak (88 keV)				Source peak				Source/Cd ratio			
	P(Se)		P(Cd)		P(Se)		P(Co)		I(Se)		I(Co)	
	S	$\sigma$	S	$\sigma$	S	$\sigma$	S	$\sigma$	S	$\sigma$	S	$\sigma$
Acid sample	0.8	0.1	0.7	0.1	0.3	0.2	0.7	0.2	0.9	0.22	0.9	0.22
Pu sample	0.6	0.1	0.5	0.1	0.4	0.2	0.3	0.2	0.7	0.22	0.9	0.22

The optimum measurement time to yield a precision in the assay result compatible with the stability of the instrument would be  $(5-6) \times 300$  s for concentrations between 100 and 200 g/L.

### 3.3.3. Practical aspects of use of equipment

There was no safety concern expressed by operators with respect to the glove box installation of the instrument. The mechanical stability of the tripod is, however, not sufficient and the risk that the instrument may tip over accidentally should be minimized by construction of an easily movable support car.

The exchange of sources causes no difficulty, and can be done in about one hour by experienced staff. At each of the installations where the K-edge densitometer was used, a sample holder was left permanently in the glove box to save inspectors and operators time and effort in decontamination and bagging-in and -out operations.

### 3.3.4. Effects of exchange of sources, detectors and MCAs on assay results

Source exchange is a normal requirement for the K-edge densitometer owing to the half-lives of the Co (271 d), Se (120 d) and Cd (463 d) sources. The resulting decrease in the count rate per day is about 0.4% for the Se, 0.2% for the Co and 0.1% for the Cd source.

After seven months, the Co and Se sources were exchanged for new sources and this opportunity was taken to check the effect of the exchange of the sources on the measured results. Other components which, for various reasons, may require replacement in the field are the Ge detector and the MCA. Therefore, these two components were also replaced to check whether any significant influence on the measurement results occurred.

The experiment was essentially carried out with two reference plutonium samples having concentrations of about 100 and 150 g Pu/L. There were two

TABLE V. EFFECT OF EXCHANGE OF INSTRUMENT COMPONENTS AND SOURCES ON VALUE OF  $r$ 

	D1, G1, SO	D1, G1, SF	D2, G2, SO	D2, G2, SF
Concentration (g Pu/L)	$r + S (\sigma)$	$r + S (\sigma)$	$r + S (\sigma)$	$r + S (\sigma)$
99.66	$1.917 \pm 0.02$ (0.01)	$1.957 \pm 0.01$ (0.015)	$1.971 \pm 0.02$ (0.015)	$1.978 \pm 0.02$ (0.02)
Normalized value	$1.000 \pm 0.01$	$1.025 \pm 0.01$	$1.028 \pm 0.01$	$1.032 \pm 0.01$
149.4	$2.667 \pm 0.02$ (0.01)	$2.733 \pm 0.03$ (0.01)	$2.749 \pm 0.03$ (0.03)	$2.795 \pm 0.03$ (0.03)
Normalized value	$1.000 \pm 0.008$	$1.025 \pm 0.01$	$1.031 \pm 0.01$	$1.048 \pm 0.01$

Davidson MCAs (D1, D2), two Ge detectors (G1, G2) and two sets of Co and Se sources (seven months old (SO) and fresh (SF)). The K-edge body and the sample holder configuration were not changed. Two samples were used for these measurements.

The results of the measurements are presented in Table V. Because the transmission ratio  $r$  (see definition in Section 3.1.3) should be invariant to these types of exchanges and also time independent, this parameter was chosen for comparison.

The following conclusions can be drawn:

- For one instrument configuration (D1, G1), the source exchange (fresh source for old source) resulted in an increase of about 2.5% for both samples. For the other configuration (D2, G2), the source exchange resulted in increases of 0.3 and 1%. The standard deviation of the measured  $r$ -values was about 1%.
- For a given K-edge body-source configuration, there was a change in  $r$  of about 3% from one MCA-detector configuration to another for the set with old sources, and of about 1–2% when the same replacement was done with the new sources in the K-edge body.
- The effects were consistent in both samples.

The change in  $r$  due to source replacement for the same instrument configuration is an effect which was also noticed in another way. When the  $\ln r$  values were plotted as a function of concentration  $\rho$  for the different installations, where the K-edge densitometer was used at different times, a parallel upward shift in the lines was observed with increasing age of the sources (Fig. 1 and Section 4). More precise measurements are, however, required to check whether this observation can be confirmed.



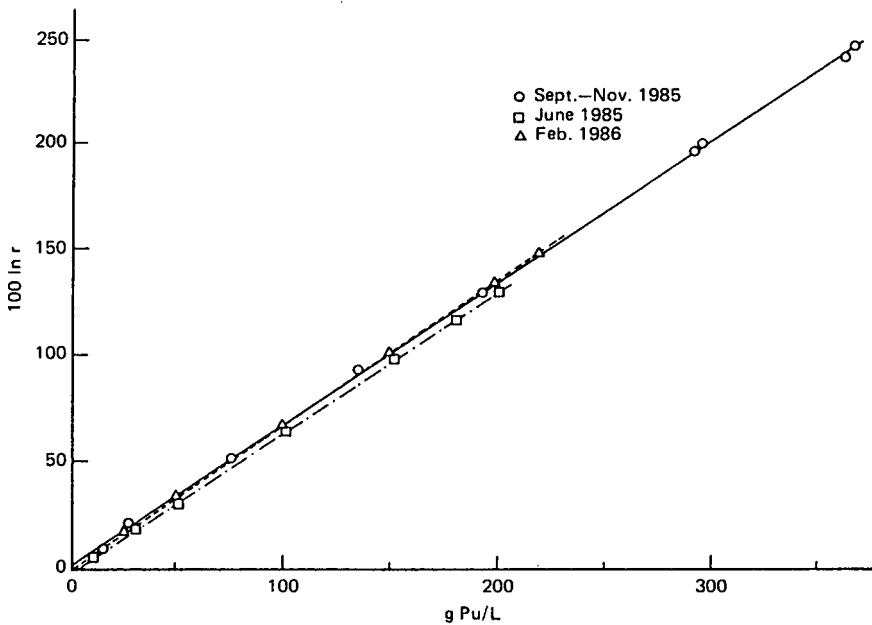


Fig. 1. Transmission ratio ( $\ln r$ ) as a function of Pu concentration.

#### 4. OVERALL EVALUATION OF DATA

Section 3 discussed the measurement data as obtained in the field, i.e. based on a calibration run with a plutonium sample which was chosen as a reference. With this procedure calibration constants varying between about 0.00637 and about 0.00690 were obtained. In order to check whether one unique calibration constant could be determined and to check the overall variation in the data, all values of  $\ln r$  were used and plotted as a function of concentration  $\rho$  (Fig. 1). Only the mean value of  $\ln r$  for a given concentration was used. A fit including all data led to quite large deviations in the measured values from the declared values.

Because there was a high correlation of the data in the individual data sets obtained within a measurement campaign, the data were subdivided and analysed in three groups as indicated in Fig. 1.

A linear regression of the data to the function

$$\ln r = K\rho + \ln r_0$$

for the data from June 1985 gave the following results:

$$K = 0.00656 \pm 1.2 \times 10^{-5} (\pm 0.2\%)$$

$$\ln r_0 = -0.019 \pm 0.001$$

The correlation coefficient was  $R^2 = 0.99998$ .

The same fit for the data obtained during September and November 1985 gave the following values:

$$K = 0.00665 \pm 7 \times 10^{-5} (\pm 1\%)$$

$$\ln r_0 = 0.014 \pm 0.01$$

$$R^2 = 0.9997$$

The fit of the data obtained in February 1986 gave:

$$K = 0.00680 \pm 1.7 \times 10^{-5} (\pm 0.25\%)$$

$$\ln r_0 = -0.013 \pm 0.002$$

$$R^2 = 0.99996$$

As can be seen from these data there is a remarkable increase in the slope (i.e. calibration constant) of the data over the period in question. The data set of February 1986 contains measurements with both instrument and source sets, whereas the measurement data in June, September and November 1985 were obtained with the same hardware configuration and sources as installed in June 1985.

The following observations can be made:

- At zero Pu concentration  $\ln r$  is often non-zero (see, for example, the acid measurement result in Table II);
- Only for the measurements in September and November 1985 is  $\ln r_0$  statistically zero;
- The use of the fitted  $K$  and  $\ln r_0$  parameters has two remarkable effects:
  - The systematic bias as observed, for example, in the measurements at the TUI (see Section 3.1.3) disappears,
  - The deviations between measured and declared Pu concentrations are smaller;
- For the data in Table II, the mean of all  $\Delta\rho$  values was  $-1.7\% \pm 3.3\%$ . Use of the values  $K = 0.00656$  and  $\ln r_0 = -0.019$  obtained from the June 1985 fit gives a mean  $\Delta\rho$  of  $-0.1\% \pm 1.8\%$ , i.e. the systematic bias and the variation are considerably reduced.

In order to show the overall spread in the data, the values of the parameters obtained from the least squares fits were taken to recalculate  $\rho$  and to plot  $\Delta\rho/\rho$ , where  $\Delta\rho$  is the measured value minus the declared value, as a function of the Pu concentration for individual  $2 \times 300$  s runs. The data are given in Fig. 2, where the dashed line corresponds to the expected standard deviation for the individual measurement.

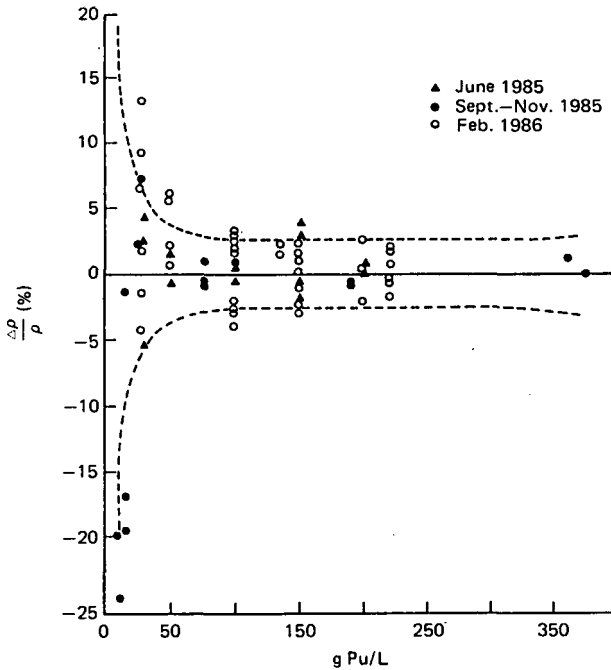


Fig. 2.  $\Delta\rho/\rho$  as a function of Pu concentration.

## 5. RECOMMENDATIONS AND CONCLUSIONS

There is a clear need for a portable K-edge densitometer in plants with plutonium nitrate solutions. The densitometer can be used to reduce the number of samples to be transported for chemical analysis. If the precision can be improved, it could replace chemical analysis to a larger extent. It is, however, required that another sample position and other software be used to permit also the measurement of the isotopic composition of plutonium.

There is a need to improve the electronic stability of the instrument, which was found to be a severe limitation for making more precise measurements by increasing the measurement time. All indications suggest that the Davidson MCA (main amplifier) needs improvements.

The large count rate variation from straight-through measurements to sample measurements with high concentrations results in a spectrum shift. The regions of interest (ROIs) are determined by the straight-through measurements with the sources and the FWHM of the photopeaks. These ROIs are then maintained for all measurements, although shifts of more than five channels were noticed when the count rate changed from sample to sample. The ROIs should be determined before each

measurement as in the case of the acid measurement (or by using a spectrum stabilizer).

The values of the calibration constant determined from the fit of  $\ln r$  as a function of Pu concentration for all data in a measurement campaign gave better consistency and smaller differences with respect to the operators' declared values. It is unclear, however, why the calibration 'constant' was not constant over the eight-month period from June 1985 to February 1986 (it increased by about 4%). This is an important aspect for in-field application which needs to be solved. The need to use the acid assay result for the  $\ln r_0$  value in the data evaluation should also be further examined.

The change of hardware components did not produce significant differences, but a better understanding of the origin of some differences is required.

The difference between measured and declared values was of the order of 1–2% in the concentration range of about 100–300 g Pu/L. This error could be decreased to slightly lower than 1% by increasing the measurement time and using the fitting procedure described in Section 4. A further improvement is only possible if the stability of the instrument is improved.

The software mentioned above should also allow for repeat measurements for straight-through and sample measurements.

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# NEUTRON MULTIPLICITY FOR NEUTRON INDUCED FISSION OF $^{235}\text{U}$ , $^{238}\text{U}$ AND $^{239}\text{Pu}$ AS A FUNCTION OF NEUTRON ENERGY\*

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## Abstract

NEUTRON MULTIPLICITY FOR NEUTRON INDUCED FISSION OF  $^{235}\text{U}$ ,  $^{238}\text{U}$  AND  $^{239}\text{Pu}$  AS A FUNCTION OF NEUTRON ENERGY.

Recent developments in the theory and practice of neutron correlation ('coincidence') counting require knowledge of the higher factorial moments of the  $P_\nu$  distribution (the probability that  $\nu$  neutrons are emitted in a fission) for the case where the fission is induced by bombarding neutrons of more than thermal energies. In contrast to the situation with spontaneous and thermal neutron induced fission, where, with a few exceptions, the  $P_\nu$  is reasonably well known, in the fast neutron energy region almost no information is available concerning the multiplicity beyond the average value  $\langle \nu \rangle$ , even for the most important nuclides. The reason for this is the difficulty of such experiments, with consequent statistically poor and physically inconsistent results. In the paper, previously unpublished data made available from a fast neutron fission multiplicity experiment have been used to extract usable multiplicity probabilities from otherwise intractable data, using smoothing techniques supplemented by mathematical and physical plausibility arguments. In contrast, it appears that attempts to apply systematics to deducing such parameters as  $P_\nu$  to the required accuracy will not work because of particular differences between even neighbouring nuclides. Thus, despite the difficulties, further experimental work is required, supplemented by data evaluation, if necessary, as in the paper.

## 1. INTRODUCTION

For many technical purposes it suffices to know only the first moment,  $\langle \nu \rangle$  ('nubar'), of the neutron multiplicity probability  $P_\nu$  that  $\nu$  prompt neutrons are emitted in a fission. Certain applications, however, do require knowledge of the higher moments of the  $P_\nu$  distribution. A case in point, and the immediate reason for the present work, are several methods for the analysis of nuclear material by what amounts to an autocorrelation performed on the pulse train from a detector exposed to the unknown sample. Then it can be shown that the second factorial moment

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\* Research carried out under the auspices of the United States Department of Energy under Contract No. DE-AC02-76CH00016.

$\langle \nu(\nu - 1) \rangle = \Sigma \nu(\nu - 1)P_\nu$  is proportional to the fission rate in the sample; and that the third factorial moment  $\langle \nu(\nu - 1)(\nu - 2) \rangle = \Sigma \nu(\nu - 1)(\nu - 2)P_\nu$  can be of use in disentangling spontaneous fission, which can be related directly to the amount of material present, from induced fission, which is only partly related to the amount of material, since it is also a function of geometry, density and other artefacts.

Calculating the effects of induced fission in the sample, i.e. fission caused by either spontaneous fission generated neutrons or those produced by  $(\alpha, n)$  processes, or, in the case of certain nuclear material assay systems, by neutrons from an external source bombarding the sample, clearly requires knowledge of these moments for neutron energies ranging from the thermal to the fast (fission) neutron energy region [1-5].

While it has been possible to glean from the published literature, or from private communications, much information (though never enough) for nuclides<sup>1</sup> undergoing spontaneous fission induced by thermal neutrons or neutrons of tens of kilo-electronvolts [6-13], we are unaware of any published  $P_\nu$  data for fission induced by neutrons in the few mega-electronvolt range, which are crucial to calculations of fast induced fission correlation.

We became aware, though, of extensive unpublished work by Fréhaud and collaborators in which neutron multiplicities were derived for the fast neutron induced fission of  $^{235}\text{U}$ ,  $^{238}\text{U}$  and  $^{239}\text{Pu}$ . These  $P_\nu$  data, despite their uniqueness and importance, were not published because they are obviously flawed, owing mainly, it is presumed, to poor counting statistics and the way in which counting statistical uncertainties propagate in the transformations connecting experimentally observed neutron multiplicities with the  $P_\nu$  distributions (see below). Certain  $P_\nu$  at some energies are negative, which is physically and mathematically impossible for a probability, and, considered as a function of energy, the  $P_\nu$  often exhibit fluctuations obviously related to poor counting statistics rather than to any real physical process.

## 2. SCOPE AND METHODOLOGY

Our task has been to salvage from these data the multiplicity  $P_\nu$  as a function of incident neutron energy  $E_n$ . The overall procedure may be described as data smoothing guided by general physical and mathematical principles. These will be described immediately below and illustrated with particular cases cited later on.

Though the data furnished cover up to  $E_n \approx 25$  MeV, results will be quoted only over the range 0-10 MeV, as in the region 10-15 MeV and certainly for the higher energies the data become prohibitively unreliable even though smoothing processes are employed. Another problem is that the furnished data only begin at  $E_n = 1.36$  MeV. Fortunately, comparatively accurate thermal neutron induced

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<sup>1</sup>  $^{257}\text{Fm}$ ,  $^{252}\text{No}$ ,  $^{242}\text{Cm}$ ,  $^{244}\text{Cm}$ ,  $^{246}\text{Cm}$ ,  $^{248}\text{Cm}$ ,  $^{246}\text{Cf}$ ,  $^{250}\text{Cf}$ ,  $^{252}\text{Cf}$ ,  $^{254}\text{Cf}$ ,  $^{236}\text{Pu}$ ,  $^{238}\text{Pu}$ ,  $^{239}\text{Pu}$ ,  $^{240}\text{Pu}$ ,  $^{241}\text{Pu}$ ,  $^{242}\text{Pu}$ ,  $^{233}\text{U}$ ,  $^{235}\text{U}$ ,  $^{238}\text{U}$ .

fission cross-sections do exist for ( $^{235}\text{U} + n$ ) and ( $^{239}\text{Pu} + n$ ); more will be said of this later.

Any of the  $P_\nu$  as a function of  $E_n$  has the appearance of a bell shaped curve or a portion of one. For the smaller  $\nu$ -values only the decreasing tail of the bell appears, starting at  $E_n = 0$ . For intermediate values of  $\nu$  the whole bell shaped curve appears except the ascending part cut off at  $E_n = 0$ ; the function rises to a maximum and then declines, presumably approaching zero for high enough  $E_n$ . The  $P_\nu$ , being probabilities, must sum to unity at any energy. Therefore, the only way for  $\langle \nu \rangle$  to increase monotonically, as it is well known to do from experimental results, is for the  $P_\nu$  to increase for higher values of  $\nu$ , with a corresponding decrease in the relative importance of those  $P_\nu$  for smaller values of  $\nu$ . There is no evidence from the data of Fréhaut et al. for more complicated behaviour of any of the  $P_\nu$  as a function of  $E_n$  than this.

Therefore, a basic premise of the fitting procedure was that each  $P_\nu$  for any  $\nu$  was considered to be a smooth function of neutron energy describable by a low order least squares fitted polynomial in  $E_n$ . In fact, in the region 0–10 MeV, as will be seen below, there was no need for polynomials higher than the fourth order. In some cases these polynomial fits are good representations of the data for energies beyond 10 MeV, though in other cases the fit rapidly becomes unusable after 10 MeV.

In the fitting procedure the attempt was made, however, to use data beyond  $E_n = 10$  MeV to help establish the trend in the data up to that point. Similarly, thermal neutron data for  $^{235}\text{U}$  and  $^{239}\text{Pu}$  were used to anchor the start of the various  $P_\nu$  curves for these nuclides, since the Fréhaut data start only at  $E_n = 1.36$  MeV. Not surprisingly, there are no thermal neutron data for ( $^{238}\text{U} + n$ ) as the cross-sections are too small below the effective threshold for induced fission. Instead, the obtained fit to the data between  $E_n = 1.36$  and 10 MeV was extrapolated to  $E_n = 0$  to yield an estimate for the multiplicity distribution between  $E_n = 0$  and 1.36 MeV for ( $^{238}\text{U} + n$ ).

After least squares fitting of the  $P_\nu$  as a polynomial in  $E_n$ , the  $P_\nu$  set at a given energy will depart slightly from the normalization condition  $\sum P_\nu = 1$ , and so they were renormalized.

The average value for  $\nu$ ,  $\langle \nu \rangle = \sum \nu P_\nu$ , has been determined as a function of  $E_n$  by independent experiments to a greater accuracy than can be determined from the  $P_\nu$ . These experiments basically determined a gross count rate  $G$  in terms of a detector efficiency  $\epsilon$  and a source strength  $q$ :

$$G = \epsilon \langle \nu \rangle q \quad (1)$$

The renormalized  $P_\nu$  at any given energy predict a  $\langle \nu \rangle = \sum \nu P_\nu$  close to but not precisely equal to the best available values for  $\langle \nu \rangle$  determined as a function of  $E_n$  from Eq. (1). The differences were reconciled by considering them formally as though they had arisen through error or uncertainties in the efficiency of a (hypothetical) detector in an experiment determining the  $P_\nu$ . In such an experiment, the

observed neutron multiplicity  $Q_n$ , i.e. that  $n$  neutrons are observed from a given fission event, is related to the  $P_\nu$  by

$$Q_n = \sum P_\nu \binom{\nu}{n} \epsilon^n (1 - \epsilon)^{\nu-n} \quad (2)$$

The  $P_\nu$  are obtained from this expression by inverting the relation:

$$P_\nu = \sum Q_n \binom{n}{\nu} \epsilon^{-n} (\epsilon - 1)^{n-\nu} \quad (3)$$

As applied in this instance, the normalized  $P_\nu$  derived from the least squares fitting of the Fréhaut data were used to obtain a hypothetical set  $Q_n$  based on an assumed value for  $\epsilon$ . The normalized set  $P_\nu$  defines a value  $\langle \nu \rangle = \sum \nu P_\nu$ . A set  $P'_\nu$  that would yield a value  $\langle \nu \rangle' = \sum \nu P'_\nu$  considered more correct can be thought of as being related to a detector efficiency  $\epsilon'$  such that  $\langle \nu \rangle' \epsilon' = \langle \nu \rangle \epsilon$ , since experimentally  $\langle \nu \rangle$  and  $\epsilon$  are inversely related. Then the hypothetical  $Q_n$  can be used to obtain the set  $P'_\nu$  corresponding to  $\epsilon'$  from

$$P'_\nu = \sum Q_n \binom{n}{\nu} (\epsilon')^{-n} (\epsilon' - 1)^{n-\nu} \quad (4)$$

This procedure then produces a normalized set of  $P'_\nu$  as a function of  $E_n$  which yields the proper value of  $\langle \nu \rangle' = \sum \nu P'_\nu$  at any given  $E_n$ , and can be considered to be related to the same set of observables  $Q_n$  as the original set  $P_\nu$  that was smoothed and renormalized.

The  $P'_\nu$  are expected to be close to the least squares fits to the  $P_\nu$ , and indeed are. In principle a new least squares fit could be made to the  $P'_\nu$  and the results renormalized and again reconciled to the best values available for  $\langle \nu \rangle$ . This process is rapidly convergent, so much so that the resultant changes in  $P_\nu$  would be a marginal improvement considering the precision of the original data.

### 3. DATA SMOOTHING (POLYNOMIAL FITTING) PROCEDURE

The statistical uncertainties in the  $P_\nu$  are naturally greater the fewer the observations of a given multiplicity. The larger  $P_\nu$  therefore have better statistical precision. Since all observed  $Q_n$  with  $n < \nu$  can be considered as stemming from a multiplicity  $\nu$  with probability  $P_\nu$  because the efficiency  $\epsilon$  is less than 1 (see Eqs (2, 3)), the error propagation from the observed  $Q_n$  to the derived  $P_\nu$  is not simple. Nevertheless there is a qualitative correspondence between the magnitude of the  $P_\nu$  and the 'smoothness' of the data points. Thus  $P_3$ ,  $P_4$  and  $P_5$ , which are comparatively large in the region 0–10 MeV, were relatively easy to fit, while  $P_0$ ,  $P_7$  and  $P_8$  were the most difficult.

$P_0$  is small to begin with ( $\sim 0.01$ ) at thermal energy and decreases rapidly to zero at roughly 10 MeV.  $P_7$  and  $P_8$  start out even smaller at thermal energies; while



they increase rapidly, another difficulty that manifests itself is an increase in the general variability of the data at higher energies which is not explicable on the basis of the number of fissions observed at a given energy, being inferior for high energies ( $\sim 10$  MeV) compared with low energies ( $\sim 2$  MeV). In fact, the higher energy data have significantly more fissions analysed; for example, for  $(^{235}\text{U} + \text{n})$  4532 fissions were analysed for  $E_n = 1.87$  MeV, but 11 374 fissions were analysed for  $E_n = 9.74$ , and these are typical of their respective neighbouring values. (Incidentally, a typical modern  $P_v$  experiment for spontaneous fissions or thermal neutron energies would involve  $10^5$  or  $10^6$  fissions. This points out the basic problem of the Fréhaut data, lack of sufficient statistics.)

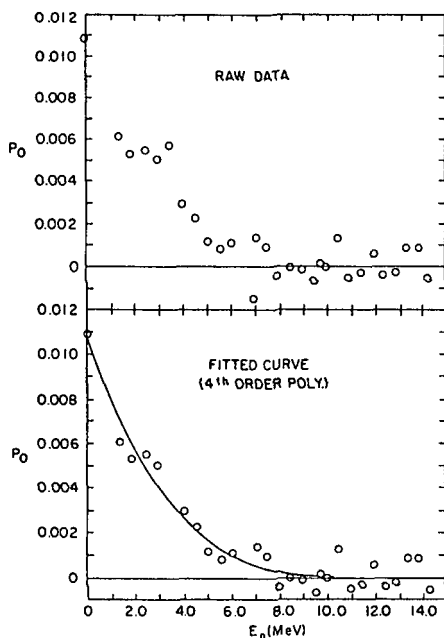
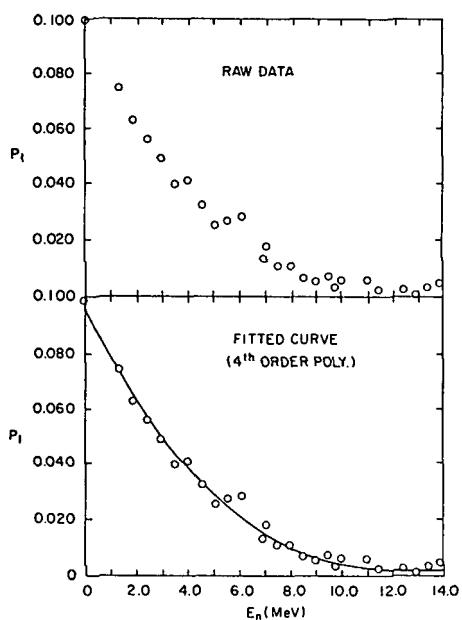
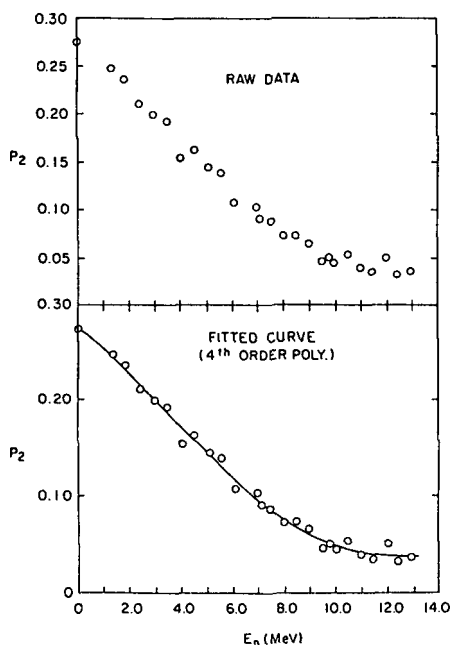
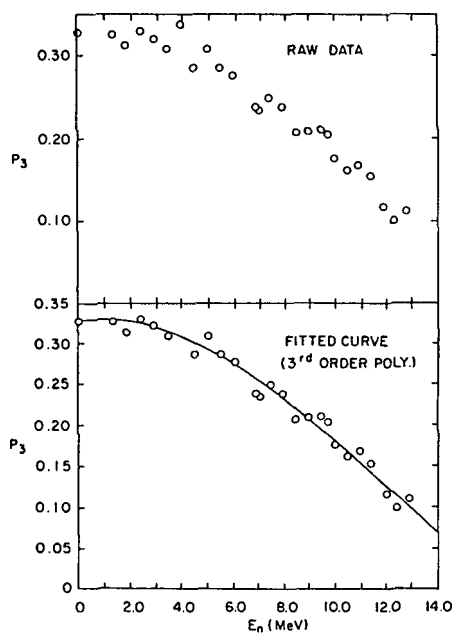
It is not practical to present every case treated, but the problems encountered and solutions adopted in reducing the data to a usable form can best be illustrated with examples, using  $(^{239}\text{Pu} + \text{n})$  for  $\nu = 0$  to 8 and  $(^{235}\text{U} + \text{n})$  for  $\nu = 7, 8$  as fairly representative.

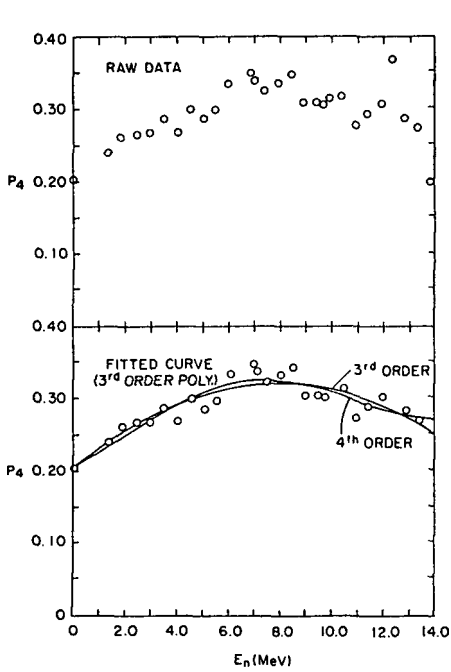
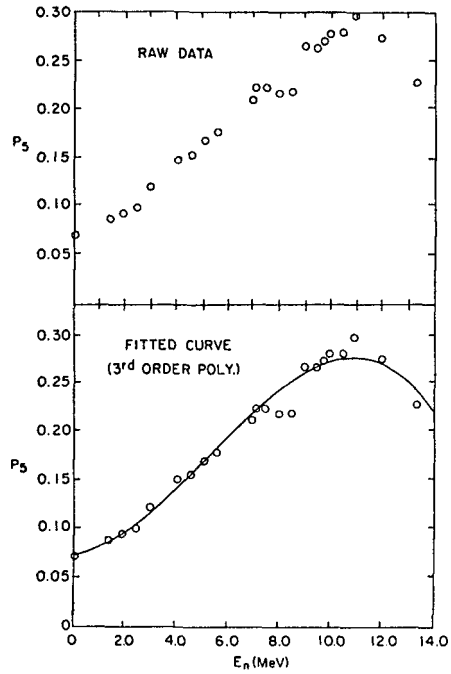
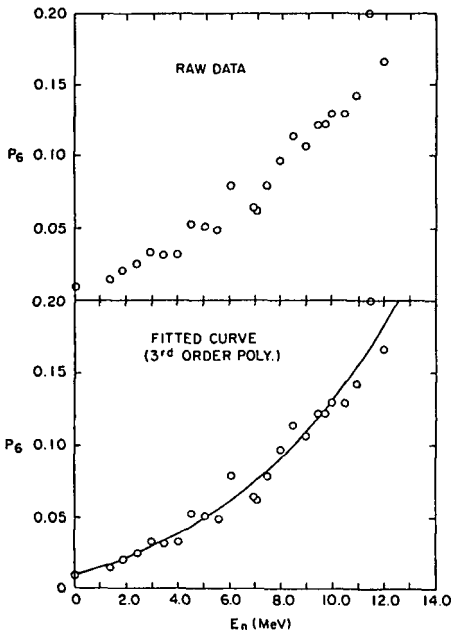
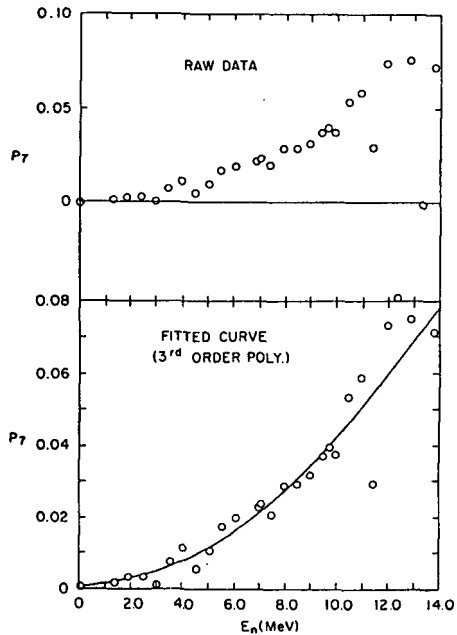
These cases are illustrated in Figs 1–10, which show the raw data (i.e. thermal plus Fréhaut data points) above and the data plus the fitted polynomial in powers of  $E_n$  below.

The polynomial used was the lowest order which would give a good fit. Although the (Hewlett-Packard 9845) software for the polynomial fitting routine also produced goodness of fit parameters, the criteria that proved practical in deciding the goodness of fit were those based in the general knowledge of how the  $P_v$  functions of energy behaved, and that the fitted curve should intercept the  $P_v$  axis close to the thermal value, since the thermal values are assumed to be one or two orders of magnitude more accurate than the typical data for  $E_n > 0$ .

The curve fitting routine did not allow for weighting the data points according to statistical uncertainty, but a simple subterfuge, entering a data pair more than once, produced the same effect. The only points weighted this way were the thermal values. It was found that giving the thermal value a weight of 5 or 10 tended to improve the agreement of the fitted curve at  $E_n = 0$ , while not noticeably affecting the goodness of fit among the Fréhaut data points. In a few cases, the choice of the order of the polynomial between otherwise equivalent fits was decided on the basis of which gave best agreement with the thermal value. However, in all cases where a thermal value was available, there was no difficulty in fitting a low order polynomial (fourth order or less) to the thermal value and the Fréhaut data, a point which will be commented on later.

Though the resulting fits are meant to be considered representations of the respective  $P_v$  only in the region 0–10 MeV, data points for  $E_n$  up to about 14 MeV were used in order to be more certain of the course of the function in the neighbourhood of 10 MeV. In some of Figs 1–10 the fitted polynomial is shown extending beyond 10 MeV. In some cases, the particular  $P_v$  continues to be represented by the fitted curve beyond 10 MeV, but in other cases the formal analytical extension of the polynomial is clearly non-physical, as the curve would depart from zero when it should remain there, or reverse direction when it should continue monotonically. This point will also be discussed later.

FIG. 1.  $P_0$  versus  $E_n$  for  $^{239}\text{Pu}$ .FIG. 2.  $P_1$  versus  $E_n$  for  $^{239}\text{Pu}$ .FIG. 3.  $P_2$  versus  $E_n$  for  $^{239}\text{Pu}$ .FIG. 4.  $P_3$  versus  $E_n$  for  $^{239}\text{Pu}$ .

FIG. 5.  $P_4$  versus  $E_n$  for  $^{239}\text{Pu}$ .FIG. 6.  $P_5$  versus  $E_n$  for  $^{239}\text{Pu}$ .FIG. 7.  $P_6$  versus  $E_n$  for  $^{239}\text{Pu}$ .FIG. 8.  $P_7$  versus  $E_n$  for  $^{239}\text{Pu}$ .

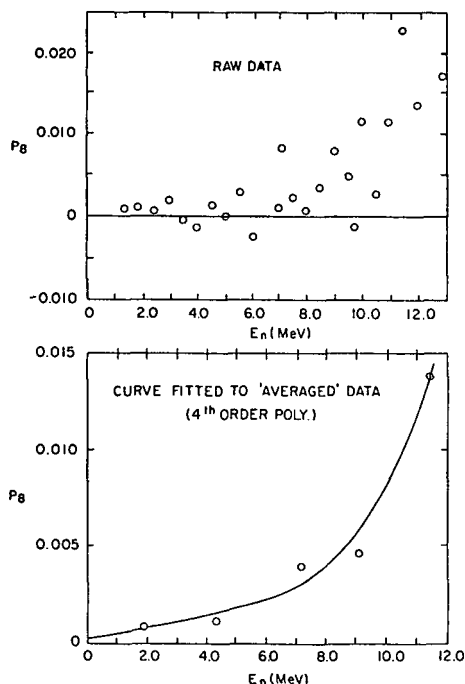


FIG. 9.  $P_8$  versus  $E_n$  for  $^{239}\text{Pu}$ .

In relatively few situations, data points were discarded when they seemed to lie off the tentatively fitted curves by what seemed to be significantly more than the typical variability in the data for that particular  $P$ , and seemed to interfere with the goodness of fit to the rest of the data.

We would like now to make some particular remarks. The curve for  $P_0$  (Fig. 1) illustrates the inadequacy of a finite order polynomial representation for more than a limited range, as the data indicate that  $P_0$  is statistically zero from about  $E_n = 10$  MeV onwards. The situation is similar for  $P_1$  (Fig. 2), where the best fit to the data over the whole region tends to change direction at  $E_n = 13$  MeV, whereas (considering the data up to  $E_n \approx 10$  MeV) it should asymptotically approach zero.

Figure 5 ( $P_4$ ) illustrates that a higher order least squares fitted polynomial, while a better fit by the least squares criterion, is not necessarily better physically, so that the third order polynomial was chosen as the better representation.

$P_8$  (Fig. 9) presented two problems: the extreme scatter of the data points, though a trend is clearly visible, and the absence of a thermal value to serve as an 'anchor point'. The effect of scatter was greatly reduced by averaging groups of points with respect to ordinate and abscissa, and fitting the curve to these points as

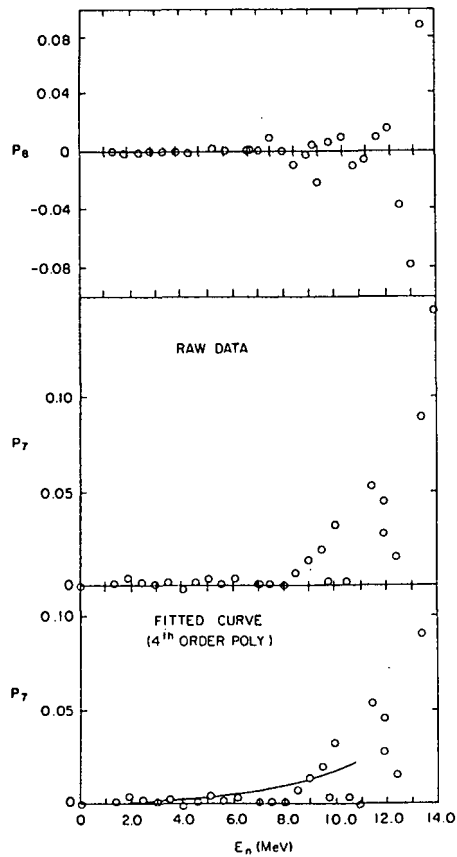
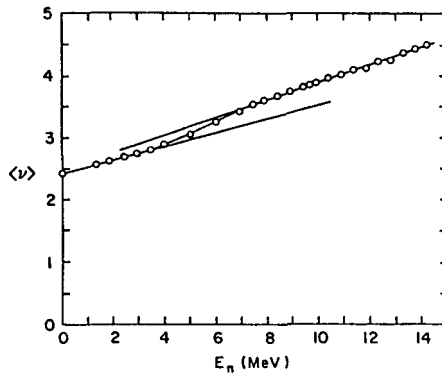
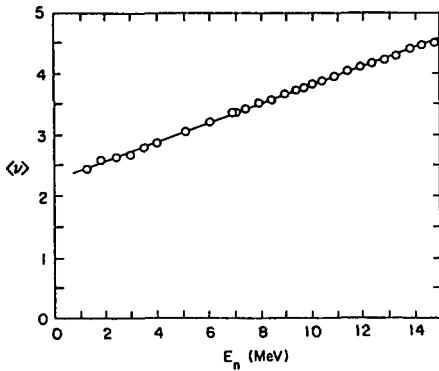
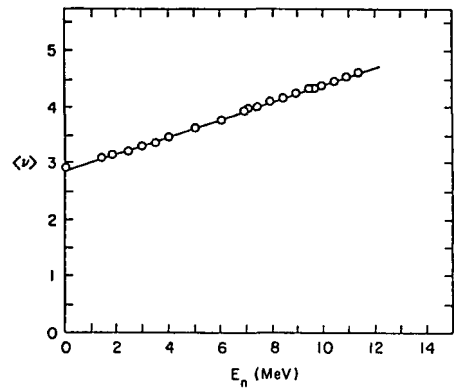


FIG. 10.  $P_7$ ,  $P_8$  versus  $E_n$  for  $^{235}\text{U}$ .

plotted in the lower part of the figure. The fitted curve for  $P_8$  predicts a small non-zero value for  $P_0$  at  $E_n = 0$ , which is not inconsistent with the zero value reported, considering the assigned standard deviations for these data.

Figure 10 illustrates  $P_7$  and  $P_8$  for  $(^{235}\text{U} + n)$ . A non-zero thermal value is reported for  $P_7$ , but not for  $P_8$ , for this system. As can be seen, the Fréhaut data for  $(^{235}\text{U} + n)$  are consistent with  $P_8 = 0$  even beyond  $E_n = 10$  MeV. However, in agreement with the non-zero thermal value for  $P_7$ , the Fréhaut data do show a small non-zero component for  $P_7$  from  $E_n = 1.36$  to about 6 MeV, followed by what seems to be an abrupt rise starting at about  $E_n = 8.5$  MeV, with scatter in the data. It was decided that the abrupt rise is an artefact due to chance occurrence of three consecutive low values in the region from  $\sim 7$  to  $\sim 8$  MeV. Therefore, a curve was fitted which agrees with the thermal values and extrapolates to an average of the Fréhaut data in the region from 10 to 11 MeV.

FIG. 11.  $\langle \nu \rangle$  for  $(^{235}\text{U} + n)$ .FIG. 12.  $\langle \nu \rangle$  for  $(^{238}\text{U} + n)$ .FIG. 13.  $\langle \nu \rangle$  for  $(^{239}\text{Pu} + n)$ .

#### 4. 'NORMALIZING' THE SMOOTHED DATA

Here, 'normalized' will be used to mean that  $\sum P_\nu = 1$  and  $\sum \nu P_\nu = \langle \nu \rangle$ , where  $\langle \nu \rangle$  is a prescribed value.

After smoothing of each  $P_\nu$  treated as a function of  $E_n$  by least squares fitting of a polynomial to the data, the  $P_\nu$  at a given energy will no longer sum to unity although the sum will be only about 1% or so off. The renormalized data are then transformed according to Eqs (2-4), subject to the condition that they produce the value of  $\langle \nu \rangle$  appropriate to that energy.

The curves of  $\langle \nu \rangle$  versus  $E_n$  used in principle could have been those evaluated from the literature. In the present situation it was thought better to ensure consistency and minimize uncertainties due to the as yet untried technique for evaluating the data

TABLE I. COMPARISON OF  $P_\nu(E_n = 0)$  FOR  $(^{235}\text{U} + n)$ , DERIVED IN DIFFERENT WAYS

$\nu$	A	B	C	SD
	$P_\nu$	$P_\nu$	$P_\nu$	
0	0.031 900 4	0.031 652 0	0.031 722 3	0.001 5
1	0.172 521 3	0.171 800 3	0.171 707 1	0.001 4
2	0.336 139 7	0.335 792 6	0.336 199 1	0.003 1
3	0.303 879 8	0.304 464 0	0.303 969 5	0.000 4
4	0.126 615 5	0.127 158 2	0.126 945 9	0.003 6
5	0.026 184 3	0.026 351 0	0.026 679 3	0.002 6
6	0.002 617 0	0.002 638 3	0.002 632 2	0.000 9
7	0.000 142 1	0.000 143 6	0.000 144 9	0.000 06
$\langle \nu \rangle$	2.410 520 0	2.414 000 0	2.414 000 0	0.007
$\langle \nu(\nu - 1) \rangle$	4.623 107 3	4.636 465 5	4.638 2	0.029 7
$\langle \nu(\nu - 1)(\nu - 2) \rangle$	6.776 987 6	6.806 381 3	6.817 6	0.168 3
$\langle \nu(\nu - 1) \rangle / \langle \nu \rangle^2$	0.795 632 5	0.795 632 5	0.795 93	0.005 10
$\langle \nu^2 \rangle - \langle \nu \rangle^2$	1.223 020 7	1.223 069 5	1.224 8	0.029 7
$\langle \nu^2 \rangle$	7.033 627 3	7.050 465 4	7.052 2	0.029 7

<sup>A</sup> Curve fitted,  $\langle \nu \rangle = 2.410\ 52$ .

<sup>B</sup> Curve fitted, normalized to  $\langle \nu \rangle = 2.414\ 00$ .

<sup>C</sup> Consensus data, normalized to  $\langle \nu \rangle = 2.414\ 00$ .

by using  $\langle \nu \rangle$  for the three nuclides as determined by the same group that furnished the  $P_\nu$  data [14].

A second choice made was to smooth the  $\langle \nu \rangle$  data rather than use the quoted experimental points. This is because away from an energy region roughly less than 1 MeV there is no indication of any structure in a plot of  $\langle \nu \rangle$  versus  $E_n$ ; the relation seems quite linear, with the exception of what seems to be a definite change between 4 and 7 MeV for  $(^{235}\text{U} + n)$  (Fig. 11). Thus, with the exceptions noted, deviations from linearity could be considered statistical (Figs 12 and 13).

The process of normalizing, in both senses, the smoothed distributions is illustrated in Tables I and II for  $(^{235}\text{U} + n)$  and  $(^{239}\text{Pu} + n)$ , respectively, using the data at  $E_n = 0$ . Column A contains the smoothed data at  $E_n = 0$  normalized only in the sense of  $\Sigma P_\nu = 1$ . Column B contains the data normalized additionally so that  $\Sigma \nu P_\nu = \langle \nu \rangle$ , where the value of  $\langle \nu \rangle$  was determined independently. Column C shows

TABLE II. COMPARISON OF  $P_\nu(E_n = 0)$  FOR  $(^{239}\text{Pu} + n)$ , DERIVED IN DIFFERENT WAYS

$\nu$	A	B	C	
	$P_\nu$	$P_\nu$	$P_\nu$	SD
0	0.010 881 9	0.010 859 3	0.010 860 1	0.000 03
1	0.099 384 8	0.099 599 4	0.099 304 4	0.002 8
2	0.274 774 7	0.274 980 0	0.274 873 7	0.000 3
3	0.326 932 2	0.326 854 9	0.327 050 0	0.004 1
4	0.204 692 2	0.204 475 4	0.204 766 0	0.008 7
5	0.072 745 8	0.072 600 4	0.072 772 0	0.013 3
6	0.009 739 5	0.009 713 8	0.009 743 0	0.002 7
7	0.000 646 3	0.000 645 2	0.000 631 0	0.000 9
8	0.000 272 6	0.000 271 6	—	—
$\langle \nu \rangle$	2.877 370 8	2.876 000 0	2.876 000 0	0.009
$\langle \nu(\nu - 1) \rangle$	6.756 962 0	6.750 525 4	6.743 5	0.018 4
$\langle \nu(\nu - 1)(\nu - 2) \rangle$	12.635 021 2	12.616 971 6	12.544 7	0.053 9
$\langle \nu(\nu - 1) \rangle / \langle \nu \rangle^2$	0.816 130 9	0.816 130 9	0.815 28	0.002 23
$\langle \nu^2 \rangle - \langle \nu \rangle^2$	1.355 070 5	1.355 149 8	1.348 1	0.018 4
$\langle \nu^2 \rangle$	9.634 332 8	9.626 525 4	9.619 5	0.018 4

<sup>A</sup> Curve fitted,  $\langle \nu \rangle = 2.877\,37$ .

<sup>B</sup> Curve fitted, normalized to  $\langle \nu \rangle = 2.876\,00$ .

<sup>C</sup> Consensus data, normalized to  $\langle \nu \rangle = 2.876\,00$ .

$P_\nu$  at  $E_n = 0$  from our earlier evaluation of thermal  $P_\nu$  data [6–13], together with the standard deviations assigned at that time.

## 5. FINAL RESULTS AND EVALUATION

The final results of this process of data evaluation are presented in tabular form (Tables III–V) and graphically (Figs 14 and 15) to show the general truncated (on the abscissa scale) bell shape. The  $E_n = 0$   $P_\nu$  for  $(^{238}\text{U} + n)$  are obtained of course by extrapolation of the fitted functions. The  $(^{238}\text{U} + n)$  curves are qualitatively similar to those for  $(^{239}\text{Pu} + n)$  and so are not presented.



TABLE III.  $P_\nu$  VERSUS  $E_n$  (MeV) FOR ( $^{235}\text{U} + n$ )

$\nu$	$E_n$	0	1	2	3	4	5	6	7	8	9	10
0	0.031 722 3	0.023 789 8	0.018 398 9	0.014 146 0	0.011 520 8	0.007 849 8	0.004 627 2	0.002 465 9	0.001 270 2	0.000 728 8	0.000 437 3	
1	0.171 707 1	0.155 552 5	0.138 489 1	0.119 483 9	0.103 262 4	0.080 201 0	0.056 332 1	0.036 095 7	0.021 609 0	0.013 487 9	0.008 011 5	
2	0.336 199 1	0.321 651 5	0.306 212 3	0.288 307 5	0.271 684 9	0.245 659 5	0.213 229 6	0.178 863 4	0.147 222 7	0.123 120 0	0.100 232 9	
3	0.303 969 5	0.315 043 3	0.321 756 6	0.326 656 8	0.328 342 6	0.330 817 5	0.329 040 7	0.321 050 7	0.308 303 2	0.294 939 0	0.277 928 3	
4	0.126 945 9	0.144 473 2	0.162 867 3	0.183 601 4	0.202 120 6	0.229 164 6	0.259 980 6	0.289 253 7	0.312 395 0	0.325 825 1	0.334 261 1	
5	0.026 679 3	0.035 601 3	0.045 597 2	0.056 911 3	0.067 445 6	0.083 691 2	0.104 597 4	0.128 257 6	0.152 254 0	0.173 187 9	0.196 610 0	
6	0.002 632 2	0.003 433 9	0.005 569 4	0.008 942 6	0.012 892 4	0.018 701 6	0.026 560 4	0.036 088 7	0.046 244 9	0.055 173 7	0.065 009 0	
7	0.000 144 9	0.000 454 6	0.001 109 3	0.001 950 4	0.002 730 7	0.003 914 8	0.005 632 2	0.007 924 4	0.010 700 9	0.013 537 6	0.017 509 9	
$\langle \nu \rangle$	2.414 000 0	2.523 670 0	2.636 820 0	2.762 340 0	2.873 840 0	3.038 699 9	3.231 609 9	3.427 280 0	3.604 190 0	3.739 590 0	3.874 980 0	
$\langle \nu(\nu - 1) \rangle$	4.638 2	5.101 375 8	6.622 988 3	6.228 194 4	6.789 247 6	7.625 491 6	8.645 780 9	9.735 712 4	10.774 870 0	11.613 322 8	12.497 055 3	
$\langle \nu(\nu - 1)(\nu - 2) \rangle$	6.817 6	8.001 220 1	9.476 469 9	11.263 753 6	12.988 228 6	15.572 621 4	18.859 627 6	22.558 617 1	26.279 115 8	29.444 447 7	32.964 597 1	
$\langle \nu(\nu - 1) \rangle / \langle \nu \rangle^2$	0.795 93	0.800 981 0	0.808 734 9	0.816 221 2	0.822 046 7	0.825 833 0	0.827 878 0	0.828 835 9	0.829 462 4	0.830 440 5	0.832 278 7	
$\langle \nu^2 \rangle - \langle \nu \rangle^2$	1.224 8	1.256 135 6	1.306 988 5	1.360 012 0	1.404 131 6	1.430 494 0	1.434 088 0	1.416 744 3	1.388 874 2	1.368 379 1	1.356 565 2	
$\langle \nu^2 \rangle$	7.052 2	7.625 045 8	8.259 808 4	8.990 534 4	9.663 087 6	10.664 191 5	11.877 390 9	13.162 992 3	14.379 060 0	15.353 912 8	16.372 035 3	

TABLE IV.  $P_\nu$  VERSUS  $E_n$  (MeV) FOR ( $^{238}\text{U} + n$ )

$\nu$	$E_n$	0	1	2	3	4	5	6	7	8	9	10
0	0.039 648 4	0.029 907 6	0.022 665 1	0.017 025 3	0.012 493 2	0.008 816 7	0.005 873 6	0.003 599 7	0.001 949 5	0.000 876 7	0.000 327 1	0.000 818 4
1	0.252 954 1	0.204 321 5	0.162 402 0	0.127 299 2	0.098 479 7	0.075 174 4	0.056 598 5	0.042 046 0	0.030 908 7	0.022 658 7	0.016 818 4	0.011 111 4
2	0.293 954 4	0.299 588 6	0.295 726 3	0.284 054 0	0.266 187 5	0.243 657 0	0.217 925 2	0.190 409 5	0.162 505 5	0.135 605 8	0.111 111 4	0.089 243 4
3	0.264 447 0	0.291 488 9	0.311 909 8	0.326 019 2	0.334 493 8	0.337 971 1	0.336 886 3	0.331 457 5	0.321 739 2	0.307 691 9	0.289 243 4	0.289 243 4
4	0.111 175 8	0.130 148 0	0.152 878 6	0.177 957 9	0.204 011 6	0.229 790 1	0.254 157 5	0.276 041 3	0.294 379 2	0.308 081 6	0.316 016 6	0.316 016 6
5	0.031 226 1	0.036 311 9	0.043 423 3	0.052 657 5	0.064 046 8	0.077 597 1	0.093 312 7	0.111 207 5	0.131 307 4	0.153 644 6	0.178 248 4	0.178 248 4
6	0.005 934 7	0.007 363 8	0.009 747 3	0.013 099 7	0.017 383 7	0.022 561 9	0.028 620 0	0.035 568 3	0.043 434 7	0.052 254 9	0.062 061 7	0.062 061 7
7	0.000 543 6	0.000 694 7	0.000 931 8	0.001 346 7	0.002 030 8	0.003 068 9	0.004 543 1	0.006 538 7	0.009 147 4	0.012 468 2	0.016 606 6	0.016 606 6
8	0.000 115 8	0.000 175 1	0.000 315 9	0.000 540 5	0.000 873 0	0.001 362 6	0.002 083 1	0.003 131 6	0.004 628 4	0.006 717 6	0.009 566 5	0.009 566 5
$\langle \nu \rangle$	2.275 378 1	2.430 563 1	2.585 748 1	2.740 933 1	2.896 118 1	3.051 303 1	3.206 488 1	3.361 673 1	3.516 858 1	3.672 043 2	3.827 228 1	3.827 228 1
$\langle \nu(\nu - 1) \rangle$	4.340 582 4	4.896 020 2	5.515 163 5	6.192 689 2	6.924 105 3	7.706 625 0	8.539 378 1	9.423 250 1	10.360 565 7	11.354 732 4	12.409 902 5	12.409 902 5
$\langle \nu(\nu - 1)(\nu - 2) \rangle$	6.993 708 8	8.139 568 0	9.617 431 <sup>3</sup>	11.422 940 9	13.551 890 8	16.008 366 2	18.808 243 9	21.979 708 5	25.562 233 6	29.604 817 1	34.163 896 7	34.163 896 7
$\langle \nu(\nu - 1)(\nu)^2 \rangle$	0.838 379 9	0.828 761 2	0.824 870 9	0.824 294 5	0.825 526 7	0.827 739 2	0.830 552 3	0.833 853 5	0.837 671 6	0.842 095 5	0.847 226 8	0.847 226 8
$\langle \nu^2 \rangle - \langle \nu \rangle^2$	1.438 615 2	1.418 946 2	1.414 818 4	1.420 908 1	1.432 723 5	1.447 477 6	1.464 300 5	1.484 077 2	1.509 132 7	1.542 874 7	1.589 455 6	1.589 455 6
$\langle \nu^2 \rangle$	6.615 960 4	7.326 583 2	8.100 911 5	8.933 622 3	9.820 223 4	10.757 928 1	11.745 866 2	12.784 923 2	13.877 423 8	15.026 775 6	16.237 130 6	16.237 130 6

TABLE V.  $P_\nu$  VERSUS  $E_n$  (MeV) FOR ( $^{239}\text{Pu} + n$ )

$\nu$	$E_n$	0	1	2	3	4	5	6	7	8	9	10
0	0.010 882 6	0.008 484 2	0.006 255 5	0.004 586 0	0.003 290 8	0.002 275 0	0.001 489 3	0.000 906 1	0.000 464 7	0.000 280 0	0.000 206 4	
1	0.099 491 6	0.079 003 0	0.061 192 1	0.047 787 9	0.037 439 0	0.029 141 6	0.022 236 9	0.016 352 8	0.011 328 3	0.007 146 0	0.003 885 6	
2	0.274 889 8	0.253 617 5	0.226 560 8	0.198 300 2	0.170 419 6	0.143 764 5	0.119 043 9	0.096 811 0	0.077 520 1	0.061 557 7	0.049 254 8	
3	0.326 919 6	0.328 987 0	0.326 063 7	0.318 466 7	0.307 186 2	0.292 800 6	0.275 629 7	0.255 852 4	0.233 592 6	0.208 981 0	0.182 207 8	
4	0.204 606 1	0.232 811 1	0.258 835 4	0.279 281 1	0.294 856 5	0.306 390 2	0.314 490 8	0.319 456 6	0.321 328 9	0.320 012 1	0.315 415 9	
5	0.072 683 4	0.080 016 1	0.095 607 0	0.115 895 0	0.139 259 4	0.164 164 7	0.189 289 7	0.213 488 8	0.235 661 4	0.254 584 6	0.268 728 2	
6	0.009 728 2	0.015 558 1	0.022 470 5	0.030 112 8	0.038 673 8	0.048 434 3	0.059 735 3	0.072 973 9	0.088 618 3	0.107 234 4	0.129 514 3	
7	0.000 630 1	0.001 176 0	0.002 594 6	0.004 847 1	0.007 870 1	0.011 615 1	0.016 082 8	0.021 333 9	0.027 489 5	0.034 725 5	0.043 265 4	
8	0.000 168 5	0.000 346 9	0.000 520 5	0.000 723 3	0.001 004 6	0.001 414 0	0.002 001 7	0.002 824 6	0.003 953 1	0.005 478 6	0.007 521 7	
$\langle \nu \rangle$	2.876 000 0	3.008 880 0	3.162 830 0	3.316 780 0	3.470 730 0	3.624 680 0	3.778 630 0	3.932 580 0	4.086 530 0	4.240 490 0	4.394 440 0	
$\langle \nu(\nu - 1) \rangle$	6.747 988 8	7.410 774 0	8.239 700 8	9.124 137 3	10.054 437 7	11.024 355 1	12.031 178 4	13.075 411 9	14.160 252 7	15.291 145 3	16.475 099 7	
$\langle \nu(\nu - 1)(\nu - 2) \rangle$	12.589 411 4	14.592 844 4	17.321 044 5	20.441 684 6	23.906 361 3	27.686 434 8	31.777 120 6	36.197 468 4	40.988 367 2	46.210 552 6	51.941 646 9	
$\langle \nu(\nu - 1)(\nu)^2 \rangle$	0.815 824 2	0.818 566 3	0.823 682 3	0.829 389 4	0.834 672 6	0.839 100 2	0.842 634 9	0.845 474 0	0.847 933 1	0.850 370 0	0.853 141 3	
$\langle \nu^2 \rangle - \langle \nu \rangle^2$	1.352 612 9	1.366 295 4	1.399 037 4	1.439 887 6	1.479 201 2	1.510 729 9	1.531 764 0	1.542 806 6	1.547 055 0	1.549 879 9	1.558 436 5	
$\langle \nu^2 \rangle$	9.623 988 8	10.419 653 9	11.402 530 8	12.440 917 3	13.525 167 7	14.649 035 1	15.809 808 4	17.007 991 9	18.246 782 8	19.531 635 3	20.869 539 7	

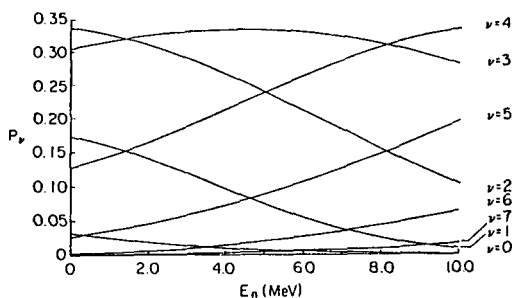


FIG. 14.  $P_\nu$  versus  $E_n$  for  $(^{235}\text{U} + n)$ .

The important question at this point is how accurately these salvaged data represent reality. There are only  $P_\nu$  data for  $E_n = 0$  and then only for  $(^{235}\text{U} + n)$  and  $(^{239}\text{Pu} + n)$ . As mentioned above, the smoothed data of Fréhaud extrapolate very well to those  $E_n = 0$  values in the two cases where such are available, even before weighting those points (although in recognition of their superior statistical precision and accuracy, they were accordingly weighted in the final fitting process). From this it can be argued that the lack of smoothness in the Fréhaud data is statistical and that there may not be any serious systematic errors. In this case a smoothing procedure which tends to level statistical fluctuations would have validity.

Another indication of validity in the procedure is that in the two cases (Tables I and II) where the thermal values obtained by the data processing procedure could be compared with  $P_\nu$  values obtained independently, the two sets are well within the uncertainties assigned to the independently obtained  $P_\nu$  values, even without the  $E_n = 0$  values being weighted.<sup>2</sup>

Finally, each  $P_\nu$  set was derived independently on at least two occasions separated widely enough in time so that some of the subjective details of the process, such as which points to drop from the fitting procedure, how far beyond 10 MeV points should be included, what was the lowest order best fit, etc., were forgotten. Nevertheless, aside from minor differences due to our acquiring improved skills at fitting, the respective derived  $P_\nu$  sets were all essentially equivalent well within the attributed uncertainties.

It would be very desirable to compare the Fréhaud data as processed above with  $P_\nu$  values derived from new experiments. However, it was the lack of such experiments together with the low probability, in the present state of reactor physics or nuclear data research, that such experiments would be performed that was a major reason for the present work.

<sup>2</sup> The data in Tables I and II do have weighting for the thermal values, which of course increases the apparent agreement.

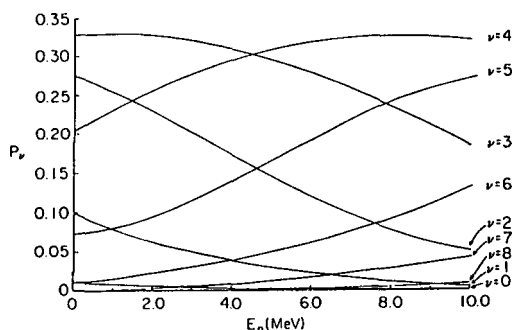


FIG. 15.  $P_\nu$  versus  $E_n$  for  $(^{239}\text{Pu} + n)$ .

All things considered, we think the present set of data will prove adequate for safeguards work, and the kind of methodology used may even indicate how to design future  $P_\nu$  experiments so as to most economically and efficiently utilize experimental time and facilities.

The lack of a thermal  $P_\nu$  set to anchor the fitted curves does, however, reduce the reliability of  $(^{238}\text{U} + n)$  compared with the others.

## 6. FURTHER REMARKS

A reason for our interest in  $P_\nu$  is the hope that some information regarding the systematics of neutron multiplicity in fission would result. This could be important both from the standpoint of improving the theory of the fission process and to compensate for the lack of experimental data in many instances.

The more we have delved into the details of  $P_\nu$ , the more it has seemed that such systematics as are noticed (e.g. Ref. [15]) are true only in a fairly approximate sense and are not necessarily accurate enough relations to be useful in many technical applications, such as neutron correlation counting.

In the present paper it is seen that for two nuclides  $\langle \nu \rangle$  is quite linear over the range  $E_n > 1$  MeV, but must be represented by two lines joined by a smooth transition for the third nuclide. That same nuclide has no discernible  $P_8$  component in the studied region, whereas the other nuclides, with only three and four nucleons more, have small but definitely non-zero  $P_8$  components.

On the other hand, a plot of the second factorial moment  $\langle \nu(\nu - 1) \rangle$  for the three nuclides reveals a curious consistency which we are at a loss to explain as being some artefact of the data smoothing process (Fig. 16). The three plots of  $\langle \nu(\nu - 1) \rangle$  each seem to be made up of two straight line segments with a smooth transition between.

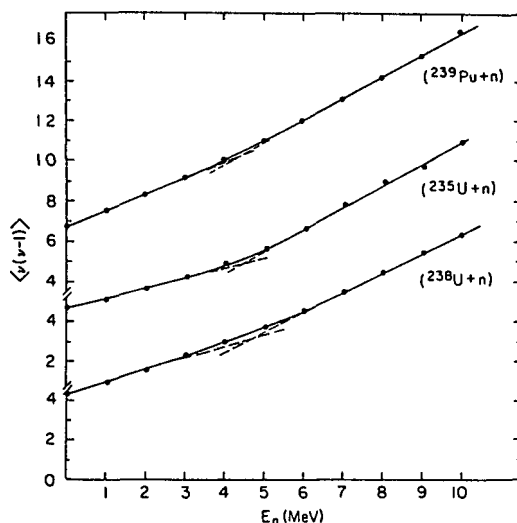


FIG. 16.  $\langle \nu(\nu - 1) \rangle$  versus  $E_n$ .

The extrapolations of these straight line portions all seem to intersect between 4 and 5 MeV.

These and other speculations can ultimately only be satisfied with more experimental work.

## ACKNOWLEDGEMENTS

We are very much indebted to J. Fréhaut and co-workers at the CEA, Centre d'études de Bruyères-le-Châtel, Montrouge, for so kindly furnishing their data.

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# MONTE CARLO CALCULATIONS OF COINCIDENCE COUNT RATES FOR VERIFYING SAMPLE DECLARATIONS

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## Abstract

### MONTE CARLO CALCULATIONS OF COINCIDENCE COUNT RATES FOR VERIFYING SAMPLE DECLARATIONS.

To verify a sample declaration the author proposes the following steps: to calculate on the basis of the declaration the total and coincidence count rates to be expected in a measurement; and to accept or reject the declaration in the light of statistical considerations based on experimental (and calculational) uncertainties. As the sample-detector configuration is very complex in geometry and composition, the Monte Carlo method may be used to evaluate the relevant quantities. Furthermore, a 'least squares adjustment' of the isotopic vector of the sample may be performed using measured count rates and the initially declared isotopic vector within one of the models which result when simplifying the exact equations for the measurement quantities. The sensitivities necessary for the adjustment procedure may also be evaluated with the Monte Carlo method.

## 1. INTRODUCTION

In this paper a proposal is made for solving the following problem. A safeguards inspector is supposed to verify the declaration that a sample contains, for example,

$Q_0$  (kg) of an  $(\alpha, n)$  emitting material

$Q_i$  (kg) of a spontaneously fissioning material of type  $i$  ( $i = 1, 2, \dots, L$ )

$Q$  (kg) of fissile material of type  $\alpha$  ( $\alpha = 1, 2, \dots, K$ )

(all materials are assumed to be 'homogeneously distributed' over the volume of the sample).

The current procedure for verifying this declaration consists in forgetting about these statements, performing some (coincidence) count rate measurements and trying to recover the actual material composition of the sample by interpreting the measurements on the basis of (very) approximate theoretical models. These approximations result from drastic simplifications of the exact theoretical equations which are known.

Full use of these exact theoretical equations could be made, however, if we used the declared sample composition and geometry as input to these equations and calculated (predicted) the results to be expected in an actual experimental measurement. The calculated (predicted) results could then be used to *verify* the actual measurements. Statistical considerations based on the experimental (and calculational) uncertainties may then decide whether the sample declaration should be accepted or rejected.

The measurement data and the initial declaration of the isotopic vector  $Q_i^{(0)}$ ,  $i = 1, 2, \dots, L$ , may also be used in another way. The measurable quantities  $X_1$  (total count rate) and  $X_2$  (coincidence rate) are, in general, complicated functions of the isotopic vector  $Q_i$ . Several measurements of  $X_1$  and  $X_2$  on the same sample in different detector facilities provide us with a set of measured data  $X^{(0)} = \{X_\ell^{(0)}, \ell = 1, 2, \dots, L\}$  and their uncertainty covariance matrix  $C_X$ . We may use these data, combined with the declaration values  $Q^{(0)} = \{Q_i^{(0)}\}$  and the corresponding uncertainty covariance matrix  $C_Q$ , to improve our knowledge of the isotopic vector  $Q = \{Q_i\}$  by performing a 'least squares adjustment', which means by choosing  $Q$  such that

$$F = (X^{(0)} - X(Q))^T C_X^{-1} (X^{(0)} - X(Q)) + (Q^{(0)} - Q)^T C_Q^{-1} (Q^{(0)} - Q) \quad (1)$$

becomes a minimum.

The sensitivity coefficients  $S_{ik} = (\partial X_i / \partial Q_k)_{Q^{(0)}}$ , necessary for the adjustment procedure, are to be evaluated with the Monte Carlo method and depend on the model used to represent the analytical relation  $X = X(Q)$  between measured quantities  $X$  and isotopic vector  $Q$ . The presence of some (amount  $Q_0$  of) moderating material (e.g. water) in the sample may be taken into account by making a first estimate  $Q_0^{(0)}$  (and its uncertainty) and adding  $Q_0$  to the vector  $Q_k$  and adjusting the complete vector  $(Q_0, Q_k)$ .

To turn this proposal into a practical recipe, we have, therefore,

- To select a set of physical quantities which might be measurable,
- To establish theoretical expressions relating the measured quantities (count rates) to the declared sample characteristics,
- To prepare these equations for numerical evaluation.

This will be done in the following sections.

## 2. SELECTION OF MEASURABLE QUANTITIES

For the purpose of identifying probe characteristics, we have, for example, the measurement of the following quantities in mind. Using one counter and randomly triggered (non-overlapping) time intervals  $I_\nu$ ,  $I_\mu$  and  $I_\lambda$ , we may measure the moments  $\overline{M_a}, \overline{M_a M_b}, \overline{M_a M_b M_c}$  ( $a, b, c = \nu, \mu, \lambda$ ), where  $M$  is the actual number

of counts in I. (To be more general, we could use two or more counters and place the intervals I on different counters.) In a similar way we may measure the moments of count numbers in non-overlapping time intervals  $I_\nu, I_\mu$  within a pulse triggered interval, which we denote by  $\overline{M}_a^{(0)}, \overline{M}_a \overline{M}_b^{(0)}$  ( $a, b = \nu, \mu$ ).

We recall that all the measurable quantities mentioned above may be expressed through the 'density' functions  $f_n(t_1, t_2, \dots, t_n)$ , where  $f_n(t_1, t_2, \dots, t_n) dt_1 dt_2 \dots dt_n$  is the probability of there being a count in each of the intervals  $\{t_i, t_i + dt_i\}$  regardless of how many counts are outside these intervals.

In the case of randomly triggered intervals, we have, for example,

$$\overline{N}_\nu = \int_{I_\nu} f_1(t) dt \quad (2)$$

$$\overline{N}_\nu^2 = \overline{N}_\nu + \int_{I_\nu} f_2(t_1, t_2) dt_1 dt_2 \quad (3)$$

$$\overline{N}_\nu^3 = \overline{N}_\nu + 3 \int_{I_\nu} f_2(t_1, t_2) dt_1 dt_2 + \int_{I_\nu} f_3(t_1, t_2, t_3) dt_1 dt_2 dt_3 \quad (4)$$

etc.

To interpret measurements using pulse triggered intervals, we introduce first conditional probabilities and put, for example,

$$\begin{aligned} f_2(t_1, t_2) &= f_1(t_1) f_2(t_2|t_1) \\ &= f_1(t_1) f_2(t_2 - t_1|0) \\ &= f_1(t_1) f_2(\tau|0); \quad \tau = t_2 - t_1 \end{aligned} \quad (5)$$

where  $f_2(\tau|0)d\tau$  is now the conditional probability of there being a count in  $d\tau$  if there was a count at  $\tau = 0$ . Similarly,

$$\begin{aligned} f_3(t_1, t_2, t_3) &= f_1(t_1) f_3(t_2, t_3|t_1) \\ &= f_1(t_1) f_3(t_2 - t_1, t_3 - t_1|0) \\ &= f_1(t_1) f_3(\tau_1, \tau_2|0); \quad \tau_1 = t_2 - t_1, \tau_2 = t_3 - t_1 \end{aligned} \quad (6)$$

where  $f_3(\tau_1, \tau_2|0)d\tau_1 d\tau_2$  is the conditional probability of measuring a count in  $d\tau_1$  and  $d\tau_2$  if there was a count at  $\tau = 0$ .

In this case the measurable moments may be expressed as:

$$\overline{N_\nu}^{(0)} = \int_{I_\nu} f_2(\tau|0) d\tau \quad (7)$$

$$\overline{N_\nu N_\mu}^{(0)} = \int_{I_\nu} d\tau_1 \int_{I_\mu} f_3(\tau_1, \tau_2|0) d\tau_2 \quad (8)$$

etc.

The density functions  $f_n$ , however, in turn may be expressed eventually by 'correlation functions'  $g_n$  which are given by the physics of the count producing process [1]. We give here only the first three relations between the  $f_n$  and  $g_n$  as higher  $f_n$  will hardly be needed:

$$f_1(t_1) = g_1(t_1) \quad (9)$$

$$f_2(t_1, t_2) = g_1(t_1)g_1(t_2) + g_2(t_1, t_2) \quad (10)$$

$$\begin{aligned} f_3(t_1, t_2, t_3) = & g_1(t_1)g_1(t_2)g_1(t_3) + g_1(t_1)g_2(t_2, t_3) + g_1(t_2)g_2(t_1, t_3) \\ & + g_1(t_3)g_2(t_1, t_2) + g_3(t_1, t_2, t_3) \end{aligned} \quad (11)$$

Thus, we arrive at the result that all our chosen measurable quantities may, finally, be expressed by (some time integrations over) the correlation functions  $g_n$ .

Our next problem is therefore to construct the  $g_n$  in terms of the physics of the process.

### 3. EVALUATION OF CORRELATION FUNCTIONS $g_n$

The physical meaning of the  $g_n$  for our problem is this:

$g_n(t_1, t_2, \dots, t_n) dt_1 dt_2 \dots dt_n$  is the probability:

- of there being a count in each of the intervals  $dt_i$ ,
- and that all neutrons (leading to this multiplet of counts) may be traced back to one common source event (spontaneous fission of some type).

To construct explicit expressions for the  $g_n$ , we restrict our considerations here to  $g_1$  and  $g_2$  ( $g_3$  is dealt with in the Appendix), and introduce  $Q_K(xs) dx ds$ ,

the mean number of fission events of type  $K$  in volume element  $dx$  and time interval  $ds$ . (We use index  $i$  for spontaneous fissions, index  $\alpha$  for induced fissions and  $K$  for fissions of any type.)

If  $\nu_K$  is the number of (fission) neutrons emitted in a fission event of type  $K$ , we define:

$$\mu_K^{(1)} = \bar{\nu}_K$$

$$\mu_K^{(2)} = \overline{\nu_K(\nu_K - 1)}$$

and  $D_K(t|xs)dt$  is the mean number of counts in the detector in time interval  $dt$  due to one fission neutron emitted in a fission event  $K$  at  $\{x, s\}$ .

For our application we may assume stationary situations, and thus:

$$Q_K(xs) = Q_K(x) \quad \text{time independent}$$

$$D_K(t|xs) = D_K(t - s|x)$$

Then we may write:

$$g_1(t_1) = \sum_i \mu_i^{(1)} \int Q_i(x) T_i(t_1|x) dx \quad (12)$$

$$g_2(t_1, t_2) = \sum_K \mu_K^{(2)} \int Q_K(x) T_K(t_1, t_2) dx \quad (13)$$

where

$$T_K(t_1, t_2, \dots, t_n|x) = \int_{-\infty}^{+\infty} \prod_{i=1}^n D_K(t_i|xs) ds \quad (14)$$

We see that the quantities needed for evaluating the expressions for  $g_1$  and  $g_2$  are:

- The space dependent induced fission densities  $Q_\alpha(x)$  (the spontaneous fission densities  $Q_i(x)$  are assumed to be known),
- The space and time dependent detector responses  $D_K(t|x)$ .

All these quantities may, in principle, be obtained by proper sampling of the relevant information in one Monte Carlo run (starting the histories with the spontaneous fission neutrons homogeneously distributed over the space of the

probe). The spatial integrations to be performed in the expressions for the  $g_i$  may be done by dividing the probe and detector volumes into (small) space cells and replacing the integrals with a summation over these cells.

We rewrite the integrals in the expressions for  $g_1$  and  $g_2$  in the form:

$$\int Q_K(x) T_K(t_1, t_2, \dots, t_n | x) dx = Q_K T_K(t_1, t_2, \dots, t_n) \quad (15)$$

where

$$Q_K = \int Q_K(x) dx$$

is the total number of K-type fissions per second and

$$T_K(t_1, t_2, \dots, t_n) = \int \frac{Q_K(x)}{Q_K} T_K(t_1, t_2, \dots, t_n | x) dx \quad (16)$$

is the spatial average of the function  $T_K(t_1, t_2, \dots, t_n | x)$  taken with the normalized fission distribution  $g_K(x) = Q_K(x)/Q_K$ . Note that  $T_K(t_1, t_2, \dots, t_n)$  (with respect to spatial averaging) is in essence the product of the space dependent detector responses. Therefore, we have to form the spatial average of the product of the D's.

The induced fission densities  $Q_\alpha(x)$  are produced by the spontaneous fissions  $Q_i(x)$ . The relation is:

$$Q_\alpha(x) = \sum_i \int Q_i(x') \mu_i^{(1)} F_{i\alpha}(x' \rightarrow x) dx' = \sum_i Q_i \mu_i^{(1)} F_{i\alpha} \quad (17)$$

where

$$F_{i\alpha}(x' \rightarrow x) dx$$

is the mean number of induced  $\alpha$ -type fissions in  $dx$  due to one  $i$ -fission neutron starting at  $x'$ , and

$$F_{i\alpha} = \int q_i(x') dx' \int F_{i\alpha}(x' \rightarrow x) dx$$

is the total mean number of induced  $\alpha$ -type fissions due to one i-fission neutron. Separating out the induced fissions in the expression for  $g_2$ , we obtain:

$$g_1(t_1) = \sum_i Q_i \mu_i^{(1)} T_i(t_1) \quad (18)$$

$$g_2(t_1, t_2) = \sum_i Q_i \left( \mu_i^{(2)} T_i(t_1, t_2) + \mu_i^{(1)} \sum_{\alpha} F_{i\alpha} \mu_{\alpha}^{(2)} T_{\alpha}(t_1, t_2) \right) \quad (19)$$

We shall now introduce some models to simplify these expressions.

#### 4. SIMPLIFIED MODELS

At first we assume the validity of the separation:

$$D_K(t|xs) = D_K(x)H(t|s) \quad (20)$$

where

$$D_K(x) = \int_{-\infty}^{+\infty} D_K(t|xs) dt$$

is the total number of counts due to one K-fission neutron and  $H(t|s)$  is a normalized time dependence (which under certain circumstances may be approximated by an exponential behaviour). This gives:

$$g_1(t_1) = \sum_i Q_i \mu_i D_i T(t_1) \quad (21)$$

$$g_2(t_1, t_2) = \sum_i Q_i \left( \mu_i^{(2)} \overline{D}_i^2 + \mu_i^{(1)} \sum_{\alpha} F_{i\alpha} \mu_{\alpha}^{(2)} \overline{D}_{\alpha}^2 \right) T(t_1, t_2) \quad (22)$$

where

$$\overline{D}_K^n = \int q_K(x) D_K^n(x) dx \quad (23)$$

$$T(t_1, t_2, \dots, t_n) = \int_{-\infty}^{+\infty} \prod_{i=1}^n H(t_i|s) ds \quad (24)$$

Furthermore, we assume the  $D_K(x)$  to be space independent and obtain:

$$g_2(t_1, t_2) = \sum_i Q_i \left( \mu_i^{(2)} \overline{D}_i^2 + \mu_i^{(1)} \sum_{\alpha} F_{i\alpha} \mu_{\alpha}^{(2)} \overline{D}_{\alpha}^2 \right) T(t_1, t_2) \quad (25)$$

At this point we may introduce several quantities characterizing

- The sample (probe) individually,
- The detector individually;

and the interaction between sample and detector (when the sample is put into its position in the detector) is then covered by balance equations. We divide the energy axis into group intervals and characterize the fission spectra by:

$X_{K\nu}$  the probability for a K-fission neutron to have an energy in group  $\nu$  ( $X$  is the matrix with elements  $X_{K\nu}$ ),

$\mu$  a diagonal matrix with elements  $\mu_{\beta}^{(1)}$ .

The isolated sample is characterized by the following quantities:

$P_{\nu\mu}^{(L)}$  the probability that a neutron produced in the (isolated) sample in group  $\nu$  ( $\nu$ -neutron) leaves the sample as a  $\mu$ -neutron ( $P^{(L)}$  is the matrix with elements  $P_{\nu\mu}^{(L)}$ ),

$P_{\nu\beta}^{(F)}$  the probability that a  $\nu$ -neutron produced in the (isolated) sample leads to an induced  $\beta$ -fission (matrix  $P^{(F)}$ ),

$P_{\nu}^{(A)}$  the probability that a  $\nu$ -neutron produced in the (isolated) sample is absorbed (in the sample).

Obviously, we have:

$$P_{\nu}^{(A)} + P_{\nu}^{(L)} + P_{\nu}^{(F)} = 1 \quad (26)$$

$$P_{\nu}^{(L)} = \sum_{\mu} P_{\nu\mu}^{(L)} \quad (27)$$

$$P_{\nu}^{(F)} = \sum_{\beta} P_{\nu\beta}^{(F)} \quad (28)$$

These probabilities are very easy to evaluate with the Monte Carlo method (on the basis of the given declaration) and serve as parameters to characterize the sample.



The multiplying properties of the isolated sample can now be expressed in terms of these probabilities and are defined by:

- $M_{\nu\mu}^{(0)} (M^{(0)})$  the mean number of  $\mu$ -neutrons leaving the (isolated) sample for one  $\nu$ -neutron produced in the sample,  
 $F_{\nu\alpha}^{(0)} (F^{(0)})$  the mean number of  $\alpha$ -type induced fissions due to one  $\nu$ -neutron produced in the sample,  
 $A_{\nu}^{(0)} (A^{(0)})$  the mean number of absorptions in a population generated by one  $\nu$ -neutron in the isolated sample.

The relations are:

$$M^{(0)} = P^{(L)} + P^{(F)} X M^{(0)} \quad (29)$$

$$F^{(0)} = P^{(F)} + P^{(F)} \mu X F^{(0)} \quad (30)$$

$$A^{(0)} = P^{(A)} + P^{(F)} \mu X A^{(0)} \quad (31)$$

The detector will be described by:

- $R_{\nu\mu}^{(D)} (R^{(D)})$  the mean number of  $\mu$ -neutrons reflected back from the detector into the sample for one  $\nu$ -neutron entering the detector from the sample,  
 $\epsilon_{\mu}$  the probability that a  $\mu$ -neutron entering the detector from the sample leads to a count (energy dependent sensitivity).

The interaction between the sample (when put in place) and the detector is then described by:

- $M_{\nu\mu} (M)$  the total mean number of  $\mu$ -neutrons leaving the sample for one  $\nu$ -neutron produced in the sample,  
 $F_{\nu\beta} (F)$  the total mean number of  $\beta$ -type induced fissions due to one  $\nu$ -neutron produced in the sample,  
 $A_{\nu} (A)$  the total mean number of absorptions in a population generated by one  $\nu$ -neutron in the sample.

The balance equations are now:

$$M = M^{(0)} + M^{(0)} R^{(D)} M \quad (32)$$

$$F = F^{(0)} + M^{(0)} R^{(D)} F \quad (33)$$

$$A = A^{(0)} + M^{(0)} R^{(D)} A \quad (34)$$

and the number of counts is related to the multiplication  $M$  by:

$$D = X M \epsilon \quad (35)$$

Note that for the evaluation of the correlation function  $g_2$ , we need  $D$  and  $F$ , and these quantities may eventually be expressed by:

- (a) The sample parameters  $P^{(L)}$ ,  $P^{(F)}$ ,  $P^{(A)}$ ,
- (b) The detector parameters  $R^{(D)}$ ,  $\epsilon$ .

With the help of the absorption probability we may formulate the overall neutron balance:

$$1 + \sum_{\beta} F_i(\mu_{\beta}^{(1)} - 1) = \sum X_{i\nu} [A_{\nu} + M_{\nu\mu} (1 - R_{\mu})] \quad (36)$$

where

$$R_{\mu} = \sum_{\lambda} R_{\mu\lambda}$$

In the case of one energy group only, these equations reduce to:

$$D_K = M\epsilon \quad (37)$$

$$M^{(0)} = \frac{P^{(L)}}{1 - \sum P_{\beta}^{(F)} \mu_{\beta}^{(1)}} \quad (38)$$

$$M = \frac{M^{(0)}}{1 - M^{(0)} R^{(D)}} \quad (39)$$

$$F_{K\beta} = \frac{F_{K\beta}^{(0)}}{1 - M^{(0)} R^{(D)}} = M \frac{P_{\beta}^{(F)}}{P^{(L)}} \quad (40)$$

$$F_{K\beta}^{(0)} = \frac{P_{\beta}^{(F)}}{1 - \sum P_{\beta}^{(F)} \mu_{\beta}^{(1)}} \quad (41)$$

In this case we have:

$$P^{(A)} + P^{(L)} + P^{(F)} = 1 \quad (42)$$

For the special case of one fissile isotope (type I) only, the overall balance equation becomes:

$$1 + F_I(\mu_I - 1) = M(1 - R) + A \quad (43)$$

which gives:

$$(\mu_I - 1)F_I = M - 1 + A - MR \quad (44)$$

The correlation function  $g_2$  is then expressed as:

$$g_2(t_1, t_2) = \sum_i Q_i \left( \mu_I^{(2)} + \mu_I^{(1)}(M - 1 + A - MR) \frac{\mu_I^{(2)}}{\mu_I - 1} M^2 \epsilon^2 \right) T(t_1, t_2) \quad (45)$$

If we put:

$$H(t|s) = \lambda e^{-\lambda(t-s)} \quad (46)$$

we obtain ( $\tau = t_2 - t_1$ ):

$$T(t_1, t_2) = T(0, \tau) = \frac{\lambda}{2} e^{-\lambda\tau} \quad (47)$$

The decay constant  $\tau$  introduced above refers to the complete system of probe plus detector and, in case it is measured, has to be measured with the probe in place. If, on the other hand, the decay constant  $\lambda_D$  of the detector only is measured, then one has the relation:

$$\lambda = \lambda_D(1 - M^{(0)}R^{(D)}) \quad (48)$$

## Appendix

### CORRELATION FUNCTION $g_3(t_1, t_2, t_3)$ [2]

For the case of using third moments in randomly triggered intervals or second moments in pulse triggered intervals, we need the third order correlation function  $g_3$ . We present the explicit expression for  $g_3$  for further study and show what approximations are involved to arrive at the one-point, one-group model.

According to the physical meaning of the correlation functions we may write the explicit expression for  $g_3$ :

$$g_3(t_1, t_2, t_3) = \sum_{\mathbf{K}} \int dx Q_{\mathbf{K}}(x) [\mu_{\mathbf{K}}^{(3)} T_{\mathbf{K}}(t_1, t_2, t_3|x) + \mu_{\mathbf{K}}^{(2)} (T_{\mathbf{K}}(t_2, t_3|x t_1) + T_{\mathbf{K}}(t_1, t_3|x t_2) + T_{\mathbf{K}}(t_1, t_2|x t_3))] \quad (49)$$

where we have introduced:

$$T_{\mathbf{K}}(bc|ax) = \int_{-\infty}^{+\infty} ds D_{\mathbf{K}}(a|xs) \int dx' \int ds' \sum_{\beta} F_{\mathbf{K}\beta}(xs \rightarrow x's') \mu_{\beta}^{(2)} \times D_{\beta}(b|x's') D_{\beta}(c|x's') \quad (50)$$

and  $F_{\mathbf{K}\beta}(xs \rightarrow x's') dx' ds'$  is the probability for a  $\beta$ -type fission in  $dx' ds'$  due to one  $\mathbf{K}$ -fission neutron starting at  $\{x, s\}$ .

Using the assumption

$$D_{\mathbf{K}}(a|xs) = D_{\mathbf{K}}(x) H(a|s)$$

made above and the further condition

$$F_{\mathbf{K}\beta}(xs \rightarrow x's') = F_{\mathbf{K}\beta}(x \rightarrow x') \delta(s' - s) \quad (51)$$

gives:

$$g_3(t_1, t_2, t_3) = \sum_{\mathbf{K}} \int dx Q_{\mathbf{K}}(x) \left( \mu_{\mathbf{K}}^{(3)} D_{\mathbf{K}}^{(3)}(x) + 3\mu_{\mathbf{K}}^{(2)} D_{\mathbf{K}}(x) \times \sum_{\beta} \int dx' F_{\mathbf{K}\beta}(x \rightarrow x') \mu_{\beta}^{(2)} D_{\beta}^2(x') \right) T(t_1, t_2, t_3) \quad (52)$$

Space independence of  $D_{\mathbf{K}}(x)$  and  $F_{\mathbf{K}\beta}(x \rightarrow x')$  eventually gives:

$$g(t_1, t_2, t_3) = \sum_{\mathbf{K}} Q_{\mathbf{K}} \left( \mu_{\mathbf{K}}^{(3)} D_{\mathbf{K}}^3 + 3\mu_{\mathbf{K}}^{(2)} D_{\mathbf{K}} \sum_{\beta} F_{\mathbf{K}\beta} \mu_{\beta}^{(2)} D_{\beta}^2 \right) T(t_1, t_2, t_3) \quad (53)$$

For further evaluation we have again to separate out the actual source (spontaneous) fissions ( $K = i$ ) and the induced fissions through

$$Q_\beta = \sum_i \mu_i^{(1)} Q_i F_{i\beta} \quad (54)$$

to obtain:

$$\begin{aligned} g(t_1, t_2, t_3) = \sum_i Q_i \left[ \mu_i^{(3)} D_i^3 + 3\mu_i^{(2)} D_i \sum_\beta F_{i\beta} \mu_\beta^{(2)} D_\beta^2 + \mu_i^{(1)} \sum_\alpha F_{i\alpha} \right. \\ \left. \times \left( \mu_\alpha^{(3)} D_\alpha^3 + 3\mu_\alpha^{(2)} D_\alpha \sum_\beta F_{\alpha\beta} \mu_\beta^{(2)} D_\beta^2 \right) \right] T(t_1, t_2, t_3) \end{aligned} \quad (55)$$

Here we may use the sample-detector separation described above.

Taking one energy group only, we obtain:

$$\begin{aligned} g(t_1, t_2, t_3) = \sum_i Q_i \left[ \mu_i^{(3)} + 3\mu_i^{(2)} \sum_\beta F_\beta \mu_\beta^{(2)} + \mu_i^{(1)} \sum_\alpha F_\alpha \right. \\ \left. \times \left( \mu_\alpha^{(3)} + 3\mu_\alpha^{(2)} \sum_\beta F_\beta \mu_\beta^{(2)} \right) \right] D^3 T(t_1, t_2, t_3) \end{aligned} \quad (56)$$

which is the expression used in the one-point, one-group superfission model.

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# IMPROVEMENT OF ISOTOPIC COMPOSITION MEASUREMENTS OF PLUTONIUM BY THE USE OF CALIBRATION SAMPLES FOR GAMMA SPECTROSCOPY

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## Abstract

IMPROVEMENT OF ISOTOPIC COMPOSITION MEASUREMENTS OF PLUTONIUM BY THE USE OF CALIBRATION SAMPLES FOR GAMMA SPECTROSCOPY.

The PuO<sub>2</sub> pilot reference samples from the Central Bureau for Nuclear Measurements at Geel, with isotopic compositions corresponding to different burnup conditions (93, 84, 70 and 61% <sup>239</sup>Pu), are almost infinitely thick (>99.9%) for gamma rays up to 165 keV. They thus allow the application of the enrichment meter technique for the determination of the isotope abundances by comparing the gamma peak intensities of the reference samples with those of sufficiently thick plutonium samples of unknown isotopic composition but with specified cladding conditions. The same reference samples have also shown to be helpful for complementary calibrations when high resolution gamma ray spectroscopy is used in conjunction with the intrinsic calibration technique to obtain isotope ratios. These complementary calibrations concern: the testing and correction of the procedures for the determination of the overall relative efficiency curve for the lower energy gamma rays; and the derivation of correction factors for the calculation of isotope ratios from characteristic (preferentially close lying) gamma lines. In this way accuracies of less than 1 % are obtained for the determination of Pu isotope ratios in Pu samples characterized by different burnup and cladding conditions.

## 1. INTRODUCTION

The determination of the isotopic composition of Pu (including <sup>241</sup>Am) by the intrinsic calibration method using high resolution gamma spectrometry is based on the following equation:

$$X_a = A/\lambda \epsilon_a B_a$$

where

- $X_a$  is the number of nuclei of the isotope considered,
- $\lambda$  is the nuclear transition probability,
- $B_a$  is the absolute branching ratio for the characteristic gamma ray considered,
- $A$  is the peak area of the gamma ray,
- $\epsilon_a$  is the absolute overall detection efficiency for the gamma ray considered.

When relative efficiencies  $\epsilon_r$  and/or relative branching ratios  $B_r$  are used, relative quantities  $X_r$  are obtained from

$$X_r = A / \lambda \epsilon_r B_r$$

The final accuracy of  $X_r$  depends not only on the uncertainties of the nuclear parameters  $\lambda$  and  $B_r$ , but also on experiment dependent parameters, such as the relative overall efficiency curve and the selected peak evaluation method. The uncertainties of all these parameters are difficult to evaluate. For given experimental conditions their effect can be reduced by complementary calibrations using reference samples whose isotopic composition is precisely known [1]. By the measurement of such reference samples correction factors  $F = R(\text{MS})/R(\gamma)$  can be determined, where  $R(\text{MS})$  is the isotope ratio obtained by mass spectrometry and a corresponding  $^{241}\text{Am}$  determination and  $R(\gamma)$  is the isotope ratio obtained from quantities  $X_r$  derived from gamma spectrometry. For calibrations the now available pilot reference samples of the Central Bureau for Nuclear Measurements (CBNM) at Geel were used (Appendix I).

These samples are infinitely thick for isotope characteristic gamma lines up to 165 keV and are thus also suited for the application of the enrichment meter technique [2]. Three of the reference samples with isotopic compositions corresponding to different burnups (61, 70 and 84%  $^{239}\text{Pu}$ ) were used for corrections. The correction factors were then applied for correcting the isotope ratios of samples from identical materials, but measured under experimental conditions different from those for the calibration measurements. The same correction factors were also applied for correcting isotope ratios determined for samples measured under conditions similar to those used for the calibration measurements with the reference samples, but made of different materials. The application of complementary calibrations for correcting systematic errors depends strongly on the good reproducibility of experimental conditions and, in particular, on the gamma peak evaluation methods and the determination of relative overall efficiency curves. Special attention was thus devoted also to these two items. In particular, different peak evaluation programs were compared (Section 2) and some attention was devoted to the determination of sample and experiment specific overall efficiency curves (Section 3).



TABLE I. RATIOS OF PEAK AREAS DETERMINED BY GSFT AND SMOOTH FOR MATERIALS WITH DIFFERENT BURNUPS

Gamma line energy (keV)	Burnup (% Pu-239)		
	61	70	84
125	1.003 96	1.006 42	1.015 32
129	1.000 03	1.000 09	1.004 53
148	1.000 99	1.001 00	1.003 29
152	1.001 76	1.001 74	1.002 47
160	1.006 50	1.005 39	0.994 88
164	1.004 70	1.004 31	1.008 21
203	0.997 99	1.002 16	1.009 27
208	0.997 26	0.997 29	1.000 80
265	1.005 24	1.005 77	1.009 14
332	1.002 39	1.002 61	1.002 71
336	1.024 94	1.033 28	1.029 81
345	1.004 53	1.004 89	1.004 48
413	0.972 17	0.982 04	0.995 46

## 2. PEAK EVALUATION

Different programs have been tested and compared for peak area integration. They belong to two groups of procedures:

- (i) *Channel by channel summation.* Two programs called PLANI and SMOOTH perform channel by channel summation but differ in the determination of the background, which is obtained either by a step-like function [3, 4] or by successive smoothing.
- (ii) *Peak fitting programs.* Besides SAMPO [5], a peak fitting program called GSFT [6] is used. Both approximate the gamma peak by an asymmetric Gaussian superimposed on a linear background. GSFT works in an interactive way and proved to be very useful especially for the fitting of complex multiplets.

Equivalent results were obtained for GSFT and SMOOTH for most of the lines (Table I), but as fitting programs are needed for the analysis of complex lines, GSFT was used for all further results discussed in the present work.

TABLE II. EXAMPLES OF EFFICIENCY RATIOS DETERMINED FOR GAMMA LINE RATIOS CORRESPONDING TO DIFFERENT ISOTOPE RATIOS

Isotope ratio	Gamma ratio (keV)	Efficiency ratio <sup>a</sup>
Pu-239/Pu-241	129.29/148.57	0.608
Pu-238/Pu-241	152.68/148.57	1.086
Pu-240/Pu-241	160.28/148.57	1.244
Pu-240/Pu-241	160.28/164.58	0.931
Pu-241/Pu-239	208.00/203.54	1.055
	332/335	0.999 92
	332/345	1.000 69
	335/345	1.000 77

<sup>a</sup> Experimental values for thick samples and a high efficiency gamma detector.

### 3. RELATIVE EFFICIENCY CURVE AND ISOTOPE RATIOS

Supposing that all nuclear parameters entering the fundamental equation of Section 1 are known, we can solve it if the relative efficiency  $\epsilon_r$  is known. As all the most used gamma rays lie in the 100–415 keV region, a strong variation of self-absorption with energy will be observed and no previously established efficiency curve can be used. Instead an overall sample and experiment specific efficiency curve taking into account absorption, self-absorption, detector efficiency and geometry has to be established. In order to minimize errors introduced by the influence of the experimental uncertainties on the slope of the efficiency curve, one generally uses characteristic gamma rays close in energy for the determination of isotope ratios. In Table II some gamma line ratios often used for the determination of isotope ratios [7] are given, as well as typical examples of the corresponding efficiency ratios measured for a high efficiency Ge spectrometer and for relatively thick samples. Strong variations are observed at lower energies while the relative efficiencies between 332 and 345 keV are almost constant. The program PLUTO [8] used takes advantage of this fact, as shown in Appendix II.

As the three different reference samples (61, 70 and 84% <sup>239</sup>Pu) are all infinitely thick up to 165 keV, equal efficiencies are expected in any case in the low energy region but to a large extent also at higher energies because samples are very similar in thickness. Almost equal efficiency values were obtained with an accuracy of less than 1% for the higher burnup sample materials, while higher deviations are observed for the low burnup material used.

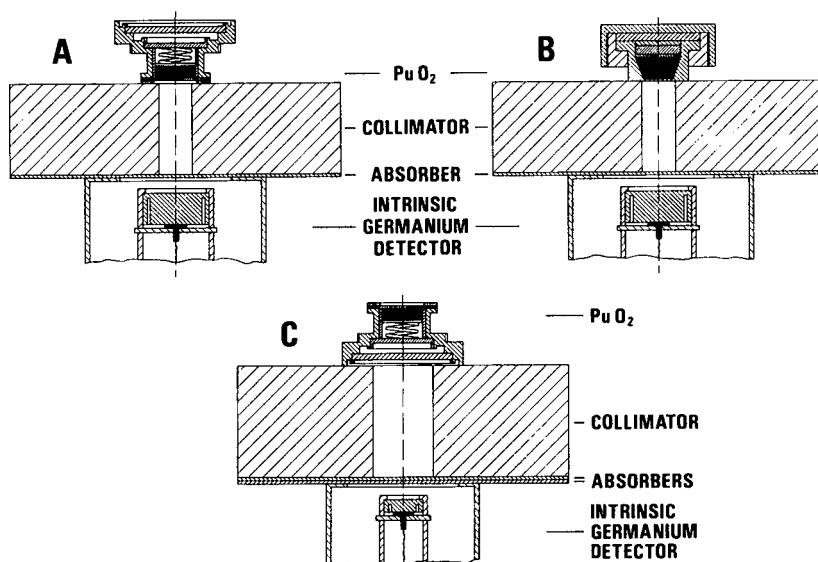


FIG. 1. Experimental set-ups.

## 4. MEASUREMENTS

Figure 1 and Table III give an overview of the different measurements performed. The correction factors  $R(\text{MS})/R(\gamma)$  for the isotope ratios  $^{238}\text{Pu}/^{241}\text{Pu}$ ,  $^{239}\text{Pu}/^{241}\text{Pu}$ ,  $^{240}\text{Pu}/^{241}\text{Pu}$  and  $^{241}\text{Am}/^{239}\text{Pu}$  were obtained from runs QA6103N, QA7003N and QA8403N. For data evaluation the Gunnink branching ratios [9] were used. In order to minimize statistical uncertainties for the determination of the gamma line emission rates long counting times were used with correspondingly large total count numbers ( $> 10^8$ ).

## 5. RESULTS

### 5.1. Correction factors

The experimentally obtained correction factors were similar for the reference samples from high burnup material (i.e. the 61 and 70% samples) within sub-1% limits (except Am), but more different for those values obtained with the help of the reference sample from low burnup material. The correction factors are given in Table IV.

TABLE III. DETAILS OF MEASUREMENTS PERFORMED WITH EXPERIMENTAL SET-UPS SHOWN IN FIG. 1

Code	Sample material (% Pu-239)	Detector	Collimator	Absorber	Total input counts (10 <sup>7</sup> )	Fig. 1
QA6103N <sup>a</sup>	61				102	A
QA7003N <sup>a</sup>	70	PGT <sup>b</sup>	Pb	Cd	69	A
QA8403N <sup>a</sup>	84	Ø25/13	Ø11/30	1.5 mm	63	A
QA0056 <sup>c</sup>	56	(mm)	(mm)		56	B
QA0078 <sup>c</sup>	78				55	B
PF6104	61	Ortec	Pb	Cd and Cu		C
PF7004	70	Ø16/5	Ø20/37	1 mm	> 10	C
PF8404	84	(mm)	(mm)	each		C

<sup>a</sup> Calibration measurements.<sup>b</sup> Princeton Gammatec.<sup>c</sup> Experimental conditions similar to those used for the reference samples.

TABLE IV. EXPERIMENTAL CORRECTION FACTORS FOR DETERMINATION OF ISOTOPE RATIOS

Isotope ratio	Correction factor			
	61 %	70 %	Mean <sup>a</sup>	84 % <sup>b</sup>
238/241	0.969	0.971	0.970	0.969
239/241	0.994	0.996	0.995	1.015
240/241	0.883	0.876	0.880	0.997
Am/241	0.944	0.971	0.958	0.896

<sup>a</sup> Mean values used for high burnup material.<sup>b</sup> For low burnup material.

TABLE V. ISOTOPIC COMPOSITION OF REFERENCE SAMPLE MATERIALS MEASURED WITH AN EXPERIMENTAL SET-UP DIFFERENT FROM THAT FOR THE CALIBRATION MEASUREMENTS: RATIO OF GAMMA TO MS RESULTS

Isotope ratio or isotope	PF6104		PF7004		PF8404	
	A	B	A	B	A	B
238/241	0.965	0.995	0.965	0.995	0.961	0.992
239/241	0.987	0.992	0.988	0.993	1.009	0.994
240/241	0.896	1.018	0.872	0.991	0.983	0.986
Am/239	0.926	0.966	0.914	0.954	0.845	0.943
Pu-238	1.002	0.996	0.999	1.002	0.956	0.999
Pu-239	1.025	0.993	1.023	1.001	1.004	1.001
Pu-240	0.930	1.019	0.903	0.998	0.983	0.978
Pu-241	1.038	1.001	1.035	1.007	0.995	1.007
Am-241	0.949	0.960	0.934	0.954	0.849	0.945

Note: The correction factors were taken from Table IV (mean for PF6104 and PF7004; 84% for PF8404).

A: Uncorrected values.

B: Corrected values.

## 5.2. Analysis of unknown samples

Runs PF6104, PF7004 and PF8404 were performed with different samples made of the same Pu material as the reference samples. A different experimental set-up was used. Individual isotope ratios were corrected with the help of the correction factors given in Table IV. (The mean and 84% values were used for the evaluation of high (PF6104, PF7004) and low (PF8404) burnup sample measurements, respectively.) The results are summarized in Table V, which gives in columns A and B the ' $\gamma$ /MS' ratios before and after application of the correction factors, respectively. Both isotope ratios and isotopic composition values are compared. Similarly, Table VI summarizes the results obtained for runs QA0056 and QA0078 using different materials measured under experimental conditions similar to those used for the calibration runs. As different PuO<sub>2</sub> materials were used, the corresponding MS values and their errors are also indicated.

TABLE VI. ISOTOPIC COMPOSITION OF HIGH AND LOW BURNUP PuO<sub>2</sub>: RATIO OF GAMMA TO MS RESULTS

Isotope ratio or isotope	MS value (%)	Uncertainty (2 $\sigma$ )	Gamma/MS	
			A	B
238/241	0.189		0.967	0.997
239/241	7.200		0.992	0.997
240/241	3.144		0.874	0.993
Am/239	0.067		0.960	1.002
Pu-238	1.484	0.20	1.007	1.001
Pu-239	56.625	0.16	1.034	1.001
Pu-240	24.724	0.20	0.910	0.997
Pu-241	7.864	0.20	1.042	1.004
Pu-242	5.514	0.20	—	—
Am-241	3.790	< 10	0.992	1.002
238/241	0.086		0.952	0.982
239/241	55.400		1.011	0.996
240/241	13.190		0.959	1.012
Am/239	0.013		0.920	1.027
Pu-238	0.122	2.00	0.952	0.983
Pu-239	78.187	0.06	1.011	0.997
Pu-240	18.619	0.21	0.959	1.013
Pu-241	0.411	0.20	1.000	1.001
Pu-242	0.642	0.20	—	—
Am-241	1.020	< 10	0.930	1.024

**Note:** The correction factors were taken from Table IV (mean for high burnup and 84% for low burnup material).

A: Uncorrected values.

B: Corrected values.

## 6. CONCLUSIONS

The applied reference samples made of different burnup materials were used to obtain two sets of correction factors (one for high and one for low burnup materials) in order to correct isotope ratios obtained by high resolution gamma ray spectroscopy. Under the same experimental conditions as those used for the calibration runs, these factors were applied with success to samples made of different

materials, as shown in Table VI. When these factors were applied to samples made of the same material but measured under experimental conditions different from those used for the calibration runs performed with the reference samples the results were slightly less good (Table V). However, in both cases the use of the correction factors led to a net increase of the accuracy of the isotope ratios determined, showing that besides instrumental problems, such as high count rate effects, other systematic errors depending on the accuracy of different parameters and of data evaluation methods, etc., are important and can be corrected by the use of reference samples.

Note also the greater difficulties encountered for low burnup samples, as illustrated by the need for a different set of correction factors and the observation of a different relative overall efficiency curve in spite of the fact that all reference samples are very similar in thickness and infinitely thick for lower energy gamma rays. This might be due to the fact that the low burnup material is poor in  $^{241}\text{Pu}$  and  $^{241}\text{Am}$ , and that consequently the analysis of the 332–345 keV quintuplet is more delicate.

The use of more recently published branching ratios [10–12] resulted in smaller correction factors and has shown that the application of calibration samples allows one to reduce the problem of incorrect nuclear data for the application of the intrinsic calibration method. One can therefore suppose that other systematic errors introduced, for instance, by less sophisticated peak integration methods could also be corrected by complementary calibrations using reference samples. Further investigations are planned also in connection with the Plutonium Isotopic Determination Inter-laboratory Exercise of the NDA working group of the European Safeguards Research and Development Association.

## Appendix I

### **$\text{PuO}_2$ PILOT REFERENCE SAMPLES FOR ISOTOPIC COMPOSITION MEASUREMENTS BY GAMMA SPECTROMETRY: CBNM SPECIFICATIONS**

#### *General*

Sets of four samples consisting of sintered  $\text{PuO}_2$  pellets in steel containers, of the following material types (%  $^{239}\text{Pu}$ ): 93, 84, 70, 61.

#### *Sintered $\text{PuO}_2$ pellets*

Areal density ( $\text{g cm}^{-2}$ )	$3.90 \pm 0.10$
Diameter (mm)	$14.75 \pm 0.17$
Flatness	$< \pm 0.02$
Density ( $\text{g cm}^{-3}$ )	$10.4 \pm 0.6$
Thickness (mm)	$3.75 \pm 0.12$
Mass (g)	$6.65 \pm 0.05$
Stoichiometry ( $\text{PuO}_{2-x}$ ) <sub>x</sub>	$< 0.05$

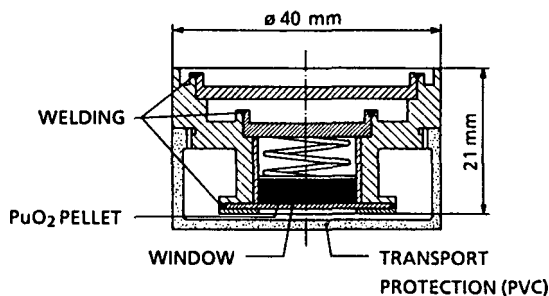


FIG. 2. Cross-section of pilot reference sample.

TABLE VII. ISOTOPIC COMPOSITION OF SAMPLE MATERIALS  
*(Pu(iso)/(Pu(tot) + Am) (at. %), preliminary values<sup>a</sup> valid for 20 June 1986)*

Sample material	Pu-238	Pu-239	Pu-240	Pu-241	Pu-242	Am-241
93	0.012	93.338	6.286	0.2212	0.039	0.1038
84	0.0706	84.214	14.130	1.0184	0.3526	0.2152
70	0.8420	72.581	18.039	5.3576	2.0307	1.1492
61	1.1889	61.771	24.992	6.5437	4.0880	1.4161

<sup>a</sup> Final specifications will include uncertainties of abundance values for a confidence level of about 95%.

TABLE VIII. CHEMICAL PURITY OF SAMPLE MATERIALS  
*(maximum total impurities without Am-241)*

Impurities ( $\mu\text{g/g}$ )	Sample material			
	93	84	70	61
For all elements, $Z \leq 30$	170	500	500	400
For low Z elements (B, Be, Li, F, Mg, Na) <sup>a</sup>	10	20	10	30
For all elements, $Z > 30$	30	800 (20) <sup>b</sup>	600 (250) <sup>b</sup>	1200 (380) <sup>b</sup>

<sup>a</sup> Elements with high  $\alpha/n$  cross-section.

<sup>b</sup> Included contributions of ingrown U-234 from the decay of Pu-238 (June 1986).



*Canning*

Container (Fig. 2) certified by the Bundesanstalt für Materialprüfung, Federal Republic of Germany (approval D/0039/S) as type A for special forms of radioactive material following IAEA specifications.

Material: stainless steel

Window thickness (mm):  $0.780 \pm 0.002$

**Appendix II****DETERMINATION OF OVERALL EFFICIENCY CURVE  
USING THE PROGRAM PLUTO**

If we use the general equation  $X_j B_{i,j} \lambda_j = A_i / \epsilon_i$ , where  $X_j$  ( $j = 1, 2, \dots, 5$ ) are the relative quantities of  $^{238}\text{Pu}$ ,  $^{239}\text{Pu}$ ,  $^{240}\text{Pu}$ ,  $^{241}\text{Pu}$  and  $^{241}\text{Am}$  and  $i=1, 2, \dots, 13$  correspond to the energies of the gamma lines measured, the following system of 13 equations and five unknowns can be written:

$^{238}\text{Pu}$	$^{239}\text{Pu}$	$^{240}\text{Pu}$	$^{241}\text{Pu}$	$^{241}\text{Am}$	
	$X_2 B_{1,2} \lambda_2$			$+ X_5 B_{1,5} \lambda_5$	$= A_{125} / \epsilon_{125}$ (1)
	$X_2 B_{2,2} \lambda_2$				$= A_{129} / \epsilon_{129}$ (2)
			$X_4 B_{3,4} \lambda_4$		$= A_{148} / \epsilon_{148}$ (3)
$X_1 B_{4,1} \lambda_1$					$= A_{153} / \epsilon_{153}$ (4)
	$X_2 B_{5,2} \lambda_2$	$+ X_3 B_{5,3} \lambda_3$	$+ X_4 B_{5,4} \lambda_4$		$= A_{160} / \epsilon_{160}$ (5)
			$X_4 B_{6,4} \lambda_4$	$+ X_5 B_{6,5} \lambda_5$	$= A_{164} / \epsilon_{164}$ (6)
	$X_2 B_{7,2} \lambda_2$			$+ X_5 B_{7,5} \lambda_5$	$= A_{203} / \epsilon_{203}$ (7)
			$X_4 B_{8,4} \lambda_4$	$+ X_5 B_{8,5} \lambda_5$	$= A_{208} / \epsilon_{208}$ (8)
			$X_4 B_{9,4} \lambda_4$	$+ X_5 B_{9,5} \lambda_5$	$= A_{263} / \epsilon_{263}$ (9)
	$X_2 B_{10,2} \lambda_2$		$+ X_4 B_{10,4} \lambda_4$	$+ X_5 B_{10,5} \lambda_5$	$= A_{332} / \epsilon_{332}$ (10)
	$X_2 B_{11,2} \lambda_2$		$+ X_4 B_{11,4} \lambda_4$	$+ X_5 B_{11,5} \lambda_5$	$= A_{335} / \epsilon_{335}$ (11)
	$X_2 B_{12,2} \lambda_2$				$= A_{345} / \epsilon_{345}$ (12)
	$X_2 B_{13,2} \lambda_2$				$= A_{415} / \epsilon_{415}$ (13)

$\lambda_1, \dots, \lambda_5$  represent the nuclear transition probabilities for the isotopes concerned and the  $B_{i,j}$  the gamma branching ratios; the  $A_i$  represent the peak areas and the  $\epsilon_i$  the corresponding relative overall efficiencies. As  $\epsilon_{332} \approx \epsilon_{335} \approx \epsilon_{345}$  preliminary efficiency values may be used. Equations (10–12) form then a system of three equations and three unknowns in  $X_2$ ,  $X_4$  and  $X_5$ . The solution may be introduced into Eqs (1–3) and (6–13) in order to calculate the corresponding relative efficiencies, through which a polynomial can be fitted.

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## A NEW TOOL FOR POST-EVALUATION OF PLUTONIUM GAMMA RAY SPECTRA

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### Abstract

#### A NEW TOOL FOR POST-EVALUATION OF PLUTONIUM GAMMA RAY SPECTRA.

A powerful means for evaluation of plutonium isotopic abundances determined by high resolution gamma spectroscopy (HRGS) has been realized on a transportable DEC PRO-380 computer. All functions required for the evaluation of the abundances up to the generation of NDA worksheets were integrated and linked together in a consistent way. So far, this tool has been used to process more than 500 spectra. The advantages that have become obvious to all users are: ease of use, high speed, small risk of transcription errors and minimum routine data handling. It was also recognized that the concept could be very useful for the inspector in the field. The paper briefly describes the authors' efforts to set up several functions on the computer to help in some HRGS related tasks at IAEA Headquarters.

### 1. INTRODUCTION

In its independent verification activities in plutonium facilities the International Atomic Energy Agency is routinely applying high resolution gamma spectroscopy (HRGS) to determine plutonium isotopic abundances as well as  $^{241}\text{Am}$  and  $^{235}\text{U}$  concentrations. These measurements complement high level neutron coincidence counting or calorimetry for Pu assay.

Staff in various support divisions of the Department of Safeguards have to provide the inspector with proper tools for applying these techniques. This means that staff at Agency Headquarters have not only to monitor instrument performance but also to study the data coming from the field. Obviously, they need proper tools for their work. However, all that is usually available is only what the inspector is currently using in the field; this applies to both hardware and software.

For any more demanding HRGS related HQ tasks this is simply not adequate. The field tools were not designed for HQ work and lack the necessary data handling support. Trying to do some HQ tasks with the field equipment is unrealistic because of the time and manpower required. It is therefore necessary to have on HQ computers more powerful counterparts of all functions now provided for the field.

HQ computers can provide the equivalent of all functions available in the field and much more. Except for the actual measurement function, all others, such as raw data reduction, estimation of measurement errors and report generation, can be done much more efficiently utilizing the computing power available at HQ.

This paper briefly describes our efforts to implement and expand HRGS field functions on a DEC PRO-380 computer to help us in some of our HRGS related HQ tasks (see Appendix).

## 2. PROTOTYPE HRGS TOOL

### 2.1. The need for the tool

We had the following HRGS related tasks:

- (a) To evaluate the performance of the various implementations of the algorithm developed at the Lawrence Livermore National Laboratory (LLNL) [1], especially with regard to mixed oxide or high burnup samples; and to identify differences from the output when using the Plutonium Isotopic Analysis Unit (PIAU, or 'Blue Box').
- (b) Preliminary work for the NDA test database. This meant storing Blue Box output and keyboard entered operators' data on disk files for subsequent loading into a database.
- (c) To investigate the suitability of a computer generated NDA worksheet for attachment to the inspection report.

We realized that with only the current field equipment we would have serious problems. For instance, working with a multichannel analyser and the Blue Box [2] is too cumbersome if many spectra are to be processed. We also do not have the manpower for all the manual data handling involved.

All three tasks would have required enhancements to the field capabilities for satisfying at least some data handling requirements. This was impossible to do on the field equipment. The alternative was to create the equivalent on a fairly powerful HQ computer and then add what we thought to be necessary for our work.

Finally, producing a computer generated worksheet for the inspection report would only have made sense if the various steps leading to it were also computer supported and easily done. For this, it was necessary not only to move various field functions on the HQ computer but also to provide some integration to achieve ease of use.

## 2.2. Functions wanted

For our purposes the following features were necessary:

- Easy loading of the raw data from Cicero or portable multichannel analyser (PMCA) cassettes into computer files;
- Unique labelling of each cassette and the individual spectra on it, because such information is not in the raw data themselves;
- Graphic display of a spectrum and those functions that are needed if the isotopic abundances and ratios are to be computed;
- The LLNL algorithms with improved messages and earlier error notification;
- Computed results stored for later retrieval, and not just a printout;
- Easy keyboard entry and storage of the operator declared values;
- Security for confidential data;
- Decay correction for the declared isotopic abundances;
- Statistical tests for operator-inspector comparisons;
- Various NDA worksheets.

In addition, we were committed to two long term requirements for NDA activity in general:

- (1) The need to standardize the evaluation procedures used for verifying Pu isotopic abundances and ratios both in the field and at HQ. This implies the capability to easily test algorithms provided by support programmes against actual Agency data and provide quick feedback. This is essential for the developer if the Agency asks him to make improvements before finalizing the code.
- (2) Preparing the way for an NDA database at HQ. This also implies storing all important data elements in a manner that will make it easy to transfer this information to other systems.

## 3. DESIGN CONSIDERATIONS

Because of limited time and manpower, our design had to stress modularity. The major tool functions had to be so self-contained that each could be completed and used independently for work on our main tasks. These tasks therefore dictated in which order individual functions had to be implemented.

Since this was to be a prototype, neither the choice of functions nor the options to be provided in each could be considered final. Therefore, the design had to allow for relatively easy addition, removal or modification of functions without requiring major rewrites. For this, common data structures had to be isolated from the main code making up any one function, and a flexible way of interfacing between the individual functions had to be found.

We also wanted to make all routine file handling operations transparent to the user, including those for passing data between the functions listed in Section 2.2. Besides helping the user, this allows tighter software control, especially for ensuring that there is consistency between the function the user selects and the files on his diskettes.

Another major concern was how to keep track of and correctly correlate the various items of data that the computer will be helping to accumulate. We wanted the software to manage this efficiently.

Finally, all functions had to be easily accessible via menus and the need to refer to manuals minimized.

#### 4. IMPLEMENTATION

As mentioned earlier, software development had to proceed in discrete steps. The order in which individual functions were completed depended on how urgently these were needed for progressing with our main tasks.

We now have a first version of the full tool, in which all functions have been completed (Fig. 1). We will briefly comment on each function. More details will be made available in Ref. [3].

##### 4.1. Loading of raw data

By use of menu selectable options raw spectrum data can be transferred from Cicero or PMCA cassettes [4,5] to computer files. The 'Greyhound' cassette reader designed by the Joint Research Centre at Ispra and implemented by Silena is being used to read Cicero cassettes. For PMCA cassettes, a PMCA itself is used as the reading device. This function is designed to load spectra into files created on the floppy disks, which can then be locked away if there is need for confidentiality. Nothing remains on the computer.

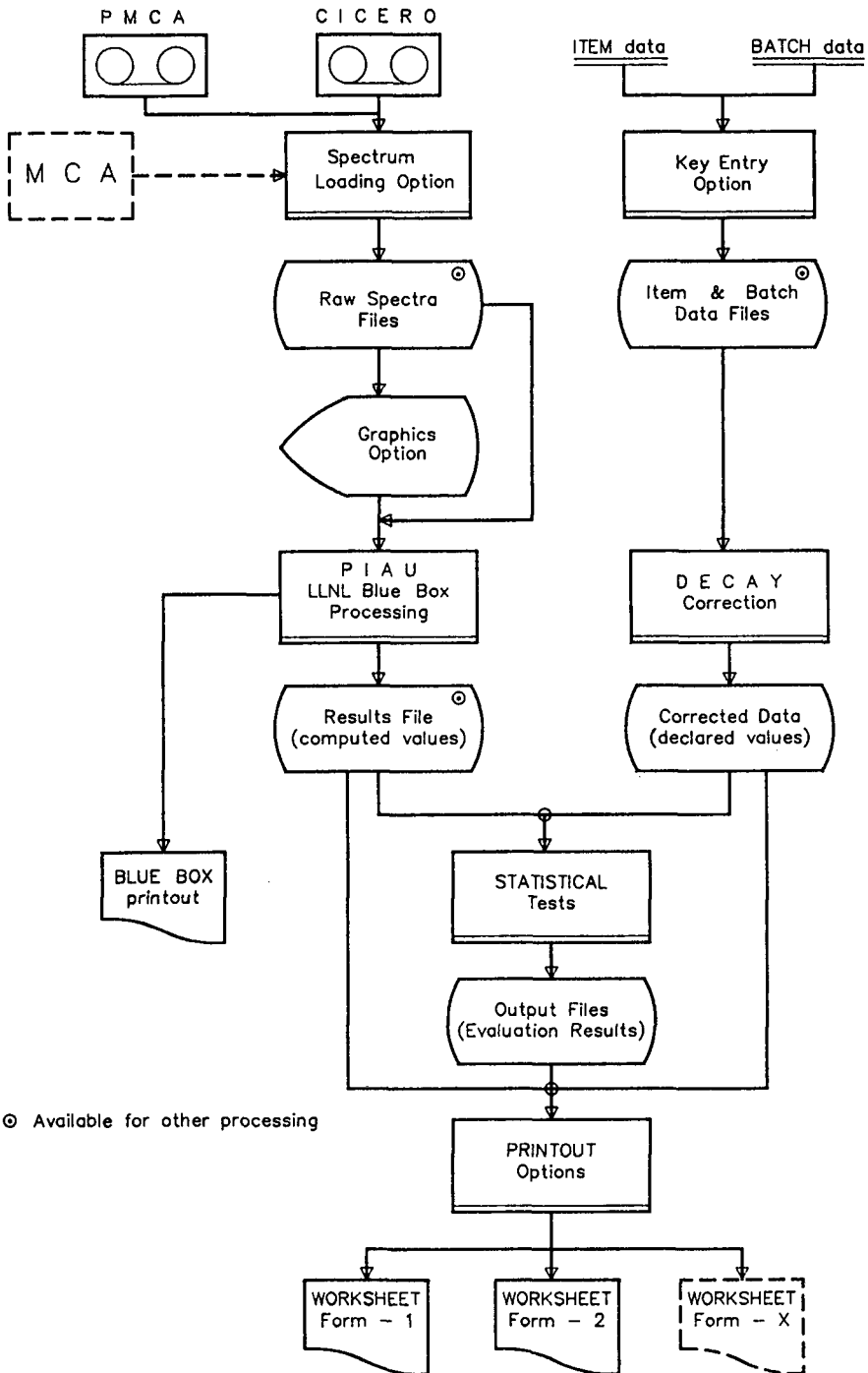


FIG. 1. Functional overview.

#### 4.2. Labelling of each cassette and individual spectra on it

Each cassette is considered to represent a 'series'. Every spectrum on the cassette is an 'item' in this series and the file containing it will have a name of the form SERIESNAM.SID, where SERIESNAM represents a nine-character series name and 'SID' is the three-character spectrum identification.

The contents of every cassette processed with this tool are mapped on to two diskettes (2 x 400 kbyte), the so-called series diskettes. When loading the first item of a series from a cassette on to a diskette, the user will be prompted for a unique series name that will then be automatically used for all the other items on that cassette. This series name is also used for all other files that are being used to store data relating to this series. The two diskettes therefore represent a compact document of all data accumulated for a series at any point of time.

Any time the user wishes to access a function, he is asked to insert the series diskettes. The software checks for consistency of the file names and does not permit further work if something is wrong.

#### 4.3. Graphics display

We have provided a choice of linear or logarithmic display. Up to 4000 channels can be currently handled. Special features for finding and selecting the peaks needed for an energy calibration have been incorporated to make this task easy. There is also a provision for viewing an expanded section of the spectrum.

The graphics functions also serve as a convenient interface to the LLNL (Blue Box) function. After selection of the necessary peaks the Blue Box processing is invoked by the pressing of a key. All LLNL error messages come back immediately to this graphics display. Reselecting the peaks and trying again in response to error messages can be done extremely fast. The spectrum data, once loaded, are retained in memory until the user decides either to quit the session or to move to another spectrum.

On-line help is available.

#### 4.4. LLNL (Blue Box) algorithms

This was the most important function to be implemented. An earlier stand-alone version was completed for test purposes



Series name: TOMC2224    Spectrum ID: 200    Field media ID: CASSETTE1986B

ITEM	DATA	ENTRY
1	100	100
2	200	200
3	300	300
4	400	400
5	500	500
6	600	600
7	700	700
8	800	800
9	900	900
10	1000	1000
11	1100	1100
12	1200	1200
13	1300	1300
14	1400	1400
15	1500	1500
16	1600	1600
17	1700	1700
18	1800	1800
19	1900	1900
20	2000	2000
21	2100	2100
22	2200	2200
23	2300	2300
24	2400	2400
25	2500	2500
26	2600	2600
27	2700	2700
28	2800	2800
29	2900	2900
30	3000	3000
31	3100	3100
32	3200	3200
33	3300	3300
34	3400	3400
35	3500	3500
36	3600	3600
37	3700	3700
38	3800	3800
39	3900	3900
40	4000	4000
41	4100	4100
42	4200	4200
43	4300	4300
44	4400	4400
45	4500	4500
46	4600	4600
47	4700	4700
48	4800	4800
49	4900	4900
50	5000	5000
51	5100	5100
52	5200	5200
53	5300	5300
54	5400	5400
55	5500	5500
56	5600	5600
57	5700	5700
58	5800	5800
59	5900	5900
60	6000	6000
61	6100	6100
62	6200	6200
63	6300	6300
64	6400	6400
65	6500	6500
66	6600	6600
67	6700	6700
68	6800	6800
69	6900	6900
70	7000	7000
71	7100	7100
72	7200	7200
73	7300	7300
74	7400	7400
75	7500	7500
76	7600	7600
77	7700	7700
78	7800	7800
79	7900	7900
80	8000	8000
81	8100	8100
82	8200	8200
83	8300	8300
84	8400	8400
85	8500	8500
86	8600	8600
87	8700	8700
88	8800	8800
89	8900	8900
90	9000	9000
91	9100	9100
92	9200	9200
93	9300	9300
94	9400	9400
95	9500	9500
96	9600	9600
97	9700	9700
98	9800	9800
99	9900	9900
100	10000	10000

Report No.:           RN1985-BLA                   MBA code:           ABCD

Measurement date: 85-04-03

Item ID: 0225ROD Batch ID: 6/07

Stratum ID: ROD Pu mass (g):  
U mass (g):

Comments :

Batch ID: 6/07

Current item: 200

B A T C H      D A T A      I N P U T

Data source: 0	MDC:	Total items	:	1
(Oper/Insp)		Total Pu mass (g):		1.000
		Total U mass (g) :		0.000

Pu isotopics Am-241

Analysis dates: 85-04-25 85-04-25

Isotopic Abundances	Errors	(smpdata)
---------------------	--------	-----------

Pu-238:	1.110	00.000	
Pu-239:	62.000	00.000	
Pu-240:	24.700	00.000	
Pu-241:	8.230	00.000	
Pu-242:	3.960	00.000	
Am-241:	57000 (ppm)	00.000	
U-235:	0.000 ←	00.000	← only for MOX

All essential elements entered. Save (Y/N)?

FIG. 2. Samples of input screen displays.

Pu High Resolution Gamma Spectroscopy

Report No.: RN1985-BLA

Facility:            MBA: ABCD

Measurement Date: 86-04-03    Tape ID.: CASSETTE1986B

ISOTOPIC ABUNDANCES (per 100 g of Pu)

Spec. ID	Item ID	Age(yrs)	Op./I	Pu-238	Pu-239	Pu-240	Pu-241	Pu-242	Am-241 Pu	U-235 Pu	Pu-240 eff
200	0225R0D	2.50	O-input	1.110	62.000	24.700	8.230	3.960	0.570	0.000	0.000
			O-nmlzd	1.110	62.000	24.700	8.230	3.960	0.570	0.000	0.000
			O-crctd	1.106	62.232	24.791	7.896	3.975	0.936	0.000	33.785
			I-blubx	1.132	61.485	26.431	7.766	3.185	0.994	0.268	34.251
			Merr (%)	0.029	1.327	2.164	0.182	3.185	0.028	0.011	5.450
			Diff (%)	-2.354	1.201	-6.618	1.647	19.861	-6.230	*****	-1.379
210			O-input	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a
			I-blubx	1.303	57.868	27.542	9.188	4.100	0.398	0.140	37.222
			Merr (%)	0.024	0.941	1.620	0.159	4.100	0.008	0.006	6.637
			Diff (%)	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a
240	4.03		O-input	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a
			I-blubx	1.437	58.219	27.561	8.445	4.337	1.807	0.244	37.949
			Merr (%)	0.027	0.906	1.546	0.156	4.337	0.042	0.008	6.982
			Diff (%)	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a
260	4.27		O-input	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a
			I-blubx	1.014	62.733	25.782	7.380	3.090	1.686	0.275	33.159
			Merr (%)	0.022	1.116	1.782	0.153	3.090	0.043	0.010	5.169
			Diff (%)	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a

FIG. 3. Sample of a worksheet.

even before the spectrum loading options were available. This version was the exact equivalent of the original PIAU function.

For this tool, the code had to be completely restructured to integrate it with the other functions that we wanted. We were careful not to modify the algorithm itself. The results obtained when activating this function are identical to those that would be obtained using the LLNL code on the PIAU (as of 1 January 1986).

Error messages have been improved and the response is much faster. The time for processing one 2000-channel spectrum has been reduced by a factor of at least 20 compared with the PIAU or the Cicero. In case of incorrectly selected peaks, a retry is possible immediately.

#### 4.5. Storage of computed isotopic abundances or ratios

The field equipment does not provide this function. For our work, however, this is essential. Several modifications had to be made in the LLNL (Blue Box) code to isolate the data elements we wanted and write them into a file. There is only one such file for each series. Following the naming convention described above, it is called SERIESNAM.MCA.

#### 4.6. Keyboard entry of data

This has been made easy by the provision of formatted screens, as shown in Fig. 2. To reduce redundancy we decided to use separate indexed files for the item and batch data. There must be item data for each spectrum in the series. Batch data can be common to several items and therefore do not need to be stored repeatedly. It is enough to provide the linkage between item data and batch data. Following our convention, the two files are called SERIESNAM.ITM and SERIESNAM.BCH.

If during item data entry the batch specified already exists, the linkage is done automatically without the user being asked to repeat the batch data entry. Because of this linkage, making modifications to existing batch data takes on a special significance. A separate subfunction is therefore provided for this purpose.

#### 4.7. Data confidentiality

The desk-top computer we used is essentially a single-user system. For such systems securing of data requires either the computer itself or the critical data media to be locked up. All functions that accept or generate data

have been designed to access only the series diskettes for storing or retrieving data. The easiest way for a user to protect data is to restrict access to his series diskettes.

#### 4.8. Decay correction and statistical tests

These two functions belong together. To test the validity of the operator's values, they have first to be decay corrected to the date of the inspector's verification measurements.

The statistical tests that will be used in the near future are currently the subject of much discussion and recommendations [6]. At the moment, however, simple difference tests are being made.

#### 4.9. Printouts

The immediate requirement was to generate a worksheet to be attached to the inspection report. A sample of this can now be generated and has been distributed to the relevant working group members for consideration (Fig. 3). Printouts similar to those from the Blue Box are also available.

### 5. CONCLUSIONS

We have described how we have used an HQ computer to make it possible to do work for supporting field activity. Without having first implemented the various functions as described above on an HQ computer we would not have been able to finish our work, although we were given strong support by both the Divisions of Operations and the Division of Development and Technical Support.

Besides fulfilling our own needs, this tool has also been useful to users from other divisions. It has already been used to process more than 500 spectra. The advantages that are becoming obvious to all who have occasion to use it are:

- Ease of use
- High speed
- Small risk of transcription errors
- Minimum routine data handling

In the course of our joint activity, it was recognized that the concept could also be useful for the inspector's work environment in the field.

We hope more HQ support work can be done in future using similar solutions.

## Appendix

### COMPUTER RESOURCES

A DEC PRO-380 was available to us for this work. The computer hardware consists of:

- A basic CPU with a 512 kbyte memory
- Memory management and floating point hardware
- A 33 Mbyte Winchester drive
- Dual 400 kbyte floppy disks
- Three-plane, bit mapped graphics hardware
- A colour monitor
- A dot matrix printer.

Auxiliary HRGS related hardware consists of:

- An Ispra 'Greyhound' cassette reader
- A Davidson PMCA.

The software tools we could use consist of:

- A P/OS multitasking operating system, Version 2.0
- Fortran-77
- A PDP-11 macro assembler
- An RMS file and record handling support
- An FMS forms package
- P/OS menu and help utilities
- A CORE graphics library.

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## NEW DEVELOPMENTS IN HIGH RESOLUTION GAMMA SPECTROMETRY MEASUREMENTS OF U AND Pu ISOTOPIC RATIOS AND ABUNDANCES

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### Abstract

#### NEW DEVELOPMENTS IN HIGH RESOLUTION GAMMA SPECTROMETRY MEASUREMENTS OF U AND Pu ISOTOPIC RATIOS AND ABUNDANCES.

A new method for  $^{235}\text{U}$  enrichment (abundance) measurement was developed. It unites intrinsic calibration in the 60–100 keV region with enrichment type measurement based on intensity ratio corrected 186 keV gamma ray intensity measurement. A corresponding computer program, URAX, was prepared. It was experimentally confirmed that for quasi-infinitely thick samples the intensities of different lines are strictly proportional to the abundances of emitting isotopes. It was found that for low enriched samples the intensity of self-excited U K X-rays is proportional to the  $^{235}\text{U}$  enrichment. It was established that the 35–60 keV region of the Pu and  $^{241}\text{Am}$  spectrum can be measured and used for precise isotopic analysis of freshly separated bulk Pu materials in 1 mm thick stainless steel containers. A suitable program for data analysis of materials with high burnup and high  $^{235}\text{U}$  concentration was prepared. The 94–122 keV region of the Pu and  $^{241}\text{Am}$  spectrum together with the region up to 208 keV is recommended for use in plant conditions for precise isotopic analysis with short measurement time.

### 1. INTRODUCTION

High resolution gamma spectrometry (HRGS) is the only non-destructive assay (NDA) technique for measurements of U and Pu isotopic ratios and abundances [1,2]. This technique still has significant development potential, particularly for field applications, e.g. for IAEA safeguards, both in improving the accuracy of the results and in reducing the time of measurement and data analysis. Some recent developments are reported in this paper.

TABLE I. SOME DATA CONNECTED WITH U308 CRM<sup>a</sup> MEASUREMENTS USING URAX PROGRAM

U308 standard	U-235 enrichment (%)	$\frac{I(63)}{E8_i}$	$\frac{I(93)}{E8_i}$	$\frac{I(98)}{E5_i}$	$\frac{I(98)}{F(E5)_i}$	$\frac{I(98)}{I(186)}$	$\frac{R(meas.)}{R(calc.)}$	$\frac{I(144)}{E5_i}$	$\frac{I(186)}{E5_i}$
1	0.3207	13.323	45.98	149.26	1.0005	5.931	1.0003	3.583	25.165
2	0.7209	13.384	45.82	75.03	0.99779	2.984	0.99833	3.635	25.141
3	1.9668	13.299	46.26	37.63	1.0000	1.495	0.9993	3.655	25.168
4	2.9852	13.331	46.54	30.2	0.99934	1.199	0.99833	3.639	25.187
5	4.5183	13.329	46.88	25.48	1.0000	1.014	1.0060	3.621	25.124
Mean		13.333	46.296		1.0005		1.0005	3.6266	25.157
SD		0.03113	0.42671		0.00259		0.00318	0.02722	0.02465
RSD (%)		0.23	0.92		0.26		0.32	0.75	0.10

Note:  $E8_i$  is the abundance of U-238 for standard i.

<sup>a</sup> CRM: certified reference material.



## 2. URANIUM MEASUREMENT AND DATA ANALYSIS

A new method and computer program for  $^{235}\text{U}$  enrichment (abundance) measurement and analysis, URAX, based on gamma spectrometric and absorptiometric measurements (GSAM) [3] was developed. It unites the positive features of two independent approaches: intrinsic calibration measurements (ICM) [4,5] using the energy range 60–100 keV and enrichment measurement [6,7] using in addition an intrinsic correction [8] for attenuation of the 185.7 keV gamma rays of  $^{235}\text{U}$ .

There are many papers connected with ICM of Pu but only a few connected with ICM of U [4,9], and there is still no suitable procedure or data analysis program for this type of U-abundance measurement. The main reason of course is the difficulty of this analysis due to the scarcity of suitable gamma and X-rays. Similar to the work described in Ref. [9] the new procedure again uses the 63.29, 92.37 and 92.79 keV gamma rays connected with  $^{238}\text{U}$  decay, the 84.24 keV gamma ray connected with  $^{235}\text{U}$  decay, and the 94.66 and 98.44 keV K X-rays, which are in practice the strongest lines in the measured spectrum of low enriched U-materials. The 92.37 and 92.79 keV gamma rays are considered as one peak with an average energy of 92.58 keV. The three ' $^{238}\text{U}$ ' lines and the two K X-ray lines are used to obtain the overall relative efficiency dependence (ORED) [5]. The ratio of the intensities of the 84.24 keV gamma ray to the double-peak, 92.58 keV gamma rays is used to determine the  $^{235}\text{U}/^{238}\text{U}$  ratio and finally the  $^{235}\text{U}$  abundance. It was essential to use the Gunnink algorithm [10] for fitting to the shape of the detected K X-ray peaks (93.35, 94.66 and 98.44 keV).

The second approach is the enrichment measurement method enhanced by an intrinsic correction of 185.7 keV gamma ray intensity attenuation using intensity ratio measurements [8]. The following ratios of strong peaks can be used: the 63.29 keV to the 92.58 keV 'doublet'; the 94.66 keV to the 98.44 keV K X-rays (the most intense lines); the 84.20 to the 185.7 keV lines; and the 143.8 to the 185.7 keV lines. The choice of ratio depends upon the thickness and gamma ray attenuation of the container wall at the measurement position. Some of the results from measurements of a set of certified reference materials (EC NRM 171), produced by the Central Bureau for Nuclear Measurements (Geel), and their analysis are collected in Table I.

The following conclusions can be drawn from these results.

- (a) The ratios of the intensities of the lines connected with  $^{235}\text{U}$  decay (e.g. 185.7 keV) to the corresponding  $^{235}\text{U}$

enrichments of the samples are constant values to within the statistical errors of the intensity measurements: this observation is correct to within the 0.1% RSD of the measurements of the 185.7 keV line. This means that the calibration line passes through the origin of the intensity versus enrichment co-ordinate system:

$$Ab(235) = K_5 I(186) \quad (1)$$

- where  $Ab(235)$  and  $I(186)$  are the  $^{235}\text{U}$  abundance and 185.7 keV line intensity of the corresponding samples.
- (b) The ratios of gamma rays connected with  $^{238}\text{U}$  decay (63.29, 92.37 and 92.79 keV lines) to the corresponding  $^{238}\text{U}$  abundances are also constant values to within the statistical errors of the corresponding intensity measurements:

$$Ab(238) = K_6 I(63) \quad (2)$$

The range of  $^{238}\text{U}$  abundances in this case was 95.5–99.7%, i.e. practically the highest possible abundances. In this indirect way (from two different isotopes,  $^{235}\text{U}$  and  $^{238}\text{U}$ ) it was experimentally confirmed that for this type of nuclear material and for quasi-infinitely thick samples the intensities of different gamma lines are strictly proportional to the corresponding isotopic abundances over their whole range. This important conclusion under corresponding conditions (quasi-infinitely thick samples) will be true for other special nuclear materials (SNMs) – Th, Pu and MOX materials.

- (c) The accuracy of this type of abundance measurement can be very high. It is determined by the statistical errors of the intensity measurements and errors connected with the determination of gamma ray attenuation by the container wall.
- (d) One partly unexpected result was the simple linear relation between the intensities of self-excited U K X-rays and the  $^{235}\text{U}$  enrichment in the low enrichment range of abundances:

$$I(98.44) = K_1 + K_2 Ab(235) \quad (3)$$

There is a simple relation also for the dependence of the intensity ratio of the 98.44 keV K X-ray line and the 185.7 keV line upon  $^{235}\text{U}$  enrichment:

$$R_1 = \frac{I(98.44)}{I(185.7)} = K_3 + K_4 \frac{1}{Ab(235)} \quad (4)$$

or

$$Ab(235) = \frac{K_4}{R_1 - K_3} \quad (5)$$

Because the enrichments in this range are determined by intensity ratios (which are relatively easily measured) and do not in practice depend upon the geometrical factors of the measurements, this simple dependence can provide a very convenient verification measurement of  $^{235}\text{U}$  enrichment.

- (e) At the same time it is well known [11] that the intensity of self-excited U K X-rays lines depends strongly on the concentration of U in the samples, so the  $R_1$  ratios in Eq. (4) will depend strongly on the U-concentration as well. Thus this dependence in connection with independent enrichment measurements (Eq. (1)) can be used for NDA verification measurements of U-concentrations in U-containing samples and items.

### 3. PLUTONIUM MEASUREMENTS AND ANALYSES

There are many papers connected with gamma spectrometric measurements of Pu and  $^{241}\text{Am}$  isotopic ratios and abundances. The main reason is the importance of this SNM. Recently we have had the opportunity to work on the following new developments.

#### 3.1. Procedure and computer program for isotopic analysis of freshly separated Pu materials

During inspection measurements of freshly separated bulk  $\text{PuO}_2$  samples it was established that even for bulk samples (more than 1 kg of material) contained in 1 mm thick steel containers the low energy part (35-60 keV) of the Pu and  $^{241}\text{Am}$  spectrum can be easily measured. It is well known [12-15] that in this part of the spectrum all Pu isotopes (with the exception of  $^{241}\text{Pu}$ ) and  $^{241}\text{Am}$  are very well represented and the corresponding gamma lines are easily measured, so that isotopic abundances and their ratios can be measured with high accuracy. In our case we used the 38.66 and 51.62 keV gamma rays of  $^{239}\text{Pu}$ , the 43.50 keV gamma rays of  $^{238}\text{Pu}$ , the 45.24 keV gamma rays of  $^{240}\text{Pu}$ , the 59.54 keV gamma rays of  $^{241}\text{Am}$ , and the 59.54 and 64.83 keV gamma rays of  $^{237}\text{U}$  (a daughter product of  $^{241}\text{Pu}$ ). In addition, the 94.66, 97.07 and 101.1 keV lines, the '111 keV' group of K X-ray lines, and the 164.6 and 208 keV  $^{237}\text{U}$  gamma lines were used for ORED calibration purposes, and the 129.3 and 148.6 keV lines for determination of the  $^{241}\text{Pu}/^{239}\text{Pu}$  ratio.

A program for this procedure has been prepared.

TABLE II. RESULTS OF ANALYSIS OF  $^{239}\text{Pu}$  AND  $^{240}\text{Pu}$  ABUNDANCES AND RATIOS FOR MEDIUM AND HIGH BURNUP MATERIALS USING PIAU PROGRAM

Spectrum No.	Pu-240 (op.)	Pu-240 (meas.)	Meas. Op.		Pu-239 (op.)	Pu-239 (meas.)	Meas. Op.		Pu-240 Pu-239 (op.)	Pu-240 Pu-239 (meas.)	R(meas.) R(op.)	
15	20.273	20.446	1.0085		74.39	74.75	1.0048		0.27252	0.27353	1.0037	
16	20.273	20.645	1.0183		74.39	74.56	1.0023		0.27252	0.27689	1.0160	
22	26.000	26.142	1.0055		68.17	67.82	0.99487		0.38140	0.38546	1.0107	
23	23.261	22.797	0.98005		73.004	73.79	1.0108		0.31863	0.30894	0.96961	
26	23.245	20.912	0.89963		71.9	74.43	1.0352		0.32330	0.28096	0.86905	
27	23.245	23.209	0.99845		71.9	71.98	1.0011		0.32330	0.32244	0.99734	
28	23.245	23.262	1.0007		71.9	71.83	0.99903		0.32330	0.32385	1.0017	
Mean			0.98732				1.0069				0.98115	
SD			0.04039				0.01342				0.05160	

13	22.656	24.189	1.0677	63.02	61.33	0.97318	0.35950	0.39441	1.0971
14	22.656	26.042	1.1495	63.02	60.29	0.95668	0.35950	0.43195	1.2015
18	24.127	24.463	1.0139	62.21	61.64	0.99084	0.38783	0.39687	1.0233
18b	24.127	25.188	1.0440	62.21	60.68	0.97541	0.38783	0.41510	1.0703
20	24.127	25.238	1.0460	62.21	61.7	0.99180	0.38783	0.40904	1.0547
24	24.336	24.259	0.99684	62.084	63.04	1.0154	0.39199	0.38482	0.98172
Mean			1.0530			0.98388			1.0714
SD			0.05353			0.02015			0.07513

Note: For the medium burnup materials (first group) neither Pu-240 abundances nor Pu-240/Pu-239 ratios indicate bias. For higher burnup materials (second group) both Pu-240 abundances and Pu-240/Pu-239 ratios indicate bias.

### 3.2. Measurements and analyses of high burnup materials and MOX with high concentrations of $^{235}\text{U}$

The computer program PIAU [16] is widely used for data analysis of gamma spectra for Pu isotopic ratio determination. For low burnup materials it gives satisfactory results. However, for high burnup plutonium materials (Table II) and for MOX materials with high  $^{235}\text{U}$  concentrations the results are not satisfactory. The reasons for these results are as follows:

- (a) Many of the  $^{235}\text{U}$  gamma ray lines were not considered in the program.
- (b) The contribution of  $^{240}\text{Pu}$  gamma rays in the complex peak at 160 keV is quite small for high burnup Pu materials, and a small error in ORED will result in a significant error in the  $^{240}\text{Pu}/^{239}\text{Pu}$  ratio.

In order to solve these problems at least temporarily a new program, PuU, was prepared which works sequentially. Using the 267-393 keV range the  $^{241}\text{Pu}/^{239}\text{Pu}$  and  $^{241}\text{Am}/^{239}\text{Pu}$  ratios are determined. In this region  $^{235}\text{U}$  does not emit any gamma rays. With these data the ORED is determined in the 122-208 keV region on the basis of the four strong peaks at 129.3, 148.6, 164.6 and 208.0 keV. Three of them are mainly connected with  $^{241}\text{Pu}$  decay. It was significant that the last three peaks are determined using Gaussian fitting on a part of the peak which is free from the influence of other peaks. The net counts per channel are of course determined over a wider range using a diffused step function [17] subtraction of the background. Special attention was paid to error propagation in order to properly determine the uncertainties of the Pu and  $^{241}\text{Am}$  ratios and abundances [18].

Some results using this program for materials that are difficult to analyse are shown in Tables III and IV. It is easy to see that there are some improvements of the results. Still we are not quite satisfied, particularly with the results for  $^{240}\text{Pu}$ , which is important for combined gamma-neutron coincidence measurements used by the IAEA. The main reason of course is that the  $^{240}\text{Pu}$  contribution in the complex 160 keV group for high burnup Pu materials is quite small. This is why efforts to use the possibilities of other energy regions are continuing.

### 3.3. Use of the 600-800 keV region

In this region  $^{238}\text{Pu}$ ,  $^{239}\text{Pu}$ ,  $^{240}\text{Pu}$  and  $^{241}\text{Am}$  are well represented. By combining measurements in the 600-800 keV region with those in the 330-393 keV region, all Pu and  $^{241}\text{Am}$  isotopic ratios and abundances can be determined. The work of

TABLE III. RATIOS OF MEASURED TO KNOWN ISOTOPIC RATIOS ON THE SAME SPECTRA USING PROGRAMS PIAU AND PuU

		$\frac{\text{Pu-238}}{\text{Pu-241}}$	$\frac{\text{Pu-239}}{\text{Pu-241}}$	$\frac{\text{Pu-240}}{\text{Pu-241}}$	$\frac{\text{Am-241}}{\text{Pu-241}}$
PuU	Mean	1.004	1.002	1.055	1.013
	SD	0.029	0.050	0.138	0.090
PIAU	Mean	1.010	0.942	1.047	1.079
	SD	0.039	0.143	0.103	0.147

TABLE IV. RATIOS OF MEASURED TO KNOWN ISOTOPIC ABUNDANCES ON THE SAME SPECTRA USING PROGRAMS PIAU AND PuU

		Pu-238	Pu-239	Pu-240	Pu-241	Pu-242	Am-241
PuU	Mean	0.970	1.005	0.998	1.008	0.862	1.010
	SD	0.086	0.012	0.041	0.065	0.082	0.109
	(G-M)% <sup>a</sup>	-2.97	+0.54	-0.19	+0.75	-13.8	+1.02
PIAU	Mean	0.954	0.991	1.068	0.960	0.802	1.040
	SD	0.063	0.012	0.036	0.046	0.098	0.109
	(G-M)% <sup>a</sup>	-4.62	-0.91	+6.85	-4.04	-19.8	+3.99

<sup>a</sup> (G-M)% = [(GS-MS)/MS] × 100%.

Ottmar et al. [19] and Fleissner et al. [20] has shown the possibilities of this region very well. In this case the high penetration of the high energy gamma rays is very attractive. There are still two weak points: the measurement time is rather long and the information for <sup>235</sup>U and for U in general is rather poor.

### 3.4. Use of the 94-122 keV region

This region is particularly attractive, since all Pu isotopes and <sup>241</sup>Am are very well represented. If a thin planar detector is used then most of the registered gamma rays will be in this region and can be used in the data analysis,

and even if the counting throughput rate is not very high, it will be satisfactory. Uranium-235 gamma rays do not in practice disturb the Pu gamma ray peaks in this region. Together with the gamma rays of the neighbouring regions up to 208 keV, all Pu,  $^{241}\text{Am}$  and  $^{235}\text{U}$  abundances and ratios can be determined with high accuracy and reliability. Gunnink has been using this region for several years. He has developed a specific algorithm [10] for correct fitting of the shape of the X-ray peaks, which is extremely important for analysis of this very complex region where gamma and X-rays are closely mixed. The complexity of this region is the main reason why it has not been used more. Improvements in detectors, related electronics and the specific software nowadays, however, make it possible to take further steps and to use this region widely in the gamma spectrometric determination of Pu and  $^{241}\text{Am}$  isotopic composition.

Even for this relatively narrow energy range the ORED for detection of gamma and X-rays with different energies is very important for the determination of isotopic ratios. Two approaches to solving this problem are possible; they can be described as follows:

The first one, developed by Gunnink, is based on general physical considerations in determining the ORED. The ORED is determined by three main factors: (i) the integration and attenuation of gamma and X-rays in the measured sample itself, (ii) the attenuation of gamma ray intensities by all absorbers between the measured sample and the detector (e.g. container wall, Cd absorber), and (iii) the energy dependence of the detector efficiency in detecting gamma rays with different energies.

In this approach, parameters for all these factors are determined separately. At the same time the parameters are determined together with different isotopic contributions for the whole range of the measured spectrum; the intensities of the individual lines in general are not determined.

The second approach is based on the intrinsic calibration method. First, the intensities of most of the peaks are determined. Then, using these values and some of the known and measured intensity ratios, the ORED is determined. Finally, all of the isotopic ratios and abundances are calculated. Thus, in this case the intensities of the single peaks are determined explicitly and can be used for GSAM [3] evaluation of the same measured spectrum.

There are two main difficulties in measuring the single peak intensities in this energy region. The peaks are quite



close in two of the complex groups: 98-100 keV (five peaks) and 102-104 keV (five peaks), and the gamma and X-rays are mixed together. The last difficulty turns out to be an advantage at the same time, since the different shapes of the gamma and X-ray peaks help significantly in solving the problem. The investigation which we have done has shown that when shapes are different even two peaks with the same energy can be resolved.

The precise determination of the ORED is favoured by the many strong lines from only two internally related groups: the group of  $^{241}\text{Pu}$  lines, including  $^{237}\text{U}$  lines (corrected for small  $^{241}\text{Am}$  contributions) in the case of aged materials, or only  $^{237}\text{U}$  lines in the case of fresh materials, and the group of three strong U K X-ray lines. For the ORED only the relative intensities of these lines are important, so they are not so strongly sensitive to a lack of homogeneity in the sample.

A simple iterative procedure is used in calculating the ORED for the whole region between 94 and 208 keV. First, the three U K X-ray peaks (94.66 and 98.44 keV and the 111 keV group) are used to determine the ORED in the 94-111 keV range, the  $^{241}\text{Am}/^{241}\text{Pu}$  ratio (from two strong and close lines: the 102.97 keV line of  $^{241}\text{Am}$  and the 103.68 keV line of  $^{241}\text{Pu}$ ) and the  $^{241}\text{Am}/^{237}\text{U}$  ratio (from the same  $^{241}\text{Am}$  line and the strong 101.07 keV line of  $^{237}\text{U}$ ). The small contribution of  $^{241}\text{Am}$  in the last peak can easily and precisely be taken into account. The  $^{237}\text{U}/^{241}\text{Pu}$  ratio for aged materials is known and fixed, so in this way the material is characterized as aged or freshly separated.

These ratios being known, the small correction for  $^{241}\text{Am}$  contributions is made in all  $^{237}\text{U}$ - $^{241}\text{Am}$  peaks, including the two strong and well separated 97.07 and 101.07 keV X-ray lines due to  $^{237}\text{U}$  and  $^{241}\text{Am}$  decays. Then, using nine lines (94.66, 98.44, 111.0, 97.07, 101.07, 103.68, 148.57, 164.60 and 208 keV), the ORED is determined in the 94-208 keV region. At the same time the K-edge discontinuity in the ORED due to either Pu or mixed Pu-U material absorption is taken into consideration and determined as an unknown value. Having peak intensities and the ORED makes it easy to determine all isotopic ratios, including that of  $^{235}\text{U}/^{241}\text{Pu}$ , from the 185.7 keV peak of U and the 208 keV peak of  $^{237}\text{U}$  for aged samples or from the 148.567 keV peak for freshly separated samples.

The specific alpha activity of the sample (the sum of alpha particle energies per gram of sample) is proportional to the specific thermal power of the sample. For homogeneous samples the ratio of Pu K X-ray (99.53 or 103.75 keV) intensities to the specific power of the sample is correlated

TABLE V. PRECISION ERRORS ( $SD/\bar{R}(\%)$ ) OF Pu ISOTOPIC RATIOS ON THE SAME SPECTRA USING PIAU AND GSAM METHODS  
(10 measurements;  $t = 600$  s)

	$\frac{\text{Pu-238}}{\text{Pu-241}}$	$\frac{\text{Pu-239}}{\text{Pu-241}}$	$\frac{\text{Pu-240}}{\text{Pu-241}}$	$\frac{\text{Pu-242}}{\text{Pu-241}}$	$\frac{\text{Am-241}}{\text{Pu-241}}$
PIAU	2.14	15.6	6.2	29.2	18.0
GSAM	1.69	1.15	6.8	7.97	2.5

TABLE VI. PRECISION ERRORS ( $SD/\bar{R}(\%)$ ) OF ISOTOPIC ABUNDANCES ON THE SAME SPECTRA USING PIAU AND GSAM METHODS  
(10 measurements;  $t = 600$  s)

	Pu-238	Pu-239	Pu-240	Pu-241	Pu-242	Am-241
PIAU	9.6	8.6	9.0	8.0	35.6	8.2
GSAM	1.7	1.4	6.8	0.7	7.8	2.3

with the Pu concentration. Similarly, the ratios of the alpha excited contributions to the U K X-ray intensities (94.66 and 98.44 keV) to the specific thermal power of the sample are correlated with the U-concentration of the sample.

This information is quantitatively reliable only if U, Pu and other materials in the sample are homogeneously mixed. Unfortunately we do not have experimental information about the performance with this approach.

Recent measurements both in Ispra and in Vienna using Gunnink's program were very impressive. It was possible to measure and analyse with high precision a single sample in only five minutes. However, we still do not have enough experimental data about the performance of this program.

### 3.5. Gamma spectrometric and absorptiometric measurements (GSAM)

In addition to intrinsic energy and ORED calibration of the measurements, GSAM also uses the information on the attenuation of gamma rays in the measured sample itself and in

the absorbers between the sample and the detectors (e.g. container wall). In the case of quasi-infinitely thick SNM samples this corresponds to the isotopic abundance (enrichment) type of measurement. As already indicated in the case of U-measurements, this approach has very attractive features in respect of simplicity and accuracy, and in the case of U it is widely used. In spite of the fact that Pu gamma spectrometry is even more suitable for this approach [21] until now this technique practically has not been in use. The main reason probably is the even greater simplicity of the intrinsic calibration method for Pu isotopic ratio determination. We think, however, that the increase in information provided by GSAM and the higher accuracy which one can expect should be used whenever they are required, bearing in mind that if these measurements are well prepared in advance (suitable hardware and precalibration) they may require even less measurement time and be simpler, e.g. measurements of  $\text{PuO}_2$  and MOX in standard containers, pellets, rods and fuel assemblies. A comparison of the precision errors obtained using ICM and GSAM on the same set of spectra is given in Tables V and VI.

#### 4. REMARKS

Owing to the efforts of the development groups working on NDA gamma spectrometric measurements of uranium and plutonium isotopic ratios and abundances, there has been significant progress in this field over the last few years. Difficulties connected with the variety of the materials to be measured were identified and ways to overcome them investigated. More information was published on the possibilities of using different energy regions of the spectrum. Probably the most significant, from a practical point of view, are the strong indications that the 94-122 keV region of the Pu and  $^{241}\text{Am}$  spectrum can be used by inspectors under plant conditions. This enables precise Pu isotopic measurements to be made with short measurement times.

It is important to finally establish the practicality of using the 94-122 keV region, and to develop corresponding procedures and data analysis programs as soon as possible, because the choice of the energy region of the spectrum to be measured determines the features of the detectors, electronics and software to be used, and once these are routinely accepted and established it is not easy to change them.

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## СТАЦИОНАРНАЯ УСТАНОВКА ДЛЯ НЕРАЗРУШАЮЩЕГО КОНТРОЛЯ ТВС ЭНЕРГЕТИЧЕСКИХ РЕАКТОРОВ

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### Abstract—Аннотация

#### FIXED FACILITY FOR NON-DESTRUCTIVE ANALYSIS OF POWER REACTOR FUEL ASSEMBLIES.

The results of work to set up the ESNART facility for research into neutron methods for the non-destructive analysis of WWER type power reactor fuel assemblies are given in brief. In order that active and passive neutron methods may be studied, the composition of the formative zone of the facility (whose configuration can be controlled operationally) includes a cyclically switched antimony-beryllium irradiator with a maximum  $^{124}\text{Sb}$  source activity of about  $4 \times 10^{11}$  Bq and a channel type detecting system with up to 44 SNM-18  $^3\text{He}$  detectors arranged in a cylindrical paraffin moderator and up to 16 in peripheral segments, with replaceable liquid fillers. The fission neutron detection efficiency is 10–20%, depending on the configuration of the formative zone. The facility is used for research on the possibility of non-destructive fuel assembly assay using full scale (except in length) assembly analogues whose  $^{235}\text{U}$  concentrations and distributions can be varied. Particular attention was given to studying the facility's response function and the components from which it is synthesized. These include the intensity of induced fission of fissile nuclides and the efficiency of fission neutron detection. The results of experiments on the ESNART facility to analyse fresh WWER-440 and WWER-1000 fuel assemblies are given. The reproducibility of the measurement results was 0.7%, which corresponds to detecting the removal of two or three fuel elements out of 312 for WWER-1000 assemblies and of one in 126 for WWER-440 assemblies. With maximum gamma source activity, some 40 000 delayed neutrons from induced fission of  $^{235}\text{U}$  in WWER-1000 fuel assemblies were detected over 10 minutes of cyclical operation of the facility.

#### СТАЦИОНАРНАЯ УСТАНОВКА ДЛЯ НЕРАЗРУШАЮЩЕГО КОНТРОЛЯ ТВС ЭНЕРГЕТИЧЕСКИХ РЕАКТОРОВ.

Кратко изложены результаты работы по созданию установки ЭСНАРТ, предназначенной для исследования нейтронных методов неразрушающего контроля (НРК) ТВС энергетических реакторов типа ВВЭР. Для изучения активных и пассивных нейтронных методов в состав формирующей зоны установки, конфигурация которой может оперативно изменяться, входят циклически включаемый сурьмяно-бериллиевый облучатель с максимальной активностью источников сурьма-124 около  $4 \times 10^{11}$  Бк и каналная детектирующая система с гелий-3 детекторами СНМ-18, размещенными в цилиндрическом парафиновом замедлителе (до 44 шт) и периферийных сегментах (до 16 шт) со сменными жидкими наполнителями. В зависимости от конфигурации формирующей зоны эффективность регистрации нейтронов деления составляет 10–20%. На установке выполнено изучение возможностей НРК ТВС с использованием их полномасштабных (кроме длины) аналогов,

обеспечивающих возможность варьировать концентрацию урана-235 и ее распределение. Основное внимание уделялось изучению функции отклика установки и функций, из которых она синтезируется. К ним относятся интенсивность вынужденных делений делящихся нуклидов и эффективность регистрации нейтронов деления. Приведены экспериментальные результаты, полученные на установке ЭСНАРТ при контроле неработавших ТВС ВВЭР-440 и ВВЭР-1000. Воспроизводимость результатов измерений составляет 0,7 %, что соответствует обнаружению изъятия двух-трех твэлов из 312 для ТВС ВВЭР-1000 и одного из 126 для ТВС ВВЭР-440. При максимальной активности гамма-источников за 10 мин циклической работы установки регистрируется около 40 000 запаздывающих нейтронов вынужденного деления урана-235 в ТВС ВВЭР-1000.

Стационарные установки для неразрушающего контроля ядерных материалов могут иметь ряд преимуществ, компенсирующих их нетранспортабельность. Отсутствие жестких ограничений на габариты, массу и электронное оборудование позволяет расширить функциональные возможности установок, повысить производительность и точность контроля, а также проводить исследования методов контроля применительно к конкретному типу объектов. Эти исследования позволяют оптимизировать условия контроля, усовершенствовать отдельные узлы с целью повышения их надежности. Прекрасным образцом является семейство установок PHONID [1, 2], развитие которых на протяжении ряда лет служит стимулом для создания стационарных установок.

Стационарная установка ЭСНАРТ предназначена для исследования нейтронных методов НРК крупногабаритных изделий ядерного топливного цикла таких, как ТВС энергетических реакторов типа ВВЭР. Целью исследований является выработка оптимальных условий, при которых контроль ТВС данного типа может быть проведен с минимальными погрешностью и временными затратами. Такие исследования предполагают подробное изучение пространственных распределений нейтронных полей в измерительной системе, включающей контролируемый объект, выявление и подавление наиболее сильных источников погрешностей, проверку метода в полномасштабном эксперименте с аналогами штатных ТВС.

Конструкция установки ЭСНАРТ обеспечивает исследование активных и пассивных нейтронных методов НРК и оптимизацию условий контроля ТВС диаметром до 280 мм. Общий вид установки приведен на рис. 1. Элементы формирующей зоны имеют сегментную конфигурацию, позволяющую собирать их в кольцевую систему с центральным измерительным каналом различного диаметра, оптимального для данного типа ТВС. Формирующая зона высотой около 30 см образована сурьмяно-бериллиевым выключаемым облучателем со свинцовым отражателем, парафиновым замедлителем, в котором на двух различных диаметрах расположены гнезда для размещения 44-х детекторов тепловых нейтронов типа СНМ-18 и периферийными сегментами с гнездами для 16-ти таких же детекторов. Эти сегменты могут быть заполнены как чистой водой, так и водными растворами поглотителей тепловых нейтронов. В промежутках между элементами формирующей зоны могут быть установлены кадмиевые экраны.

Конструкция установки обеспечивает циклическое включение с быстрым (за 1 с) выключением облучателя путем подъема 16-ти гамма-источников сурьма-124 из свинцового контейнера и ввода их в бериллиевый конвертор, расположенный на высоте



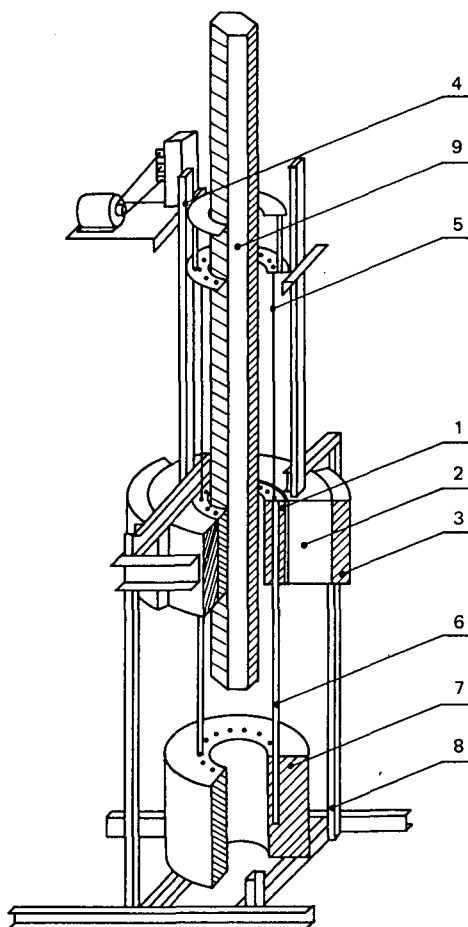


Рис. 1. Установка ЭШАРТ: 1 — сурьмяно-бериллиевый облучатель; 2 — парафиновый замедлитель; 3 — периферийный сегмент; 4 — устройство транспортировки источников; 5 — подвес источников; 6 — транспортный канал источника; 7 — свинцовый контейнер; 8 — опорная рама; 9 — ТВС.

около 1 м над контейнером. При включенном облучателе формируется поток нейтронов, вызывающих деление делящихся нуклидов (уран-235, плутоний-239), регистрируются мгновенные нейтроны деления и мониторируется облучающий поток. После выключения облучателя регистрируются запаздывающие нейтроны в различных временных интервалах.

Детектирующая система установки имеет каналную структуру, в которой каждый из детекторов обеспечен индивидуальными усилителем, дискриминатором и формирователем. Система регистрации позволяет фиксировать информацию как от каждого из детекторов, так и от различных их сочетаний. Такая схема помимо обеспечения мак-

симальной надежности дает возможность обнаруживать локализованные неоднородности распределения ядерного материала в ТВС.

На установке выполнено изучение возможностей неразрушающего анализа ТВС, причем для определения содержания урана-235 использовался активный, а для урана-238 — пассивный нейтронные методы. При исследованиях необлученного топлива ВВЭР-440 использовался аналог ТВС, геометрические размеры которого и конструкционные материалы аналогичны штатным ТВС. Конструкция аналога длиной 50 см позволяет оперативно изменять как общее содержание урана-235, так и его пространственное распределение. Такое изменение производится заменой твэлов, содержащих топливные таблетки с различным (1,6; 2,4 и 3,6%) обогащением и с размещением в твэлах ампул, содержащих порошок высокообогащенной окиси урана. Между топливными таблетками твэлов можно размещать комплекты слюдяных детекторов (КСД) для измерения потока облучающих нейтронов внутри твэлов. КСД состоит из слоя ( $1 \text{ мг/см}^2$ ) высокообогащенного урана и слюдяного детектора осколков деления. Сверху и снизу комплект фиксируется алюминиевыми или кадмиевыми дисками, толщиной 1 мм. Внутри твэлов могут также размещаться специально изготовленные полупроводниковый детектор делений (ПДД), калифорниевый или америций-литиевый источники нейтронов.

При исследованиях необлученного топлива ВВЭР-1000 использовался укороченный (1,2 м) вариант штатной ТВС с дополнительным набором твэлов, заполненных ураном с обогащением 2,0; 2,4; 2,8; 3,2 и 3,6%.

Перечисленные средства позволяют проводить исследование пространственного распределения облучающих нейтронов и нейтронов деления в формирующей зоне установки с контролируемым объектом (ТВС). Интенсивность делений урана-235 в облучающем потоке в любой точке ТВС оперативно может быть измерена с помощью ПДД, а одновременно в нескольких (до 50) — с помощью КСД. Предусмотрена также возможность измерения кадмиевого отношения.

Эффективность регистрации мгновенных и запаздывающих нейтронов, испускаемых из различных точек ТВС, изучалась при размещении внутри твэлов калифорниевых и америций-литиевых источников соответственно. Проводилось также и прямое исследование эффективности регистрации запаздывающих нейтронов при облучении ампул с высокообогащенным ураном вне установки и затем быстрой (за 7 с) доставки и размещения их в заданной позиции в ТВС.

Отклик системы на локальное изменение концентрации урана-235 измерялся при послойной замене твэлов в ТВС, когда в заданном слое равномерно размещались твэлы с пониженным или с повышенным обогащением.

Физические обоснованные функции связи между контролируемой и регистрируемой величинами, определенные во всей измерительной системе и зависящие от ее нейтронно-физических характеристик, могут быть получены следующим образом.

Отклик  $R$  анализирующей системы, характеризующий вероятности регистрации единицы массы делящихся нуклидов, может быть описан двумя величинами [3–5]. Одна из них  $F$  — интенсивность делений делящихся нуклидов в потоке облучающих нейтронов, другая  $S$  — эффективность регистрации актов вынужденных делений.

Функциональная зависимость величин  $R$ ,  $F$ ,  $S$  от пространственных координат может быть представлена в виде:

$$R(\vec{r}, \kappa_1, \kappa_2) = F(\vec{r}, \kappa_1) \cdot S(\vec{r}, \kappa_2), \quad (1)$$

где  $\vec{r}$  — радиус-вектор,  $\kappa_1$  и  $\kappa_2$  — параметры, зависящие от нейтронно-физических характеристик системы.

Основные процессы, протекающие в системе с активным нейтронным методом, можно описать взаимодействием с нуклидами среды двух независимых потоков нейтронов, один из которых, первичный, создается внешним источником нейтронов, а второй, вторичный — нейтронами, испускаемыми из анализируемого объекта в актах вынужденного деления. Каждый из этих потоков может быть охарактеризован некоторой средней энергией, причем для первичного потока она приходится на тепловую и эпитепловую область, а для вторичного — составляет около 2 МэВ. Соответственно этим средним энергиям параметры  $\kappa_1$  и  $\kappa_2$  для одной и той же системы будут различными.

Аналитические выражения для функций  $R$ ,  $F$  и  $S$  могут быть получены в диффузионном приближении с решением диффузионного уравнения по методу, изложенному в [6] для элементарной ячейки цилиндрического реактора. Однако, учитывая диаметрально противоположные задачи, решаемые при описании активной зоны реактора и системы контроля, решения, полученные в [6], не могут быть использованы непосредственно. Требуется модификация граничных условий, заключающаяся в том, что экстраполируемая граница  $H'$  в аксиальном направлении должна быть устремлена к бесконечности. Это связано с характером распределений функций  $R$ ,  $F$  и  $S$  в системах контроля протяженных объектов, таких как ТВС. Используя метод решения диффузионного уравнения для реактора с источником [7], можно построить решение уравнения Гельмгольца в виде суммы:

$$G(m, \sigma, z) = \sum_{n=0}^{\infty} a_n \cos \alpha_n \cdot z. \quad (2)$$

Функция нормального распределения (Гаусса) с параметрами  $m$  и  $\sigma$  может быть получена, если коэффициенты  $a_n$  в (2) получены разложением функции  $G$  в ряд Фурье по косинусам.

Модифицированные решения диффузионного уравнения для функций  $F$ ,  $S$  и  $R$  с учетом аксиальной симметрии, имеют вид:

$$F_i(r, z) = A_i I_0(\kappa_i, r) \cdot G(m_i, \sigma_i, z), \quad (3)$$

где  $F_i$  — обозначает одну из функций  $F$ ,  $S$  и  $R$ ;

$$\kappa_i = \frac{\sum a_i}{D_i} + \frac{1}{a_n^2}, \quad \frac{1}{a_n^2} = \frac{\sum a_n^2 \cos \alpha_n \cdot z}{\sum a_n \cos \alpha_n \cdot z};$$

$m_i$  и  $\sigma_i^2$  — центр и дисперсия нормального распределения, соответственно.

Учитывая, что при решении задач НРК искомой величиной является масса контролируемых нуклидов, целесообразно видоизменить макросечение взаимодействий следующим образом:

$$\Sigma_i(\vec{r}, E) = \rho(\vec{r}) \cdot \frac{\sigma_i(E) \cdot N_0}{A} = \rho(\vec{r}) \Sigma^m(E), \quad (4)$$

где  $E$  — энергия нейтронов;  $\rho(r)$  — плотность;  $\sigma_i(E)$  — микросечение взаимодействия типа  $i$ ;  $N_0$  — число Авогадро;  $A$  — массовое число нуклидов. Величина  $\Sigma^m(E)$ , которую можно назвать массовым макросечением, имеет размерность  $L^2 M^{-1}$ . Используя (4) можно связать эту величину с плотностью потока  $\Phi$  облучающих нейтронов:

$$F(\vec{r}) = \int_E \Sigma^m(E) \cdot \Phi(\vec{r}, E) dE. \quad (5)$$

Результат измерения массы делящихся нуклидов можно записать в виде:

$$\begin{aligned} N &= \int R(\vec{r}, \kappa_1, \kappa_2) \cdot \rho(\vec{r}) \cdot dv = \\ &= 2\pi A_R \iint I_0(\kappa_R, r) \cdot G(m_R, \sigma_R, z) \cdot \rho(\vec{r}, z) \cdot dE \cdot r \cdot dr, \end{aligned} \quad (6)$$

где  $N$  — скорость регистрации нейтронов деления;  $\kappa_R^2 \approx \kappa_f^2 + \kappa_S^2$ .

В общем случае решение выражения (6) можно получить в виде бесконечного степенного ряда, однако в некоторых случаях его можно свести к выражению с функциями Бесселя первого ряда, нулевого и первого порядка.

Если  $\rho(r, z) = \rho_0$ , то используя результаты, полученные в [8] для бесконечного поглощающего цилиндра, можно получить:

$$N = A_0 \cdot m \cdot \bar{F} = A_0 \cdot F_0 \cdot m \cdot \frac{2I_1(\kappa_R, r_0)}{\kappa_R r_0 I_0(\kappa_R, r_0)} \cdot \frac{f_0}{f_S}, \quad (7)$$

где  $A_0$  — калибровочный коэффициент,  $m$  — линейная плотность контролируемых нуклидов;  $F$  — интенсивность делений, усредненная по сечению объекта;  $F_0$  — интенсивность делений с невозмущенным потоком внутри объекта;  $r_0$  — радиус окружности;  $f_S$  и  $f_0$  — коэффициенты самоэкранирования и полного самоэкранирования, соответственно;  $I_0$  и  $I_1$  — функции Бесселя.

ТАБЛИЦА I. РАСПРЕДЕЛЕНИЯ ФУНКЦИЙ В ТВЦ ВВЭР-440 (БЕЗ КАДМИРОВАНИЯ)

r	0	1.22	2.44	3.66	4.88	6.10	7.32
S	3.05	3.05	3.06	3.07	3.08	3.09	3.10
F	52.67	51.4	54.0	60.6	71.1	85.7	104
R	7.51	7.35	7.78	8.79	10.4	12.8	15.8

ТАБЛИЦА II. РАСПРЕДЕЛЕНИЯ ФУНКЦИЙ В ТВС ВВЭР-440 (С КАДМИРОВАНИЕМ)

r	0	1.22	2.44	3.66	4.88	6.10	7.32
S	2.87	2.87	2.88	2.88	2.89	2.90	2.91
F	15.4	15.8	16.2	16.6	17.8	17.8	17.8
R	2.02	2.08	2.14	2.20	2.26	2.32	2.39

ТАБЛИЦА III. ЗНАЧЕНИЯ КОЭФФИЦИЕНТА  $\kappa_i$ 

Функции	Характеристика формирующей зоны	
	без кадмирования	с кадмированием
S (отн. ед)	0.035	0.032
F (отн. ед)	0.347	0.103
R (отн. ед)	0.357	0.112

Так как  $\kappa_R$  пропорционально  $\Sigma_a$ , то из (7) видно, что с ростом поглощающих свойств объекта скорость регистрации нейтронов деления падает в основном из-за уменьшения величины среднего по сечению потока облучающих нейтронов. Сечение поглощения  $\Sigma_a$  является суммой сечений всех элементов, находящихся в ТВС, включая как делящиеся нуклиды, так и неделяющиеся поглотители. Таким образом в (7) учитываются эффекты матрицы, составляющие одну из серьезных проблем при НРК [9]. Если поглощающие средства объекта не учитываются при облучении тепловыми нейтронами, регистрируемая величина может оказаться в несколько раз меньше, чем должна была быть при невозмущенном потоке. Это особенно важно при контроле ТВС с выгорающими поглотителями или с осколками деления. Чтобы избежать большого отклонения  $F$  от  $F_0$  необходимо уменьшать величину  $\kappa_R$ . Этого можно добиться, исключив тепловые нейтроны из потока внутри объекта, например, путем введения кадмиевого экрана, окружающего ТВС [1].

Результаты измерений распределений и определения величины  $\kappa_i$  для функций  $R$ ,  $F$ ,  $S$  в ТВС ВВЭР-440 приведены в табл. I–III, иллюстрирующих изложенное выше.

В табл. I и II приведены радиальные распределения функций, полученные при интегрировании их по оси  $z$ . Значения функций в каждом слое твэлов были получены

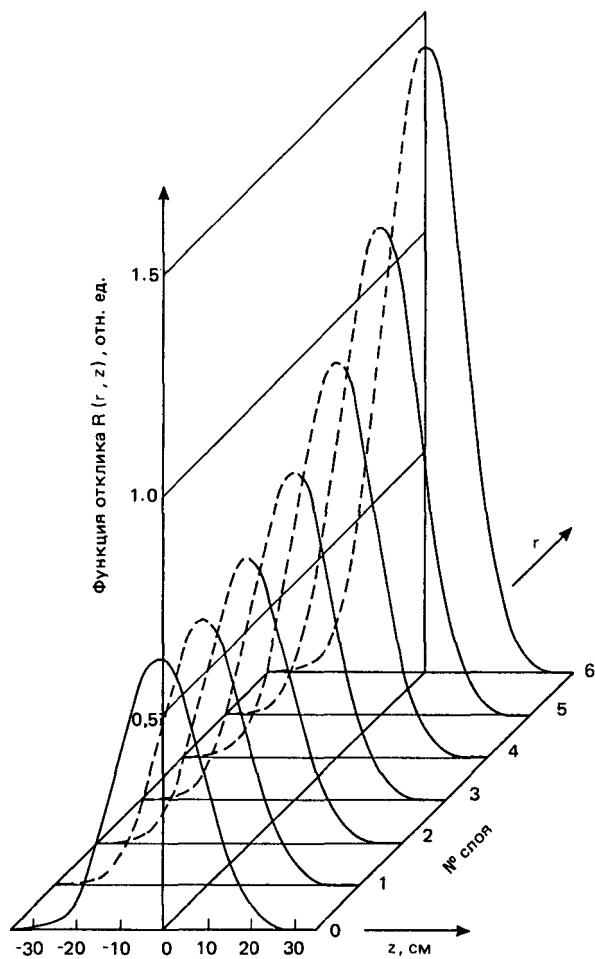


РИС. 2. ТВС ВВЭР-440 (без кадмиевого фильтра).

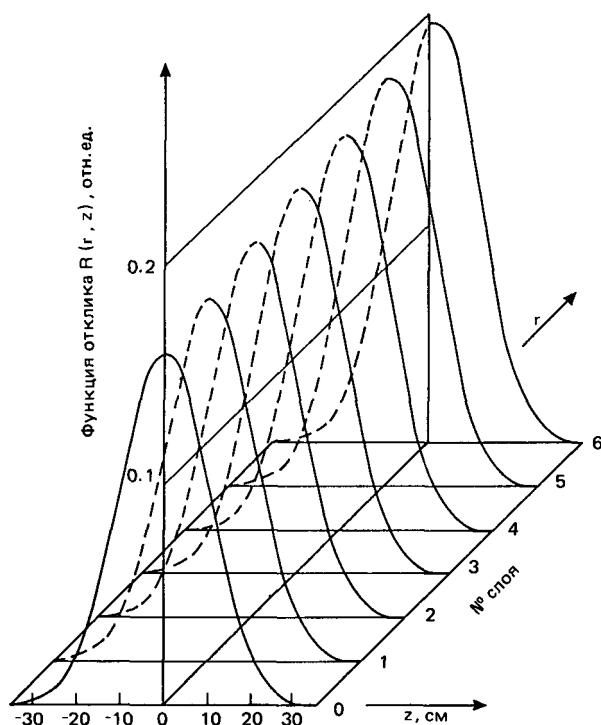


РИС. 3. ТВС ВВЭР-440 (кадмиевый фильтр).

интегрированием соответствующих аксиальных распределений, параметры которых были получены при обработке методом наименьших квадратов (МНК) экспериментальных результатов с аппроксимирующей функцией нормального распределения. Погрешность определения этих величин не превышает 5%.

В табл. III приведены значения величин  $\kappa_i$ , полученных при обработке МНК данных из табл. I и II с аппроксимирующими функциями Бесселя  $I_0(\kappa_i r)$ . Значения  $\kappa_i$  находились по критерию минимума остаточной суммы квадратов отклонений экспериментальных и расчетных значений. Погрешность определения величин  $\kappa_i$  не превышает 20%. В табл. I и II значения радиуса  $r$  приведены в сантиметрах, а значения функций  $S$ ,  $F$ ,  $R$  — в относительных единицах. Расчетные кривые пространственных распределений функции  $R(r)$  для ТВС ВВЭР-440 и ВВЭР-1000 приведены на рис. 2–4. Данные для расчетов этих кривых получены из измерений значений функций  $F$  и  $S$ . Аппроксимация кривых при обработке МНК проводилась с функциями Гаусса и Бесселя для аксиальных и радиальных распределений, соответственно.

Оптимизация условий контроля ТВС энергетических реакторов позволила получить воспроизводимость результатов контроля не превышающую 0,7%, что соответствует обнаружению изъятия двух–трех твэлов из 312 для ТВС-1000 и одного из 126 для

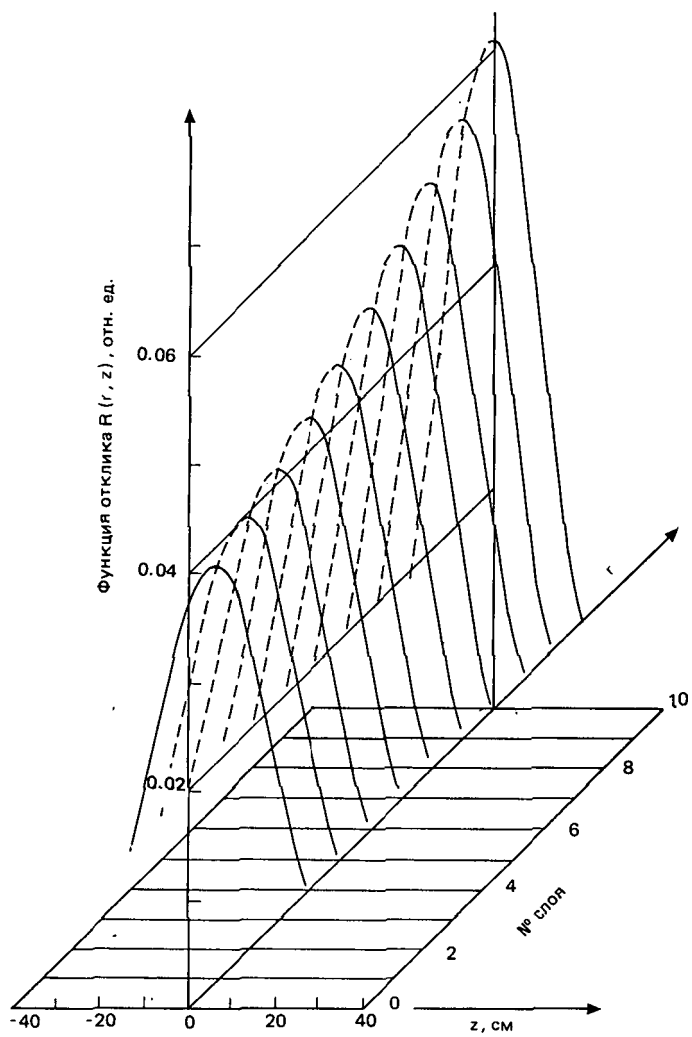


РИС. 4. ТВС ВВЭР-1000 (кадмиевый фильтр).



ТВС ВВЭР-440. Для максимальной активности гамма-источников за 10 мин циклической работы установки регистрируется около 4000 запаздывающих нейтронов вынужденного деления урана-235 в ТВС ВВЭР-1000.

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## POSTER PRESENTATIONS

IAEA-SM-293/99P

### FIELD TEST OF THE LASER SURVEILLANCE SYSTEM (LASSY) AT THE PAKS NUCLEAR POWER PLANT

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In March 1986 the IAEA made a request to the Paks Nuclear Power Plant to test the Laser Surveillance System (LASSY) at Paks. The management of the Paks NPP gave permission and promised to ensure the necessary conditions for installation of LASSY.

Because of the scheduled modification of the spent fuel cooling pool of the first unit at the Paks NPP, in July 1986 it was necessary to transfer 114 spent fuel assemblies to the second unit. It was decided that LASSY could be installed in the cooling pool of the second unit and be tested during the transfer.

To facilitate installation, the staff at Paks designed and constructed two supporting masts for fixing the two LASSY 'eyes' in adjacent corners of the pool. Each part occupied the position of a fuel assembly or hermetic container above the level of the first layer of spent fuel. It was required that the eyes be about 50 cm higher than the tops of the fuel assemblies and in fixed positions relative to the wall. It was also required that each mast be so designed that it would be stable and could be fitted to the assembly gripper and the mechanical equipment of the pool, as well as to the eyes.

Chemical analysis proved that there was no interaction between the boric acid solution in the pool and those parts of LASSY that were submerged. Before fuel transfer, the LASSY electronic equipment was set up next to the pool, and the masts needed some modification so that the equipment was fitted properly to the hermetic container and the eyes were properly fixed. The modified design could be a basis for the final version of the mast for use in cooling pools of WWER-440 type units with a compact method of storage.

The main conclusions arising from the LASSY field test are as follows:

- (a) LASSY has proved its ability to fulfil its task under operating conditions.
- (b) There is a need for some modification of the LASSY software system.
- (c) During the design of the final version of LASSY and with respect to serial production, various aspects should be taken into account:
  - Integration of the construction;
  - Installation of the masts and establishment of the electronics and computer system in accordance with the specifications of the reactor units concerned;

- Reduction of sensitivity to disturbance;
- Reduction of the time required for checking LASSY;
- Replacement with stainless steel of those parts which are in contact with water.

The field test, which lasted four months, with organizational and preparatory work included, was considered to have been successful.

IAEA-SM-293/163P

## SAFEGUARDS CHARACTERISTICS OF PRESSURIZED HEAVY WATER REACTOR FUEL

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Safeguards characteristics of KANUPP (PHWR) irradiated fuel bundles were investigated theoretically by performing detailed summation calculations of fission product and actinide inventories (AHMED, I., Rep. KANUPP-STR-85-6, Karachi Nuclear Power Plant (1986)) and experimentally through non-destructive gamma ray spectrometry (AHMED, I., HODA, Q., Rep. KANUPP-STR-81-3, Karachi Nuclear Power Plant (1981)).

The irradiation histories of three fuel bundles of low, moderate and high burnups were processed using the ORIGEN computer code (BELL, M.J., Rep. ORNL-4628, Oak Ridge Natl Lab., TN (1973)), modified suitably to treat PHWR irradiated fuel. The appropriate spectral indices necessary for defining the prevalent KANUPP neutron spectrum and subsequently averaging the neutron cross-sections were determined using the lattice code WIMS-D (ARSHAD, M., Pakistan Institute of Nuclear Science and Technology, private communication, 1984). The calculated results, which include such parameters of safeguards relevance as burnup as well as inventories of some important fission products and actinides, were found to be in good agreement with the results obtained with a fuel management computer code.

The experimental studies were conducted by measuring the fuel decay gamma spectra at different burnups and cooling times. The measured relative activities of  $^{137}\text{Cs}$  and  $^{134}\text{Cs}/^{137}\text{Cs}$  ratios were correlated with theoretically calculated burnups. Furthermore, the use of the relative activity of  $^{137}\text{Cs}$  as a measure of plutonium content in the discharged bundles was investigated.

The results of gamma scanning measurements were found to be consistent with earlier work on CANDU spent fuel. These point towards the possibility of employing empirical relationships observed between measurements and spent fuel characteristics for verification and accountability purposes.

It could be concluded from the studies that the decay characteristics of a PHWR fuel bundle depend strongly on its irradiation history and particularly on whether the bundle was irradiated in the inner or outer reactor zone, following a single or double bundle shift criterion. It has been observed that direct proportionality between the burnup and activity of the fuel is not established until after about 200 days of cooling since the last irradiation received by it. The activity or decay heat at discharge has, however, been found to be directly proportional to the exposure received by the bundle at the last irradiation position in the reactor core.

It has been verified that most of the information of safeguards relevance is contained in the energy range 400–900 keV of the decay gamma spectra of the fuel bundles. This energy range is dominated by gamma ray emissions from  $^{95}\text{Zr}$ ,  $^{95}\text{Nb}$ ,  $^{106}\text{Rh}$ ,  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$ , all of which are extensively used in the interpretation of safeguards related parameters, such as burnup, cooling time and Pu concentration in the irradiated fuel assemblies.

It has been determined that for PHWR fuel,  $^{137}\text{Cs}$  is an ideal monitor of fission product burnup and Pu content. This is particularly so because its concentration does not seem to depend on the location of the fuel in the reactor core or the power history. Moreover, gamma scanning performed to determine the  $^{137}\text{Cs}$  distribution over the fuel bundle length suggests that errors associated with its use are minimal.

IAEA-SM-293/89P

**LATEST DEVELOPMENTS AND IMPLEMENTATIONS  
OF INSTRUMENTS FOR URANIUM ASSAY  
BY DELAYED NEUTRON TECHNIQUE**

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The principle of the delayed neutron technique for uranium assay is well known. Described below are implementations of the technique with field instrumentation designed for safeguards inspections.

Two devices, SIGMA and DUCA, have been studied and developed in recent years at the Joint Research Centre, Ispra, for the Euratom Safeguards Directorate. With these devices, measurements can be made for the assay of  $^{235}\text{U}$  in uranium-thorium oxide contained in fuel pebbles (60 mm diameter graphite spheres) used in high temperature reactors (SIGMA; see CUYPERS, M., VAN DER STRICHT, E., BOURSIER, M., CORBELINI, M., in Safeguarding Nuclear Materials (Proc. Symp. Vienna, 1975), Vol. 2, IAEA, Vienna (1976) 521); and in uranium oxide as powder or pellets contained in a cylindrical capsule with a diameter of 18 mm and length of 83 mm (DUCA).

These instruments consist of:

- An irradiation zone in which four  $^{252}\text{Cf}$  sources supply a neutron flux for active interrogation of the sample.
- A counting zone which houses the  $^3\text{He}$  detectors in a polyethylene block; these measure the delayed neutron count rate from the sample.

In the DUCA device the two functions (irradiation and counting) are performed in the same zone; a bar carrying the  $^{252}\text{Cf}$  sources is moved in or out of the zone (shuffler principle).

SIGMA has been in use for more than ten years but recently some improvements were made to produce a device that is easier for an inspector to use, more reliable and able to improve the measurement accuracy to better than 0.3%. The improvements are as follows:

- All operations are made from floor level;
- The possibility of automatic measurement repetition increases accuracy;
- A modern instrument control system is used to increase the reliability of the measurements;
- The self-diagnosis system greatly facilitates instrument checking and maintenance;
- Corrected mean values, including statistical data, are computed;
- The automatic measurement evaluation includes error propagation.

A fast neutron counting chain has been developed to reduce the counting losses, above all in the first 2 s after irradiation when the count rate is high. The dead time of the system is about 1  $\mu$ s.

The hardware of the control and data handling system consists of:

- A microcomputer (SIGMA:  $\mu$ MAC 5000 analogue device; DUCA: Intel 8030) on which dedicated software has been implemented;
- An intelligent terminal (IRIS) linked to the microcomputer;
- A neutron counting chain;
- A synoptic panel;
- A low voltage power supply unit.

For DUCA a calibration formula has been developed to correlate the net total counts (I) with the content of  $^{235}\text{U}$  ( $U_5$ ) and  $^{238}\text{U}$  ( $U_8$ ). This formula takes into account the thermal flux depression in the matrix of the sample, the thermal fissions in  $^{235}\text{U}$ , fast fissions in  $^{235}\text{U}$  and fast fissions in  $^{238}\text{U}$ . The algorithm is:

$$I = aU_5^2 + bU_5 + cU_8 + dU_5U_8$$

where the calibration constants a, b, c and d have been optimized with a best fit method of linear regression for powders and pellets (with enrichment from natural to 6.4%). The agreement between declared and measured values was in every case better than 0.5% for pellets and 0.6% for powders using a simple measurement cycle. (It is planned to implement pellet and powder measurement also on SIGMA.)

In conclusion:

- (a) The system will give a very small verification time and require minimum inspector effort at the plant.
- (b) The operating system provides automatic self-diagnosis of the instrument.
- (c) The new design for some subsystems yields maximum reproducibility of results and improves the measurement accuracy.
- (d) The devices have two different fields of application:
  - SIGMA, permanently located at HOBEG (NUKEM GmbH, Hanau), has been dedicated to inspections on pebbles of the AVR and THTR reactors;
  - DUCA, which is transportable, will be used for inspections on powder or pellets of LWRs, with sampling of bulk quantities.

IAEA-SM-293/121P

**AN ASSAY SYSTEM FOR FUEL ASSEMBLIES  
FROM MATERIAL TESTING REACTORS**

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Material testing reactor (MTR) type fuel assemblies have attracted worldwide safeguards interest because they contain significant quantities of high enriched uranium which can be separated with elementary chemical processing. They are moreover employed in a research oriented atmosphere not normally associated with high security, and therefore have been considered a high risk from the safeguards/terrorist threat standpoint.

Acting under directives of the United States Department of Energy, we have developed a non-destructive assay system which can quickly determine ( $\sim 10$  min) with a degree of assurance approaching certainty (approaching  $5-6\sigma$ ) whether an amount of material equivalent to one plate of an element (about 5% of the total fuel loading) is missing from the element. The assay of a new fuel loading for a research type reactor can be completed within a few hours of fuel receipt, thus minimizing exposure before the elements are put into the (relatively much more safeguards secure) reactor core. The system moreover operates under the handicap that no standard or reference fuel element is available at the time the new fuel elements are being assayed. The assay of the new fuel elements takes place during the same period allotted to mechanical and visual quality assurance acceptance testing, and the two activities complement rather than intrude on one another.

The assay is based on a fast ( $\sim 20$  ns resolution time) true coincidence (not correlation) formed between three plastic scintillator-photomultiplier detectors which monitor the prompt radiation from fissions induced in the MTR element by neutrons from an uncorrelated subthreshold ( $\alpha$ , n) source (AmLi,  $\sim 10^6$  neutrons/s). Standard nuclear instrumentation module (NIM) electronics are used to process the fast photomultiplier signals ( $\sim 5$  ns) into logical quantities which yield the true triple and double coincidences, the singles, and the chance triple and double coincidences. Apart from high voltage bias and scaler modules, only four multiple function NIMs are required. The scalers (two quad units) allow direct interfacing to micro- or minicomputers. The triple chance coincidence background is typically less than 2% and the double chance coincidence less than 7% of the corresponding true count rates. Background (i.e. count rates with only a dummy element present) is negligible.



While the basic scenario involves detection of about 5% of the fuel missing, because there is so much shielding and self-shielding inherent in the way MTR elements are constructed (about twenty 'flat' plates, approximately 50 cm  $\times$  10 cm, stacked parallel with about 2.5 mm spacing between them), there is actually only a roughly 3% effect visible externally when a fuel plate is replaced with a dummy plate. Establishing a roughly 3% deficit reliably requires measurement to within a fraction of that, i.e. less than 1%. Thus a large effort was put into minimizing sources of variance such as geometric positioning and electronic drift.

To realize the advantages of photomultiplier detectors (high count rate capability, the possibility of using fast coincidences to reduce background, and relative noise immunity), their tendency to drift had to be overcome. It was found possible to do this by referencing all measurements to a small  $^{252}\text{Cf}$  source ( $\sim 370$  Bq (0.01  $\mu\text{Ci}$ )), which produced a true count about ten times that of an element. This offered a means both to compare the standard fuel element with any new element and to monitor the behaviour of the electronics over a long period independently of the AmLi source.

Drift involves changes in amplification, bias settings, etc., which cause change in the effective detector efficiency. It was therefore of interest to test the 'invariant ratio' technique of using as assay signatures quantities which are, to first order, independent of efficiency. This would provide a way of reducing the variance due to drift.

In particular, a quantity similar to the ratio of the product of the three double coincidences to the square of the triple coincidences was compared with the triple coincidences. Preliminary results indicate that this quantity,  $T_2^3/T_3^2$ , was at least as good as, and sometimes about 2.5 times better (i.e. smaller  $\sigma$ ) than the triple coincidences,  $T_3$ , alone. However, the nuclear material accounting results are based on  $T_3$  alone.

Two independent expressions for the pulse width (resolution) in the logic system electronics were derived in terms of the scaler readings. These can be compared with the pulse width that was set for the electronics when the initial adjustment was made. This furnishes a check on the system performance independent of sample, standard or normalization source.

IAEA-SM-293/107P

## A NEW NON-DESTRUCTIVE MEASUREMENT TECHNIQUE FOR ACCURATE NUCLEAR SAFEGUARDS ANALYSIS

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A new non-destructive measurement technique based on neutron activation delayed fission neutron counting has been developed at the Demokritos Nuclear Research Center for accurate analysis of nuclear safeguards samples. The technique is a combination of cyclic activation, long intermediate cooling of a sample and simultaneous irradiation of the following samples, and special data processing (PAPADOPOULOS, N.N., in Proc. 7th Int. Conf. on Modern Trends in Activation Analysis, Copenhagen, 1986, Risø Natl Lab., Roskilde (1986) 1351; in Proc. Int. Mtg on Nuclear Analytical Methods, Orléans, 1986).

Cyclic activation improves counting statistics, without increasing the count rate, to prevent pulse pile-up, and reduces sample irradiation position uncertainty and timing inaccuracy. Long intermediate sample storage allows for enough cooling to prevent radiation buildup and, again, pulse pile-up. During this period other samples can be measured to shorten the overall analysis time. By use of subsamples the weighing and inhomogeneity error can be reduced and the total sample mass increased for better counting statistics without exceeding the activity limits.

Instead of mixing the signals from the  $\text{BF}_3$  detectors to one counting chain (preamplifier-amplifier-discriminator-counter), the use of more counting chains will reduce the count rate of each channel. Thus the total counts can be increased without dead time losses, improving again the counting statistics and hence the precision of the measurements.

Subsamples and substandards are measured alternately. The counts from the partial chains and from the subsamples and substandards are added to obtain the total counts from each sample and standard separately. Then the total sample and standard counts of the measurement cycles are added separately and compared for absolute determination. By this procedure also long range flux variation errors are eliminated. Thus a precision (relative standard deviation) of less than 0.03% and an accuracy of better than 0.1% have finally been achieved, and there is the possibility of further improvement.

Besides the small measurement uncertainty, which is competitive with that of other destructive methods such as mass spectrometry, this non-destructive analytical technique has all the advantages of the neutron activation delayed neutron counting method, e.g. high analytical speed, negligible matrix interference, wide concentration range determination, simplicity of sample preparation, a great possibility for automation, low personnel requirements and low operating cost.

Since sensitivity is usually not a problem in the analysis of nuclear safeguards samples, the efficiency of the system might be sufficient for portable neutron sources such as  $^{252}\text{Cf}$  or Am-Be to be used for sample activation, instead of a nuclear reactor. Thus the technique could also be used in field applications and in general in cases where a reactor is not available, although usually samples can easily be sent to reactor installations (PAPADOPOULOS, N.N., J. Radioanal. Chem. **72** (1982) 463; in Proc. 1st Balkan Conf. on Activation Analysis, Varna, 1985; in Proc. Int. Symp. on Nuclear Analytical Chemistry, Halifax, Canada, 1985; in Significance and Impact of Nuclear Research in Developing Countries (Proc. Symp. Athens, 1986), IAEA, Vienna (1987) 339).

Thus it is believed that this new non-destructive technique will fill a gap in accurate nuclear safeguards analysis, as well as in nuclear reference material certification, where also high accuracy is required.

IAEA-SM-293/13P

# USE OF $^{137}\text{Cs}$ CALIBRATION SOURCE IN EVALUATION OF BOILING WATER REACTOR FUEL BURNUP

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A method for evaluating the burnup (BU) of boiling water reactor (BWR) spent fuel was investigated by using a novel type of  $^{137}\text{Cs}$  calibration source. The source is constructed to fit in the fuel handling fixtures of all the BWR type power plants in Sweden and Finland. It can be used also in the interim storage facilities for spent fuel, CLAB in Sweden and TVO-KPA-STORE in Finland.

The source is covered by a watertight steel cylinder which is fixed inside a 0.65 m long piece of ASEA-ATOM type BWR fuel channel. Inside the cylinder there is a 37 GBq  $^{137}\text{Cs}$  pellet fixed to a wagon which can be driven up and down by means of a stepping motor. By moving the source, the repeatability of the geometric positioning is attained. The amplitude and scanning speed are controlled by a remote control unit. The apparatus, shown in Fig. 1, is easy to handle and decontaminate.

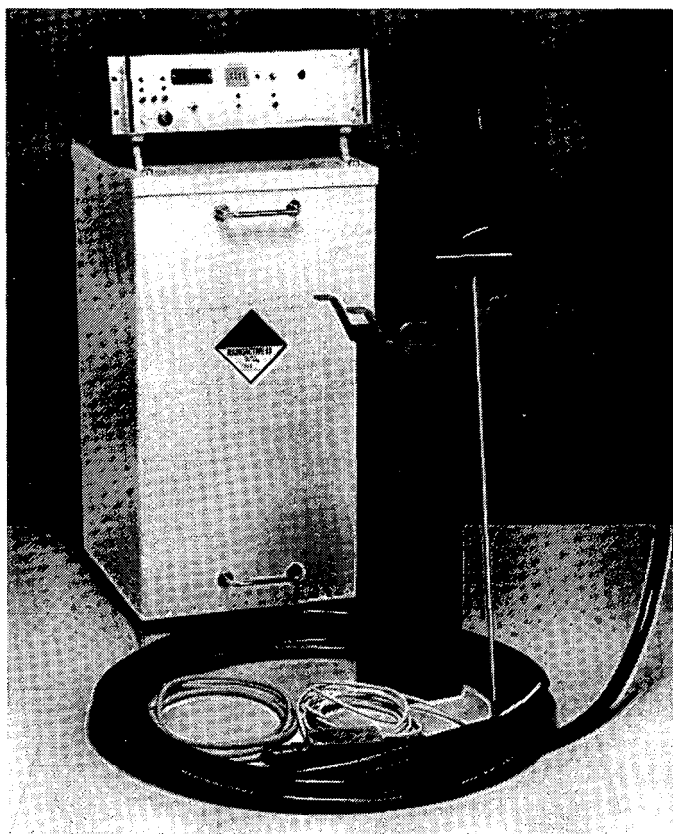


FIG. 1. The  $^{137}\text{Cs}$  calibration apparatus. Right: source inside a piece of BWR fuel channel; left: remote control unit (top) and transport box.

The source can be transported in a custom made box (45 cm  $\times$  45 cm  $\times$  70 cm) under Category II — Yellow.

In repeated measurements the precision of the new calibration source was found to be  $\pm 1.7\%$ . Use of this source makes it possible to calibrate the whole measurement chain and to compare the data measured in different geometries. A typical calibration time is 15 min, including source handling.

In recent measurements, a Westphal loss free counting (LFC) system was used in connection with an ND66 multichannel analyser for scanning of fuel assemblies. By use of LFC, a real time correction of counting losses is performed.

For BU verification 21 assemblies with mean BUs from 14 to 31 MW  $\cdot$  d/kg U and cooling times from 200 to 1500 d were scanned on each of the four corners. The total time needed per assembly was typically half an hour. The measured  $^{137}\text{Cs}$  data

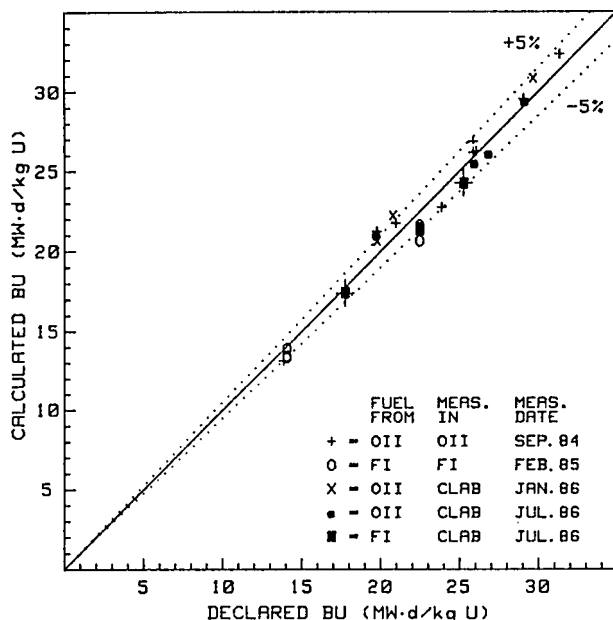


FIG. 2. Operator declared BU compared with calculated BU based on  $^{137}\text{Cs}$  activity measurements (FI: Forsmark Unit I; OII: Oskarshamn Unit II).

were corrected for radioactive decay, self-absorption and inhomogeneous Cs distribution. The BU was calculated by use of the arithmetic mean for the four corners and the earlier defined relation between BWR fuel BU and  $^{137}\text{Cs}$  activity (AF EKENSTAM, G., in Proc. 6th ESARDA Annual Symp. on Safeguards and Nuclear Material Management, Venice, 1984, CEC Joint Res. Centre, Ispra (1984) 327).

The BU calculated from the measured data is shown plotted against the declared BU in Fig. 2. Error bars reflect the precision of measurements for single assemblies. The  $\pm 5\%$  deviation lines are also plotted. A more detailed description of the method will be published elsewhere (AF EKENSTAM, G., TARVAINEN, M., Rep. STUK-A52, Finnish Centre for Radiation and Nuclear Safety, Helsinki (in press)).

The measurement system has been developed for use by the Swedish and Finnish national safeguards authorities for verification of spent fuel BU.



**SAFEGUARDS FOR  
HEAVY WATER  
(Session 13)**

**Chairman  
T. BEETLE  
IAEA**





# SELECTION OF A SAFEGUARDS APPROACH FOR THE ARROYITO HEAVY WATER PRODUCTION PLANT

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## Abstract

### SELECTION OF A SAFEGUARDS APPROACH FOR THE ARROYITO HEAVY WATER PRODUCTION PLANT.

In preparing for the application of IAEA safeguards at the Arroyito Heavy Water Production Plant in Argentina the Secretariat evaluated several alternative safeguards approaches from the aspects of safeguards effectiveness, intrusiveness, availability of instruments and efficiency. It was concluded that a safeguards approach based on deuterium material balance closing, combined with the monitoring of some key process parameters, can be expected to provide the required safeguards effectiveness in the most economical and least intrusive way. The deuterium intake is measured in the ammonia flow before the first enrichment tower. The flow of pure  $D_2O$  is measured on the product side. Key plant parameters are used to determine plant configuration, losses and changes of the deuterium inventory. All necessary information is obtained from in-line measurement equipment specially installed for safeguards purposes, or from plant instrumentation. The information is collected and processed in a dedicated on-site safeguards computer on a continuous basis. The concept and equipment will be developed, tested and installed in close co-operation with the plant designer and the plant operator in a multi-phase programme that is harmonized with the plant construction and commissioning.

## 1. INTRODUCTION

On 14 October 1981 the IAEA and Argentina concluded an Agreement for the Application of Safeguards in Relation to the Heavy Water Production Plant at Arroyito. Since this is the first agreement with a State on the application of IAEA safeguards at a heavy water production plant, and since the Arroyito plant contains a combination of technical design features and components which is unique to IAEA safeguards, the implementation of the provisions of the Agreement requires considerable development effort. In particular, a safeguards approach is to be developed in the light of:

- (1) Requirements for effective safeguards
- (2) Prudent management practices for the economic and safe construction and operation of the plant
- (3) Availability of instrumentation and equipment for measurement and data handling

- (4) Anticipated schedule for plant construction and operation
- (5) Efficient use of Agency and Argentinian resources.

This report describes the safeguards approach envisaged for the Arroyito plant.

## 2. PROCESS DESIGNATION

The plant is designed to produce 250 t of fully enriched heavy water annually. It is located at Arroyito near Neuquen, nearly 1000 km southwest of Buenos Aires.

In the Arroyito plant deuterium depleted ammonia is equilibrated with purified natural water (Step I), thereby increasing the deuterium concentration in the ammonia. In several sequential enrichment towers the deuterium concentration in the ammonia is further increased in a monothermal counter-current exchange process with gaseous hydrogen (Step II). The enriched ammonia is cracked and most of the deuterium-hydrogen mixture is returned to the enrichment towers. The product from the last enrichment tower is fully enriched  $\text{ND}_3$  which is cracked to  $\text{D}_2$  (and  $\text{N}_2$ ). A small fraction of the  $\text{D}_2$  is withdrawn from the process and converted with dry air to  $\text{D}_2\text{O}$  product (Step III). The rest of the  $\text{D}_2$  is refluxed into the enrichment process. The depleted  $\text{H}_2$  coming from the enrichment towers is used to synthesize (depleted) ammonia which is returned to Step I.

## 3. REQUIREMENTS FOR EFFECTIVE SAFEGUARDS

The requirements for effective safeguards were established as follows:

(1) The Agency should be able to provide assurance that there were no unrecorded significant withdrawals or removals of "heavy water". According to the agreement, "heavy water" means water, hydrogen and hydrogen compounds in which the ratio of deuterium atoms to hydrogen atoms exceeds 1 to 5000. Since the ratio of deuterium atoms to hydrogen atoms already exceeds 1 to 5000 in the first enrichment tower, the safeguards approach would have to cover all intermediate flows.

(2) For planning purposes, 20 t  $\text{D}_2\text{O}$ , or its equivalent, would be considered a significant quantity and a detection probability of 90% would be considered adequate.

(3) The Agency should be able to draw conclusions at least annually. To identify differences between the results of the safeguards system and the operator's statements at an early time, and to avoid the sudden appearance of large anomalies, preliminary conclusions should be available more frequently.

#### 4. REQUIREMENTS FOR ECONOMIC AND SAFE CONSTRUCTION AND OPERATION OF THE PLANT

According to the agreement the safeguards procedures should be implemented "in a manner designed to be consistent with prudent management practices required for the economic and safe conduct" of plant construction and operation. The safeguards approach, therefore, should use instruments and techniques which do not negatively influence the production rate or costs, and which do not jeopardize the safety of the plant. The Agency has engaged the services of the designer of the plant and is co-operating with the Argentinian authorities to evaluate and minimize the potential impact of safeguards measures on the economy and safety of the plant.

To the extent possible the Agency will use data available from plant instrumentation or from non-intrusive "clamp on" flow and density meters. Some safeguards equipment will be installed into by-pass lines, so that the plant operation will not be hindered in the event of a failure or leakage in safeguards equipment. The safety standards used for the safeguards equipment are identical to those being used for the rest of the plant.

Co-operation with Argentina is essential. During the development period there has been close contact with the relevant Argentinian experts and authorities, and these authorities have agreed to the installation of the required equipment. This co-operation will continue, and the progress of the project and results of tests will always be jointly evaluated. Only after the approach has been successfully tested and demonstrated will a permanent facility attachment be negotiated.

#### 5. AVAILABILITY OF INSTRUMENTS AND CO-ORDINATION WITH PLANT CONSTRUCTION

Whereas some of the information required for the safeguards approach can be obtained from plant instrumentation, several important parameters are to be determined by independent safeguards instruments. This occurs where the relevant information is not available from plant instruments,

or where the quality or authenticity of operational information is insufficient. Safeguards equipment will be installed to measure flows of gases and liquids, densities of liquids and the deuterium concentration in ammonia. Feasibility studies have shown that, while instruments necessary to establish the required information are in principle commercially available, modifications and adaptations will be necessary.

The construction of a chemical plant of the size and complexity of the Arroyito plant in a developing country is a difficult task requiring a high degree of flexibility in time schedules. Whereas it would be unwise to install the safeguards equipment at an early phase of the plant construction, certain provisions for the later connection of safeguards equipment are necessary prior to leak and pressure testing of the pipework and before the plant is filled with hydrogen and ammonia. The Secretariat has secured the financing of the safeguards equipment for the Arroyito plant through a waiver by the IAEA Board of Governors of the requirement for an annual budget and through the establishment of a special multi-year fund for this project.

## 6. THE SAFEGUARDS APPROACH

To be able to select an efficient and effective safeguards approach for the Arroyito plant a number of alternative safeguards approaches were studied:

### 6.1. Verification only of the product material in Step III (deuterium-heavy water converter)

This approach would measure the deuterium inputs and outputs of the heavy water converter. It is not able to detect the withdrawal of partially or fully enriched deuterium gas ( $D_2$ ) or partially or fully enriched ammonia ( $ND_3$ ) prior to the measurement of the input to the Step III. Thus, this approach was rejected because, on the basis of the measurements of the inputs and outputs of this step, the Agency would not be in a position to verify any statement concerning the operation of the most sensitive parts of the plant (enrichment towers).

### 6.2. Verification of the product material only in Step III plus the application of seals and surveillance to those other parts of the plant that handle fully enriched material

To mitigate the weakness of the first approach, the possibility was considered of adding seals and a surveillance

system sufficient to provide assurance that no fully enriched material was removed from the plant without the Agency's knowledge. Three factors led to the rejection of this option:

- (i) Fully enriched material is contained in many plant units besides the deuterium-heavy water converter and enrichment towers, and these units are dispersed over a large area of the plant. The installation of surveillance systems over such a wide zone would be an extremely difficult task.
- (ii) Sealing of sampling points, drains, and other locations where material could be removed would require numerous seals and the presence of an inspector whenever they needed to be opened. The inspection effort associated with this would be prohibitive.
- (iii) The approach would have no capability of detecting the removal of material enriched to 1-20% deuterium concentration.

### 6.3. Material balance accounting

As applied to a heavy water plant, the purpose of material balance accounting would be to compare the amount of deuterium extracted from a given feed stream with the deuterium contained in the product, with corrections for losses and changes in the inventory. The losses and the inventory changes would be determined on the basis of process monitoring data.

The Agency investigated numerous material balance alternatives in which the feed streams of interest are:

- (i) The water feed to the plant minus a depleted water stream;
- (ii) The ammonia feed stream to the first enriching tower, minus a depleted ammonia stream;
- (iii) The ammonia feed stream to the second or third enriching tower, minus a depleted stream consisting of a mixture of nitrogen, hydrogen, and ammonia vapour.

For each of these cases, there is no theoretical reason why a material balance approach could not result in obtaining the desired level of safeguards effectiveness. However, practical reasons of economics, instrument availability, plant design, and plant safety considerations led to the rejection of all alternatives except for (ii), the ammonia feed stream to the first enriching tower.

The Agency was unable to find an acceptable technical solution to measurement problems posed by the other possible material balances. The difficulties arise from the complexity

of some of the streams considered, in particular of the flow of synthesis gas out of the material balance area and the flows of catalyst containing ammonia into and out of the material balance area. For the synthesis gas stream, the measurement is further complicated by the fact that it contains a significant amount of ammonia. Thus, the stream is a mixture of three components, two of which carry deuterium with different deuterium concentrations.

#### 6.4. Process monitoring

Here the basic concept is that the Agency should use plant control signals together with plant design and the laws of physics and chemistry to establish how much deuterium is extracted from the feed stream at the plant input, what the plant inventories are, and what the plant losses are.

Analysis indicated that process monitoring could be implemented with a minimum of additional installations. The plant designer evaluated the expected performance of a process monitoring system and determined that it could achieve performance levels compatible with the goals established by the Agency. Similar analyses for process monitoring evaluations restricted to the second and third stage enrichment towers indicated that, for plausible plant operating conditions, these process monitoring evaluations could lead to statistical uncertainties that approach the Agency's targets in the best case, but that could be three to four times larger for other operating modes. Thus, the key process monitoring system is considered to be that established about the ammonia water equilibration (Step I).

The Agency did not consider it reasonable to use process monitoring as a sole safeguards technique because to do so would require a large number of signals to be transmitted to the Agency's computer, and the Agency to be able to establish the authenticity of all these signals. In addition, the process monitoring system would require highly sophisticated evaluation algorithms, which could only be developed on the basis of several years of plant operation experience and may only be expected to provide reliable results when the plant is operating in a stable mode.

#### 6.5. Combination of material balance accounting and process monitoring

To provide an independent system for safeguards verification and to compensate for the weakness of the process monitoring system in non-stable operating modes, the Agency has

selected as its base case safeguards approach a material balance system that is complemented by some process monitoring.

The combination of material balance evaluation and process monitoring provides the Agency with a robust safeguards system, key elements of which may be independently authenticated, and which is expected to provide satisfactory results for most plant operating modes. If the instrumentation selected performs according to specifications, and if the plant operates in a reasonably stable mode, the Agency should be able to attain the safeguards goals established for the plant. If the plant does not operate in a reasonably stable mode, the safeguards system is nonetheless expected to provide results that are acceptable despite its degraded performance. The reason is that unstable operation will result in smaller production capacity. For example, if the plant only operates for half the year, the safeguards system can afford a relative uncertainty that is twice as high as for full production since the goal quantity is a fixed amount.

The combination of material balance accounting and process monitoring is not a redundant combination, since the material balance evaluation requires the use of process monitoring data in order to ascertain process inventories (normally well below 20 t) and the process losses. The capability of the process monitoring data to provide an estimate of the plant's extraction is an additional benefit. There are also additional benefits in terms of increasing the safeguards system's robustness and in providing early warning regarding possible discrepancies, thereby avoiding situations where investigations need to be carried out that might affect the plant's operation. The Agency is fully aware that there exists no specific operating experience for some of the main components and for the complete system and that adjustments to the approach may be necessary at a later date.

## 7. CHARACTERISTICS OF THE APPROACH BASED ON MATERIAL BALANCE ACCOUNTING AND PROCESS MONITORING

During routine, steady state plant operation, the Agency will have available to it a set of data generated by instrumentation installed especially for safeguards purposes and data generated for plant control purposes and made available to the Agency. These data will be transmitted to a dedicated Agency computer. The computer will contain physical constants, evaluation algorithms and plant characteristics that are sufficient to perform the following evaluations:

- (1) Data screening program - designed to detect data that are out of pre-established ranges and sudden jumps for any of the

data collected. The program will also identify the plant's operating configuration and select the appropriate process monitoring evaluation scheme.

(2) Plant stability analysis - detecting operating periods during which the main parameters of the plant are stable and can be used for calibration or recalibration purposes. The program will establish estimates of the losses and the extraction for the plant as a whole and for each of the 2nd and 3rd stage units.

(3) Calibration of vessels for deuterium inventory determination - part of the program will be used to establish the actual inventory of certain vessels at the time of plant startup. Afterwards these data will be used together with levels, volumes, and concentrations to establish inventory estimates for the plant that are valid for a particular time.

(4) Process monitoring, extraction and loss forecast - based on measurements made at the time this program will compute, on a real-time basis, the extraction and the loss estimates.

(5) Isotopic balance extraction forecast - based on measurements of the flow and concentration of the equilibrated ammonia and the depleted ammonia.

(6) Consistency checks - a variety of intercomparisons are made between data made available to the Agency computer from the plant computer and from Agency instruments.

(7) Executive summary program - this program will compute MUF and sigma-MUF over a selected period. The target of the program is to provide an output corresponding to the Agency's requirements in normal, routine situations as well as to detect situations that need investigation.

The period over which the summary program will compute MUF and sigma-MUF is expected to be around one to two weeks. Thus, in routine operation, the Agency will have available a weekly or bi-weekly comparison between the amount of heavy water measured at the output and the amount that should have been produced on the basis of determinations of extraction and losses.

Because of the interrelationship of the material balance and process monitoring determinations, if part of the safeguards system fails the safeguards system is expected to continue to operate on the basis of the remaining element.

Leak rates by themselves, whether high or low, are not factors that should affect significantly the performance of the



safeguards system described above. The plant designer has identified a procedure by which the Agency may ascertain the plant leak rates during test runs and isolate the fraction of the total leak which arises from different enrichment zones in the plant. A sudden significant increase in the leak rate would be detected by the periodic material balances, or by their sequential analysis, or by jumps in operating data. Sudden increases in the leak rate could indicate a diversion or a pipe break. A key rationale for frequent material balance and process monitoring MUF evaluations is to identify such changes at an early time in order to permit the Agency to investigate discrepancies before they accumulate to a significant amount.

While it is premature to predict the accuracy of the proposed safeguards system in detecting changes of operating parameters and leak rates, it is foreseen that the input and output measurements will be performed with relative uncertainties of 1 to 2%. This corresponds to a determination of the inputs and outputs over the course of a month to better than 1 t of heavy water equivalent. If the inventory can be determined to better than 10%, then the Agency should be in a position to detect changes in the leak rate, or removals from the plant, over the course of one month of 3 t of heavy water equivalent.

It is foreseen that at each monthly inspection the Agency would have records that have been produced by the Agency computer showing the material balances for preceding months as well as recent abnormal events or discrepancies that could accumulate to a significant quantity when extrapolated over a longer future time period. It has been proposed that the Agency computer would provide a message to the operator when a large discrepancy was detected. This message could be transmitted to the Agency and lead to inspection at short notice.

It may be seen from the description above that the selection of a base-line approach for planning of safeguards at the heavy water plant has resulted from the comparison of specific vulnerabilities together with the evaluation of technical feasibility.

## 8. IMPLEMENTATION

To implement IAEA Safeguards at the Arroyito plant the IAEA has adopted a 4-phase programme. At present, phase 1 is being carried out, during which all modifications to the

pipework of the plant necessary for the later installation or connection of safeguards equipment will be made. During the second phase (1987/88) the equipment required for the test and demonstration programme will be procured and prepared. As soon as the plant starts operating (1988) the concept, programmes and instruments will be tested and demonstrated (phase 3). At the end of this phase the results of the tests will be jointly evaluated with the Argentinian authorities and decisions will be made on the composition of the final system. It is expected that the final system will be highly instrumented and automated. After agreement has been reached on the permanent safeguards system, the installations will be completed and improved, the tamper resistance will be increased and routine procedures will be implemented (phase 4). It is expected that the permanent safeguards system will require not more than one inspection visit every month.

## EVALUATION OF HEAVY WATER (D<sub>2</sub>O) MATERIAL BALANCE IN POWER REACTORS

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### Abstract

#### EVALUATION OF HEAVY WATER (D<sub>2</sub>O) MATERIAL BALANCE IN POWER REACTORS.

The experience of implementing an integrated safeguards scheme for heavy water inventories in power reactors is reported. This scheme has elevated the safeguards status of D<sub>2</sub>O to a level similar to nuclear material safeguards. Selected results of the techniques used to verify the different components of the material balance are displayed. These verification techniques include volume determination; weight determination; non-destructive assay; sampling and analysis; and criticality check. They are combined in both attribute and variable mode according to a preset strategy. Over the material balance period the systematic and random components of error associated with the quantities forming the material balance equations were determined. The results of the final integration of the inspectors' and operators' measurement uncertainties and their propagation to estimate the limits of accuracy are reported.

### 1. INTRODUCTION

Heavy water (D<sub>2</sub>O) in production plants and power reactors is subject to IAEA safeguards under certain agreements. The NPT type agreements, however, place no importance on this material and require no measures to be taken when implementing NPT attachments in nuclear facilities that might use D<sub>2</sub>O. This is because, in NPT situations, all nuclear material is subject to safeguards, and D<sub>2</sub>O is only strategically significant when used with the appropriate type of nuclear material. The safeguards requirements to satisfy the relevant non-NPT agreements that might deal with D<sub>2</sub>O started with records auditing. As safeguards philosophies developed and the importance of integrated fuel cycles activities became apparent, the general trend shifted towards the necessity of physical inventory verification of D<sub>2</sub>O in situations where some of the fuel cycle facilities are not under safeguards [1].

This paper aims to describe and report the results of the different techniques used to achieve independent quantitative statements about heavy water in the different components of the

material balance. The present work also attempts to establish reference values for the measurement uncertainties for closing a material balance for D<sub>2</sub>O, which are considered achievable in practice.

## 2. SAFEGUARDS AND STATISTICAL PARAMETERS

Unlike nuclear material, the main parameters which define the safeguards objectives, i.e. significant quantity and detection time, are not defined for heavy water. It could be assumed, however, that the significant quantity is about 20 t. This quantity of D<sub>2</sub>O is enough to achieve criticality in a small heavy water power reactor capable of producing one significant quantity of plutonium annually. The detection time for D<sub>2</sub>O in this case will be one year.

One physical inventory verification (PIV) per year coupled with several interim inspections to verify the other components of the material balance (i.e. shipments, receipts, shipper/receiver (S/R) difference, and losses) will, therefore, meet the safeguards objectives. This is the case for a reactor that constantly operates between two PIVs (with the exception of occasional short shutdown periods), provided that the operation mode of the reactor is established during interim inspections. This activity is necessary to ensure the presence of the previously verified amount and quality of D<sub>2</sub>O in the system at all times. For reactors with extended shutdown periods, other considerations exist related to the concept of material misuse (D<sub>2</sub>O in this case). Stepping up the activities in some of the interim inspections to include quantitative verification of the amounts of heavy water in the shutdown reactor system is an adequate measure against material misuse.

The inventory of D<sub>2</sub>O in the reactor is determined by measuring the quantities in all the reactor vessels. The inventory of D<sub>2</sub>O in containers outside the reactor is determined from measurements on random samples of containers. The parameters for sample size calculations are:

Detection probability:  $1 - \beta \geq 0.90$   
False alarm probability:  $\alpha = 0.05$   
Significant quantity: SQ = 20 000 kg D<sub>2</sub>O  
Accounting verification goal:  $AVG = 2.93 \sigma A$   
 $\sigma$  = relative standard deviation of MUF  
A = maximum of annual throughput or inventory  
Number of containers: N  
Precision of measurement methods.

Sample sizes for attribute tests are based on SQ, and sample sizes for variables tests are based on AVG.

### 3. COMPONENTS OF THE MATERIAL BALANCE

#### 3.1. Inventory

Heavy water inventory in a power plant is the sum of the D<sub>2</sub>O quantities in the following systems or locations:

- moderator system (MS)
- primary heat transport system (PHTS)
- upgrading system
- downgraded D<sub>2</sub>O stocks
- reactor grade D<sub>2</sub>O store.

This situation can be regarded as similar to bulk handling facilities in which nuclear material inventories in liquid form appear in process containers or storage tanks.

Usually the moderator and primary heat transport systems have D<sub>2</sub>O with different isotopic compositions and tritium levels. A wide spectrum of D<sub>2</sub>O isotopic composition will form the inventory of the upgrading system and the downgraded D<sub>2</sub>O stocks. Reactor grade D<sub>2</sub>O will have fairly consistent isotopic composition at 99.8%, either as fresh receipts or as a result of the upgrading process.

Inspector's values for inventories are compared with the operator's values in statistical tests.

#### 3.2. Shipments

Occasionally, downgraded D<sub>2</sub>O collected from the moderator or primary heat transport systems might be sent for upgrading outside the plant. Shipments may also take the form of reactor grade heavy water for use in other heavy water power plants under safeguards.

#### 3.3. Receipts

Receipts in the form of reactor grade fresh D<sub>2</sub>O to compensate for operational losses are the normal transaction in this component of the material balance. Some receipts from outside the plant upgrading system might also occur.

### 3.4. Shipper/receiver (S/R) difference

S/R values are established for shipments and receipts. For the current analysis S/R differences were taken as zero because the operator accepts the shipper's values for received D<sub>2</sub>O.

### 3.5. Losses

The losses are D<sub>2</sub>O that is downgraded to the point where it is not economical to upgrade it, and D<sub>2</sub>O lost through evaporation. When the latter is not measured it is a component of MUF. Some unmeasured losses in liquid form due to leakage may also occur. Reference values for annual losses from similar reactors are considered in assessing the magnitude of MUF.

## 4. ATTRIBUTE VERIFICATION

The safeguards scheme is a combination of records auditing, comparison of records with reports, and the standard Agency verification methods as applied to flow and inventory verification.

For records auditing, a set of records capable of providing the following information must be established:

- (a) Accounting records showing quantities of heavy water (D<sub>2</sub>O) received at, or shipped out of, the facility;
- (b) Accounting ledger(s) and its (their) supporting operating records showing the quantities of D<sub>2</sub>O in the systems and in the store, and documenting all intersystem transactions (including upgrading system);
- (c) Results of isotopic composition analysis of D<sub>2</sub>O in the different locations at the facility;
- (d) Detailed data of physical inventory taking (PIT), e.g. data related to PIT procedures, calibration charts, measurements and calculations, including corrections for temperature, pressure and isotopic composition;
- (e) Data related to losses, MUF and S/R difference;
- (f) Data related to the accuracy of the operator's measurement systems, and the limits of accuracy of his quantitative physical inventory results.

The verification methods are combined in a way that allows both variable and attribute verifications of D<sub>2</sub>O inventories during PIVs. They are also used during interim inspections to ensure the continuous operation of the reactor and to verify inventory changes.

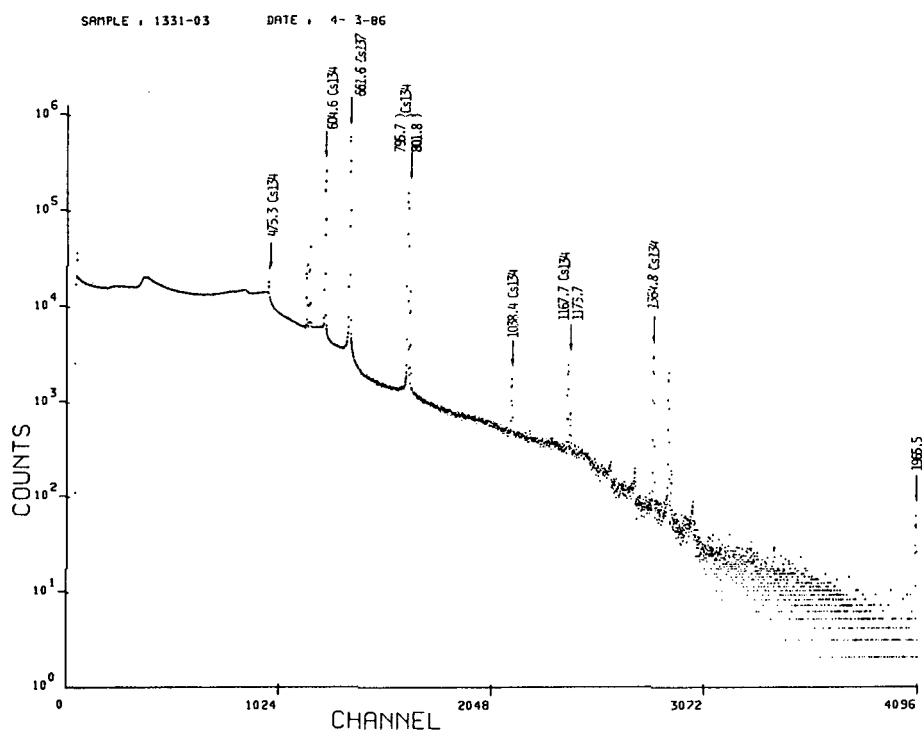


FIG. 1.  $D_2O$  gamma spectrum (PHTS).

For attribute verifications the following methods are used:

- (i) For the moderator and primary heat transport systems: Non-destructive assay, criticality check for verification if the difference between the minimum quantity required for criticality and the quantity stated by the operator is less than 1 SQ, and power check.
- (ii) For other locations (including downgraded  $D_2O$ , stored reactor grade  $D_2O$  and upgrading system): Item counting, tag check, and non-destructive assay, on a random sample of containers.

Non-destructive assay through analysing the  $\gamma$ -signatures of heavy water is an area which proved to be very effective in safeguarding  $D_2O$ . Figures 1 and 2 show the  $\gamma$ -spectrum emitted from  $D_2O$  collected from the inventories of the primary heat transport system (PHTS) and from the

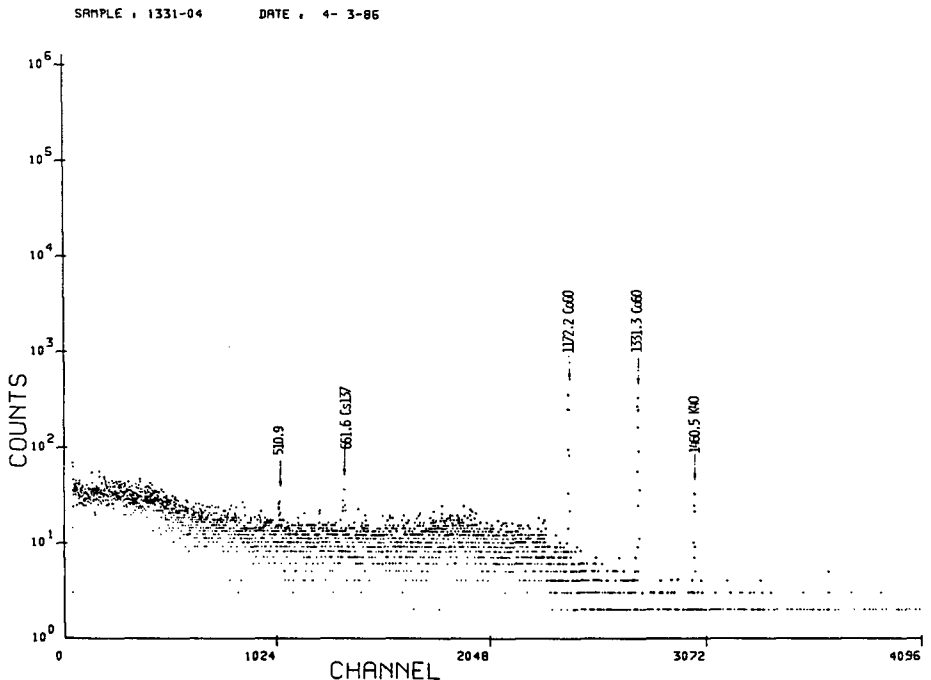


FIG. 2.  $D_2O$  gamma spectrum (moderator system).

moderator system, respectively. In Fig. 1 the existence of the irradiated fuel fission product is due to the PHTS  $D_2O$  being in close contact with the fuel and therefore the fission products will easily migrate to the  $D_2O$ . On the other hand, in the moderator system, the dominant energy lines of Fig. 2 are those emitted from the corrosion isotopes since the residence time for that inventory is much higher than that of the PHTS. The attractiveness of this technique stems from the fact that the spent fuel measurement equipment and software analyses could be utilized without many difficulties. The future intention is to develop this technique as a tracking mechanism for  $D_2O$  transactions.

Tritium and  $\beta$ -activities have also been measured (Table I). These measurements can distinguish between  $D_2O$  samples collected from the moderator or primary heat transport systems. For the PHTS the  $\beta$ -activity is much higher than for the moderator system, while the tritium activity shows the opposite trend.



TABLE I. RESULTS OF  $\beta$ - AND TRITIUM MEASUREMENTS

	$\beta$ -activity (Bq/g)	T-activity (Bq/g)
Moderator system (MS)	1.8	$13.7 \times 10^7$
Primary heat transport system (PHTS)	1250	$7.16 \times 10^7$

The reason is that, in the first case the high  $\beta$ -activity is due to the closer contact of D<sub>2</sub>O with the fuel, while in the second case the higher tritium activities are due to longer residence (or irradiation) time.

It is planned to introduce soon the acoustic velocity method for field measurements. The heavy water contained in drums will be verified by using this technique. The estimated accuracy of this method is about 0.5%.

The Agency uses installed instrumentation to establish criticality conditions and power levels during PIVs. This verification method is of utmost importance in safeguarding against material misuse.

## 5. VARIABLE VERIFICATION

The following verification techniques are used to carry out variable tests and to produce quantitative estimates of the different components of the material balance:

- (a) For the moderator and primary heat transport system: Volume determination; sampling and analysis.
- (b) For other locations: Weight determination; sampling and analysis; and volume determination.

Volume determination is the most important verification activity when verifying D<sub>2</sub>O in power reactors. To achieve independent verification of heavy water in different systems, the Agency should, as part of the design information verification, participate during the plant commissioning stage

in the calibration of all D<sub>2</sub>O system containers. For commissioned reactors where the Agency has missed this opportunity, independent verification might be achieved by

- (i) Comparing the operator's calibration charts with a reference reactor system of similar design; or
- (ii) Confirming the operator's charts by verifying a few points on the calibration curves.

It should be noted that the conditions under which the calibration curves were generated should be maintained during the verification activities. If the D<sub>2</sub>O volume is to be determined under different conditions, knowledge of the magnitude of correction to be introduced to reflect the actual situation should be established.

Volume determination coupled with sampling and analysis produces a quantitative statement about D<sub>2</sub>O inventories in the moderator system and primary heat transport system. Sampling and analysis are being carried out for D<sub>2</sub>O concentration measurements. The Agency uses a very effective density meter, type PAAR-DMA-35. The principle of operating it is to relate the resonance vibrations to the liquid density and, through calibration, to D<sub>2</sub>O concentration. The engineering of this principle resulted in a very light and portable instrument with an accuracy of  $1 \times 10^{-3}$  g/cm<sup>3</sup> with a sample size of 1 ml. It is considered ideal for inspection work where numerous on-site samples are taken. A more advanced version is the PAAR-DMA-46 with an accuracy of  $1 \times 10^{-4}$  g/cm<sup>3</sup>, but this, however, requires a more permanent location. More advanced methods for sampling and analysis are infrared spectroscopy and mass spectrometry. At present, the Agency is using one of the Network of Analytical Laboratories (NWAL) to carry out infrared spectroscopy and refractometry on the collected D<sub>2</sub>O samples.

Table II shows a comparison between the measurements performed using densitometry, refractometry and infrared spectroscopy. The densitometry measurements have been done at the IAEA Safeguards Analytical Laboratory (SAL), while the other types of measurement have been done at one of the Network of Analytical Laboratories (NWAL).

Weighing is carried out exactly in the same way as for nuclear material accountancy, either by using the plant operator's weighing machines with appropriate authentication, or by using an adaptation of the load cell system, at present used for UF<sub>6</sub> cylinders. The operator's scales are calibrated periodically and the random and systematic components of error are estimated for each scale.

TABLE II. SAMPLING AND ANALYSIS RESULTS

Stratum	SAL	NWAL	
	Densitometry (%)	Refractometry (%)	Infrared (%)
Moderator system	99.6	98.7	99.1
Primary heat transport system	98.6	98.7	98.5
Reactor grade D <sub>2</sub> O (store)	99.5	99.7	99.2

Over the material balance period, the systematic and random components of errors associated with quantities forming the material balance equation are calculated. The propagation of the operator's measurement uncertainties results in the following value of the accuracy limits of MUF expressed as percentage of the plant inventory during the PIV:

$$\sigma_{\text{MUF}} = 0.5\%$$

Unlike nuclear material, there are no reference values for the measurement uncertainties expected for closing a material balance for D<sub>2</sub>O which are considered achievable in practice.

The authors believe that the above quoted figure is the first available estimate of the measurement uncertainty for MUF for power reactors.

#### REFERENCE

- [1] MORSY, S., et al., "Heavy water physical verification in power plants", Proc. 27th INMM Annual Mtg, New Orleans, 1986, Nucl. Mater. Manage. 15 (1986).



# **GENERAL STATISTICS**

**(Session 14)**

**Chairman**

**T. BEETLE**  
**IAEA**



# CRITERIA FOR SPECIFICATION OF STATISTICAL TESTS FOR MATERIALS SAFEGUARDS

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## Abstract

### CRITERIA FOR SPECIFICATION OF SEQUENTIAL TESTS FOR MATERIALS SAFEGUARDS.

In recent years many approaches have been proposed for analysis of sequences of material balance data. It is widely recognized that there is a central role for sequential evaluation of such data. However, near real time materials accountancy is not routinely used in practice; and there is, both inside and outside the nuclear community, misunderstanding of the aims and objectives attached to the use of these techniques. The paper reconsiders the suggestion that the problem is caused by the way in which detection and inspection goals were first specified by the IAEA. The result of this is that the evaluation of all tests is based on the traditional criteria of the power ultimately to detect a specified alternative hypothesis for diversion. It is suggested here that these criteria overlook the need for timely detection, and the need to guard against a wide range of possible diversion scenarios. Moreover, it is important to have a clear picture of the capability of any particular techniques to detect a range of types of anomaly in a data stream. The authors outline the benefits to be gained by the use of Page's test with the standardized MUF residuals, and develop this idea with reference to 'detectable quantity', which for a test for a given diversion scenario in a given measurement system gives the quantity of material which can be detected with a given probability.

## 1. INTRODUCTION

What are the aims of nuclear materials accountancy and why has the topic occupied such an important place in research for the last twenty years? In broad terms the answer lies with the politicians who wish to assure each other that efforts have been made to ensure

"... the timely detection of diversion of significant quantities of *nuclear material* from peaceful nuclear activities to the manufacture of nuclear weapons or of other nuclear explosive devices or for purposes unknown, and deterrence of such diversion by the risk of early detection" [1].

The justification for this effort is shown in the annual safeguards implementation reports which contain a statement to the effect that the IAEA detected no anomaly which would indicate the diversion of a significant amount of safeguarded nuclear material.

Three important questions remain about how such a statement can be made, what degree of confidence can be attached to it and what numerical techniques were used. The last ten years have brought a dramatic increase in automatic instrumentation and computerization with the consequent possibility of developing a near real time materials accountancy (NRTMA) system. Such a system would enable the 'timely' detection of anomalies in a sequential data stream to be more than a mirage. Yet, despite the considerable research effort, such a system is not in routine operation as a safeguards tool, though perhaps as a management tool. We must ask why this should be and what can be done to rectify the situation. This paper reviews some aspects of the present state of the techniques for NRTMA in the light of the stated IAEA guidelines, with a brief comment on their shortcomings. We note that experts from industry feel, as we do, that research results over the last five years point towards a consensus view of what is required in practice, but which lacks a set of detailed and mutually accepted aims. We attempt to outline such a set of aims and to show how they could be put into effect by referring to some plant data which have been made available to us [2]. Our aim is not to say, "We know how to do it — do it our way", but to provide a framework within which to stimulate discussion and to initiate action.

## 2. ACCOUNTANCY METHODS AND CRITERIA

A vast literature is now available on statistical methods in nuclear materials accountancy, and a feature of recent work is the similarity being achieved by independent workers. A detailed bibliography can be found in the paper by Speed and Culpin [3], which was recently read to the Royal Statistical Society in the United Kingdom. However, a feature of that paper and of other work in the last ten years is an obsession with the ideas of power of detection and the need to specify the nature of the alternative hypothesis. The problem here is that an alternative hypothesis is synonymous with the strategy of the diverter and as such is 'unknown'. Thus, if a methodology is to be proposed for use it must be robust to a range of different diversion scenarios, the extremes of which are a large abrupt diversion and a small protracted diversion.

Whether this is accepted or not the concept of power is still a problem. This concept crept into use as one of the standard components of statistical inference. The difficulty is that power and timeliness are incompatible. Power in this context is by definition 'the probability of ultimate detection of a diversion of a given type', and as such the 'power one test' has a comforting ring about it. We would suggest that it is small consolation to be sure of detecting a diversion if by the time of detection



a considerable amount of material has been diverted and the diversion has been going on for a long time. Two points should be made. The first is that it may be preferable to use a test with a lower *ultimate* power, if in the early stages of a diversion the probability of detection is greater. The second is that the traditional concept of power demands that a loss scenario be known and then that the probability of ultimate detection of this alternative hypothesis be calculated. There are several deficiencies in this approach:

- (a) The concept of probability is one which is both feared and not well understood by the people who will use the tests;
- (b) When a test is used as a plant evaluation tool the plant can only "pass" or "fail" the test, and the extent of the failure, for example, is not easy to assess;
- (c) It does not immediately provide a measure of what a given accountancy system applied to a given plant is capable of detecting.

In the paper we gave at the IAEA safeguards symposium in 1982 [4] we outlined the concept of detectable quantity which, like power, involves probability statements about detection. However, these are couched in a form which makes them directly relevant to the practitioner. We define  $DQ_\alpha$  as the quantity of material which can be diverted before there is an  $\alpha\%$  chance of detection.

This quantity is most easily expressed in units of the MUF standard deviation. We contend that this measure should form part of a new set of safeguards guidelines. The beauty of the idea is that the benefits of changing the specifications of the test statistic (to accept, for example, a higher rate of false alarms) or of improving the measurement precision of inventories or throughputs can immediately be evaluated. In the use of Page's test, Jones [5] has shown how such calculations can be performed rapidly rather than by the use of extensive Monte Carlo simulations. In the discussion on the paper of Speed and Culpin [3], Jones remarked that the fundamental plant information is contained in the ratio of the inventory to throughput standard deviations. These ideas together make possible the concept of dynamic plant design.

### 3. ELEMENTS IN A SAFEGUARDS PACKAGE

We recognize that two statisticians outside industry who are arrogant enough to attempt to frame a set of safeguards guidelines are liable to be branded as blinkered idealists unaware of the real problems. So we are, but we are realists inasmuch as we feel it is better to be positive and to define guidelines which will state what *can* be achieved rather than to be depressed because of our inability to meet unrealistic target goals. We see five components to a package for NRTMA.

- (a) It is necessary to control false alarms on both practical and economic grounds. In the specification of statistical tests it is most appropriate to think of a campaignwise false alarm rate and not to attempt to use a rate per balance

period which has less meaning in the context of testing a sequential data stream. It is also important to remember that a false alarm is not synonymous with a false accusation. As such a false alarm rate can be set for a given plant, it being recognized that a higher false alarm rate will provide a greater sensitivity in the detection of diversion.

- (b) It is important that all tests should be two-sided, thus providing an opportunity to detect a positive bias in a measuring system as well as genuine loss. Failure to do so could mean that a genuine loss within a plant with a positive measurement bias would go undetected.
- (c) A system must guard against a range of diversion scenarios. In the sense that the value of MUF in a given balance period is unable to provide any information about the existence of a protracted diversion, it is important to combine tests relevant to a range of types of loss. Herein lies the dilemma. With enough tests it should be possible to detect any type of loss, but at the price of a combined false alarm rate which is unacceptably high. However, when the tests are modified to give an acceptable combined false alarm rate, the danger is that individual tests have unacceptably low detection capabilities. With care, it is possible to provide a single test sensitive to both abrupt and protracted losses.
- (d) For a given test used on a given plant configuration it is important to be able to state the detectable quantities for suitably chosen probability levels.
- (e) If in a sequential data stream an anomaly is detected, it is important to have a simple means of assessing the type of the anomaly and the period in which it started. A more complex problem which requires further research effort is to estimate the amount of material missing given that an alarm has occurred.

#### 4. AN ILLUSTRATIVE EXAMPLE

It has been mentioned elsewhere [3] that no real data sets relating to NRTMA are available in the open literature. We have said many times that there are likely to be plant specific aspects to every problem and these need to be incorporated into the statistical techniques which are applied. The structure of the plant data we have seen is such that, as well as substantial negative correlations between MUFs in successive periods, there are also appreciable correlations between MUFs two periods apart. The recognition of this fact and its inclusion in a test procedure is an important component of the statistical evaluation of the data. However, although we can learn much from study of such 'historical' data sets, they are of limited value in the long term. In an on-line evaluation of a sequential data stream an anomaly, once detected would, it is hoped, be followed up and its cause isolated. This is particularly true of any anomalies caused by systematic errors in measuring instruments. When a complete campaign is analysed retrospectively it is likely that either no corrective action has been taken, thereby possibly masking

TABLE I. DETECTABLE QUANTITIES  $DQ_\alpha$  FOR 35-PERIOD CAMPAIGN: PROTRACTED LOSS FROM PERIOD 1

$\alpha$	Loss rate ( $\times \sigma_{MUF}$ )								
	0.15	0.20	0.30	0.40	0.50	0.60	0.70	0.80	1.00
50	3.15	2.80	2.70	2.80	3.00	3.00	2.80	3.20	3.00
75	4.80	4.00	3.60	3.60	3.50	3.60	3.50	4.00	4.00
90	—	5.20	4.20	4.00	4.00	4.20	4.20	4.00	5.00
Maximum loss possible	5.25	7.00	10.50	14.00	17.50	21.00	24.50	28.00	35.00

TABLE II. DETECTABLE QUANTITIES  $DQ_\alpha$  FOR 35-PERIOD CAMPAIGN: ABRUPT LOSS IN PERIOD 5

$\alpha$	Detection in period 5	Detection by period 6
50	2.75	2.00
75	3.25	2.40
90	3.75	2.85

other anomalies, or it has been taken at arbitrary points in the data stream unconnected with any formal analysis.

However, in this paper we use the data from a 35-period campaign to illustrate the guidelines set out in Section 3. We used a false alarm rate of 5% over the campaign period and performed a two-sided test aimed at detecting gains and losses. We used Page's test with the SITMUF statistic and, in an attempt to provide a single test sensitive to both protracted and abrupt diversions, we chose the parameter values as  $h = 2.500$ ,  $k = 0.985$ . Tables I and II give detectable quantities in units of  $\sigma_{MUF}$  for protracted losses of varying sizes starting in period 1, and for an abrupt loss occurring in period 5. To calculate these values we have taken as plant indices the average inventory and throughput standard deviations for the whole campaign.

TABLE III. PAGE'S TEST ANALYSIS OF 35-PERIOD CAMPAIGN, SHOWING THE TEST STATISTIC FOR LOSSES

Period	h = 13.720 k = 0.000		h = 2.500 k = 0.985		h = 0.000 k = 3.186	
	SITMUF	TEST	SITMUF	TEST	SITMUF	TEST
1	-0.445	0.000	-0.445	0.000	-0.445	0.000
2	6.319	6.319	6.319	<u>5.334</u>	6.319	<u>3.133</u>
3	-2.693	3.626	-3.392	0.000	-3.392	0.000**
4	-0.774	2.852	-1.634	0.000**	-1.773	0.000
5	-0.746	2.106	1.190	0.205	-2.228	0.000
6	-6.138	0.000	-5.153	0.000**	-6.725	0.000**
7	-5.494	0.000*	-5.492	0.000**	-5.492	0.000**
8	-2.836	0.000	-2.836	0.000	-2.836	0.000
9	-2.661	0.000	-2.661	0.000*	-2.661	0.000
10	-2.910	0.000	-0.720	0.000	-2.910	0.000
11	-2.544	0.000	-0.729	0.000	-2.544	0.000
12	-1.976	0.000	-0.223	0.000	-1.976	0.000
13	-3.927	0.000*	-2.335	0.000	-3.927	0.000**
14	-0.733	0.000	-1.421	0.000	-0.733	0.000
15	-5.592	0.000	-7.608	0.000*	-5.592	0.000**
16	10.286	10.286	11.204	<u>10.219</u>	11.204	<u>8.018</u>
17	-0.213	10.073	0.000	0.000	0.000	0.000
18	1.945	12.018	2.119	1.134	2.119	0.000
19	2.248	<u>14.266</u>	2.367	<u>2.516</u>	2.367	0.000

From Table I the fascinating result emerges that, although the maximum possible loss over the campaign rises steadily with the rate of loss, the detectable quantities are remarkably stable. Table II shows that, if delay in detection by one balance period is acceptable, then appreciably smaller abrupt diversions can be detected. Table III gives the values of the test statistic for this data set and it is clear how these can be used to investigate the type of anomaly which has arisen. Symbol a shows the occurrence of an abrupt loss in that a significant value of the statistic is preceded by a zero. Symbol b, on the other hand, shows a substantial loss starting in period 24 which, although not significant, was followed by further losses in the succeeding three periods and which then gave an alarm. Immediately prior to period 24 there was no evidence of any loss occurring.

TABLE III. (cont.)

Period	h = 13.720 k = 0.000		h = 2.500 k = 0.985		h = 0.000 k = 3.186	
	SITMUF	TEST	SITMUF	TEST	SITMUF	TEST
20	-1.973	0.000	-1.973	0.000	-0.686	0.000
21	-1.353	0.000	-1.353	0.000	-0.081	0.000
22	-3.064	0.000	-3.064	0.000*	-2.328	0.000
23	-2.394	0.000	-1.437	0.000	-2.345	0.000
24	10.197	10.197 <u>b</u>	10.223	<u>9.238</u> <u>a</u>	10.196	<u>7.010</u>
25	1.145	11.343	1.167	0.182	1.167	0.000
26	1.444	12.786	1.463	0.660	1.463	0.000
27	1.293	<u>14.079</u>	1.312	0.987	1.312	0.000
28	-1.367	0.000	-0.492	0.000	-0.492	0.000
29	2.747	2.747	3.992	<u>3.007</u>	3.992	<u>0.806</u>
30	-0.427	2.320	-0.622	0.000	-0.622	0.000
31	1.507	3.827	1.255	0.270	1.255	0.000
32	-0.168	3.659	-0.371	0.000	-0.371	0.000
33	-1.343	2.316	-1.459	0.000	-1.459	0.000
34	-1.555	0.761	-1.677	0.000	-1.677	0.000
35	-4.560	0.000	-4.604	0.000*	-4.604	0.000*

\* a protracted gain signalled in this period.

\*\* an abrupt gain signalled in this period.

a, abrupt loss.

b, substantial loss starting in period 24.

## 5. CONCLUSIONS

What of the future? One of the conclusions from the recent technical workshop on near real time material accounting for reprocessing plants [6] was that

“... consensus on the best statistical tests to use seems to be emerging among the experts in order to reach an acceptable combination of sensitivity and timeliness; clearer definition of safeguards boundary conditions needs to be established”.

We agree. The time is now ripe for genuine international collaboration on a single package of methods for the design of plant measurement systems and analysis of sequential data streams.

We have illustrated in this paper how such a package might be designed and the way in which one particular technique could be used in practice. In doing this we are well aware that we are guilty of the parochial attitude to our own methods which has pervaded the development of much of this work. We must emphasize that it is unimportant which techniques are finally chosen, but it is important that they are able to meet adequate boundary conditions. But they must be internationally agreed upon and accepted. In this there is an organizational and a financial role for the IAEA, to act not just as an honest broker but as a catalyst for action.

To proceed it is clear that a set of boundary conditions must be agreed upon. It is important that these conditions differ radically in form from those which evolved out of INFCIRC/153 [1, 7]. It is of no value to know that NRTMA applied to a given plant *cannot* meet internationally agreed guidelines. It is more important to know what levels of anomaly *can* be detected and within what period of time of their occurrence. Couched in these terms the materials accountancy package has a role to fulfil in plant design and licensing as well as in routine monitoring and safeguards. We believe an opportunity for significant advance is now within the grasp of industry. With vision and a willingness for international co-operation and compromise, the benefits of this opportunity can be realized.

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# A GENERALIZED APPROACH TO DETERMINING INSPECTION SAMPLE SIZES

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## Abstract

### A GENERALIZED APPROACH TO DETERMINING INSPECTION SAMPLE SIZES.

In general terms, it is assumed that for a given stratum there are  $h$  methods available for verification measurements. With the error standard deviation denoted by  $\delta_i$  for method  $i$ , it is assumed that  $\delta_1 > \delta_2 \dots > \delta_h$ . For inspection planning, it is assumed that inspection is by variables measurements applied in the attributes mode. The problem is to select the sample size for each measurement method to provide specified assurance that some given goal amount will be detected. Detection consists of finding one or more 'defective' items in the sample, where an item is declared to be defective if the operator-inspector difference exceeds some quantity selected to control the false alarm rate. The solution to this problem is presented for certain fixed parameter values. A more general solution is referenced. An example is presented to illustrate the approach. The detection probability is then studied as a function of diversion strategy.

## 1. INTRODUCTION

The inspection planning problem from the aspect of determining inspection sample sizes in a given stratum may be stated in its simplest terms as follows: "There are a given number of measurement methods. How many items should be measured by each method?"

Existing sample size formulas that deal with this question are based on the methodology described in a 1975 IAEA symposium paper [1]. This methodology involves calculating three sample sizes to guard against gross defects ( $n_a$ ), partial defects ( $n_{v_1}$ ) and small biases ( $n_{v_2}$ ). Developed under the assumption that there are two measurement methods, called an attributes tester and a variables tester, the sample size is  $n_a$  for the attributes tester and the larger of  $n_{v_1}$  and  $n_{v_2}$  for the variables tester.

Sample sizes  $n_a$  and  $n_{v_1}$  are calculated using simple formulas whereas the calculation of  $n_{v_2}$  is complex and realistically requires the use of a computer. Usually, experience has shown that  $n_{v_1} > n_{v_2}$  and in many applications the sample size  $n_{v_2}$  need not be calculated.

The problem of sample size determination has been reconsidered by a number of authors in recent years [2-8]. These contributions have been motivated by improvements in NDA such that the originally conceived attribute tester may now be regarded as a variables tester, but one that is applied in the attributes mode. This development has resulted in some new inspection features that have to be taken into account:

The definition of a partial defect needs to be re-evaluated;

The variables tester in a two-tester situation may not detect a partial defect with certainty as was originally assumed;

There are often three measurement methods (two NDA and one DA) rather than two, and conceptually there could be more.

Further, if it is assumed that errors of measurement are constant on a relative basis rather than an absolute basis, it is necessary to make a distinction between positive operator-inspector differences (overstatements) and negative differences (understatements).

This paper reports on a sample size determination procedure that takes into account these identified features. It is essential that the procedure be simple enough to permit reasonably simple application, and yet be based on a realistic model. The proposed procedure satisfies these criteria.

It will be noted that the sample size formulas require that goal quantities be given in terms of the amount of element or isotope, and also that all measurement errors are assumed to be random, i.e. systematic errors are ignored. With respect to the first point, it is noted that many NDA instruments must be used in connection with other data (e.g. bulk measurements) to determine amounts of element or isotope. Strictly speaking, it is the entire measurement process for which the sample size formulas apply, but in the case of NDA it is the NDA error that dominates and the sample sizes can be based on NDA errors alone. In some cases one can accept the operator's statements about some features of the data (e.g. U-concentration) and only verify other features (e.g. enrichment). With respect to the random error assumption, the assumption is made only for planning purposes. With internal standards and emphasis on checking for internal consistency, the assumption for planning purposes does not appear unreasonable. Systematic errors should be taken into account in the data analysis.



## 2. SUMMARY

The following summary statements are helpful in understanding the formulas given in this paper.

Sample sizes are calculated separately for each stratum;

Sample sizes are based on inspection in the attribute mode for planning purposes, although at least a part of the data is applied in the variables mode;

The number of measurement methods,  $h$ , for a given stratum is not fixed;

The relative error standard deviation for method  $i$  is  $\delta_i$ , and  $\delta_1 > \delta_2 > \dots > \delta_h$ ;

The only requirement on  $\delta_1$  is that it must detect with high probability the existence of a gross defect equal in size to the declared value. No requirements are placed on  $\delta_i$  for  $i > 1$ , except that  $\delta_h$  must be small enough to permit detection of small biases;

For any given measurement, the reject level is fixed at three standard deviations;

Whenever an operator-inspector difference exceeds the critical value, the item is remeasured and declared to be a defect only if the remeasured value also exceeds the critical value;

A distinction is made between operator declared overstatements and understatements;

The sample size formulas are given by Eqs (1) and (13).

## 3. ASSUMPTIONS

The following assumptions are made:

For a single measurement, the random error is of dominant importance relative to the systematic error;

The occurrence of one or more defects by any measurement method is equivalent to "detection" of an unacceptable situation, and the probability of detection is one of the criteria for determining sample sizes;

The size of the level  $i$  defect is related to  $\delta_{i-1}$  and is chosen such that it will be declared a defect with small probability, 0.025, if measured by method  $(i-1)$ ;

Measurement error standard deviations are expressed on a relative basis. In the event that such errors are constant on an absolute basis, the theory can be readily adapted;

The "amounts" specified in defining some of the parameters are amounts of either element or isotope but are all in the same units (kg U; g Pu; g  $^{235}\text{U}$ , etc.).

#### 4. NOTATION

The results apply to a given stratum. The subscript  $i$  refers to a measurement method level;  $i = 1, 2, \dots, h$ .

- $M$ , goal amount to be detected
- $\beta$ , probability of failing to detect the diversion of an amount  $M$
- $x_0$ , nominal or average amount per item
- $N$ , number of items in stratum
- $\alpha$ , probability that a given method will detect one or more defects when none exist.
- $\delta_i$ , relative error standard deviation for method  $i$   
( $\delta_1 > \delta_2 > \dots > \delta_h$ )
- $x_{ai}$ , actual amount in defective item for level  $i$  defect
- $d_i$ ,  $x_0 - x_{ai}$
- $\hat{d}_i$ , operator value minus inspector value = estimate of  $d_i$
- $R_i$ ,  $\delta_i/\delta_{i-1}$
- $\gamma_i x_0$ , size of level  $i$  defect =  $|d_i|$
- $m_i$ ,  $M/\gamma_i x_0$  = number of level  $i$  defects
- $p_i$ , probability that  $\hat{d}_i$  exceeds the critical value for a single measurement when  $E(\hat{d}_i) = d_i$
- $n_i$ , sample size for method  $i$
- $E$ , expected value; and  $\hat{d}_i$ , always = estimate of  $d_i$ .

(In application, sample sizes should be embedded with  $n_i - n_{i+1}$  items measured by method  $i$  for  $i = 1, 2, \dots, h-1$  and  $n_h$  by method  $h$ . The total number of items measured is then  $n_1$ .)

- $q$ , probability that a level  $i$  defect will be detected as a defect by method  $(i-1)$  with a single measurement.

## 5. RESULTS

For measurement method 1, the well known sample size formula applies:

$$n_1 = N(1 - \beta^{x_0/M}) \quad (1)$$

In developing the sample size formulas for  $i > 1$ , it is first necessary to define a level  $i$  defect.

For level  $i$ ;  $i > 1$ , all items that are defective are defective by an amount  $\gamma_i x_0$ , where

$$\gamma_i = t\delta_{i-1} \quad (2)$$

and where the diverter is free to choose  $\gamma_i$  and the value of  $t$  is chosen by the inspector.

This problem of choosing  $\gamma_i$  is now considered separately for overstatements in which the declared value,  $x_0$ , is larger than the actual value,  $x_{a1}$ , and for the understatement case in which  $x_0 < x_{a1}$ . The quantity  $d_1$  is the defect size.

### Overstatement

$$d_1 = \gamma_1 x_0 = t\delta_{1-1} x_0 \quad (3)$$

$$x_{a1} = x_0 - d_1 = x_0(1 - t\delta_{1-1}) \quad (4)$$

The diverter chooses  $t$  such that the probability is small of classifying a level  $i$  defect as a defect when measured by the level  $(i-1)$  method. That is, he wants a high probability of escaping detection. For a single observation, and with a critical value of three standard deviations, the probability of detection, denoted by  $q$ , is

$$q = P(\hat{d}_1 > 3\delta_{1-1}x_0 \mid E(\hat{d}_1) = t\delta_{1-1}x_0) \quad (5)$$

where  $\hat{d}_1$  is the measured discrepancy.

Equation (5) reduces to

$$q = P\left(Z > \frac{3-t}{1-t\delta_{1-1}}\right) \quad (6)$$

where  $Z$  is a standardized random variable, normally distributed with mean zero and unit standard deviation. The parameter  $t$  is now chosen to make  $q$  small. Since an item is declared to be a defect only if two successive differences exceed the critical value, which occurs with probability  $q^2$ , we choose  $t$  such that  $q^2$  is less than some value. A convenient reasonably small value for  $q^2$  is 0.025. This is convenient because if  $q = \sqrt{0.025} = 0.158$ , then from a table of the normal distribution, the right hand side of the inequality in Eq. (6) is 1.0, and the simple solution for  $t$  is

$$t = \frac{2}{1 - \delta_{i-1}} \quad (7)$$

Thus, from Eq. (2), the diverter will select a level  $i$  defect of size

$$d_i = \gamma_i x_0 = \frac{2\delta_{i-1}x_0}{1 - \delta_{i-1}} \quad (8)$$

Now, for a defect of size  $\gamma_i x_0$  as given by Eq. (8) we find the probability that it will be declared to be a defect, which occurs when the critical value  $3\delta_i x_0$  is exceeded on the two successive measurements. Letting  $p_i$  be the probability that the measured discrepancy for an item with a defect of size  $d_i$  will exceed its critical value, the expression for  $p_i$  reduces to

$$p_i = P\left(Z > \frac{3R_i(1 - \delta_{i-1}) - 2}{R_i(1 - 3\delta_{i-1})}\right) \quad (9)$$

where

$$R_i = \delta_i / \delta_{i-1} \quad (10)$$

If normally and independently distributed random errors are assumed,  $p_i$  can be read from a table of the normal

distribution, and  $p_i^2$  is the probability of declaring the item in question to be a defect.

Since the defect is of size  $\gamma_i x_0$  given by Eq. (8) there are

$$m_i = \frac{M}{\gamma_i x_0} = \frac{M(1 - \delta_{i-1})}{2\delta_{i-1}x_0} \quad (11)$$

level  $i$  defects. Let us choose the sample size  $n_i$  such that the probability of detection, i.e. of finding one or more defects in the sample, is  $(1 - \beta)$ . The problem is more easily solved if we set the probability of finding zero defects equal to the complementary probability,  $\beta$ .

Now, zero defects are found if:

- (a) there are zero defects in the sample of size  $n_i$ , or
- (b) there is one defect but it escapes detection, or
- (c) there are two defects but both escape detection, etc.

The probability of  $x$  defects is represented by the binomial probability function in which there are  $n_i$  trials and in which  $m_i/N$  is the probability of a defect on any one trial.

Also, given  $x$  defects, and keeping in mind the assumption of independence in measurement errors, the probability that no defects are identified as such is  $(1 - p_i^2)^x$ . Combining this with the binomial PDF, and equating to  $\beta$ , we can solve for  $n_i$  the required sample size. The equation for  $\beta$  reduces to

$$\beta = (1 - m_i p_i^2 / N)^{n_i} \quad (12)$$

Solving Eq. (12) for  $n_i$  gives the solution

$$n_i = \frac{\ln \beta}{\ln(1 - m_i p_i^2 / N)}, \quad i > 1 \quad (13)$$

#### Understatements

By a similar development, the corresponding key results for the understatement case are as follows:

$$p_i = P \left( Z < \frac{-3R_i(1 + \delta_{i-1}) + 2}{R_i(1 + 3\delta_{i-1})} \right) \quad (14)$$

$$\gamma_i x_0 = \frac{2\delta_{i-1} x_0}{(1 + \delta_{i-1})} \quad (15)$$

The sample size formula is the same as for the overstatement case.

The sample size  $n_1$  in Eq. (13) is a function of  $\beta$ ,  $M$ ,  $\delta_{1-1}$ ,  $\delta_1$ ,  $N$ , and  $x_0$ . In application, tables have been prepared which make calculation of the sample size a very simple process.

In this paper, two important parameters were fixed for simplification. One is the definition of the critical value, i.e. the size of the discrepancy between the declared value and the inspector's value that defines a defect. The critical value was fixed at three standard deviations. The other parameter is the size of the level 1 defect created by the diverter. This was chosen to be of a size that would escape detection with a probability of 0.975 if measured by method (1-1). In the IAEA Safeguards Technical Report that forms the basis for a part of this paper [9], these parameters were not fixed in the Report's Annex 4 so as to permit the user greater flexibility in application. Also, some results were given in Annex 5 of the Report relating to the effect on the sample size of varying the defect size.

## 6. EXAMPLE

Let us consider the UO<sub>2</sub> powder stratum 1 of the low enriched uranium (LEU) fuel fabrication facility that is used as an example facility throughput in the IAEA Safeguards Technical Manual [10]. This being an input stratum, assume that the diverter will understate the amount of uranium. The input parameter values are as follows:

$$\text{goal amount: } M = \frac{75 \text{ kg } ^{235}\text{U}}{0.03 \text{ kg } ^{235}\text{U} / \text{kg U}} = 2500 \text{ kg U}$$

$$\text{number of containers of UO}_2 \text{ powder: } N = 12\,000$$

$$\text{non-detection probability: } \beta = 0.10$$

$$\text{nominal amount of U per container: } x_0 = 20 \text{ kg U}$$

We assume three measurement methods with the following relative standard deviations:

$$\delta_1 = 0.12, \quad \delta_2 = 0.05, \quad \delta_3 = 0.002$$

The sample size for method 1 is computed from Eq. (1):

$$n_1 = 12\,000(1 - 0.10^{20/2500}) = 219$$

To find  $p_1$  from Eq. (14), we first calculate the ratio  $R_1$  for levels 2 and 3:

$$R_2 = 0.05/0.12 = 0.417; \quad R_3 = 0.040$$

Then

$$p_2 = P \left( Z < \frac{-3(0.417)(1.12) + 2}{(0.417)(1.36)} = 0.855 \right)$$

$$p_3 = P \left( Z < \frac{-3(0.040)(1.05) + 2}{(0.040)(1.15)} = 1.000 \right)$$

The sample sizes are then calculated from Eq. (13):

$$n_2 = \frac{\ln 0.10}{\ln[1 - (583)(0.855)^2/12\,000]} = 64; \quad n_3 = 20$$

Thus, the number of items measured by method 1 would be  $219-64 = 155$ ; by method 2,  $64-20 = 44$ ; and by method 3, 20. The total number of items measured would be 219.

## 7. NON-DETECTION PROBABILITY

The non-detection probability is found for a given stratum as a function of the defect size. We assume that the diversion strategy is to create  $m_0$  defects of size  $\gamma_0 x_0$ , so that in order to divert the goal amount  $M$  the relationship is

$$m_0 = M/\gamma_0 x_0 \quad (16)$$

Then, from Eq. (12), the probability of finding zero defects with method 1 is

$$\beta_1 = (1 - m_0 q_1^2/N)^{n_1 - n_1 + 1} \quad (17)$$

for  $i = 1, 2, \dots, h$ , where  $n_{h+1} = 0$  by definition. In Eq. (17),  $q_1$  is the probability that the defect of size  $\gamma_0 x_0$  will be declared a defect by method 1 on a single measurement. Specifically, for overstatements,

$$q_1 = P \left( Z > \frac{3\delta_1 - \gamma_0}{(1 - \gamma_0)\delta_1} \right) \quad (18)$$

with the signs in the numerator and on  $\gamma_0$  in the denominator reversed for understatements.

TABLE I. SAMPLE SIZES FOR LEU PLANT

$\delta_2$	Method 1	Method 2	Method 3
0.11	0	176	43
0.10	0	180	39
0.09	0	184	35
0.08	0	187	32
0.07	45	146	28
0.06	118	77	24
0.05	155	44	20
0.04	170	33	16
0.03	173	34	12
0.02	173	38	8
0.01	173	43	3

The non-detection probability,  $Q$ , is simply the product of the  $\beta_i$ 's.

Because of the many parameters, it is difficult to evaluate the non-detection probability in general terms. However, some calculations have been made to cover the typical situation in which there are three measurement methods. Method 1 is the gross defects tester with a typical standard deviation of about 10%, and method 3 is a bulk sampling analytical method with a typical standard deviation of less than 1%. In the investigations done to date, method 2 is assumed to have a standard deviation varying between that of methods 1 and 3 to study the effect on  $Q$ .

The results of one typical investigation are now presented.

LEU fabrication plant of Section 6 example:

$$\delta_1 = 0.12$$

$$\delta_3 = 0.002$$

and  $\delta_2$  is varied from 0.01 to 0.11 in steps of 0.01.

It was found that for  $\delta_2 \geq 0.08$ ,  $n_2$  from Eq. (13) is greater than  $n_1$  of Eq. (1). In this event, do not use method 1; make  $n_1 = n_3$  measurements with method 2 and  $n_3$  with method 3. Table 1 gives the sample sizes for the various cases.



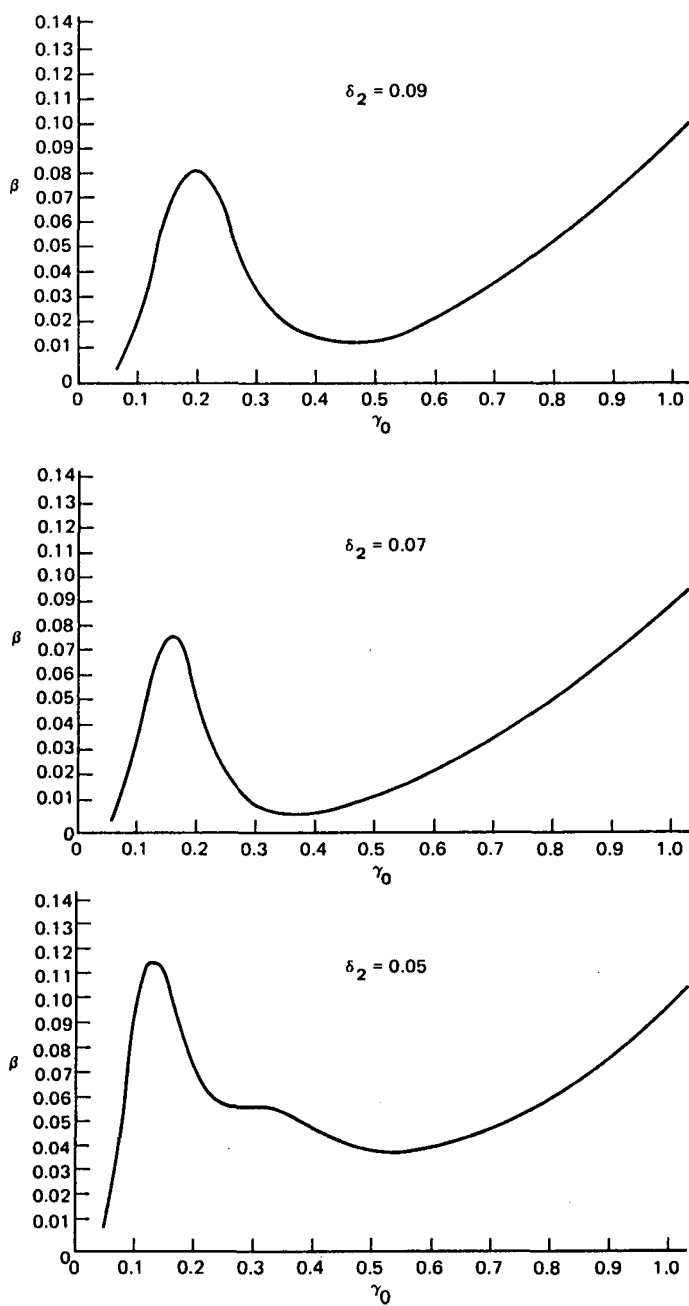


FIG. 1.  $\beta$  versus  $\gamma_0$  for  $\delta_2 = 0.09, 0.07$  and  $0.05$ .

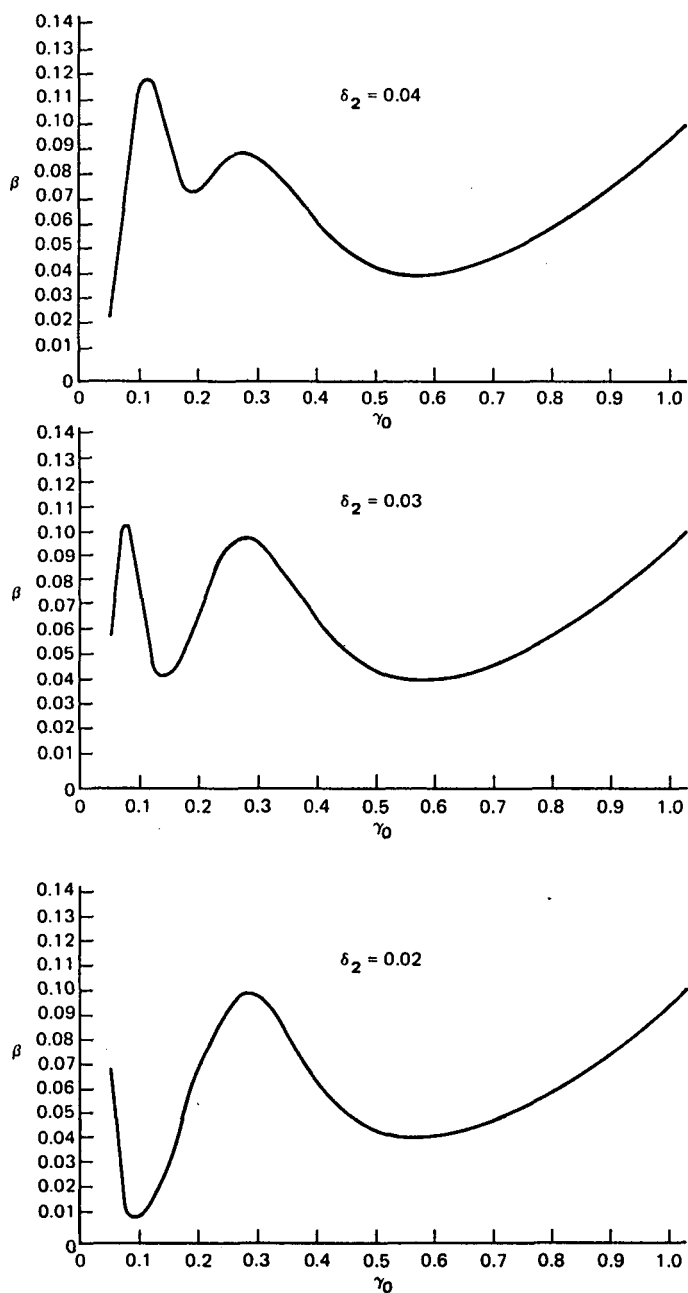


FIG. 2.  $\beta$  versus  $\gamma_0$  for  $\delta_2 = 0.04, 0.03$  and  $0.02$ .

Figures 1 and 2 show  $\delta_2$  plots of  $\beta$  versus the defect size  $\gamma_0$  for some specific values of  $\delta_2$ . It is noted that for some combinations of  $\delta_2$  and  $\gamma_0$ ,  $\beta$  exceeds the design value of 0.10. The degree to which the design value of 0.10 is exceeded is not great, and  $\beta > 0.10$  only within a very small region of the defect size  $\gamma_0 \times \delta_2$ . For this and other cases studied, a small increase of 10% in sample size  $n_3$  reduces  $\beta$  to 0.10 or smaller for all defect sizes, and this 'rule of thumb' has been incorporated as part of the procedure for determining inspection sample sizes.

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# RELIABILITY MODELLING FOR CONTAINMENT AND SURVEILLANCE DEVICES

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## Abstract

### RELIABILITY MODELLING FOR CONTAINMENT AND SURVEILLANCE DEVICES.

In connection with the development, testing and implementation of various containment and surveillance (C/S) devices, test criteria are established and inferences are made about reliability based on the test data. It is important that workers in this field be aware of the mathematical models used, of the assumptions behind them and of how invalid assumptions can affect the conclusions drawn. Following a brief introduction of the descriptive functions used in reliability applications, the exponential model is discussed. The assumptions behind the model are presented, and minimum test requirements are set forth to demonstrate a given required reliability. The effect of inspection frequency on reliability is introduced, the random variable of interest in this case being the downtime of the C/S device. A section on the exponential model deals with component redundancy to enhance reliability. For the exponential model, the parameter  $\lambda$  is a constant, the same for all units or items, and also over time for a given unit. In the event that this parameter is a random variable which may take on different values for different units, then the probability density function is a two-parameter function related to the F density function. This function is derived and methods for estimating the parameters are given. Using a set of data generated from this two-parameter distribution, the consequences of assuming incorrectly that the exponential function applies are set forth. Goodness of fit tests are also presented and applied to choose between the models on the basis of the data. The final section briefly mentions the Weibull density function, an appropriate function when the conditional probability of failure is not constant. This would be the case, for example, if wearout mechanisms exist.

## 1. INTRODUCTION

Reliability modelling is receiving increasing attention at the IAEA and elsewhere in connection with the development and implementation of various containment and surveillance (C/S) devices. In discussions of test requirements between developers and users, and in evaluating the performance of a given C/S device, it is important that all parties be aware of the assumed models and of the consequences of invalid assumptions. This paper provides a discussion of reliability modelling to enhance the understanding of reliability concepts and improve communications on this subject.

## 2. BASIC RELIABILITY CONCEPTS

In reliability modelling, the random variable of interest is either the time to failure measured on some time-scale or else the number of failures per unit time. The two random variables are obviously related, and this relationship will be demonstrated for the exponential model in Section 3. This paper focuses on time to failure as the random variable.

There are various probability functions that characterize time to failure:

The probability density function (PDF) is denoted by  $f(t)$ , and  $f(t)dt$  is the probability of failure in the interval  $(t, t + dt)$ .

The cumulative distribution function (CDF) is denoted by  $F(t)$ , and is the integral of  $f(t)dt$  from 0 to  $t$ .  $F(t)$  is the probability of failure before time  $t$ .

The survival function is denoted by  $S(t)$ ;  $S(t) = 1 - F(t)$ .

The hazard function is denoted by  $h(t)$ , and

$$h(t)dt = f(t)dt/S(t) \quad (1)$$

is the conditional probability of failure in the interval  $(t, t + dt)$  given that the item has survived to time  $t$ .

The mean time to failure (MTTF) or mean time between failures (MTBF) is the expected value of  $t$ :

$$MTTF = \int_0^{\infty} tf(t)dt \quad (2)$$

When  $h(t)$  is plotted versus  $t$ , a classic curve is the so-called bathtub curve in which  $h(t)$  decreases monotonically from  $t = 0$  to  $t = t_0$ , is flat from  $t = t_0$  to  $t = t_1$ , and then increases monotonically, beginning at  $t = t_1$ . Infant mortality is described in the initial part of the curve while wearout failure begins to occur after  $t = t_1$ . A common assumption for C/S devices is that the applicable portion of the curve is between  $t_0$  and  $t_1$ , i.e. in the region where  $h(t)$  is independent of  $t$ . In this event, the appropriate PDF is the exponential PDF, discussed in Section 3.

## 3. EXPONENTIAL PDF

The exponential density function has been justified in reliability work on strictly empirical grounds; it has been

demonstrated to be applicable in many situations. It is also known to be an appropriate model for complex systems, i.e., for systems with a large number of in-series components, none of which individually contributes very heavily to the total failure probability. (This result corresponds to the central limit theorem which establishes the normal PDF as the appropriate model in many non-life test situations). However, the simplicity of the theory and the ease of application should not delude one into believing that the exponential model is universally applicable. Whether or not it applies in the case of C/S devices is a question of importance. As will be demonstrated in later sections, the consequences of using an invalid model can be quite serious.

With this caveat in mind, the exponential PDF is briefly reviewed here. The relevant functional forms are as follows:

$$f(t) = \lambda e^{-\lambda t} \quad (3)$$

$$F(t) = 1 - e^{-\lambda t} \quad (4)$$

$$h(t) = \lambda e^{-\lambda t} / e^{-\lambda t} = \lambda, \text{ independent of } t \quad (5)$$

$$\text{MTTF} = E(t) = 1/\lambda \quad (6)$$

The exponential PDF is related to the Poisson PDF in that if Eq. (3) applies, then the probability of  $x$  failures occurring before time  $t$  is given by the Poisson PDF.

$$P(x) = e^{-\lambda t} (\lambda t)^x / x! \quad (7)$$

Note that for  $x = 0$ ,  $P(x) = e^{-\lambda t}$ , which is simply the survival function  $S(t)$ .

The parameter  $\lambda$  is estimated very simply from test data. The estimate, designated by  $\hat{\lambda}$ , is given by

$$\hat{\lambda} = F/T \quad (8)$$

where  $F$  is the total number of failures and  $T$  is the total accumulated test time. In Eq. (8), it is important to note that given the exponential PDF, it does not matter how many test units are involved. One unit tested for 50 weeks has the same  $T$  value as 10 units each tested for five weeks or 50 units each tested for one week, say. Clearly, one must have great faith in the exponential PDF as being applicable, especially when either the number of test units or the test time per unit is small. In Section 5, a test of the goodness of fit of the model is discussed.

Although Eq. (8) provides a point estimate of  $\lambda$ , it gives no indication of the uncertainty in that estimate. Intuitively one would expect that the greater the number of

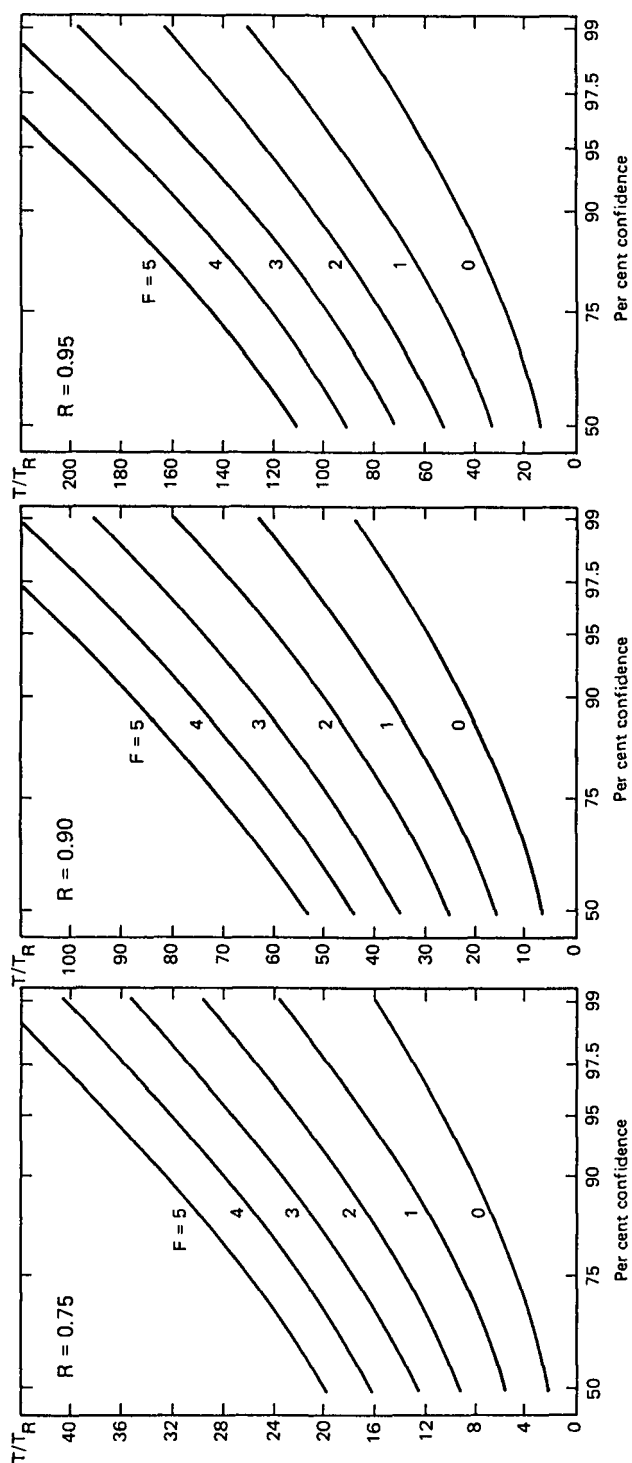


FIG. 1. Test requirements to establish a reliability  $R$  at exposure  $T_k$



failures, the smaller the uncertainty in  $\hat{\lambda}$ . The following result provides a measure of the relationship between  $F$  and the uncertainty in  $\hat{\lambda}$ .

The  $100(1-\alpha)$  % upper confidence limit on  $\lambda$  is  $\lambda_u$ , given by

$$\lambda_u = \chi^2_{(1-\alpha), \gamma} / 2T \quad (9)$$

where  $\gamma = 2(F + 1)$  and where  $\chi^2_{(1-\alpha), \gamma}$  is the value exceeded by a chi-square variate with probability  $\alpha$ , where the variate has  $\gamma$  degrees of freedom.

This result establishes minimum test requirements for testing C/S devices. By specifying  $\alpha$  and the required upper limit on  $\lambda$  (or equivalently, lower limit on MTTF), the minimum requirements expressed as the pair  $(F, T)$  are established.

For example, if the minimum reliability of some C/S device must be 0.90 for three months, then solving Eq. (4) for  $t = 3$  gives the required upper limit on  $\lambda$ , expressed in months<sup>-1</sup>.

$$e^{-3\lambda_u} = 0.90 \Rightarrow \lambda_u = 0.0351/\text{month}$$

Equivalently, the required lower limit on MTTF =  $1/\lambda_u$  = 28.5 months. If this reliability is to be established with 95% confidence ( $\alpha = 0.05$ ), and since minimum test time occurs when  $F = 0$ , the minimum test time comes from solving Eq. (9) for  $T$  with  $\lambda_u = 0.0351$ ,  $\alpha = 0.05$ , and  $\gamma = 2$ .

$$T = 5.99 / (2 \times 0.0351) = 85.3 \text{ months}$$

Figure 1 gives some general results that are useful in planning acceptance tests for C/S devices. This figure gives total test time,  $T$ , required to establish a reliability,  $R$ , with a given per cent confidence when  $F$  failures occur among the test units. The abscissa of Fig. 1 gives the per cent confidence and the ordinate gives the ratio  $T/T_R$ , where  $T_R$  is the exposure on some scale for which the reliability,  $R$ , is to be achieved.

For example, if a reliability of 0.90 is to be achieved for a  $T_R$  of 4 weeks, and if this reliability is to be demonstrated with 95% confidence, then the following combinations of  $F$  and  $T$  will provide this confidence:

$$\begin{array}{ll}
 F = 0, T/TR = 28 & \Rightarrow T = 112 \text{ weeks} \\
 F = 1, T/TR = 45 & \Rightarrow T = 180 \text{ weeks} \\
 F = 2, T/TR = 60 & \Rightarrow T = 240 \text{ weeks, etc.}
 \end{array}$$

One other reliability problem of interest to the IAEA in implementing C/S devices concerns the probability distribution of the device downtime given a specified time interval between inspections. The first four moments that characterize this density function have been found [1], and the reference indicates how to make appropriate probability statements. An important descriptive parameter is the expected value of the downtime, which, expressed in fractional form, is shown in [1] to be

$$E(\text{downtime}) = \frac{e^{-\lambda H} - 1}{\lambda H} + 1 \quad (10)$$

where  $H$  is the time between inspections expressed in the same exposure units as  $\lambda^{-1}$ . For example, if  $\lambda$  is  $0.02 \text{ weeks}^{-1}$  (MTTF = 50 weeks) and if  $H = 26$  weeks, then the expected fractional downtime is

$$\frac{e^{-(0.02)(26)} - 1}{(0.02)(26)} + 1 = 0.220, \text{ or } 22\%$$

For  $H = 13$  weeks this expected fraction is 0.119, and for  $H = 4$  weeks it is 0.039.

#### 4. REDUNDANCY

When redundancy is built into a given system, then failure occurs when the  $k$  identical components fail. One can envisage one operating component and  $(k - 1)$  standby units, with one standby unit called into service when the operating unit fails. This process continues until the  $k$ -th unit fails, at which time system failure occurs. Alternatively, for the assumed exponential PDF, with its hazard function independent of time in service, the system can be perceived as having  $k$  identical units in operation initially, with system failure occurring when all  $k$  units have failed.

For this redundant system, the system survives if the number of failures,  $x$ , at time  $t$  is less than or equal to  $k-1$ . The distribution function is the complementary probability. Therefore, from Eq. (7),

$$F(t) = 1 - \sum_{x=0}^{k-1} e^{-\lambda t} (\lambda t)^x / x! \quad (11)$$

TABLE I. EFFECT OF REDUNDANCY ON FAILURE PROBABILITY  $F(t)$ 

$\lambda t$	$k = 1$	$k = 2$	$k = 3$	$k = 4$
0.2	0.181	0.018	0.001	0.000
0.4	0.330	0.062	0.008	0.001
0.6	0.451	0.122	0.023	0.003
0.8	0.551	0.191	0.047	0.009
1.0	0.632	0.264	0.080	0.019
1.5	0.777	0.442	0.191	0.006
2.0	0.865	0.594	0.323	0.143
2.5	0.918	0.713	0.456	0.242
3.0	0.950	0.801	0.577	0.353
4.0	0.982	0.908	0.762	0.567
5.0	0.993	0.960	0.875	0.735

The right-hand side of Eq. (11) is the incomplete gamma function

$$F(t) = \int_0^t \frac{\lambda t^{k-1} e^{-\lambda t}}{\Gamma(k)} dt \quad (12)$$

and hence the PDF for the redundant system is simply the integrand in Eq. (12).

Table I illustrates the effect of redundancy for  $k \leq 4$ . It gives  $F(t)$ , the CDF, as a function of the dimensionless quantity  $\lambda t$ . Physically,  $\lambda t$  is the operating time divided by the MTTF for a single component.

## 5. MTTF IS A RANDOM VARIABLE

Thus far in the discussion, the parameter  $\lambda$  has been regarded as a constant, the same value applying to all items in the population. We now consider the PDF for time to failure if  $\lambda$  is a random variable. A reasonable model for the PDF of  $\lambda$  is the gamma PDF which can assume a wide variety of shapes. Specifically, write

$$g(\lambda) = \frac{\alpha \lambda^{\alpha-1} e^{-\alpha \lambda}}{\Gamma(\alpha)} \quad (13)$$

Then, for an item selected at random, the PDF of  $t$ , the time to failure is, from Eqs (3) and (13),

$$f(t) = \frac{\alpha n}{\Gamma(n)} \int_0^{\infty} \lambda^n e^{-\lambda(\alpha+t)} d\lambda \quad (14)$$

By making the substitution

$$v = (\alpha + t)\lambda \quad (15)$$

and performing the integration, we obtain the following:

$$f(t) = \frac{n\alpha^n}{(\alpha + t)^{n+1}} \quad (16)$$

The corresponding CDF is

$$F(t) = 1 - \left( \frac{\alpha}{\alpha + t} \right)^n \quad (17)$$

and the MTTF is

$$E(t) = \frac{\alpha}{n-1} \quad (18)$$

Before discussing the practical implications of the model, we note a rather interesting and somewhat curious relationship between  $f(t)$  given by Eq. (16) and the PDF for the well-known F-distribution. Denoting the F-distributed random

TABLE II. TIME TO FAILURE DATA  
(Values of  $t$ : arbitrary units)

16.7	1.4	84.7	2.7
14.1	38.5	5.9	17.7
3.1	88.2	4.3	0.1
14.6	3.7	112.7	5.2
15.1	3.1	5.2	4.1
1.3	2.1	11.3	2.6
9.8	33.9	12.2	7.1
3.7	4.2	4.4	7.8
15.1	43.1	19.2	1.7
8.3	0.1	607.3	15.8

variable by  $w$ , its PDF for 2 and  $2\eta$  degrees of freedom is simply written:

$$h(w) = \frac{\eta}{(1+w)^{\eta+1}} \quad (19)$$

Comparing this with Eq. (16) shows that they are identical when  $w$  and  $t$  are related by the scale factor  $\alpha$ . That is, for  $t = \alpha w$ ,  $f(t)dt = h(w)dw$ .

If, in fact, the correct model is Eq. (16) with  $\lambda$  a random variable, but we assume that it is Eq. (3) with constant  $\lambda$ , the inferences about time to failure can be quite misleading. To demonstrate this, the 40 time to failure data points in arbitrary units given in Table II were generated by a random number generator based on the model (16) for  $\alpha = 12.5$  and  $\eta = 1.25$ , thus, the actual MTTF is 50 units, from Eq. (18).

If it is assumed that the exponential PDF applies, then the estimate of  $\lambda$  is given by Eq. (8), and for these data the result is

$$\hat{\lambda} = 40/1252.1 = 0.03195$$

$$\hat{MTTF} = 1/\hat{\lambda} = 31.3 \text{ units}$$

For the model whose PDF is given by (16) (henceforth referred to as the variable parameter, or VP model), the maximum likelihood estimates of  $\alpha$  and  $\eta$  are found by solving simultaneously the following equations:

$$\eta = \frac{N}{-N \ln \alpha + \sum \ln(\alpha + t_i)} \quad (20)$$

$$\frac{N\eta}{\alpha} = (\eta + 1) \sum (\alpha + t_i)^{-1} \quad (21)$$

where  $N$  is the number of data points. These equations are solved by trial and error, the solutions being given to the nearest integral value of  $\alpha$ :

$$\hat{\alpha} = 12 \quad \hat{\eta} = 1.365 \quad \hat{MTTF} = 32.9$$

A large sample goodness of fit test may be performed to see if the exponential model provides an adequate fit to the data, the comparison being with the VP model. The logarithmic likelihood functions for the two models are:

$$\text{Exponential: } L(\hat{w}) = N(\ln \hat{\lambda} - 1) = -177.74$$

$$\text{VP: } L(\hat{\Omega}) = N[\ln \hat{\eta} + \hat{\eta} \ln \hat{\alpha}] - (\hat{\eta} + 1) \sum \ln(\hat{\alpha} + t_i) = -156.19$$

The chi-square value with one degree of freedom is  $2 \times 21.55$  or 43.10, and the model fits are quite significantly different.

In life testing situations, the data are often censored, i.e. the test is terminated before all units have failed in order to shorten the test time. Naturally, such a practice requires that greater faith be placed on the validity of the assumed model.

To illustrate the effect of censoring, the data in Table II were progressively censored, first at 35 units in which the 6 largest data points were lost (except to note that the items had survived to 35 units without failure), and then at 15 units, in which case 13 data points were lost. The estimate of  $\lambda$  is still given by Eq. (8). The results are:

For censoring at 35 units,  $\hat{\lambda} = 0.0697$  ;  $\hat{MTTF} = 14.3$   
 For censoring at 15 units,  $\hat{\lambda} = 0.0796$  ;  $\hat{MTTF} = 12.3$

For the VP model, the estimating equations to solve simultaneously for the censored data are as follows, where  $t_0$  is the censoring time.

$$n = \frac{F}{-N \ln \alpha + (N-F) \ln (\alpha + t_0) + \sum_i \ln (\alpha + t_i)} \quad (22)$$

$$\frac{[(N-F)\alpha + N t_0]n}{\alpha(\alpha + t_0)} = (n + 1) \sum_i (\alpha + t_i)^{-1} \quad (23)$$

TABLE III.  $F(t)$  VERSUS  $t$  FOR SIX MODELS

t	VP model			Exponential model		
	$\alpha=12$ $\eta=1.365$	$\alpha=15$ $\eta=1.650$	$\alpha=20$ $\eta=2.022$	$\lambda=0.0319$	$\lambda=0.0697$	$\lambda=0.0796$
5	0.378	0.378	0.363	0.147	0.294	0.328
10	0.563	0.570	0.560	0.273	0.502	0.549
15	0.669	0.681	0.677	0.380	0.648	0.697
20	0.738	0.753	0.754	0.472	0.752	0.796
30	0.819	0.837	0.843	0.616	0.876	0.908
40	0.865	0.883	0.892	0.721	0.938	0.959
50	0.894	0.911	0.921	0.797	0.969	0.981
75	0.933	0.948	0.957	0.909	0.995	0.997
100	0.953	0.965	0.973	0.959	0.999	1.000

In Eqs (22) and (23), the indicated sums are only for the failed items.

For the VP model, the results are:

For censoring at  $t_0 = 35$  :  $\hat{\alpha} = 15$  ;  $\hat{\eta} = 1.650$ ;  $\hat{M}TTF = 23.1$   
 For censoring at  $t_0 = 15$  :  $\hat{\alpha} = 20$  ;  $\hat{\eta} = 2.022$ ;  $\hat{M}TTF = 12.6$

In performing the likelihood ratio tests as was done for the full data set, the likelihood equations are altered to account for the censoring. The equations for the exponential and VP model respectively are as follows:

$$\text{Exponential: } L(\hat{W}) = F(\ln \hat{\lambda} - 1) = \begin{cases} -124.56 & \text{for } t_0 = 35 \\ -95.33 & \text{for } t_0 = 15 \end{cases} \quad (24)$$

$$\begin{aligned} \text{VP: } L(\hat{\Omega}) &= F \ln \hat{\eta} + N \hat{\eta} \ln \hat{\alpha} - (N-F) \hat{\eta} \ln(\hat{\alpha} + t_0) - (\hat{\eta} + 1) \sum_1 \ln(\alpha + t_i) \\ &= -122.44 \quad \text{for } t_0 = 35 \\ &= -94.94 \quad \text{for } t_0 = 15 \end{aligned} \quad (25)$$

Note that for censoring at  $t_0 = 35$ , the chi-square value is

$$-2(-124.56 + 122.44) = 4.24$$

which is barely significant at the 5% level of significance. For  $t_0 = 15$ , the chi-square value is only 0.78, and there would be no reason to conclude that the exponential model is invalid, even though it is known to be so. Thus, as the data become more and more censored, one must have greater faith in the model, a statement that is rather obvious.

In comparing the various models and parameters, it is noted that for the VP model, the estimates  $\hat{\alpha}$  and  $\hat{\eta}$  seem to be quite dependent on the degree of censoring. However,  $\hat{\alpha}$  and  $\hat{\eta}$  are closely correlated, and it is more important to compare the estimated  $F(t)$  values than the individual estimates.  $F(t)$  is given in Table III for the six models estimated from these data. It is noted that  $F(t)$  is quite independent of the degree of censoring for the valid VP model, but is heavily dependent on the censoring when the exponential model is assumed.

If, in fact, time to failure has the exponential PDF and one attempts to fit the data with the VP model, then the estimate of  $\alpha$  would tend to infinity. The data would fit the model equally well for any number of combinations of the parameters,  $(\alpha, n)$ , with large  $\alpha$ .

## 6. WEIBULL PDF

So far in the discussion, the exponential parameter  $\lambda$  has been assumed to be constant over exposure time for a given physical unit. For some C/S devices,  $\lambda$  may be a function of the exposure time. That is, the conditional probability of failure within a given time interval may depend on the previous exposure time of the unit in question (non-constant hazard function). The same may be true for other C/S devices once a certain exposure time has been attained and wearout failures take over. In this event, the Weibull PDF applies.

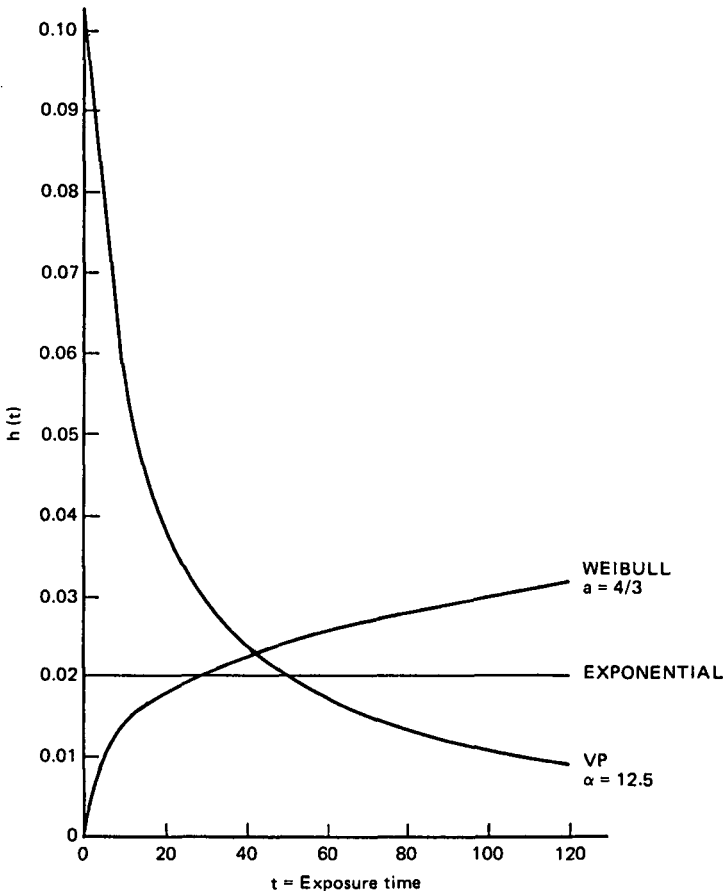


FIG. 2. Hazard function for three models. All with MTTF = 50 units.  $t$  = exposure time.



The Weibull PDF has associated with it the following functions:

$$\text{PDF: } f(t) = a\lambda t^{a-1} e^{-\lambda t^a} \quad (26)$$

$$\text{CDF: } F(t) = 1 - e^{-\lambda t^a} \quad (27)$$

$$\text{hazard: } h(t) = \frac{a\lambda t^{a-1} e^{-\lambda t^a}}{e^{-\lambda t^a}} = a\lambda t^{a-1} \quad (28)$$

$$\text{MTTF: } E(t) = (1/\lambda)^{1/a} \Gamma(1/a + 1) \quad (29)$$

By comparing the results with the corresponding results for the exponential PDF, Eqs (3) to (6), it is seen that for  $a = 1$  the Weibull PDF is the same as the exponential PDF. Also, if exposure time is measured in  $t^a$  units rather than in  $t$  units, the PDF of  $t^a$  is exponential. This assumes an a priori knowledge of the parameter  $a$ .

For a complete data set, it is easy to estimate the parameters  $a$  and  $\lambda$  by graphical methods. Note from Eq. (27) that

$$Z = \ln\{1 - F(t)\} = \ln \lambda + a \ln t \quad (30)$$

so that by plotting  $Z$  versus  $\ln t$  a straight line is found, and the slope estimates the parameter  $a$  while the intercept estimates  $\ln \lambda$ . In calculating  $Z$ , the quantity  $F(t)$  for the  $i$ -th ordered point is simply  $(i - 0.5)/N$ .

Estimation of the parameters can also be accomplished by maximum likelihood, either for censored data or for complete data. In either event, the solution of two simultaneous non-linear equations is required, similar to the estimation procedure for the VP model.

Unfortunately, it can be very difficult to determine from a limited set of data, part of which may well be censored, just what is the appropriate model. The more limited the data, the more one must rely on a priori information, including judgment as to which set of assumptions would appear to be more valid. It is important to recognize the assumptions that are implied for a given model, and to exercise some caution in the interpretation of limited test data where the statistical analysis is based on poorly founded assumptions.

Figure 2 illustrates how three models, all with the same MTTF, can differ appreciably with respect to their hazard functions.

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## UNCERTAINTY ESTIMATES FOR CALIBRATION MEASUREMENTS

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### Abstract

#### UNCERTAINTY ESTIMATES FOR CALIBRATION MEASUREMENTS.

The volume of liquid in a process tank is typically determined from a calibration function that relates tank volume to some measure of liquid height. This calibration equation is determined from data acquired during one or more calibration runs. In the paper, solutions are proposed for two statistical problems that arise when the calibration function is derived and used. The first is that of computing plausible variance estimates if the data exhibit statistically significant run-to-run differences. The second is that of computing confidence intervals that hold simultaneously for an arbitrary number of volume determinations. When statistically significant run-to-run differences are ignored and standard statistical procedures are applied to data, resulting variance estimates can seriously underestimate actual measurement variability. In the proposed procedure, the variance of a predicted new response is estimated as the sum of two components: one due to run-to-run differences and one due to the fitted model. Depending upon the nature of the model(s) required to fit the data (a single function, parallel functions or individual non-parallel functions), estimates of the response variance components take one of three forms. For linear functions, estimates of component variances are derived from sums of squares that are computed during an analysis of covariance. The estimated calibration function is ultimately used to determine the volumes that correspond to observed liquid heights. Thus, confidence intervals are required that hold simultaneously for arbitrarily many volume determinations. A procedure is presented for computing simultaneous discrimination intervals. It employs variance estimates computed as described above in Scheffé-type simultaneous prediction intervals, which are combined with tolerance intervals on the response variable (liquid height) to obtain equations that can be solved to obtain the desired discrimination intervals for volume determinations.

## 1. INTRODUCTION AND SUMMARY

Accurate volume measurements are an essential component of any system to control and account for nuclear materials that are processed or stored in a liquid medium. The volume of liquid in a process tank is determined from a calibration equation that relates volume to some measure of liquid height. This calibration equation is determined from data obtained during one or more calibration runs. During each run, carefully measured increments of liquid are added to the tank. These volume measurements, together with observations of the corresponding liquid heights, are the basic data used to derive the calibration equation. In practice, liquid height is often measured indirectly - via pressure, for example. For convenience, however, we shall use the term "liquid height" to refer to the tank's measurement system response. A detailed discussion of the RITCEX tank calibration study is given by Franssen [1].

In this paper, we present solutions to two problems that frequently arise during the estimation and subsequent use of a calibration function. The first is that of obtaining correct variance estimates when the calibration data exhibit statistically significant run-to-run differences. In a study of the RITCEX data, Sellinschegg et al. [2] allude to this problem, but give no solution. The second problem is that of computing uncertainty estimates for volume measurements when liquid height is taken as the response variable. The use of standard textbook results to "solve" this problem often produces unrealistic estimates.

With many calibration systems, liquid height and volume measurements can be made with such precision that

run-to-run differences, presumably due to uncontrolled ambient conditions, are statistically greater than within-run measurement errors. When these differences are not significant from a physical point of view, they are usually ignored and the data from all runs are combined. However, when these differences are ignored, the use of standard statistical methods can result in serious underestimates of measurement variability. In Section 3 we present a method that yields acceptable variance estimates in the situation just described. For linear calibration functions, the estimates are based upon sums of squares that are computed to test for equal slopes and equal intercepts as part of an analysis of covariance (See [3], p. 376, for example). Similar estimates can be computed for polynomial calibration functions of higher degree (See [4], for example).

For data obtained by a calibration system that measures volume more precisely than liquid height, liquid height is usually taken as the response variable when a calibration equation is fit to the data<sup>1</sup>. However, the fitted calibration equation is used in reverse: Volume measurements are computed for observed liquid height measurements. Likewise, uncertainty estimates for volume measurements are obtained by "inverting" those obtained for liquid height measurements during model-fitting. A method for constructing and inverting simultaneous confidence statements is presented in Section 4. The method combines the variance estimates proposed in Section 3 and tolerance intervals on the response variable with Scheffé-type simultaneous prediction intervals.

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<sup>1</sup> It is noted that Carroll and Spiegelman [5] make a strong case for using errors-in-variables models under these circumstances.

Before a calibration function can be developed, the raw calibration data usually require adjustment to a standard set of conditions (because of temperature differences observed during calibration, for example). Moreover, the data may require "alignment" to compensate for differences in the volume of liquid in the tank at the start of each calibration run. It is assumed that all necessary data standardization has previously been accomplished. Reasons for alignment are given by Sellinschegg et al. [2]. A recent paper by Jones [6] discusses data normalization for tanks equipped with pressure measurement systems. Methods for aligning data and checking for internal consistency are presented in a paper by Goldman and Liebetrau [7].

## 2. MODEL-FITTING AND ESTIMATION

The variance estimates given in Section 3 are based on the sums of squares derived from the model-fitting steps discussed in this section. Let  $X_{ij}$  be the standardized cumulative volume of the first  $i$  ( $i = 1, 2, \dots, n$ ) increments of calibration liquid added to the tank during run  $j$  ( $j = 1, 2, \dots, r$ ), and let  $Y_{ij}$  be the corresponding standardized liquid height. We assume for simplicity that the number of volume increments is the same for all runs.

When liquid height ( $Y$ ) is the response variable and volume ( $X$ ) is the control variable, the calibration function is estimated by fitting an equation of the form  $Y = f(X; \beta_0, \beta_1, \dots, \beta_k)$  to the standardized calibration data. We assume that classical least-squares regression is appropriate for estimating the unknown parameters  $\beta_0, \beta_1,$

...,  $\beta_k$ , and further, that all measurement errors have normal (Gaussian) distributions. These assumptions must be verified for each calibration, but they are plausible for the data with which the authors are familiar. Because nearly all experimenters use only linear and quadratic functions, we restrict attention to these cases and use the linear case for illustration.

A calibration function is typically estimated piecewise, with a first or second degree polynomial being fit to each segment. Because it is not necessary to identify particular segments for our purpose, all references to segment number are suppressed to simplify notation.

For computing variance estimates, the sums of squares from certain model-fitting steps in an analysis of covariance are required. It is first necessary to select the functional form of the model to be fitted, either linear or quadratic. At this point, the reader may wish to refer to a textbook that gives formulas for analysis of covariance (See [3], for example).

Step 1: Fit the selected function individually to the data of each calibration run. Denote the corresponding total pooled sum of squared residuals by  $SSE(1)$ .

Step 2: Fit "parallel" functions to the data, one function for each calibration run. In this context, the word "parallel" refers to functions for which all coefficients, except intercepts, are estimated jointly from the pooled data from all runs. Let  $SSE(2)$  denote the corresponding pooled sum of squared residuals. The difference

$$D_1 = SSE(2) - SSE(1)$$

is a measure of the increased lack-of-fit, relative to that for Step 1, when parallel functions are fit to the data.

Step 3: Fit a single function to the pooled data from all runs, and let  $SSE(3)$  denote the pooled sum of squared residuals. The difference

$$D_2 = SSE(3) - SSE(2)$$

measures the increased lack-of-fit, relative to that for parallel functions, when a single function is fit to the data.

The degrees of freedom associated with  $SSE(1)$ ,  $SSE(2)$ , and  $SSE(3)$  are, respectively,

$$df(1) = r(n-k-1) ,$$

$$df(2) = (n-1)r-k = nr-(k+r) ,$$

$$df(3) = nr-(k+1) ,$$

where

$r$  = numbers of runs,

$n$  = number of data pairs per run,

$k$  = degree of the polynomial being fitted.

Note that  $k=1$  for a linear function,  $k=2$  for a quadratic function, and that the number of parameters to be estimated in either case is  $k+1$ .

The statistic

$$T_1 = \left[ \frac{SSE(2)-SSE(1)}{df(2)-df(1)} \right] \div \left[ \frac{SSE(1)}{df(1)} \right] \quad (1)$$



can be used to test the hypothesis that parallel functions adequately fit the data. Under the assumption that errors are normally distributed,  $T_1$  has an F-distribution with parameters  $df(2)-df(1) = k(r-1)$  and  $df(1) = r(n-k-1)$ . Similarly, if it is determined that parallel functions adequately fit the data, i.e., if  $T_1$  is not significant, then the statistic

$$T_2 = \left[ \frac{SSE(3)-SSE(2)}{df(3)-df(2)} \right] \div \left[ \frac{SSE(2)}{df(2)} \right] \quad (2)$$

can be used to test the hypothesis that a single function adequately fits the data. The statistic  $T_2$  has an F-distribution with parameters  $df(3)-df(2) = r-1$  and  $df(2) = rn-(k+r)$  under the stated assumptions. The significance of the statistics  $T_1$  and  $T_2$  is used to determine the form of the variance estimates given in the next section.

### 3. VARIANCE ESTIMATES

#### 3.1. Preliminaries

Let  $Y_0 = \hat{f}(X_0; b_0, b_1, \dots, b_k)$  be the predicted mean liquid height for a given volume  $X=X_0$ , where  $\hat{f}$  is obtained by fitting the pooled data from all runs, and  $b_0, \dots, b_k$  are the least-squares estimators of  $\beta_0, \dots, \beta_k$ . The variance of a new individual future predicted liquid height measurement  $Y$ , given  $X=X_0$ , is

$$\sigma^2(Y|X_0) = \sigma_M^2(Y_0|X_0) + \sigma_E^2(Y|X_0) , \quad (3)$$

where  $\sigma_M^2$  is the variance due to  $\hat{f}(X_0; b_0, \dots, b_k)$  and  $\sigma_E^2$  is the variance of the (conditional) random variable  $Y|X_0$ . For the linear least-squares regression model,

$$\sigma^2(Y|X_0) = \sigma^2 \left[ \frac{1}{nr} + \frac{(X_0 - \bar{X})^2}{S_2} \right] + \sigma^2, \quad (4)$$

where  $S_2 = \sum_{ij} (X_{ij} - \bar{X})^2$  and  $\bar{X} = \sum_{ij} X_{ij} / (nr)$ . We write  $\sigma_E^2(Y|X_0) = \sigma^2$  in this case because  $\sigma_E^2(Y|X_0)$  does not depend upon  $X_0$ . The variance  $\sigma^2$  is usually estimated by the residual ("error") mean square, which is given by

$$S^2 = \text{SSE}(3)/\text{df}(3) \quad (5)$$

An expression analogous to (4) can be written for polynomial equations of higher degree.

Typically,  $\sigma^2(Y|X_0)$  is estimated from (5) without regard to the question of whether the calibration data contain statistically significant run-to-run differences. The result is that true measurement variability may be seriously underestimated.

### 3.2. Estimation of Component Variances

In this section, we present estimators of  $\sigma^2$ , the "pure error" factor of model variance, and  $\sigma_E^2(Y|X_0)$ , the variance of the response variable, that are appropriate for the situation just described. The estimators, denoted by  $S^2$  and  $S_E^2(Y|X_0)$  respectively, depend upon the significance of the test statistics  $T_1$  and  $T_2$ . There are three cases to consider.

Case 1: Neither  $T_1$  nor  $T_2$  is significant. A single function fits the pooled data in this case. Equivalently,

run-to-run variability is not significantly greater than random measurement variability.

Case 2: The statistic  $T_2$  is significant, but  $T_1$  is not. In this case, parallel functions fit the pooled data, but a single function does not. Equivalently, there is significant run-to-run variability, but it is due solely to differences in intercepts.

Case 3: The statistic  $T_1$  is significant. Individual (non-parallel) functions are required to adequately fit the data in this case, and lack of parallelism in the individual calibration runs contributes significantly to run-to-run differences.

For each of these three cases, estimates of  $\sigma^2$  and  $\sigma_E^2(Y|X_0)$  are given in Table I, together with appropriate degrees of freedom. Note that the usual least-squares estimators are used only in Case 1, where run-to-run differences are not significant.

In Case 3,  $\sigma_E^2(Y|X_0)$  depends upon  $X_0$  because the individual lines are not parallel. This dependence is modeled as follows: First,  $\sigma_E^2(Y|\bar{X})$ , the variance of the response variable at  $X=\bar{X}$ , is estimated by

$$s_E^2(Y|\bar{X}) = \frac{1}{n} \left[ \frac{SSE(2)-SSE(1)}{df(2)-df(1)} + \frac{SSE(3)-SSE(2)}{df(3)-df(2)} \right] = \frac{1}{n}(A+B) \quad (6)$$

Next, this estimator is multiplied by an appropriate scale factor to estimate  $\sigma_E^2(Y|X_0)$  at other values  $X=X_0$ . The resulting estimator is

$$s_E^2(Y|X_0) = c^2 s_E^2(Y|\bar{X}) \quad (7)$$

Finally, note that the terms A and B in (6) may be significantly different. Consequently, the degrees of

TABLE I. ESTIMATORS OF VARIANCE COMPONENTS

Case	Model	Variance estimator		Degree of freedom	
		$S^2$	$S_E^2(Y X_0)$	$S^2$	$S_E^2(Y X_0)$
1. $T_1$ not significant $T_2$ not significant	Single function	$\frac{SSE(3)}{df(3)}$	$\frac{SSE(3)}{df(3)}$	$rn - k - 1$	$rn - k - 1$
2. $T_1$ not significant $T_2$ significant	'Parallel' functions	$\frac{SSE(2)}{df(2)}$	$\frac{SSE(3) - SSE(2)}{n[df(3) - df(2)]}$	$rn - (r + k)$	$r - 1$
3. $T_1$ significant	Individual function each run	$\frac{SSE(1)}{df(1)}$	$\frac{C^2}{n}(A + B)$	$r(n - 2)$	$\frac{(A + B)^2}{\frac{A^2}{df(A)} + \frac{B^2}{df(B)}}$

$$A = \frac{SSE(2) - SSE(1)}{df(2) - df(1)}, \quad df(A) = (r - 1)k$$

$$B = \frac{SSE(3) - SSE(2)}{df(3) - df(2)}, \quad df(B) = r - 1$$

$C$  is an appropriate scale factor (see text)

freedom for (7) is computed using Satterthwaite's formula [3]. This procedure gives the correct degrees of freedom even though A and B are not significantly different. Unfortunately, the choice of a scale factor may involve some trial-and-error. If the standard deviation of the residuals is approximately proportional to volume, for example, then  $C = X_0/\bar{X}$  is a suitable choice.

### 3.3. Estimation of Response Variance

With the aid of appropriate estimates of  $\sigma^2$  and  $\sigma_E^2$ , the variance of a predicted liquid height measurement  $Y$  is estimated by analogy with (2):

$$S^2(Y|X_0) = S^2 \left[ \frac{1}{nr} + \frac{(X_0 - \bar{X})^2}{S_2} \right] + S_E^2(Y|X_0) \quad (8)$$

In (8),  $S^2$  estimates the "pure error" factor of the model variability component, and  $S_E^2$  estimates the variance of the response variable  $Y|X_0$ .

Unless estimators  $S^2$  and  $S_E^2(Y|X_0)$  are determined according to Case 1, they are significantly different. Thus, in Cases 2 and 3, the degrees of freedom for (8) are estimated by applying Satterthwaite's formula to the components involving  $S^2$  and  $S_E^2(Y|X_0)$ . For Case 2, these two estimators have degrees of freedom  $nr-(r+k)$  and  $(r-1)$ , respectively. For Case 3, the estimator  $S^2$  has degrees of freedom  $r(n-2)$ , while the degrees of freedom for  $S_E^2(Y|X_0)$  is determined by Satterthwaite's formula (see Table I).

The interpretation of  $S_E^2(Y|X_0)$  in (8) is somewhat different from the initial interpretation of  $\sigma_E^2(Y|X_0)$  in (3). The variance  $\sigma_E^2(Y|X_0)$  was initially interpreted as the variance of an observation from the distribution of  $Y$ . In (8), however, the estimator  $S_E^2(Y|X_0)$  is essentially a measure of the between-run variance of the calibration data. The initial interpretation holds only in Case 1, where within-run and between-run variances are not significantly different. To emphasize this point, we shall henceforth use the notations  $S_R^2(Y|X_0)$  and  $\sigma_R^2(Y|X_0)$  instead of  $S_E^2(Y|X_0)$  and  $\sigma_E^2(Y|X_0)$ .

### 3.4.Example

Figure 1 shows a profile plot of an actual set of aligned calibration data. A profile plot is simply a plot of the residuals (versus  $X$ , and connected for each run)

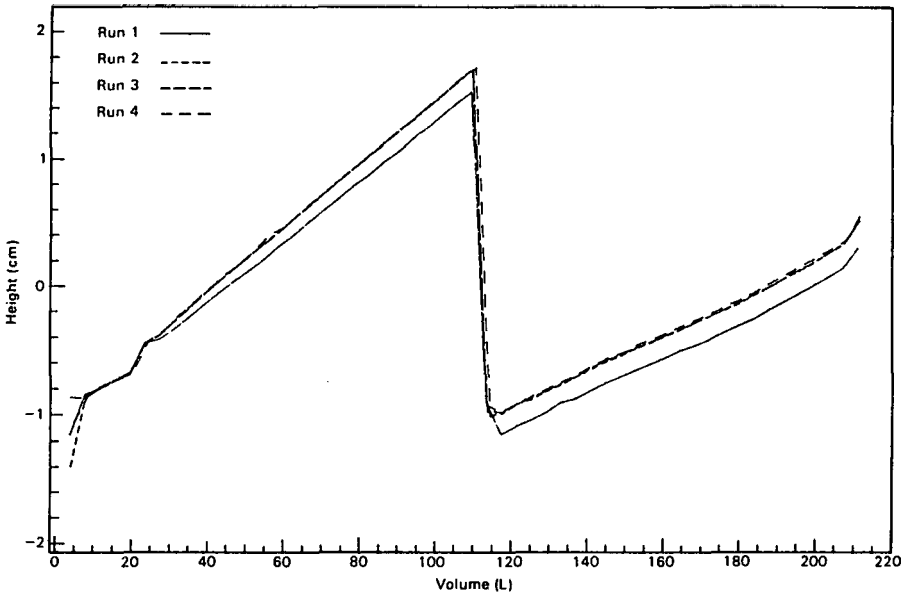


FIG. 1. Profile plot of liquid height measurements.

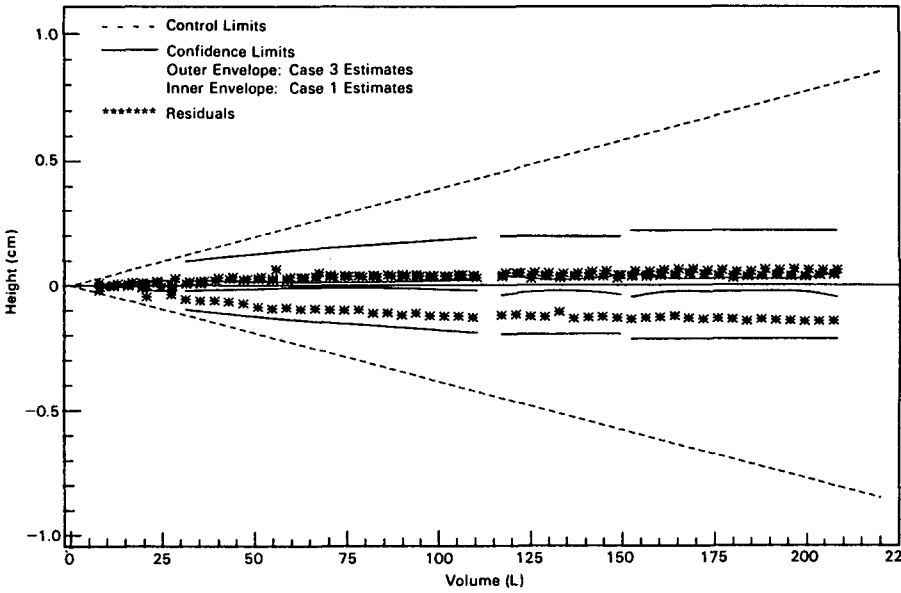


FIG. 2. Control limits, confidence limits and residuals from fitted calibration function.

TABLE II. SUMS OF SQUARES (SSE) AND DEGREES OF FREEDOM (df) FOR THE SEGMENT CONSISTING OF INCREMENTS 8-28

Model	SSE	df
Individual functions, each run	$SSE(1) = 0.0033303$	$df(1) = 76$
Parallel functions, each run	$SSE(2) = 0.0166503$	$df(2) = 79$
Single function, all runs	$SSE(3) = 0.2839689$	$df(3) = 82$

resulting from a straight line fit to the entire set of calibration data. Thus, a profile plot clearly shows the shape (profile) of the calibration function. It is apparent from Figure 1 that the residuals from Run 1 are smaller than those from the other three runs. The discrepancy between Run 1 and the others is a major source of variability.

After examining the residuals more closely, it was decided that the data could be adequately fit in three major segments. The three segments consist of increments 8-28 (30-110 L, approximately), increments 29-38 (114-150 L) and increments 39-53 (150-208 L). The first two segments were fit with linear functions, but the third required a quadratic polynomial. Residuals from these fits are shown in Figure 2, together with those from fits for several smaller segments (not discussed in this paper). As in Figure 1, it is evident that the residuals from the first run deviate from those for other runs. Figure 2 also shows the confidence limits for individual measurements derived by the methods of the preceding section; computational details for the first segment are given below. The V-shaped envelope indicates measurement accuracy criteria which were established for this tank.

For the segment consisting of increments 8-28, Table II shows the numerical values of the sum of squares

identified in Section 2. For these data, the values of (1) and (2) are  $T_1 = 101.33$  and  $T_2 = 1067.42$ . Since  $T_1$  is significantly large, the Case 3 estimates in Table I are appropriate. For illustrative purposes, variance estimates are calculated at the mean ( $\bar{Y}$ ,  $\bar{X}$ ). The estimate of random measurement variance is

$$s^2 = 0.0033303/76 = 0.0000438 \quad (9)$$

and the estimate of run-to-run variability is

$$\begin{aligned} s_R^2(\bar{Y}|\bar{X}) &= \frac{1}{21} \left[ \frac{0.0166503 - 0.0033303}{3} \right] + \left[ \frac{0.2839689 - 0.0166503}{3} \right] \\ &= 0.0044546 \end{aligned} \quad (10)$$

From Satterthwaite's formula, the degrees of freedom for  $s_R^2(\bar{Y}|\bar{X})$  is 3.30. Substitution of (9) and (10) into (8) yields

$$s^2(\bar{Y}|\bar{X}) = 0.0000438 \left( \frac{1}{84} \right) + 0.0044546 = 0.004455 \quad (11)$$

The degrees of freedom for  $s^2(\bar{Y}|\bar{X})$  is computed to be 3.36. For the choice  $C = X/\bar{X}$ , the 90% confidence limits computed from (8), (9), and (10) yield the outer envelope shown in Figure 2.

Had run-to-run differences been ignored, and had Case 1 estimates been used (incorrectly) to compute  $s^2(\bar{Y}|\bar{X})$ , the result would be

$$s^2(\bar{Y}|\bar{X}) = \frac{0.2839689}{82} \left( \frac{1}{84} \right) = 0.00004123 \quad (12)$$

with corresponding degrees of freedom 82. By comparing (11) and (12), it is clear that ignoring run-to-run differ-



ences results in overestimation of the first term in (8) and underestimation of the second. When the second term dominates, measurement variability is underestimated, and this example shows that the error can be extremely large. Corresponding confidence intervals are even more disparate than variance estimates because of the large differences in degrees of freedom. The discrepancy may be seen in Figure 2 by comparing the outer envelope of confidence intervals (Case 3) with the inner envelope, the latter obtained by using Case 1 estimates of  $\sigma^2$  and  $\sigma_R^2(Y|X)$ .

#### 4. DISCRIMINATION INTERVALS FOR VOLUME DETERMINATIONS

A calibration function, once developed, is used repeatedly to make volume determinations. Thus, a method is required for computing prediction intervals that hold simultaneously with specified probability for an indeterminate number of volume determinations. Methods exist (see [3] or [8], for example) for computing simultaneous confidence bands about the regression line. However, if liquid height is the chosen response variable, (typically the case when volume is measured more precisely than liquid height), then these confidence limits must somehow be inverted to give the required prediction intervals for volume measurements.

The procedure described here is a modification of one recommended by Miller [8] for computing simultaneous discrimination intervals. The procedure combines variance estimates from the previous section and tolerance intervals on the response variable  $Y$  with Scheffé-type simultaneous prediction intervals. The discussion is restricted to linear calibration functions  $Y = \beta_0 + \beta_1 X$ .

#### 4.1. Prediction Intervals about the Regression Line

The first step is to derive simultaneous prediction intervals about the estimated regression line. Unlike those proposed by Miller [8], however, the prediction intervals used here employ the variance estimates derived in the previous section. Thus, we assume that the confidence band on the regression line

$$\beta_0 + \beta_1 X \in b_0 + b_1 X \pm \left( 2 F_{2,v}^{\alpha/2} \right)^{1/2} \left[ S^2 \left( \frac{1}{nr} + \frac{(X-\bar{X})^2}{S_2} \right) + S_R^2(Y|X) \right]^{1/2} \quad (13)$$

holds (approximately) with probability  $(1-\alpha/2)$  for all  $X$ . In (13),  $F_{2,v}^{\alpha/2}$  is the  $(1-\alpha/2)$  percentile of the  $F$ -distribution with 2 and  $v$  degrees of freedom,  $S^2$  and  $S_R^2(Y|X)$  are computed by the methods of the previous section, and  $v$  is the degrees of freedom for the quantity  $S^2(Y|X)$  in brackets in (13) [see Table I and Equation (8)].

#### 4.2. Tolerance Intervals on the Response Variable

The second step is to derive simultaneous tolerance intervals on the response variable  $Y$ . Let

$z_{1-p/2}$  = the  $(1-p/2)$  percentage point of the standard normal distribution,

$S'$  = the estimated standard deviation of future observations on  $Y$ ,

$v'$  = the degrees of freedom associated with  $S'$ ,

and let

$\chi_{v'}^{\alpha/2}$  = the lower  $(\alpha/2)$  percentile of the chi-square distribution with  $v'$  degrees of freedom.

Then the following family of tolerance intervals hold simultaneously with probability  $(1-\alpha/2)$ :

$$Y \pm z_{1-p/2} \left( v' / \alpha/2 \chi_{v'}^2 \right) S' \quad (14)$$

The tolerance intervals have the following interpretation: All intervals in the family given by (14) cover 100p percent of the probability distribution of  $Y$  with probability  $(1-\alpha/2)$ . By the Bonferroni inequality, (13) and (14) hold simultaneously with probability at least  $(1-\alpha)$ .

The estimation of  $\sigma'^2$ , the variance of a future response variable measurement, requires comment. In cases where the method for making future liquid height measurements is the same as that used for calibration,  $\sigma'^2$  can be estimated from the calibration data. In these cases, the "pure error" factor of the model component is used to estimate  $\sigma'^2$ . Thus,  $S' = S$ , where  $S^2$  is defined in Section 3. The computational form for  $S^2$  and the associated degrees of freedom are given in Table I. In Case 1, for example,  $S'^2 = \text{SSE}(3)/(nr-2)$  and  $v' = nr-2$ . The results in this case are identical to those given by Miller [8].

In the volume calibration context, however, it may well happen that liquid height measurements are made differently during process operation than during calibration. In this case,  $\sigma'^2$  should be estimated from a sample of new liquid height measurements  $Y_1, \dots, Y_m$  that are taken with the process method. In this case,

$$S'^2 = \sum_{i=1}^m (Y_i - \bar{Y})^2 / (m-1) \quad \text{and} \quad v' = m-1$$

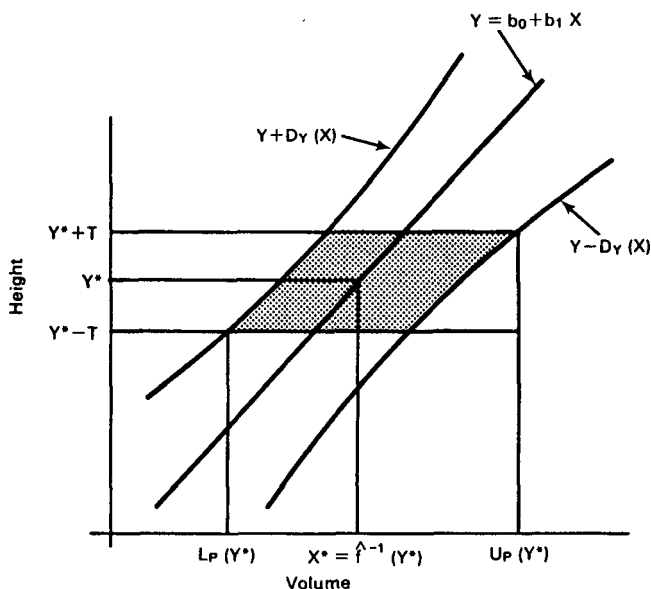


FIG. 3. Determination of discrimination intervals.

**Note:**  $D_Y(X) = (2F_{2,\nu}^{\alpha/2})^{1/2} \left[ S^2 \left( \frac{1}{nr} + \frac{(X - \bar{X})^2}{S_2} \right) + S_R^2(Y|X) \right]^{1/2}$

[see (13)]

$$T = z_{1-p/2} (\nu' / \alpha / 2 \chi_{\nu'}^2)^{1/2} S'$$

[see (14)]

#### 4.3. Discrimination Intervals for the Control Variable

Finally, the intervals (13) and (14) are combined to obtain discrimination intervals for volume determinations. For a given liquid height  $Y=Y^*$ , a discrimination interval about  $X^* = \hat{f}^{-1}(Y^*)$  is obtained by projecting the intersections of the tolerance intervals (14) centered at  $Y^*$  with the confidence bands (13) about the line  $Y = b_0 + b_1 X$  onto the  $X$ -axis as shown in Figure 3. If  $b > 0$ , the lower limit  $L_P(Y^*)$  is the  $X$ -value that satisfies the equation

$$\begin{aligned} b_0 + b_1 X + D_Y(X) \\ = Y^* - z_{1-p/2} (\nu' / \alpha / 2 \chi_{\nu'}^2)^{1/2} S' \end{aligned} \quad (15)$$

where

$$D_Y(X) = \left( 2 F_{2,v}^{\alpha/2} \right)^{1/2} \left[ S^2 \left( \frac{1}{nr} + \frac{(X-\bar{X})^2}{S_2} \right) + S_R^2(Y|X) \right]^{1/2}$$

and the upper limit  $U_p(Y^*)$  is the solution to the equation

$$\begin{aligned} b_0 + b_1 X - D_Y(X) \\ = Y^* + z_{1-p/2} \left( v' / \alpha/2 \chi_{v'}^2 \right)^{1/2} S' \end{aligned} \quad (16)$$

If  $b < 0$ , then the solutions to (15) and (16) yield the lower and upper discrimination limits, respectively; this case is not relevant to the volume calibration problem. The discrimination intervals  $[L_p(Y^*), U_p(Y^*)]$  obtained as solutions to Equations (15) and (16) have the following interpretation: For each calibration function estimated from the data, the probability is  $(1-\alpha)$  that 100p percent of the discrimination intervals will contain the true  $X$ 's, i.e., the true volumes. If  $\alpha = 0.95$  and  $p = 0.99$ , for example, there is a 95% chance that 99% of the discriminations made with the estimated regression line will cover the true volumes.

The solution of Equations (15) and (16) may become somewhat complicated for cases in which  $S_R^2(Y|X)$  depends upon  $X$ . It is convenient to replace  $S_R^2(Y|X)$  by  $S_R^2(Y^*|X^*)$  in these cases. It has been the authors' experience that this substitution introduces negligible error and simplifies calculations.

#### 4.4. Example

We continue with the example of Section 3.4, and specifically consider the segment consisting of increments 8-28 (30-110 L, approximately). For the interval

TABLE III. QUANTITIES REQUIRED TO COMPUTE DISCRIMINATION INTERVALS

Quantity	Value	How obtained
$\bar{X}$	70.786	From calibration data
$S_2$	47145.5	From calibration data
$nr$	84	From calibration data
$S^2$	0.00004382	Method of Section 3 (See Table I)
$S_R^2(100 X)$	$\frac{53.847}{70.786}(0.0044551)$	Method of Section 3 (See Table I)
$= \frac{X^*}{X} S_R^2(\bar{Y} \bar{X})$	$= 0.0033859$	
$\nu$	3.36	Method of Section 3 (See Table I)
$F_{2,\nu}^{0.975}$	39.2	Tables of the F-distribution
$S'$	S	From calibration data or independent experiment
$\nu'$	76	From calibration data or independent experiment
$0.025 \chi_{\nu'}^2$	53.8 (approx.)	Tables of the $\chi^2$ -distribution
$z_{0.99}$	2.33	Tables of the standard normal distribution

$27.6 \leq X \leq 110.6$ , or  $47.5 \leq Y \leq 213.5$ , the following equation was fitted to the calibration data:

$$Y = 1.998672 * X - 7.621975$$

Now, suppose the liquid height  $Y^* = 100$  is observed using the same measurement method that was used for calibration. The corresponding volume determination is  $X^* = 53.847$ . A 95% discrimination interval with 98% coverage is required. Table III identifies the quantities required to set up Equations (15) and (16), gives their values for this example, and summarizes how they are

obtained. The lower discrimination limit  $L_{0.98}(100) = 53.82$  is obtained by solving the following equation for  $X$ :

$$1.998672 * X - 7.621975 + (2*39.2)^{1/2} \left[ 0.000043820 \left( \frac{1}{84} + \frac{(X-70.786)^2}{47145.5} \right) + 0.0033859 \right]^{1/2} = 100 - 2.33 (76/53.8)^{1/2} (0.00004382)^{1/2}$$

Similarly,  $U_{0.98}(100) = 53.87$

## 5. CONCLUDING REMARK

Procedures have been presented for dealing with two problems frequently encountered in the development and use of a tank volume calibration function, namely that of obtaining reasonable variance estimates when run-to-run differences are present and that of deriving simultaneous "confidence" intervals for volume measurements. Although the methods proposed here have their origins in well-established and widely-used statistical theory, they must of themselves be regarded as somewhat empirical. Nevertheless, the authors have found that the methods work quite well for calibration data with which they are familiar.

## ACKNOWLEDGEMENTS

The authors wish to acknowledge D. J. Bates for assistance with computations and S. L. Cammann for her efficient preparation of this manuscript.

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## THE DoD METHOD OF MEASUREMENT DATA EVALUATION

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### Abstract

#### THE DoD METHOD OF MEASUREMENT DATA EVALUATION.

Theoretical considerations and practical applications of the DoD method of measurement data evaluation are presented. The method proves to be a reasonable way of estimating meaningful standard deviations without the necessity of rejecting outliers. It therefore always leads to the same values, regardless of the statistician who performed the evaluation. Graphical display of the method is helpful for judging measurement discrepancies and can be used as a basis for data verification purposes.

### 1. INTRODUCTION

During recent years the DoD method was developed at KfK primarily as an empirical approach of measurement data evaluation [1-3]. With this method the absolute differences of the results of repetitive measurements are used as the basis for statistical data treatment. Recent theoretical studies have shown that the DoD method delivers solid estimates for the standard deviations of normally distributed groups of data [4, 5].

The main advantage of the method is that no application of outlier criteria for the data analysis becomes necessary. Other than in the conventional computing method, the value of the estimate derived for the standard deviation of a group of data is influenced above all by the number, and very little by the quality of existing outliers.

This feature is of particular importance for evaluating analytical inter-laboratory experiments. It is unsatisfactory to suppress analytical measurement values for merely statistical reasons; however, using evaluation techniques such as variance analysis, outlier rejection is indispensable for obtaining sufficiently homogeneous data material.

Furthermore, the selection of the outlier criterion among the many methods described in the literature, as well as the arbitrariness in fixing the threshold for data elimination, introduce a considerable ambiguity in the statistical estimation of expectations. Being independent of the handling of outliers, the estimation

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of standard deviations by the DoD method always leads to the same figures, regardless of the statistician who performed the evaluation. For safeguards in particular, this is of practical value.

## 2. MATHEMATICAL-STATISTICAL BACKGROUND

The DoD method delivers a robust estimate for the standard deviation of a normally distributed random variable  $X$ . In the following, the mathematical-statistical basis for the estimation method is presented.

It is assumed that there is a simple random sample of random variables  $X_1, X_2, \dots, X_n$ . The  $X_i$  are normally distributed with identical mean values and a given standard deviation  $\sigma$ . Furthermore, it is assumed that  $n$  is an even number, i.e.  $n = 2m$ ; and that a new random sample is defined as

$$Z_k = X_{2k} - X_{2k-1}, \quad k = 1, 2, \dots, m. \quad (1)$$

If the sample has an odd number, the last value is neglected. Now  $Z_1, Z_2, \dots, Z_m$  are also independent identically distributed random variables. For the distribution of  $Z_k$ , we get

$$H(z) = \begin{cases} 2\Phi(z/(\sigma\sqrt{2})) - 1, & \text{for } z \geq 0 \\ 0, & \text{otherwise,} \end{cases} \quad (2)$$

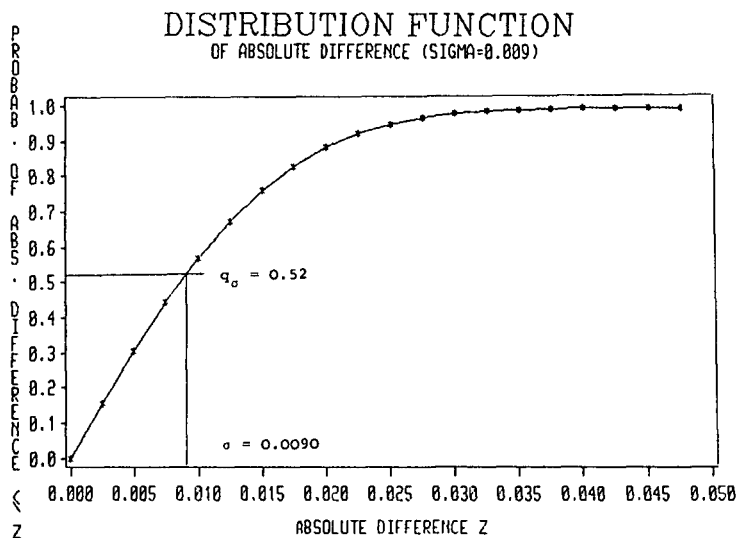


FIG. 1. Example of the distribution function  $H(z)$  relating to differences.

where  $\Phi(\cdot)$  denotes the normal distribution function with expectation value 0 and standard deviation 1. From Eq. (2) we get at  $\sigma$

$$q_\sigma = H(\sigma) = 2\Phi(1/\sqrt{2}) - 1 \approx 0.52 \quad (3)$$

Summarily it can be stated that  $H(z)$  is a strictly increasing distribution function (see also Fig. 1) and that  $\sigma$  is the  $q_\sigma$  quantile of  $H(z)$ . According to Fig. 1 a suggestion for the estimation of  $\sigma$  is the inverse function of  $H$  evaluated at  $q_\sigma = 0.52$ , i.e.  $H^{-1}(q_\sigma)$ .

Because  $\sigma$  is unknown (it has to be estimated),  $H^{-1}(q_\sigma)$  cannot be evaluated. So the empirical distribution  $H_m^*(z)$ , which is defined as

$$H_m^*(z) = (1/m) \sum_{i=1}^m I(Z_i \leq z) \quad (4)$$

may be used, where  $I$  stands for the indicator variable. The inverse function of  $H_m^*$  is defined as

$$H_m^{*-1}(u) = \inf\{z : H_m^*(z) \geq u\} \quad (5)$$

where  $0 \leq u \leq 1$ .

In our case, this line of reasoning leads to the following definition for an estimation function for  $\sigma$  [4]:

$$\text{DODU} = H_{m=n/2}^{*-1}(q_\sigma) \quad (6)$$

In this connection, it is very helpful that  $H_m^{*-1}(u)$  can be written as order statistics. If we have the simple random sample  $Z_1, Z_2, \dots, Z_m$ , then the sample function  $Z_{(k)}$ ,  $1 \leq k \leq m$ , denotes the  $k^{\text{th}}$  position of the ordered sample, i.e.

$$Z_{(1)} \leq Z_{(2)} \leq \dots \leq Z_{(k)} \leq \dots \leq Z_{(m)} \quad (7)$$

Using Eq. (7) the estimate in Eq. (6) may be written as

$$\text{DODU} = Z_{([q_\sigma \cdot m] + 1)} \quad (8)$$

where  $[q_\sigma \cdot m]$  is the largest integer less than or equal to  $q_\sigma \cdot m$ . The estimate DODU is asymptotically normally distributed with mean  $\sigma$  and variance  $[q_\sigma(1 - q_\sigma)/(n/2)]/h(\sigma)$  where  $h(z)$  is the density function of  $H(z)$ . Furthermore, there is

$$\lim_{n \rightarrow \infty} P\{|\text{DODU} - \sigma| > \epsilon\} = 0 \quad \text{for all } \epsilon > 0$$

That means DODU is a consistent estimator for  $\sigma$ .

TABLE I. FUNCTION T DEFINING THE NUMBERS OF GROUPS TO WHICH EACH DIFFERENCE  $Y_{ij} = |X_i - X_j|$ ,  $i < j$ , IS ATTACHED;  $n = 10$  or  $9^a$

$\begin{matrix} \rightarrow j \\ \downarrow i \end{matrix}$	1	2	3	4	5	6	7	8	9	10
1		2	3	4	5	6	7	8	9	1
2			4	5	6	7	8	9	1	3
3				6	7	8	9	1	2	5
4					8	9	1	2	3	7
5						1	2	3	4	9
6							3	4	5	2
7								5	6	4
8									7	6
9										8

<sup>a</sup> For  $n = 9$  omit the last column.

In Eq. (1) there is a somewhat arbitrary choice for the  $Z_k$ 's. Also, the random sample is split in half. Another aspect is that, for graphical estimation, it is more suitable to apply all absolute differences. In sum these considerations led to the idea of using all differences for the estimation of  $\sigma$ . Defining

$$Y_{ij} = |X_i - X_j| \text{ for all } i < j \tag{9}$$

$n(n - 1)/2$  absolute differences are obtained which have to be ordered in ascending order  $Y_{(1)} \leq Y_{(2)} \leq \dots \leq Y_{(n(n - 1)/2)}$ . The variables  $Y_{ij}$  are generally not pairwise

independent. So, not all the theoretical considerations can be applied for the  $Z_k$  variables. Analogous to the estimate DODU we define the estimate

$$\text{DODA} = Y_{([q \cdot n(n-1)/2] + 1)} \quad (10)$$

that is based on all absolute differences.

A further possibility to apply the idea of estimating the standard deviation with a quantile is to divide all  $n(n-1)/2$  absolute differences into  $n-1$  (in the case of even  $n$ ) groups of  $n/2$  stochastically independent differences, where in each of the  $n-1$  groups every original value is considered exactly once. (If  $n$  is an odd figure,  $n$  groups of  $(n-1)/2$  stochastically independent differences are formed, where in each of the groups exactly one of the original values is omitted; thus, in total each of the  $n$  values appears  $(n-1)$  times.) [6, 7].

The procedure which gives the groups may be defined as follows: For every pair of measurements ( $X_i, X_j$  with  $i < j$ ), the value of function  $T$  gives the group to which each of the absolute differences has to be attached; if  $n$  is an odd figure, one has to add a dummy variable. For that even-numbered  $n$  one gets for  $i < j < n$ :

$$T(i, j) = \begin{cases} i + j - 1, & \text{for } i + j \text{ less than or equal to } n \\ i + j - n, & \text{for } i + j \text{ greater than } n. \end{cases} \quad (11a)$$

And should no dummy variable need be applied (originally even-numbered  $n$ ) for  $i < j = n$ :

$$T(i, n) = \begin{cases} 2i - 1, & \text{for } 2i \text{ less than or equal to } n \\ 2i - n, & \text{for } 2i \text{ greater than } n \end{cases} \quad (11b)$$

Table I illustrates this procedure for  $n = 10$  as well as for  $n = 9$ .

Now the  $n-1$  (or  $n$ , if  $n$  was an odd figure) estimates  $\text{DODU}_1, \text{DODU}_2, \dots, \text{DODU}_{(n-1)}$  (or  $\text{DODU}_n$ ) can be calculated and combined by

$$\text{DODM} = 1/(n-1) \sum_{i=1}^{n-1} \text{DODU}_i, \quad \text{even } n \text{ case, or} \quad (12a)$$

$$\text{DODM} = 1/n \sum_{i=1}^n \text{DODU}_i, \quad \text{odd } n \text{ case} \quad (12b)$$

to a single estimate of the standard deviation  $\sigma$ . It is obvious that DODM is a consistent estimate of  $\sigma$  and has all the other statistical features of DODU. Up

TABLE II. MONTE CARLO SIMULATION FOR DODU, DODA AND DODM BASED ON 10 000 RUNS AND STANDARD NORMAL SAMPLES (i.e.  $\sigma = 1$ )

Sample Size  n	esti- mate  $\sigma$	DODU stand. devia- tion	theo- retical value <sup>a</sup>	DODA		DODM	
				esti- mate  $\sigma$	stand. devia- tion	esti- mate  $\sigma$	stand. devia- tion
6	1.04	0.58	0.66	1.05	0.39	1.03	0.38
10	1.01	0.47	0.51	1.05	0.28	1.01	0.27
20	1.09	0.36	0.35	1.01	0.19	1.09	0.20
40	1.02	0.25	0.25	1.01	0.13	1.02	0.13
80	1.03	0.16	0.16	1.00	0.08	not determined	

<sup>a</sup> For great sample sizes, the theoretical value of the standard deviation of the DODU estimate amounts to

$$\{ [q_{\sigma}(1 - q_{\sigma})/(n/2)]/h(\sigma) \}^{(1/2)}.$$

to now nothing can be said about its variance. But the Monte Carlo experiment in Table II shows very good results for DODM, and it should be preferred for computerized numerical calculations. If a graphical evaluation is done, as discussed in Section 3, the estimate DODA has to be used.

3. PRACTICAL APPLICATION

Practical application of the DoD method is illustrated with the following example:

In an analytical interlaboratory experiment [8]  $n = 9$  laboratories determined the  $^{238}\text{Pu}$  isotopic content (wt%) of the same sample material by mass spectrometry with the following results<sup>1</sup>:

$x_1 = 0.2043$	$x_4 = 0.1706$	$x_7 = 0.2108$
$x_2 = 0.2070$	$x_5 = 0.2152$	$x_8 = 0.2019$
$x_3 = 0.2061$	$x_6 = 0.2062$	$x_9 = 0.2175$

The value  $x_4 = 0.1706$  is obviously an outlier.

<sup>1</sup> Data taken from Ref. [9], Evaluation Sheet 55, p. 150.

Conventional estimation of the standard deviation of this data group without rejection of  $x_4$  would result in

$$s_9 = [(1/(9-1)) \sum_{i=1}^9 (x_i - \bar{x})^2]^{1/2} = 0.0137 \quad (13)$$

and after rejection of  $x_4$  in  $s_8 = 0.0054$ .

Applying the DoD method, the  $n(n-1)/2 = 36$  absolute difference of type  $Z_{(k)} = Y_{ij} = X_i - X_j$  for all  $i < j$  are calculated and arranged according to:

$$\begin{array}{lll} Z_{(1)} = Y_{36} = 0.0001; & Z_{(2)} = Y_{26} = 0.0008; & Z_{(3)} = Y_{23} = 0.0009; \\ Z_{(4)} = Y_{13} = 0.0018; & Z_{(5)} = Y_{16} = 0.0019; & Z_{(6)} = Y_{59} = 0.0023; \\ Z_{(7)} = Y_{18} = 0.0024; & Z_{(8)} = Y_{12} = 0.0027; & Z_{(9)} = Y_{27} = 0.0038; \\ Z_{(10)} = Y_{38} = 0.0042; & Z_{(11)} = Y_{68} = 0.0043; & Z_{(12)} = Y_{57} = 0.0044; \\ Z_{(13)} = Y_{67} = 0.0046; & Z_{(14)} = Y_{37} = 0.0047; & Z_{(15)} = Y_{28} = 0.0051; \\ Z_{(16)} = Y_{17} = 0.0065; & Z_{(17)} = Y_{79} = 0.0067; & Z_{(18)} = Y_{25} = 0.0082; \\ Z_{(19)} = Y_{78} = 0.0089; & Z_{(20)} = Y_{56} = 0.0090; & Z_{(21)} = Y_{35} = 0.0091; \\ Z_{(22)} = Y_{29} = 0.0105; & Z_{(23)} = Y_{15} = 0.0109; & Z_{(24)} = Y_{69} = 0.0113; \\ Z_{(25)} = Y_{39} = 0.0114; & Z_{(26)} = Y_{19} = 0.0132; & Z_{(27)} = Y_{58} = 0.0133; \\ Z_{(28)} = Y_{89} = 0.0156; & Z_{(29)} = Y_{48} = 0.0313; & Z_{(30)} = Y_{14} = 0.0337; \\ Z_{(31)} = Y_{34} = 0.0355; & Z_{(32)} = Y_{46} = 0.0356; & Z_{(33)} = Y_{24} = 0.0364; \\ Z_{(34)} = Y_{47} = 0.0402; & Z_{(35)} = Y_{45} = 0.0446; & Z_{(36)} = Y_{49} = 0.0469. \end{array}$$

The estimate DODA for the standard deviation of the nine measurement values is given by Eq. (10). Because there is always  $q_\sigma = 0.52$  and in this example  $n(n-1)/2 = 36$  (total number of differences), there is

$$\text{DODA} = Z_{([0.52 \cdot 36] + 1)} = Z_{([18.72] + 1)} = Z_{(19)} = 0.0089$$

Compared to the conventional estimate for the same population  $s_9 = 0.0137$ , this value lies closer to the 'true' estimate  $s_8 = 0.0054$  derived by conventional calculation after outlier rejection. In particular, please note that – different from the calculation by Eq. (13) – this DODA estimate is completely independent of the actual value of the outlier  $x_4$ : As can be verified by the compilation of differences above, only the last eight values,  $Z_{(29)}$  to  $Z_{(36)}$  are influenced by the outlier  $x_4$ , whereas the estimate of the standard deviation is given by  $Z_{(19)}$ <sup>2</sup>.

<sup>2</sup> If  $x_4$  is rejected, DoD evaluation results in the differences,  $Z_{(1)}$  to  $Z_{(28)}$ , and the estimate for the standard deviation is then given by  $Z_{(15)} = 0.0051$  in good agreement with the estimate  $s_8 = 0.0054$  obtained by conventional calculation.

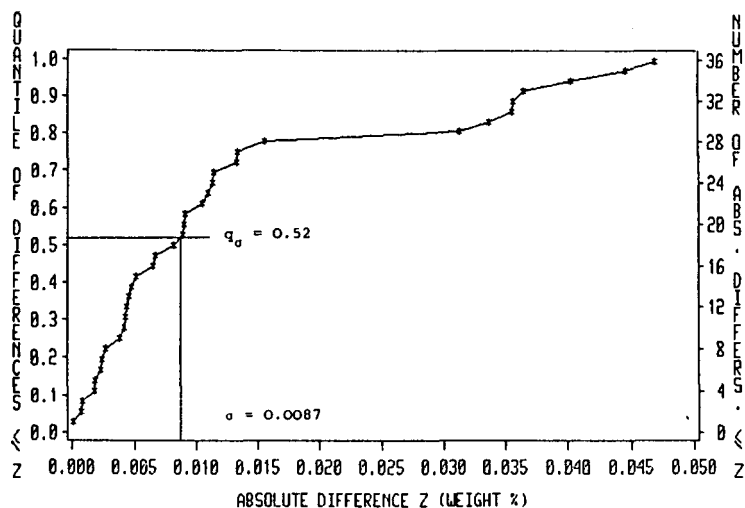


FIG. 2. DoD display of mass spectrometric determinations of  $^{238}\text{Pu}$  abundances;  $n = 9$ ,  $n(n-1)/2 = 36$ .

In Fig. 2, the cumulative distribution of the 36 absolute differences is plotted in increasing order ('DoD display'). It is helpful for judging a difference observed between the results of two laboratories, e.g. those of plant operator and control authority, or of a shipper and receiver. The corresponding ordinate value gives the probability of occurrence of a difference equal to or smaller than the observed one in the analytical assay in question. After threshold values are fixed for considering observed differences as 'acceptable', 'suspicious' or 'unacceptable', such DoD displays may be used as a tool to verify safeguards data [10].

The estimate of the standard deviation of the nine measurement results is given by the abscissa value corresponding to the 52% ordinate value as indicated<sup>3</sup>. According to the curve drawn empirically, a value of 0.0087 is found with this graphical method compared to 0.0089 calculated above. This discrepancy diminishes with increasing number  $n$  of measurement data.

The horizontal part in the shape of the curve indicates the existence of the outlier value  $x_4$ . The eight data points of the right section of the curve originate from differences calculated with this outlier value. Plateaus of this type produced by more than one outlier value may indicate a typical source of error.

<sup>3</sup> In earlier publications the differences were plotted in decreasing order. In such a case, the estimate of the standard deviation is given by the abscissa value corresponding to the 48% ordinate value, the complement to the 52% quantile.



To illustrate the calculation of the appropriate DODM estimate, now

- the  $n = 9$  subgroups (odd case) of independent differences will be created according to Table I and arranged in increasing order of their amounts and
- the  $n = 9$  DODU estimates are picked up according to Eq. (8). As  $n$  is an odd number,  $m$  is given by  $m = (n - 1)/2 = 4$  and therefore  $\text{DODU} = Z_{([q_{\sigma} \cdot m] + 1)} Z_{([0.52 \cdot 4] + 1)} = Z_{([2.08] + 1)} = Z_{(2 + 1)} = Z_{(3)}$  as follows:

Subgroup 1: <sup>4</sup>

$$Z_{(11)} = Y_{38} = 0.0042$$

$$Z_{(12)} = Y_{56} = 0.0090$$

$$Z_{(13)} = Y_{29} = 0.0105$$

$$Z_{(14)} = Y_{47} = 0.0402$$

$$\text{DODU}_1 = Z_{(13)} = 0.0105$$

Subgroup 2:

$$Z_{(21)} = Y_{12} = 0.0027$$

$$Z_{(22)} = Y_{57} = 0.0044$$

$$Z_{(23)} = Y_{39} = 0.0114$$

$$Z_{(24)} = Y_{48} = 0.0313$$

$$\text{DODU}_2 = Z_{(23)} = 0.0114$$

Subgroup 3:

$$Z_{(31)} = Y_{13} = 0.0018$$

$$Z_{(32)} = Y_{67} = 0.0046$$

$$Z_{(33)} = Y_{58} = 0.0133$$

$$Z_{(34)} = Y_{49} = 0.0469$$

$$\text{DODU}_3 = Z_{(33)} = 0.0133$$

Subgroup 4:

$$Z_{(41)} = Y_{23} = 0.0009$$

$$Z_{(42)} = Y_{59} = 0.0023$$

$$Z_{(43)} = Y_{68} = 0.0043$$

$$Z_{(44)} = Y_{14} = 0.0337$$

$$\text{DODU}_4 = Z_{(43)} = 0.0043$$

Subgroup 5:

$$Z_{(51)} = Y_{78} = 0.0089$$

$$Z_{(52)} = Y_{15} = 0.0109$$

$$Z_{(53)} = Y_{69} = 0.0113$$

$$Z_{(54)} = Y_{24} = 0.0364$$

$$\text{DODU}_5 = Z_{(53)} = 0.0113$$

<sup>4</sup> In  $Z_{(ik)}$  the index  $i$  refers to the subgroup according to Table I, and  $k$  gives the ordered numbering of the differences within the subgroup.

Subgroup 6:

$$Z_{(61)} = Y_{16} = 0.0019$$

$$Z_{(62)} = Y_{79} = 0.0067$$

$$Z_{(63)} = Y_{25} = 0.0082$$

$$Z_{(64)} = Y_{34} = 0.0355$$

$$\text{DODU}_6 = Z_{(63)} = 0.0082$$

Subgroup 7:

$$Z_{(71)} = Y_{26} = 0.0008$$

$$Z_{(72)} = Y_{17} = 0.0065$$

$$Z_{(73)} = Y_{35} = 0.0091$$

$$Z_{(74)} = Y_{89} = 0.0156$$

$$\text{DODU}_7 = Z_{(73)} = 0.0091$$

Subgroup 8:

$$Z_{(81)} = Y_{36} = 0.0001$$

$$Z_{(82)} = Y_{18} = 0.0024$$

$$Z_{(83)} = Y_{27} = 0.0038$$

$$Z_{(84)} = Y_{45} = 0.0446$$

$$\text{DODU}_8 = Z_{(83)} = 0.0038$$

Subgroup 9:

$$Z_{(91)} = Y_{37} = 0.0047$$

$$Z_{(92)} = Y_{28} = 0.0051$$

$$Z_{(93)} = Y_{19} = 0.0132$$

$$Z_{(94)} = Y_{46} = 0.0356$$

$$\text{DODU}_9 = Z_{(93)} = 0.0132$$

According to Eq. (12b) the  $n = 9$  DODU estimates are now combined to

$$\text{DODM} = 1/9 \sum_{i=1}^9 \text{DODU}_i = 0.0095$$

With respect to the variances given in Table II, this shows that the DODM estimate is in good agreement with both the numerical and the graphical DODA estimates derived above<sup>5</sup>. For practical purposes the DODA estimate is more convenient since it requires fewer calculations, and no distinction is necessary with respect to even or odd numbered  $n$ .

If the  $x_i$  are repetitive measurement results obtained within one laboratory, the estimate of the standard deviation derived by the DoD method describes the within laboratory repeatability. If such data exist from different laboratories,

<sup>5</sup> The small discrepancy between DODA and DODM diminishes with increasing number  $n$  of measurement data.

the absolute differences may be calculated for each laboratory separately and then pooled to one DoD display. From this, an average value for the within laboratory repeatability of the analytical method in question can be estimated.

#### 4. EVALUATION OF IDA-80 DATA BY THE DoD METHOD

An extended comparison of DoD evaluation results with those obtained by conventional data treatment was made for the IDA-80 measurement evaluation programme [11]<sup>6</sup>. This comparison was of particular interest, since the handling of outliers had played a substantial role in the evaluation performed by conventional methods.

The data material of the IDA-80 programme consists of concentration values for uranium and plutonium determined in samples of different solutions and of isotopic compositions determined for these elements. Each assay was made by a group of 24 to 30 laboratories.

In Table III, three estimates are given for the (relative) standard deviation of the measurement results of each assay:

- the DoD estimate 'DODA' (column 5),
- the conventional estimate without outlier rejection (column 6)
- the conventional estimate after outlier rejection as performed in the official evaluation of the IDA-80 programme (column 7)<sup>7</sup>.

The number of outliers excluded in the latter case is entered in column 8, the 99% confidence limits of that estimate in column 9.

As demonstrated by the figures presented in column 10 of Table III, the conventional estimates calculated without application of outlier criteria are higher than the DoD values. In many cases the differences amount to several hundred per cent. This confirms the expectation that the values estimated by conventional calculation without outlier rejection are meaningless, whereas the DoD values are little affected by extreme values so long as their fraction is below about 20%.

The deviations of the conventional results obtained after outlier rejection from the DoD estimates are equally as often negative as positive (see column 11). This means that the outlier criteria have been applied 'reasonable' in conventional evaluations; thus, the results scatter around the DoD values and do not deviate systematically.

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<sup>6</sup> This analytical measurement programme relates to the most recent mass spectrometric isotope dilution analysis of uranium and plutonium in input solutions of a reprocessing plant for spent nuclear fuels.

<sup>7</sup> The estimate DODM discussed above is given in Ref. [11].

TABLE III. ESTIMATIONS OF RELATIVE STANDARD DEVIATIONS (RSDs) OF THE IDA-80 PROGRAM

1	2	3	4	5	6	7	8	9	10	11	12
	Type of determination	Part of IDA-80 program <sup>a</sup>	No. of labs	'DOD' s <sub>DODA</sub> (%)	Estimate of RSD 'All data' s <sub>ALL</sub> (%)	'IDA-80 eval.' s <sub>IDA</sub> (%)	No. of labs excluded	99% confidence limits of s <sub>IDA</sub> (%)	Rel. dev. of s <sub>ALL</sub> from s <sub>DODA</sub> (%)	Rel. dev. of s <sub>IDA</sub> from s <sub>DODA</sub> (%)	s <sub>DODA</sub> Within s <sub>IDA</sub> confid. limits?
CONCENTRATION	U-Element	1.11	30	0.59	1.72	0.72	2	0.53 ; 1.09	+ 192	+ 22	YES
	"	1.12	27	0.78	2.70	1.03	2	0.75 ; 1.60	+ 246	+ 32	YES
	"	1.2	30	0.73	3.02	0.53	5	0.38 ; 0.83	+ 314	- 27	YES
	"	2.1	28	0.82	2.64	0.48	6	0.34 ; 0.77	+ 222	- 41	NO
ISOTOPIC ABUNDANCE	U-234	1.11	27	0.47	0.82	0.34	4	0.25 ; 0.53	+ 74	- 28	YES
	U-235	2.1	28	0.58	0.93	0.40	4	0.29 ; 0.64	+ 60	- 31	YES
	U-236	1.11	27	4.55	6.28	4.01	1	2.93 ; 6.16	+ 38	- 12	YES
	U-236	2.1	25	5.62	9.74	6.08	1	4.40 ; 9.56	+ 73	+ 8	YES
CONCENTRATION	PU-ELEMENT	1.11	28	0.97	1.90	0.82	2	0.60 ; 1.26	+ 96	- 15	YES
	"	1.12	26	1.95	3.04	2.56	2	1.85 ; 4.03	+ 56	+ 31	YES
	"	1.2	29	0.65	3.77	0.35	6	0.25 ; 0.56	+ 480	- 46	NO
	"	2.1	26	1.34	3.20	1.25	3	0.90 ; 1.99	+ 139	- 7	YES
ISOTOPIC ABUNDANCE	PU-238	1.11	26	0.43	0.66	0.35	3	0.25 ; 0.55	+ 53	- 19	YES
	PU-238	2.1	27	0.49	0.52	0.50	3	0.36 ; 0.80	+ 6	+ 2	YES
	PU-239	1.11	26	5.24	13.7	7.01	2	5.07 ; 11.02	+ 161	+ 34	YES
	PU-239	2.1	24	5.17	16.2	6.21	4	4.36 ; 10.30	+ 213	+ 20	YES
ISOTOPIC ABUNDANCE	PU-240	1.11	29	0.11	0.16	0.13	3	0.095 ; 0.20	+ 45	+ 18	YES
	PU-240	2.1	27	0.08	0.09	0.09	0	0.066 ; 0.14	+ 13	+ 13	YES
	PU-241	1.11	29	0.15	0.32	0.14	2	0.10 ; 0.21	+ 113	- 7	YES
	PU-241	2.1	29	0.11	0.14	0.14	0	0.10 ; 0.21	+ 27	+ 27	YES
ISOTOPIC ABUNDANCE	PU-242	1.11	27	0.46	0.54	0.56	4	0.41 ; 0.87	+ 17	+ 22	YES
	PU-242	2.1	27	0.57	0.62	0.62	0	0.46 ; 0.94	+ 9	+ 9	YES
	PU-242	1.11	29	1.07	1.48	1.17	1	0.86 ; 1.77	+ 38	+ 9	YES
	PU-242	2.1	27	1.61	3.18	1.26	2	0.91 ; 1.96	+ 98	- 22	YES

<sup>a</sup> See Ref. [8].

Finally, in column 12 of Table III it is stated whether or not the DoD estimate lies within the 99% confidence interval of the conventionally calculated value after outlier exclusion. In about 85% of the cases, the answer is 'yes'. For the cases where this does not apply, more detailed studies have revealed that this is very probably due to exclusion of the outliers. If the DoD value occurs below the confidence interval, the last extreme value considered narrowly missed the condition for exclusion, or vice versa<sup>8</sup>.

## 5. CONCLUSIONS

The theoretical considerations and the applications of actual data suggest that the DoD method is a reasonable way to estimate the standard deviation, especially where the data include outliers. Being free from the arbitrariness associated with the use of outlier criteria, the DoD method always leads to the same estimates, regardless of the statistician who performed the evaluation. This is of particular interest for safeguards applications. Graphical DoD display is helpful for judging measurement discrepancies and can be used as a basis for data verification purposes.

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<sup>8</sup> For more details, please see Ref. [11].

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## REPROCESSING INPUT TANK ACCOUNTANCY

### *Analysis of measurement errors by simulation methods*

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#### Abstract

#### REPROCESSING INPUT TANK ACCOUNTANCY: ANALYSIS OF MEASUREMENT ERRORS BY SIMULATION METHODS.

Determination of the quantities of uranium and plutonium contained in irradiated fuels, as performed at the input accountancy tank of the reprocessing plant, is based on a complex measurement process involving several distinct steps, different instrument types, and extensive physico-chemical manipulations of the material to be measured. To assess the quality of the results obtained, a quantitative analysis of the influence of every identified error source is needed. To reach this target, the authors have started the simulation of all measurement operations carried out for reprocessing input accountancy purposes by the volume concentration method. Such simulation is performed by methods of numerical analysis based on random sampling of the actual errors, combination and propagation of such errors, and iteration of the calculation for a number of times sufficient to generate a converged probability function. This is then analysed to produce an estimate of the accuracy and precision of the actual measurement. The program can be adapted to different plant and operating conditions. The simulation program allows the different error sources to be separately tested, and can therefore be used to explain the eventual lack of reproducibility in the results of some procedures. The authors discuss the merit of the simulation approach, describe the simulation model as applied to the measurement of the volume of nitric solution containing the dissolved fuel materials, and present a set of results.

## 1. INTRODUCTION

In current industrial practice the quantities of uranium and plutonium contained in irradiated nuclear fuels can be accurately determined only when the assemblies are submitted to chemical reprocessing. Then, such determination is performed by the plant operator to establish the materials accountancy in his plant. Part of the

operator's measurements are replicated by inspectors of the safeguards authorities, as required by domestic regulations and/or international agreements or treaties, to verify the use of nuclear materials.

There are basically two methods by which the operator carries out his determination — 'volume concentration', and the 'gravimetric' method.

In the volume concentration method the amounts of U and Pu are calculated from the determination of: (i) the volume  $V$  of the nitric solution containing the dissolved materials, (ii) the density  $\rho$  of the solution, and (iii) the concentrations  $C_i$  of the relevant chemical elements in the solution. Masses  $M_i$  of uranium and plutonium in the solution are then calculated as the product of the three preceding quantities:

$$M_i = V \cdot \rho \cdot C_i$$

By the gravimetric method the operator determines (i) the plutonium to uranium mass ratio (Pu/U) in the solution and (ii) the average burnup of the dissolved fuel. The Pu and U masses are then calculated from simple considerations of mass balance in the fuel cycle.

In both cases, the measurement process is complex, since it involves many instruments (from scales to a mass spectrometer), remote handling of most operations, sampling of the solution and extensive conditioning of the samples to be analysed, use of reference materials of various kinds and origins, etc.

In assessing the quality of the results obtained, consideration must be given to numerous factors which may affect the determination, such as:

- instrument performance
- accuracy of calibration procedures
- stability of calibrations
- uncertainties in the reference materials and in their use
- performance of the procedures
- capability of controlling the physical parameters which influence the result
- effect of neglecting the influence of ambient conditions
- extensive manipulation and conditioning of the materials to be assayed
- effectiveness of the statistical and mathematical models used for interpreting the raw data
- effectiveness of the procedures for comparing the results of measurements performed in different laboratories, often by different methods, procedures and/or instrumentation.

For obvious reasons, both the operator and the inspector need to know the performance of the whole measurement process in terms of precision and accuracy. They also need to know by which error sources the performance is affected, and how, in order to be able to explain possible anomalies in the results and to take appropriate corrective actions.



The experimental determination of the accuracy of the whole measurement process is not easy, owing to the complexity of the procedures, the high cost of repetitions, the possible lack of full control of some ambient and physical parameters, the impossibility of replicating some of the measurements in other laboratories, and the necessity of not interfering with routine industrial production.

In such conditions, a physico-mathematical model describing the whole measurement process and allowing both analysis and synthesis of its performance, may prove of great assistance in understanding the statistical behaviour of the measurement results and in tracing the causes of possible anomalies.

Unfortunately, the use of classic statistical formulas which combine errors (and therefore allow error propagation to be studied through a system), leads to a very cumbersome analytical problem, owing to the large number of possible error sources, the covariance of many of them and the complex error propagation pattern.

Owing to these difficulties it seems appropriate to try, as an alternative to the analytical approach, a numerical approach based on the Monte Carlo method, in which the actual value of the error (or uncertainty) generated by each operation, or imposed by any external source, is drawn at random from an appropriate probability distribution. Combination and propagation of the actual error values do not result in the mathematical difficulties inherent in the analytical model.

By repeated simulation of the complete procedure for a sufficient number of times, a statistical distribution of the result is obtained similar to that which could be obtained from the actual repetition of the measurement. By this means, the performance of the measurement process can be determined, and even hypothetical variants to the existing system can be studied.

Of course, the accuracy of the model should be proved by extensive application to real cases, and comparisons with experimentally obtained conclusions.

An application of the Monte Carlo approach to the reprocessing input measurements has been worked out in a study promoted by the Ispra Establishment of the CEC Joint Research Centre and carried out in conjunction with the Dipartimento di Energetica, Università degli Studi di Roma 'La Sapienza'. The model developed so far concerns the volume concentration method only. However, the model (and the relevant program) are open ended, and the addition of new procedures, techniques and instruments is not restricted.

This report describes the part of the simulation program already completed and gives examples of calculations.

## 2. GENERAL DESCRIPTION OF THE PROGRAM

A computer program is being developed for simulating all measurement operations carried out by the volume concentration method for reprocessing input accountancy. At present, only that part dealing with tank volume calibration and solution volume measurement has been completed and is presented here. The final program

will, however, also include simulation of the following measurement system and procedures:

- densitometer calibration
- liquor density measurement
- sampling
- sample conditioning
- isotopic dilution analysis (IDA)
- calibrations and materials relevant to the IDA technique
- evaluation of plutonium and uranium masses in the input solution.

The description of the measurement process incorporated in the simulator is very detailed, including instruments, procedures, physical parameters and environmental conditions liable to affect the input accountancy (and its verification) through the uncertainties inherent in their use or control. As an example, the possible error sources considered in the simulation of the tank volume calibration are:

- scale
- air buoyancy
- calibration fluid impurities
- temperature of the calibration fluid (after introduction)
- positioning of the dip tubes
- gas flow in the dip tubes
- bubbling
- thermal expansion of the tank and the dip tubes
- films on the walls of the tank and of the lines
- air pressure and density in the tank
- temperature of the manometric fluids
- reading of the manometers
- electromanometer.

The simulator is apt to solve easily several problems related to the activities of the plant operator and the inspector, but can also be valuable in many research and development areas of measurement techniques. Typical uses of the program are:

- (1) Determination of the performance of the measurement system, or of specific parts of it;
- (2) Analysis of the importance of the various error sources;
- (3) Analysis of the effect of changing selected physical parameters (or the ambient conditions) on the precision and accuracy of the measurement system;
- (4) Comparison of the precisions and accuracies achievable under different procedures or with different instrumentation.

By the use of these possibilities it is possible to:

- Determine whether the measurement results are in line with the expected performance of the system;

- Investigate possible causes of detected anomalies; and
- Suggest possible actions for improving the measuring system and test their potential.

The simulation is performed with methods of numerical analysis based on the random sampling of all errors and uncertainties affecting the measurement; the simulation is repeated many times to obtain a statistical distribution of the results (Monte Carlo method). The distribution is then analysed to estimate the precision and accuracy of the simulated measurement system.

The simulation program is written in Fortran-77. Two versions are now being developed, one for IBM compatible PCs and one for the IBM mainframe computer. The latter is totally conversational and helps the user to make his way easily and consistently through the numerous available options.

### 3. DESCRIPTION OF THE PROGRAM OPERATION

The rationale of the simulation program is described in Fig. 1. The statistical distributions of errors (or uncertainties) affecting the various quantities are contained in a database attached to the program. The program starts the calculation by determining the 'true' value of all the physical quantities involved in the problem, as they should be in the absence of any errors. The determination is then repeated, but considering that an error affects each step of the process, the errors are automatically combined and propagated as the calculation progresses. The values of the errors to be used in the calculation are randomly drawn from the appropriate density function stored in the database.

At the end of this process, a simulated 'measured' value (i.e. affected by the errors which are likely to occur in true operation) is produced.

The whole calculation is repeated frequently (1000–10 000 iterations) so as to produce a distribution which simulates the statistical density function of the actual measurements and allows their precision and accuracy to be estimated.

Application of the above concepts to solution volume measurements is illustrated in Fig. 2.

Volume calibration of the tank and solution volume measurement are dealt with in two separate routines. In the first the process of calibration is carried out by subsequent additions of known fluid weights. The geometric shape of the tank, the pneumatic system design and the pressure sensor system can be described by the program user. Operating conditions which can be imposed by the user include air and calibration fluid temperatures, air flow in the dip tubes, air density and pressure in the tank, status of the lines (dry/wet), status of the tank (dry/wet) and introduction of the fluid (from top/bottom). The sizes of the fluid additions are assigned by the user. Instruments for level reading can be chosen from U-tubes with water and U-tubes with the

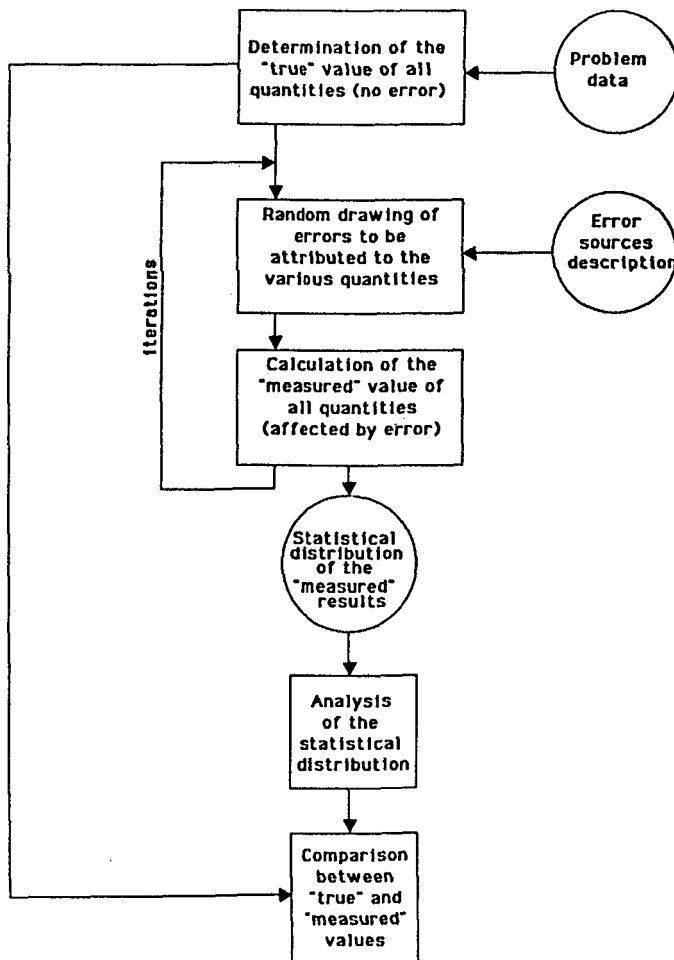


FIG. 1. Rationale of the simulation.

TBE, a Ruska electromanometer. The program automatically assigns errors to all physical and operating parameters included in the calculation.

From the calibration, some volume level points are obtained, which are interpolated to derive a calibration curve. This can be either a straight line or a polyline. By repeating the calculation, an average calibration curve is produced, together with its statistical parameters.

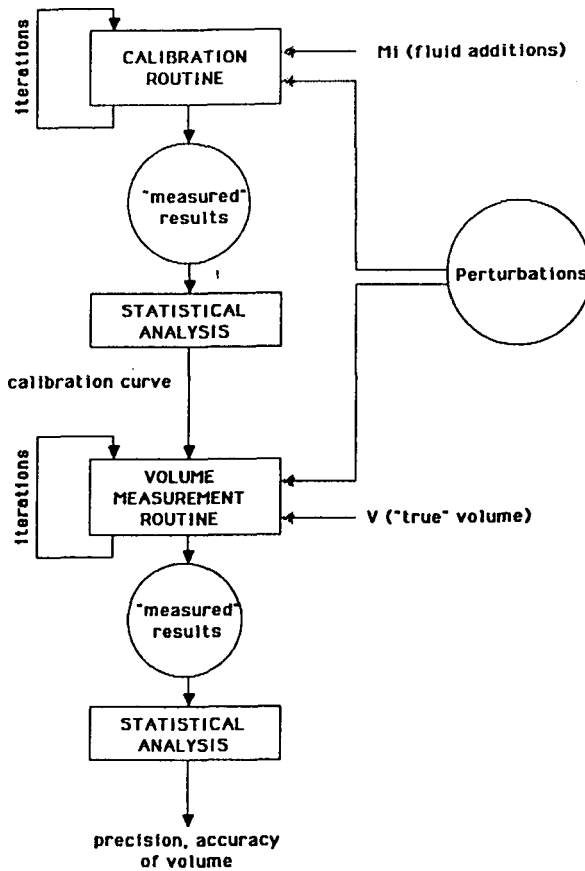


FIG. 2. Operation scheme of the routines simulating the calibration and the solution volume measurement.

In the second routine (volume measurement), the 'measured' solution level is first calculated by simulation — a different level reading instrument than that used in calibration can be used in this step. The 'measured' volume is then derived from the average calibration curve previously calculated. Iteration of this process allows a statistical distribution of the 'measured' volumes to be generated, which can subsequently be analysed to derive estimates of the precision and accuracy of the actual measurements.

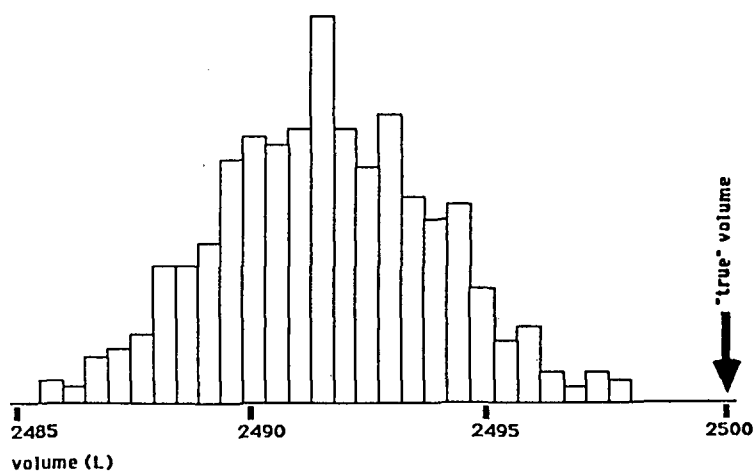


FIG. 3. Probability density function of the 'measured' volume (1000 iterations only).

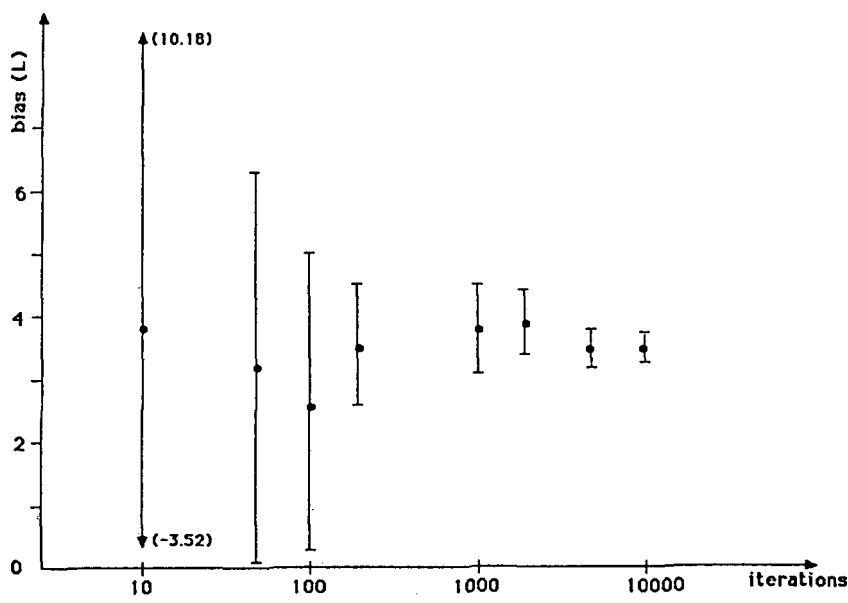


FIG. 4. Testing the convergence of the program (simulation of measurement of 3000 L).

TABLE I. EFFECT ON THE CALIBRATION CURVE OF  
DIFFERENT PROCEDURES

Description of the test calculation	Bias of the volume corresponding to 3181 mm (L)	Standard deviation of the volume (L)
Calibration by additions of 300 kg each	4	0.9
Calibration by additions of 30 kg each	8	0.5
Use of Ruska TBE electromanometer for measuring levels	4	0.9
Use of a U-tube for measuring levels	9	2.4

#### 4. EXAMPLES OF APPLICATION

Up to now the program has been tested only on artificially created problems to show its potentials, but application to real cases is planned. The examples given below indicate that the aims of the program can be achieved.

The test calculations were carried out on a cylindrical tank of 3500 litres' capacity, using 3000 litres of water for calibration and 2500 litres of a heavier fluid for operation.

In Fig. 3 a probability density function of the simulated 'measured volume', as produced by the program, is presented. Analysis of this function allows estimates of the precision and accuracy of the actual measurement to be derived.

In Fig. 4 the results of a convergence test are presented. The simulated volume converges fairly well after a few hundred iterations. The bars, which include the calculated points, represent the 95% confidence level of the result.

The results of other tests performed with the calibration routine are summarized in Table I. These tests have been performed under different operating conditions, described in the table. The results are quoted in terms of bias and standard deviation of the volume corresponding to the 3181 mm level (which should theoretically be 2500 litres).





**SAFEGUARDS PHILOSOPHY  
AND CONCEPTS**

**(Session 15)**

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**German Democratic Republic**



## ОСОБЕННОСТИ ПРИМЕНЕНИЯ ГАРАНТИЙ МАГАТЭ ДЛЯ ЯДЕРНОГО ТОПЛИВНОГО ЦИКЛА, ВКЛЮЧАЮЩЕГО ЛЕГКОВОДНЫЕ РЕАКТОРЫ И ДОЛГОВРЕМЕННЫЕ ХРАНИЛИЩА ОТРАБОТАВШЕГО ТОПЛИВА

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### Abstract—Аннотация

SPECIAL FEATURES OF APPLICATION OF IAEA SAFEGUARDS TO NUCLEAR FUEL CYCLE, INCLUDING LIGHT WATER REACTORS AND LONG TERM SPENT FUEL STORES.

The further improvement and optimization of the safeguards system are of great importance for the effective application of IAEA procedures with regard to nuclear materials whilst making the most economical use of the Agency's resources. One way to achieve both of these goals is to take into account the special features of the nuclear fuel cycle of individual States in planning and carrying out safeguards activities. Allowing for the presence or absence of 'sensitive' nuclear facilities within a State and for the international interdependence of the fuel cycle in such States may occasionally simplify Agency safeguards procedures, as well as increasing the effectiveness of safeguards planning and implementation. In the paper the 'open' nuclear fuel cycle of a non-nuclear-weapon State is examined; the cycle consists of several WWER type reactors and long term spent fuel stores, with one and the same nuclear weapon State supplying the fresh fuel and taking back the spent fuel. These features of the nuclear fuel cycle reduce the probability of diversion of nuclear materials. On the basis of existing recommendations for the individual facility types and taking into account the features of the nuclear fuel cycle in question, possible ways are examined of improving safeguards procedures and reducing manpower expended on the Agency's safeguards activities. The proposed approach could be extended to the group of States having this type of nuclear fuel cycle, which together with the State supplying the fresh fuel and taking back the spent fuel could appropriately be viewed as constituting a regional nuclear fuel cycle.

ОСОБЕННОСТИ ПРИМЕНЕНИЯ ГАРАНТИЙ МАГАТЭ ДЛЯ ЯДЕРНОГО ТОПЛИВНОГО ЦИКЛА, ВКЛЮЧАЮЩЕГО ЛЕГКОВОДНЫЕ РЕАКТОРЫ И ДОЛГОВРЕМЕННЫЕ ХРАНИЛИЩА ОТРАБОТАВШЕГО ТОПЛИВА.

Важное значение для эффективного применения контрольных процедур МАГАТЭ в отношении ядерного материала с обеспечением наиболее экономичного использования ресурсов Агентства имеют вопросы дальнейшего совершенствования и оптимизации системы гарантий. Одним из путей дости-

жения эффективного применения гарантий МАГАТЭ с обеспечением наиболее экономичного использования ресурсов Агентства является учет особенностей ядерного топливного цикла (ЯТЦ) государств при планировании и осуществлении контрольной деятельности. Учет наличия или отсутствия в государстве "чувствительных" ядерных установок и международной взаимозависимости ЯТЦ такого государства позволяет в некоторых случаях упростить процедуры гарантий МАГАТЭ, а также повысить эффективность планирования и осуществления деятельности по гарантиям. В настоящей работе рассматривается "открытый" ЯТЦ неядерного государства, состоящий из нескольких реакторов типа ВВЭР и долговременных хранилищ отработавшего топлива (ДХОТ), причем поставка свежего топлива и прием отработавшего топлива осуществляется одним и тем же ядерным государством. Такие особенности рассматриваемого ЯТЦ снижают вероятность переключения ядерного материала. На основе ранее разработанных рекомендаций для отдельных типов установок и с учетом особенностей рассматриваемого ЯТЦ обсуждаются возможные пути совершенствования процедур гарантий и снижения трудозатрат на контрольную деятельность МАГАТЭ. Предложенный подход может быть распространен на группу государств с рассматриваемым типом ЯТЦ, которую совместно с государством-поставщиком свежего и получателем отработавшего топлива целесообразно рассматривать в качестве регионального ЯТЦ.

Важное значение для эффективного применения контрольных процедур МАГАТЭ в отношении ядерного материала с обеспечением наиболее экономичного использования ресурсов МАГАТЭ имеют вопросы дальнейшего совершенствования и оптимизации системы гарантий. Одним из путей достижения этого является учет особенностей ЯТЦ государств при разработке процедур гарантий, а также при планировании, осуществлении и оценке деятельности по гарантиям. С этой целью МАГАТЭ считает целесообразным, как отмечено в [1], разработку альтернативных подходов к применению гарантий с учетом особенностей ЯТЦ (например, взаимозависимость между собой ядерных установок, наличие или отсутствие в государстве "чувствительных" установок и т.д.), а также с учетом международной взаимозависимости ЯТЦ государств.

Представляет интерес рассмотреть несколько ЯТЦ государств, состоящих в основном из легководных реакторов типа ВВЭР и долговременных хранилищ отработавшего топлива (ДХОТ), причем поставка свежего топлива и прием отработавшего реакторного топлива осуществляется одним государством. Такую совокупность нескольких ЯТЦ можно рассматривать в качестве регионального ядерного топливного цикла (РЯТЦ) (рис. 1).

Примером такого РЯТЦ может являться группа государств—членов ДНЯО, развивающих свою ядерную энергетику на основе реакторов ВВЭР при обеспечении поставок свежего и приема отработавшего топлива СССР — ядерным государством, одним из депозитариев ДНЯО. В настоящее время в такую группу входят шесть государств (табл. I), к 1990 г. их число возрастет до десяти, а к 2000 г. еще более увеличится.

Рассматриваемый РЯТЦ состоит из наиболее простых и распространенных ЯТЦ отдельных государств, которые в данном случае тесно взаимосвязаны с государством, поставляющим им ядерные установки и свежее топливо для них, а также забирающим отработавшее реакторное топливо на переработку. Между системами учета и контроля ядерного материала отдельных государств может осуществляться оперативный обмен

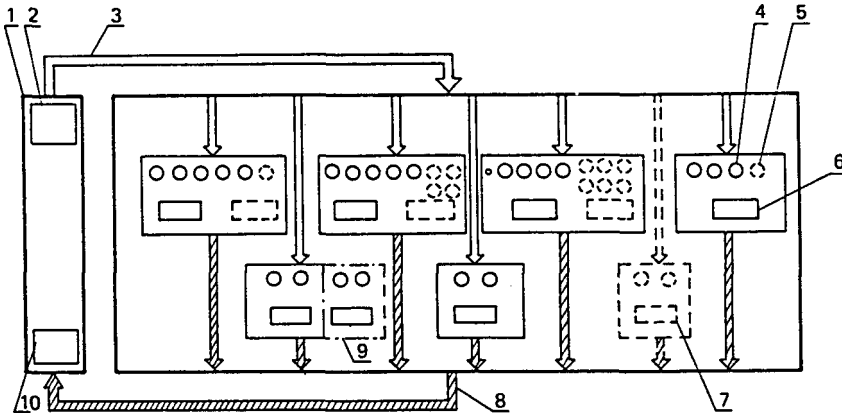


РИС. 1. Схема РЯТЦ с поставкой свежего реакторного топлива из одного (ядерного) государства: 1 — ЯТЦ ядерного государства; 2 — завод по производству реакторного топлива; 3 — поток свежего топлива реакторов ВВЭР; 4 — действующая АЭС с реактором ВВЭР; 5 — строящаяся АЭС с реактором ВВЭР; 6 — действующее ДХОТ; 7 — строящееся или планируемое ДХОТ; 8 — поток отработанного топлива реакторов ВВЭР; 9 — часть ЯТЦ государства имеющего АЭС с другими типами реакторов; 10 — завод по химической переработке отработанного топлива.

Примечание: у других государств РЯТЦ имеется только АЭС с реакторами ВВЭР и долговременные хранилища отработанного топлива (ДХОТ).

информацией о ядерном материале на магнитных носителях или по телекоммуникационным каналам.

Ниже проводится анализ возможности снижения трудозатрат МАГАТЭ на осуществление процедур гарантий в ЯТЦ, являющимся частью рассматриваемого РЯТЦ.

В работе используются критерии гарантий, принятые МАГАТЭ в настоящее время для осуществления практической деятельности, и предполагается, что возможные пути переключения ядерного материала для рассматриваемого ЯТЦ\* учтены в рекомендациях по осуществлению гарантий на отдельных установках.

Рассмотрим ЯТЦ государства, состоящий из  $n$ —реакторов типа ВВЭР и долговременного хранилища отработанного топлива (ДХОТ). Возможные процедуры контроля и трудозатраты МАГАТЭ для проведения инспекционных проверок на установках такого типа были описаны и оценены в работах [5–8]. Общие трудозатраты МАГАТЭ на инспекционную деятельность в рассматриваемом ЯТЦ можно представить в виде:

$$T = \sum_{i=1}^{i=n} T_{r,i} + T_s \quad (1)$$

В свою очередь процедуры контроля на ВВЭР и соответствующие трудозатраты на них МАГАТЭ можно разделить на проверку потока свежего и отработанного топлива

\* Подробный анализ возможных путей переключения для рассматриваемого типа ЯТЦ проведен в ряде работ, например в [4].

ТАБЛИЦА I. ПЕРСПЕКТИВЫ РАЗВИТИЯ ЯДЕРНОЙ ЭНЕРГЕТИКИ ГОСУДАРСТВ, ИСПОЛЬЗУЮЩИХ ДЛЯ ЭТОЙ ЦЕЛИ РЕАКТОРЫ СОВЕТСКОЙ КОНСТРУКЦИИ ТИПА ВВЭР [2, 3]

№№	Государство	Количество реакторов типа ВВЭР в государстве*			
		1984 г.	1985 г.	1986 г.	1990 г.
1.	СССР	12 + 4*	12 + 7*	12 + 18*	14 + 45*
2.	ВНР	2	3	3	4
3.	ГДР	(1) + 4	(1) + 4	(1) + 4	(1) + 8 + 2*
4.	НРБ	4	4	4 + 1*	4 + 2*
5.	ЧССР	4	6	6	9
6.	Финляндия	2 + (2)	2 + (2)	2 + (2)	2 + (2)
7.	Куба				2
8.	ПНР				2
9.	СРР				(3)
10.	Ливийская Джамахирия				—
11.	СРВ				
12.	КНДР				
Итого: реакторов ВВЭР		33	39	51	93
Всего в мире PWR			189		289
Всего в мире LWR = PWR + BWR			268		398
Всего в мире реакторов			355		528

\* Количество реакторов ВВЭР-1000 дано числом со знаком "\*\*", в скобках указано число реакторов другого типа.

и на проверку инвентарного количества ядерного материала на АЭС (условные обозначения даны в табл. II) :

$$T_r = (T_r^F + T_r^S) + T_r^I = m \cdot \sum_{j=1}^{j=4} t_{r,j} + k \cdot \sum_{j=5}^{j=11} t_{r,j} + \sum_{j=12}^{j=18} t_{r,j} , \quad (2)$$

где  $m$  и  $K$  — число проверок в год потоков свежего и отработавшего топлива на АЭС с ВВЭР, соответственно (обычно  $m, K \leq 4$ ).

Аналогичное выражение можно записать для соответствующих трудозатрат МАГАТЭ для ДХОТ, разделив их соответственно на проверку потока топлива (получение и отправление) и проверку инвентарного количества (условные обозначения даны в табл. II).

$$T_s = nk \sum_{j=1}^{j=6} t_{s,j} + \ell \sum_{j=7}^{j=12} t_{s,j} + \sum_{j=13}^{j=16} t_{s,j} , \quad (3)$$

где  $\ell$  — число отправлений в год отработавшего топлива из ДХОТ за границу.

Очевидно, что предложенное выше формализованное представление трудозатрат МАГАТЭ на инспекционную деятельность будет также способствовать стандартизации процедур контроля и учету особенностей рассматриваемого ЯТЦ в выборе критериев для оценки эффективности осуществления гарантий.

Подобное представление трудозатрат МАГАТЭ поможет проведению на ЭВМ, с помощью несложных программ, оптимальной координации инспекционных усилий с учетом графиков эксплуатации установок рассматриваемого ЯТЦ.

Учитывая полную взаимозависимость между государством с рассматриваемым ЯТЦ и ядерным государством-поставщиком свежего и получателем отработавшего топлива, а также принимая во внимание возможность проведения проверок МАГАТЭ по выбору на одной из смежных установок ЯТЦ, можно предложить следующие рекомендации по сокращению некоторых процедур проверки МАГАТЭ.

(а) Прием свежего топлива из-за границы на АЭС производится только на основании данных отправителя. Предлагается ограничить контроль поступления свежего топлива только проверкой документации и осуществить его подсчет и идентификацию один раз в год во время проверки проведения физической инвентаризации (т.е. во время перегрузки реактора). В этом случае экономия трудозатрат составит:

$$\frac{nm \sum_{j=2}^{j=4} t_{r,j}}{T} \times 100\% , \quad (4)$$

(б) Проверка отработавшего топлива на АЭС производится в момент его выгрузки из реактора в бассейн выдержки, во время его хранения на станции в течение нескольких лет (обычно 3 года и более), в момент его отгрузки и во время приема контейнера с отработавшим ТВС в ДХОТ. Предлагается контроль отправления отработавшего топлива на АЭС ограничить проверкой документации и проводить проверку такого топлива во время его приема в ДХОТ. В этом случае экономия трудозатрат составит:

$$\frac{nk \sum_{j=6}^{j=8} t_{r,j}}{T} \times 100\% , \quad (5)$$

ТАБЛИЦА II. ПРОЦЕДУРЫ КОНТРОЛЯ МАГАТЭ И ТРУДОЗАТРАТЫ НА ПРОВЕДЕНИЕ ИНСПЕКЦИОННЫХ ПРОВЕРОК ДЛЯ РЕАКТОРОВ ТИПА ВВЭР И ДХОТ

Вид инспекционной проверки	ВВЭР		ДХОТ		
	Проверка потока свежего топлива	Проверка потока отработавшего топлива	Проверка инвентарного количества	Проверка потока отработавшего топлива	Проверка инвентарного количества
Проверка документации					
Подсчет и идентификация ТВС	$t_r, 1$ $t_r, 2$	$t_r, 5$ $t_r, 6$	$t_r, 12$ $t_r, 13$	$t_s, 1$ $t_s, 2$	$t_s, 13$ $t_s, 14$
Выборочные измерения содержания ядерного материала или его качественная проверка методами НРА	$t_r, 3$	$t_r, 7$	$t_r, 14^*$	$t_s, 3$	$t_s, 15^*$
Проверка пломб и печатей и их применение для проверки топлива, элементов контеймента, технических средств МАГАТЭ	$t_r, 4$	$t_r, 8$	$t_r, 15$	$t_s, 4$	$t_s, 16$
Обслуживание и снятие показаний аппаратуры для оптического наблюдения (теле- и кинокамеры)	-	$t_r, 9$	$t_r, 16$	$t_s, 5$	-
Обслуживание и снятие показаний монитора перемещения ТВС в/из бассейна выдержки	-	$t_r, 10$	$t_r, 17$	$t_s, 6$	-
Проверка показаний и обслуживание монитора мощности реактора	-	$t_r, 11$	$t_r, 18$	-	-
Сумма трудозатрат:	$T_r^F = \sum_{j=1}^{j=4} t_{r,j}$	$T_r^S = \sum_{j=5}^{j=11} t_{r,j}$	$T_r^I = \sum_{j=12}^{j=18} t_{r,j}$	$\sum_{j=1}^{j=6} t_{s,j}$	$\sum_{j=7}^{j=16} t_{s,j}$

\* Качественная проверка на "облученность" отработавшего топлива, например, регистрации излучения Чернобыля-Вавилова в воде от ТВС или использование миниатюрного дозиметра.



(с) При условии регулярных проверок проведения физической инвентаризации в ДХОТ, надежной работы аппаратуры для осуществления мер по сохранению и наблюдению в течение времени хранения отработавших ТВС в хранилище, а также в случае действия системы оперативного оповещения ядерного государства о получении отработавшего топлива и о содержании в нем ядерного материала, можно предложить ограничить процедуры проверки в ДХОТ при отправке ТВС за рубеж только проверкой документации и их подсчетом и идентификацией. В этом случае экономию трудозатрат МАГАТЭ можно оценить как

$$\frac{\sum_{j=9}^{j=12} t_{s,j}}{T} \times 100\%, \quad (6)$$

Для примера проведем количественную оценку экономии трудозатрат МАГАТЭ для ЯТЦ, состоящего из 4 реакторов ( $n = 4$ ) и предположим, что на этих реакторах проводится один раз в год проверка потока свежего ( $m = 1$ ) и четыре раза в год — отработавшего топлива ( $K = 4$ ). Учитывая рекомендации работ [6–8], примем следующие значения трудозатрат для рассматриваемой инспекционной деятельности:

$$\left. \begin{aligned} T_r &= 20 \text{ чел.-сут./инспекц. (или 16 сут./инспекц.)} \\ T_s &= 65 \text{ чел.-сут./инспекц. (или 39 сут./инспекц.)} \end{aligned} \right\}$$

$$\left. \sum_{j=2}^{j=4} t_{r,j} = 1 \text{ чел.-сут./инспекц. (или 1 сут./инспекц.)} \right\}$$

$$\left. \sum_{j=6}^{j=8} t_{r,j} = 3 \text{ чел.-сут./инспекц. (или 3 сут./инспекц.)} \right\}$$

$$\left. \sum_{j=9}^{j=12} t_{s,j} = 49 \text{ чел.-сут./инспекц. (или 23 сут./инспекц.)} \right\}$$

Общие трудозатраты для рассматриваемого примера составят:

$$T = n T_r + T_s = 145 \text{ чел.-сут./инспекц. (или 103 сут./инспекц.)}$$

Подставляя значения величин (7) в выражения (4) – (6), получим следующие оценки экономии трудозатрат МАГАТЭ для рассматриваемого ЯТЦ (табл. III).

ТАБЛИЦА Ш. ОЦЕНКИ ЭКОНОМИИ ТРУДОЗАТРАТ МАГАТЭ

Рассматриваемый вариант	Величина абсолютной и относительной экономии трудозатрат			
	чел.-сут/инспекц.	сут/инспекц.	%	%
a	4	(4)	2,8	(9,9)
b	48	(48)	31,1	(46,6)
c	49	(23)	33,8	(22,3)
a + b	52	(52)	35,9	(50,5)
a + b + c	101	(75)	69,7	(72,8)

На повышение эффективности контрольных процедур МАГАТЭ в целом для рассматриваемого примера РЯТЦ и для отдельных его элементов – ЯТЦ могут оказать влияние следующие факторы, зависящие в основном от операторов взаимодействующих ядерных установок.

Во-первых, тщательное и рациональное планирование получения и отправления ядерного материала на установках рассматриваемого ЯТЦ позволит осуществить оптимальное распределение инспекционных трудозатрат МАГАТЭ для проверок соответствующего потока ядерного материала. Например, получение свежего топлива на АЭС непосредственно перед перегрузкой реактора позволит совместить его контроль с проверкой осуществления физической инвентаризации на станции.

Во-вторых, размещение на одной площадке АЭС с несколькими блоками реакторов типа ВВЭР и ДХОТ позволит совмещать инспекционные проверки на смежных установках, что приведет к экономии времени на поездки инспекторов и на транспортировку оборудования МАГАТЭ.

В-третьих, размещение нескольких установок в одном месте позволит оборудовать помещение для аппаратуры и технических средств МАГАТЭ, в котором также может производиться первичная обработка результатов инспекционной проверки, включая использование для этой цели микро-ЭВМ (персональный компьютер).

В-четвертых, при согласии государств в проектах типовых реакторов ВВЭР и ДХОТ для всех ЯТЦ в рассматриваемом РЯТЦ можно учитывать требования, связанные с применением гарантий МАГАТЭ. Кроме этого, такие установки могли бы в будущем укомплектовываться при их поставке и строительстве приборами и техническими средствами, необходимыми для осуществления процедур гарантий МАГАТЭ (например, мониторами для регистрации тепловой мощности реактора, для регистрации перемещений отработавших ТВС, стендами и приборами для измерений ТВС методами НРА, оптической аппаратурой для наблюдения и т.д.).

В-пятых, размещение нескольких установок в одном месте позволяет создать для них высокоэффективную систему учета и контроля ядерного материала с использованием ЭВМ. Это также позволит повысить эффективность проверки документации на ядерный материал и оперативность представления отчетной документации в МАГАТЭ.

Представляется, что выводы и рекомендации настоящей работы можно рассматривать в качестве одного из конкретных примеров для оптимизации распределения инспекционных ресурсов МАГАТЭ, например, как это было предложено в работе [9].

Снижение трудозатрат МАГАТЭ на инспекционные процедуры в ЯТЦ, имеющих только легководные реакторы и ДХОТ, позволит МАГАТЭ сконцентрировать свои усилия по проверке на тех стадиях топливного цикла государств, где переключение ядерного материала может непосредственно вести к созданию ядерных взрывных устройств в наиболее короткие сроки. Такой подход будет также способствовать более экономному использованию органических ресурсов МАГАТЭ.

В дальнейшем результаты распределения государств на группы с одинаковыми ЯТЦ могли бы быть использованы в общей количественной схеме распределения ресурсов МАГАТЭ с учетом, например, "фактора переключения" [10], учитывающего развитость топливного цикла.

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# NUCLEAR MATERIAL DETECTION AND INSPECTION GOALS FOR A STATE AS A WHOLE

*Discussion of technical problems involved*

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## Abstract

NUCLEAR MATERIAL DETECTION AND INSPECTION GOALS FOR A STATE AS A WHOLE: DISCUSSION OF TECHNICAL PROBLEMS INVOLVED.

On the basis of detection and inspection goals of individual material balance areas (MBAs) the possible statements on a system of MBAs and thus on a State as a whole are investigated. The false alarm and non-detection probabilities of this system are calculated from the corresponding probabilities of the individual systems. The expected accountancy capability for the system of MBAs is calculated, taking into account the optimal diversion strategy. These results are compared with those of a 'global' concept of formulating the detection goals for a system of MBAs which is not based on the results in the individual MBAs. It is shown by way of example that the calculations of the expected accountancy capability based on the two different concepts lead to nearly the same results.

## 1. INTRODUCTION

In accordance with the provisions of INFCIRC/153 type Safeguards Agreements nuclear material accountancy includes the striking of material balances at certain intervals for the safeguarded nuclear material. In addition, the Agreements demand that the IAEA make statements on the conclusions it has drawn from its inspection activities, in particular on the amount of material unaccounted for (MUF) at the end of each verified material balance period, giving the limits of accuracy of the amounts stated.

Of still greater interest, however, are statements which hold for the State as a whole and not only for individual MBAs. The IAEA's annual Safeguards Implementation Report contains an analysis of inspection goal attainment for

individual States, but in contrast with those for individual MBAs these goals are not given in quantitative terms.

Without consideration of the time component, the detection goals for individual MBAs are characterized by the following three parameters:

- The accountancy verification goal quantity  $M$ ,
- The non-detection probability  $\beta$ ,
- The false alarm probability  $\alpha$ .

Specific numerical values of the parameters represent the detection and inspection goals, which can be either identical or different for a given MBA [1].

In contrast,

“the inspection goal for each State as a whole is to have the inspection goals attained at least partially for all facilities with regard to direct-use material outside cores of operating reactors ...” [2].

So far, no statements have been made on the values of  $M$ ,  $\alpha$  and  $\beta$  for a system of MBAs and thus for a State as a whole.

The object of this paper is to contribute to the solution of the problem and to make statements on the goals which can be attained in a system of MBAs, taking into account the goals for individual MBAs. The following considerations are restricted to nuclear material accountancy measures.

## 2. MUF EVALUATION FOR AN MBA

First, the known procedure of evaluating the amount of MUF in an MBA should be recalled:

$$MUF = BI + R - S - EI$$

where BI is the beginning physical inventory of the material balance period, R is the sum of increases to inventory and S the sum of decreases from inventory during the material balance period, and EI is the ending physical inventory of this period. Such equations have to be established separately for the individual nuclear material categories. Assuming that the terms in the equation are independent and that their variances are known, then

$$\sigma_{MUF}^2 = \sigma_{BI}^2 + \sigma_R^2 + \sigma_S^2 + \sigma_{EI}^2$$

The main question to be answered when evaluating MUF is: is a specific MUF value exclusively due to measurement errors associated with the individual components of the MUF equation or are there any additional factors involved?

This question is usually answered by a statistical test procedure. Here, the hypothesis

$H_0$ : No nuclear material was diverted,  
i.e.  $E(MUF) = 0$ ,

has to be tested against the alternative hypothesis

$H_1$ : A quantity not less than  $M$  was diverted,  
i.e.  $E(MUF) \geq M$ .

$E(MUF)$  denotes the expectation value for MUF.

Introducing an at first unknown significant quantity  $M_s$ , it is decided that

$H_0$  is correct if  $MUF < M_s$ ,  
 $H_1$  is correct if  $MUF \geq M_s$ .

It may turn out, however, that a correct hypothesis is rejected whereas a wrong one is accepted. The corresponding probabilities are the parameters  $\alpha$  and  $\beta$  mentioned above. From

$$\alpha = \text{prob} \{MUF \geq M_s | H_0\}$$

then with

$$MUF \sim N(0, \sigma_{MUF}^2)$$

it follows that

$$1 - \alpha = \phi \left( \frac{M_s}{\sigma_{MUF}} \right)$$

where  $\phi$  denotes the distribution function of the standard normal distribution. If the argument of  $\phi$  corresponding to the functional value  $1 - \alpha$  is designated as  $Z_{1-\alpha}$  it follows that

$$M_s = Z_{1-\alpha} \sigma_{MUF}$$

Starting from the assumption that  $\sigma'_{MUF}$  corresponds to the international standard of the facility type under consideration, the value of  $M_s$  is determined exclusively by  $\alpha$ . In the case where the real MUF value is greater than or equal to  $M_s$  and thus the decision is made in favour of the alternative hypothesis  $H_1$ , the question arises about the expected value of the distribution corresponding

to this hypothesis. This value can be calculated using the definition of non-detection probability. From

$$\beta = \text{prob}\{\text{MUF} < M_s | H_1\}$$

then with

$$\text{MUF} \sim N(M, \sigma_{\text{MUF}}^2)$$

it follows that

$$\beta = \phi\left(\frac{M_s - M}{\sigma_{\text{MUF}}}\right)$$

and

$$1 - \beta = \phi\left(\frac{M}{\sigma_{\text{MUF}}} - Z_{1-\alpha}\right)$$

Hence it follows that

$$M = (Z_{1-\alpha} + Z_{1-\beta})\sigma_{\text{MUF}}$$

or

$$M = (Z_{1-\alpha} + Z_{1-\beta})\sigma'_{\text{MUF}} I$$

In the equation  $\sigma'_{\text{MUF}}$  represents the international standard of accountancy and  $I$  the larger of the throughput or inventory of a material balance.  $M$  is the material quantity corresponding to a given  $\sigma_{\text{MUF}}$ , the diversion of which can be just about detected with a detection probability of  $1 - \beta$ . The detection probability of a diversion compared with a given  $1 - \beta$  will increase if the real MUF becomes greater than  $M$  and will decrease if MUF remains smaller than  $M$ .

The relationships discussed are illustrated in Fig. 1. The denotation of  $M$  as the expected accountancy capability of a control system is, however, an unfortunate choice of terminology, because the accountancy capability is not the better, the greater  $M$ . Just the opposite is the case. Expected accountancy incapacity would have been a more appropriate name.

### 3. MUF EVALUATION FOR A SYSTEM OF MBAs

Starting from the assumption that the detection goals  $M_1$ ,  $\alpha_1$  and  $\beta_1$  are known or given for the individual MBAs of a State, it shall be investigated what



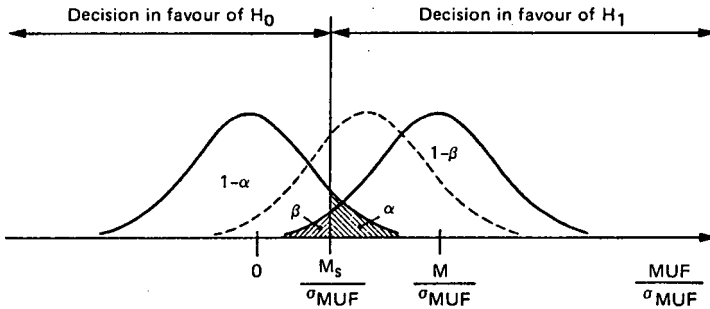


FIG. 1. Representation of the relationships between  $\alpha$ ,  $\beta$ ,  $1 - \alpha$  and  $1 - \beta$  of the null and alternative hypotheses.

statements are possible with respect to the values  $1 - \alpha$ ,  $1 - \beta$  and  $M$  for a system of MBAs. The systems  $S_i$  are considered to be  $N$  partial systems of an entire system  $S$ , i.e.  $i = 1, \dots, N$ .

The problem can again be solved as a test problem. For this purpose the hypothesis

$H_0$ : No nuclear material was diverted in the system if no nuclear material was diverted from one of the partial systems,  
i.e.  $E(MUF_i) = 0$  for all  $i = 1, \dots, N$ ,

is to be tested against the alternative hypothesis

$H_1$ : Nuclear material was diverted in the system if at least in one partial system  $i$  a quantity of at least  $M_i$  was diverted,  
i.e.  $E(MUF_i) \geq M_i$  for at least one  $i = 1, \dots, N$ .

The next step is to calculate the system parameters  $1 - \alpha$  and  $1 - \beta$ . The confidence level  $1 - \alpha$ , decisive for acceptance or rejection of the null hypothesis, is calculated by using the false alarm probabilities  $\alpha_i$  which are assumed to be known for all partial systems:

$$1 - \alpha = \text{prob} \{ \text{the decision that } MUF_i < M_{s_i} \text{ for all } i \mid \text{on condition that } E(MUF_i) = 0 \text{ holds for all } i \}$$

Hence it follows that

$$\begin{aligned} 1 - \alpha &= \text{prob} \{ MUF_1 < M_{s_1} \mid E(MUF_1) = 0 \text{ and } MUF_2 < M_{s_2} \mid E(MUF_2) = 0 \dots \\ &\quad \text{and } MUF_N < M_{s_N} \mid E(MUF_N) = 0 \} \\ &= (1 - \alpha_1)(1 - \alpha_2) \dots (1 - \alpha_N) = \prod_{i=1}^N (1 - \alpha_i) \end{aligned}$$

i.e. that with an increasing number of MBAs the confidence level of statements on the system decreases and the false alarm probability

$$\alpha = 1 - \prod_{i=1}^N (1 - \alpha_i)$$

increases. This result agrees also with intuition. Since the statements on every individual system are made with a confidence level smaller than 1, i.e. they are not completely certain, the false alarm probability of the system increases with the number of partial systems on which these statements are made.

The calculation of the non-detection probability  $\beta$  is more complicated since a whole class of hypotheses has to be considered as an alternative. For this reason an upper bound for the non-detection probability is given.

Non-detection occurs if in all  $N$  systems  $MUF_i < M_{s_i}$  is determined but in at least one system  $j, j \in \{1, \dots, N\}$ ,  $E(MUF_j) \geq M_j$  is true. The derivation of an upper bound for the non-detection probability can be restricted to the alternatives of the partial systems

$$E(MUF_j) = M_j, \quad j = 1, \dots, N$$

and it can be assumed that the most unfavourable case for detection is diversion in only one system.

For the case where such a diversion is not detected,

$$\tilde{\beta}_j = \text{prob}\{MUF_1 < M_{s_1} | E(MUF_1) = 0 \dots \text{and } MUF_j < M_{s_j} | E(MUF_j) = M_j \dots \\ \text{and } MUF_N < M_{s_N} | E(MUF_N) = 0\}$$

$$\tilde{\beta}_j = (1 - \alpha_1) \dots (1 - \alpha_{j-1}) \beta_j (1 - \alpha_{j+1}) \dots (1 - \alpha_N)$$

The upper bound for the non-detection probability is obtained from

$$\beta = \max_{j=1, \dots, N} (\beta_j) = \beta_{j^*}$$

where  $j^*$  is that index for which

$$\frac{\beta_{j^*}}{1 - \alpha_{j^*}} \geq \frac{\beta_j}{1 - \alpha_j} \quad \text{for all } j = 1, \dots, N$$

The question now to be discussed is: what can be stated when applying the procedure of test series at a given total detection probability  $1 - \beta$  for a total expected accountancy capability  $M$ ? When answering this question a new

formulation of the null and alternative hypotheses seems to be more adequate. More correctly formulated, the hypotheses are:

$H_0$ : No nuclear material was diverted in the system,  
i.e.  $E(MUF_i) = 0$  for all  $i = 1, \dots, N$ ,

$H_1$ : At least a total quantity  $M$  of nuclear material was diverted in the system,  
i.e.  $\sum_{i=1}^N E(MUF_i) = \sum M_i = M$ .

A solution of this problem is not possible without taking into account the diversion strategy.

It can hardly be expected that a diverter will divert the material quantity  $M$  wanted from only one MBA, since this would mean running a high risk of detection. It is therefore assumed that he draws this quantity from small diversions in the individual systems. The question of interest here is: if the tests made in all of the MBAs lead to acceptance of the null hypothesis, what total diversion  $M$  can be ruled out with a probability of  $1 - \beta$ ?

To answer this question, first the most favourable strategy for the diverter should be analysed. This evidently consists in dividing the total diversion  $M$  into the individual systems in such a way that the total non-detection probability  $\beta$  becomes a maximum. Every deviation from this strategy would increase the probability of detecting the diversion.

With allowance being made for non-detectable diversion from all individual MBAs, the non-detection probability for the system is

$$\beta = \text{prob}\{\text{the decision that } MUF_i < M_{s_i} \text{ for all } i \mid \text{although } E(MUF_i) = M_i \text{ is true for all } i\}$$

$$= \text{prob}\{MUF_1 < M_{s_1} \mid E(MUF) = M_1 \dots \\ \text{and } MUF_2 < M_{s_2} \mid E(MUF) = M_2 \dots \\ \text{and } MUF_N < M_{s_N} \mid E(MUF) = M_N\}$$

$$= \beta_1 \beta_2 \dots \beta_N = \prod_{i=1}^N \beta_i$$

To simplify the following mathematical derivations, instead of the distributions

$$MUF_i \sim N(0, \sigma_{MUF_i}^2) \text{ and } N(M_i, \sigma_{MUF_i}^2)$$

the distributions

$$\frac{\text{MUF}_i}{\sigma_{\text{MUF}_i}} = \text{MUF}^* \sim N(0, 1) \text{ and } N\left(\frac{\text{M}_i}{\sigma_{\text{MUF}_i}} = \text{M}_i^*, 1\right)$$

are introduced. Then for an individual system

$$Z_{1-\alpha_i} = \text{M}_{s_i}^* \text{ and } \beta_i = \phi(\text{M}_{s_i}^* - \text{M}_i^*)$$

and for the total system

$$\beta = \prod_{i=1}^N \beta_i = \phi(Z_{1-\alpha_1} - \text{M}_1^*) \dots \phi(Z_{1-\alpha_N} - \text{M}_N^*)$$

To calculate the maximum of  $\beta$ , weighting factors  $\gamma_i$  are introduced. With

$$\text{M}_i^* = \gamma_i \text{M}^* \text{ and } \sum_{i=1}^N \gamma_i = 1$$

it follows that

$$\text{M}^* = \gamma_1 \text{M}^* + \gamma_2 \text{M}^* \dots + \gamma_N \text{M}^*$$

Using the relation  $\gamma_N = 1 - \sum_{i=1}^N \gamma_i$ , the equation for  $\beta$  can be put in logarithmic form, and the partial derivations

$$\frac{\partial}{\partial \gamma_1} \ln \beta, \quad \frac{\partial}{\partial \gamma_2} \ln \beta, \dots, \quad \frac{\partial}{\partial \gamma_{N-1}} \ln \beta$$

formed and put equal to zero. Allowing for the fact that the derivations of logarithms are equal if their arguments are equal, a linear equation system is obtained to determine the weighting factors:

$$\begin{array}{rcl}
 M^* \gamma_1 & & -M^* \gamma_N = Z_{1-\alpha_1} - Z_{1-\alpha_N} \\
 M^* \gamma_2 & & -M^* \gamma_N = Z_{1-\alpha_2} - Z_{1-\alpha_N} \\
 & & \vdots \\
 & & M^* \gamma_{N-1} - M^* \gamma_N = Z_{1-\alpha_{N-1}} - Z_{1-\alpha_N} \\
 \\ 
 M^* \gamma_1 + M^* \gamma_2 + \dots & + M^* \gamma_N = & M^*
 \end{array}$$

From this the values of  $\gamma_i$  are calculated as

$$\begin{aligned}
 \gamma_i &= \frac{M^* + (N-1)Z_{1-\alpha_i} - \sum_{j=1, j \neq i}^N Z_{1-\alpha_j}}{NM^*}, j = 1, \dots, N \\
 &= \frac{M^* + N Z_{1-\alpha_i} - \sum_{i=1}^N Z_{1-\alpha_i}}{NM^*}
 \end{aligned} \tag{1}$$

Since  $\gamma_i$  can take positive or negative values, two cases have to be distinguished:

*Case A:*  $\gamma_i \geq 0$  for all  $i = 1, \dots, N$

By insertion of the calculated  $\gamma_i$  values into the above arguments of the normal distribution function  $Z_{1-\alpha_i} - \gamma_i M^*$ , all arguments will become equal. From this it follows that, for a given non-detection probability  $\beta$  of the system, the optimal diversion strategy calls for an equal non-detection probability in all partial systems, i.e.

$$\beta_i = \tilde{\beta} = \beta^{1/N}$$

With the optimal diversion strategy being taken into account, the expected accountancy capability  $M$  is calculated. Inserting the  $\gamma_i$  values into the arguments of the normal distribution yields

$$Z_{1-\alpha_i} - \gamma_i M^* = \frac{1}{N} \left( \sum_{i=1}^N Z_{1-\alpha_i} - M^* \right)$$

and

$$\tilde{\beta} = \phi \left[ \frac{1}{N} \left( \sum_{i=1}^N Z_{1-\alpha_i} - M^* \right) \right]$$

With

$$Z_{\tilde{\beta}} = -Z_{1-\tilde{\beta}}$$

then

$$M^* = \sum_{i=1}^N Z_{1-\alpha_i} + N Z_{1-\tilde{\beta}}$$

Finally,

$$M = \sum_{i=1}^N M_i = \sum_{i=1}^N M_i^* \sigma_i = M^* \sum_{i=1}^N \gamma_i \sigma_i$$

$$M = \left( \sum_{i=1}^N Z_{1-\alpha_i} + N Z_{1-\tilde{\beta}} \right) \sum_{i=1}^N \gamma_i \sigma_i$$

Inserting the value for  $M^*$  into (1) gives

$$\gamma_i = \frac{Z_{1-\alpha_i} + Z_{1-\tilde{\beta}}}{\sum (Z_{1-\alpha_i} + Z_{1-\tilde{\beta}}) \sigma_i}$$

and thus

$$M = \sum_{i=1}^N (Z_{1-\alpha_i} + Z_{1-\tilde{\beta}}) \sigma_i \quad (2)$$

The diversion quantity  $M$  which can be ruled out in the total system with a probability of at least  $1 - \beta$  is the sum of the diversions from individual systems when taking into account the optimal diversion strategy, i.e.  $\beta_i = \tilde{\beta} = \beta^{1/N}$ .

Case B:  $\gamma_i \leq 0$  for some  $i \in \{1, \dots, N\}$

The results obtained so far hold for the case where the condition  $\gamma_i \geq 0$  is met for all  $i$ . As can be seen from (1), this need not be so for highly different  $\alpha_i$  values in the partial systems. What is now the optimal diversion strategy to obtain a quantity  $M$ ? If not all  $\gamma_i \geq 0$ , it is assumed, without restriction of generality, that

$$Z_{1-\alpha_1} \leq Z_{1-\alpha_2} \dots \leq Z_{1-\alpha_N} \quad (3)$$

From Eq. (1) for  $\gamma_i$  it follows that  $\gamma_i \geq 0$  if

$$M^* + N Z_{1-\alpha_i} - \sum_{i=1}^N Z_{1-\alpha_i} \geq 0$$

and  $\gamma_i < 0$  if

$$M^* < \sum_{i=1}^N Z_{1-\alpha_i} - N Z_{1-\alpha_i}$$

For this case a smallest  $i = i^* \geq 2$  exists for which

$$M^* \geq \sum_{i=i^*}^N Z_{1-\alpha_i} - [N - (i^* - 1)] Z_{1-\alpha_{i^*}}$$

Under these conditions a diverter will try to make the diversion in such a way that the variation of non-detection probabilities  $\beta_i$  remains as small as possible. In diversion of the quantity  $M$  the optimum for  $\beta$  is attained if

$$\gamma_1 = \gamma_2 = \gamma_3 \dots = \gamma_{i^*-1} = 0$$

and

$$\gamma_i = \frac{M^* + [N - (i^* - 1)]Z_{1-\alpha_i} - \sum_{i=i^*}^N Z_{1-\alpha_i}}{[N - (i^* - 1)]M^*}, i = i^*, \dots, N \quad (4)$$

i.e. the diversion is reduced to the partial systems having the greatest non-detection probabilities [3].

The optimal  $\beta_i$  values of the partial systems can be calculated by proceeding from the  $\beta$ -value of the entire system in the following way. For at least one partial system, allowing for (3), it is true that

$$\phi(Z_{1-\alpha_1}) < \beta^{1/N}$$

Then it is checked whether the condition

$$\phi(Z_{1-\alpha_2}) \geq \left( \frac{\beta}{\phi(Z_{1-\alpha_1})} \right)^{1/(N-1)}$$

is met. If this is not the case, it is further checked whether

$$\phi(Z_{1-\alpha_3}) = \left( \frac{\beta}{\phi(Z_{1-\alpha_1})\phi(Z_{1-\alpha_2})} \right)^{1/(N-2)}$$

holds, etc. This is to be continued up to  $i = i^*$ :

$$\phi(Z_{1-\alpha_{i^*}}) \geq \left( \frac{\beta}{\prod_{i=1}^{i^*-1} \phi(Z_{1-\alpha_i})} \right)^{1/[N-(i^*-1)]} =: \beta_{i^*}$$

With

$$\phi(Z_{1-\alpha_i} - \gamma_i M^*) = \phi \left[ \frac{1}{N - (i^* - 1)} \left( \sum_{i=i^*}^N Z_{1-\alpha_i} - M^* \right) \right] = \beta_{i^*}$$

$M^*$  is given by

$$M^* = \sum_{i=1}^N Z_{1-\alpha_i} + [N - (i^* - 1)]Z_{1-i^*}$$



Replacing  $M^*$  with  $M$ , one obtains by means of the same transformations as in Case A:

$$M = \sum_{i=i^*}^N (Z_{1-\alpha_i} + Z_{1-\beta_{i^*}}) \sigma_i \quad (5)$$

So far, the evaluation of a system of MBAs has started from an evaluation of its individual systems. A system evaluation, however, can also be attained by a more global procedure. For this purpose the global test parameter  $MUF = \sum_{i=1}^N MUF_i$  with

$$E(MUF) = \sum_{i=1}^N E(MUF_i) \text{ and } \sigma_{MUF}^2 = \sum_{i=1}^N \sigma_{MUF_i}^2$$

is formed, from which

$$MUF \sim N\left(0, \sum_{i=1}^N \sigma_{MUF_i}^2\right) \text{ and } MUF \sim N\left(\sum_{i=1}^N E(MUF_i), \sum_{i=1}^N \sigma_{MUF_i}^2\right)$$

The expected accountability capability is calculated formally in the same way as for an MBA, resulting formally also in the same final equation:

$$M = (Z_{1-\alpha} + Z_{1-\beta}) \sigma_{MUF} \quad (6)$$

However, for the global system the probabilities  $\alpha$  and  $\beta$  have to be agreed to separately, they cannot be calculated from  $\alpha_i$  and  $\beta_i$  values of the individual MBAs. Certainly this agreement will be a compromise between the desirable reliability of the inspection results and economic constraints.  $\sigma_{MUF}$  can be represented as

$$\sigma_{MUF}^2 = \sum_{i=1}^N \sigma_{MUF_i}'^2 I_i^2$$

where the  $\sigma_{MUF_i}'$  are the international standards of accountability [4] and  $I_i$  denotes the larger of the inventory or throughput for the material balance.

In this context it has to be recognized that national transfers of nuclear material do not contribute to the  $\sigma_{MUF}$  of the system. Therefore, the inventory

will dominate the throughput and  $\sigma_{\text{MUF}}$  in the global concept will tend to be lower than in the previous concept based on individual MBAs.

From comparison of the results of the two concepts it can be stated that, with the system parameters  $\alpha$  and  $\beta$  being left as constant, the expected accountancy capability does not differ much if the system contains only a relatively small number of MBAs. With an increasing number of MBAs in the system the global expected accountancy capability (Eq. (6)) becomes significantly lower than in the case of the individual MBA concept (Eq. (2)).

The above considerations are limited to measured material balances. To use them for evaluations of real national fuel cycles, two extensions of the concept are required. First, as nuclear fuel cycles consist of bulk and item MBAs, the latter have to be included; and second, apart from the above mentioned differences between the two concepts a far more comprehensive investigation is necessary before one can decide which concept should be given preference.

A further extension towards an overall evaluation would be the assessment of the influence on the inspection goal attainment of additional inspection activities, such as containment and surveillance, and of the measures and methods for the treatment of anomalies.

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## FUEL CYCLE SAFEGUARDS APPROACHES

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### Abstract

#### FUEL CYCLE SAFEGUARDS APPROACHES.

A review is presented of the status of studies recently performed in the United States of America on alternative safeguards approaches for light water reactor fuel cycles under the safeguards system of the Non-Proliferation Treaty; and a new method is described for consistent allocation of limited inspection manpower resources. The studies concern the advantages and disadvantages of using safeguards approaches for groups of nuclear facilities that interact to form a nuclear fuel cycle, rather than an approach that is facility specific, treating each facility as an isolated entity. The allocation methodology makes use of inspection evaluation criteria used by the IAEA, as well as a procedure to provide consistency in allocation of limited resources to individual facilities of any fuel cycle while considering all other facilities in the fuel cycle.

### 1. INTRODUCTION

Much has been done by the International Atomic Energy Agency to improve safeguards utilization and effectiveness without increasing the size of the IAEA safeguards staff, including improvement of the concentration of available inspection efforts on direct-use material [1]. Member States and the IAEA have encouraged studies of ways to improve the efficiency and reduce the burden of safeguards. Many past studies have considered facility specific safeguards, treating each facility as an isolated entity. In December 1984, the IAEA convened a Consultants Meeting [2, 3] to consider whether facility specific safeguards might be modified to reflect the relationships between facilities in the fuel cycle of a State under the safeguards system of the Non-Proliferation Treaty (NPT) as defined in IAEA INFCIRC/153 [4]. In connection with this meeting, the United States of America performed the fuel cycle studies for light water reactor (LWR) fuel cycles discussed below.

These LWR fuel cycle safeguards depart from facility specific safeguards in various ways. In the best understood fuel cycle safeguards approach, three zones

would be defined among multiple facilities that are interconnected as shipper-receiver pairs, with the three individual zones including (1) nothing but low enrichment uranium (LEU), or (2) irradiated fuel assemblies containing plutonium, or (3) all recovered plutonium. Each zone is an extended material balance area (MBA) for material accounting purposes. Verification of some or all interfacility, intrazone nuclear material transfers would be eliminated, but assurance would exist that all material in the zone is verified only once while an inventory is made of the entire zone. Other safeguards approaches, for instance random inspections, have little or no intrinsic dependence on fuel cycle relationships.

Experts also studied the design of a safeguards approach for a fuel cycle by introducing weighting factors with a measure of objective validity for use in establishing the relative safeguards significance of the kind of material in a zone and the corresponding amount of inspection effort required.

The IAEA now expends about 8000 inspector man-days per year at about 500 installations. This is substantially less than the total effort the IAEA has the right to apply under existing facility attachments, but reflects a serious effort to achieve maximum use of available staff resources. Allocation of inspection manpower resources is a complex decision making process that has a strong influence on the quality of inspections and subsequent evaluation of safeguards effectiveness. We think that the resource allocation procedure presented in this paper would be useful for application to any type of fuel cycle and be fully consistent with established safeguards practices as currently applied under INFCIRC/153, while minimizing other subjective judgements and extraneous considerations involved in assigning inspection effort. It is not an intrinsic fuel cycle safeguards approach in that it can be applied to any collection of facilities, but it does inherently reflect the presence of direct-use material in the different facilities.

## 2. THE ZONE APPROACH

For the fuel cycle studies summarized here, fuel cycles which might exist in advanced NPT States were postulated with two or three LEU conversion and fabrication plants, about 20 LWRs, a reprocessing plant and a smaller mixed oxide (MOX) fuel fabrication plant. One study considers five different versions of the safeguards approach for the LEU in zone 2, ranging from the discrete facility by facility approach to treating the LEU zone (conversion, fabrication and reactor fresh fuel store) as one extended MBA [5, 6]. Using available estimates for time required per inspector activity, the number of inspector man-days expended per year for this extended MBA approach to the LEU zone was calculated as well as the number for the facility specific approach with full verification of receipts and shipments at each facility.

The reductions in man-days that result when departures are made from the facility specific approach do not reflect a potential for future additional savings. They are

more comparable to the difference that would exist between the actual routine inspection effort (ARIE) of such a fuel cycle and the smaller number of man-days that would be implemented by the IAEA, as has been the practice in recent years at facilities under safeguards. In this regard, the more complete the elimination of verification of intrazone shipments and receipts, the greater the reduction in specificity of verified information about the disposition of nuclear material within the fuel cycle. Loss of this information makes it more difficult to detect diversions concealed by keeping on hand material declared to be in transit, or by concealing diversion at one facility within the measurement errors of another facility in the same zone.

Inspector availability and the feasibility of scheduling inventory taking and verification are major considerations because physical inventory verification is to be essentially simultaneous throughout the LEU zone, to ensure that all material in the zone is verified only once at inventory time. The practicality of this simultaneous verification remains to be demonstrated for the LEU zone.

Another study of LWR fuel cycle zones was performed by three senior safeguards experts from the United States of America and the Federal Republic of Germany. The study considers a roughly comparable fuel cycle of LEU bulk handling facilities and nuclear power plants and includes explicit consideration of NPT safeguards for a reprocessing plant and MOX fabrication facility [7]. In this analysis, the inspection effort expended in each zone is compared to the quantity of nuclear material in the zone expressed in terms of its safeguards significance. Weighting factors introduced by the IAEA in 1980 are used to adjust the quantity of material in a way that reflects the decreased safeguards significance of material that is harder to convert to directly usable weapons material. The weighting factors used are 0.33 for fresh MOX fuel, 0.1 for plutonium in irradiated fuel and 0.02 for LEU, relative to the safeguards significance of plutonium metal. They are the reciprocals of the approximate conversion times measured in weeks. We consider the calculated number of inspector man-days per weighted significant quantity for the low enrichment zone, 4.2, to be appropriate because it is comparable to the 5.9 calculated for the zone containing separated plutonium. We reached this level of comparability by omitting all verification of internal transfers between facilities in the low enrichment zone in order to obtain the desired balance between safeguards significance and inspection effort across the fuel cycle zones. We suggest several ways to ease somewhat the requirement for simultaneous inventories.

The studies described above provide rationales for low intensity in the safeguards approach for the LEU portion of the fuel cycle, intended to help rationalize the balance and improve safeguards efficiency and credibility. The IAEA is already concentrating available inspection resources on 'sensitive facilities' such as enrichment plants, reprocessing plants and other facilities with large amounts of direct-use material [1], and a further reduction in the resources allocated for LEU verification is not being recommended. However, the results of the three-expert study depend on the reasonableness of the weighting factors. Since 1980, when the above mentioned factors were introduced, effective technologies for enrichment of uranium

have been developed and disseminated. If this trend continues, the weighting factor for LEU may have to be increased somewhat in the future, and this would tend to increase the inspection effort on LEU.

### **3. ALLOCATION OF RESOURCES USING SAFEGUARDS IMPLEMENTATION REPORT CRITERIA**

#### **3.1. Introduction**

This resource allocation method is applicable to various fuel cycles and reflects fully the criteria for evaluating inspection goal attainment used by IAEA staff in preparing their annual Safeguards Implementation Report (the SIR criteria). These criteria reflect an implicit ranking of material types according to their safeguards importance. In fact, the criteria reflect judgements on inspection activities necessary or useful for the various categories of material in various types of facility. The criteria are intended for use in evaluating the attainment of safeguards goals. Four levels of attainment are recognized: partial or full attainment of nuclear material verification, and almost or fully timely completion of the verification.

Allocation methods reflecting the SIR criteria promise several advantages:

- (a) They make use of a careful and informed effort by the IAEA Department of Safeguards to construct an effective and defensible structure for relating the objective importance of specific inspection activities to inspection goals.
- (b) Allocations of effort reflecting the SIR criteria ensure an overall consistency in the amount of inspection effort expended on the various materials inspected.
- (c) Methods using the qualitative values embodied in the SIR criteria avoid the process of obtaining consensus on quantifying subjective judgements as to the relative importance of different types of material, facility and inspection activity.

In addition to these advantages, an easily used allocation method of this type can be useful for evaluating different approaches to planning and evaluating inspections.

A significant feature of such an allocation method is the consistent allocation of resources to verification of material by categories, even though the categories may be distributed with complete variability among different types of facility.

#### **3.2. Inspection activities at individual facilities**

The SIR criteria reflect a ranking in safeguards importance for the material categories and require corresponding quality and timeliness of inspection activities for each material category. Direct-use material, plutonium and highly enriched uranium, is of highest importance, but has somewhat reduced importance when in spent fuel. Low enriched and natural uranium are of lesser importance. In general, the

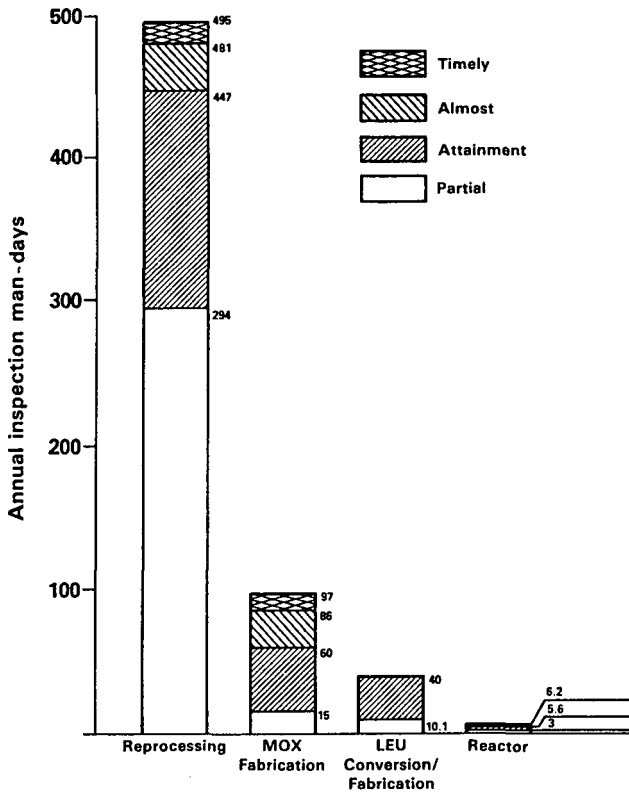


FIG. 1. Inspection man-days for SIR attainment levels at reference facilities.

inspection activities for material of higher safeguards significance deal with smaller goal quantities, more frequent inventory verification and a higher probability of detecting defects. For each material category in a reactor, or other type of facility, the SIR criteria state the inspection activities to be performed and grade the effectiveness of these activities in the four attainment levels according to their quality and timeliness. The annual inspection man-days shown in Fig. 1 for four reference facilities, the types considered in this paper, represent the inspection effort required to reach the four attainment levels established in the SIR criteria.

The number of inspector man-days associated with each inspection activity required under the SIR criteria for the various attainment levels was obtained by employing commonly used lists of inspection activities for each function performed at each facility type and corresponding estimates of the man-days required for the listed activities, as has been done in other studies [7-9].

### 3.3. Allocation of inspection effort

A new rationale is presented for allocating inspector effort to an arbitrary group of facilities when the need for allocation is due to a short-fall in available man-days. The rationale is consistent with safeguards goals, avoids quantification of subjective considerations and is applicable in a consistent way to any such group of facilities. The proposed allocation method uses an algorithm that requires relative and qualitative judgements rather than quantitative judgements. The orders of preference used are consistent with those reflected in the current structure of the SIR criteria.

We formulate the problem of assigning inspection resources to a group of facilities as a constrained optimization problem in which resources are allocated to maximize attainment according to the SIR criteria subject to limitations on available inspection resources. Because our approach to assigning inspection resources emphasizes material categories rather than facilities, we do not distinguish between material of the same category at different facilities. Thus, we require resource allocations which achieve a uniform attainment level across a material category in a fuel cycle.

We denote the possible attainment levels for each material category by the variables  $A_{DU}$  for direct-use material other than spent fuel,  $A_{SF}$  for spent fuel and  $A_{IU}$  for indirect-use material. These variables take one of the four previously defined values: partial attainment, attainment, almost timely attainment and timely attainment. To determine each allocation of inspection effort, we associate an attainment triplet  $(A_{DU}, A_{SF}, A_{IU})$  that is a non-quantitative measure of the effectiveness of the allocated inspection effort.

Consistent with the SIR criteria, we introduce the additional constraint that every inspection allocation should result in attainment levels satisfying

$$A_{DU} \geq A_{SF} \geq A_{IU} \quad (1)$$

In addition, the limitation on available inspection man-days that can be provided by the IAEA suggests a constraint

$$\text{cost}(A_{DU}) + \text{cost}(A_{SF}) + \text{cost}(A_{IU}) \leq \text{total man-days} \quad (2)$$

where, for example,  $\text{cost}(A_{DU})$  indicates the inspection effort in man-days required to achieve a given attainment level  $A_{DU}$  for all direct-use material other than spent fuel in the reference facilities.

Because we cannot yet maximize the attainment levels for the three material categories simultaneously, we establish an additional constraint, namely preference orders on the attainment triplets  $(A_{DU}, A_{SF}, A_{IU})$  that are consistent with the priorities expressed in the SIR criteria and that allow the identification of a most preferred allocation of inspection resources.

Among the many possible preference orders for judging the relative worth of a possible attainment triplet  $(A_{DU}, A_{SF}, A_{IU})$ , two represent the extremes in alloca-



tion rationales. Preference order I prefers the triplet  $(A_{DU}^1, A_{SF}^1, A_{IU}^1)$  to  $(A_{DU}^2, A_{SF}^2, A_{IU}^2)$  when

$$A_{DU}^1 > A_{DU}^2 \quad (3)$$

or

$$A_{DU}^1 = A_{DU}^2 \quad \text{and} \quad A_{SF}^1 > A_{SF}^2 \quad (4)$$

or

$$A_{DU}^1 = A_{DU}^2, \quad A_{SF}^1 = A_{SF}^2 \quad \text{and} \quad A_{IU}^1 > A_{IU}^2 \quad (5)$$

This preference ordering leads to allocations of inspection effort that tend to provide higher attainment levels for the material categories with greater safeguards significance, at the expense of reduced attainment for other categories.

Preference order II prefers triplet  $(A_{DU}^1, A_{SF}^1, A_{IU}^1)$  to triplet  $(A_{DU}^2, A_{SF}^2, A_{IU}^2)$  when the minimum of the three attainment levels  $A_{DU}^1, A_{SF}^1, A_{IU}^1$  is strictly greater than the minimum of the three attainment levels  $A_{DU}^2, A_{SF}^2, A_{IU}^2$ . To avoid ties when these two minima are equal, we prefer the first triplet to the second triplet if one of (3), (4) and (5) is satisfied. This preference ordering results in resource allocations producing nearly uniform attainment levels, with preference given to the more safeguards significant material when complete uniformity cannot be achieved.

The optimization problem becomes: find the most preferred triplet  $(A_{DU}, A_{SF}, A_{IU})$  that is attainable within the constraints of (1) and (2) when the attainment triplets are put in order of priority using preference order I or II.

### 3.4. Examples of groups of facilities

We illustrate the application of these resource allocation procedures with three reference groups of facilities (fuel cycles). Fuel cycle I consists of 20 LWRs, each with a capacity of 1000 MW(e) and with fresh fuel imported and spent fuel exported. Fuel cycle II consists of the 20 LWRs plus three LEU conversion/fabrication plants each with a 400 t throughput of uranium hexafluoride per year from an external source, and export of all spent fuel. Fuel cycle III consists of the 20 LWRs and the three LEU conversion/fabrication plants plus a reprocessing facility with a 240 t heavy metal throughput per year and a MOX fabrication plant with a 500 kg plutonium dioxide throughput per year, import of uranium hexafluoride and export of a portion of the spent fuel.

### 3.5. Application of allocation procedure

This section illustrates the use of the above described adaptive allocation procedures. The total number of man-days theoretically available for a group of facilities

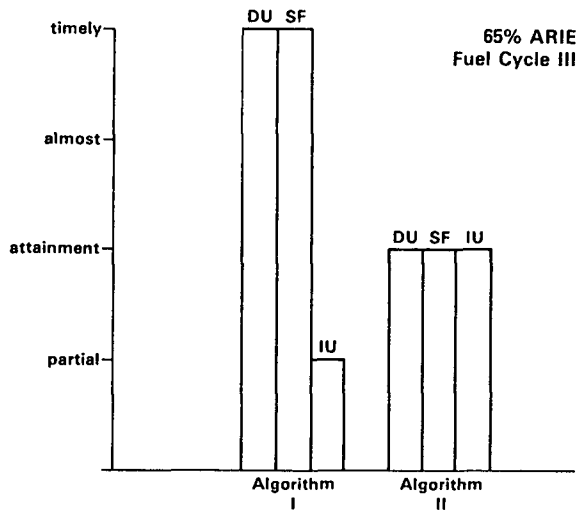


FIG. 2. Dependence of attainment level on allocation algorithm for different material categories.

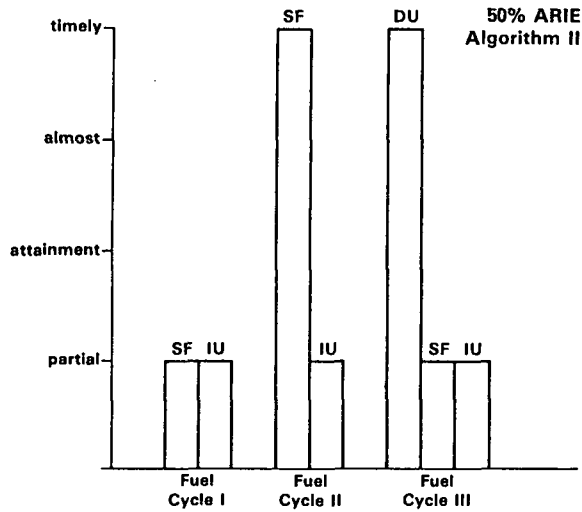


FIG. 3. Dependence of attainment level on fuel cycle context for different material categories.

is derived from the total across all the facilities of the assumed ARIEs for the individual facilities. The figures used for the individual facilities are 10 inspector man-days per year for an LWR, 80 man-days per year for an LEU conversion/fabrication plant, 180 man-days per year for a MOX fabrication plant and 600 man-days per year for a reprocessing plant. These assumed ARIE values reflect those used

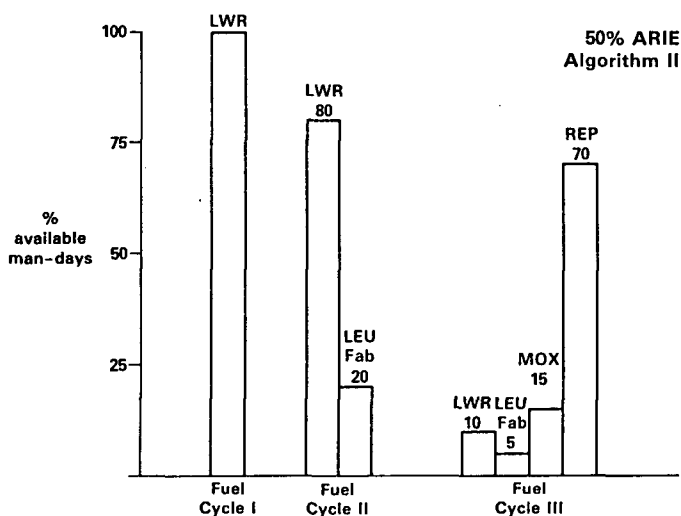


FIG. 4. Dependence of inspection effort allocation on fuel cycle context.

previously by others and are believed to be consistent with IAEA values [5,8,9].

Resource limitations on inspector man-days are reflected in the examples by reducing to 50% or 65% the number of inspector man-days for the total ARIE for a fuel cycle, to obtain the man-days implemented. These percentages are considered to be reasonably close to the values actually assigned by the IAEA in 1982 [8], but are used for illustrative purposes in this paper.

Allocation algorithms I and II represent reasonable extremes among rationales for assigning inspection resources. For an assumed resource of 65% ARIE applied to fuel cycle III, Fig. 2 shows the differences in attainment levels for the various material categories under the two algorithms. Clearly algorithm I favours the more safeguards significant material at the expense of the less significant, the goal for a fuel cycle safeguards approach. Algorithm II seeks a uniform attainment across all material categories. By calculating these two extreme allocations, an analyst can obtain upper and lower limits on the achievable attainment levels.

The influence of the SIR criteria on levels of attainment for the three material categories in fuel cycle contexts is illustrated in Fig. 3, which limits the total man-day resource to 50% ARIE and uses algorithm II. As the size and complexity of the fuel cycle increase, the additional resources that become available are employed to improve attainment for the more significant material. Thus, in fuel cycle II, the attainment level for spent fuel improves at the expense of that for indirect-use material, and similarly in fuel cycle III, direct-use attainment is increased over that for other material. A complementary view of this same effect is represented in Fig. 4, which shows the percentage of the available man-days allocated to each facility type.

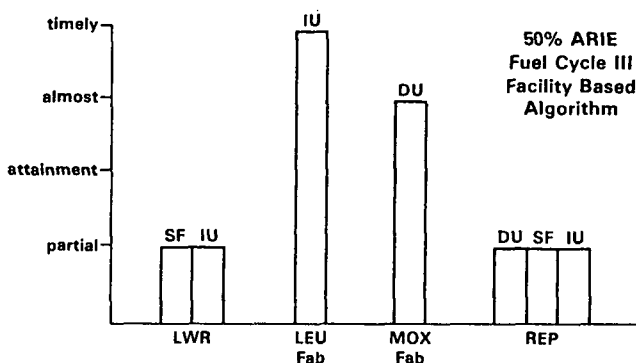


FIG. 5. Attainment levels by facility type under facility based algorithm.

The allocation procedure described in this paper assigns effort to a particular facility while considering the other facilities to be inspected to within the capabilities of the available resources. This procedure contrasts starkly with the conceptual alternative of assigning essentially the same effort to facilities of the same type determined as a percentage of the ARIE, regardless of the other facilities competing for resources. The difference in attainment of safeguards goals is illustrated by comparing Figs 5 and 6. In Fig. 5, attainment levels for material categories at each facility are shown, it being assumed that the effort at each facility is 50% of the ARIE for that facility type. In Fig. 6, algorithm II results in timely attainment for all direct-use material other than spent fuel, with partial attainment for spent fuel and indirect-use material. These results illustrate two important differences between these allocation procedures. First, the allocation procedure reflecting the SIR criteria improves the attainment level for direct-use material relative to the facility based procedure. Second, the attainment level for a material category is uniform using the SIR criteria, whereas the facility based procedure results in a wide variation in attainment level within a material category.

#### 4. CONCLUSIONS

A fuel cycle safeguards study calculated the possible reduction in inspection effort for the fresh fuel zone of a model LWR fuel cycle due to reduction or elimination of verification of intrazone flow between facilities. This does not suggest a further reduction in IAEA inspection effort on LEU but is similar in some respects to a well known difference between the ARIE and the lesser number of inspector man-days implemented in recent years. The reduction in verifications of flows between facilities results in decreasing specificity of verified information about the disposition of nuclear material among the facilities and a material balance uncertainty for the zone that is larger in absolute terms than otherwise would be found for any

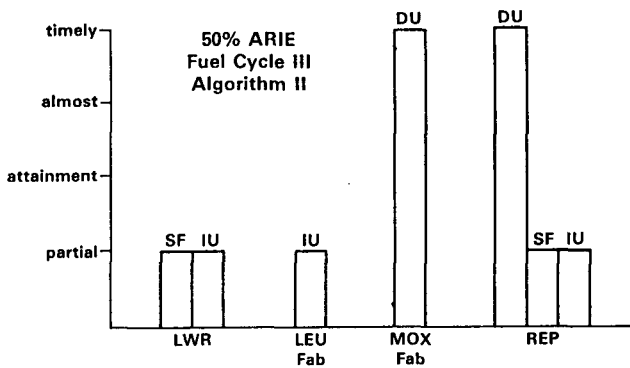


FIG. 6. Attainment levels by facility type under algorithm II.

individual facility. Further study of the feasibility of reductions in verification of intrazone transfers would have required the evaluation of the specific significance of paths for possible diversion known to be introduced by the reductions, as well as specific measures, such as near simultaneous inventories at facilities within a zone to ensure that all material in the zone is verified only once at inventory time. Although helpful and informative, the study was not conclusive as to the disadvantages associated with elimination of intrazone verification of flows.

Another study extended consideration to include the back end of a comparable LWR fuel cycle, including reprocessing and MOX fabrication [7]. The inspection effort expended in each zone was normalized to the quantity of nuclear material in each zone, weighted to reflect its safeguards significance. The results supported elimination of verification of intrazone transfers between facilities. However, the results depend on the acceptability of weighting factors for the relative significance of nuclear material. The values of these weighting factors cannot be demonstrated conclusively to be correct for all time and are not universally accepted. Making choices regarding the acceptability of increased diversion opportunities in selecting weighting factors and obtaining a consensus on them would quite possibly not be achievable and in fact may not be acceptable in such an approach.

Commitments have been made for recycling plutonium in about twenty power reactors and fuel cycle facilities over the next ten years under NPT type safeguards in non-nuclear-weapon States. Plans exist to build and operate MOX fuel fabrication facilities. With plutonium recycling, increases in inspection effort on fresh fuel tend to offset reduced effort on fresh LEU fuel in the LWR fuel cycle.

The new allocation method presented in this paper for use in distributing limited inspection manpower resources incorporates the value judgements reflected in the SIR criteria, including a ranking of material types according to their importance. The method requires an adaptive allocation procedure reflecting preferred attainment levels for material in a fuel cycle. In this way, resources are allocated consistently

for verification of each material category, even though the material may be distributed with complete variability among different types of facility. Algorithms for preference ordering provide for either maximum attainment levels for more safeguards significant material or uniform attainment levels for all material, where resources are too limited to satisfy fully the SIR criteria for an ideal, facility specific regime. Examples were given of the application of this allocation method to three fuel cycles. The examples show the different responses of two algorithms when the numbers and types of facilities are increased. They also show that this allocation procedure assigns effort to a particular facility with consideration given for the other facilities to be inspected to within the capabilities of the available resources.

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## A FRAMEWORK FOR FUEL CYCLE APPROACHES TO IAEA SAFEGUARDS\*

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### Abstract

#### A FRAMEWORK FOR FUEL CYCLE APPROACHES TO IAEA SAFEGUARDS.

A framework is presented for comparing various safeguards verification approaches. Each inventory change, inventory and material balance for each nuclear facility reported by a State may be verified. Verification approaches are compared by listing which of the reports would be verified and to what degree for each approach as it might be applied to a State with a closed fuel cycle. The comparison indicates that the extended material balance area (or zone), the information correlation and the randomization over facilities approaches make more efficient use of IAEA resources than the facility oriented approach for States with large nuclear power programmes. In contrast, any advantages of randomizing inspections over inspection activities within facilities are, in terms of the percentage change in the number of required inspections, relatively independent of the size of a State's nuclear programme.

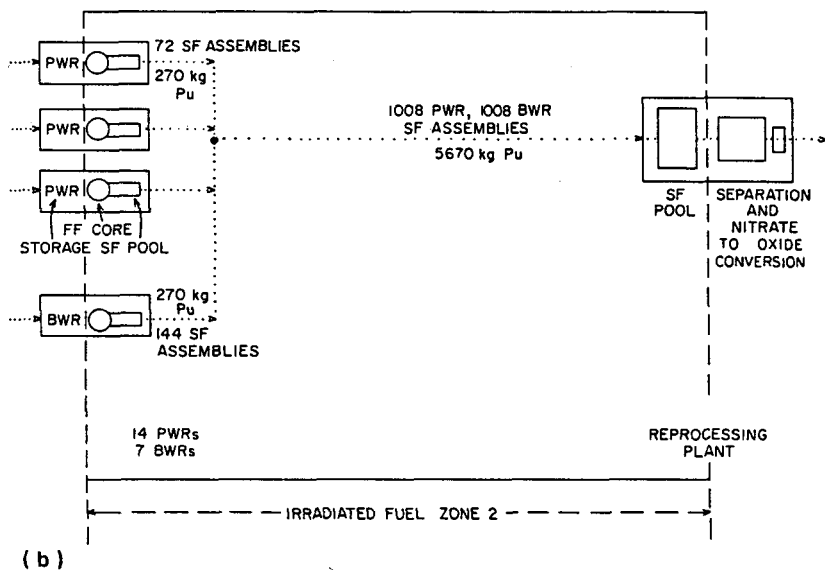
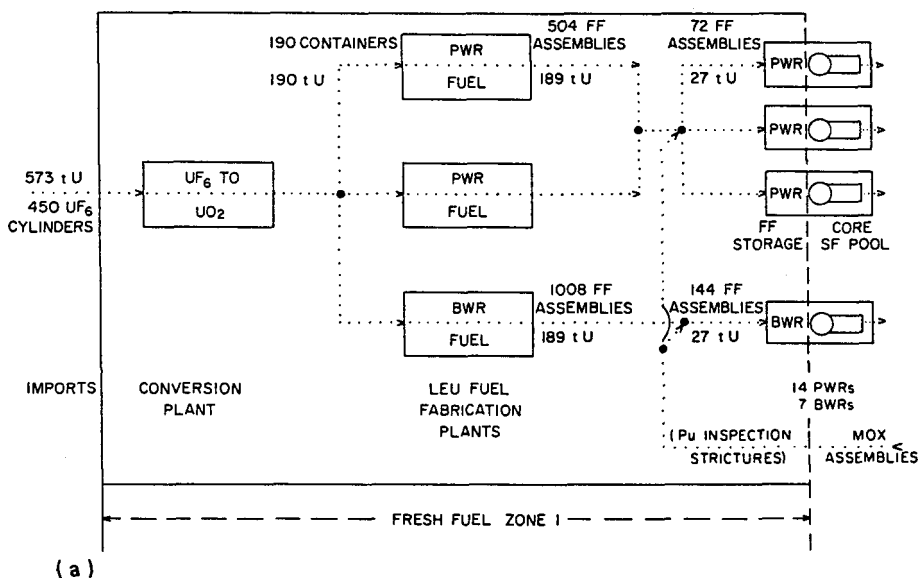
### 1. INTRODUCTION

An International Atomic Energy Agency (IAEA) Consultants Meeting on the Application of Safeguards to Multiple Facility Fuel Cycles was held in December 1984. The working paper, prepared by the Agency staff, described a number of safeguards approaches which might be applied to nuclear facilities in States with nuclear programmes (fuel cycles) of differing sizes and complexity [1]. The consultants recommended [2] that three of the proposals discussed (the extended material balance area (MBA) (or zone), the information correlation and the randomization approaches) deserved further study. The underlying goal is to enhance the efficiency or effectiveness of IAEA safeguards inspections, particularly for large fuel cycles.

The key aspect of a fuel cycle approach is whether the safeguards verifications at a particular type of facility would differ qualitatively from the verifications applied under the

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\* Research carried out under the auspices of the United States Department of Energy under Contract No. DE-AC02-76CH00016.





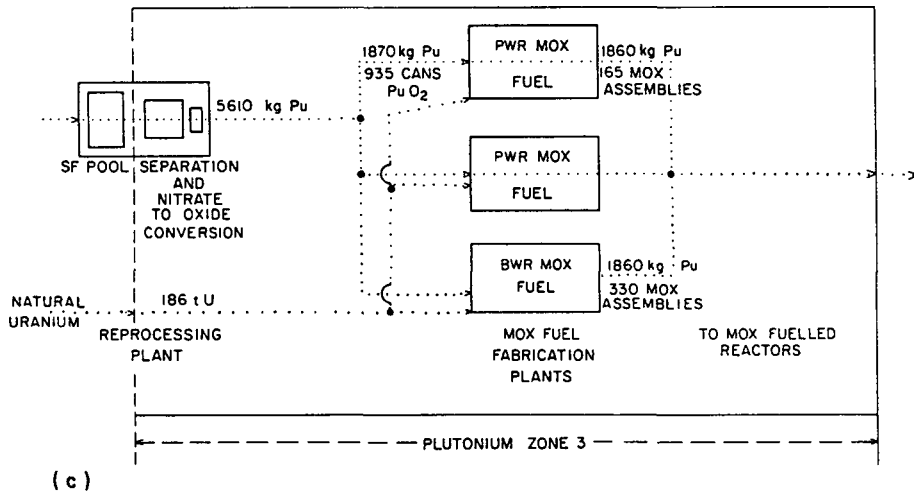


FIG. 1. Zone structure and major annual nuclear material flows for the closed reference fuel cycle. This fuel cycle includes a separate plant for conversion of  $UF_6$  to  $UO_2$ . (a) Fresh fuel, Zone 1, (b) irradiated fuel, Zone 2, and (c) plutonium, Zone 3. The presentation of this fuel cycle in zones in no way prejudices the analysis of safeguards approaches in favour of or against the zone approach.

conventional facility oriented approach because of the size or complexity of the fuel cycle in which the facility is located.

Subsequent to the Consultants Meeting, the framework described below was developed to compare alternative safeguards approaches.

## 2. A CLOSED FUEL CYCLE

The fuel cycle analysed here (Figs 1 (a-c)) includes 14 pressurized water reactors (PWRs), 7 boiling water reactors (BWRs) and facilities to convert low enriched uranium (LEU) hexafluoride to oxide, to fabricate fresh LEU fuel assemblies for all of the reactors, to reprocess all of the spent fuel from the reactors, yielding plutonium oxide, and to fabricate mixed oxide (MOX) fuel for use in 5 PWRs and 3 BWRs. No attempt has been made to calculate steady state balanced flows of LEU and MOX fuels for the fuel cycle. Also, MOX fuel is used in some of the reactors; this forces the imposition of plutonium inspection structures on what otherwise would be areas reserved for LEU.

The safeguards approach notwithstanding, the facilities of the fuel cycle fall into three zones that contain nuclear material with different safeguards significance and different timeliness goals: unirradiated LEU, irradiated fuel assemblies and unirradiated plutonium.

### 3. SAFEGUARDS APPROACHES

#### 3.1. General assumptions

Several assumptions underlie all of the safeguards approaches under study. First, the approaches must adhere to the guidelines of the document INFCIRC/153 [3]. In particular, all nuclear material in the fuel cycle under consideration is subject to IAEA safeguards verifications and there are no unsafeguarded peaceful nuclear facilities.

Second, the IAEA reports annually on its safeguards verification activities. By assumption, therefore, an annual physical inventory verification (PIV) of all nuclear material in the fuel cycles shall be included in any approach considered.

Third, ad hoc inspections to verify design information and the initial inventory of nuclear material and special inspections to verify the information contained in special reports shall be part of all approaches.

Fourth, collusion among different States to carry out or conceal a diversion is assumed not to occur. On the other hand, a low but non-zero probability of diversion anywhere is assumed, so that safeguards verifications at each facility must take this into account. Furthermore, the possibility of undeclared facilities, undeclared nuclear material or unknown technical skills shall not be excluded.

Fifth, material accountancy (MA) supplemented by containment and surveillance (C/S) is the basic safeguards measure for all approaches. Thus, all routine accounting reports specified in INFCIRC/153 would be submitted for each facility containing nuclear material.

Sixth, no new technology shall be considered for any of the safeguards approaches, i.e. all proposed verification activities must already have been demonstrated.

Seventh, only technical factors may be exploited to obtain fuel cycle dependent verifications; political factors

are not considered. Thus, similar fuel cycles in different States must be similarly safeguarded.

Eighth, certain inspection activities involving nuclear material flows across zone boundaries (Fig. 1) within the fuel cycle shall be part of all of the inspection approaches. These include verifications of PWR and BWR refuelling (fresh and spent fuel transfers) and of reprocessing plant dissolver accountability batches.

Ninth, what are presented here for later comparison are theoretical safeguards inspection approaches, of which the facility oriented approach based on facility specific published reports prepared by or for the IAEA is presumably closest to current IAEA objectives.

### 3.2. Facility oriented approach

According to the conventional, facility oriented approach, MA applied to each MBA within each facility is the fundamental safeguards technique (see para. 29 of INFCIRC/153). Thus, each component of the material balance as given in the material balance equation for material unaccounted for (MUF) or related statistics should be verified by the IAEA for each MBA. The equation for a single material balance period is [4]:

$$\text{MUF} = \text{PB} + \text{X} - \text{Y} - \text{PE}$$

where PB is the beginning physical inventory, X is the sum of increases and Y is the sum of decreases to the inventory (encompassing flows), and PE is the ending physical inventory. If sufficient data are given, MUF can be compared with its estimated uncertainty as one MA test to determine if there is a significant, inexplicable deviation from zero, requiring an investigation.

Physical inventories are to be performed by facility personnel at least annually and verified then by Agency inspectors. Physical inventories are to be verified annually at reactors and also whenever reloading occurs. Physical inventories are to be verified annually at the bulk handling facilities (BHF's), and interim inventory verifications are to be performed quarterly at reactors and monthly at facilities in the plutonium zone.

In verifying the inventory of nuclear material at facilities, inspectors collect data that can help rule out diversions concealed by 'borrowing' substitute nuclear material from other facilities or from transit (see Appendix E of

Ref. [5] for a summary and references). Specific measures to preclude such concealments include item identification, C/S measures, and simultaneous inspections at related facilities.

As defined here and as analysed in Section 5, the facility approach for a series of facilities would involve double flow verifications, i.e. at the shipper's and at the receiver's facility.

Because of the absence of a method for measuring the nuclear material content of fuel assemblies, the existing safeguards approach for LEU fabrication plants has not been based on full flow verification [6]. It is assumed here that the neutron collar method [7,8] suffices for this measurement and for the analogous measurement at MOX fabrication plants [9].

### 3.3. Fuel cycle approaches

#### 3.3.1. Information correlation approach

Agency-State safeguards agreements require a State to provide the Agency with reports on inventory changes (regarding receipts and shipments) for each MBA monthly and on MBA inventories annually. Also, inspectors are permitted to obtain up to date book data for each MBA when visiting a facility. The Agency routinely reviews such data to confirm that material shipped is received and that shipper-receiver (S/R) differences are insignificant, and to plan inspections. Given the predictable flows in a fuel cycle, detailed analysis of such information plus information obtained on inspections and from other sources assist in planning verification activities efficiently. Identifying and analysing all of the possibilities and their consequences with respect to needed verification activities are beyond the scope of this study [10-12].

Here, only one obvious conclusion of information correlation is illustrated: the elimination of redundant verification activities, or, in other words, the verification of each transfer either as a shipment or as a receipt.

#### 3.3.2. Zone approach

##### 3.3.2.1. General description

According to the zone or extended MBA approach used here, MA is applied to a collection of facilities or parts thereof in a fuel cycle, instead of to a single facility or part thereof [1,5,10,12-14]. The essence of the zone approach is then the elimination of measurement verifications of

interfacility, intrazone nuclear material flows and the determination of verified inventories for all of the nuclear material within the zone for the beginning and end of the zone material balance period. Interzone flows would continue to be verified.

Since MA would be verified for the zone material balance but not for the material balance for each facility, the particular facility at which there is a nuclear material discrepancy could not in theory be determined from information verified by the IAEA.

### 3.3.2.2. Zone physical inventory verifications

A determination of a zone MUF requires that the physical inventory of the zone be verifiably determined at the beginning and end of the material balance period. A conceptually simple way to do this is by carrying out simultaneous (or nearly simultaneous) PIVs at all the facilities within the zone [1,5,13]. They must extend over a time sufficiently long to allow all the material in transit between the facilities in the zone to be received for verification. An additional benefit of simultaneous PIVs is the prevention of concealment of diversion of nuclear material at one facility by the 'borrowing' of substitute material from another. The importance of gaps in the verified zone inventory due to a lack of simultaneity in facility PIVs [14] depends on the amount of unsealed material (generally, bulk material in process in the facilities), its safeguards significance (e.g. LEU versus Pu), the plausibility of diversion paths (DPs) involving this material, and the timeliness and patterns of information about the nuclear material flows [15].

### 3.3.3. Randomization

#### 3.3.3.1. Over activities

With respect to inspection activities at a single facility, randomization means that the IAEA inspectors would be prepared to carry out many activities but would only carry out some [16,17]. Probably the group of activities subject to random selection would be supplementary to a group that is always carried out. For example, audit reviews would probably always be done, while the decision to observe the Cerenkov glow around spent fuel assemblies in reactor pools could be subject to randomization.

Randomization over activities is presented in two variants. According to the first, the inspection effort is

lower than in the facility oriented approach; the same number of DPs is covered but at a lower probability of detection for each compared with the probability in the facility oriented approach. According to the second, the effort is equal to that in the facility oriented approach but there is a change in the intensities of verification activities from those in the facility oriented approach to put more emphasis on some DPs and less on others.

### 3.3.3.2. Over facilities

With respect to a collection of facilities of a given type, randomization means that IAEA inspectors would be scheduled to carry out inspections at all of them, not necessarily simultaneously, but would only do them at some fraction [18,19] of scheduled visits; the omitted facilities must not be known in advance to the subject State and facilities. Also, operator declarations of nuclear material content must be made before the selection of facilities to be inspected becomes known [20]. Given these conditions, facility randomization could mean, for example, that IAEA inspectors would only appear at fuel fabrication facilities to verify flows some fraction of the number of times they have the permission to do so.

TABLE I. COMPARISON OF APPROACHES FOR CONVERSION PLANT

Information class	Facility oriented	Information correlation	Zone	Random-ization over activities (I) <sup>a</sup>	Random-ization over activities (II) <sup>a</sup>	Random-ization over facilities
Conversion feed <sup>b</sup>	X	X	X	<X	Y	X
Conversion inventory	X	X	X	<X	Y	X
Conversion wastes	X	X	X	<X	Y	X
Conversion product	X			<X	Y	X
Change in DP coverage	Basis	—	—	0 <sup>c</sup>	0	0
Change in effort	Basis	—	—	—	0	0
Conv.-Fab. flow verifications	2/48	1/36	0/0	2/48	2/48	2/<48

TABLE II. COMPARISON OF APPROACHES FOR LEU FUEL FABRICATION PLANTS

Information class	Facility oriented	Information correlation	Zone	Random-ization over activities (I) <sup>a</sup>	Random-ization over activities (II) <sup>a</sup>	Random-ization over facilities
Fabrication feed	X	X		<X	Y	<#
Fabrication inventory	X	X	X	<X	Y	#
Fabrication wastes	X	X	X	<X	Y	<#
Fabrication product	X	X	h	<X	Y	<#
Change in DP coverage	Basis	0	—	0 <sup>c</sup>	0	0 <sup>c</sup>
Change in effort	Basis	0	—	—	0	—
Fab.-Reac. flow verifications	2/120	2/120	1/84	2/48	2/120	2/<120

#### 4. INFORMATION AS FRAMEWORK FOR COMPARING SAFEGUARDS APPROACHES

The three classes of information that can serve as the framework for distinguishing safeguards approaches from one another are inventory data, flow data and S/R data. Roughly speaking, verified inventory and flow data allow material balances to be drawn. Inventory verifications encompass both PIVs and interim inventory verifications. Verified S/R data, depending on the diversion assumptions made, serve as redundant checks on or substitutes for the flow data or as verifications of transportation links.

The accounting information is supplied to the IAEA in regular reports about the nuclear material in all peaceful activities within each State as specified in INFCIRC/153. This information comes from nuclear facilities through each State.

#### 5. FUEL CYCLE DESCRIPTION OF SAFEGUARDS APPROACHES ACCORDING TO INFORMATION FRAMEWORK

##### 5.1. General remarks

Based upon the reference fuel cycle and the types of information available to the IAEA for verification, Tables I-V present a qualitative comparison of the fuel cycle approaches.

TABLE III. COMPARISON OF APPROACHES FOR REACTORS (LEU AND MOX)

Information class	Facility oriented	Information correlation	Zone	Random-ization over activities (I) <sup>a</sup>	Random-ization over activities (II) <sup>a</sup>	Random-ization over facilities
Fresh fuel inventory <sup>d,e</sup>	X + $\delta$	X + $\delta$	X + $\delta$ + $\Delta^h$	< X + $\delta$	Y + $\delta$	< # <sup>i</sup>
Core refuelling <sup>f</sup>	X	X	X	< X	Y	#
Spent fuel inventory <sup>d</sup>	X	X	X	< X	Y	< # <sup>i</sup>
Spent fuel shipments <sup>g</sup>	X			< X	Y	< #
Change in DP coverage	Basis	0	0	0 <sup>c</sup>	0	0 <sup>c</sup>
Change in effort	Basis	0	+	—	0	—
Reac.-Repr. flow verifications	2/108	1/84	0/0	2/108	2/108	2/<108

TABLE IV. COMPARISON OF APPROACHES FOR REPROCESSING PLANT

Information class	Facility oriented	Information correlation	Zone	Random-ization over activities (I) <sup>a</sup>	Random-ization over activities (II) <sup>a</sup>	Random-ization over facilities
Spent fuel receipts	X	X	j	< X	Y	X
Spent fuel inventory	X	X	X	< X	Y	X
Spent fuel dissolution	X	X	X	< X	Y	X
Product, process inventory	X	X	X	< X	Y	X
Wastes	X	X	X	< X	Y	X
Product (Pu, U) shipments	X	X	j	< X	Y	X
Change in DP coverage	Basis	0	—	0 <sup>c</sup>	0	0
Change in effort	Basis	0	—	—	0	0
Repr.-Mfab. flow verifications	2/60	1/24	0/0	2/60	2/60	2/<60



TABLE V. COMPARISON OF APPROACHES FOR MOX FUEL FABRICATION PLANTS

Information class	Facility oriented	Information correlation	Zone	Random-ization over activities (I) <sup>a</sup>	Random-ization over activities (II) <sup>a</sup>	Random-ization over facilities
MOX feed	X	j	j	<X	Y	<#
MOX inventory (PIV)	X	X	X	<X	Y	<#
MOX inventory (interim)	X	X	X	<X	Y	<#
MOX wastes	X	X	X	<X	Y	<#
MOX product	X	X	<sup>h,j</sup>	<X	Y	<#
Change in DP coverage	Basis	—	—	0 <sup>c</sup>	0	0 <sup>c</sup>
Change in effort	Basis	—	—	—	0	—
MOX-Reac. flow verifications	2/120	2/120	1/84	2/120	2/120	2/<120

The columns of the tables represent the different safeguards approaches. The rows represent, first, the classes of verifiable safeguards information from safeguards inspections and, second, the relative effort and effectiveness of the approaches. Symbols represent the status of the row item according to the particular safeguards approach and are defined below:

- X represents the verification activities of the facility oriented approach.
- Y represents different intensities of verification activities than X, but encompassing the same effort per facility.
- δ, Δ represent additional inspection activities.
- <X means fewer activities than X.
- A blank box means that no activities are conducted.
- # means that activities are conducted at the number of facilities within the fuel cycle.
- <# means that activities are conducted at fewer than the number of facilities within the fuel cycle.
- +, -, 0 describe facility specific changes in the number of DPs covered and in inspection effort with respect to the DP coverage and effort in the facility oriented approach ('Basis').

n/N gives the number of flow verification inspections for a single shipment (n) per facility pair (e.g. one conversion and one fabrication plant) and for a year's worth of shipments (N) for all such pairs in the fuel cycle. For the latter, the following assumptions apply: 12 feed and product shipments occur annually at the conversion and LEU and MOX fabrication plants; 4 each occur at the LWRs; and 24 each occur at the reprocessing plant. The nature and intensity of these verifications vary over plant types and safeguards approaches.

#### Footnotes to Tables I-V

- a Variants I and II of the randomization over activities approach describe respectively the cases of reduced overall effort and equal but differently distributed overall effort with respect to the effort required by the facility oriented approach.
- b The verification of conversion plant feed could be eliminated within any approach by relying on international interdependence.
- c Lower probability of detection than in the facility oriented approach for each DP covered.
- d Reactor fresh fuel receipts and spent fuel inventories are verified during both PIVs and interim inventory verifications.
- e The interim inventory period for MOX fuelled reactors is governed by the timeliness criterion for plutonium in unirradiated MOX assemblies in the fresh fuel store. This requires more frequent verifications ( $\delta$ ) in all approaches.
- f Core refuelling information includes verification during a PIV of fresh fuel going into the core, spent fuel coming out and irradiated fuel remaining. If an annual refuelling does not occur, the refuelling verification is replaced by a check of the core containment seal where possible.
- g Sealing only of partially filled spent fuel casks sent from reactors.
- h Including neutron collar verification of fresh fuel assemblies. This could be done at the LEU and MOX fabrication plants within the zone approach if the assemblies are then sealed. Effectiveness suggests doing the verification as late as possible before insertion into the reactor core, and zone efficiency suggests eliminating flow verifications at the fabrication plants. Of course, the collar verification could be done at the reactors within the facility approach.

- i Verified at all reactors once a year during the PIV.
- j Inspection effort savings from the elimination of flow verifications at facilities processing plutonium must be evaluated in the light of the need to conduct interim inventory inspections to satisfy timeliness goals.

## 5.2. Comparison of approaches for a closed fuel cycle

According to the facility oriented approach, there are two verifications of the  $UO_2$  shipped from the single conversion plant (Table I) to the fabrication plants (Table II) as well as two verifications of the assemblies shipped from the fabrication plants to the reactors (Table III). For all flows subject to double verification and especially for fuel assemblies, one verification could involve a measurement and the other a seal verification. Flow verifications are done under this approach for the feed, product and waste streams of the single reprocessing plant (Table IV) and the three MOX fuel fabrication plants (Table V). Because of the stringent timeliness criterion for plutonium, frequent interim inventory inspections are performed at these four facilities as well as at reactors when MOX fresh fuel assemblies are stored (δ in Table III). For MOX fuelled reactors, increases or decreases in the number of verifications due to randomization or the fuel assembly collar measurement within the zone approach refer to this base of extra activity.

According to the information correlation approach, flows between BHF's within the fuel cycle are verified once only. All fabrication plant flows are verified, eliminating the need to verify the product from the conversion plant, but also resulting in fewer DPs covered. The elimination of fresh fuel verifications at reactors lacks great practical significance in terms of inspection effort because such verifications are not made at the time of receipt under the facility oriented approach, but rather at an interim verification or core opening PIV. Under the information correlation approach, therefore, the reactors and fabrication plants undergo the same inspections and thus have the same DP coverage as under the facility oriented approach. All reprocessing plant flows are verified under this approach. Verification of the feed to the MOX plants is based on reprocessing plant product verification, while MOX plant product assemblies are verified at the MOX plants.

For the zone approach, the feed entering the fresh fuel zone is verified as the feed to the conversion plant, the product leaving the zone is verified as the fresh fuel entering the reactor cores, and no major flow need be verified at the

fuel fabrication plant. To compensate for the elimination of product fuel assembly verification at fabrication plants by neutron collar measurement, fresh fuel assemblies are verified by using the collar at the reactors ( $X+\delta+\Delta$  in Table III for reactor fresh fuel inventory). This imposes extra effort at reactors with respect to the qualitative (attributes or seals) verification likely in the facility oriented approach and yields greater fresh fuel DP coverage. Alternatively, the collar verification could occur at the fabrication plants, with the assemblies sealed there and verified at the reactors.

Since irradiated fuel shipments, as intrazone transfers, are not checked as flows but only as inventory changes at PIVs or interim inventory verifications, some spent fuel DP coverage is lost at the reactors. (Overall, no net change in DP coverage at reactors is ascribed to the zone approach.) The process area of the reprocessing plant and the entirety of the MOX fuel fabrication plants undergo PIVs on a zone basis. While the transfers between zones at the head end of the reprocessing plant are verified, the intrazone transfers between the reprocessing plant and the MOX plants are not. The MOX plant product assemblies are verified at the reactors, in analogy with the situation for LEU assemblies in the LEU zone described. The fresh fuel storage areas of these MOX fuelled reactors in effect are part of the plutonium zone. Alternatively, MOX assembly verification could be done at the MOX plant.

Variant I of the randomization over activities approach involves the same activities as the facility oriented approach, but with a lower effort ( $<X$ ). Variant II involves different intensities of activities ( $Y$ ) than the facility oriented approach but the same total effort.

The randomization over facilities approach also involves the same verified activities as the facility oriented approach, but with the omission of all activities at certain facilities ( $<\#$ ).

## 6. DISCUSSION

### 6.1. Intrinsic and extrinsic fuel cycle dependence

Among fuel cycle oriented approaches, a useful distinction that becomes apparent from Tables I-V is that of intrinsic versus extrinsic fuel cycle dependence, as shown in the following classification:

- A. Intrinsic fuel cycle dependence:  
Zone  
Randomization over facilities  
Information correlation
- B. Extrinsic fuel cycle dependence:  
Randomization over activities (variants I and II)
- C. No fuel cycle dependence (by the strict definition of this paper; compare with Ref. [19]):  
Facility oriented.

The information correlation, zone and randomization over facilities approaches are intrinsically fuel cycle dependent because their representation in Tables I-V for some facility types is qualitatively different (e.g. by inclusion of blank entries) from their representation under the facility oriented approach. The randomization over activities approach (both variants) is extrinsically fuel cycle dependent because the table representations for all facility types differ only quantitatively from their representation under the facility oriented approach. More simply stated, intrinsically fuel cycle dependent approaches lack entire classes of verifications that occur under the facility oriented approach because their very definition suggests fuel cycle dependence.

While quantitative analysis is needed to substantiate this conclusion, it is evident that both intrinsically and extrinsically fuel cycle dependent approaches reduce safeguards inspection effort for complex fuel cycles as compared with the facility oriented approach by eliminating verifications.

However, percentage reductions in effort due to extrinsically fuel cycle dependent approaches (e.g. activity randomization) do not depend on the size or complexity of the fuel cycle.

## 6.2. Effort and effectiveness considerations

### 6.2.1. Effort

In addition to displaying the classification of the safeguards approaches according to the verifications they encompass, Tables I-V present qualitatively the effort requirements of the approaches with respect to the effort required to conduct the verifications is summarized in the tables at the facility level. The potential savings in inspection effort according to some approaches due to the

decrease in the number of verifications of flows must be evaluated in the light of the number of interim inventory verifications conducted. Thus, savings are potentially large in the Zone 1 facilities (Tables I and II) because there are no interim inventory verifications to satisfy timeliness goals. Savings are potentially small in Zone 3 because there are monthly interim inventory inspections which encompass verifications of the flows of  $\text{PuO}_2$  and MOX assemblies at the MOX fabrication plants and monthly interim inventory checks and biweekly verifications of the flows at the reprocessing plant.

#### 6.2.2. Effectiveness

Effectiveness, a more elusive concept, is summarized in Tables I-V by two measures, the change in the number of DPs covered at each facility type and the number of interfacility flow verifications carried out. Other quantitative measures of the effectiveness of the safeguards approaches at the facility or zone level include the accountancy verification goal (AVG [4]) and the ratio of the annual inspection effort required to implement the approach to the amount of nuclear material (possibly weighted) thereby safeguarded [14].

Quantitative measures do not, however, capture all aspects of the effectiveness question. In the zone approach, MA is only verified completely at the zone level. The AVG for a zone with many facilities would generally be greater than that for a single facility. At the same time, the AVG for a single facility within the zone would generally be less, if operator flow measurements were accepted without verification (so operator-inspector differences would not contribute to the measurement uncertainty), than if IAEA verifications of the facility flows occurred. However, facility conclusions would be drawn without complete IAEA verification of flows under a zone approach.

The unpredictability of the approaches based on randomization may have subjective safeguards value beyond that conveyed by the quantitative measures in the tables [18].

Finally, the zone approach provides for the minimum number of MA verifications of the fuel cycle (cf. Ref. [10]), the facility oriented approach provides for the maximum number, and the information correlation approach as illustrated here provides for an intermediate number.

## ACKNOWLEDGEMENT

This paper constitutes a synopsis of work [21] for the United States Program for Technical Assistance to IAEA Safeguards.

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# TRIAL APPLICATION OF A SAFEGUARDS PERFORMANCE EVALUATION METHODOLOGY FOR LIGHT WATER REACTORS

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## Abstract

TRIAL APPLICATION OF A SAFEGUARDS PERFORMANCE EVALUATION METHODOLOGY FOR LIGHT WATER REACTORS.

A methodology for evaluating the performance of safeguards implementations under INFCIRC/153 type agreements has been developed and tested using inspection data for nine light water reactor facilities. The methodology makes use of logic programming techniques to allow for both qualitative and quantitative components in the overall assessments. The trial application indicated that the methodology is especially useful as a diagnostic tool, i.e. for pinpointing causes of non-attainment of inspection goals. Routine use would require an efficient interface to IAEA safeguards databases.

## 1. AIMS OF THE TRIAL APPLICATION

In December 1984 a meeting of consultants was held at the Headquarters of the IAEA in Vienna to discuss progress in development of methodologies for the assessment of the effectiveness of international safeguards. During that meeting, experts from the Federal Republic of Germany presented the basic concepts of a Safeguards Performance Evaluation System (SPESY) intended to assist the Agency in a posteriori evaluations of its routine inspection activities. A prototype implementation of the system on a personal computer was also demonstrated. A number of recommendations were made with regard to SPESY. Among these were:

- modify the existing program to accommodate real inspection data;

- perform a trial application on all LWR inspections undertaken during a calendar year under appropriate controls on "safeguards confidential" data;
- examine the potential use of SPESY as an easily understandable tool for use in the design of safeguards approaches and for inspection planning.

The above recommendations have in part been incorporated into the Federal German support programme for the IAEA and the present paper describes progress made so far in their fulfillment. Specifically, trial applications of SPESY to real inspection data have been carried out with the aid of LWR data kindly made available in declassified form by the IAEA.

## 2. REVIEW OF THE METHODOLOGY

### 2.1 General Principles

A detailed description of the motivations and philosophy of SPESY can be found in /1/ and these will be touched on only briefly here.

SPESY measures safeguards performance in terms of degree of inspection goal attainment, subject to constraints which ensure that manifestly peaceful nuclear activities of States will not be impeded. These constraints are introduced implicitly into the evaluation scheme by restricting consideration to a set of inspection activities negotiated and formalized in the relevant Facility Attachments.

The methodology then proceeds from the basic assumption that the best possible evaluation criterion for goal attainment is the diversion detection capability as derived from material accountancy and its verification. In the spirit of the material accountancy principle set out in INFCIRC/153, such a detection capability measure is understood to be meaningful only when related to

- a) a specific time interval (e.g. between two physical inventories or two interim inspections),
- b) a specific material balance area, and
- c) a specific category of nuclear material.

If data for the statistical analysis of material accountancy procedures are routinely available (operator and inspector measurement error variances and statistical sampling plans for all components of the material balance equation for the MBA and time period of interest), then SPESY is superfluous: detection capabilities can be

formulated rigorously in terms of classical hypothesis test probabilities and these can be calculated objectively and exactly using well known methods /2/.

If, however, such data are not available, SPESY offers a simple and general scheme for performance evaluation. It maintains the basic philosophy of material balance verification while allowing for and giving credit to the application of safeguards measures which are not, or are only partly, amenable to quantification.

SPESY evaluations are hierarchic. At the top level, goal attainment is equated to the degree to which the components of the material balance equation, expressed in their most general terms, have been "verified". The justification for this is simple:

If verification is adequate in some agreed sense, then the goal of being able to make a definitive statement regarding the continuing presence of safeguarded material in the MBA, in the sense of Art. 30, INFCIRC/153, is attained.

At this level of abstraction, all MBAs, whether associated with item or bulk handling facilities, are treated uniformly. The concept of inventory and inventory change verification then provides an interface to a lower level, which is facility specific. This structure (see Fig. 1) is realized in our PROLOG implementation of SPESY using

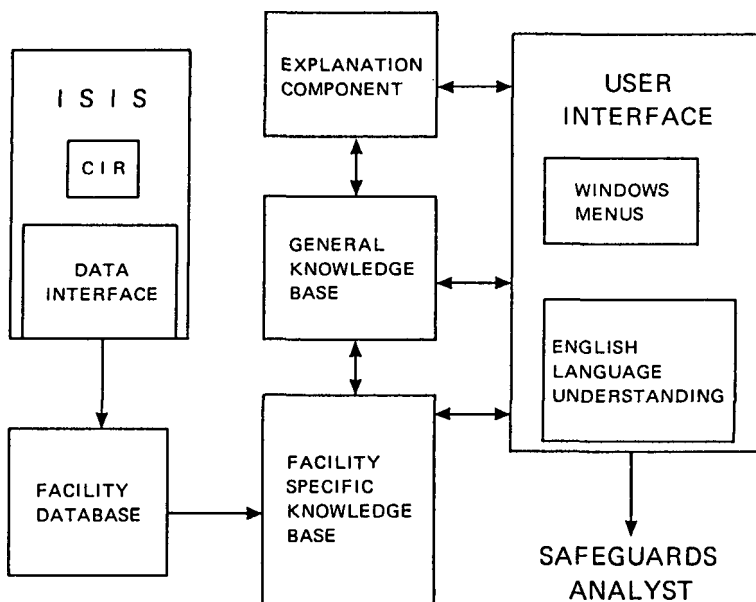


FIG. 1. Safeguards performance evaluation system.

the so-called general and facility specific knowledge bases /1/. The former encodes an overall definition of goal attainment, while the latter defines for a given facility and associated Facility Attachment precisely what is understood as "adequate verification" of the material balance components. Taken together, the modules constitute a specification of the logical structure of the safeguards approach (declarative interpretation of the PROLOG language) and, at the same time, a logic program for evaluating the consistency and completeness of activities carried out within that approach (procedural interpretation of PROLOG).

Reference /3/ gives an extensive description of the techniques used to program LWR performance evaluations within SPESY as well as a discussion of the inspection data acquisition interface used in the trial application.

## 2.2 Departures from Two-Valued Logic

In technical terms, the language PROLOG is a realization of a resolution theorem prover for the first order predicate logic. This means that all propositions are normally two-valued (i.e. true or false), a disadvantage for evaluations where one might like to draw qualified conclusions (e.g. "partial" goal attainment).

SPESY therefore uses a method which deviates from the strict two-valued logic. Conclusions regarding quantity goal attainment are assigned "confidence values" between 0 and 100 which approximate to overall detection probabilities for the diversion of the goal quantity. If we consider for example an item facility, the top level definition of goal attainment in SPESY can be paraphrased as (see also /1/):

The quantity component of the facility inspection goal for material category X over interval T1 to T2 is attained with confidence P if

- the material of category X on inventory at T1 is verified with confidence P1
- and the material of category X on inventory at T2 is verified with confidence P2
- and transfers of material category X across the boundaries of the material balance area over the interval T1 to T2 are verified with confidence P12
- and  $P = \min(P1, P2, P12)$

Minimizing over the confidence values of a conjunction of conditions, as has been done here, is common practice in rule based models for inexact reasoning (see for example /4/), but is often rather difficult to justify rigorously. However, a simple justification for the safeguards situation was suggested to the authors /5/. The argument runs as follows:

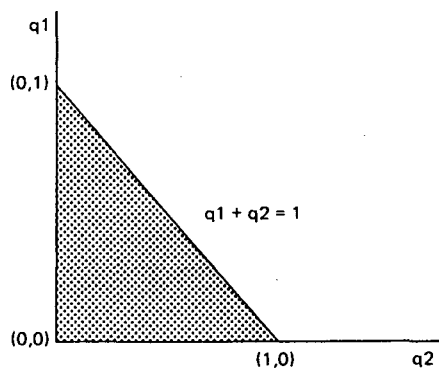


FIG. 2. Permitted domain of the objective function  $P(q_1, q_2)$ .

Let us suppose that the operator of the item facility diverts a goal quantity by falsification of beginning inventory, ending inventory or material transfer data, but not a combination of these. Denote the corresponding (and a priori unknown) probabilities of falsification by  $q_1$ ,  $q_2$  and  $q_{12} = 1 - q_1 - q_2$ , respectively. If the individual confidence values  $P_1$ ,  $P_2$  and  $P_{12}$  can in fact be interpreted as detection probabilities for the diversion/falsification for one goal quantity (this of course depends on how they are defined in the program), then the overall detection probability for the closed material balance is simply

$$P(q_1, q_2) = P_1 q_1 + P_2 q_2 + P_{12}(1 - q_1 - q_2)$$

subject to the boundary conditions

$$\begin{aligned} q_1 &\leq 1 \\ q_2 &\leq 1 \\ q_1 + q_2 &\leq 1 \end{aligned}$$

The guaranteed detection probability (i.e. the detection probability for the diverter's optimal choice of  $q_1$  and  $q_2$ ) is the solution of the problem

$$P^* = \min_{q_1, q_2} P(q_1, q_2)$$

where the objective function  $P$  is confined to the region shown in Fig. 2. Standard linear optimization theory tells us that the optimal solution must occur at a "corner" of this region, i.e. for  $(q_1^*, q_2^*) = (1, 0)$ ,  $(0, 0)$  or  $(0, 1)$ . For example, if  $P_2 < P_1 < P_{12}$ , then  $(q_1^*, q_2^*) = (0, 1)$  and  $P^* = P_2 = \min(P_1, P_2, P_{12})$  as claimed.

### 2.3 Cost Effectiveness, Missing Activities and Explanations

For resource management purposes it can be of interest to associate inspection costs (expressed e.g. in man-days) with effectiveness (i.e. goal attainment) on a material specific or plant specific basis. Furthermore, in the event that planned inspection activities cannot be carried out (with the result that safeguards goals are not attained), an indication of responsibility and/or a qualitative description of associated circumstances at the facility would be desirable both for evaluation and for reporting.

SPESY provides a simple facility dealing with both of these requirements. Associated with each inspection activity predicate /3/ is a 2-member list variable. The first component may be set to the amount of effort expended in performing the activity in question. In concluding goal attainment for a specific material category and time interval, SPESY will sum up all efforts for the activities of relevance. This aggregate effort can have various uses, for instance in making facility to facility efficiency comparisons, establishing a history of effort expended to meet specific goals, examining effort as a function of safeguards significance of material, choosing optimal verification strategies in the event of redundant inspection activities, etc.

If an activity was planned for, but not actually performed, the user can nevertheless enter it into the facility database with the effort variable set to a so-called non-execution code. The alternatives are:

NE1 Operator responsibility

NE2 Force majeure

NE3 Agency responsibility

in order of decreasing significance (from the standpoint of the safeguarding authority). The existence of one or more non-execution codes in the proof chain for goal attainment causes the overall confidence value to be set to 0 and the total effort to be replaced by the most significant non-execution code in the chain. Of course such a clear designation of responsibility will often simply not be possible. The second component of the list variable is therefore reserved as a pointer (possibly nil) to a short descriptive indicating any special problems associated with the activity. All pointers occurring in a particular proof chain are appended together and appear in the answer to the posed query. Taken together, they point to that set of comments in the database which is of relevance to the conclusion reached. These comments are displayed automatically by the system.

The above features provide SPESY with a rudimentary explanation capability. In addition, the user can resort to the "How" and "Whynot" facilities. These allow a detailed exploration of the reasoning followed (or attempted) in answering specific queries.

## 2.4 Are Discrepancies Good or Bad?

One final remark on the evaluation philosophy: as will be apparent in the next section, the existence of one or more significant, unresolved discrepancies recorded in the facility database will prevent the conclusion of goal attainment, i.e. performance was bad. The observation of discrepancies would of course be indicative of GOOD safeguards performance if a diversion had actually taken place. However, it is more reasonable to adopt the point of view that covert diversions are rare occurrences indeed, and that unresolved discrepancies in inspector findings rather reflect deficiencies in the system, whether due to lack of operator cooperation, bad equipment or procedures, insufficient inspector training or whatever. SPESY interprets all discrepancies negatively.

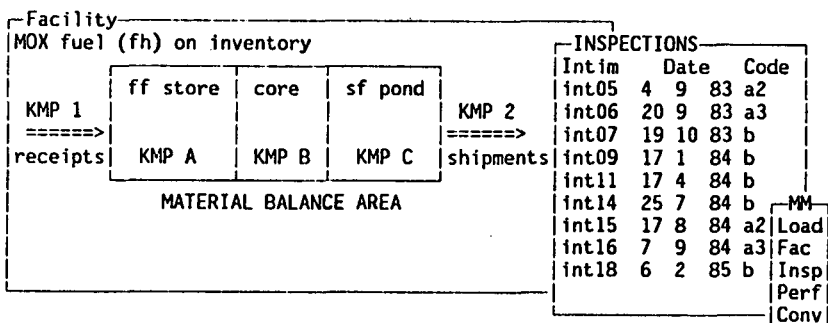
## 3. PERFORMANCE EVALUATIONS

Safeguards performance is understood in SPESY to be synonymous with attainment of the IAEA's internal inspection goals, subject to the constraints of the Facility Attachments. Both the quantity and the timeliness components of the inspection goals are material category specific and we therefore discuss in the following the performance evaluation results for each material category separately.

Since the purpose of the present paper is to demonstrate the methodology itself, we have presented the analyses for the individual facilities in considerable detail so that the reader may form his or her own opinion as to SPESY's merits.

As explained in /3/ SPESY operates within a "window" environment. The relevant windows have been integrated into the text of this section so that a flavor for the actual evaluation sessions may be conveyed. Explanations for the various windows are given as they are encountered. In all, nine reactors were evaluated /3/. For reasons of space, only a small fraction of the actual evaluation is presented here.

## 3.1 Facility R001



The window labeled "Facility" shows the KMP structure of the material balance area and is in fact the same for all 9 reactors evaluated. The "INSPECTIONS" window displays the dates of all routine inspections recorded in the facility database as well as their SPESY internal reference (int--) and a code describing inspection type:

- a1 preparation for a physical inventory verification (PIV)
- a2 actual PIV inspection
- a3 follow-up to PIV, reattachment of seal
- b interim inspection

The evaluation period chosen for this facility was from the first PIV (int05) to the final PIV (int15).

For the category "fresh low" (fl) the following result was obtained:

SUMMARY			EXPLAIN Return Summary How Whynot
OVERVIEW OF GOAL ATTAINMENT			
Quantity goal for fl over (int05 int15)			
*attained confidence 100 code 0			
* fl inv vfd int05	* fl inv vfd int15	* fl flow vfd	
* fl rclds audit	* fl rclds audit	* int05 to int07	
*no disc acc reports	*no disc acc reports	* int07 to int09	
*no disc acc oprec	*no disc acc oprec	* int09 to int11	
*no disc acc inspbil	*no disc acc inspbil	* int11 to int14	
*no disc acc fl	*no disc acc fl	* int14 to int15	
* fl insp at A	* fl insp at A		
* fl accessible	* fl accessible		
No (more) results to explain			



The above "SUMMARY" window presents an overview of the justification for conclusions of goal attainment in the form of an inverted "AND-Tree" with attainment at the root. The "leaves" or terminal nodes of the tree are all marked with an asterisk, indicating that they were successfully proved. As can be seen, the left branch refers to the verification of the beginning physical inventory of fresh fuel at int05. The abbreviations used are:

fl inv vfd: inventory of category fl verified

fl rcds audit: operating records for fl audited

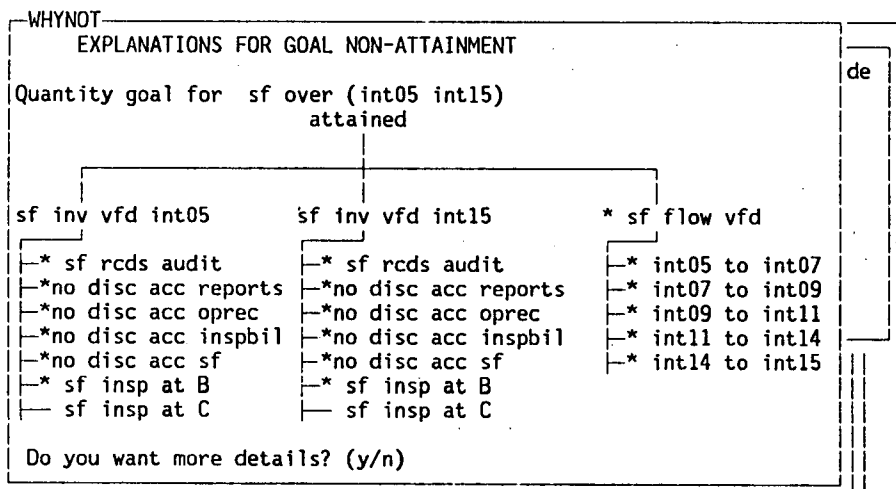
no disc acc reports (oprec, inspbil): no discrepancies between accounting records and reports (operating records, inspector's book inventory listing)

fl insp at A: inventory of fl at KMP A inspected

fl accessible: less than one goal quantity of fl was inaccessible to verification

Note that category fl was present only at KMP A and was inspected. Moreover, the recorded material was accessible to verification. Similarly, the middle branch refers to the final PIV, while the right branch deals with flow verification. In the latter the leaves refer to the consecutive intervals between interim inspections over the whole balance period being queried. The quantity goal is evaluated as attained with 100% confidence.

The query regarding quantity goal attainment for spent fuel did not produce an answer. The "Whynot" command was then issued to explain this failure:



As can be seen, the "WHYNOT" window has the same format as the "SUMMARY" window. However, now one or more leaves could not be proved (missing asterisk), resulting (since this is an "AND Tree") in the overall failure of goal attainment. Evidently a valid inspection at KMP C was not carried out at either PIV.

Answering the prompt at the bottom of the "WHYNOT" window with "y" initiated a trace which led to the following explanation:

Whynot Trace	Insp
(inspected (C sf 88) ----- int05 ----- (?? _8FA6)) fails because	Perf
it cannot be shown that	Conv
identification of sample of _818A items	Expl
of sf at KMP C available at int05 , code _E8B6	Move
more... or nondestructive assay of sample of _DA10 items	Clr
of sf at KMP C available at int05 , code _E44E	Prlg
more...	Data
	DOS
WHYNOT (inspected (C sf 91) ----- int15 -----) ? (y/n)	

The database contained no information as to why neither item identification nor NDA was performed during the PIV inspections.

### 3.2 Facility R003

Performance was evaluated from int04 (first PIV) to int09 (last PIV).

The quantity goal for the spent fuel category was not attained. Here, in addition to lack of adequate inspection at KMP C during both PIVs, problems were diagnosed with flow verification over the period from int08 (an interim inspection) to int09 (final PIV). A request for more details yielded:

Whynot Trace
(verf1 sf (int08 int09) _9DFE (?? _936C)) fails because
it cannot be shown that
sf shipments were verified over (int08 int09) conf _AA58 code _CEA6
more...

The trouble thus originated with the verification of spent fuel shipments. Because of the break in the logical chain caused at this stage, a deeper trace is not possible. It is possible, however, to go into the "Conversation" mode (see /3/) in which SPESY accepts commands and queries posed in normal English. Since sf shipment verification requires proof of continuous successful surveillance, it

was suspected that one of the CHECK database facts might point to the cause:

CONVERSATION

q. will return you to the menu.  
SPESY is listening ...  
Show all comments for the CHECK predicate.

info

CS device sf-camera examined for (int08 int11) , code 0  
expended effort: 0 man-days  
(Inconclusive for KMP B. PIV was on 840812 and canal gate was replaced on 840814 Therefore no change of core inventory can be confirmed.  
Inconclusive for C because 2 partially filled shipping flasks were not verified . (manpower problems)) more..  
(On 840912 and 840919 20 slightly irradiated . EASI were shipped from B to A in part. filled flasks. Due to scheduling and manday problems no verification . Reverification of spent fuel (new night vision device) foreseen during next inspection .)

We see here that a break in the continuity of knowledge for the material in the spent fuel pond did indeed occur. SPESY would have given credit to a reverification of pond inventory to prove flow verification, and the comments indicate that such an activity was planned for a later inspection. Further probing yielded the following dialogue:

CONVERSATION

SPESY is listening ...  
Were spent fuel elements in the spent fuel pond inspected at int13?  
No (more) answers  
SPESY is listening ...  
Were spent fuel elements measured in the pond?  
(0 items of sf at KMP C were assayed at int13 code NE1)  
more?(y/n)y  
No (more) answers  
SPESY is listening ...  
Show all comments for the NDA relation.

info

nondestructive assay of sample of 0 items  
of sf at KMP C available at int13 , code NE1  
not executed, code: NE1  
(Pond was covered by plastic sheet . Therefore it was not possible to verify the spent fuel inventory . Pond must be reverified .) more..

A reverification was not thus possible and it was again postponed, now presumably to a time beyond the horizons of our facility database.

### 3.3 Facility R006

The evaluation period was from int05 (first PIV inspection) to int19 (final PIV).

In this facility the quantity goal for spent fuel was attained. The way in which SPESY proved goal attainment is interesting:

```

HOW
(attained (qg sf) (int05 int19) 99 0)

1  A routine inspection took place on int05
2  A routine inspection took place on int19
3  int05 is earlier than int19
4  Inventory of sf was verified at int05 , conf 99 code 0
5  Inventory of sf was verified at int19 , conf 99 code 0
6  Flow of sf was verified over (int05 int19) , conf 100 code 0
7  Quantity goal for sf is 2 items
8  Operator error in MUF for sf over (int05 int19) is 0 Kg
11
Enter # to trace, q to quit, b to backtrack .

```

First it is noted that the confidence values for beginning and ending verifications are equal (99%). Further tracing established that the initial spent fuel pond inventory of 126 items was counted but not identified or measured. At the final PIV, however, a sample of 119 spent fuel elements was identified. On the basis of continuous surveillance, this information was projected backwards in time to the first PIV and used by SPESY to deduce a detection probability of 99% at time int05.

This trick is obviously not valid for proving timeliness goal attainment (if the beginning inventory in the pond had been falsified that fact would have been detected only at the final PIV). Here SPESY correctly concludes that the timeliness goal for sf was never attained over the queried interval:

```

PERFORMANCE
Timeliness goal attainment for sf
interval    0%      100%
int09 int12 ]      0 no_solution
int12 int15 ]      0 no_solution
int15 int18 ]      0 no_solution
int18 int19 ]      0 no_solution

```

It should be remarked that, had the database contained evidence that a valid spent fuel pond verification had taken place prior to int09, both the timeliness and quantity components of the inspection goals would have been evaluated as attained.

#### 4. CONCLUSIONS

Prior to the availability of the trial data for testing SPESY, it had been necessary to resort to fictive inspection data, more or less defined by the evaluation procedure itself. The LWR trial application has made possible the essential step of tailoring the methodology to real data collected in the field. This step has been invaluable. In the course of the work described in the present paper, and on the basis of demonstrations and discussions with IAEA personnel, a number of advantages as well as problems and possible solutions have been identified. These will be summarized here.

The most serious and obvious problem is that of data acquisition. The present system is makeshift. Although the PROLOG interpreter used is surprisingly powerful and comfortable for a PC implementation, it lacks direct access to the relevant IAEA databases, so that one must resort to the tedious and impractical device of generating computerized inspection reports, typing them into a microcomputer database handling package and, finally, translating the lot into PROLOG clauses for evaluation. The solution clearly would be to interface SPESY directly to ISIS (the IAEA Safeguards Information System) with, for instance, the currently used database query language NATURAL. A mainframe implementation of PROLOG (such as MPROLOG) might then also be considered as an alternative to decentralized evaluations on microcomputers.

For large scale performance assessments, such as those carried out annually for the Safeguards Implementation Report (SIR) to the Board of Governors, inspection data for about 2000 inspections must be processed. A batch processing component would be required for SPESY to perform evaluations on this scale. This poses no serious technical problems, although a PROLOG compiler would probably be needed. Interactive question and answer type sessions could still be advantageous, for example to provide States, on request, with detailed explanations for SIR conclusions.

The SPESY user interface is presently equipped with a simple natural language parser which enables the user to pose his or her queries in ordinary English (discussed in /3/). Since the domain of discourse is quite well defined and simply structured, this feature, at present only rudimentary, could be refined to a very high degree. PROLOG is ideally suited for such a task. This would provide detailed system access for all levels, from inspectors to management, irrespective of computer training or expertise.

The "Whynot" trace facility of SPESY is one of its most important features and at the same time the most difficult to program. The interpreter works its way through the logic program/database using a "depth first" backtracking search algorithm. The "Whynot" trace is a sort of "meta-program" which

tries to pinpoint the critical stage of the search where a failure originated. More effort is needed to perfect this procedure and make it a reliable diagnostic tool.

SPESY can keep track of inspection effort and associate with any derivation of goal attainment the amount of effort expended specific to that goal. At present, inspectors are not asked to estimate the amount of time they spend performing specific verification tasks, so that this feature could not be used in the trial application (the `_e` variable was always set to 0). Although such detailed information may not be readily obtainable, it could prove invaluable for cost effectiveness studies. The effort variable remains a potentially useful component of the methodology.

SPESY has no preconceived notions of minimum detection probabilities necessary for goal attainment. On the contrary, if a detection probability can be calculated legitimately, it is taken as a MEASURE of goal attainment. The interpretation of that measure should not, indeed cannot be divorced from the fuel cycle context in which it was derived and SPESY does not presume to make such an interpretation. This is a point of view which seems to differ strongly from the current IAEA evaluation procedures, but it is also one which, we believe, would be favored by many Member States.

Variants of the LWR specific knowledge base for different Facility Attachments and agreed inspection procedures need to be written and tested with real data. The same is true for radically different safeguards implementations such as for on-load reactors, research reactors, bulk handling facilities, etc. The strength of the SPESY methodology is that it provides a general, standard prescription for creation of all of these evaluation modules.

In the present paper we have only considered using SPESY for a posteriori performance evaluations on a static database of inspection activities and results. It would be relatively straightforward to expand the program into an "expert system" which could be used, for example, to plan inspections in advance /6/. Thus temporary, fictive updates of an existing facility database could be made, and the system queried as to whether the intended activities, if performed, would meet inspection goals. Alternatively, the program could suggest an inspection plan which would meet the current goals and at the same time minimize associated inspection effort. This would be a first step toward fulfillment of the last recommendation of Section 1.

SPESY is clearly still only a prototype, requiring considerable development effort before it could be implemented at Agency Headquarters as a routine tool for safeguards performance assessment. Nevertheless, our experience so far has been encouraging. It has been

established not only that a qualitative, material balance oriented evaluation methodology can be formally specified and then automated (i.e. programmed), but also that it has the potential of being a practical, convenient and, above all, easily understandable tool for routine use. It is obviously in the Agency's interest to have standard evaluation procedures which find the general acceptance of Member States. We feel that an approach like SPESY, which is coupled closely to the principal formal basis for safeguards implementation, INFCIRC/153, stands the best chance of meeting this requirement.

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# ASSESSMENT OF THE EFFECTIVENESS OF SAFEGUARDS

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## Abstract

### ASSESSMENT OF THE EFFECTIVENESS OF SAFEGUARDS.

Assessments of the effectiveness of nuclear material safeguards have so far been mainly verbal and qualitative. Such assessments cannot give any clear indication of the probability that undetected diversion of material occurs, or indicate what limit is placed (or should be placed) upon this risk. Attempts to develop quantitative assessment methods have concentrated attention upon the probability of detection of diversion, should diversion occur. This approach could lead to some false conclusions, because detection probability is not a complete measure of the effectiveness of safeguards. Deterrence of diversion attempts is also important. Attention is drawn in the paper to a range of factors which could influence the risk of diversion, and a mathematical model is proposed to illustrate their possible effects. This model indicates an approach of the type which would have to be taken in order to evaluate and assess the effectiveness of safeguards in explicit, quantitative terms. The general approach is referred to as probabilistic assessment of safeguards effectiveness (PASE). The value which probabilistic assessment has already demonstrated in the field of safety suggests that this may be a fruitful line of development in safeguards work. However, some problems of application are clear. In particular: (i) the effectiveness of safeguards is strongly influenced by human decision processes; (ii) PASE contains factors which can only be subjectively evaluated. PASE appears to be most readily applicable to optimisation studies. For compliance assessments, it should be used initially to supplement rather than replace existing methods.

## 1. INTRODUCTION

A central problem in defining and hence assessing the effectiveness of nuclear material safeguards is the variety of views on their objectives. Differences arise

- . between the points of view of designers, operators and inspectors of plant,
- . between the essential positions of the IAEA and individual nations, and
- . between nations, depending upon whether they are principally suppliers or users of nuclear materials and upon their positions in relation to the development of nuclear weapons.

It is considered, in this paper, that the overall objective of safeguards is to facilitate the peaceful uses of atomic energy by providing reassurance that nuclear materials are not being diverted to improper uses (such as the clandestine manufacture of nuclear weapons). The effectiveness of nuclear material safeguards is considered to be the extent to which this objective is achievable by a design or proposal, or achieved in practice. Assessment of effectiveness is considered to have three components:

- (i) determination of the level of effectiveness;
- (ii) determination of whether resources are allocated to achieve the optimum overall effectiveness;
- (iii) application of criteria for judgement of adequacy.

The different groups mentioned above have different boundary conditions placed upon their views of the overall objective of safeguards because of their various requirements, functions or legal constraints. It can also be viewed in the broader context of non-proliferation and the avoidance of nuclear war. However the objective of safeguards is viewed, the essential means whereby it is achieved are

- . the ability to detect any attempt which is made to divert nuclear material, before the material can be used for a clandestine purpose, and
- . deterrence of such attempts through the risk to the potential diverter of early detection.

In this context, deterrence is achieved wholly because of the risk of timely detection. Therefore detection is fundamental to the effectiveness of safeguards. However, safeguards can be completely effective without total assurance of detection, depending upon other factors which are considered later in this paper. Hence, detection is not synonymous with effectiveness.

Deterrence is also achieved in other ways, e.g. sociologically. It is to be expected that the existence of effective safeguards influences social attitudes but the mechanisms of such feedback are outside the scope of this paper.

## 2. IAEA SAFEGUARDS

The terms of the Agreements, Subsidiary Arrangements and Facility Attachments, which determine the potential effectiveness of IAEA safeguards, are decided by negotiation and compromise between the Agency and Member States. The implementation of these terms is modified by budgetary constraints on the Agency.

The principal method used by the Agency to judge the adequacy of safeguards implementation is the application of criteria stated in its Safeguards Implementation Report (SIR). These are deterministic

requirements relating to the performance of the specified inspection activities. The measure of effectiveness is whether the criteria are not met, partially met or fully met.

Failure to meet the SIR criteria in full would not necessarily mean that diversion occurs. On the other hand, it is recognised that, should diversion occur, the probability of detection might be significantly less than one even if all the criteria are fully met. Hence, the results of these assessments do not indicate specific levels of diversion risk or imply that any particular level of risk is unacceptable.

An alternative method of assessment called SEAM (safeguards effectiveness assessment methodology) has been developed and tested by the IAEA, with the help of consultants from Member States [1, 2]. Essential features of SEAM are as follows:

- (i) As complete a listing as possible of potential diversion paths, assigning to each a classification of its technical complexity level as A (low), B (intermediate) or C (high). The term "diversion path" may include some or all of the following information: nuclear material type and location, physical routes for removal of material and/or introduction of undeclared material, diversion rate and concealment methods.
- (ii) Identification of the anomalies which would be created in each diversion path if diversion occurred in that path. "Examples (of anomalies) include excessive quantities of material unaccounted for, inconsistencies in records, damaged seals....."[3]. Although anomalies may have innocent causes, it appears to be axiomatic in SEAM that diversion would cause at least one anomaly. The probability of detecting an anomaly might, however, be low or even zero if an inspection activity capable of detecting that anomaly is not performed.
- (iii) Estimates of the probability of detection of each anomaly (if it occurs), based on quantitative data, if possible, or subjective judgement.
- (iv) Specification of follow-up actions capable of resolving whether each anomaly has actually been caused by diversion of nuclear material.
- (v) Presentation of results in the form of
  - (a) bar charts showing the probability which detection of anomalies would have in each diversion path, and
  - (b) histograms showing the number of diversion paths in which anomaly detection probabilities would be zero or in ranges from "very low" ( $<0.1$ ) to "very high" ( $>0.9$ ).

Consideration has also been given to the presentation of SEAM results as an aggregate measure. A form of aggregate measure has been proposed, consisting of two factors: an index made up largely of

subjectively determined weighting factors summed over all diversion paths, and the number of diversion paths with zero detection probability. No consensus has been reached on the use of the aggregate measure. Its use is not advocated in this paper but the subjective inputs to its formulation have proved valuable in developing the approach outlined in Section 3.

The intent for SEAM was not to provide an absolute measure of effectiveness, but rather to provide measures which could be used to compare alternatives and to study the effects of varying safeguards inputs. Criteria of acceptability are not stated (except in the case of the aggregate measure).

The only parameter considered in SEAM to characterise the effectiveness of Agency safeguards is the conditional probability of detection of anomalies, should diversion occur. This appears to limit the value of SEAM even for comparative evaluations. Detection probability, by itself, is not a measure that evaluates the effectiveness of safeguards. If it were,

- (i) the existence of even one diversion path with zero detection probability would mean that safeguards were entirely ineffective, which is clearly not so; and
- (ii) the potential existence of diversion strategies which have not been taken into account in planning safeguards measures could invalidate the whole safeguards approach, even though such strategies might be too expensive, complex and/or dangerous to warrant implementation by a potential diverter.

Criteria are therefore needed for ranking the importance of diversion paths on the basis of the likelihood of their use, as well as the probability of detection of anomalies caused by diversion in paths which are used. There are two components to this likelihood. Although these are inter-related, they may be considered separately.

The first of these components is the likelihood that diversion occurs. Diversion path analysis depends on the implicit assumption that diversion may occur. The likelihood of the occurrence depends on decisions by the potential diverter and the extent to which diversion may be deterred (as discussed in Section 1). These are national considerations, essentially outside the role of the Agency, and cannot therefore be included in the Agency's assessments.

The second component is the likelihood that a particular strategy would be adopted, should diversion occur. This depends upon the diversion strategy chosen by the diverter. To the extent that choice is independent of national characteristics, it does not appear to be outside the scope of legitimate consideration. Indeed, the SEAM aggregate measure reflects such considerations, as the value to the Agency of covering particular diversion paths.

### 3. PROBABILISTIC ASSESSMENT OF SAFEGUARDS EFFECTIVENESS (PASE)

#### 3.1 General considerations

Independent characteristics displayed by assessment methods may be described in the following terms:

- . subjective or objective,
- . qualitative or quantitative,
- . deterministic or probabilistic.

These pairs of terms in fact represent ranges of characteristics within which the description of a particular method would lie. Experience with assessments of safety has demonstrated a need for the characteristics of a method to correspond as closely as possible with the second alternative in each of the above cases. This cannot always be fully achieved, and objections have even been raised to doing so because it makes certain problems too explicit, e.g. the uncertainties in the assessments and the limitations of technology. In the technical context, these objections are spurious because the problems are inherent in safety, not created by the assessment method, and are merely obscured by less explicit methods. Objective, quantitative, probabilistic methods are uniquely valuable in identifying problems and directing proper attention to them.

Uncertainties arise through incomplete knowledge, inadequate data on rare events and statistical distribution of the available data. In safeguards assessments, because of the pervasive importance of human actions and the high degree of subjectivity in many of the evaluations, uncertainties are likely to be at least as great as in safety assessments. However, probabilistic assessment does not necessarily have to be accurate to be useful. Probabilistic safety assessment (PSA) has helped to establish the significance of potential safety issues without placing high reliance on numerical estimates. Even the demonstration of overall compliance with safety goals may not require great accuracy. Applications of PSA have spread to cover many (perhaps most) of the important areas of concern in nuclear reactor safety, including on-line assistance to inspection and operations staff. There appears to be no reason, on the grounds of inherent uncertainties, why probabilistic assessment should not also have wide application to nuclear material safeguards.

One of the principal benefits from using the systematic fault tree and event tree logic of PSA [4] is that it provides a thorough method for identifying sets of conditions which constitute significant accident scenarios. Thus the entirely qualitative aspects of PSA provide valuable insights into safety problems which may be more important than the quantitative estimates. Similar methods should be capable of identifying sets of information which describe potential diversion scenarios and possibilities for concealment of anomalies, thus providing a parallel to the "diversion path" approach and a check on the completeness of current methods of assessment.

They may also provide insights into the interrelation between factors relevant to safeguards which are not revealed by other methods.

The problem of technological limitation in safety is that zero risk is not achievable. PSA therefore seeks to answer the questions "how safe is an industry?" and "is it safe enough?" Assessments based on deterministic concepts cannot address this problem. Rules and procedures expressed in deterministic terms are necessary for a safe industry but compliance with them is not (by itself) sufficient to demonstrate any particular level of safety. A similar problem exists in safeguards, where the relevant questions are "how effective are safeguards?" and "are they effective enough?"

### 3.2. An explicit approach to the problem

Although the basic questions in the assessment of safety and safeguards are similar, there is a fundamental difference between the two problems. In PSA, the cause of an accident is usually treated as a random occurrence. In PASE, a parallel assumption cannot be made. An attempted diversion would involve deliberate choice by the diverter and would be anything but a random event. In this respect, it is similar to sabotage, which is not well handled by quantitative PSA.

It has already been observed that detection probability, by itself, is not an adequate criterion for PASE. If it is used, it should be in conjunction with other factors which would influence a decision to attempt diversion and the choice of diversion strategy. Within the context of national diversion for a technical purpose (a nuclear weapons programme) it is assumed that relevant factors would be rational<sup>1</sup>. For example, although potential diverters would see advantage in taking unexpected courses of action, they would be more likely to divert highly enriched uranium (HEU) than natural uranium (NU) if both were equally available. A full list of such factors would include

- (i) the perceived need for diversion of the material;
- (ii) the perceived consequences of being detected;
- (iii) the probability of being detected;
- (iv) the utility of the material;
- (v) the cost, health risk and technical difficulty of diversion; and
- (vi) the relative cost, risks and technical difficulty of obtaining the material (or nuclear weapons) some other way;

and the relative importance which a potential diverter would attach to each factor.

---

<sup>1</sup> An implied assumption is that irrationality is unlikely to lead to significant weapons proliferation, although it might lead to isolated acts of terrorism.

The criteria for decision and choice cannot be known with certainty. However, reasonable assumptions or propositions can be made about them on the basis of objective information and subjective judgements. After all, it is only an assumption (albeit a reasonable assumption) that the probability of detection would be important to a potential diverter.

The following equation is given in order to illustrate a quantitative approach to expressing the dependence of diversion psychology upon these factors. It makes use of subjective factors and concepts developed for SEAM [1] in order to show the effects which the various factors might be considered to have. For example, the value of detection in a given diversion path (k) is assumed proportional to  $[P_k(d/D)]^n$ , where  $P_k(d/D)$  is the conditional probability of detection (d) of diversion given that diversion (D) occurs in path k.

$$F_k(D) = C_1(W_k - C_2)[C_3 - P_k(d/D)]^n \quad (1)$$

in which  $F_k(D)$  is the likelihood of diversion attempts in path k (with units of frequency). Thus,  $F_k(D)$  reflects the attractiveness of path k to the potential diverter. Only positive or zero values of  $(W_k - C_2)$  and  $[C_3 - P_k(d/D)]$  are relevant.

$W_k$  is a weighting factor, with value between 0 and 100, which was developed for SEAM [1]. It reflects the importance of factors (iv) and (v) above. Subjectively determined values of  $W_k$  are given in Table I.

TABLE I. IMPORTANCE WEIGHTS  $W_k$  FOR NUCLEAR MATERIAL TYPE AND TECHNICAL COMPLEXITY LEVELS: SUBJECTIVE JUDGEMENTS ON A SCALE OF 1 TO 100 [1]

Nuclear material type	Technical complexity level		
	A	B	C
	Low	Intermediate	High
Pu, HEU or U-233 as metals	100	58	24
Pu, HEU or U-233 separated from fission products	82	47	15
Pu, HEU or U-233 in spent fuel	45	20	6
LEU, NU, DU, Th	25	12	4

Notes: HEU: high enriched uranium (20% or more U-235).

LEU: low enriched uranium (less than 20% U-235).

NU: natural uranium.

DU: depleted uranium.

$n$  is a subjectively determined index, greater than zero. A value of  $n = 0.3$  was suggested for SEAM [1].

$C_1$  is a parameter, greater than zero, which would depend upon factors (i) and (ii) within a given national context. It has units of frequency.

$C_2$  reflects the balance between alternative proliferation paths on the same scale as  $W_K$ . Its value tends towards 100 for a nation having an overtly thriving nuclear arms industry with adequate, non-safeguarded sources of material supply dedicated to it.  $C_2$  would be greater than 4 (see Table I) for any nation which would find it more attractive to extract natural uranium (NU) from sea water, in secret, rather than engage in diversion strategies at a safeguarded facility.

$C_3$  reflects the maximum risk of detection that a potential diverter would be prepared to incur. A nation which would divert in the face of certain detection ( $C_3 > 1$ ) would presumably not be in the non-proliferation regime.  $C_3$  would be zero for a nation with no intention of diversion but such a nation would wish to receive, and might feel obliged to give, credible reassurance of non-diversion. Therefore, values of  $C_3$  less than 0.5 may not be important.

Equation (1) applies to one diversion path and needs to be related to the whole system. A second fundamental difference between PSA and PASE is relevant here. In PSA, many potential combinations of failures contribute to the total accident risk. In PASE, one diversion path may contribute the total risk, i.e. the one which is most attractive to the potential diverter. Hence, the likelihood of a diversion attempt,  $F(D)$ , is not the sum of values of  $F_K(D)$  for all paths (as it would be by direct analogy with PSA). As a first approximation, it is the maximum likelihood indicated for any one path (path  $K$ ). If several diversion paths are involved, this could be regarded as a representative value.

Assuming that equation (1) is appropriate, the total likelihood of attempted diversion for the whole system is thus given by

$$F(D) = C_1(W_K - C_2)[C_3 - P_K(d/D)]^n \quad (2)$$

in which  $W_K$  and  $P_K(d/D)$  are the values in path  $K$ , where  $F_K(D)$  is maximum. The likelihood of detection of diversion,  $F(d)$ , is given by

$$F(d) = F(D)P_K(d/D) \quad (3)$$

Therefore the likelihood of successful diversion is

$$F(D) - F(d) = C_1(W_K - C_2)[1 - P_K(d/D)][C_3 - P_K(d/D)]^n \quad (4)$$



The formulation of these equations ignores a number of complicating factors, such as the creation of more than one anomaly in a diversion path and the possibility of false alarms. In order to deal with these factors, it is possible that PASE would be better approached from first principles by fault tree/event tree analysis rather than diversion path analysis. However, existing data have already been processed into the diversion path format. It is also possible that decision theory and/or games theory could be usefully incorporated into further development of the approach.

### 3.3. Assessment criteria

In PSA, answers to the question "how safe is safe enough?" can be based upon three concepts [5]:

- (i) A limit beyond which risks are unacceptable under any circumstances. This is the minimum objective of safety.
- (ii) A maximum practical objective, beyond which efforts to improve safety would not be warranted. This is sometimes regarded as the "de minimis" level of risk, below which risks are acceptable or of no concern to normal people.
- (iii) Cost effectiveness optimisation of the system within the range between the minimum and maximum objectives.

In safeguards, it is clear that 100% assurance of detection is not achievable. In fact, a detection probability of 0.7 is regarded as high in SEAM [1]. When deterrence and national intentions are taken into account, it is at least theoretically possible for safeguards to be entirely effective ( $F(D)-F(d) = 0$ ), although uncertainties would be such that this might be impossible to demonstrate. Nevertheless there must be a level of detection probability at which attempts to improve the safeguards system any further would be unwarranted. This may be termed the "maximum objective" of safeguards and might not be reached in practice because of:

- . conflict of safeguards with the process requirements of design and operation;
- . national objections to the intrusiveness of safeguards;
- . constraints of cost and manpower upon the Agency; and
- . limitations of safeguards technology.

However, there must also be a level of effectiveness below which the risk of diversion would, by common consensus, be incompatible with the non-proliferation regime. This therefore represents the limit of acceptability, and may be termed the "minimum objective" of safeguards. In relation to public safety, the question of acceptable risks has been approached in a number of ways, for

TABLE II. SOME VALUES OF FACTOR  $C_d$  USED TO DESCRIBE THE RELATIVE EASE WITH WHICH AN ANOMALY COULD BE EXPLAINED AS HAVING AN INNOCENT CAUSE  
(subjective judgements on a scale of 1 to 10<sup>a</sup>)

Anomaly description	$C_d$
Fuel assembly fails attributes test by non-destructive assay	1
Camera seal tampered with	2
Fuel assembly carries incorrect identification number	3
Item count inconsistent with records	4
Missile shield seal tampered with	5
Missile shield seal broken or damaged	6
Facility records inconsistent with reports	7
Shipper-receiver difference for spent fuel	8
Facility book inventory inconsistent with Agency record	9
Surveillance record incomplete	10

Note: 1: impossible to explain ("caught red handed"); 10: easy to explain (honest mistake).

<sup>a</sup> From discussion with J.A. Powers, IAEA Division of Safeguards Evaluation.

example by studying levels of risk already accepted by society. In relation to diversion of nuclear material, it would appear that the answer must be politically determined.

Maximum and minimum objectives relate to absolute levels of effectiveness. Optimisation involves the balancing of cost against effectiveness and the allocation of resources<sup>2</sup> to the most effective areas for reducing the likelihood of diversion, and therefore requires detailed consideration and intercomparison of individual diversion paths and detection probabilities.

Equation (1) is modified for this purpose as follows. The parameter  $C_1$  may be considered to consist of two factors,  $C'$  and  $C_d$ , so that  $C_1 = C'C_d$ .  $C_d$  (for consequences of detection) is a measure of the relative ease with which an anomaly could be explained as having innocent causes. This would be a significant factor in the deterrence potential of anomaly detection, because a potential diverter would perceive different consequences associated with the detection of different anomalies. Some consequences would be easy to represent as

<sup>2</sup> Relevant resource types would include inspection man-days, research and development, equipment availability, and political resources directed towards the renegotiation of Facility Attachments.

having innocent causes. Others would be tantamount to being "caught red handed" and would be impossible to explain away. This could be expected to influence the choice of diversion strategy.  $C_d$  is assigned values from 1 (for an anomaly which would be impossible to explain as having innocent causes) to 10 (easy to explain; honest mistake). Some subjectively determined values of  $C_d$  are given in Table II.

$C'$  accounts for all the other factors in the perceived need for diversion and consequences of detection, and is considered to be constant in a given national context. Setting  $C' = 1$  therefore changes  $F_k(D)$  to become the relative likelihood of diversion in path  $k$ , designated  $L_k(D)$ . Hence

$$L_k(D) = C_d(W_k - C_2)(C_3 - P_k(d/D))^n \quad (5)$$

#### 4. APPLICATION OF PASE

Clearly there are impediments to the full implementation of probabilistic assessment, including some inherent problems discussed in Sections 4.1 and 4.2. Similar obstacles have been overcome in the introduction of PSA during the past 15-20 years. Nevertheless probabilistic methods should be introduced with care, tested against existing methodologies and used extensively as "in-house" assessment tools before consideration is given to public uses such as reporting performance.

##### 4.1. Subjective judgements

Assignment of values to the parameters  $C_1$ ,  $C_2$ ,  $C_3$  and  $n$  presents problems. There is no record of diversion and hence no possibility of estimating actual frequencies from data obtained under known conditions. Experience (if it existed) would be only indicative, in any case, as different decision makers could react differently to a given set of circumstances. Therefore evaluations can be made only on the basis of subjective judgement. Values of the variables  $W_k$  and  $C_d$  are also subjective and there are likely to be strong elements of judgement in some estimates of  $P_k(d/D)$ . Nevertheless the assignment of values is essential if arguments are to be brought out of the verbal and into the numerical realm.

Some of these evaluations, if they are to be realistic, could involve discriminating between Member States, which the Agency is constrained not to do. In order to avoid this problem, all assessments could be made applicable to the most extreme national scenario which can be envisaged. However, this would be onerous in less extreme scenarios, and States might be asked to accept what seems to them (for their own cases) an unnecessary burden in order to achieve a common good. This might not be generally acceptable to them.

It is difficult to see how any other approach, taking all relevant factors into account, could achieve total objectivity and/or avoid discriminating between States. Putting this another way: an approach which does not have these problems would not provide an

explicit, quantitative basis for assessing the effectiveness of safeguards.

#### 4.2. Uncertainties in assessments of compliance

Uncertainty could be the greatest problem in the application of PASE.  $C_1$  is perhaps the most uncertain parameter, as there is no obvious basis for assigning even a range of values to it.  $C_1$  would be needed for determining absolute values of likelihood, and would therefore be relevant to judging compliance with the maximum and minimum objectives discussed in Section 3.3.

Although, in principle, these objectives must exist, there is no record of their having been evaluated. The highest level of confidence would be needed in demonstrating compliance with any minimum objective (i.e. limit of acceptable diversion risk) which might be specified. Political difficulties of canvassing the fact that successful diversion is a real possibility may have inhibited the development of appropriate criteria, particularly expressed in terms which would make it difficult to demonstrate unequivocal compliance. A similar problem in relation to the risk of accidents has been encountered, and not fully overcome, in the safety field [5].

#### 4.3. Comparisons and optimisation studies

Experience of safety assessment has shown that problems of uncertainty become less significant when relative risks are considered, because errors tend to be similar between options which are being compared. This is also indicated by arguments presented in Section 3.3 that the main uncertainties associated with  $C_1$  can be eliminated from evaluation of the relative likelihood of diversion. Hence, optimisation using equation (5) may be the most readily available application of PASE.

To test this hypothesis, equation (5) is being used to re-analyse some of the 1983 PWR inspection data which have already been analysed using SEAM<sup>3</sup>. The validity of the results of this re-analysis depends upon the validity of the PASE model, which is intended only to illustrate the method. Subject to this limitation the following tentative conclusions are indicated:

- (i) The method is capable of concentrating attention upon only a few diversion paths that warrant concern.
- (ii) The likelihood of diversion appears to be strongly dependent on  $C_2$ .
- (iii) The likelihood of diversion is not highly sensitive to  $C_3$  or  $n$  over their expected ranges of variation.

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<sup>3</sup> Work carried out with the assistance of J.A. Powers and D. Wallace, IAEA Division of Safeguards Evaluation.

## 5. CONCLUSIONS

A realistic quantitative method of effectiveness assessment is needed to assist in ensuring the adequacy of the safeguards system and the optimum allocation of available resources. Methods and goals which are commonly in use for assessing the effectiveness of safeguards are either qualitative or relate to incomplete quantitative definitions of effectiveness, and thus have limitations for this purpose.

It is concluded that a probabilistic approach to assessment is potentially capable of overcoming these limitations. There are practical constraints upon the development and use of such a method. For example:

- (i) Quantitative assessments of effectiveness would contain important elements of subjective judgement and uncertainty. These are inherent in the problem and not created by the assessment method.
- (ii) Some essential parameters in an assessment would depend on social, economic and political conditions and could therefore vary between national scenarios.

Unless these issues are faced, consideration of safeguards effectiveness will either remain in the purely verbal realm or will develop along spurious quantitative lines which may lead to false conclusions. There is no fundamental reason why subjective evaluations should not be used in the way which is envisaged, with the aim of arriving at a structured judgement of effectiveness. However, this approach should supplement rather than replace deterministic methods, while experience of its use is gained.

Relative values of diversion risk and safeguards effectiveness may be estimated with greater confidence than absolute values and should be used to assess the relative importance of various safeguards measures, to identify those areas with the highest potential for reducing diversion risk, and to allocate resources and priorities of work in the optimum cost effective manner. For gaining insights into some of these matters, it is anticipated that the qualitative aspects of analysis may be more important than numerical estimates.

## ACKNOWLEDGEMENT

The author would like to acknowledge the assistance gained from discussions with IAEA staff and consultants on alternative safeguards assessment methods.

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# CONTRIBUTION OF A STATE'S SYSTEM OF ACCOUNTING FOR AND CONTROL OF NUCLEAR MATERIAL TO SAFEGUARDS EFFECTIVENESS

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## Abstract

### CONTRIBUTION OF A STATE'S SYSTEM OF ACCOUNTING FOR AND CONTROL OF NUCLEAR MATERIAL TO SAFEGUARDS EFFECTIVENESS.

The number of nuclear facilities (mainly power plants) under safeguards is increasing; therefore, the burden for both a State's system of accounting for and control of nuclear material (SSAC) and the IAEA is also increasing. In seeking measures for an effective approach one solution seems to be standardization of the accountancy systems at the operator level. When creating standardized systems at nuclear facilities, the SSAC has to give attention to the accommodation of the systems to IAEA requirements. For example, the data presented by the operator during IAEA inspection must fit the IAEA's forms for computerized inspection reports. The compatibility of the SSAC, at operator level as well as State level, with the IAEA requirements is useful for all who participate in safeguarding nuclear material. Computerization can also help considerably to increase safeguards effectiveness. In this respect, the types of computer and software are not so important as the means of data transmission. Magnetic tapes and diskettes are used for reporting to the IAEA but are not yet applied during inspection activities. In principle, inspectors could be provided with all data needed for their inspection reports on diskette instead of in the form of hard copies. With computers these data could be processed rapidly and without error during inspection, and the inspector could concentrate on the verification activities. On the basis of these considerations it is possible to state that the quality of the SSAC has an important influence on safeguards efficiency. A positive role is also played by close co-operation between the SSAC and the IAEA, especially when based on flexible technical approaches.

## 1. INTRODUCTION

Most national nuclear facilities are covered by IAEA safeguards, and a large number belong to an open nuclear fuel cycle. Each new facility increases the IAEA burden; therefore, new approaches must be introduced or safeguards efficiency will deteriorate because of the limitations of IAEA resources. Future IAEA inspection efforts cannot be proportional to the present efforts per facility without sufficient growth of the inspectorate.

One of the new safeguards approaches is fuel cycle oriented safeguards, which could replace existing facility oriented safeguards. However, up to now no one has determined criteria to enable the new ideas to be applied in practice.

Likewise, we are unable to establish those criteria here. However, this paper is not intended to outline new safeguards approaches, but attempts to point out possibilities for increasing safeguards effectiveness for countries with an open fuel cycle.

A typical open fuel cycle constitutes nuclear power reactors, research reactors and laboratories. The cycle excludes large bulk facilities, as well as numerous diversion possibilities. Most of the nuclear material is in the form of discrete fuel assemblies which create a typical batch. Batch integrity is maintained on national territory and therefore batch follow-up procedures are easy to implement. Therefore, a typical open fuel cycle comprises inherent safeguards features, which give us the chance to use them to improve safeguards effectiveness.

## 2. STATE LEVEL OF SSAC

States create their SSAC to fulfil their own legal requirements for nuclear material utilization as well as to enhance IAEA safeguards. It should be stressed that the SSAC must first enable the IAEA to carry out independent verifications as expressed in INFCIRC/153. When concluding the Safeguards Agreement with the IAEA, each State undertook to allow IAEA inspection at its facilities. Of course, it is in a State's interest to shorten and to facilitate these inspections as much as possible so long as the inspection results prove a State's adherence to the Safeguards Agreement. Thus, to fulfil the IAEA requirements is one of the most important goals for the SSAC in its effort to co-operate with the IAEA.

For really effective co-operation with the IAEA, not only does a State need the political will but its SSAC must also have a sufficient technical level to understand the IAEA requirements. An SSAC able to perform independent verification activities can serve as a reliable source of information for the IAEA. Indeed, providing all the information needed by the IAEA in the correct form and in time is one of the main SSAC tasks in supporting the attainment of the IAEA inspection goals. Nevertheless, it is at the IAEA's discretion to use or not to use the SSAC results.

The SSAC obtains the information mainly from its reporting system. The information or data flow is regulated by national accountancy regulations and its timeliness and content shall correspond to the IAEA requirements. Regardless of the type of data processing system (manual or computerized), the SSAC must check incoming data for consistency and content and translate them into the form required by the IAEA. However, it is not possible to check all the information directly from the SSAC headquarters. Not only the IAEA but also the State authority must carry out inspection at the facilities. Apart from tasks corresponding to specific State requirements the SSAC inspection activities should concentrate on the preparedness of the facility to fulfil IAEA requirements.



Thus, the SSAC receives sufficient information on the operator's accountancy system, its quality and its compatibility with IAEA requirements.

The IAEA inspection effort is given by the pertinent Facility Attachments, which limit this effort in scope and time. The same limitations are not acceptable for the SSAC's inspection activities. The SSAC must go into more detail and cannot be limited by the number or duration of inspections. Therefore, the SSAC usually carries out more inspections than the IAEA and has more detailed access to the operator's records and reports system and to the material itself. However, the SSAC also seeks ways of obtaining objective information with the minimum effort, and of optimizing the paperwork to obtain more time for verification activities at material locations.

The inspection effort, whether that of the IAEA or the State, is usually 30–40% for paperwork, about 30% for material verification, and the rest for containment and surveillance measures. Exact figures vary with facility and inspection type and depend, of course, on the problems faced during inspections. One error in the operating record, e.g. one wrong tag on the batch during the physical inventory, can double the time for a given activity, which means that it becomes necessary to shorten other inspection activities.

In analysing reasons for anomalies and why inspection goals are not reached, we find that the main cause is operator error. Such errors are part of each accounting system and can be reduced to a minimum, but they are difficult to prevent completely. Therefore, the SSAC plays a very important role in creating a buffer between the operator system and the IAEA requirements. Many problems can be solved at the State level, so that IAEA inspections can be carried out with a minimum of problems. The SSAC must teach the operators how to understand the IAEA requirements, and explain how to present the information in order to facilitate the inspector's task.

The safeguards system checks that significant amounts of nuclear material are not missing. For this it is not necessary to check the nuclear material to the last gram. In reality both NDA and DA verification methods are limited in their precision and therefore the control body must accept certain differences caused by measurement quality. On the other hand, the accountancy is very precise. Each gram of nuclear material can be accounted for, which means that the precision of the accountancy is always higher than that of the measurement techniques. The question arises — can the control body accept fast and less precise accountancy checks without compromising the reliability of its conclusions? It can if the precision of the accountancy check is not lower than the precision of the material verification. To lose time seeking one missing gram in the records makes no sense if the measurement method gives results with a hundred-gram difference.

To support these views we now present the results of one experiment carried out during an inspection at one of our PWRs (WWER-440 type). In 1984 we calculated the average  $^{235}\text{U}$  weight for the group of fuel assemblies with 3.6%

TABLE I. COMPARISON OF OPERATOR'S AND INSPECTOR'S DATA

Element	Number of fuel assemblies	Operator's data (g)	Inspector's data (g)
U-235, fresh fuel	422	1 819 627.6	1 819 495.2 ±10 296.8
Pu, spent fuel	204	233 859.52	232 784.4 ±7 915.2

enrichment ( $4311.6 \pm 24.4$  g), and for the same fuel assembly type we established the average plutonium weight after three years ( $1141.1 \pm 38.8$  g). These figures were applied for checking the physical inventory in 1986. The operator's precise figures were compared with the figures calculated from the average weight and number of fuel assemblies counted by inspectors (Table I).

The differences of 132.4 g for  $^{235}\text{U}$  (fresh fuel) and 1075.12 g for plutonium (spent fuel) are low compared with the inspection goals or with significant quantities. They confirm that PWR operation is very stable and represent one of the inherent safeguards features of this reactor type. This example is not meant to introduce simplified operator accountancy. It is meant to show that the SSAC can find and offer the IAEA fast inspection methods even with paper-work.

Of course, in the area of material verification more possibilities exist for the SSAC to present an efficient inspection method to allow independent IAEA verification. It is convenient if the inspection methods are based on the facility operation, because then they do not introduce new elements into the operation or disturb the operators. If the inspection method depends directly on the main facility activities then it is more difficult for the operator to find the diversion paths which could be concealed from the control bodies. At PWRs these operation dependent inspection methods could be based on power range detector readings, total thermal power calculations, standardized methods for burnup and isotopic concentration calculations, loading machine log-books, etc. To realize the ideas mentioned above it is necessary to combine the active technical SSAC approach and a flexible operation oriented IAEA approach.

### 3. SSAC FACILITY LEVEL

During their inspections at facilities IAEA inspectors experience the SSAC mainly at the facility level. Therefore, it is necessary to ensure that also at this level all IAEA requirements will be fulfilled. The basic measures for creating a

facility accountancy system compatible with the IAEA system are well known and will not be repeated here. However, experience shows that another very important feature which can influence the safeguards effectiveness exists, i.e. the complexity of the facility accountancy system.

A State with an open fuel cycle usually has two kinds of facility. Some are facilities with a limited nuclear material throughput and inventory, for which simple and logical accountancy systems are a basic condition for effective safeguards. Then, there are the nuclear power plants (NPPs), which need a complex accountancy system. The approach that can be applied to this group of facilities is a standardization of the facility accountancy system. National facilities are inspected by a limited group of IAEA inspectors. If they are faced with only a few types of facility system, or with systems similar to each other, they have no problems in having to adapt to new forms, log-books, cards, etc. It is very important for the SSAC to establish standardized accountancy systems at facilities, including at different types of facility. Of course, it is not possible to create an identical system for a PWR and a bulk facility.

However, as well as the inspection activities and the inspection working papers (e.g. computerized inspection reports (CIRs)) being standardized, it is possible to standardize forms for accountancy data presentation. The operator can and should arrange the data in such a way that the inspector may have easy access to the information needed to attain the inspection goal. This is feasible mainly in those facilities where computerized accountancy systems have been introduced. If the operator uses the computer for process management it is only a question of suitable software to apply the computerized system for nuclear material accountancy. In Czechoslovakia identical software is being used for computerized accountancy systems at all NPPs. This means that at all NPPs the inspector obtains exactly the same printouts with an identical set of information. These printouts are tailored to IAEA requirements and contain data already prepared for CIRs.

The amount of information transmitted to the inspector during inspection is high, especially at large facilities such as an NPP or bulk facility. To collect all the necessary data is time consuming. However, the inspector does not only collect the data, but also has to compare them with the data already transmitted to the IAEA and to verify if they are self-consistent and consistent with facts. This task demands high precision and skill. It can also be a source of errors which are difficult to solve later at IAEA Headquarters. Therefore, it is necessary to facilitate this part of the inspector's task by applying new techniques.

In addition to report transmission, it may be possible to use diskettes for data transmission. This method would exclude errors caused by data transcription — it is fast and the amount of paperwork would be minimized. In addition, the inspector can bring a pertinent set of the data from IAEA Headquarters, enabling him to carry out fast, precise comparison of the information gained during the inspection with the data sent to the IAEA earlier in the form of

reports. Of course, to do this job he must have the relevant tools. Budgetary questions now arise — how many personal computers are needed and what is the cost?

Introduction of the proposals mentioned should not cost a great deal. The basic condition for the use of new data transmission techniques during inspection would be the operator's application of a computerized accountancy system. Then the operator's hardware system could be applied using the IAEA software. Thus, in most cases the question of the relevant tool would be limited to suitable software. Then cost limitation would be focused on the possibility of standardizing software.

Not all facilities have a computerized accountancy system: for facilities with low throughput and inventory it is not really necessary to introduce a computerized accountancy system. It could be more difficult to feed the computer with data and to keep the system running than to apply a manual accountancy system. But here also the SSAC plays an important role by creating simple and logical accountancy systems. A further feature could positively influence safeguards effectiveness. In many cases the IAEA still lacks information on the operator's measurement uncertainties needed to determine the error limits associated with the material unaccounted for (MUF). This is very important, particularly for large bulk facilities. The SSAC can also play a very important role here by encouraging the operators to solve this problem.

#### 4. CONCLUSION

Nuclear energy utilization is now industrial in character. To maintain the efficiency of the safeguards it is necessary to keep track of rapid nuclear technology development. To do this it is necessary to establish closer co-operation between the SSAC and the IAEA and to base this co-operation on a flexible technical approach. All participants in safeguards should support the introduction of those approaches that reflect the real technical situation and cover practicable diversion paths in a given facility. In this area it is not possible to substitute the role of the SSAC, which can help the IAEA to understand the detailed features of each facility and offer fast and simple inspection methods. It can also establish standardized accountancy systems at the facilities and present its own inspection results in order to increase safeguards efficiency.

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