Gamma, X-Ray and Neutron Techniques for the Coal Industry

PROCEEDINGS OF AN ADVISORY GROUP MEETING
VIENNA, 4–7 DECEMBER 1984

INTERNATIONAL ATOMIC ENERGY AGENCY, VIENNA, 1986
The cover picture shows, at right-centre, the 300 MW(e) high-temperature demonstration reactor, Hamm-Uentrop, Federal Republic of Germany, and at the left a coal plant built alongside. By courtesy of Hochtemperatur-Reaktorbau, Mannheim.
GAMMA, X-RAY
AND NEUTRON TECHNIQUES
FOR THE COAL INDUSTRY
The following States are Members of the International Atomic Energy Agency:

<table>
<thead>
<tr>
<th>Afghanistan</th>
<th>Guatemala</th>
<th>Paraguay</th>
</tr>
</thead>
<tbody>
<tr>
<td>Albania</td>
<td>Haiti</td>
<td>Peru</td>
</tr>
<tr>
<td>Algeria</td>
<td>Holy See</td>
<td>Philippines</td>
</tr>
<tr>
<td>Argentina</td>
<td>Hungary</td>
<td>Poland</td>
</tr>
<tr>
<td>Australia</td>
<td>Iceland</td>
<td>Portugal</td>
</tr>
<tr>
<td>Austria</td>
<td>India</td>
<td>Qatar</td>
</tr>
<tr>
<td>Bangladesh</td>
<td>Indonesia</td>
<td>Romania</td>
</tr>
<tr>
<td>Belgium</td>
<td>Iran, Islamic Republic of Iraq</td>
<td>Saudi Arabia</td>
</tr>
<tr>
<td>Bolivia</td>
<td>Ireland</td>
<td>Senegal</td>
</tr>
<tr>
<td>Brazil</td>
<td>Israel</td>
<td>Sierra Leone</td>
</tr>
<tr>
<td>Bulgaria</td>
<td>Italy</td>
<td>Singapore</td>
</tr>
<tr>
<td>Burma</td>
<td>Jamaica</td>
<td>South Africa</td>
</tr>
<tr>
<td>Byelorussian Socialist Republic</td>
<td>Japan</td>
<td>Spain</td>
</tr>
<tr>
<td>Cameroon</td>
<td>Jordan</td>
<td>Sri Lanka</td>
</tr>
<tr>
<td>Canada</td>
<td>Kenya</td>
<td>Sudan</td>
</tr>
<tr>
<td>Chile</td>
<td>Korea, Republic of Kuwait</td>
<td>Sweden</td>
</tr>
<tr>
<td>China</td>
<td>Lebanon</td>
<td>Switzerland</td>
</tr>
<tr>
<td>Colombia</td>
<td>Liberia</td>
<td>Syrian Arab Republic</td>
</tr>
<tr>
<td>Costa Rica</td>
<td>Libya Arab Jamahiriya</td>
<td>Thailand</td>
</tr>
<tr>
<td>Cote d'ivoire</td>
<td>Liechtenstein</td>
<td>Tunisia</td>
</tr>
<tr>
<td>Cuba</td>
<td>Luxembourg</td>
<td>Turkey</td>
</tr>
<tr>
<td>Cyprus</td>
<td>Madagascar</td>
<td>Uganda</td>
</tr>
<tr>
<td>Czechoslovakia</td>
<td>Malaysia</td>
<td>Ukrainian Soviet Socialist Republic</td>
</tr>
<tr>
<td>Democratic Kampuchea</td>
<td>Mauritius</td>
<td>United Arab Emirates</td>
</tr>
<tr>
<td>Democratic People's Republic of Korea</td>
<td>Mexico</td>
<td>United Kingdom of Great Britain and Northern Ireland</td>
</tr>
<tr>
<td>Denmark</td>
<td>Monaco</td>
<td>United Republic of Tanzania</td>
</tr>
<tr>
<td>Dominican Republic</td>
<td>Mongolia</td>
<td>United States of America</td>
</tr>
<tr>
<td>Ecuador</td>
<td>Morocco</td>
<td>Uruguay</td>
</tr>
<tr>
<td>Egypt</td>
<td>Namibia</td>
<td>Venezuela</td>
</tr>
<tr>
<td>El Salvador</td>
<td>Netherlands</td>
<td>Viet Nam</td>
</tr>
<tr>
<td>Ethiopia</td>
<td>Nicaragua</td>
<td>Yugoslavia</td>
</tr>
<tr>
<td>Finland</td>
<td>Niger</td>
<td>Zaire</td>
</tr>
<tr>
<td>France</td>
<td>Nigeria</td>
<td>Zambia</td>
</tr>
<tr>
<td>Gabon</td>
<td>Norway</td>
<td></td>
</tr>
<tr>
<td>Germany, Federal Republic of</td>
<td>Pakistan</td>
<td></td>
</tr>
<tr>
<td>Ghana</td>
<td>Panama</td>
<td></td>
</tr>
<tr>
<td>Greece</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

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

© IAEA, 1986

Permission to reproduce or translate the information contained in this publication may be obtained by writing to the International Atomic Energy Agency, Wagramerstrasse 5, P.O. Box 100, A-1400 Vienna, Austria.

Printed by the IAEA in Austria
March 1986
GAMMA, X-RAY AND NEUTRON TECHNIQUES FOR THE COAL INDUSTRY

PROCEEDINGS OF AN ADVISORY GROUP MEETING ON GAMMA, X-RAY AND NEUTRON TECHNIQUES FOR THE COAL INDUSTRY
ORGANIZED BY THE INTERNATIONAL ATOMIC ENERGY AGENCY AND HELD IN VIENNA, 4–7 DECEMBER 1984

INTERNATIONAL ATOMIC ENERGY AGENCY
VIENNA, 1986
FOREWORD

Nuclear analytical techniques have great potential to improve the efficiency of raw materials exploration, extraction and processing, with savings in energy and materials, and could therefore contribute to national economies and development programmes. Their advantages are that they are rapid, often specific or multi-element, simple to use, and, for some applications, can be used in places where no other techniques can, for example in hot, dusty or aggressive environments or where control measurements must be made through the walls of vessels. Also, because they can sample all or most of a process stream they can give more representative results than analyses based on individual samples. Most importantly, results can be obtained in near real time, thus enabling the measurements to be used for on-line process control applications.

Recent years have seen rapid developments, and some commercialization, of these techniques applied to the coal industry. Potential applications include on-line process measurement of coal ash, moisture, calorific value, sulphur and elemental analyses. In addition, improved nuclear borehole logging instruments make possible the in situ determination of ash content and coal seam delineation for resource assessment and for mine development planning.

In December 1984 the IAEA organized an Advisory Group Meeting to review the latest developments in this area and to make recommendations on the most promising areas and priorities for future research and development activities. Participants represented a wide cross-section of the coal industry from researchers, coal producers, coal users and instrument manufacturers. Considerable enthusiasm was shown for the value of those nuclear techniques which are already well developed and available for use in all stages of the coal industry, and also for the even greater potential of some of the newer nuclear techniques under development. This report records the proceedings of that Advisory Group Meeting. It will be of interest to all with responsibilities for coal quality control in the exploration, mining, processing and use of coal, including geophysicists, mining engineers, quality control chemists, metallurgists and power station operations engineers as well as to researchers in this field.

The Agency wishes to record appreciation for the suggestions of J.S. Watt and C.G. Clayton which initiated this first meeting on this topic and greatly helped in its organization. Dr. Clayton also made a significant contribution to the success of the meeting by his able chairmanship. P. Bull and I.F. Vovk were responsible for co-ordinating and compiling this work.
EDITORIAL NOTE

The papers and discussions have been edited by the editorial staff of the International Atomic Energy Agency to the extent considered necessary for the reader's assistance. The views expressed and the general style adopted remain, however, the responsibility of the named authors or participants. In addition, the views are not necessarily those of the governments of the nominating Member States or of the nominating organizations.

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

The mention of specific companies or of their products or brand names does not imply any endorsement or recommendation on the part of the IAEA.

Authors are themselves responsible for obtaining the necessary permission to reproduce copyright material from other sources.
CONTENTS

Current developments and applications of nuclear techniques in the coal industry ................................................................. 1  
C.G. Clayton, C.F. Coleman

Discussion ......................................................................................................................................................................................... 22

Borehole elemental analysis of coal using fast neutrons .............................................................. 25
J.L. Mikesell, F.E. Senftle, A.B. Tanner

Discussion ......................................................................................................................................................................................... 41

In situ borehole determination of ash content of coal using gamma-gamma and neutron-gamma techniques .............................................. 43
J. Charbucinski, M. Borsaru, P.L. Eisler, S.F. Youl

Discussion ......................................................................................................................................................................................... 64

Current developments in nuclear techniques for coal logging in Canada ......................................................................................... 67
P.G. Killeen, C.J. Mwenifumbo

Discussion ......................................................................................................................................................................................... 77

Development and application of nuclear techniques for on-line coal analysis in India ........................................................................... 81
A.S. Prasad

Discussion ......................................................................................................................................................................................... 101

Nuclear techniques for coal quality assessment in the United Kingdom coal industry ........................................................................... 103
D. Page

Discussion ......................................................................................................................................................................................... 110

Determination of ash content of coal on-line on conveyors and in-stream in coal slurries ....................................................................... 113
J.S. Watt

Discussion ......................................................................................................................................................................................... 128

Determination of ash, moisture and specific energy of coal .......................................................................................................... 131
B.D. Sowerby

Discussion ......................................................................................................................................................................................... 147

Experiences with coal quality monitoring systems in Poland ........................................................................................................ 149
S. Cierpisz

Discussion ......................................................................................................................................................................................... 159

Physics of recent applications of PGNAA for on-line analysis of bulk minerals (abstract only) ........................................................................... 161
T. Gozani

Discussion ......................................................................................................................................................................................... 162
<table>
<thead>
<tr>
<th>Experience with nuclear instrumentation techniques in the preparation of hard coal</th>
<th>165</th>
</tr>
</thead>
<tbody>
<tr>
<td>G. Fauth, D. Leininger, H. Lüdke</td>
<td></td>
</tr>
<tr>
<td>Discussion</td>
<td>178</td>
</tr>
<tr>
<td>Nuclear techniques for the on-line analysis of coal in electricity generating stations</td>
<td>181</td>
</tr>
<tr>
<td>P.L. Surman</td>
<td></td>
</tr>
<tr>
<td>Discussion</td>
<td>189</td>
</tr>
<tr>
<td>Correlation methods in calorific value measurements of coal</td>
<td>191</td>
</tr>
<tr>
<td>T. Cywicka-Jakiel, J. Łoskiewicz</td>
<td></td>
</tr>
<tr>
<td>Discussion</td>
<td>202</td>
</tr>
<tr>
<td>Summary and Conclusions</td>
<td>205</td>
</tr>
<tr>
<td>List of Participants</td>
<td>215</td>
</tr>
</tbody>
</table>
CURRENT DEVELOPMENTS AND APPLICATIONS OF NUCLEAR TECHNIQUES IN THE COAL INDUSTRY

C.G. CLAYTON, C.F. COLEMAN
Nuclear Geophysics Group,
Nuclear Physics Division,
Atomic Energy Research Establishment,
Harwell, Didcot, Oxfordshire,
United Kingdom

Abstract

CURRENT DEVELOPMENTS AND APPLICATIONS OF NUCLEAR TECHNIQUES IN THE COAL INDUSTRY.

The natural heterogeneity of coal conflicts with the need for a homogeneous product, which is an intrinsic requirement if coal-burning furnaces are to operate under optimum design conditions. In consequence, increasing effort is now being given to monitoring coal quality in the search, production, preparation, transport and combustion sequence. Equipment based on nuclear techniques is becoming an important part of monitoring and control procedures. A number of nuclear techniques are well established for coal analysis and others are being developed. In particular, equipment based on neutron interactions and capable of giving a near-total elemental analysis is now being installed on-line. The paper includes a short review of existing techniques and identifies some approaches to coal analysis which have been developed recently. The advantages and limitations of neutron techniques for total elemental analysis of coal are briefly reviewed.

1. INTRODUCTION

Coal is one of the most important sources of energy in the world. Coal resources are a significant factor in the economy of many industrialized countries and in recent years substantial reserves have been discovered in a number of developing countries which should eventually contribute to their economic evolution. Related to the geographical spread of coal deposits is a variation in their geological origin, in the local climatic and topographical conditions which prevailed at their formation and in subsequent metamorphism. Thus, coals in different regions show variations in their combustion properties, in the concentrations of associated minerals and in the type and thickness of adjacent strata, some of which may be so thin as to be spatially inseparable during mining operations.

The intrinsic heterogeneity of coal deposits and the variation in the quality of mined coal conflict strongly with the current economic emphasis on highly efficient coal-burning furnaces which demand a homogeneous input. As a consequence, coal preparation (washing and blending) is receiving increased attention
and this implies continuous monitoring of coal quality. Quality monitoring is now also being introduced into the transport and combustion phases of coal handling and utilization.

The essential characteristics of nuclear techniques, based on X-ray, γ-ray and neutron interactions, encompass many of the requirements for monitoring the composition of coal. The range and elemental specificity of the techniques largely overcome variations in spatial inhomogeneity and are reasonably compatible with operational conditions, so that analyses of acceptable accuracy can be derived in many of the situations encountered in practice.

Although the control of coal quality is now beginning to be an important consideration for coal suppliers and users, the degree of control required, and hence the specification of the analytical equipment, may vary significantly from one application to another. For example, it will sometimes be sufficient merely to indicate the arrival at a washery of an occasional batch of coal of high mineral content in what is an otherwise acceptable product. If the region of high mineral content can be recognized and removed, significant simplification in the coal preparation procedures can be introduced. For this purpose relatively simple equipment is required. On the other hand, in the operation of some coal combustion plants, it is important to exercise a high degree of control on the concentrations of slagging elements (Al, Ca, Si, Fe) at the input to the furnace. This requires rapid, on-line analytical techniques and at present the achievement of an acceptable solution is not a simple matter, especially if the coal is supplied from several different sources.

The use of nuclear techniques for coal analysis now has a history of more than 30 years, during which time great advances have been made. From early attempts to indicate the mineral content of dried, crushed coals in the laboratory by β-backscattering techniques, equipment is now being introduced on-line which is based on neutron interactions and which can provide a near-total elemental analysis from which operational parameters, such as ash content and specific energy, can be derived.

This is not to say that further development is not required. There are still a number of important and urgent problems to be resolved before the full potential of nuclear techniques can be utilized and before some important operational requirements can be satisfied, particularly those on which complete analytical data are required very rapidly.

The full use of available information from primary and secondary neutron interactions is presently limited by the inability of single, high-resolution Ge detectors to operate for long periods in an intense fast neutron flux without deterioration in energy resolution. Moreover, the analysis of complex spectra is handicapped by overlapping peaks and by accumulating low energy background counts to a degree which may result in high statistical uncertainties in the measurement of peak intensities in certain energy regions. Apart from the need for an improvement in intrinsic detector performance, current effort in the application of multi-
detector assemblies, such as Compton-suppression systems and pair-spectrometers, holds out the promise of some improvement in the future, although it is not yet clear whether the rate of information accumulation will be sufficient for the most demanding operational requirements.

This brief review is not intended to give a comprehensive account of techniques which have previously been considered for coal analysis, but rather seeks to indicate those which are of current interest, along with their possible areas of application and attempts to identify some of the problems which, when resolved, will intensify the application of nuclear techniques to coal analysis in the future.

2. TECHNIQUES AND APPLICATIONS

2.1. Use of high-, medium- and low-energy gamma-ray techniques

Several techniques based on different $\gamma$-ray interactions have been developed for a variety of applications. The most important of these are summarized below.

2.1.1. Measurement of bulk density

Variations in mineral content reflect directly in the bulk density of coal, provided the composition is reasonably constant. Thus the measurement of bulk density is an important requirement in borehole logging and especially in deposits which are highly stratified and interspersed with multiple bands of mineralization. It is also important in several on-line techniques for generating corrections factors to the principal measurement.

The method is based on the use of a radioisotope $\gamma$-ray source, such as $^{137}\text{Cs}$ (662 keV) or $^{60}\text{Co}$ (1173 keV and 1332 keV), and a scintillation detector, the two being separated by a heavy metal shield so as to restrict $\gamma$-rays from the source reaching the detector directly. The output from the scintillation detector is normally arranged to be above some energy threshold (approximately 100 to 300 keV) which is chosen to ensure that only Compton-scattered $\gamma$-rays are recorded.

Determination of the bulk density $\rho_b$ of the coal is based on the relationship

$$\rho_e = \rho_b N_A \sum_i w_i \left( \frac{Z_i}{A_i} \right)$$  \hspace{1cm} (1)

where $\rho_e$ is the electron density, $N_A$ is Avogadro's number and $w_i$ is the weight fraction of the $i$th element of mass number $A$ and atomic number $Z$. Since Compton processes involve $\gamma$-ray/electron interactions only, then, if the detector
responds only to Compton-scattered \( \gamma \)-rays, the output from the detector is directly related to electron density and \( \rho_b \) can be derived provided

\[
\sum_i w_i \left( \frac{Z_i}{A_i} \right)
\]

is constant.

Significant natural variations in \( (Z/A) \) occur in coal from variations in the hydrogen content. Anthracite typically contains about 2 wt\% hydrogen while in some brown coals hydrogen concentrations may approach about 12 wt\%. In a specific type of deposit, such as a bituminous coal, variations in hydrogen content are largest when the coal seams are interrupted by mineralized strata. Intermediate values of hydrogen content occur when banding is severe and the band thickness is small (e.g. 1 mm), the limits of the range being associated with the hydrogen contents of the combustible material and the mineral matter. Mineralization of this type imposes a limit on the \( (\gamma-\gamma) \) method which appears at present to be outside any simple calibration procedure.

2.1.2. Ash-in-coal

A suitable model for considering the mode of interaction of low-energy \( \gamma \)-rays \( (E_\gamma < 40 \text{ keV approximately}) \) is to consider coal as a two-component material comprising the combustible elements with mean atomic number \( Z \approx 6 \) and the mineral component with \( Z \approx 12 \); apart from the iron content, for which \( Z = 26 \). By using X-rays from \( ^{238}\text{Pu} \) with an energy of 14–17 keV, it has been shown [1] that a high sensitivity to the mineral content is achieved and, with a suitable thickness of aluminium filter over the detector window, or with detector energy discrimination, compensation for variations in the iron content can be obtained. A number of instruments based on this method are now in use, worldwide.

The major disadvantages of the method are that, for high accuracy, the largest coal particle size must be less than about 80 mm and that, since crushing is required, only a small proportion of a mine output can be sampled. In addition, although compensation for moisture variations can be included in the techniques, it has been found that coals with a high moisture content (> 15\% approximately) cannot be easily accommodated in the sample presentation equipment.

2.1.3. Ash-in-coal in a tailings stream

Recently increased awareness of the need to conserve energy has focused attention on the fact that on-line measurements of the ash content of coal in a tailings stream provide a means of controlling the operational efficiency of a
coal washing plant. An X-ray backscatter technique which has been developed for this purpose [2] is now operating successfully on-line.

For this application the coal can be regarded as a three-component system; the combustible elements ($Z \approx 6$), the mineral elements ($Z \approx 12$) and water.

If suffixes c, a and w refer to coal, ash and water respectively, r is the weight fraction of ash in the coal and S is the weight fraction of solids in the slurry, the saturation backscatter intensity $n$ is given by

$$n = k \frac{\sigma_a r s + \sigma_c (1 - r) s + \sigma_w (1 - s)}{\mu_a r s + \mu_c (1 - r) s + \mu_w (1 - s)}$$  \hspace{1cm} (2)

The sensitivity $S$ of the measurement is defined as

$$S = \frac{\partial n/n}{\partial r/r}$$  \hspace{1cm} (3)

By differentiating Eq.(2) and substituting in Eq.(3), a value for $S(r,s)$ is obtained.

Following the results of a series of sensitivity calculations for various exciting radiation energies, and taking account of the conflicting requirement for a relatively high energy to penetrate the container wall and a low energy to give a high value for $S$, a $^{109}$Cd source (22 keV) is preferred.

If the solids density in the slurry is known, the ash content in the slurry can be converted into ash content of the solids. The density can be determined by using a radioisotope transmission gauge based on a $^{137}$Cs source (660 keV).

In the theoretical model two components are considered, coal and water, and the transmitted $\gamma$-ray intensity $Z$ is given by

$$Z = Z_0 \exp \left( -\mu_c \rho_c x_c + \mu_w x_w \right)$$  \hspace{1cm} (4)

for $x_c, x_w$ being the respective path lengths in coal and water and $\mu_c, \mu_w$ the corresponding mass attenuation coefficients. The density of the coal is $\rho_c$ and the density of the water is taken as unity.

Assuming that the weight fraction of the solids is proportional to their respective path lengths and putting $x = x_c + x_w$, Eq.(4) becomes

$$Z = Z_0 \exp \left[ -\frac{x \rho_c}{\rho_c (1 - s) + s} \left( s \cdot \mu_c + (1 - s) \mu_w \right) \right]$$  \hspace{1cm} (5)

With the further assumption that $\mu_c = \mu_w = \mu$, Eq.(5) simplifies to

$$Z = Z_0 \exp \left[ -\frac{\mu x \rho_c}{\rho_c (1 - s) + s} \right]$$  \hspace{1cm} (6)
A possible solution to the problem of interaction between the measurement of solids content and ash content is to carry out a series of calibrations using a number of slurries of known solids content and then to apply an iterative method to establish ash and solids contents independently from the relevant equations.

Boyce [2] has developed an alternative approach which allows on-line equipment to be calibrated over a wide range of slurry/solids contents without requiring a knowledge of the actual values of the solids contents. Only 'dry-basis' ash values are required for calibration. The method has been applied to slurries with solids contents varying from 18% to 35% and ash contents between 48% and 66%, the accuracy derived for the 'dry basis' ash measurement in a 500 s measurement time being ± 4% at the 95% confidence limit.

Lyman and Cheshire [3] have also proposed a solution to the same problem based on the simultaneous measurement of the attenuation of γ-rays of different energies (60 keV and 662 keV) when the slurry is passed through a flow cell of known dimensions. A novel feature of this equipment is the avoidance of the effects of entrained air by increasing the pressure to between 250 and 300 lbf/in\(^2\) to collapse any bubbles which might be present.\(^1\)

2.1.4. Sulphur in coal

In recent years, since sulphur has been identified as a potential source of atmospheric pollution, on-line measurement of the sulphur content of coal has become of increasing concern. The possibility of measuring the 2.308 keV S K X-ray has been discussed by several workers [4-6] and reasonable success was achieved, when adequate corrections for inter-element effects were made. However, the measuring equipment incorporates a high-resolution X-ray spectrometer and, to minimize particle size effects, the samples must be dried and ground to <0.2 mm top size.

Page and Piggins [7] have demonstrated that, for British coals, some of these problems can be overcome by exploiting the correlation between sulphur and iron in the coals and measuring the Fe K X-ray (E_{\text{K}\alpha_1} = 6.403 keV). This allows the particle size to be increased to 5 mm top-size with measurement by a low-resolution proportional counter. A typical energy spectrum is shown in Fig. 1.

The underlying philosophy is that the concentration of organic sulphur which is chemically combined within the coal substance is reasonably invariant for a known coal type from a particular geographical area. Since, in British coals, sulphate sulphur is normally present only in very low concentrations, variations in sulphur content which do occur are principally due to variations in the pyritic sulphur (FeS\(_2\)). Hence, monitoring the iron content is a satisfactory indicator of sulphur concentration variations, providing FeS\(_2\) is the main source of iron in the coal.

\(^1\) 1 lbf/in\(^2\) = 6895 Pa.
The results of circulating seven suites of coal four times each through a commercial on-line monitor\footnote{National Coal Board/Atomic Energy Research Establishment Phase 3A coal ash monitor.} and counting for a total time of three minutes are given in Table I. A linear regression analysis of sulphur content on the iron and backscatter peak count rates was made for each suite and gave values of $\sigma$ between 0.02 and 0.16% sulphur. The backscatter peak was included to give a possible matrix correction factor, but only a very slight reduction in standard deviation was obtained by its inclusion.

It was concluded that the sulphur content can be determined to a reasonable accuracy provided the ash content does not vary by more than $\pm 4\%$ and the non-pyritic sulphur and non-pyritic iron contents do not vary by more than $\pm 0.3\%$. In general, low-ash, washed coals give a calibration uncertainty within $\pm 0.1\%$ sulphur ($1\sigma$).

2.1.5. Measurement of ash-in-coal by pair-production techniques

The main limitations of the method referred to above are the rate at which the coal is analysed and the requirement for a crusher to reduce the particle distribution to $-5$ mm. A preferred arrangement is to measure the ash content when the coal is on a main conveyor, thus avoiding sampling and at the same time improving the reliability of the measurement.
TABLE I. ACCURACY OF SULPHUR CONTENT DETERMINATION USING 5 mm-O COAL ON AN ‘ON-LINE’ ANALYSER

<table>
<thead>
<tr>
<th>Type of coal</th>
<th>Ash (%)</th>
<th>Sulphur (%)</th>
<th>Standard deviation (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Washed</td>
<td>3–7</td>
<td>0.8–1.2</td>
<td>0.02</td>
</tr>
<tr>
<td>Washed</td>
<td>5–7</td>
<td>1.1–1.4</td>
<td>0.05</td>
</tr>
<tr>
<td>Washed</td>
<td>5–7</td>
<td>1.8–1.9</td>
<td>0.03</td>
</tr>
<tr>
<td>Washed</td>
<td>7–9</td>
<td>1.2–1.5</td>
<td>0.03</td>
</tr>
<tr>
<td>Washed</td>
<td>2–8</td>
<td>0.8–2.5</td>
<td>0.16</td>
</tr>
<tr>
<td>Blended</td>
<td>11–20</td>
<td>1.8–2.3</td>
<td>0.06</td>
</tr>
<tr>
<td>Blended</td>
<td>12–25</td>
<td>1.3–1.5</td>
<td>0.07</td>
</tr>
</tbody>
</table>

The pair-production technique reported by Millen et al. [8] provides a solution to this requirement. In this method the coal is irradiated by \( \gamma \)-rays with an energy above the pair-production threshold at 1.022 MeV: the preferred source is \(^{226}\text{Ra}\), which emits 1.76 MeV and 2.20 MeV \( \gamma \)-rays. The resultant electron-positron pairs annihilate and the intensity of the resultant 0.511 MeV \( \gamma \)-rays is measured. Since the pair-production probability is proportional to the average \((Z^2/A)\) of the coal, a high specific sensitivity to the mineral fraction is obtained.

In practice, apart from measuring the intensity of the 0.511 MeV peak, the Compton-scattered radiation is also measured, presumably to give a correction to variations in bulk density.

In one arrangement installed on a conveyor belt (Fig. 2) the 0.511 MeV \( \gamma \)-rays are in backscatter geometry and the intensity of the transmitted radiation is used to determine the mass per unit area of the coal on the belt.

From field trials on a re-circulating coal loop, provided the coal depth > 60 kg/m², then for seven 2-tonne samples containing ash in the range 7.5 to 33 wt%, the RMS deviation between the pair-production gauge results and the chemical laboratory ash determinations was 1.07 wt% ash for individual 300 s determinations and 0.45 wt% ash using the mean ash determined for each sample.

2.2. Neutron interaction techniques

Neutron techniques have the very important advantage that they are elementally specific. In principle, this means that any element which generates a \( \gamma \)-ray following a particular neutron interaction can be identified and, provided there exists an appropriate calibration, the concentration of the element can be determined. However, in practice, variations in neutron transport resulting from
variations in the physical state of the coal (changes in density and moisture content) and variations in the concentration of certain elements (strong absorbers such as chlorine or boron) can introduce serious errors in the result unless techniques can be found which overcome these difficulties. In addition, the multiplicity of $\gamma$-rays and their interactions in the sample and in the detector result in the build-up of a background in the $\gamma$-ray energy spectrum which increases the statistical error on any required $\gamma$-ray peak, especially at the lower energies. Also, high-resolution Ge detectors are sensitive to fast neutron damage which
results in a decrease in energy resolution and this imposes a constraint on the
efficacy of on-line equipment when a rapid analytical result is required.

Some methods of overcoming these problems are briefly discussed.

Fortunately, there are a number of applications where concentrations of
only a few elements are required and to some extent this simplifies the equip­
ment. In addition, other information intrinsic to the problem, particularly the
type and uniformity of the composition of the coal to be analysed and the
stoichiometric relationships which exist, can also be useful input to achieving
acceptable solutions. Some options are described below.

2.2.1. Total elemental analysis

The total elemental analysis of coal derived from measurement of the intensi­
ties of prompt and delayed \( \gamma \)-rays generated by neutron interrogation requires
that fast as well as thermal neutron reactions must be utilized since, in particular,
ox oxygen cannot be identified from the radiative capture of thermal neutrons.

The analytical model developed by Wormald and Clayton [9] assumes that,
within the coal volume interrogated, an equilibrium neutron flux space-energy
distribution exists such that the neutron energy spectrum resembles a Maxwellian
thermal group plus a \( 1/E \) distribution and a fast neutron group which is invariant
in shape and closely resembles the neutron source spectrum. It is also assumed
that elements in the combustible and mineral components are intimately mixed
and, in the first place, that the thermal neutron capture cross-sections of all
elements have the same \((1/v)\) form and that they can be identified by thermal
neutron capture \( \gamma \)-ray spectrometry.

Oxygen can be included using a method based on measuring the intensity
of the 6.13 MeV \( \gamma \)-ray from the \(^{16}\text{O}(n,n'\gamma)\) reaction and linking this measure­
ment to another fast neutron reaction from an element which also emits a \( \gamma \)-ray
from thermal neutron capture. The preferred link element is silicon, but carbon
is also a candidate.

The scattering probabilities for oxygen, silicon and carbon rise steeply
above the threshold energies to some significant values and then fluctuate about
those values over an energy range of many MeV, while the dominant \( \gamma \)-ray has
an energy just equal to the threshold energy. The method for including oxygen
assumes an invariance in the fast neutron energy spectrum above the \(^{28}\text{Si}(n,n'\gamma)\)
or the \(^{12}\text{C}(n,n'\gamma)\) reaction thresholds.

To establish the validity of the fast-thermal neutron reaction link requires
a knowledge of the fast neutron flux distributions produced by the various types
of neutron source as functions both of energy and position. To start with, one
may assume that in homogeneous materials the fast neutrons from the source
are unselectively moderated and thermalized, i.e. in moving away from the source
the fast neutron spectrum simply becomes weaker, its shape remaining almost
TABLE II. OXYGEN-SILICON RESPONSE RATIOS FOR A 14 MeV NEUTRON SOURCE IN A SPHERE OF RADIUS 100 cm

<table>
<thead>
<tr>
<th>Boundary radius (cm)</th>
<th>A</th>
<th>B</th>
<th>C</th>
<th>D</th>
<th>% SD</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.0</td>
<td>0.864</td>
<td>0.832</td>
<td>0.890</td>
<td>0.847</td>
<td>1.4</td>
</tr>
<tr>
<td>5.0</td>
<td>0.491</td>
<td>0.485</td>
<td>0.518</td>
<td>0.492</td>
<td>1.9</td>
</tr>
<tr>
<td>10.0</td>
<td>0.363</td>
<td>0.365</td>
<td>0.393</td>
<td>0.369</td>
<td>2.4</td>
</tr>
<tr>
<td>15.0</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>20.0</td>
<td>0.266</td>
<td>0.262</td>
<td>0.286</td>
<td>0.263</td>
<td>3.4</td>
</tr>
<tr>
<td>25.0</td>
<td>0.222</td>
<td>0.200</td>
<td>0.247</td>
<td>0.215</td>
<td>4.9</td>
</tr>
<tr>
<td>30.0</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>35.0</td>
<td>0.211</td>
<td>0.171</td>
<td>0.216</td>
<td>0.189</td>
<td>6.9</td>
</tr>
<tr>
<td>40.0</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>50.0</td>
<td>0.178</td>
<td>0.166</td>
<td>0.188</td>
<td>0.176</td>
<td>10.7</td>
</tr>
<tr>
<td>70.0</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

constant. By assuming that the shape of the fast neutron spectrum is position-independent, at least for energies above the silicon inelastic scattering threshold, and that the coal components are intimately mixed, expressions for the relative intensities of the main elements, including oxygen, can be derived in terms of the $\gamma$-ray yields from inelastic scattering from the elements O, C and Si and of the yields from radiative capture.

To check the assumption that the ratios of the $\gamma$-ray yields from oxygen, carbon and silicon are the same for every volume element in the coal, Clayton and Coleman [10] have carried out a series of Monte Carlo calculations of the neutron flux distributions from 14 MeV, $^{241}$Am-Be and $^{252}$Cf neutron sources in spherically symmetric and slab configurations for several types of bituminous coal.

The results indicate that the initial assumption in the Wormald-Clayton model was an oversimplification. For the lower-energy neutron sources the ratios of the $\gamma$-ray yields show a dependence on position which becomes very marked for the 14 MeV source so that the very simple expressions for relative elemental concentrations derived from the model cannot be used without caution. However, the calculations also show that the ratios of the specific $\gamma$-ray yields, i.e. the yields per unit density of the various elements, are almost independent of coal
type, although they do vary from one configuration to another. It is therefore concluded that expressions for the relative elemental concentrations similar to those derived by Wormald and Clayton [9] can be employed, but that calibration factors will have to be calculated for a coal of intermediate type in the required configuration, and that the first order estimates of elemental concentrations may require minor corrections if they are widely different from those used to calculate the calibration factors.
Table II gives the calculated ratios of the specific γ-ray yields from oxygen and silicon within a succession of spherical shells surrounding a 14 MeV neutron source. Coal types A, B, C and D refer respectively to an almost ash-free coal, coal with a large ash content, coal with a large water content, and coal of intermediate composition. The very small dependence of these ratios on coal type and the strong dependence on distance from the source are clearly evident.

Several other observations emerged from the calculations.

Thus, Fig. 3 shows, for a point source of 14 MeV neutrons in a coal of intermediate composition, the calculated fluxes in two spherical shells at 10–15 cm.
and 23–30 cm from the source expressed as the ratio $\lambda_0$ of the flux in an energy band of range $(e/1)$ to the thermal flux. The troughs in energy bands at 1.77–2.37, 3.16–4.22, and 7.5–10.0 MeV appear systematically for a range of sources and coal types, and are apparently associated with large changes in the neutron scattering and absorption cross-sections for carbon in these energy regions. Below 10 keV the slowing-down spectrum approximates very closely to a $1/E$ spectrum. The low value ($\sim 0.02$) for $\lambda_0$ indicates the excellent moderating properties of the coal.
The slab calculations were made in terms of the integrated uncollided γ-ray fluxes reaching a detector on the side of the slab remote from the source for a range of source and detector stand-off distances. They provide striking evidence for the build-up of the thermal neutron flux over a distance of 10 cm or so into the slab, and allow the concentrations to the integrated flux from various volume elements within the slab to be determined. Figure 4 shows the relative γ-ray yields from inelastic scattering by silicon for a source stand-off of zero cm and a detector stand-off distance of 5 cm. Some 25% of the response comes from a hemisphere of diameter 10 cm directly in front of the source. The figure is a section of the slab by a plane through the positions of the (point) source and detector. The yields are expressed as relative yields from toruses of unit cross-sectional area obtained by rotating the figure about the source-detector axis. Figure 5 shows similar results for a source stand-off of 20 cm and a detector stand-off of 5 cm, a combination which gives reasonably uniform contributions over the whole width of the slab.

2.2.2. Registration and analysis of complex spectra

At the points of emission the energy spectrum of γ-rays generated by coal consists of a series of isolated narrow lines whose energies are accurately known. The derivation of elemental concentrations is based on their relative intensities. In the passage through the material the isolated γ-rays undergo some changes to their relative intensities, and a significant continuum develops which forms a background to the isolated lines.

A typical γ-ray detector responds to a flux of monoenergetic γ-rays with energies below ~ 2 MeV by producing a spectrum of pulses (referred to as the response function) with a single broadened peak corresponding to the full γ-ray energy, and a more or less uniform continuum extending from below the peak to very low energies. However, this continuum does have some significant features and, in particular, the Compton edge can produce difficulties in data analysis. If the γ-ray energy is significantly greater than 2 MeV the spectrum exhibits three peaks, corresponding to the full energy, the full energy less 0.511 MeV and the full energy less 1.02 MeV, the extra peaks being associated with the generation of positrons within the detector crystal by pair creation. The shapes of the response functions depend on the γ-ray energy, on the type and shape of the detector and on the spatial distribution of the γ-rays near the detector. These shapes can be determined experimentally, or they can be calculated, for example by Monte Carlo procedures. The larger the detector crystal the larger the fraction of the spectrum that appears in the peaks. For both technical and economic reasons the largest NaI(Tl) crystals in use have volumes several times greater than those of the largest germanium crystals.

The broadening of the peaks depends on the type of detector employed. For γ-rays with energies above 2 MeV the broadening ranges from a few per cent
for scintillation counters using NaI(Tl) or bismuth germanate to a few keV for high-resolution Ge detectors. Against the much better resolution of the latter must be set their high cost, relatively low efficiency and loss of resolution when used at high counting rates. There is also a progressive loss of resolution as a result of radiation damage.

Several factors give rise to problems in analysing the spectrum. The first is that the lines in a coal spectrum are relatively closely spaced because of the number of contributing elements and the effective spacing is further reduced above 2 MeV by the three-peak detector response at high energies. Some of the spacings are uncomfortably close even with the resolution of HPGe detectors, and in general it is hardly worth using scintillation counters unless sufficient is known about the concentration patterns in the coal to allow energy intervals spanning groups of lines to be used for analysis. A related factor is that a peak may be on or near a Compton edge. An additional factor arises because the contributions to the pulse height spectrum continuum from all higher energy $\gamma$-rays add up to lower energies. Thus, the backgrounds at the lower energies are often much larger than the peaks under which they lie, and the statistical uncertainties in the integrated peak counts then become disproportionately large.

These problems can be minimized by employing complex detector systems which considerably reduce the detector response function for a monoenergetic $\gamma$-ray at pulse heights below that corresponding to the main peak, usually at the cost of a large drop in counting rate. Auxiliary detectors are employed for this purpose. At intermediate energies commercially available Compton suppression systems are reasonably effective. At higher energies pair spectrometers using two auxiliary detectors are under development, their outputs being processed to ensure that only the lowest of the three peaks in a typical high-energy response function is actually registered. The response function for a complex system tends to depend on the way in which the system is set up, so that it is difficult to calculate.

Since the heterogeneity in coal composition relates to no more than about ten elements, the energy regions containing the key $\gamma$-ray peaks are actually quite limited. Hence, the simplest form of analysis is to determine the integrated counts in those peaks which are well isolated and to use a limited amount of curve fitting to analyse groups of peaks which are poorly resolved. In practice such an analysis is limited to spectra obtained from high-resolution Ge detectors and is complicated by the presence of Compton edges in the continuum. If the detector response functions are available, then in principle a full deconvolution of the original energy spectrum from the detector pulse height spectrum can be performed, but such a procedure is unlikely to give useful results.

A much more promising approach is to exploit the fact that the bulk of the detector spectrum arises from about one hundred lines generated by approximately ten elements present in the coal and in the detector environment. Allowing 4 or 5 parameters to specify the relatively smooth continuous $\gamma$-ray background permits an approximately fifteen parameter fit to the whole spectrum, which is
certainly a reasonable target. Such a procedure could be used with the somewhat poorer resolution spectra obtained from Ge detectors at very high counting rates and might even allow spectra obtained using scintillation counters to be analysed successfully.

From the above considerations and from comparisons of the present rates of development of detector technology and of computer processing power we are led to conclude that an important future trend in coal analysis will be the use of increasingly sophisticated and comprehensive procedures in numerical analysis.

2.2.3. Measurement of the specific energy of coal

It has been established that the specific energy of coal correlates very well with the carbon content \[11, 12\], showing a deviation as small as 0.3 kcal/kg. The carbon content can be determined from the yield of the 4.43 MeV $\gamma$-rays produced by the inelastic scattering of fast (\(> 5\) MeV) neutrons from this element. Sowerby \[11\] used a $^{238}$Pu-Be neutron source and indicates a sampling depth approaching 20 cm and makes the point that measurements on coal carried by conveyor belts are feasible.

The background beneath the 4.43 MeV $\gamma$-ray peak is usually five or more times more intense than the peak itself so that long counts are required to obtain acceptable statistical accuracies. Nevertheless, Sowerby achieved useful results with a 15 cm diameter 10 cm NaI(Tl) detector. He emphasizes the effect of density variations on the measured yield and describes a $\gamma$-ray backscattering system which can be substituted for the neutron system to obtain a measurement of bulk density. It is designed to have a similar sampling pattern in the coal.

Cywicka-Jakiel et al. \[12\] describe a technique for reducing the relative background counts by exploiting the fact that each neutron emitted from a $^{238}$Pu-Be source is followed almost immediately by a 4.43 MeV $\gamma$-ray and that inelastic scattering events involving carbon atoms usually occur within 1 $\mu$s after the neutron is emitted, whereas the radiative capture of thermal neutrons may be delayed by as long as 1 ms. In the experimental system one detector is mounted close to the source to detect the $\gamma$-ray from the \((\alpha,\text{Be})\) reaction and the other, which is shielded from direct radiation, detects the $\gamma$-ray from inelastic scattering on the carbon in the coal and measures the time correlation between the two outputs (effectively coincidences between the outputs within a time of 1 $\mu$s). The procedure worked effectively, but it involves a considerable loss of counting rate and statistical factors limit the improvement that can be obtained simply by increasing the activity of the neutron source.

2.2.4. Measurement of the concentrations of selected elements

(a) Measurement of ash content

For many applications measurement of ash content is the dominating requirement. As stated above, low-energy X-ray techniques have serious limitations
operationally and although equipment based on the pair-production technique can be mounted directly on-line it is still in the early stages of development. An alternative option is to measure the Al and Si concentrations in the coal and to rely (by previous analytical confirmation) on a strong correlation between the total concentration of these two elements and the ash content.

Using a $^{241}$Am-Be neutron source, the $^{27}$Al$(n,\gamma)$ and $^{28}$Si$(n,p)$ reactions both result in the emission of the 1.779 MeV $\gamma$-ray from $^{28}$Mg, the half-life of the parent state ($^{28}$Al) being 2.4 min. Variations in bulk density arising from changes in packing density and from moisture content can affect the result. However, it has been shown that a separate measurement of hydrogen content can be used to normalize this effect [11].

One of the merits of this method is that, since delayed $\gamma$-rays are being measured, counting equipment can be mounted away from the source and hence it is divorced from the associated neutron and prompt $\gamma$-ray flux. In consequence, a high-resolution detector is not required.

This approach is limited by variations in iron content. If these have to be allowed for, a prompt $\gamma$-ray technique is required but this defeats the attraction of using a scintillation counter.

(b) Measurement of sulphur content

Sulphur can be identified through the 5.420 MeV $\gamma$-ray from thermal neutron capture and sulphur concentrations are now of interest (or required) for reasons of pollution control. An isolated measurement of sulphur content can be made to an acceptable accuracy provided that large corrections for variations in neutron transport are not required. Generally a high-resolution detector is needed for the measurement.

(c) Use of stoichiometric relationships

In many situations where coal analyses are required the mineral composition is reasonably stable, as is the composition of the combustible component. Where this situation applies it is questionable whether oxygen needs to be measured, since it could be obtained from all other elements by the known stoichiometry. The Wormald-Clayton model would still apply, but in a simplified form without the fast neutron coupling component. The merit of this approach is that the detector could be more isolated from the fast neutron flux, with a resultant improvement in lifetime and $^{252}$Cf could also be employed with a lower fast neutron component than $^{241}$Am-Be, for example.

An interesting possibility for the application of a stoichiometric relationship is the measurement of the moisture content of coals which is an important but particularly difficult measurement.
In many deposits the carbon/hydrogen ratio of the coal substance is adequately constant and can be measured accurately by classical methods. Since the mineral component of the coal is generally low in hydrogen, a measurement of total hydrogen (via the 2.23 MeV $\gamma$-ray) in a wet coal can be used to infer the moisture content.

(d) Corrections for the hydrogen effect in bulk density measurement by $\gamma$-ray scattering

In Section 2.1.1 it was stated that, in principle, variations in bulk density can be made to high accuracy by ($\gamma$--$\gamma$) techniques provided that Compton-scattered $\gamma$-rays only are recorded and the hydrogen content of the coal/mineral mixture is invariant.

When a high accuracy is required, such as in borehole logging during the early assessment of marginal deposits, corrections for hydrogen variations can be made in dry boreholes by direct measurement of the 2.23 MeV $\gamma$-ray from thermal neutron capture. With a suitable design it is convenient to mount the hydrogen monitor in the same probe as the ($\gamma$--$\gamma$) density equipment.

(e) Measurement of the solids weight fraction in coal slurries

Watt [13] has proposed a method to determine the solids weight fraction of mineral matter of coal in slurries which is not affected by variations in voidage due to air entrainment. The method depends on the correction between the solids weight fraction and the concentration of hydrogen in the slurry. It is determined by combining neutron moderation measurements for hydrogen per unit volume with $\gamma$-ray transmission measurements for bulk density. These measurements can be combined with low energy X-ray measurements to give a more accurate measurement of the solids weight fraction and the mineral component than would be otherwise possible.

3. A CALIBRATION FACILITY

Laboratory and on-line equipment, either for the measurement of elemental concentrations or for the direct determination of some defined parameter such as ash content, requires calibration, both during installation and at regular intervals during the operational lifetime. Similarly, during the development of new techniques it is frequently required to test the response of a proposed system in an orderly manner and against a selected range of coal properties. In addition, a potential user needs strong assurance that a proposed new installation will perform to specification and it is generally unsatisfactory to predict performance
from results obtained on a 'different' coal at a different site. A temporary installation to prove a new option can be expensive and tedious to construct so that there now seems to be a need for a flexible, mobile arrangement which can be easily transported to the equipment and used to give an early indication of performance with the types of coal which it might be expected to handle in the future.

3.1. The unit calibration block

A standardizing facility has been developed which meets the needs of situations identified above (Fig. 6). It is fabricated from two sizes of elementary hollow container which can be filled with real or artificial coals to a well-defined density and moisture content. The shape of the main block is hexagonal, as shown in Fig. 7, and there is also a secondary half-hexagonal block which enables the standardizing facility to have a flat top and bottom. The hexagonal cross-section is used to prevent neutron and γ-ray streaming and the blocks are constructed to a very close tolerance to give a high precision in dimensional reproducibility. When assembled the shapes give constructional stability.

The dimensions chosen enable the facility to be constructed in incremental steps of 2 cm.

The blocks can also be formed into an array for calibrating borehole logging probes. The hexagonal cross-section and dimensions selected result in a near-circular inner surface.

3.2. Fabrication and construction

Each block is made of a carbon-fibre-nylon composite with close similarity in composition to the combustible component of bituminous coal. The wall
thickness is approximately 0.5 mm and the shell and cap are formed by injection moulding. Each block is filled with one of a series of artificial coals and sealed. The density of each block and a signature which indicates the composition of the contents are inscribed on the outside. The signature refers to a certificate which fully describes the contents.

4. CONCLUSIONS

Several nuclear techniques are now available and a number of new, potentially attractive techniques are being developed to meet an increasing range of applications in the coal industry.

The trend is towards full on-line analysis. High-energy γ-ray and neutron techniques, because of their range, have characteristics which suggest a good match to the basic requirements.

A number of laboratories are giving considerable attention to the development of equipment for providing a near-total on-line analysis. Stoichiometric correlations and the simultaneous use of more than one technique are being employed to improve measurement accuracy and to simplify the instrumental analytical requirements.
Although much has been achieved, the requirement for a rapid, highly accurate, total analysis of the primary elements in coal has not yet been met and the fundamental limitations of γ-ray detectors and the associated electronics are now being approached. Several options to reduce the instrumental problems are being examined based on multiple-detector systems, spectrum analysis and fast data-handling techniques.

REFERENCES


DISCUSSION

P.L. SURMAN: On the point about calibration I think the other half of the difficulty is that the traditional chemical methods of sampling and analysis themselves have problems and the nuclear techniques are getting to a stage now where they are equalling and perhaps bettering the combined sampling and analysis conventionally available to us. That is another reason why we identified a need for an absolute standard against which we could investigate the nuclear techniques. It was not just the unavailability of coal locally to the instrument.
J.S. WATT: Where do you see the applications of the neutron techniques coming in? We have simple ash gauges and we have more complicated techniques for elemental abundance. Could you give some indication of where the more complex techniques are necessary to solve problems in the coal industry?

C.G. CLAYTON: I think the question has to be put in terms of the accuracy of the techniques which do not use neutrons and the particular requirements which vary worldwide. If ash content only is required, then this can be derived by a gauge which does not include neutron techniques. On the other hand, if ash is required along with some elemental concentration, such as sulphur, then that information cannot be extracted from an ash gauge. If specific energy, ash content and some elemental concentrations are required, for furnace control, for example, more complex equipment is needed.

Much depends on your point of view. The producer is governed by the requirements of the user. He will not measure the quality of his coal unless he has to do so. My own impression is that there is more and more pressure coming from the users to measure several parameters, particularly from power station operators in order to control boiler efficiency. In this situation there is no other option but to go for techniques which will give you the elemental parameters that you want, including ash.

T. GOZANI: As a general comment, you have discussed a lot of techniques and you spent proportionately more time on the fast neutron measurement of, presumably, mostly oxygen through calibration with silicon. This is only a certain aspect and it is an issue whether you even need oxygen. I think one result of this meeting ought to be a clear understanding of where the problems are, and I will put my opinion that the problems are not in the physics area, they are in the applications area, i.e. in the power plant, the coal mining areas, etc. The problem is how to use the techniques.

Also we have to make the measurements either less dependent on unknown assumptions, or state clearly what are the assumptions or approximations involved in each technique. For specific items, you apparently have a great love for Monte Carlo methods. This is a very expensive technique. I wonder if you recommend it to everybody, or is it just a prerogative of Harwell? Most of the calculations you could probably do with a transport calculation and achieve similar results.

Thirdly when you talk about the issue of the uniformity of the results between carbon, oxygen and silicon, the key is what is the leakage rate? I saw no leakage rate in your results, just flux as a function of position in an infinite medium. That might be quite different because of the gradient towards the outside of the sphere.

C.G. CLAYTON: I thought you were going to say that the problems are concerned more with equipment limitations, which is what we think. A particular requirement is the need to handle higher input data rates.
I do agree that we need input on what the customers require. In the United Kingdom we have some benefits from being a small country. We can interrogate our suppliers and users quite rapidly. We know what they say they want. Therefore, as far as we are concerned, the principal question is whether we can satisfy their needs.

As to why we use Monte Carlo, this depends on whether you have these codes readily available, and we have. But there are other factors. When considering point source infinite geometry, Monte Carlo is not expensive and the codes are well developed. It is the cheapest way of solving the problem. You can certainly use transport calculations but when you include particular equipment, such as borehole probes asymmetrically situated in a borehole with boundaries in which the absorption cross-sections are very different, then you come up against a situation where transport calculations cannot be used with an acceptable accuracy. It is in these realistic situations that Monte Carlo codes are more important but then they can be expensive if high statistical accuracy is required for small volumes in regions of low flux. We now use coupled Monte Carlo and diffusion codes which have diffusion coefficients determined by benchmark Monte Carlo. Essentially the diffusion codes give us an accurate indication of the overall trend by varying a particular parameter, few Monte Carlo runs being used to set the absolute levels. This combination gives very comprehensive information without being very expensive.

T. GOZANI: I agree that my view is influenced mainly by the American situation, but I still maintain that the major problems are not technical, are not interpretational, but lie with the users who do not know to the full extent how to use these techniques.
BOREHOLE ELEMENTAL ANALYSIS OF COAL USING FAST NEUTRONS

J.L. MIKESSELL, F.E. SENFTLE, A.B. TANNER
United States Geological Survey,
Reston, Virginia,
United States of America

Abstract

BOREHOLE ELEMENTAL ANALYSIS OF COAL USING FAST NEUTRONS.

A borehole generator for producing 14 MeV neutrons has been constructed, using the 'zetatron' tube design developed by Sandia National Laboratories. The generator can be joined to a high-purity germanium or a sodium iodide detector for measuring neutron-induced gamma radiation in a coal seam. The neutrons cause various nuclear reactions to produce prompt and decay gamma rays by which one may measure C, H, O, N, S, and ash in coal. Spectra made in coal during a 10 μs time window coincident with the burst of fast neutrons are shown. The decay gamma rays following the activation of oxygen by means of the (n, p) reaction provide a practical means of measuring oxygen, which cannot be measured using $^{252}\text{Cf}$.

1. INTRODUCTION

High-resolution prompt capture gamma-ray spectroscopy of coal, measured in a borehole, was first demonstrated in 1978 [1]. The experimental technique involved the use of a germanium detector mounted in a special cryostat that cooled the detector crystal with a melting propane cryogen [2–4]. A $^{252}\text{Cf}$ fission source, mounted below the detector, provided neutrons for activating the coal. A method suggested by Fanger et al. [5] in their work on scheelite ores was modified and used to extract from the spectral data a whole-coal ultimate analysis: a measurement of the mass fractions of the elements C, H, N, S, O, and the ash fraction.

The method of Fanger et al. requires that one measure the intensity of at least one gamma ray from each element present in the sample. This was not possible for oxygen, which is not detectable by means of the low-energy neutrons emitted by $^{252}\text{Cf}$. Although some minor metals that are constituents of the ash fraction may not be detected in a spectrum made under field conditions, an adequate determination of ash may be made from the measurements of silicon, aluminium, and iron. Clayton et al. [6] have recently made analyses of coal in a simulated borehole in the laboratory using essentially the same method. They concluded that fast neutron inelastic reactions must be used.
### TABLE I. DETECTABLE ELEMENTS IN AN 'AVERAGE' US COAL

<table>
<thead>
<tr>
<th>Concentration rank</th>
<th>Element</th>
<th>Concentration, f, (whole-coal basis) (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Carbon</td>
<td>64.1</td>
</tr>
<tr>
<td>2</td>
<td>Oxygen</td>
<td>16.4</td>
</tr>
<tr>
<td>3</td>
<td>Hydrogen</td>
<td>5.1</td>
</tr>
<tr>
<td>4</td>
<td>Silicon</td>
<td>2.6</td>
</tr>
<tr>
<td>5</td>
<td>Sulphur</td>
<td>2.0</td>
</tr>
<tr>
<td>6</td>
<td>Iron</td>
<td>1.6</td>
</tr>
<tr>
<td>7</td>
<td>Aluminium</td>
<td>1.4</td>
</tr>
<tr>
<td>8</td>
<td>Nitrogen</td>
<td>1.1</td>
</tr>
<tr>
<td>9</td>
<td>Calcium</td>
<td>0.54</td>
</tr>
<tr>
<td>10</td>
<td>Potassium</td>
<td>0.18</td>
</tr>
<tr>
<td>11</td>
<td>Magnesium</td>
<td>0.12</td>
</tr>
<tr>
<td>12</td>
<td>Chlorine</td>
<td>0.1</td>
</tr>
<tr>
<td>13</td>
<td>Titanium</td>
<td>0.08</td>
</tr>
<tr>
<td>14</td>
<td>Sodium</td>
<td>0.06</td>
</tr>
<tr>
<td>15</td>
<td>Manganese</td>
<td>0.01</td>
</tr>
<tr>
<td>16</td>
<td>Boron</td>
<td>0.005</td>
</tr>
<tr>
<td>17</td>
<td>Cadmium</td>
<td>0.00013</td>
</tr>
</tbody>
</table>

In this paper the problem is examined on an element by element basis and a description is given of some preliminary laboratory experiments designed to study the potential of a neutron generator for fast-neutron analysis of coal in a borehole.

### 2. LIMITATIONS OF THERMAL CAPTURE

Table I lists in order of concentration seventeen of the elements found in an 'average' US coal. The mass fractions listed are taken from Swanson et al. [7], and represent an average over 799 published whole-coal elemental analyses. Table II uses the same data, but lists the elements in order of the expected strength of their photopeaks in a prompt capture spectrum. Cross-section and intensity data in Table II are taken from Rasmussen et al. [8]. With the exception
of hydrogen and boron, the elements produce only a weak response. An accurate measurement of the photopeaks of carbon, the element of highest concentration, requires that spectral data be collected for periods of 20 minutes to an hour. Several ash constituents, particularly calcium and potassium, are quite hard to detect by capture gamma-ray analysis, nitrogen is still more difficult, and oxygen is impossible to detect.

**TABLE II. THE ELEMENTS OF TABLE I, HERE LISTED IN ORDER OF RELATIVE PHOTOPEAK STRENGTH USING THE PHOTOPEAK OF HIGHEST INTENSITY AND THE AVERAGE FRACTIONAL CONCENTRATIONS OF TABLE I.**

*A is the atomic weight. Cross-section and intensity data from Ref. [8].*

<table>
<thead>
<tr>
<th>Concentration rank</th>
<th>Element</th>
<th>( \sigma ) (capture)</th>
<th>Strongest usable photopeak (keV)</th>
<th>( I ) (( \gamma )-rays/100 captures)</th>
<th>Relative photopeak strength (( \sigma )If X 100)/A</th>
</tr>
</thead>
<tbody>
<tr>
<td>3</td>
<td>Hydrogen</td>
<td>0.33</td>
<td>2223</td>
<td>100</td>
<td>168.3</td>
</tr>
<tr>
<td>16</td>
<td>Boron</td>
<td>752</td>
<td>478</td>
<td>479</td>
<td>163</td>
</tr>
<tr>
<td>1</td>
<td>Carbon</td>
<td>0.034</td>
<td>1261</td>
<td>29.2</td>
<td>5.2</td>
</tr>
<tr>
<td>12</td>
<td>Chlorine</td>
<td>33.2</td>
<td>1165</td>
<td>25.7</td>
<td>2.4</td>
</tr>
<tr>
<td>5</td>
<td>Sulphur</td>
<td>0.512</td>
<td>841</td>
<td>54.3</td>
<td>1.74</td>
</tr>
<tr>
<td>4</td>
<td>Silicon</td>
<td>0.160</td>
<td>3539</td>
<td>79.6</td>
<td>1.18</td>
</tr>
<tr>
<td>7</td>
<td>Aluminium</td>
<td>0.235</td>
<td>1779</td>
<td>88.2</td>
<td>1.07</td>
</tr>
<tr>
<td>6</td>
<td>Iron</td>
<td>2.62</td>
<td>352</td>
<td>10.9</td>
<td>0.82</td>
</tr>
<tr>
<td>13</td>
<td>Titanium</td>
<td>6.09</td>
<td>1381</td>
<td>65.5</td>
<td>0.67</td>
</tr>
<tr>
<td>17</td>
<td>Cadmium</td>
<td>3260</td>
<td>559</td>
<td>79.7</td>
<td>0.33</td>
</tr>
<tr>
<td>9</td>
<td>Calcium</td>
<td>0.43</td>
<td>1943</td>
<td>52.5</td>
<td>0.3</td>
</tr>
<tr>
<td>10</td>
<td>Potassium</td>
<td>2.1</td>
<td>771</td>
<td>31.2</td>
<td>0.3</td>
</tr>
<tr>
<td>8</td>
<td>Nitrogen</td>
<td>0.075</td>
<td>1888</td>
<td>27.5</td>
<td>0.16</td>
</tr>
<tr>
<td>15</td>
<td>Manganese</td>
<td>13.3</td>
<td>7243</td>
<td>12.</td>
<td>0.029</td>
</tr>
<tr>
<td>14</td>
<td>Sodium</td>
<td>0.534</td>
<td>2027</td>
<td>19.81</td>
<td>0.028</td>
</tr>
<tr>
<td>11</td>
<td>Magnesium</td>
<td>0.063</td>
<td>2828</td>
<td>35.7</td>
<td>0.011</td>
</tr>
<tr>
<td>2</td>
<td>Oxygen</td>
<td>0.00018</td>
<td>–</td>
<td>–</td>
<td>–</td>
</tr>
</tbody>
</table>
TABLE III. THERMAL AND 14 MeV NEUTRON REACTIONS FOR THE ELEMENTS IN COAL.

The cross-section data are from Ref. [13] and the gamma-ray data are from various sources [15-17]

<table>
<thead>
<tr>
<th>Reaction</th>
<th>$\sigma$ (mb)</th>
<th>$\sigma$ (mb)</th>
<th>$T_{1/2}$</th>
<th>Gamma-ray energies (keV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Th</td>
<td>322</td>
<td>-</td>
<td>-</td>
<td>Stable</td>
</tr>
<tr>
<td>14Tl/2</td>
<td>425</td>
<td>82</td>
<td>prompt</td>
<td>474</td>
</tr>
<tr>
<td>9Be</td>
<td>474</td>
<td>47</td>
<td>7.13 s</td>
<td>Stable</td>
</tr>
<tr>
<td>14N</td>
<td>430</td>
<td>9.98 min</td>
<td>511</td>
<td></td>
</tr>
<tr>
<td>160</td>
<td>491</td>
<td>prompt</td>
<td>841, 5420, 2380</td>
<td></td>
</tr>
<tr>
<td>14N</td>
<td>252</td>
<td>prompt</td>
<td>513</td>
<td></td>
</tr>
<tr>
<td>160</td>
<td>500</td>
<td>prompt</td>
<td>2230, 4282</td>
<td></td>
</tr>
<tr>
<td>14N</td>
<td>110</td>
<td>Stable</td>
<td></td>
<td></td>
</tr>
<tr>
<td>1779</td>
<td>232</td>
<td>2.25 min</td>
<td>1779</td>
<td></td>
</tr>
<tr>
<td>28Si</td>
<td>160</td>
<td>prompt</td>
<td>6380, 4934, 3539, 2093</td>
<td></td>
</tr>
<tr>
<td>28Si</td>
<td>524</td>
<td>prompt</td>
<td>1779, 2838</td>
<td></td>
</tr>
<tr>
<td>28Si</td>
<td>233</td>
<td>2.25 min</td>
<td>1779</td>
<td></td>
</tr>
</tbody>
</table>
### Table III (cont.)

<table>
<thead>
<tr>
<th>Reaction</th>
<th>σ (mb)</th>
<th>T_1/2</th>
<th>Gamma-ray energies (keV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>56Fe(n,γ)^57Fe</td>
<td>2260</td>
<td>-</td>
<td>Stable</td>
</tr>
<tr>
<td>56Fe(n,n'γ)^56Fe</td>
<td>-</td>
<td>749</td>
<td>prompt</td>
</tr>
<tr>
<td>56Fe(n,p)^56Mn</td>
<td>-</td>
<td>125</td>
<td>2.58 h</td>
</tr>
<tr>
<td>56Fe(n,2n)^55Fe</td>
<td>-</td>
<td>432</td>
<td>prompt</td>
</tr>
<tr>
<td>40Ca(n,γ)^41Ca</td>
<td>430</td>
<td>-</td>
<td>1943, 6420</td>
</tr>
<tr>
<td>40Ca(n,n'γ)^40Ca</td>
<td>-</td>
<td>243</td>
<td>prompt</td>
</tr>
<tr>
<td>40Ca(n,p)^40K</td>
<td>-</td>
<td>215</td>
<td>770</td>
</tr>
</tbody>
</table>

### 3. FAST-NEUTRON REACTIONS

The use of fast neutrons, such as those produced by the T(d, n)^4He reaction, makes analysis possible by a number of reactions that cannot be achieved with thermal neutrons. The gamma rays from these reactions often appear during definite 'time windows' which can be exploited to simplify the spectra [9]. Table III lists the elements in coal that have appreciable concentrations and the reactions that they undergo when irradiated with 14 MeV neutrons.

#### 3.1. Hydrogen

The cross-section of hydrogen for capture at 14 MeV is negligible. As no reactions other than capture are possible, the 2223 keV thermal capture photopeak is the only usable line.

#### 3.2. Carbon

Although it is the principal element in coal, carbon is difficult to detect, due to its low cross-section for the thermal capture reaction. The cross-section at 14 MeV for inelastic scattering is adequate (425 mb) and the required neutron energy is low enough so that the reaction is possible even with an Am-Be neutron source, whose neutron energies range to approximately 10 MeV. The 4439 keV photopeak can pose problems for quantitative analysis, because it is Doppler-broadened from recoil and may require special treatment during spectral data...
reduction. The sharp, single escape of the 4945 keV thermal capture photopeak (4434 keV) and the broad inelastic peak interfere with each other. If a continuous high-energy neutron source, such as an $\alpha$-Be isotopic source, is used, the data analysis is complicated because these two peaks are then superimposed in the spectrum. With a pulsed-neutron generator the two peaks can be resolved temporally, with one spectrum made during the neutron burst, containing only the inelastic photopeak and one, made during a thermal-neutron capture time window, containing only the capture peak. The $(n, p)$ reaction, while possible with 14 MeV neutrons, has a very small cross-section at that energy. The $(n, \alpha)$ reaction is unlikely to produce significant $\gamma$-radiation and is the inverse of the $\alpha$-Be reaction used to generate neutrons.

3.3. Oxygen

Oxygen also has a low thermal capture cross-section, and can be measured in a borehole only with fast neutrons. The inelastic scattering cross-section of 474 mb is appreciable, and the low neutron threshold energy of 6.4 MeV makes this reaction possible with neutrons from $\alpha$-Be sources as well as with 14 MeV neutrons. This reaction, together with the inelastic scattering reaction on carbon, has been used for a number of years in borehole logging by the petroleum industry [10]. The 6128 keV photopeak is strong, and the much weaker 7117 keV and 6919 keV peaks may also be seen. Because the half-life of the 6128 keV state of oxygen is long (18 ps) the gamma ray from this state is not Doppler-broadened by the recoil of the oxygen nucleus, as are the 7117 keV and 6919 keV gamma rays, which originate from states of half-lives of 8 and 4.2 fs, respectively.

The $(n, p)$ reaction, with a minimum neutron energy of 10.3 MeV, has a cross-section that is ten times lower than that for the $(n, n'\gamma)$ reaction. The gamma rays produced are a consequence of the beta decay of $^{16}$N nuclei. Because these gamma rays are seen in decay, and because the time to achieve saturation activation is so short ($T_{1/2} = 7$ s), the photopeaks from this reaction have a good signal-to-noise ratio with a pulsed generator in a time window following the die-away of thermal neutrons.

The neutron generator tube used in the experiments described below is cooled by immersion in a fluorocarbon dielectric fluid. The activation of this fluorocarbon via the reaction $^{16}$F$(n, \alpha)^{16}$N(25 mb) produces the same nuclide, hence the same decay gamma rays, as the oxygen $(n, p)$ reaction. Thus, if oxygen is to be measured by means of the $(n, n'\gamma)$ or $(n, p)$ reactions, the neutron generator must be provided with a well-designed shadow shield for attenuating gamma rays, so that gamma rays from the dielectric fluid do not reach the gamma-ray detector.

The $(n, \alpha)$ reaction has a low threshold (2.36 MeV) and a reasonable cross-section, but the resulting $^{13}$C nucleus is stable.
3.4. Nitrogen

Nitrogen in coal is difficult to measure because of its low concentration and low cross-section. The measurement is feasible using the high-energy capture gamma ray (10.8 MeV) because of the low background in that part of the spectrum. It is difficult to extract from the spectral data an efficiency function that can be extrapolated to such high energies because the only other high-energy gamma ray from coal is that of iron at 9298 keV. An alternative approach to measuring nitrogen would be useful.

Although the inelastic reaction has a substantial cross-section, the resulting gamma rays are in a part of the spectrum where the background is high; the signal-to-background ratio is consequently better for capture gamma rays. In the laboratory, the $^{14}$N(n, 2n)$^{13}$N reaction is frequently used [11], producing annihilation radiation in decay. For borehole work the cross-section for this reaction is too small. The (n, p) reaction, which is the source of the $^{14}$C in the earth’s atmosphere, does not produce any radiation useful in borehole work.

3.5. Sulphur

The 5420 keV thermal capture photopeak can usually be used to measure sulphur, provided one is not working in a low-sulphur coal such as that found in the western USA. The prompt photopeaks from the (n, p) and (n, n'$\gamma$) reactions might also prove useful in high-sulphur coal.

3.6. Aluminium

Aluminium may be measured using the 7724 keV thermal capture photopeak. The 1779 keV decay photopeak, which is a good peak when using $^{252}$Cf, cannot be used with 14 MeV neutrons because the (n, p) reaction on $^{28}$Si produces the same 1779 keV peak, with the same half-life. The inelastic scattering reaction has a good cross-section, except that the 844 keV photopeak is quite close to the 847 keV iron (n, n'$\gamma$) peak and the 2210 keV peak may have some interference from the hydrogen thermal capture photopeak at high pulse repetition rates when thermal neutrons are presented during the neutron burst.

3.7. Silicon

The thermal capture reaction is usually sufficient for measuring silicon, using the 3539 keV photopeak. The (n,p) reaction produces the same product as the (n,$\gamma$) reaction on aluminium; hence the source of the 1779 keV photopeak seen in decay is made ambiguous. The inelastic reaction can produce, in addition to a prompt 1779 keV peak, the broad Doppler-shifted 2839 keV prompt peak.
3.8. Iron

Prompt iron photopeaks from both thermal capture and inelastic scattering are prominent in a coal spectrum. Because the \((n, p)\) reaction produces the same isotope \(^{56}\text{Mn}\) as the \((n, \gamma)\) reaction on manganese, the decay gamma rays and the half-life of the activity are the same for both reactions. To add further ambiguity to these gamma rays, they are also seen as prompt gamma rays by the inelastic reaction on iron. However, the inelastic reaction can be distinguished from the decays following the \((n, p)\) and \((n, \gamma)\) reactions by using the 1238 keV photopeak. Because the beta decay of \(^{56}\text{Mn}\) does not populate the 2085 keV state of \(^{56}\text{Fe}\) [12], the 1238 keV photopeak originating at this state is not seen in the iron decay spectrum. If a peak of this energy is seen in the coal decay spectrum, it is due instead to the natural uranium background.

Because of the short range of gamma rays in coal (\(\approx 10\) cm) the borehole detector and its housing are irradiated at the same time as the surrounding coal. If this housing contains iron, as is common, the intensity measured for the iron peaks will be at least in part due to the presence of the measuring tool, resulting in an overestimate of iron concentration in the coal. This can be compensated for in at least four ways. The response of the system to the iron in the tool can be determined in advance. One can use an ash function that does not contain iron as a dependent variable. A detector can be built that does not contain iron. If the tool is made of a stainless steel of known composition, the chromium or nickel photopeaks may be used to apply a correction to the measured iron peaks by subtracting the contribution made to them by the iron in the tool.

<table>
<thead>
<tr>
<th>Element</th>
<th>Concentration (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>C</td>
<td>61.22</td>
</tr>
<tr>
<td>H</td>
<td>6.67</td>
</tr>
<tr>
<td>O</td>
<td>18.84</td>
</tr>
<tr>
<td>S</td>
<td>3.44</td>
</tr>
<tr>
<td>N</td>
<td>1.45</td>
</tr>
<tr>
<td>ash</td>
<td>8.38</td>
</tr>
</tbody>
</table>
3.9. Calcium

Gamma rays from other elements in coal interfere with the most intense thermal capture gamma rays of calcium. The cross-sections for \((n, n'\gamma)\) and \((n, p)\) are only half as great as those for the thermal capture reaction. These reactions produce prompt gamma rays that are free of interference and may be useful in measuring calcium.

3.10. Other reactions

As discussed by Volborth [11], secondary reactions, such as those induced by protons, may be detectable. The proton is produced either as a recoil when hydrogen is struck by a fast neutron or by an \((n, p)\) reaction. Several \((p, \gamma), (p, n)\) and \((p, \alpha)\) reactions are possible, albeit fairly improbable.

4. EXPERIMENTAL WORK

4.1. New equipment

The United States Geological Survey (USGS) and US Bureau of Mines have worked together since 1982 on a project to demonstrate the uses of a 14 MeV neutron generator coupled to a borehole germanium detector for mineral logging. The USGS has outfitted a logging truck for supporting such measurements, and has constructed borehole neutron generators based on the design developed by Sandia National Laboratories [13]. This type of neutron generator uses a sealed 'zetatron' tube that features a Penning-type ion source. Both the ion source and the 110 kV accelerating voltage are pulsed. The full neutron output of this tube is typically \(3 \times 10^8/s\) and the tube life is in excess of 250 hours. The generator, in its pressure housing, is 6 cm in diameter and 150 cm long and uses 125 W of power at full neutron output. The generator is completely self-contained and requires only a source of 500 VDC to produce neutrons.

The logging truck uses a LeCroy 3500M multichannel analyser for data acquisition. The LeCroy analyser supports two CAMAC crates that contain the spectral data processing devices. These include: a 5 \(\mu\)s successive approximation analog-to-digital converter that can simultaneously collect two 8000-channel spectra during different time windows; a time-to-digital converter that can make time-of-arrival histograms; and various programmable timing gates, multichannel scalers and deadtime correction modules.

This equipment is now undergoing laboratory tests before the truck is sent out on field experiments.
4.2. Sample

In order to test the borehole sonde under controlled conditions, a large laboratory coal sample was prepared. The sample consisted of a high-moisture bituminous coal from the Herrin No. 6 seam from Illinois of approximately 'buckwheat' size. A chemical analysis of the sample is given in Table IV. The coal sample is contained in cylindrical steel drums, 150 cm high and 120 cm in diameter. A 10 cm diameter steel pipe completely penetrates the cylinder along its axis to simulate a borehole. The filling ports of the drums are gasketed to provide a hermetic seal.

4.3. Shadow shields

Lead or tungsten shadow shields placed between source and detector are normally used in borehole logging to prevent gamma rays produced by or near the source from reaching the detector. This is particularly important if, as discussed above, a fluorocarbon coolant is used in the neutron generator. If fast neutrons are being used, a shadow shield can also serve to deflect primary neutrons away from the detector to prevent crystal damage. Figure 1 shows the total 14 MeV cross-section of the elements as a function of atomic number as reported in the ENDF/B-V tables [14]. The heavier elements have a higher total cross-section, due to the increase of the elastic scattering cross-section as the size of the nucleus increases. Tungsten is quite effective at deflecting 14 MeV neutrons, the mean free path of a neutron in solid tungsten being about 3 cm. A shadow shield, 10 cm in diameter and 30 cm long, filled with fine tungsten powder, was used in the experiments reported here.

![Graph showing total cross-section of elements for 14 MeV neutrons. Elements with Z > 60 are best for constructing fast-neutron shadow shields [13].](image-url)
4.4. Initial experiments

The neutron generator sonde could be fitted with either a sodium iodide detector or a borehole cryostat containing a high-purity germanium crystal. In each case the charge from the detector was first integrated and then amplified and fed to a cable-driving amplifier. These tail pulses were received and analysed by a Tracor Northern TN1700 multichannel analyser in the logging truck after transmission over approximately 200 m of logging cable.

The generator was configured so that each pulse of neutrons was produced in response to a trigger signal generated in the truck and sent down the cable, superimposed on the 500 VDC that power the generator. This trigger pulse was the time reference used for producing the timing gates used in collecting the spectra.

The neutron output per pulse could be changed by removing the neutron generator from its pressure housing and changing a storage capacitor. Changing the voltage powering the generator would also have caused the neutron output per pulse to change [12]. The 500 volts were also used to power a DC to DC converter that provided the low voltages used by the generator circuitry, and this converter did not tolerate wide variations in the supply voltage.

A proton recoil detector for detecting fast neutrons and a BF$_3$ thermal neutron detector were placed near the gamma-ray detector and were used in setting up the timing windows. The initial response of the gamma-ray detector was found to be coincident with the arrival of fast neutrons at the detector to within the timing accuracy attainable with these detectors ($\pm 1$ $\mu$s).

Three timing windows were used. The first, extending for 10 $\mu$s from the first arrival of fast neutrons, was used to measure the prompt gamma rays from fast-neutron reactions. The second, extending from 0.1 ms to 2 ms, was used to detect prompt gamma rays from thermal-capture events. The third, extending from 4 ms to 9 ms, after most of the thermal neutrons were gone, was used to measure delayed-activation gamma rays.

Measurements during the first window posed considerable experimental difficulties. At a pulse repetition rate of 100 pulses/s, and an average neutron emission rate of $3 \times 10^7$/s, the neutron output during each 5 $\mu$s burst is $3 \times 10^5$, for a peak neutron emission rate within the neutron burst of $6 \times 10^{10}$/s. This is the equivalent of a $^{252}$Cf source of 26 mg, much larger than those normally used for borehole logging. At the source-to-detector distances typically used in logging, the detector would be completely overloaded by pulse pile-up and by the charge released by fast-neutron elastic collisions within the detector crystal. To obtain high-quality spectra within this window, the neutron generator output was reduced by a factor of two and the source-to-detector distance was increased to a distance (about 100 cm) at which the gamma-ray counting rate within the window was 0.08 counts per burst. At this rate there was virtually no pulse pile-up or evidence of high background due to direct fast-neutron interactions.
Figure 2 shows a spectrum made during the fast neutron window, using several large blocks of paraffin wax as a sample. The experiment was done with a 60 cm$^3$ high-purity germanium detector and with a 51 cm diameter $\times$ 51 cm thick NaI(Tl) detector. The dominant peaks seen are the full-energy and escape peaks of the 4439 keV inelastic gamma ray of carbon and the 847 keV and 1238 keV inelastic peaks, due to iron in the sonde. Although there is no oxygen in the wax sample, weak oxygen photopeaks are seen in the spectrum. The oxygen in the concrete floor upon which the wax was resting is undoubtedly the source of these peaks. Because of the Doppler-broadening of the carbon peak, little improvement in measuring this peak is to be gained by using a germanium detector rather than the NaI(Tl) scintillator.

Figures 3a, 3b, and 3c show a similar spectrum made with the germanium detector in the large bituminous coal sample described above. There is essentially no evidence in this spectrum of the presence of thermal neutrons, and the broad carbon and sharp oxygen inelastic photopeaks are quite prominent.

The spectra made during the thermal-capture window are quite similar to those measured using a $^{252}$Cf source, and are not shown here.

Figure 4 shows the region near the oxygen photopeaks in a coal spectrum made during the decay window. The oxygen 6128 keV ($T_{1/2} = 7$ s) decay peaks are quite prominent. The iron capture peaks also seen in this spectrum are due to some thermal neutrons that survive longer than 5 ms.
FIG. 3. High-resolution coal spectra made in a laboratory borehole during a 10 μs window coincident with the burst of 14 MeV neutrons. The pulse repetition rate was 100 pulses/s. (a) Spectrum from 0 to 2.5 MeV, (b) spectrum from 2.5 to 5 MeV, (c) spectrum from 5 to 7.5 MeV.
4.5. Discussion

The clock time required to collect a useful spectrum in the fast-neutron window is exceptionally long. The reason for the long collection time is that at the 100 pps neutron burst repetition rate, the effective gamma ray counting rate was only 8 counts/s. This rate could obviously be increased simply by increasing the neutron burst repetition rate and simultaneously reducing the number of neutrons emitted in each pulse. However, this has the effect of mixing the three kinds of spectra. Even though thermal neutrons are present during the fast-neutron window, only a small number of the gamma rays produced by them are counted in the window because it is narrow in comparison with the repetition period. Because of the short lifetime of fast neutrons, the capture spectrum is always essentially free of prompt lines due to fast neutrons. Decay lines, however, are always present in the capture spectrum. Even at 100 pulses/s the decay spectrum shows evidence of prompt capture lines. At rates of $10^3$ pulses/s and above, the capture and decay spectra are completely mixed. This mixing is particularly detrimental to the decay spectrum, since the photopeaks in that spectrum are often of low energy and intensity and the capture spectrum admixture introduces a strong low-energy Compton background.

A complete ultimate analysis of coal necessarily involves photopeaks measured in spectra made in more than one of these time windows. Counting two or more of these spectra simultaneously poses additional problems. As mentioned above, to collect a spectrum in the fast-neutron window one must use an inordinately low counting rate. The total number of thermal-capture gamma rays produced in the sample during the thermal-capture window is considerably less than the number of gamma rays from fast-neutron reactions during the fast-neutron window.
Thus, the counting rate in the spectra made during both the fast-neutron and thermal-capture windows must necessarily be quite low.

A combination of a fast-neutron spectrum and a thermal-neutron capture spectrum can measure oxygen and carbon by inelastic scattering, with the rest of the elements measured in the thermal-capture spectrum. However, to measure sulphur and nitrogen accurately in a high-resolution thermal-capture spectrum usually requires that the spectrum contain from $1 \times 10^6$ to $4 \times 10^6$ counts. To acquire so many counts at the low counting rates that are required of a simultaneously acquired fast-neutron spectrum calls for an acquisition time that is not practical for field work. If less information is required, such as a measurement of just oxygen, carbon and hydrogen, then the number of counts needed in each spectrum is considerably reduced and the method becomes practical at high neutron pulse repetition rates.

Simultaneously measuring a prompt thermal-capture spectrum and a decay spectrum is much simpler. Here the technique would be to keep the neutron pulse repetition rate low, but to increase the neutron output per pulse or shorten the source-to-detector distance until the counting rate during the capture window approaches pulse pile-up. The detector would be saturated during the fast-neutron window, rendering the fast-neutron spectrum useless, but an adequate capture spectrum could be measured in a practical period of time, while still obtaining a measurement of oxygen via the $(n, p)$ reaction detected during the decay window. In this technique the crystal is exposed to a much higher fluence rate of fast neutrons, but because the time required to acquire a spectrum is less than that for the fast-neutron spectrum as discussed above, the total fast-neutron damage to the crystal would be about the same in each case.

Because sulphur, nitrogen and hydrogen do not have useful decay lines, and because the decays from $\text{Al}(n, \gamma)$ and $\text{Si}(n, p)$ yield gamma rays of the same energy, measuring spectra in the fast-neutron and decay windows does not provide sufficient information to calculate an ultimate analysis.

5. CONCLUSIONS

The use of a 14 MeV neutron generator in borehole logging for coal makes possible a number of fast-neutron reactions that cannot be done with thermal neutrons. Gamma ray spectral information from such reactions can supplement the thermal-capture spectral data normally used for measuring the elemental composition of the coal. With interpulse time gating of the data as they are acquired by a high-resolution borehole detector, separate spectra can be obtained of the gamma rays from fast-neutron, thermal-neutron, and decay events.

A practical method of obtaining an ultimate analysis under field conditions is to use the generator at high neutron output and a pulse repetition rate of about 100 pulses/s and to record two spectra: one made during the thermal neutron capture
window (0.1 to 2 ms after the burst), and the other during the decay window of 5–10 ms. Oxygen can be determined from the latter data and the other elements from the former. Determining how the oxygen data can be used to calculate an accurate ultimate analysis is a subject for further work.

ACKNOWLEDGEMENTS

We thank Asghar Ghias and Clinton Merritt for their assistance in performing the experiments; Eugene Alban for the long hours he spent building hardware components for the neutron generators; Therese Lloyd and Dan Dotson for their help during the construction of the generator; and Franklin Woodbury and Paul Needham for their support. This work was supported in part by the US Bureau of Mines as part of a co-operative project with the US Geological Survey.

REFERENCES

DISCUSSION

T. GOZANI: To my knowledge, the oxygen \((n, n')\) is not Doppler-broadened.

J.L. MIKESSELL: No, the oxygen \((n, n')\) is but the \((n, p)\) is not. The \((n, p)\) is a decay line so if one sees a sharp oxygen 6128 line, that automatically means it was not due to \((n, n')\). All light element \((n, n')\) are Doppler-broadened.

T. GOZANI: I thought that the lifetime of that state was much longer.

J.L. MIKESSELL: The lifetime of the oxygen line, I believe, is a few femtoseconds and the lifetime of the carbon is a few picoseconds. In either case it is not long enough for either nucleus to come to rest after the neutron interaction.

J.S. WATT: Could you comment on what the US coal industry wants for borehole analysis? One thing you could want is elemental analysis, another is straight ash. Thirdly, washability data, which you cannot get by any borehole technique. Do you have any idea of what percentage of holes you would want for elemental analysis, compared with ash?

J.L. MIKESSELL: If private industry has a hard time knowing what the coal industry wants, government agencies have even less chance. I don’t know what the coal industry wants. The work that we do is primarily to support the scientific research of the US Geological Survey.

J.S. WATT: I wonder if anyone from the coal industry of the United Kingdom or the Federal Republic of Germany has a comment on that point.

D. PAGE: I don’t think I can give you proportions but I do know that within the UK coal industry our prime interest in exploration work is to get information on the coal which is beyond what can be obtained from instrumental...
analysis. We are interested in coking properties and washability, as you suggest. Consequently we don’t employ borehole logging at all for coal quality assessment. We rarely use it, except gamma/gamma equipment, for seam delineation. For the most part we use 6-inch drills to recover cores for examination in the laboratory, for other properties than just straight elemental analysis.

C.G. CLAYTON: Every hole which is drilled in the UK has a core. So I think the question was aimed at the wrong country. If we consider developing countries, where there might be no easy access to specialist analytical laboratories then borehole logging becomes much more important.

A.S. PRASAD: In India logging is a problem in the sense that we have to know the exact identity of the coal at different places because there are two natures of coal. We have non-coking coal in abundance and we have very little coking coal. So unless we know whether the coal at the site is coking or non-coking we cannot develop it further; it becomes uneconomical. Logging is necessary, for two reasons; we want to know the ash content of the coal and whether it is coking or non-coking.

J. CHARBUCINSKI: What is the normal procedure in the case of a seam which has a thickness of the same order as your source-to-detector distance or shorter? The shape of anomaly which you get from such seams depends mainly on the position of your point-by-point logging. Did you try a deconvolution technique to get an anomaly which would represent such a narrow seam? And in such cases how narrow are the seams you are able to analyse?

J.L. MIKESELL: The particular case shown was done point by point, but our present analyses are done either continuously or over much shorter intervals. We have found that the response of the detection system is correlated with the position of the detector, and that at the relatively large source-to-detector distances that we typically use, 30 cm to 1 m, the neutron distribution is fairly uniform at the detector, so it is mostly the detector position. But once you know where the centre is, you have to ask what is the spatial resolution, and this of course is limited by the technique itself. That is, you are trying to measure gamma rays that have a mean free path in coal, and I would expect that the spatial resolution is not ever going to be better than perhaps 10-15 cm, simply because gamma rays travel so far through coal.
IN SITU BOREHOLE DETERMINATION OF ASH CONTENT OF COAL USING GAMMA-GAMMA AND NEUTRON-GAMMA TECHNIQUES

J. CHARBUCINSKI, M. BORSARU, P.L. EISLER, S.F. YOUL
Commonwealth Scientific and Industrial Research Organization,
Division of Mineral Physics,
Port Melbourne, Australia

Abstract

IN SITU BOREHOLE DETERMINATION OF ASH CONTENT OF COAL USING GAMMA-GAMMA AND NEUTRON-GAMMA TECHNIQUES.

During the past decade, borehole logging technology based on nuclear geophysics has found wide application in the Australian coal-mining industry. In response to the need for further improved accuracy in coal ash measurements, the Commonwealth Scientific and Industrial Research Organization, Division of Mineral Physics, has developed two new alternative techniques, which are both spectrometric. The spectrometric gamma-gamma technique is based on the existence of a simple correlation between the ash content and the equivalent atomic number and density of coal. The technique is spectrometric in that it records and uses the count rates in several windows of the backscatter spectrum. These count rates and their selected ratios describe the changes in spectral shape which are due to ash content variations. The spectrometric neutron-gamma method is suitable where probe responses are required for specific elemental contents. Consequently, the method tolerates larger variations in ash composition for accurate measurement than does the gamma-gamma method. Both methods have been tested at several coal deposits in New South Wales and Queensland. For both techniques, RMS deviations between nuclear assay and chemical analysis are typically 2% ash in the range 5 to 40% ash. Both techniques are currently undergoing commercial development under the name of SIROLOG. The SIROLOG technology is designed to accommodate logging speeds up to 4 m·min⁻¹. However, the gamma-gamma probe uses gamma-ray sources of strength two orders of magnitude smaller than that of sources used in commercial probes. The logging system provides information on ash content in 5 cm intervals if required, although the vertical resolution of the probes is 30–35 cm.

1. INTRODUCTION

The economic value of coal deposits, the mine planning and the production schedules are determined by a number of factors. These include several geological parameters which can be determined by borehole logging techniques, namely, the thickness of the coal seams, their depth below the surface and the ash content in the coal. The above first two parameters are simply and accurately determined
using borehole logging technology based on non-spectrometric natural gamma and gamma-gamma methods. However, the borehole logging methods routinely used for ash determination generally have insufficient accuracy to satisfy the industry’s requirements for mine development and production.

Currently, in situ raw coal determinations depend on the existence of a strong correlation between the gamma-gamma density log response and the coal ash content. However, this correlation is not always high. Significant variations are reported in the relationship between density and ash content for coal deposits in Australia [1], South Africa [2], North America [3] and England [4]. The results shown in Fig. 1 of an analysis of correlation between ash content and density, which was carried out with the core data from the English [4] coal deposit, gave an RMS deviation of 3.1% ash. Consequently, this value represents a threshold error for ash content determinations using ‘density’ gamma-gamma logging measurements for that deposit.

Current commercial borehole logging technology for coal ash determination based on the density gamma-gamma method has the following shortcomings:

![Figure 1: Correlation between specific gravity and ash content in cores obtained from an English coal deposit.](image)
(a) The relationship between ash content and coal density is not universal. It varies for different coal deposits and, to a lesser extent, for different seams within the same deposit.

(b) Measurements carried out in strata of brecciated or fractured coal will have reduced accuracy; they will indicate the existence of a higher quality coal than is actually the case.

(c) Because the eccentralized probes, in commercial practice use small detectors (rarely greater than 25 mm in diameter) and strong collimation (required with side-wall operation), intense gamma-ray sources (~2 to ~5.5 GBq) are generally required. This necessitates stringent procedures for radiation safety of personnel.

(d) The gamma-gamma probes used routinely provide only non-spectrometric information. Consequently, the ash content can be correlated with only a single probe parameter.

The prompt neutron-gamma method provides an alternative borehole logging technique for ash determination. This method is less sensitive than the gamma-gamma method to either borehole rugosity or the difficulties of correlating probe response with ash content. Nargolwalla et al. [5] and Senftle [6] described the application of this method to borehole logging for coal. While several recent papers [7-10] reported the potential of the spectrometric neutron-gamma borehole logging method for coal ash measurements, the routine use of the method in this application has not been reported elsewhere.

The neutron capture process produces photons with energies ranging up to 10 MeV. In fact, most of the gamma rays that characterize the constituents of ash in coal have energies greater than 3 MeV. This contrasts with photons measured in gamma-gamma techniques where photon energies are largely below 1 MeV. The greater penetration of the neutron capture photons enables the neutron-gamma logging method to be used to sample larger volumes in the coal strata than the gamma-gamma methods. It also renders the neutron-gamma method the least sensitive to diameter variations in fluid-filled boreholes.

Another important advantage of the neutron-gamma method is that it is direct. It is direct in the sense that quantitative information on ash content is based on the individual interactions between the thermalized neutrons and nuclei of the major ash constituents (i.e. Si, Al, Fe). Consequently, the neutron-gamma method provides scope to determine the contents of silica, alumina and iron in the ash. The ash content can be determined from the above information because the correlation between total raw ash in coal and the sum of the contents of SiO$_2$, Al$_2$O$_3$ and Fe$_2$O$_3$ is very high. A crossplot of total raw ash content in coal against the combined contents of SiO$_2$, Al$_2$O$_3$ and Fe$_2$O$_3$ is shown in Fig. 2 for 174 Australian coals located in different seams and deposits. The RMS deviation of 0.67% for this correlation represents the threshold error for a method of ash determination based on an accurate knowledge of SiO$_2$, Fe$_2$O$_3$ and Al$_2$O$_3$ contents.
FIG. 2. Correlation between combined contents of Al, Si and Fe in coal and ash content. (174 coals from various Australian coal deposits.)

2. GAMMA-GAMMA METHOD

The spectrometric gamma-gamma technique is a suitable alternative to the 'density' gamma-gamma method, because it is inherently less sensitive to fluctuations in the physical properties of the coal.

The relationship between the ash content and the chemical composition of coal (expressed in $Z_{eq}$ units and an average $Z/A$) forms the basis for this technique. The coal ash content varies, not only with density, but also with the average chemical composition of the coal. A higher ash content, caused by increased concentrations of Si, Al, Fe and Ca (which have higher Z than carbon) produces a higher $Z_{eq}$ for the coal.
For a unique relationship between ash content and $Z_{eq}$, it should be possible to determine ash content by measuring changes in $Z_{eq}$. However, because Fe, Ti and Ca have a greater influence on $Z_{eq}$ than the major elemental ash constituents, Si and Al, some significant variation is anticipated in the correlation between ash content and $Z_{eq}$ of coal (adjusted for the average $Z/A$) for a wide range of deposits. Figure 3 shows the crossplot between ash content and values of $Z_{eq}$ and $\beta = (Z/A)_{\text{average}}$ for the above 174 Australian coals.

A comparison of the crossplots in Figs 2 and 3 indicates that the neutron-gamma method has the more general application geographically, and that the gamma-gamma method should be used regionally where coal ash composition is relatively uniform.

The methods most commonly used to measure the changes of $Z_{eq}$ are the measurement of spectral intensity at low energy and the measurement of the
P₂ factor, which is the ratio between count rates recorded in two broad spectral windows at high and low energies. However, the successful application of these methods for ash determination depends on the uniformity of the chemical composition of the ash. Consequently, if Fe, Ti and Ca concentrations vary considerably in the ash of samples from different areas of a deposit, a gamma-gamma logging method is desired in which the sensitivity of the response to the heavy element concentration is lower than either P₂ or the spectral intensity at low energies. This method is based on measurements of the shape of backscattered gamma-ray spectra [11, 12]. The parameters of spectral shape, which form the basis of simple linear models for predicting ash content [13], are the co-ordinates for the maximum of the spectrum and the angles corresponding to the spectral slopes at low and high energies (i.e. Eₐ, Iₐ, θ, P₂). These spectral shape parameters are illustrated in Fig. 4. In fact, the P₂ ratios are also spectral shape parameters because they describe the variations between the high- and low-energy parts of the spectrum.

The gamma-gamma method based on the utilization of different spectral shape parameters is referred to below as the spectrometric gamma-gamma method to differentiate it from non-spectrometric, 'density' gamma-gamma methods.

In order to develop the spectrometric gamma-gamma method, two sets of borehole logging tests were carried out in the laboratory. The purpose of the first set of measurements was to determine the optimal probe configuration. This configuration provided the best sensitivity for measuring the coal ash contents that was compatible with adequate spatial resolution. For these tests, probe separations of 19, 29 and 39 cm were considered. The angles of source collimation, which were varied, were 90, 65, 25 and 15° to the longitudinal axis of the probe.

The measurements were carried out in seven borehole models, each containing about 300 L of crushed coal. These coal samples were blended from two larger
samples having ash contents of 55 and 10%. As a consequence of the blending procedures used, the chemical composition of the ash in the models varied only slightly.

The regression analyses of tests carried out indicated that the smallest values of RMS deviation (expressed as % ash) were obtained under the most rigid source-collimation conditions. While the 39 cm probe separation gave the highest sensitivity for measuring ash content, the probe configuration that was considered optimal, and that was chosen for the second test, used a probe separation of 29 cm and a source collimation angle of 90°. This probe separation was a compromise, chosen to give adequate bed resolution. In fact, where criteria for probe design are governed by stringent requirements of bed resolution, the choice for probe separation is 19 cm.

The aim of the second set of laboratory tests was to evaluate the performance of different spectrometric gamma-gamma methods for ash determination in coal samples which varied considerably in ash composition. Forty samples of natural, unblended coals were used for the tests. There were two groups of samples differing significantly, mainly as regards the level of Fe content in the ash.

The first group of samples represented coals with fairly low and uniform iron content in the ash. The mean Fe content in the ash was 4.2% and the standard deviation was 1.2% Fe. The first group were obtained from the collieries of Vales Point and Liddell, denoted by VP & L.

The second group of coal samples, obtained from the Utah-Blackwater mine, contained significantly more Fe in the ash than the VP & L samples. The variation of Fe content in the ash was also much greater in the Utah samples. The mean value of the Fe in the ash was 9.0% and the standard deviation was 3.9% Fe for this second group of samples.

Although the ratio of events which are due to photo-electric and Compton interactions in the backscattered gamma ray spectrum is governed by the initial photon energy, the accuracy for determining ash content is also dependent on the type of gamma-ray source used. Consequently, ¹³⁷Cs, ¹³³Ba and ⁷⁵Se were tested for their suitability as primary radiation sources.

However, the prime aim of the measurements was to select the most suitable spectral parameters and the regression model for coal ash measurement. Table I shows a selection of results from relevant regression analyses. These were carried out with the data from measurements of the two groups of samples described above. In order to compare the suitability of the different spectral parameters for probe calibration, the values of correlation coefficient, \( R_c \), and RMS deviation, \( \sigma \), were used as the criteria.

One matter, clearly indicated by the data in the table, is that ⁷⁵Se is the least suitable source for ash determination for coals varying significantly in chemical composition. The reason for this is that the absorption of the low-energy gamma rays emitted by this source is highly sensitive to the concentration of Fe and other heavy elements.
TABLE I. SUMMARY OF RESULTS OBTAINED FROM THE LABORATORY TESTS OF THE SPECTROMETRIC GAMMA-GAMMA METHOD

<table>
<thead>
<tr>
<th>Response variable</th>
<th>$^{75}$Se</th>
<th></th>
<th></th>
<th>$^{133}$Ba</th>
<th></th>
<th></th>
<th>$^{137}$Cs</th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Utah</td>
<td>VP &amp; L</td>
<td></td>
<td>Utah</td>
<td>VP &amp; L</td>
<td></td>
<td>Utah</td>
<td>VP &amp; L</td>
<td></td>
</tr>
<tr>
<td></td>
<td>$R_c$</td>
<td>$\sigma$</td>
<td>$R_c$</td>
<td>$\sigma$</td>
<td>$R_c$</td>
<td>$\sigma$</td>
<td>$R_c$</td>
<td>$\sigma$</td>
<td>$R_c$</td>
</tr>
<tr>
<td>$E_a$</td>
<td>0.894</td>
<td>3.3</td>
<td>0.967</td>
<td>1.2</td>
<td>0.919</td>
<td>3.0</td>
<td>0.963</td>
<td>1.3</td>
<td>0.900</td>
</tr>
<tr>
<td>$I_a$</td>
<td>0.911</td>
<td>3.0</td>
<td>0.898</td>
<td>2.2</td>
<td>0.949</td>
<td>2.4</td>
<td>0.900</td>
<td>2.1</td>
<td>0.951</td>
</tr>
<tr>
<td>$\theta_1$</td>
<td>0.919</td>
<td>2.9</td>
<td>0.893</td>
<td>2.2</td>
<td>0.955</td>
<td>2.3</td>
<td>0.903</td>
<td>2.1</td>
<td>0.887</td>
</tr>
<tr>
<td>$\theta_2$</td>
<td>0.915</td>
<td>3.0</td>
<td>0.885</td>
<td>2.3</td>
<td>0.954</td>
<td>2.3</td>
<td>0.904</td>
<td>2.1</td>
<td>0.914</td>
</tr>
<tr>
<td>$P_1 = 60-250/6-59$</td>
<td>0.901</td>
<td>3.2</td>
<td>0.963</td>
<td>1.3</td>
<td>0.914</td>
<td>3.1</td>
<td>0.914</td>
<td>2.0</td>
<td>0.909</td>
</tr>
<tr>
<td>$P_2 = 100-250/6-99$</td>
<td>0.910</td>
<td>3.0</td>
<td>0.964</td>
<td>1.3</td>
<td>0.926</td>
<td>2.9</td>
<td>0.926</td>
<td>1.8</td>
<td>0.919</td>
</tr>
<tr>
<td>$P_3 = 45-100/6-40$</td>
<td>0.893</td>
<td>3.3</td>
<td>0.963</td>
<td>1.3</td>
<td>0.913</td>
<td>3.1</td>
<td>0.959</td>
<td>1.4</td>
<td>0.905</td>
</tr>
<tr>
<td>$P_4 = 70-110/6-40$</td>
<td>0.888</td>
<td>3.4</td>
<td>0.963</td>
<td>1.3</td>
<td>0.907</td>
<td>3.2</td>
<td>0.954</td>
<td>1.5</td>
<td>0.899</td>
</tr>
<tr>
<td>$P_5 = 35-80/6-34$</td>
<td>0.890</td>
<td>3.3</td>
<td>0.966</td>
<td>1.3</td>
<td>0.915</td>
<td>3.1</td>
<td>0.965</td>
<td>1.3</td>
<td>0.905</td>
</tr>
<tr>
<td>ROI 1 = 6-30</td>
<td>0.884</td>
<td>3.4</td>
<td>0.944</td>
<td>1.6</td>
<td>0.927</td>
<td>2.8</td>
<td>0.945</td>
<td>1.6</td>
<td>0.933</td>
</tr>
<tr>
<td>ROI 2 = 31-50</td>
<td>0.914</td>
<td>3.0</td>
<td>0.909</td>
<td>2.6</td>
<td>0.949</td>
<td>2.4</td>
<td>0.913</td>
<td>2.0</td>
<td>0.945</td>
</tr>
<tr>
<td>ROI 3 = 100-250</td>
<td>0.894</td>
<td>3.3</td>
<td>0.723</td>
<td>3.4</td>
<td>0.943</td>
<td>2.5</td>
<td>0.742</td>
<td>3.3</td>
<td>0.931</td>
</tr>
<tr>
<td>ROI 4 = 150-250</td>
<td>0.716</td>
<td>5.1</td>
<td>0.704</td>
<td>3.5</td>
<td>0.917</td>
<td>3.0</td>
<td>0.769</td>
<td>3.1</td>
<td>0.905</td>
</tr>
</tbody>
</table>

Above limits shown in channels. Energy calibration: 1.5 keV/channel.
Table I also indicates that $^{137}$Cs and $^{133}$Ba are comparably suitable primary radiation sources.

The smallest RMS deviations between the nuclear and chemical assays (referred to below as accuracies) were about 1.4% ash for the VP & L samples and about 2.3 to 2.4% ash for the Utah samples. We selected $^{137}$Cs as the radiation source for field applications because of its longer half-life and the greater penetration of its emitted photons.

While Table I indicates that $P_2$ and the selected spectral intensity parameters gave the best accuracy for measuring ash content where the ash is of relatively uniform chemical composition, it also shows that $I_a$ was the best parameter for samples varying widely in the heavy element concentration of the ash. This suggests that the spectral parameter based directly on spectrum shape ($I_a$) is less dependent on variations in ash composition, and should be used for those conditions.

The spectrometric gamma-gamma method was tested at several coal deposits in New South Wales and Queensland. The tests were carried out in both open and cored holes, and also in water-filled and dry holes. The diameter of holes tested varied between 100 and 160 mm.

Table II shows the details of the borehole characteristics, the types of borehole logging probes used, the number of independent data points, the range of ash content and the standard deviation of the ash. The table is relevant to both the gamma-gamma and neutron-gamma methods tested.
<table>
<thead>
<tr>
<th>Logging site</th>
<th>Type of holes</th>
<th>Water-filled holes or dry</th>
<th>Number of data points</th>
<th>Range of ash (%)</th>
<th>Standard deviation of ash (%)</th>
<th>Type of gamma-gamma probe used (source, std^a distance)</th>
<th>Type of neutron-gamma probe used (detector, std^a distance)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1. Queensland 1</td>
<td>open</td>
<td>dry</td>
<td>22</td>
<td>7.0–38.1</td>
<td>5.6</td>
<td>Cs, 29 cm</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>water-filled</td>
<td>35</td>
<td>7.0–28.1</td>
<td>5.4</td>
<td>Cs, 29 cm</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>water-filled</td>
<td>25</td>
<td>7.0–24.8</td>
<td>3.8</td>
<td>Cs, 29 cm</td>
<td>BGO, 23 cm</td>
</tr>
<tr>
<td>2. German Creek &amp; Gregory South</td>
<td>cored</td>
<td>water-filled</td>
<td>26</td>
<td>7.2–33.0</td>
<td>7.7</td>
<td>Cs, 19 cm</td>
<td>BGO, 15 cm</td>
</tr>
<tr>
<td>3. Middlemount &amp; Lake Lindsay</td>
<td>cored</td>
<td>water-filled</td>
<td>28</td>
<td>11.2–44.7</td>
<td>12.2</td>
<td>Cs, 19 cm</td>
<td>BGO, 15 cm</td>
</tr>
<tr>
<td>4. Singleton</td>
<td>cored</td>
<td>water-filled</td>
<td>30</td>
<td>7.7–50.3</td>
<td>12.1</td>
<td>Cs, 19 cm</td>
<td>BGO, 15 cm</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Cs, 19 cm</td>
<td>BGO, 23 cm</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Co, 19 cm</td>
<td>BGO, 33 cm</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Co, 29 cm</td>
<td>Nal(Tl), 19 cm</td>
</tr>
<tr>
<td>5. Blackwater</td>
<td>cored</td>
<td>dry</td>
<td>35</td>
<td>4.2–38.1</td>
<td>7.5</td>
<td>Cs, 29 cm</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Co, 19 cm</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>23</td>
<td>4.1–37.7</td>
<td>8.5</td>
<td>Co, 29 cm</td>
<td></td>
</tr>
<tr>
<td></td>
<td>and</td>
<td>(new &amp; old)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>open</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>6. Saxonvale</td>
<td>cored</td>
<td>water-filled</td>
<td>19</td>
<td>15.9–47.5</td>
<td>10.4</td>
<td>Nal(Tl), 18 cm</td>
<td></td>
</tr>
<tr>
<td>7. Queensland 1 &amp; Queensland 2</td>
<td>cored</td>
<td>water-filled</td>
<td>35</td>
<td>7.0–31.0</td>
<td>7.3</td>
<td>BGO, 23 cm</td>
<td></td>
</tr>
</tbody>
</table>

^a^ std = source-to-detector.
CREATE LITHOLOGICAL CHARTS FROM RAW DATA (5 CM SPLITS)

DETERMINE SEAM BOUNDARIES FROM THE CHARTS

JOIN RAW DATA INTO ONE SPECTRUM CORRESPONDING TO THE DETERMINED SEAM BOUNDARIES

CREATE SPECTRAL WINDOWS (NORMALIZED TO LIFE TIME) AND SPECTRAL RATIOS FOR PRESELECTED WINDOWS

DO A SUBSET ANALYSIS TO SELECT THE BEST ONE, TWO AND THREE VARIABLE MODELS

ANALYSE THESE MODELS FOR:
  PHYSICAL MEANING,
  SENSITIVITY
  STATISTICAL SIGNIFICANCE

CHOOSE A MODEL TO BE USED FOR CALIBRATION

FIG. 6. System of data analysis.

A schematic diagram of the borehole logging system is shown in Fig. 5.

The borehole logging probe was of a modular construction, which permitted changes of source, source collimators and source-to-detector distances. The probe was fabricated from aluminium tubing of 60 mm o.d. and wall thickness 3.2 mm. The electronics module comprised a 76 X 38 mm NaI(Tl) scintillation detector of integral construction and premium grade, a high-voltage power supply, an AC coupled charge-sensitive preamplifier and a cable driver of low impedance. The preamplifier was capable of driving, with negligible loss of energy resolution, a 300 m cable link from the probe to a spectral analysis system housed in the logging truck (Ford F100). All measurements were conducted with the probe located centrally in the borehole. Two flat, star-shaped centralizers fabricated from a flexible, heavy-duty plastic, Lurethane, were used satisfactorily for centralization.
### TABLE III. SUMMARY OF ASH DETERMINATIONS USING THE SPECTROMETRIC GAMMA-GAMMA METHOD

<table>
<thead>
<tr>
<th>Location</th>
<th>RMS deviation (% ash)</th>
<th>Type of source and source-to-detector distance</th>
<th>Number of variables in regression equation</th>
</tr>
</thead>
<tbody>
<tr>
<td>1. Queensland:</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>dry holes</td>
<td>2.1</td>
<td>Cs, 29 cm</td>
<td>1</td>
</tr>
<tr>
<td>water-filled holes</td>
<td>2.6</td>
<td>Cs, 29 cm</td>
<td>1</td>
</tr>
<tr>
<td>2. Singleton</td>
<td>1.8</td>
<td>Cs, 19 cm</td>
<td>2</td>
</tr>
<tr>
<td>3. German Creek &amp;</td>
<td>2.5</td>
<td>Cs, 19 cm</td>
<td>3</td>
</tr>
<tr>
<td>Gregory South</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>4. Middlemount &amp;</td>
<td>2.0</td>
<td>Cs, 19 cm</td>
<td>3</td>
</tr>
<tr>
<td>Lake Lindsay</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>5. Blackwater:</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>all holes</td>
<td>2.0</td>
<td>Co, 29 cm</td>
<td>3</td>
</tr>
<tr>
<td>new holes</td>
<td>1.7</td>
<td>Co, 29 cm</td>
<td>2</td>
</tr>
</tbody>
</table>

**FIG. 7. Comparison of coal ash content determined by chemical analysis and the spectrometric gamma-gamma method. Coal deposit in Hunter Valley area.**

\[ R_c = 0.988 \]
\[ \text{s.d.} = 1.81 \]
The logging operations were carried out with the probe moving continuously in the borehole. The full energy spectra accumulated along each 5 cm of a borehole were transferred to a microcomputer and stored on hard disc. These spectra represented the raw data for further analyses. Figure 6 shows schematically the system of data analysis used, not only for the gamma-gamma method, but also for neutron capture logging.

Table III summarizes the best results obtained during the field tests of the spectrometric gamma-gamma method. It shows the accuracies obtained for ash determination, together with the logging probe characteristics and the regression models used.

It is important to note that the RMS values shown in Table III intrinsically contain the errors of both sampling and laboratory assaying. Therefore, the error of the radiometric method is always smaller than the RMS deviation shown in the table.

Figure 7 shows the crossplot of assays obtained by the spectrometric gamma-gamma method against chemical assays for a coal deposit in the Singleton area.

Figure 8 shows the results of 'density' gamma-gamma borehole logging measurements carried out in the same boreholes of the deposit used for Fig. 7,
### TABLE IV. ACCURACY OF ASH MEASUREMENTS OBTAINED FROM THE FIELD TEST OF THE GAMMA-GAMMA METHOD IN BLACKWATER

<table>
<thead>
<tr>
<th>Regression model</th>
<th>Cored holes (all)</th>
<th>Open holes (all)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$^{137}$Cs</td>
<td>$^{60}$Co</td>
</tr>
<tr>
<td>1 variable</td>
<td>3.7</td>
<td>2.4</td>
</tr>
<tr>
<td>2 variables</td>
<td>2.4</td>
<td>2.1</td>
</tr>
<tr>
<td>3 variables</td>
<td>2.2</td>
<td>2.0</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th></th>
<th>Cored holes (new)</th>
<th>Open holes (new)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$^{137}$Cs</td>
<td>$^{60}$Co</td>
</tr>
<tr>
<td>1 variable</td>
<td>2.3</td>
<td>1.7</td>
</tr>
<tr>
<td>2 variables</td>
<td>1.8</td>
<td>1.7</td>
</tr>
<tr>
<td>3 variables</td>
<td>1.7</td>
<td>1.3</td>
</tr>
</tbody>
</table>

The values in the above Table represent the RMS deviations between gamma-gamma results and laboratory ash determinations, expressed as % ash.

and using the same equipment as for the spectrometric measurements. A comparison of Figs 7 and 8 clearly indicates the superiority of the spectrometric gamma-gamma method.

At all but one of the deposits (Blackwater), chemical assaying of the core samples used for calibration preceded the borehole logging trials by a considerable period (1 to 5 years). Consequently, the core samples taken for ash analysis were not always of the same thickness as the geophysical thickness of the corresponding coal seams. The reason for this is that routine geological sampling is usually carried out to provide an ash value for a seam which can be economically mined. Coal strata which might be economically mined may be selected, depending on a number of factors, either to include non-coal material adjacent to the seam proper, or to exclude splits of low-grade coals. During field trials at Blackwater, several holes used were specially drilled to coincide with the test programme. The bore cores obtained from these holes were then sampled on the basis of the geophysical profiles obtained from the geophysical logs. The results of regression analyses using the data from only the freshly drilled holes (i.e. new holes) showed significantly better accuracy than the calibrations based on data from all the holes. The values of accuracy shown in Table IV, which summarize the performance of different probes using $^{60}$Co and $^{137}$Cs in various regression
models, support the thesis that when boreholes are sampled on the basis of the profiles of the geophysical logs, the goodness of fit is greatly improved, irrespective of the regression model used for calibration.

3. NEUTRON-GAMMA METHOD

The interactions between neutrons and nuclei give rise to a number of nuclear reactions. The occurrence of particular reactions depends on the neutron energy and the nuclei involved. The energies of the gamma rays emitted by the nuclei following the neutron reactions characterize the nuclei and the nuclear reactions which occur. On this basis, the occurrence of spectral peaks in particular energy windows of a multichannel pulse height analyser indicates the nature of the chemical elements involved in the reaction. If the thermal neutron flux is constant, then, for a fixed probe configuration, the intensity of the radiation (specific to a particular element and recorded in an energy window encompassing the spectral peak characteristic of that element) is proportional to the number of nuclei of that element in the volume unit of the coal (or rock) being logged. This prompt neutron-gamma method will also be referred to as the neutron capture method.

The neutron capture method \((n, \gamma)\) is based on thermal neutron reactions which have large cross-sections for many elements. Because coal has a high hydrogen content, it is an excellent medium for the neutron capture technique. The neutrons emitted by the neutron source are rapidly thermalized in their collisions with the hydrogen nuclei and are subsequently captured by the various nuclei in the coal. Table V gives the neutron capture gamma-ray lines for the principal chemical elements in coal ash.

The probe response for capture gamma rays emitted with energy \(i\), after a collision with nuclei \(j\), is written [9]:

\[
R_{ij} = \int \int_{\text{En} \text{ V}} \phi(E_n, \vec{r}) \cdot \sigma_{ij}(E_n) \cdot \nu_j(\vec{r}) \cdot G(E_{\gamma}, \vec{r}) \cdot dE_n \cdot dV
\]

where \(R_{ij}\) is the intensity of radiation in the gamma-ray peak of energy \(i\), \(\sigma_{ij}(E_n)\) is the gamma-ray production cross-section, \(E_n\) is the neutron energy, \(\vec{r}\) is the radius vector with reference to the detector, \(\phi(E_n \vec{r})\) is the neutron flux at \(\vec{r}\) of energy \(E_n\), \(\nu_j\) is the number of nuclei of element \(j\) per unit volume at \(\vec{r}\), \(G(E_{\gamma}, \vec{r})\) is the detection probability for a gamma ray of energy \(E_{\gamma}\) at point \(\vec{r}\) and \(V\) is the volume of coal in which neutron capture occurs. In practice, the determination of the elemental composition of coal is complicated because the space-energy distribution of the neutron flux is perturbed by the inherent variations in bulk density, hydrogen content and elemental composition of the coal. It is also perturbed by the variations in the physical condition of the borehole.
TABLE V. NEUTRON CAPTURE PARAMETERS FOR ALUMINIUM, SILICON, IRON AND TITANIUM

<table>
<thead>
<tr>
<th>Element</th>
<th>Cross-section $\sigma$ (barn)</th>
<th>Major gamma rays (MeV)</th>
<th>Intensity (I) (photons/100 neutrons)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Aluminium</td>
<td>0.23</td>
<td>7.72</td>
<td>20.10</td>
</tr>
<tr>
<td>(26.98)</td>
<td></td>
<td>7.69</td>
<td>4.17</td>
</tr>
<tr>
<td>Silicon</td>
<td>0.16</td>
<td>7.20</td>
<td>7.2</td>
</tr>
<tr>
<td>(28.09)</td>
<td></td>
<td>6.38</td>
<td>12.6</td>
</tr>
<tr>
<td></td>
<td></td>
<td>4.93</td>
<td>70.5</td>
</tr>
<tr>
<td></td>
<td></td>
<td>3.54</td>
<td>79.6</td>
</tr>
<tr>
<td>Iron</td>
<td>2.62</td>
<td>7.65</td>
<td>22.1</td>
</tr>
<tr>
<td>(55.85)</td>
<td></td>
<td>7.63</td>
<td>27.2</td>
</tr>
<tr>
<td>Titanium</td>
<td>6.1</td>
<td>1.38</td>
<td>65.5</td>
</tr>
<tr>
<td>(47.90)</td>
<td></td>
<td>4.88</td>
<td>5.7</td>
</tr>
<tr>
<td></td>
<td></td>
<td>6.42</td>
<td>36.5</td>
</tr>
<tr>
<td></td>
<td></td>
<td>6.56</td>
<td>6.5</td>
</tr>
<tr>
<td></td>
<td></td>
<td>6.76</td>
<td>54.0</td>
</tr>
</tbody>
</table>

The accuracy of determination of elemental concentrations can be improved by normalizing the peak intensity to the intensity of the 2.22 MeV hydrogen peak, which is proportional to the neutron flux.

The most common isotopic neutron sources used in borehole logging are Am-Be and $^{252}$Cf. The fission neutron spectrum of $^{252}$Cf has a mean energy of about 2.2 MeV, which is appreciably lower than the energy of neutrons emitted by Am-Be source. The neutrons from $^{252}$Cf have energies too low sufficiently to excite 4.43 MeV gamma rays from the inelastic scattering reaction with carbon to interfere with the 4.93 MeV gamma rays arising from silicon capture. Therefore, $^{252}$Cf is the more suitable neutron source for neutron capture logging.

For coal ash content determinations, scintillation detectors are preferred to solid-state detectors. The high-resolution, solid-state Ge detector is being used for such applications where detailed elemental analysis is required. However, an important drawback of this detector is its low efficiency for high-energy gamma rays. This is particularly important with neutron capture reactions where the gamma rays released as an effect of the capture process have energies up to 10 MeV (see Table V). Another disadvantage of the Ge detector for neutron-gamma logging is that it is prone to neutron radiation damage. This places
constraints not only on the choice of neutron source strength but also on the probe design with regard to the source-to-detector distance. The shorter source-to-detector distance which is desirable for better vertical resolution in the borehole cannot be used because of the increased rate of detector damage. The third drawback in the borehole application of a Ge detector is that it operates at cryogenic temperature and consequently needs a stable supply of liquid nitrogen.

NaI(Tl) detectors have the widest usage in borehole logging applications. Although the energy resolution is almost two orders of magnitude broader than for the Ge detector, the NaI(Tl) scintillation detector has the following important advantages:

(1) Operation is at normal ambient temperature;
(2) Because the NaI(Tl) scintillators are commonly grown as crystals of much larger volume than Ge, they have by far the greater efficiency for detection of gamma rays;
(3) NaI(Tl), like other scintillators, is not prone to radiation damage.

An alternative scintillation detector, CsI(Na), has a higher density (4.51 g·cm⁻³) than NaI(Tl) (3.67 g·cm⁻³), and is therefore a more efficient detector of high-energy gamma rays. On the other hand, its energy resolution is inferior even to that of NaI(Tl).

A recently developed scintillator, bismuth germanate (Bi₄Ge₃O₁₂), which is commonly referred to as BGO, has both a high density (7.1 g·cm⁻³) and very high effective atomic number, Zₑq (70). Consequently its detection efficiency, especially for high-energy gamma rays, is greater than for either NaI(Tl) or CsI(Na) [14]. Further, it has two other advantageous characteristics for borehole logging, compared with other scintillators; it is mechanically rugged and not hydroscopic. The disadvantageous feature of this detector is that the energy resolution is much broader than that of either NaI(Tl) or CsI(Na).

The borehole logging tests of the neutron capture method were carried out using the same borehole logging instrumentation as shown in Fig. 5. Two types of scintillation detectors were used: 76 × 38 mm NaI(Tl) and 51 × 51 mm BGO. Both crystals were covered by a thin cylindrical aluminium housing coated with a thin layer (~16 mg·cm⁻²) of.SimpleDateFormat

The hydrogen capture gamma-ray peak (2.22 MeV) was used for monitoring the thermal neutron flux in the vicinity of the detector.

Three different probe configurations were tested for their suitability in quantitative ash analysis. The source-to-detector distances used in the trials were 15, 23 and 33 cm. Consequently, to maintain sufficient counting statistics it was necessary to use ²⁵²Cf sources of different strengths: 0.7, 2.5 and 7.0 μg. Bismuth shielding was placed between source and detector. The hydrogen capture gamma-ray peak (2.22 MeV) was used for energy stabilization during logging.
In field trials, the applications of the neutron-gamma method were tested both for the delineation of coal seams and plies, and for the determination of ash content in coal.

The position and thickness of coal plies can be determined by monitoring the spectral response of the probe to the Si, Al, Fe and Ti constituents in both the coal and the interply and interseam sediments. The energy window from 3 MeV to 10 MeV was selected for monitoring the stratigraphy. Figure 9
TABLE VI. SUMMARY OF ASH DETERMINATIONS USING THE SPECTROMETRIC NEUTRON-GAMMA METHOD

<table>
<thead>
<tr>
<th>Location</th>
<th>RMS deviation (% ash)</th>
<th>Type of detector and std\textsuperscript{a} distance</th>
<th>Number of variables in regression equation</th>
</tr>
</thead>
<tbody>
<tr>
<td>1. Queensland 1</td>
<td>1.4</td>
<td>BGO, 23 cm</td>
<td>3</td>
</tr>
<tr>
<td>2. Queensland 1 &amp; Queensland 2</td>
<td>2.8</td>
<td>BGO, 23 cm</td>
<td>3</td>
</tr>
<tr>
<td>3. Saxonvale</td>
<td>2.5</td>
<td>NaI(Tl), 18 cm</td>
<td>2</td>
</tr>
<tr>
<td>4. German Creek &amp; Gregory South</td>
<td>1.7</td>
<td>BGO, 15 cm</td>
<td>3</td>
</tr>
<tr>
<td>5. Middlemount &amp; Lake Lindsay</td>
<td>2.8</td>
<td>BGO, 15 cm</td>
<td>2</td>
</tr>
<tr>
<td>6. Singleton</td>
<td>1.9</td>
<td>BGO, 15 cm</td>
<td>3</td>
</tr>
<tr>
<td></td>
<td>2.3</td>
<td>NaI(Tl), 19 cm</td>
<td>3</td>
</tr>
</tbody>
</table>

\textsuperscript{a} std = source-to-detector.

illustrates the correspondence between the geophysical log (obtained with this spectral window setting) and the geological log obtained by inspection of core samples from the same hole. From the figure, it appears that the geophysical log is accurate both for defining the position of the coal strata and for measuring their width. In general, reduced source-to-detector distance gives improved delineation of coal plies and intraseam sedimentary strata.

From considerations given earlier in the paper (Fig. 2), concerning the mineral content of Australian black coals, the variation of ash content in these coals is predominantly determined by the chemical concentrations of Si, Al and Fe. Consequently, we considered it unnecessary to consider the regression analysis models based on variables corresponding to the energy peaks of Ti, Ca and other elements.

The variables used in the regression model were the total count rates in spectral windows corresponding to the elemental neutron capture responses, the count rates in the windows for the 0.478 MeV peak of $^{10}\text{B}$, and for the 2.22 MeV peak of H, and also the ratios between the count rates in these various windows.

Table VI summarizes the results obtained in the field tests of the spectrometric neutron-gamma method. It is worth noting that the coal deposits denoted in the table as ‘Queensland 1’ and ‘Queensland 2’ were more than 200 km apart.
FIG. 10. Comparisons of ash contents determined by chemical analysis and neutron-gamma SIROLOG probe responses.

FIG. 11. Comparison of coal ash content determined by chemical analysis and the neutron-gamma method. The same coal deposit as for Fig. 10. The logging probe with NaI(Tl) detector.
Figures 10 and 11 show crossplots between assay values obtained for coal ash content by neutron-gamma logging and by laboratory analyses of core samples from boreholes in the two coal deposits. The nuclear assays for Fig. 10 were obtained using a borehole logging probe fitted with the BGO detector, while Fig. 11 shows results based on probe measurements using the NaI(Tl) detector.

4. SUMMARY AND CONCLUSIONS

The work carried out, both in the laboratory and in the field trials, indicated that spectrometric gamma-gamma and neutron-gamma methods are suitable for the delineation of coal plies and seams, and also for the determination of ash content in coal. The commercially used ash logging methods, based on measurements of a single parameter, have not sufficient reliability for accurate grade control applications because the chemical composition, density and mechanical properties of coal strata vary unpredictably.

In response to the need for further improved accuracy in coal ash measurements, the Commonwealth Scientific and Industrial Research Organization (CSIRO) Division of Mineral Physics has developed two new alternative techniques, which are both spectrometric. The techniques for coal derive from the Division’s earlier research applied to mineralized orebodies, and these are currently undergoing commercial development (known as SIROLOG) through collaborative licence agreements.

The spectrometric gamma-gamma method uses the correlation between the ash content and $Z_{eq}$ and density of coal. A regression equation based on a linear combination of spectral shape parameters is used for ash determination. This approach proved to be superior to the single parameter gamma-gamma methods.

The BGO detector, which has a relatively large peak efficiency and also a large peak-to-Compton ratio, was found to be particularly advantageous for the spectrometric neutron-gamma method.

The relatively low energy distribution of primary neutrons emitted by $^{252}\text{Cf}$, in comparison with Am-Be, gives a response to the silicon content in the coal which has minimal interference from carbon. This makes $^{252}\text{Cf}$ the most suitable primary neutron source.

Both methods were tested at several coal deposits for coal strata varying in ash content from 5–50%, and the RMS deviations were between 1.4 and 2.8% ash.

Tests for the reproducibility of the measurements showed that the precision of those techniques was 0.6–0.8% ash, which could not have been a determining factor for the accuracies obtained.
REFERENCES


DISCUSSION

T. GOZANI: Did you use a 5 microgram californium source to get the capture gamma ray?

J. CHARBUCINSKI: Most of the results shown were with 1 microgram. We used 5 micrograms only for the longer spacings and we found that going from 30 cm to 19 cm (our shortest distance) we improved our accuracy.

T. GOZANI: How did you do the calibration, because you get a broad spectrum? Do you calibrate on some standard which you have outside, or do you do it on one hole?

J. CHARBUCINSKI: We are doing our calibration on such a number of holes as will give us about 25 to 35 independent data points. So if we have one hole which has enough seams to give this many data points that is fine. Usually we use three to five holes for calibration. Our recommendation is that when going into
a new deposit about which not much is known, it is much easier to ask the company to provide you with four or five drilled holes, analysed and sampled very carefully. If you leave the sampling to the geologist he will do it as he always does — regarding the mining possibilities of the seam. Sometimes when he has 20 cm of shale he will still include it because he is not able to exclude it with the mining machine. But for our purposes we want to exclude it, to have a uniform point for the calibration. So you have to be present during the sampling.

T. GOZANI: But in your basic model you are fitting about five or six regions?

J. CHARBUCINSKI: Regions of the spectrum, yes. In the case of neutron capture spectra it was two regions at low energies, one boron, the second hydrogen, plus three to five regions in the higher-energy part of the spectrum, a very broad region. Using BGO detectors our spectral resolution is even worse than for sodium iodide. On the other hand we get much better efficiency. For example around hydrogen we get five times better efficiency for the same size detector of sodium iodide.

P.G. KILLEEN: As you point out, one of the problems with calibrating is knowing exactly what has been sampled by the geologist for comparison with your measurements. In addition, there sometimes are inhomogeneities in nature which will account for some of the statistical scatter on your calibration graph. I wonder if it would be better to try to make artificial calibration standards, maybe five of them, where they are accurately known, and give five points on a graph where you have control over the standard — rather than using a real hole?

J. CHARBUCINSKI: We think it might be better to decrease the number of required data points using real holes, to get quicker calibration. But still, when you go to the mine you do not have five or six blocks with very well known ash content or coal composition or whatever. Usually you have only existing holes. The most you can get from the company for calibration is that they will drill three or four more holes and analyse these again in the ways suggested. If you had such facilities as three or five data points which were well known then these could also be used. But one problem is that if you have differing chemical composition of ash in the drill holes and the blocks, then your calibration would not be very good. So the advantage of using a real area, and logging, say, for 20 points out of the 2000 which geologists will have to sample later, is that you have the cross-section of the chemical composition of coal which really appears in this mine, which you cannot model in the laboratory. What we are trying to do now in our laboratory is similar to this idea of creating large models for testing. We have constructed a big hole, about 4 metres deep, with doughnut-shaped containers, 90 cm and 45 cm deep, which we fill with different types of coal and put one on top of another to obtain an artificial profile. By changing the properties of these coals, and by changing the combination and thicknesses of these doughnuts we can obtain information about the calibration, and also about the vertical resolution. The big problem is always when you reach very thin seams of coal. Without applying a deconvolution method (which according to all people who have tried it is not
very good for nuclear logging), we find you are in trouble if you come to coal seams with thickness comparable with the source-to-detector distance. This is now a very important problem, not only for calibration but also for measuring ash content in thin seams.

J. Łoskiewicz: You need a normalization to the real ash content in the given mine. This is not a standardization but a normalization to the composition of ash.

J. Charbucinski: Yes. But usually the information about the chemical composition of ash is based on the information obtained from the hole you are going to log, anyway. So we found that, instead of normalizing, just spending one day for extra logging for calibration gave us good enough information.

C. G. Clayton: Why do you use centralized probes?

J. Charbucinski: In the case of neutron-gamma probes the decentralized probe would be no good because you could not avoid the neutrons going into the hole. You would not be able to shield the neutron source from both sides of the hole. In the case of gamma-gamma probes decentralized logging is easy. You can put a lead shield around the detector and press the probe against the sides. But we tested one of the gamma-gamma probes used by commercial logging companies and the construction is such that actually the micro-roughness, micro-cavities, have much higher influence on the output than in the case of a centralized probe. With a centralized probe you get trouble when the hole changes diameter from say 5 to 6 or 7 inches, but you are almost independent of the micro-roughness in the 2 to 3 mm range. When going into bigger diameters we are normally using, in our gamma-gamma method, compensation by applying this S factor, the ratio of intensities measured in two energy windows, both above 500 keV. This gives you almost intrinsic correlation on borehole diameter variation.
CURRENT DEVELOPMENTS
IN NUCLEAR TECHNIQUES
FOR COAL LOGGING IN CANADA

P.G. KILLEEN, C.J. MWENIFUMBO
Geological Survey of Canada,
Ottawa, Ontario,
Canada

Abstract

CURRENT DEVELOPMENTS IN NUCLEAR TECHNIQUES FOR COAL LOGGING IN CANADA.

Nuclear logging techniques for coal currently in use in Canada are briefly reviewed. Current research and development in this area is largely conducted by government laboratories. New developments at the Geological Survey of Canada include construction of a multiple-zone logging research calibration facility in Ottawa. Progress towards establishment of the field calibration facilities in eastern and western Canada is described, including the installation of two fibreglass tanks in Nova Scotia to house the model boreholes. The western calibration facility is expected to be installed during the summer of 1985 in Calgary. Construction of cylindrical calibration zones for the model boreholes will be an ongoing activity.

1. INTRODUCTION

Research and development (R&D) work in Canada in the field of nuclear borehole logging has been conducted by the logging service companies, the geophysical instrument industry, and government laboratories. Currently, the only active R&D related to nuclear logging for coal is carried out by the Geological Survey of Canada (GSC). The lack of R&D by the service and geophysical companies reflects the present state of the world economy and the slump in coal mining activities.

1.1. Current technology

In Canada, a typical coal logging programme might include neutron, density, natural gamma-ray, resistivity and caliper logs.

The neutron-neutron logs are often used for lithological correlation in the coal fields of the foothills of the Rocky Mountains. They are not commonly used to derive quantitative information (e.g. ash, sulphur content) about the coal.

The neutron capture-prompt gamma ray logging technique developed by Scintrex Limited (Toronto) has been applied only to a limited extent for the
delineation of coal seams, and measurement of ash and sulphur content. This system, termed the 'Metalog System', is able to determine the sulphur content of coal seams to within a standard error of 0.24% S in the range of 1 to 7.5% sulphur, and the ash content to within a standard error of 1.2% ash in the range of 4 to 24% ash (when logging at 7 metres/hour) [1]. The system is based on the use of a californium-252 neutron source and a sodium iodide (NaI(Tl)) detector. The sulphur content is determined from the 5.42 MeV prompt gamma emission, and the ash content is determined from the iron, silicon, and aluminium gamma rays at 8.78 MeV, 4.934 MeV and 7.724 MeV respectively. Although successful field tests have been carried out in coal fields, the system has not yet achieved popular acceptance.

The gamma-gamma density log is widely used in coal exploration in both eastern and western Canadian coal fields for identifying coal and determining the thickness of coal seams. Coal quality information can also be derived from density logs acquired with carefully calibrated density tools. Often, selected sections of the borehole may be relogged at slower speeds for more detail. Sometimes more than one density log with different source/detector spacings are acquired to obtain both bulk density and high spatial resolution information. Caliper logs provide information to correct density logs for borehole conditions. The natural gamma-ray log is primarily used for lithological identification, and is also the most commonly used method.

2. NEW DEVELOPMENTS

Since the introduction in Canada of digital magnetic tape recording of borehole logs for coal exploration around 1975, and the subsequent application of computer processing of the logs, there has been an increasing capability to make quantitative measurements. This requires development of increasingly accurate methods of calibrating the logging equipment. The emphasis on density logs as a source of quantitative information on coal quality and geotechnical data when combined with other logs led to a demand for establishing coal logging calibration facilities, in particular for density logging. In response to this need, the GSC initiated a research project in 1982 to establish calibration facilities in western Canada for the coal fields of Saskatchewan, Alberta and British Columbia and in eastern Canada for the coal fields of Nova Scotia and New Brunswick.

At the same time the need for training exploration personnel in various aspects of geophysical logging for coal was identified by the Coal Mining Research Centre in Edmonton as an essential requirement for progress. As a result, a 270 page handbook on Geophysical Borehole Logging for Coal Exploration was prepared and published in 1982 [2].
2.1. The GSC coal logging project

The coal logging project initiated in 1982 has several components. One component, still in its very early stages, is the study of the application of induced polarization (IP) logs to coal quality evaluation.

A second component is the development of a research-oriented gamma-gamma spectral logging tool. This consists of the construction of a source and source holder assembly to be attached to a GSC gamma ray spectral logging tool. The source holder is being designed by Atomic Energy of Canada Limited (AECL) to meet the Atomic Energy Control Board (AECB) safety requirements as well as GSC specifications for variability in source-detector spacing, source collimation angle, source energy, and several other parameters. The gamma-gamma spectral logging tool will be used for density, coal quality, and other investigations.

The third component of the project is the design and construction of calibration facilities. This is part of an ongoing programme to provide facilities for testing and evaluating borehole logging instrumentation and development of new borehole geophysical techniques [3, 4]. It is being done in three stages: (1) construction of a research calibration facility in Ottawa; (2) construction of a field calibration facility in eastern Canada; and (3) construction of a field calibration facility in western Canada.

2.1.1. Research calibration facility in Ottawa

A multiple-zone model borehole has been constructed for calibration of logging probes, and for testing and evaluation of the calibration zones to be used in future field calibration models. The new calibration facility, located in Ottawa, consists of a series of zones with concrete cylinders placed in contact with each other in a horizontal configuration as shown in Fig. 1. Each cylinder represents a 'zone' with specific known physical properties for which the logging tool response can be evaluated. The horizontal model borehole is produced by the alignment of the axial holes in the individual zones. Logging tools will be calibrated by pulling them through the successive zones.

The Ottawa primary facility has provision for saturating the model borehole with water to simulate most field conditions. The horizontal configuration of the model borehole makes it a versatile facility allowing removal and placement of different calibration zones during testing and evaluation.

Initial construction of the calibration facility includes a series of calibration zones made of concrete with different densities covering the range of interest for coal logging, as shown in Fig. 2. These zones, when tested for homogeneity and accurately measured for density, will be shipped to field calibration facilities to be established in western and eastern Canada for coal logging.
FIG. 1. Configuration of cylindrical concrete zones used for calibration of borehole logging probes. Each zone will have specific known physical properties. The horizontal configuration makes it easy to interchange zones using an overhead crane assembly.
FIG. 2. The multiple-zone horizontal model borehole in its below-ground tank, located in the Borehole Geophysics Laboratory at Aberdeen Street, Ottawa.

The Ottawa facility will also be used to develop standardized calibration zones for other physical properties such as porosity, or magnetic susceptibility. These will be added to the field calibration facilities as they become available.

2.1.2. Field logging calibration facilities: model boreholes

Considerable progress has been made towards the establishment of calibration facilities in eastern and western Canada. Both sites have been selected and a major step towards the completion of the facility for eastern Canada was made in late March 1985, with the in-ground installation of two fiberglass tank assemblies at the site of the Nova Scotia Research Foundation Corporation (NSRFC) in Dartmouth. The tank assemblies designed by the Borehole Geophysics Section of the GSC will eventually contain two model boreholes to permit calibration for several physical property measurements.

The vertical stack of calibration zones which form the field calibration facilities will be contained in a below-ground tank as shown in Fig. 3 with provision for water saturation to simulate field conditions. The central hole in each cylindrical zone is aligned such that the stack provides a continuous hole through all the zones, which is further aligned with a run pipe protruding below the tank. Some calibration measurements require that a steel drill rod (or pipe) be temporarily suspended in the model borehole to simulate logging inside the drill
FIG. 3. Schematic diagram of the tank assembly used in the field calibration facilities showing a vertical stack of calibration zones aligned with the run pipe in the below-ground tank. The optional drill rod is shown suspended from the cover inside the model borehole formed by the zones. The reducer at the bottom allows smooth transition from the below-tank run pipe to the zone borehole.
rod. For this reason the insulated fibreglass cover for the tank was designed to support the weight of two drill rod sections as well as a maximum of four workers. During the calibration procedure, logging tools will normally be lowered into the model borehole on a cable which runs over a pulley suspended on an overhead gantry assembly. Continuous logging at a uniform speed through the entire simulated borehole during the calibration procedure is ensured by the ‘run pipe’ beneath the tank.

Two essential requirements for the facility were: (1) that it be watertight to prevent seepage of groundwater into an empty tank or of water from a full tank into the environment, and (2) that it be free of metallic material to avoid electrical or magnetic interference during calibration of logging tools which measure electrical or magnetic properties of rocks. Consequently, a fibreglass tank with circular cross-section was selected.

Fibreglass has several advantages: it is non-magnetic, strong, tough, resilient, waterproof and weathertight, capable of withstanding extreme weather conditions and rough handling and needs little maintenance. Also, construction problems associated with concrete forming on-site are minimized since the only requirement is for a level concrete base for the tanks. Freezing is precluded through the use of insulation around the upper section of the tank, eliminating the need for anti-freeze in the tank water. Components are designed and constructed at the fibreglass plant, shipped to the site and assembled with relative ease. Possible future damage to the fibreglass tank or run pipe is prevented by the following precautions: (1) a concrete base is installed within the tank to absorb the shock during positioning of calibration zones or the impact of an inadvertently dropped object, such as a logging tool; (2) the bottom of the run pipe is filled with a 60 cm layer of silica sand to cushion the blow from a falling tool.

The fibreglass tank design takes into consideration the particular conditions of each site, such as the environment, type of material to be excavated, depth to bedrock, hydrostatic conditions, frost susceptibility of the soil, backfill pressures, settlement, tendency for flotation or leakage of tanks, ability to vary the borehole diameter in future, worker safety, and security against possible vandalism. Four fibreglass tanks comprising tank, cover, run pipe, reducer and overhead gantry were required, i.e. two each for eastern and western Canada.

The optimum borehole diameter was determined, based on common regional coal drilling practice, to be 100 mm in western Canada and 75 mm in eastern Canada, requiring an appropriate reducer allowing smooth transition from below-tank run pipe (200 mm diameter) to the borehole.

2.1.3. Installation of the eastern Canada tank assemblies

The eastern site at the Nova Scotia Research Foundation Corporation (NSRFC) was selected for three main reasons: (1) it is the location of a group actively involved in coal logging and interested in the use of the facility;
(2) a suitable tract of land was available and the NSRFC was willing to allocate it for the facility; and (3) the NSRFC was willing to maintain security and provide access to the site for people desiring to calibrate logging probes. The location is ideal, having nearby electricity and water supplies, and a security fence surrounds the property. Preliminary subsurface soil test reports had indicated the area (a) is underlain by glacial till with extensive preconsolidation, (b) has a low water-table, and (c) has its bedrock well below the required excavation depth.

At the site, a large hole for the two tanks was excavated to a depth of about 6 m. On the downhill side of the excavation a ramp was formed by excavating one wall of the hole. A truck-mounted drill rig was backed down the ramp into
FIG. 5. A crane was used for positioning the tank over the run pipe and lowering it onto the concrete base.

the excavation to drill the holes and install the run pipes for the two tank assemblies (see Fig. 4). Upon completion and removal of the drill rig from the excavation, a single continuous concrete base was prepared for the two tanks, being poured around the two protruding fibreglass run pipes. The tanks, which had an appropriate hole in the bottom, were positioned over the run pipes and lowered into place (see Fig. 5). The fibreglass tank bottom was bonded to the run pipes. The additional layer of protective concrete was poured inside each tank and a fibreglass lip was bonded to the tank wall above it to prevent any possible upward buckling movement of the tank bottom. Backfilling, spreading and levelling were completed and the remaining fibreglass components (cover, overhead gantry, etc.) were assembled as shown in Fig. 6.
2.1.4. Progress on the western Canada coal logging facility

The western site is to be located in Calgary, at the Institute of Sedimentary and Petroleum Geology (a division of the GSC), adjacent to calibration pads for portable gamma-ray spectrometers which were constructed in 1977 [5]. At this location, the water-table is variable and the ground is unstable, requiring an engineering site evaluation with appropriate recommendations prior to installation of the new fibreglass tanks. Although the details of the procedure for installation remain to be determined, the western tanks have already been designed and constructed to withstand the condition of saturated soil surrounding them to the top. These tanks are 60 cm longer than the eastern tanks to allow a greater amount of concrete inside for additional load against uplift. It is expected that the Calgary set of in-ground tanks will be installed in the summer of 1985.

2.1.5. Calibration zones

One of the most difficult problems in the project is the control of the homogeneity of mixtures used in calibration zones, regardless of the physical
parameter being considered. Initial construction and testing of density calibration zones has been conducted with the Civil Engineering Department of the University of Ottawa, and this work is continuing. Some calibration zones for both logging facilities will likely be delivered and installed late in 1985. Additional calibration zones will be added as they become available.

**REFERENCES**


**DISCUSSION**

J.L. MIKESSELL: We have a vertical stacking system almost identical to the one you showed. Our samples don’t have a steel jacket. We have a precision mould into which we pour the concrete mixtures. We also have precision containers into which we can put crushed ore without the addition of cement. The problem we have had with that is that there is settling and it does leave an air gap, so I would not recommend it unless you find it absolutely necessary to use a loose sample of that sort.

P.G. KILLEEN: We tried to get as much information as we could in advance, and one of the other places that had used a vertical stack with loose unconsolidated material was Syntrax in Toronto, when they developed their so-called metal logger. It used presses to attempt to press the material into the mould as consistently as possible. As you point out, the more you handled it the more settling or changes in it you got. So we may someday use some unconsolidated calibration cylinders but I doubt if they would be the kind that we would ship for use by industry.

C.G. CLAYTON: Are these for both density measurement and for neutron-gamma?

P.G. KILLEEN: We might have to make a larger diameter for neutron-gamma. I am not sure what the mean free path is, from the hole out and back.
We have about 50 cm radius of material around the hole. That might be sufficient. We figured it was for any gamma-gamma work. But we thought we would build more for density and for magnetic susceptibility. And then probably for porosity, as there is a demand for that in Canada too. Other than that if we consider any elemental work we might try mixtures of copper, lead and zinc ores.

C.G. CLAYTON: The question of calibration is becoming of strong interest, particularly in relation to the oil industry. Certainly for neutron-gamma work it is important and as physicists we should attempt to make it as accurate as possible. The principal question is whether natural materials are adequate and whether impurities of high macro-absorption cross-section will give uncertainties in a calibration facility. Our attitude to this is to use reconstituted natural materials, to choose the purest rocks, and reconstitute these and remove the impurities.

There is also a strong argument for using an artificial coal for calibration. At least you can then avoid the question of whether or not your sample is representative.

P.G. KILLEEN: I think the concept of using artificial coals is pretty well accepted, for example, aluminium blocks for density, where you know the electron density very accurately to give one point on a calibration curve. Certainly artificial models are used. The density models I discussed are probably no good for Charbucinski's gamma-gamma spectral ash determinations, because I could tell him the ash content, but he would say, "yes, but what about the iron, alumina and silica?" So again you have to go back and get something where those are known.

J. CHARBUCINSKI: In my view the problem with models which you construct for calibration is that when you start you never know how far it will get you. You start with five or six models. Later you find that it would be good to know what happens when the chemical composition changes, so you build another ten models with different chemical composition. Later you would like to know what happens when the borehole diameter changes, so you build another five or six models. Most laboratories have limited space. We don't have an area where we could store such models. You can only build limited facilities, and it is a very long and costly business. It is also a problem, before starting up such facilities, how to plan so that they will be fairly universal, so that one model can serve several purposes.

T. GOZANI: I read the paper by Clayton and Butler, and I think this is the right approach. You cannot do millions of combinations or configurations, so you need a transport or Monte Carlo code that can simulate the situations. You can then do a few measurements to confirm the calculations. We also need to build techniques which are more robust, giving results that do not change suddenly when somebody sneezes, when any minor parameter changes. Then I think the combination of a minimum number of calibration standards confirming
the calculation, and using calculation to extend it, is the right approach. That is the approach that Clayton is taking.

C.G. CLAYTON: Thank you very much. Sufficient to say that the starting point for all that is to have a calibration facility that you really believe in.

P.G. KILLEEN: The key word is the combination. I would be much against a completely theoretical modelling and I can see how many others here would be against a completely empirical modelling, because it can answer only a few of your questions. The ideal thing is a combination of real empirical models and theoretical modelling to fill in all the points on the curve in between and at the ends.
DEVELOPMENT AND APPLICATION OF NUCLEAR TECHNIQUES FOR ON-LINE COAL ANALYSIS IN INDIA

A.S. PRASAD
The Tata Iron & Steel Co. Ltd,
Jamshedpur, Bihar,
India

Abstract

DEVELOPMENT AND APPLICATION OF NUCLEAR TECHNIQUES FOR ON-LINE COAL ANALYSIS IN INDIA.

In view of the meagre reserves of coking coal, which are restricted to some parts of the country only, whereas comparatively much larger reserves of non-coking coal are found throughout the country, it is essential to consider seriously the rational utilization of coking coal resources and increased use of substandard coking coals and non-coking coals in coke-making for the growing iron and steel industry in India. Techniques such as preheating, selective preparation, briquette blending, pellet coking and briquette coking are some of the steps which are being adopted for increased utilization of substandard coking coals. Rational utilization of coking coal resources in India and successful implementation of the newly developed techniques for increased utilization of substandard coals call for fully foolproof quality control at all levels of operation and continuous assessment of coking potentialities of individual coals and their mixtures prior to their use in coking plant. Nuclear analytical techniques have been extensively applied in India to medicine, fertilizers, chemicals, polymers, and the iron and steel industries, but not much work has been done to apply the technique in the coal industry. The paper gives an overview of Indian coal reserves and describes efforts made towards adopting indigenously developed nuclear techniques in coal- and coke-handling plants. The complications and problems attendant with the use of this relatively new technique are also briefly discussed. The future programme as envisaged today in India is touched upon. The industrial application of isotopes definitely holds great promise, especially in the coal industry, and can be gainfully utilized to solve many intriguing problems hitherto not fully understood.

1. INTRODUCTION

Energy and power are essential prerequisites for the industrial development of a country and these are largely derived in India from the primary commercial energy sources coal, oil and natural gas. Because of the extensive reserves of fossil fuel compared to other conventional sources, coal continues to play the major role as a source for supply of commercial energy in India. Of the total energy of about 193 million tonnes of coal equivalent (mtce), the energy from non-commercial sources are: coal, 63 mtce; oil, 21 mtce; hydro, 10.8 mtce and
there is a small contribution from the nuclear sources. Of the total 63 mtce of coal, the railways consumed a little less than 30%, the iron and steel industry about 20%, the domestic sector and collieries nearly 5% each, and the balance was accounted for by cotton textiles, paper and other miscellaneous consumers. Each of the consuming sectors mentioned requires coal of specified type and quality, the supply of which, particularly to the iron and steel industry and railways, poses a difficult problem. This problem of availability of coal of preferred grades and qualities is likely to assume much greater proportions in the years to come unless adequate technological changes are envisaged to reduce the growing dependence of the different consumers on coals only of best quality.

The metallurgical and engineering industries generally require coke as fuel. For this, coals with certain characteristic properties are needed. Based on these, coal resources can be broadly classified into two groups: (i) coking and (ii) non-coking. While India has extensive iron ore resources of fairly good quality, the opposite is the case with coking coals. Apart from the limited reserves, a disconcerting factor is the very high ash content of the Indian coking coals and its distinct tendency to increase as seams deeper down are worked, which is inevitable when the output has to be increased. Besides, the nature of dispersal of the ash in coal is such that ordinary beneficiation methods, normally applied with great success in other countries, need to be considerably modified under Indian conditions.

As regards the quantity, Tables I, II and III give an idea of the estimated reserves of coking coals. While there may be some basic difference between the

<table>
<thead>
<tr>
<th>COUNTRY</th>
<th>ECONOMICALLY RECOVERABLE HARD COAL</th>
<th>ECONOMICALLY RECOVERABLE LOWER COAL</th>
<th>MEASURED RESERVES (ALL RANKS)</th>
<th>TOTAL RESERVES (ALL RANKS)</th>
<th>PERCENT OF WORLD RESOURCES</th>
</tr>
</thead>
<tbody>
<tr>
<td>USA</td>
<td>123 443</td>
<td>58 338</td>
<td>363 562</td>
<td>2 924 503</td>
<td>27.1</td>
</tr>
<tr>
<td>USSR</td>
<td>165 800</td>
<td>107 400</td>
<td>273 204</td>
<td>5 713 681</td>
<td>53.0</td>
</tr>
<tr>
<td>CANADA</td>
<td>4 195</td>
<td>1 342</td>
<td>9 034</td>
<td>108 777</td>
<td>1.0</td>
</tr>
<tr>
<td>CHINA</td>
<td>101 300</td>
<td>200 500</td>
<td>1 011 700</td>
<td>9.4</td>
<td></td>
</tr>
<tr>
<td>GERMANY, Fed. Rep.</td>
<td>30 000</td>
<td>9 571</td>
<td>99 521</td>
<td>287 054</td>
<td>2.7</td>
</tr>
<tr>
<td>UK</td>
<td>3 871</td>
<td>98 877</td>
<td>162 814</td>
<td>1.5</td>
<td></td>
</tr>
<tr>
<td>AUSTRALIA</td>
<td>13 770</td>
<td>10 555</td>
<td>74 341</td>
<td>198 567</td>
<td>1.9</td>
</tr>
<tr>
<td>INDIA</td>
<td>10 643</td>
<td>897</td>
<td>23 139</td>
<td>82 937</td>
<td>0.8</td>
</tr>
</tbody>
</table>

REFERENCE: WORLD COAL, NOV '78
TABLE II. COAL RESERVES IN INDIA (million tonnes)

<table>
<thead>
<tr>
<th>TYPE</th>
<th>PROVED</th>
<th>INDICATED</th>
<th>INFERRED</th>
<th>GROSS</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>A. NON-COKING COAL</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>12 306</td>
<td>22 310</td>
<td>26 180</td>
<td>60 796</td>
</tr>
<tr>
<td><strong>B. COKING COAL</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>a) PRIME COKING</td>
<td>9 059</td>
<td>8 449</td>
<td>2 646</td>
<td>20 154</td>
</tr>
<tr>
<td>b) MEDIUM COKING</td>
<td>3 850</td>
<td>4 309</td>
<td>1 272</td>
<td>9 431</td>
</tr>
<tr>
<td>c) BLENDABLE</td>
<td>1 559</td>
<td>2 600</td>
<td>914</td>
<td>5 073</td>
</tr>
<tr>
<td><strong>TOTAL</strong></td>
<td>21 365</td>
<td>30 759</td>
<td>28 826</td>
<td>80 950</td>
</tr>
</tbody>
</table>

REFERENCE: MINISTRY OF ENERGY, DEPT. OF COAL, INDIA.

TABLE III. ESTIMATE OF THE POTENTIAL OF THE METALLURGICAL COAL RESERVES IN INDIA (million tonnes)

<table>
<thead>
<tr>
<th>CATEGORY</th>
<th>GROSS RESERVE</th>
<th>NET AVAILABILITY AFTER MINING AND WASHING TO OPTIMUM ASH</th>
</tr>
</thead>
<tbody>
<tr>
<td>PRIME COKING COAL</td>
<td>5 753</td>
<td>2 312</td>
</tr>
<tr>
<td>MEDIUM COKING COAL</td>
<td>7 380</td>
<td>1 752</td>
</tr>
<tr>
<td>BLENDABLE COKING COAL</td>
<td>1 372</td>
<td>688</td>
</tr>
<tr>
<td><strong>TOTAL</strong></td>
<td>14 505</td>
<td>4 752</td>
</tr>
</tbody>
</table>

actual and the estimated figures, the overall reserves after beneficiation are extremely limited and could be considered critical, in view of the extensive iron ore deposits. The total coal reserves in India are about 80 billion\(^1\) tonnes which comprise only 20 billion tonnes of coking coals. Of the gross reserves of 20 billion tonnes, prime coal constitutes 5.6 billion tonnes only and after mining and beneficiation (washing) works out to the alarmingly low figure of 2.3 billion tonnes. To smelt the entire workable reserves of iron ore of the order of 16 billion tonnes in India, 13.5 billion tonnes of coking coal, comprising

\(^1\) A billion = 10\(^9\).
TABLE IV. BLEND PERCENTAGES OF STEEL PLANT COKE OVENS IN INDIA

<table>
<thead>
<tr>
<th>STEEL PLANTS</th>
<th>PRIME COKING COAL</th>
<th>MEDIUM COKING COAL</th>
<th>SEMI- AND WEAKLY COKING COAL</th>
</tr>
</thead>
<tbody>
<tr>
<td>BHILAI</td>
<td>61</td>
<td>31</td>
<td>8</td>
</tr>
<tr>
<td>ROURKELA</td>
<td>50</td>
<td>40</td>
<td>10</td>
</tr>
<tr>
<td>DURGAPUR</td>
<td>75</td>
<td>15</td>
<td>10</td>
</tr>
<tr>
<td>BOKARO</td>
<td>58</td>
<td>42</td>
<td>-</td>
</tr>
<tr>
<td>TISCO</td>
<td>59</td>
<td>32</td>
<td>9</td>
</tr>
<tr>
<td>ISCO</td>
<td>71</td>
<td>20</td>
<td>9</td>
</tr>
</tbody>
</table>

RANGE: 50 - 75  15 - 42  0 - 10  AVERAGE: 62  31  7

about 6.5 billion tonnes of prime coal, would be required — indicating a deficit of about 4 billion tonnes of prime coal.

Nature has been more generous in respect of the reserves of the non-coking coals.

Whereas in most other countries, coking coals used for blast furnaces have an ash content of 8—10%, the run-of-mine coals in India, before washing, have an overall ash content of well over 24%. Thus, even after washing, the ash level tends to be far higher. Currently, the problem is further aggravated by the fluctuations in the ash content. The inference is obvious: concerted efforts have to be made for the conservation of higher grade metallurgical coals by adopting the most efficient beneficiation process, which will result in maximum recovery from all fractions and utilization of blends for coking which will maximize the use of weakly-coking and even non-coking coals. Table IV gives an idea of the blend percentage at the coke ovens of various iron and steel plants in India.

Even after beneficiation, the proportion of reserves of prime, medium- and weakly-coking coals works out to be 49:37:14, whereas the consumption pattern has been 62:31:7. The first step is to aim at a consumption pattern, after beneficiation, which will more or less conform to the reserve pattern, so that there is no undue depletion of the prime coking coals. To reach this goal, the washing/beneficiation processes of coking coals need to be optimized by adopting established processes applicable to medium- and weakly-coking coals. Techniques enabling the direct use of a substantial percentage of medium- and weakly-coking coals in the blends, without any adverse effect on the output and quality of coke, need also be adopted. Over and above all these steps, the utilization of non-coking coals as a direct reductant has to receive priority attention, because of its specific advantages to India.
The increasing demand of coals for coking has to be met primarily by increasing production of raw coals, which, therefore, will call for mechanized mining of selected coal seams at depth, often under adverse geological conditions. The adoption of mechanized mining produces more fines. This necessitates the beneficiation of fines separately. In the case of high-ash Indian coals containing finely disseminated mineral matter throughout the coal mass, washing poses a great problem and froth-flotation may not be the ideal process. Further, even if the technique is adopted, the yields are low and the high moisture content of the dewatered product makes handling difficult. The oil-agglomeration technique may prove more economical and commercially successful.

The most challenging task before the coal preparation technologists in India today is economically to prepare coking coal in the existing washeries and to upgrade those that are difficult to wash.

The geographical confinement of coal resources to certain areas of the country, Fig. 1, introduces its own problem. With the growth of Indian national economic activities in the coming decades, the development of in situ conversion techniques of coals into more useful forms and transporting these products will also need careful consideration.

2. NUCLEAR TECHNIQUES

It is becoming imperative as well as a challenging task for the technologists in India to develop on-line control systems, so that the limited reserve of prime coal is preserved and the non-blendable and medium grade coals are utilized to the optimum level, keeping in mind the end-use of the coke thus produced.

There are a number of on-line process control instruments available and though these are being used in various places in the Indian coal-handling plants, the nuclear techniques have an added advantage over the conventional system in that these instruments are capable of withstanding the severe environmental conditions prevailing in the coal/coke-handling plants. From an engineering, safety and maintenance viewpoint, a nuclear source, when used as a signal transmitter, represents the ultimate in simplicity. This statement is based on the fact that it is simply a small piece of solid matter generating its own power, requiring no maintenance. However, the small delay in applying nuclear devices in the early days was due to the lack of suitable electronic instruments capable of withstanding the brutal environmental conditions, namely, corrosion, abrasion, highly reactive material, severe vibrations and dust. The situation is changing fast with developments in the electronics hardware.

An attempt has been made here to highlight the various possible applications to which the radioisotopes can be put in a coal/coke handling plant by giving illustrations of the installations in India. Also briefly discussed are the complications and problems attendant with the use of this relatively new technique. The future programme as envisaged today in India is briefly touched upon.
To begin with, in the collieries, normally two processes are used — (i) underground mining and (ii) open-cast mining. In both cases it is necessary to know the quality of coal under the ground before the actual mining operation is taken to make the entire operation economically feasible. As such, on-spot determination of ash content, and, if possible, the complete proximate analysis of coal is essential. A portable X-ray fluorescence analyser is being used in India, Fig. 2. This instrument has been developed by the Electronics Corporation of India with the technical know-how obtained from Bhabha Atomic Research Centre. The principle of its working is well known and it is not necessary to go into detail of its functioning. It uses a 30 millicurie $^{238}\text{Pu}$ source and a NaI detector.
Gunsons & Sortex of England have supplied an on-line ash meter to one of our coal washeries. This carries two 10 millicurie plutonium-238 encapsulated sources, uses a proportional counter as detector and is based on backscatter technique. The amount of radiation absorbed and backscattered depends on the elemental composition of the coal sample. Elements of low atomic number (the combustible elements) backscatter well and absorb little of backscattered X-rays. Conversely, elements of high atomic number (the ash elements) backscatter less and absorb relatively more of the radiation. By measuring the backscattered radiation, a determination of the ash content of the coal sample can be made. However, the test results are not very encouraging. Perhaps the variation in the measured and analytically determined values of ash in coal is due to the peculiar quality of the Indian coal.

A system, Fig. 3, using 3.5 millicurie $^{137}$Cs backscatter density and a 40 millicurie Pu-Be neutron moisture gauge for locating aquifer regions in boreholes is being used in India. It would be advisable to go for a similar set-up for borehole coal sample logging for the classification of coals.

The coals excavated from various seams differ widely in characteristics and it becomes necessary to blend them at the pit head itself in order to make them economically viable for end-use. In this regard, the Indian Institute of Technology, Bombay, has developed an on-line nuclear weighing system (Fig. 4), which has been marketed by the Electronics Corporation of India and is being very successfully used. This is designed for coal being conveyed at the rate of 250 tonnes per hour by a conveyor of 600 mm width running at about 90 to 100 metres per hour. It contains a 20 millicurie line source of $^{137}$Cs and has an
FIG. 3. Schematic diagram of the neutron moisture probe and logging set-up (Atomic Mineral Division, Hyderabad, India).

FIG. 4. On-line nuclear weighing system (ECIL, India).
TABLE V. ASH CONTENT OF COALS AT VARIOUS SIZES OF CRUSHING

<table>
<thead>
<tr>
<th>Jharia coals</th>
<th>Size of crushing —top size— (mm)</th>
<th>Ash (%)</th>
<th>VM (%)</th>
<th>S (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Prime coking</td>
<td>75</td>
<td>15—30</td>
<td>22—23</td>
<td>0.4—0.8</td>
</tr>
<tr>
<td></td>
<td>13</td>
<td>18—24</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>0.42</td>
<td>16—18</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Medium coking</td>
<td>75</td>
<td>24—35</td>
<td>18—22</td>
<td>0.3—0.6</td>
</tr>
<tr>
<td>(Low volatile)</td>
<td>13</td>
<td>25—28</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>0.42</td>
<td>25—27</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Medium coking</td>
<td>75</td>
<td>19—35</td>
<td>32—39</td>
<td>0.3—0.5</td>
</tr>
<tr>
<td>(High volatile)</td>
<td>13</td>
<td>20—32</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>0.42</td>
<td>19—29</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

argon gas-filled ionization chamber, one litre in volume, for the detection of transmitted radiation. The accuracy is reported to be ±1%.

The mechanized operation produces fines and the fines thus produced have to be beneficiated. Also, the removal of ash by beneficiation requires crushing of coal to a size below 10 mm. With the size fraction of coals, the ash content also varies, Table V. Indian coals pose problems in washing. A typical coal washing circuit for one of the coal washeries of Tata Steel is shown in Fig. 5. In this plant, the media density is controlled by a set of nuclear density gauges, Fig. 6. These on-line density gauges are directly mounted on the process pipes and the transmitted radiation is received by a radiation detector. The signal thus produced is utilized for the control system operation, Fig. 7. Initially a number of on-line density gauges had been procured from Kay-Ray Incorporated, USA, but now indigenously developed instruments are successfully being used. Level gauges, based on the same radiation transmission principle, have also been installed to monitor the overflow of cyclones, Fig. 8.

The washed coal contains a very high percentage of moisture and it becomes absolutely necessary to monitor as well as to know the exact moisture content of coal. Moisture gauges developed by Tata Steel are being used for indication of moisture in the coal-washing circuit at one of the coal washeries.

The washed coals are stored in bunkers before they are transported either by road or by rail to the coke ovens for coking. The high moisture content adversely affects the flow of coal through the bunkers. The bunkers used for storing the coal are either of mass-flow or core-flow types. The core-flow types pose greater problems. The materials frequently stick to the bunker walls and result in permanent ‘dead’ masses giving a false indication of the bunker being
FIG. 5. Coal washing plant circuit at Tata Steel.
FIG. 6. Coal-washing plant circuit showing nuclear density gauges.

FIG. 7. Specific gravity control of medium in a coal-washing plant.
FIG. 8. Density sensor in a heavy media cyclone.

full, Fig. 9. However, this difficulty was partly solved by providing hollow pipes projecting inside, Fig. 10, up to the depth of build-up. This arrangement is working satisfactorily except that the pipes, even if they have a hard facing, wear away by erosion. This requires constant surveillance, as once the pipe is eroded, the possibility of the dislodgement of the isotope capsule from its seating cannot be ruled out.

The requirements of on-line control systems of coke ovens at coal handling plants are of a similar nature. Instruments which are being used in coal plant at coal pit-heads are equally suitable for the coal-handling plant as well and a number of such instruments are in use.

For the coal to be used for coking, it is essential for the coal mix to have an optimum bulk density. The bulk density, though dependent upon moisture content, Fig. 11, is also dependent upon the blend from various mines as well as on the various fractions of prime, medium and blendable coals from the same mine. Often it has been found possible to control the bulk density, at a particular
FIG. 9. 'Dead mass' on bunker wall giving false signals.

FIG. 10. Arrangement to overcome the false signal due to material build-up.
moisture level, by adding oil. The flow of oil has to be controlled depending upon the coal mass being conveyed on a conveyor belt. A trial set-up of a nuclear weighing system, coupled with a flowrate meter, has been installed in a coal-handling plant of Tata Steel and the results to date have been very encouraging.

Level control of the coal feed in a coke oven is necessary. This is monitored by a number of level gauges on the larry car, Fig. 12. Once the coke is ready to be taken out of the oven after the carbonization cycle is complete, it has to be pushed out by a ram and the whole of the coke cake is pushed out through the rear opening of the oven. To prevent the coke falling on the track, the quenching car, coke guide and the pusher must be in alignment. Though at present none of the coke ovens in India have this type of nuclear alignment systems, Tata Steel has worked out a plan to incorporate this system in the coke oven proposed to be commissioned shortly. A part of this alignment system is being procured from abroad and the electrical interlocks are being indigenously developed. Based on the performance of the system, further such systems will be incorporated in other existing coke ovens.

The coke is red-hot when it is pushed out. It is water quenched and transferred by conveyor to the storage bins at the blast furnaces.

When coke is charged into a blast furnace on the basis of its weight, an increase in the moisture content means a decrease in the weight of coke fed. Additional coke is then required to ensure stability in the furnace operation. The first attempt in India to develop and install an on-line neutron moisture gauge has been made by Tata Steel.
Figure 13 shows the schematic arrangement of an installation for the measurement of moisture in coke in a blast furnace charging hopper. An americium/beryllium source emitting fast neutrons is placed beside a slow-neutron detector ($^3$He), and enclosed with a pre-amplifier in a steel tube to form a moisture-measuring probe. The response from the slow-neutron detector is a measure of the hydrogen content of the material and, thus, the water content, if the bound hydrogen is ascertained earlier.

To establish that the gauge is sufficiently accurate to measure the moisture in coke, 100 kg coke samples, each weighing about 50 kg, were drawn from the skip-charging-hopper in which the neutron gauge is installed. A comparison between the moisture percentage obtained by the weight loss method and neutron gauge readings showed a fairly good relationship, Fig. 14. The gauging accuracy was found to be ±1.1%.

When the calibration curve for coke for on-line neutron moisture gauge is compared with the theoretically calculated curve, Fig. 15, it appears that the combined hydrogen in Indian coke has a pronounced effect at lower moisture content, and it becomes insignificant at higher moisture content of the coke. To increase the sensitivity in the lower moisture range, the fast neutron source was wrapped with a hydrogenous material, such as wax or Perspex. So long as the coke size remains constant, the variation in bulk density does not affect the results significantly.
FIG. 13. Neutron moisture gauge system developed at Tata Steel for coke charging in the blast furnaces.

FIG. 14. Relationship between the percentage moisture in coke as determined by nuclear moisture gauge and as analysed by the loss-in-weight method on a dry coke basis.
On-line determination of ash content in coke has also been posing problems to technologists, perhaps because of the very high ash content of Indian coke, Table VI. As mentioned earlier, in all these backscatter nuclear ash meters, the assumption of a very good correlation between the ash percentage and one of the constituents, e.g. SiO₂, Al₂O₃, Fe₂O₃, is made, Table VII. For the coke being used as Tata Steel, calculations were made to find out whether any such correlation exists. Findings are plotted in Fig. 16 for SiO₂ and in Fig. 17 for Al₂O₃ contents of coke as related to the ash content. A correlation coefficient of 0.781 was found in case of SiO₂, whereas it was 0.622 in case of Al₂O₃. The respective regression equations were found to be:

\[
Y \text{ (coke ash %)} = 0.611 \times (\% \text{ SiO}_2 \text{ in coke ash}) + 17.711
\]

\[
Y \text{ (coke ash %)} = 0.413 \times (\% \text{ Al}_2\text{O}_3 \text{ in coke ash}) + 23.660.
\]

This goes against the earlier assumptions of a good correlation. The English company Analex has also tried to analyse the ash samples of Indian coke assuming a correlation with the Fe content of ash. The readings of the Lab-X instrument are given in Table VIII. The inference is obvious – no correlation exists. Thus, on-line or off-line ash analysis of Indian coal or coke using nuclear techniques still remains a challenge to Indian technologists.
TABLE VI. ANALYSIS OF ASH IN COKE

<table>
<thead>
<tr>
<th>ELEMENT</th>
<th>PERCENTAGE</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fe₂O₃</td>
<td>2.57</td>
</tr>
<tr>
<td>SiO</td>
<td>15.60</td>
</tr>
<tr>
<td>Al₂O₃</td>
<td>6.33</td>
</tr>
<tr>
<td>CaO</td>
<td>0.90</td>
</tr>
<tr>
<td>MgO</td>
<td>0.61</td>
</tr>
</tbody>
</table>

TABLE VII. ANALYSIS OF COKE

<table>
<thead>
<tr>
<th>ELEMENT</th>
<th>PERCENTAGE</th>
</tr>
</thead>
<tbody>
<tr>
<td>SiO₂</td>
<td>10.0 - 16.0</td>
</tr>
<tr>
<td>CaO</td>
<td>0.5 - 1.5</td>
</tr>
<tr>
<td>MgO</td>
<td>TRACE - 1.5</td>
</tr>
<tr>
<td>Al₂O₃</td>
<td>5.0 - 9.0</td>
</tr>
<tr>
<td>Fe₂O₃</td>
<td>1.50 - 2.50</td>
</tr>
<tr>
<td>S</td>
<td>0.35 - 0.70</td>
</tr>
<tr>
<td>MOISTURE</td>
<td>1.50 - 10.00</td>
</tr>
</tbody>
</table>

FIG. 16. Correlation between SiO₂ content of ash and percentage of ash in coke.
3. FUTURE PLANS

There are many other areas where isotopes could possibly play an important part. It is planned to undertake the following:

(1) In situ wear studies — preferably using the thin layer activation method — of the components used in coal/coke-handling plants with a hope to replace the fast-wearing materials by wear-resistant materials and reduce the down-time of the plant.

(2) Continuous on-line monitoring of the ash content of coal and coke (the conventional nuclear meters developed in India as well as in other countries have not been found suitable for Indian coals and coke).

(3) Development of on-line slurry flowmeters with continuous indication of percentage of suspended particles.

(4) Investigation of tracers — especially for understanding the kinetics of S and P transfer from coal to coke during the carbonization of coal.

4. REMARKS

World coal deposits represent the largest reservoir of energy still available to mankind. Useful utilization of this is the demand of the day.

With the increasing demand (Fig. 18), and the problem of obtaining prime coking coal in adequate quantities and qualities commensurate with the growing needs
TABLE VIII. MEASURED INTENSITIES FOR COKE SAMPLES USING ANALEX LTD LAB-X INSTRUMENT

<table>
<thead>
<tr>
<th>Sample</th>
<th>% Ash</th>
<th>0</th>
<th>10</th>
<th>25</th>
<th>30</th>
<th>40</th>
<th>50</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>17.71</td>
<td>119 850</td>
<td>108 514</td>
<td>96 688</td>
<td>91 920</td>
<td>89 788</td>
<td>79 469</td>
</tr>
<tr>
<td>B</td>
<td>18.79</td>
<td>125 138</td>
<td>114 045</td>
<td>101 165</td>
<td>96 104</td>
<td>94 068</td>
<td>82 879</td>
</tr>
<tr>
<td>C</td>
<td>20.50</td>
<td>95 454</td>
<td>86 380</td>
<td>76 483</td>
<td>72 680</td>
<td>71 280</td>
<td>64 989</td>
</tr>
<tr>
<td>D</td>
<td>21.30</td>
<td>95 554</td>
<td>85 454</td>
<td>75 845</td>
<td>71 874</td>
<td>69 994</td>
<td>62 663</td>
</tr>
<tr>
<td>E</td>
<td>21.88</td>
<td>252 259</td>
<td>224 608</td>
<td>192 979</td>
<td>181 819</td>
<td>175 745</td>
<td>147 356</td>
</tr>
<tr>
<td>F</td>
<td>22.89</td>
<td>266 200</td>
<td>238 516</td>
<td>204 180</td>
<td>191 036</td>
<td>183 944</td>
<td>153 763</td>
</tr>
<tr>
<td>G</td>
<td>24.44</td>
<td>86 993</td>
<td>79 806</td>
<td>72 279</td>
<td>69 368</td>
<td>68 128</td>
<td>62 082</td>
</tr>
<tr>
<td>H</td>
<td>25.21</td>
<td>87 979</td>
<td>80 997</td>
<td>74 530</td>
<td>71 460</td>
<td>70 098</td>
<td>64 958</td>
</tr>
<tr>
<td>I</td>
<td>26.00</td>
<td>86 656</td>
<td>80 126</td>
<td>72 787</td>
<td>70 419</td>
<td>70 036</td>
<td>63 909</td>
</tr>
<tr>
<td>J</td>
<td>27.07</td>
<td>240 678</td>
<td>214 450</td>
<td>184 499</td>
<td>171 017</td>
<td>165 999</td>
<td>138 198</td>
</tr>
<tr>
<td>K</td>
<td>27.99</td>
<td>244 108</td>
<td>215 782</td>
<td>183 879</td>
<td>172 361</td>
<td>166 003</td>
<td>139 073</td>
</tr>
<tr>
<td>L</td>
<td>28.88</td>
<td>84 118</td>
<td>76 408</td>
<td>69 345</td>
<td>66 455</td>
<td>64 406</td>
<td>59 328</td>
</tr>
<tr>
<td>M</td>
<td>29.65</td>
<td>82 239</td>
<td>76 258</td>
<td>69 382</td>
<td>66 248</td>
<td>65 926</td>
<td>59 906</td>
</tr>
<tr>
<td>N</td>
<td>30.48</td>
<td>79 298</td>
<td>73 256</td>
<td>66 220</td>
<td>63 664</td>
<td>62 263</td>
<td>57 297</td>
</tr>
<tr>
<td>O</td>
<td>32.89</td>
<td>81 681</td>
<td>73 988</td>
<td>66 248</td>
<td>62 930</td>
<td>61 904</td>
<td>55 704</td>
</tr>
</tbody>
</table>

Instrumental parameters

Source: 30 mCl 338Pu
Detectors: Ar/CH4
Pulse height analyser: Fe peaked at 3
Threshold: 2
Counting time: 20 s.

of the iron and steel industry in India, a plan and strategy for a short- and long-term solution is in the offing. This includes judicious control of the production of coal from inferior lower seams and conservation of the prime coking coals by proper blending with medium- and non-coking or weakly-coking coals.

Nuclear techniques for on-line process controls can be of an immense help to the coal technologists in their endeavour to achieve this goal.

In conclusion, the coal industry in India has an immense and challenging future and nuclear technologists from India and elsewhere will certainly rise to meet it.

Nuclear techniques definitely hold great promise in industrial applications and can be gainfully utilized to solve many of the problems mentioned earlier.
FIG. 18. Increasing demand for prime coking coal (million tonnes) for hot metal production based on present blend composition.

ACKNOWLEDGEMENTS

The author is obliged to the Government of India, the Department of Atomic Energy and Tata Steel for their confidence in him, evidenced by nominating him to the Advisory Council.

The author is indebted to Dr. V.K. Iya, Director, Isotopes Group, BARC, India, for his help, encouragement and valuable suggestions from time to time, without which it would not have been possible for Tata Steel to pioneer the use of this technique in India.

The assistance provided by the Bhabha Atomic Research Centre, Electronics Corporation of India, colleagues and associates of the Research and Development Division, Tata Steel, is gratefully acknowledged.

DISCUSSION

B.D. SOWERBY: Regarding the analysis of moisture in coke, the accurate operation of a neutron moisture gauge for coke depends on there being a constant hydrogen content in the coke. There has been some work recently reported from the USA that indicated in some coke ovens hydrogen variations give significant problems. In fact you find that something like a 0.1 weight per cent change in hydrogen in coke will give about a 0.7 weight per cent error in moisture content. You quoted a number of 1.1 weight per cent RMS error for the moisture meter. Do you know how much of that error is due to hydrogen variation?
A.S. PRASAD: This error is the average value we have given. But in the coal the accuracy is still better, about 1 per cent. With coke, if you take a mass sample and use a small container of 20 cm radius, in which you put the coke, when you measure the moisture content it is all right. But once it goes to the plant, where there is a hopper, it has to be measured by probe or it has to be in backscatter form. We have tried this gauge in probe form hanging inside the hopper so that all the time the probe is surrounded by a coke mass, so that the minimum calculated slowdown distance of 20 cm is always maintained. Then the accuracy is quite good.

B.D. SOWERBY: What is the hydrogen content of your coke and how much does it vary?

A.S. PRASAD: The hydrogen content varies from 0.05 to 0.5% and in some cases it has reached 1.2% maximum.

B.D. SOWERBY: That is a very high level.

A.S. PRASAD: Regarding the moisture gauge for coal we have also found difficulties and we are not using it for coal. We use it for coke in the blast furnaces and there we find it quite useful.

T. GOZANI: Under the assumptions that Sowerby mentions, that the hydrogen does not change, or you accept the error and the error can be very serious.

A.S. PRASAD: Yes, that is the assumption that we make, that the combined hydrogen remains constant. For that reason we have to take samples of coke every morning for analysis of hydrogen content, and on that basis we use the neutron moisture meters.
NUCLEAR TECHNIQUES FOR COAL QUALITY ASSESSMENT IN THE UNITED KINGDOM COAL INDUSTRY

D. PAGE
Headquarters Scientific Control, National Coal Board, Harrow, Middlesex, United Kingdom

Abstract

NUCLEAR TECHNIQUES FOR COAL QUALITY ASSESSMENT IN THE UNITED KINGDOM COAL INDUSTRY.

Analytical requirements for coal quality assessment are outlined with specific reference to the United Kingdom coal industry, giving due consideration to the accuracy of measurement and speed of response required to meet planning, commercial and operational needs. A review is made of current applications of nuclear techniques which shows that measurement of ash content is predominant. Developments at present in progress in the National Coal Board are mainly in the field of ash content measurement for operational purposes. Comment is made on the direction of future work which may be required to meet potential changes in operations or markets.

1. INTRODUCTION

The United Kingdom coal industry has been involved in the development and application of nuclear techniques for coal quality assessment for more than 25 years. Interest in the application of suitable processes remains strong and this paper looks at the current situation and indicates possible directions for the future.

2. UK COAL PRODUCTION

Currently, the UK coal industry has a capacity of 100–120 million tonnes per year, of which some 88% is deep-mined coal. About 95% of the coal produced is sold to the home market; 75% of this is used in the public power supply industry and the remainder is fairly evenly divided between the coking, domestic and industrial heating markets.

UK coal-fired power stations are designed, generally, to utilize coal with an ash content of 15–20%. Consequently, coal preparation is primarily structured to meet this requirement. With deep-mined coal ash contents normally falling in the range of 25–40%, the most common preparation process is one which involves
washing three-quarters of the raw coal input (usually the larger particle size material) and blending the resulting washed product with the remaining untreated coal to obtain the specified level of ash content. Domestic coals, which are essentially larger particle size clean coal, and industrial coals are usually prepared at the same plants as part of the overall scheme of beneficiation. Coking coals are fully washed products prepared at a limited number of plants where coal with suitable carbonization properties is available.

3. ANALYTICAL REQUIREMENTS

Analytical data on coal are required in the UK coal industry for three principal purposes:

(a) data on unworked deposits and current production for planning and management
(b) data for commercial evaluation of the products
(c) data which will assist in optimizing the operations in coal production at any stage from the coalface to the final saleable product.

Depending upon the intended purpose, there are differing constraints of accuracy and analysis time which apply. In cases (a) and (b) good accuracy is required but the time is not critical. Prices for coal supplied to power stations, for instance, are usually agreed on the basis of average analyses arrived at over a three-month period. Consequently, the traditional system of sampling, preparation and analysis which may take more than 24 hours can be followed, giving maximum accuracy. For operational purposes, however, such delays are unacceptable and response times ranging from less than one minute to a few hours may be required but, fortunately, in most applications a lower level of accuracy is acceptable.

The choice of which parameters are measured in generally determined by the end-use of the product. Over 90% of British coals are utilized in a combustion process where the prime indicator of quality is heat content. Secondary factors are ash and sulphur contents while limits may also be placed on some other parameters such as moisture, chlorine content, ash fusion and particle size. Similarly the quality of the remaining coals, used for carbonization, having first been selected by reference to coking properties, is defined by heat content, with ash, moisture, sulphur and phosphorus placing limits.

4. PLANNING AND COMMERCIAL ASSESSMENTS

Data acquired for these purposes are subject to similar accuracy and time constraints and are normally obtained by British Standard (BS) methods [1]
at central laboratories. Direct measurement of quality in exploration boreholes using down-hole logging tools is not employed, preference being given to the analysis of recovered cores.

Since the present procedures are acceptable, both from the point of view of accuracy and time delay, introduction of nuclear techniques in the laboratory would only command merit if it made the tasks less labour-intensive. Such techniques would, of course, need to be able to offer the same level of accuracy as conventional methods. Many of the properties required to be analysed, such as coking properties, volatile matter and moisture content, are not readily amenable to nuclear techniques but X-ray fluorescence (XRF) is a possible candidate for some elemental constituents and is under examination within the National Coal Board (NCB) using a vacuum-path, wavelength-dispersive, multichannel spectrometer [2]. Results to date indicate that the technique can offer a reasonable accuracy for sulphur, chlorine, phosphorus and major ash elements, although it does not yet reach the level of the BS methods for all these analyses. This work is being extended and the direct determination of iron pyrites by X-ray diffraction is also under study.

5. ASSESSMENT FOR OPERATIONAL PURPOSES

Time, rather than accuracy, is the key factor in operational applications and it is in this area that the NCB has made most effort in the application of nuclear techniques. There are a number of potential points in the flow of coal from coalface to final product at which knowledge of coal quality could assist in the efficient operation of the process either as input to general management data or even to provide signals directly for automatic control.

UK coals are primarily assessed in terms of heat content (calorific value) which, on a dry, mineral-matter-free basis, is reasonably constant within a limited range of coal composition. Coal mining practice in the UK is such that, generally, a given mine produces only coal within such a limited range. The heat content of the output from a particular mine is then inversely related to the sum of moisture and mineral matter contents. Moisture content range tends to be limited by the operating conditions prevailing at a given point in the process so that the major variable is the mineral matter, which correlates quite well with ash content. Other specifying parameters are usually met by careful selection of the source coal (e.g. for coking properties, sulphur, chlorine and phosphorus contents and for ash fusion characteristics) or by selection within the process stream (e.g. for particle size) where the limits can be controlled by plant design. Consequently, in UK coal practice most operational needs for coal quality assessment can at present be met by the measurement of a single parameter, ash content. One exception is a limited requirement for on-line sulphur content determination.
5.1. Ash content measurement

Without 'real time' measurement of ash content some degree of monitoring and control of the quality of either specific components of a process stream or of the final product can be obtained by setting up, on site, small laboratories in which, using accelerated techniques for sample preparation and ash content determination by incineration, data are obtained in hours rather than days. These procedures may result in some loss of measurement precision but do permit a limited measure of control based on the forward prediction of recent historical data. They also offer economic benefits in the case of those products where strict ash limits necessitate pre-despatch confirmation of analysis by reducing expensive demurrage or product return costs.

With the advent of nuclear techniques offering response times in the order of minutes and the prospect of automatic control, the NCB invested in a number of instruments, particularly for the control of blending for power station fuel. Initially the Cendrex [3] and NCB/AERE Phase 1A [4] instruments, based on the backscatter of low-energy X-rays, were put into use. These instruments offered an accuracy ($\pm 2\sigma$) of about 10% relative, but with the necessity for drying and grinding to 0.5 mm maximum particle size, the effective response times were at least 15 minutes. These delays and the occurrence of severe handling problems in the preparation stages eventually proved unacceptable and no instruments of these types are now operational.

The problems encountered led to the concept of a monitor which could handle larger particle size coal and resulted in the development of the NCB/AERE Phase 3A (Sortex) Ash Monitor [5, 6].

This instrument is designed for continuous operation as an integral part of an automatic sampling system and is fed at a maximum rate of 18 kg/minute with increments taken from the main coal stream. It will accept coal up to 5 mm top size and approximately 10% free moisture content. This moisture limit is somewhat variable, depending upon particle size distribution and to some extent coal rank; with anthracites it is reduced to 7–8%. The measurement principle is that of X-ray backscattering, using approximately 15 keV radiation from $^{238}$Pu. The measurement procedure also includes an intrinsic compensation technique for iron content variations [7]. Given proper attention to the design of the sampling and preparation system, response times of less than 5 minutes can be achieved.

More than 20 of these ash monitors are at present operating; about 15 of them on a regular basis. The major problem that has arisen is the loss of availability due to handling difficulties which have occurred mainly as a result of increases in the fines and moisture contents of mined coal in recent years. All but one monitor are assessing the quality of power station blends, providing data for manual or, in a few cases, automatic control of blending. The exception is one instrument which monitors the quality of a fully washed product for carbonization.

An extension of the use of the ash monitors has been undertaken at five installations where they have been modified to take a microwave moisture meter
At two installations calorific value is determined on a continuous basis by way of the dry, ash-free value. Data obtained here are used for product quality management and to provide early warning to customers of unavoidable deviation from target quality.

As an alternative to the use of a continuous monitoring system which may not always be suitable for technical or economic reasons, a reduction of the time required for analysis in on-site laboratories can be obtained by replacing the incineration method for determining ash content, which takes about one hour, by an X-ray technique. This approach has been adopted in some UK coalfields and approximately 40 low-resolution energy dispersive XRF spectrometers are in regular use. The system is based on X-ray backscattering from $^{238}$Pu with iron compensation as in the Phase 3A Monitor. It uses small (30 g) samples of 0.2 mm particle size coal. The average accuracy ($\pm 2\sigma$) is about 8% relative and the measurement time two minutes.

A second approach used was the adaptation of the Phase 3A monitoring system to an instrument designed for batch analysis of 5-0 mm wet coal. This instrument, the NCB Rapid Ash Analyser [10] is a free-standing self-contained unit in which a 12 kg sample of coal is transported from an inlet hopper, by means of a small flat-bed conveyor belt underneath a suitably adapted Phase 3A measuring head. Measurement time is less than one minute and accuracy comparable to the Phase 3A Monitor. Although this instrument was designed to avoid the necessity for drying and fine-grinding of the sample, it is subject to the same handling problems as the Phase 3A Monitor and only four are in regular use.

Data on final product quality in a plant producing power station coal offers a means of controlling the actual blending process; a similar 'feedback' system of control may also be applied to the cleaning of fine coal by froth flotation. In this particular case the efficiency of the process may be judged from a knowledge of the ash content of the dirt in the effluent (tailings). On-site laboratory measurements by incineration have been widely used in the past and the NCB is currently testing the prototype of a nuclear technique, developed by the Atomic Energy Research Establishment, Harwell, for the direct determination of ash content in the tailings solids [11]. This process is applied directly to the liquid effluent concentrate and uses a $^{137}$Cs transmission gauge in combination with a $^{109}$Cd backscatter system. The process measures the ash content of the solids in the tailings to $\pm 4\%$ absolute.

5.2. Sulphur content measurement

There is only a limited call in the UK coal industry for the measurement of sulphur content for operational purposes. At present there are two installations where on-site sulphur content measurement is made. Data obtained are used to ensure that the products meet the sulphur content limits specified for a premium carbonization market or are diverted to a second outlet with less stringent requirements. The technique used involves the generation and measurement of iron-K
fluorescent radiation which can be correlated with the total sulphur content of coal [9, 12]. The accuracy of sulphur content measurement of washed coals by this technique is of the order of ±0.1%. At one installation a Phase 3A Ash Monitor has been modified and measures ash and sulphur content on a continuous basis; at the second a Rapid Ash Analyser is used.

6. CURRENT DEVELOPMENTS

Within the NCB, developments are continuing in the field of instrumentation for on-line ash monitoring, both to counter the limitations found in existing systems and to meet possible future requirements.

An alternative system of coal presentation to the Phase 3A Monitor has been developed and a prototype is under test. This monitor uses the same kind of measuring head as the Phase 3A but employs a positive feed mechanism which can handle wet coal up to 25 mm particle size.

The majority of products made have a maximum particle size of about 50 mm and it would be preferable if they could be interrogated without any sampling or preparation. Instruments for measurement of coal ash directly on a conveyor belt in both transmission [13, 14] and backscatter geometries [15] have been developed elsewhere. Similar gamma transmission [16] and backscatter instrumentation has been developed within the NCB and is under test; the problem of compensation for iron content variations in ash composition is also under study. Finally, a system which measures the natural gamma radiation from shales and which is capable of distinguishing qualitatively between coal and stone has been developed [17]. The possibility of adapting this system for the approximate determination of the ash content of raw coal is being considered.

One other area in which active research work is in progress is that of prompt gamma neutron activation analyses. A laboratory facility has been set up and a study programme on neutron/gamma interactions in coal is being pursued [18].

7. FUTURE REQUIREMENTS

The emphasis on improvements in ash-measuring techniques in current NCB developments has been illustrated and will remain a dominant feature for some time. Improvements being sought are not so much those of increased accuracy and shorter response times, although these could be necessary for some applications, but of simplicity in application and low capital requirements. A number of other potential requirements have been identified for operational or even commercial purposes.

One potential growth area in the UK coal industry in the near future is that of the 'small user' industrial market. It will be vital in order to gain and retain
customers that the quality of relatively small consignments, perhaps even as little as 30 tonnes, is within specification. This would entail rapid and accurate pre-despatch analysis for ash, moisture and/or calorific value. Sulphur content may also be a limiting requirement.

Knowledge of the ash content of run-of-mine coal could usefully be integrated with other mining operations data to assist in the optimization of coalface operations. This application would not require high accuracy but would need rapid response from the direct interrogation of the product on the mine output conveyor. Similar constraints would apply to the raw coal input to a washery for 'feedforward' control of plant operation.

Future limitations on environmental pollution could require the identification of power station fuels high in sulphur and other pollutants to allow blending with cleaner coals or the routing of supplies to combustion units specifically designed to handle them. Such restrictions may necessitate rapid response instruments for pre-despatch analysis.

New utilization procedures for coal, such as gasification or liquefaction, may call for tight specifications for ash or some ash elements which are not at present routinely measured.

On the commercial side, changes in the pricing structure for power station fuel or tighter specification limits on parameters such as ash content to obtain better consistency in quality, could mean a wider deployment of monitors to control blending processes. The possibility of using data from on-line monitors rather than the present conventional sampling and analysis procedures for commercial evaluation has been suggested [19]. If the present accounting procedures for power station fuel continued, although high short-term measurement precision and rapid response would not be required, it would be essential to obtain high accuracy over the longer accounting periods.

8. CONCLUSIONS

A variety of nuclear techniques for coal quality assessment is in use in the UK coal industry, with the emphasis being on obtaining data for operational purposes. Ash content is the most widely required parameter and current developments are concentrated on instrumentation for its measurement, but the possibility of other requirements arising from changes in market or operational needs is recognized.

REFERENCES

DISCUSSION

S. CIERPISZ: I have a question in relation to the measurement of ash content in large volumes of coal loaded into train wagons. As you showed, ash content in coal varies and probably the amount of coal in the main stream varies with time. Do you think you could get better results applying, in parallel to your ash monitor, a belt weigher for average values of ash content in large volumes over, say, several minutes?

D. PAGE: Yes, indeed. In fact that is one of our proposals, particularly for development of the natural gamma system and possibly the two gamma energies system.

S. CIERPISZ: What is your opinion about the application of ash monitors to control washing processes?

D. PAGE: We have not used them for washery control. The applications we have made have been for blending control, and there we find we can get them to work but the time required with our present systems, about 10 or 15 minutes' response time,
is perhaps a little too long to give us good feedback control on a blending scheme. We would like to reduce that measurement time.

T. GOZANI: In our rapid ash meter we are weighing the results with the flow rate so you get the weighted ash value. On the issue of controlling a washery there are several programmes that are planning this. One is in Homer City, using the facilities they have right now to look at each unit, heavy media or cyclone, and using basically the density.

D. PAGE: Indeed we do that. The reason I did not mention it is that I did delineate that I was talking about coal quality measurement and I said that measurement of density is an operational application which falls outside the scope of my paper. We do use density meters in slurries on some washery operations.

J.S. WATT: You talked about work on coal slurries, and particularly putting the gauge in the tailings stream. What use can you make of that information for control? Because as far as I can see you have a tailings measurement, you don’t know what has come into the flotation bank and so it seems very hard to know what to do with the information.

D. PAGE: The technique that has been used quite widely for many years enabled the operation of a fine coal cleaning screen, froth flotation essentially, making the necessary adjustment to the frothing conditions, etc., to make sure that you were taking the coal out at the optimum cut point between ash in the froth-cleaned coal and the ash in the waste going out.

J.S. WATT: Are those measurements taken not only from the tailings stream but also from the feed or the concentrate?

D. PAGE: Yes. But this particular application was designed for the tailings stream.
DETERMINATION OF ASH CONTENT OF COAL ON-LINE ON CONVEYORS AND IN-STREAM IN COAL SLURRIES

J.S. WATT
Commonwealth Scientific and Industrial Research Organization,
Division of Mineral Physics,
Lucas Heights Research Laboratories,
Sutherland, New South Wales,
Australia

Abstract

DETERMINATION OF ASH CONTENT OF COAL ON-LINE ON CONVEYORS AND IN-STREAM IN COAL SLURRIES.

Australian work on the determination of ash content of coal on conveyors, based on the combination of measurements of transmission of low- and high-energy γ-rays, is reviewed. Root mean square differences between gauge and chemical assay are 0.3-0.45 wt% for low-ash coal, and usually 0.7-1.5 wt% for as-mined coal of high ash content. The gauges are in routine use in the Australian coal industry. Techniques have been developed for the determination of the solids weight fraction (W) and ash content of coal in slurries of variable voidage. The technique is based on the correlation between solids weight fraction and hydrogen concentration (wt/wt) of the slurry. The technique combines measurements of neutron moderation, γ-ray transmission, X-ray backscatter and iron K X-ray excitation. In laboratory experiments with coal slurries of 5-20 wt% solids, 21-30 wt% ash, and voidage 0-4 vol.%, RMS differences between gauge and conventional assays were 0.54 wt% solids and 0.78 wt% ash.

1. INTRODUCTION

Continuous determination of the ash content of coal on conveyors and in slurries is required for the better control of coal washeries. Control based on this information should lead to an increased yield of coal at constant product ash content. Savings to Australia, assuming a 1% increase in yield, would be about A $30 million per year for export coal alone.

Coal consists of coal matter (mainly carbon with some hydrogen, oxygen and nitrogen) and mineral matter (mainly aluminium and other silicates with some iron). Coal ash is the oxidized incombustible residue from the combustion of coal and is closely correlated with the mineral matter content.

Coal ash gauges using X- and γ-ray interactions [1-6] are based on the dependence of these interactions on atomic number Z and the fact that the mean Z of ash is considerably higher than that of the coal matter.
The Commonwealth Scientific and Industrial Research Organization (CSIRO) Division of Mineral Physics has developed two ash gauges: low-energy \textit{$\gamma$}-ray transmission (LET) and pair production (PP). These SIROASH gauges are also called, respectively, the Coalscan through-belt ash monitor and the Coalscan by-line ash monitor. These gauges, which are in routine use in the Australian coal industry, are manufactured and marketed by Coalscan Pty Ltd\textsuperscript{1}.

The Division is developing nuclear techniques for determining the solids weight fraction and ash content of coal in slurries\textsuperscript{7—9}. These techniques are to be applied to slurry streams in the flotation circuit of coal washeries which normally have variable voidage due to entrained air.

This paper reviews the Australian work on the SIROASH LET gauge for on-line determination of ash content of coal on conveyors. It also describes the technique and the laboratory testing of the SIROASH coal slurry gauge being developed for the in-stream determination of solids weight fraction and ash content of coal in slurries.

2. ON-LINE CONVEYOR

2.1. Technique

In the LET gauge (Fig. 1), ash content is determined by combining measurements of intensities of narrow beams of low- and high-energy \textit{$\gamma$}-rays transmitted through the coal\textsuperscript{2, 3}. The transmission of both low- and high-energy \textit{$\gamma$}-rays depends on weight per unit area of coal in the \textit{$\gamma$}-ray beam; the transmission of low-energy \textit{$\gamma$}-rays also depends on atomic number of the coal.

The ash content $C_{\text{ash}}$ is given by

$$C_{\text{ash}} = a_1 \frac{\log (I/I_0)'}{\log (I/I_0)''} + a_2$$

(1)

where $I$ and $I_0$ are intensities detected with and without coal being present, superscripts $'$ and $''$ refer respectively to the low- and high-energy \textit{$\gamma$}-rays, and $a_1$ and $a_2$ are constants. Errors in ash determination are introduced when the coal weight per unit area varies considerably during the measurement time, e.g. when coal is on a moving conveyor. This is overcome by determining the ratio of the log terms in Eq. (1) over short periods, e.g. tens of milliseconds, and averaging these ratios over much longer periods, e.g. tens of seconds\textsuperscript{2}.

\textsuperscript{1} Coalscan Pty Ltd, PO Box 64, Unley, South Australia 5061.
FIG. 1. The low-energy γ-ray transmission (LET) gauge which determines ash content of coal on conveyors by combining measurements of intensities of low- and high-energy γ-rays transmitted through the coal.

2.2. The ash gauge

A schematic diagram of the on-line SIROASH LET gauge is shown in Fig. 1. The radioisotope γ-ray sources are $^{241}\text{Am}$ (60 keV) and either $^{133}\text{Ba}$ (356 keV) or $^{137}\text{Cs}$ (662 keV). The detected intensities of γ-rays at these energies are determined separately by pulse height analysis of the output pulses from the scintillation detector. The sources and detector are mounted at opposite ends of a C-frame about the conveyor belt. The C-frame is swung under computer control so that the γ-ray beam either traverses coal at the centre of the belt, or is beyond the edge of the belt during standardization and calibration.

The gauge records ash content averaged over any required period: tens of seconds, an hour, a shift or a day. The ash content is independent of coal thickness.
FIG. 2. Calibration curve of the LET gauge installed on the coking coal product line of the Stockton Borehole washery. Samples of both coking and steaming coal product were used to calibrate the gauge.

and of the vertical segregation of coal on the belt. The gauge averages ash content over large quantities of coal, e.g. one tonne per 100 seconds, assuming a belt speed of \(3\,\text{m}\cdot\text{s}^{-1}\) and about 100 mm thickness of coal traversed by the \(\gamma\)-ray beam.

The gauge is calibrated using approximately one kg samples of coal which have been assayed for ash content by conventional methods. This calibration is undertaken with the C-frame swung to the 'off-belt' position. Figure 2 shows the calibration for the on-line gauge in routine use at the Stockton Borehole washery near Newcastle, New South Wales. The gauge is mounted about the coking coal product conveyor. Samples from both the coking and the steaming product lines have been used in the calibration to widen the range of ash contents.

The gauge is standardized automatically using a standard absorber to intersect the \(\gamma\)-ray beam when in the off-belt position. The C-frame can be swung off-belt at regular intervals so that the calibration equation can be automatically corrected to compensate for any drift in the electronic equipment.

2.3. Accuracy of ash determination [3]

Errors in the determination of ash content of high-ash coals are mainly caused by variations in the concentration of constituents, particularly \(\text{Fe}_2\text{O}_3\), in the ash. This is also an important cause of error for low-ash coals but, since the absolute error in ash is proportional to ash content, the magnitude of this error is much smaller. The error in measurement of count rate can introduce significant errors
in ash determination when the weight per unit area of coal (in the \(\gamma\)-ray beam) is low, e.g., 5 g cm\(^{-2}\).

The errors in ash determination due to a 0.1\% error in measurement of count rate are shown in Fig. 3. It can be seen that the absolute error (wt\% ash) varies little with ash content and is somewhat lower when 356 keV \(\gamma\)-rays are used. The error in ash decreases rapidly with increasing weight per unit area (\(\rho x\)) and is very small when \(\rho x \geq 10\) g cm\(^{-2}\). There is a lower limit to \(\rho x\) where the error in ash is too great for the specific application. This lower limit depends on several factors: the absolute error acceptable in ash, the error in measurement of count rate, and the composition of ash in the coal. Current practice in Australia for low-ash coals is to reject ash determination when \(\rho x \leq 5\) g cm\(^{-2}\). If \(\rho x\) is frequently less than 5 g cm\(^{-2}\), the conveyor speed should be reduced to allow the coal to be more deeply packed on the conveyor. Further work is being undertaken to determine the accuracy which can be obtained for 3 g cm\(^{-2}\) \(\leq \rho x \leq 5\) g cm\(^{-2}\).

The effect of varying the concentration of water in coal has been shown by calculation [3] to be small; a change from 5 to 10 wt\% water in the coal results in an error of less than 0.3 wt\% ash over the ash range of 8 to 20 wt\%.

Variations in concentration in the ash of Fe\(_2\)O\(_3\), and occasionally of barium and CaO, are the main cause of errors in ash determination due to changes in ash composition. For example, errors resulting from the addition of 1 wt\% Fe\(_2\)O\(_3\) to the ash at the expense of silica are 0.50 and 1.26 wt\% ash respectively at 8 and 20 wt\% [3]. The magnitude of this error depends on the mean Fe\(_2\)O\(_3\) concentration of the ash which was assumed, in this case, to be 5 wt\%.

A knowledge of variations of constituent concentrations in the ash is necessary to assess errors in ash determinations for any specific application of on-line gauging.
## TABLE I. DETAILS OF COMPOSITION OF COAL SAMPLES AND RMS DIFFERENCES BETWEEN
LET GAUGE MEASUREMENTS AND CHEMICAL ASSAY OF ASH

<table>
<thead>
<tr>
<th>Samples from</th>
<th>Type of coal</th>
<th>Feed or product</th>
<th>Source of coal</th>
<th>No. of samples</th>
<th>Ash (wt%)</th>
<th>RMS difference (wt% ash)</th>
<th>Fe$_2$O$_3$ (wt% in ash)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Vales Point, NSW</td>
<td>Steaming</td>
<td>Feed</td>
<td>Conveyor</td>
<td>22</td>
<td>16-23</td>
<td>0.99</td>
<td>3.0-6.3</td>
</tr>
<tr>
<td>Liddell, NSW</td>
<td>Steaming</td>
<td>Feed</td>
<td>Conveyor</td>
<td>18</td>
<td>22-33</td>
<td>1.20</td>
<td>4.7-9.3</td>
</tr>
<tr>
<td>Leigh Creek, SA</td>
<td>Steaming</td>
<td>Feed</td>
<td>Conveyor</td>
<td>24</td>
<td>16-50</td>
<td>1.59</td>
<td>1.6-6.9</td>
</tr>
<tr>
<td>Blackwater, Qld</td>
<td>Coking &amp; Steaming</td>
<td>Feed</td>
<td>Mine</td>
<td>24</td>
<td>5-29</td>
<td>2.24</td>
<td>4.3-27</td>
</tr>
<tr>
<td>Newcastle, NSW</td>
<td>Coking</td>
<td>Product</td>
<td>Sampler</td>
<td>68</td>
<td>10-13</td>
<td>0.36</td>
<td>3.7-5.7</td>
</tr>
<tr>
<td>Port Kembla, NSW</td>
<td>Coking</td>
<td>Product</td>
<td>Sampler</td>
<td>48</td>
<td>11-14</td>
<td>0.36</td>
<td>2.8-5.4</td>
</tr>
<tr>
<td>West Cliff, NSW</td>
<td>Coking</td>
<td>Feed</td>
<td>By-line</td>
<td>94</td>
<td>12-21</td>
<td>0.36</td>
<td>2.8-5.4</td>
</tr>
<tr>
<td>West Cliff, NSW</td>
<td>Coking</td>
<td>Product</td>
<td>By-line</td>
<td>27</td>
<td>9.7-10.6</td>
<td>0.31</td>
<td>3.3-4.3</td>
</tr>
<tr>
<td>Gregory, Qld</td>
<td>Coking</td>
<td>Product</td>
<td>Sampler</td>
<td>72</td>
<td>7.4-9.4</td>
<td>0.40</td>
<td>~6</td>
</tr>
</tbody>
</table>
The magnitude of these variations depends on the coal seam, the section of seam from which the samples are taken, and the weight of each sample. Since the LET gauge averages ash over multitonne quantities of coal on a conveyor belt, knowledge of variations in concentration of ash constituents from one multitonne sample to another is required over an extended period, e.g. several months. The coal industry does not have this information. In practice, errors in ash have been determined experimentally both in the laboratory using coal samples, and on-line in plants.

2.4. Laboratory measurements on coal samples

Measurements of $^{241}$Am and $^{133}$Ba $\gamma$-ray transmission have been made on each coal sample of the sets of samples detailed in Table I. Samples were collected from mines and power stations in several parts of Australia. The Blackwater samples were obtained from over a wide area of the mine site. The other sets were obtained over periods of one to six months from a conveyor or sample by-line. Samples from the first four locations (Table I) were subsampled to 100 kg, placed in a plastic container, and then scanned by the $\gamma$-ray beams to determine average ash content over 50 kg of coal [3]. The coal particle size of these large coal samples was $-25$ mm. Samples from the last four locations were crushed to $-3$ mm, representatively sampled, and measurements averaged over 1 kg of coal sample.

For the large samples, RMS differences between gauge ash and the chemical assay for ash vary between 1 and 2.2 wt% ash (Table I). The high error of 2.2 wt% ash for the Blackwater samples was caused by the unusually wide range of 4 to 27 wt% $\text{Fe}_2\text{O}_3$ in the ash. The Leigh Creek coal is of poor quality, and most of the error in ash determination is caused by variations of both $\text{Fe}_2\text{O}_3$ and CaO in the ash. The Liddell and Vales Point samples are more typical of steaming coals in Australia: RMS errors are 1.2 and 1.0 wt% ash.

Root mean square differences between gauge ash and chemical assay for ash for the sets of small coal samples are in the range 0.3 to 0.4 wt% ash (Table I). A significant part of this error is due to sampling and chemical assay, i.e. not to a gauge error. For the combined sets of Newcastle and Port Kembla samples, the inherent error of the gauge technique has been estimated to be $0.25 \pm 0.05$ wt% ash [3]. These errors (Table I) are small because of the low ash content of the samples and because of the small range of variations of $\text{Fe}_2\text{O}_3$ in the ash.

2.5. On-line ash determination

Table II summarizes the results of on-line measurements of coal on conveyors. The first was a plant trial with measurements being made over two months in 1981 [2]. The Stockton Borehole installation was an advanced prototype LET gauge. The others have the Coalscan through-belt ash monitor, i.e. a LET gauge (Fig. 4). Gauge results have been compared with chemical assay of samples taken from the
<table>
<thead>
<tr>
<th>Installation at</th>
<th>Type of coal</th>
<th>Coal on conveyor</th>
<th>Ash (wt%)</th>
<th>RMS difference in ash (wt%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>BHP Minerals washery near Gregory, Qld</td>
<td>Coking</td>
<td>Washery product</td>
<td>7.4–9.4</td>
<td>0.45</td>
</tr>
<tr>
<td>Stockton Borehole washery near Newcastle, NSW</td>
<td>Coking</td>
<td>Washery product</td>
<td>6–9</td>
<td>0.32</td>
</tr>
<tr>
<td>Hebden Mining Co. near Liddell, NSW</td>
<td>Steaming</td>
<td>As mined</td>
<td>26 (mean)</td>
<td>1.3</td>
</tr>
<tr>
<td>Muswellbrook Coal near Muswellbrook, NSW</td>
<td>Steaming</td>
<td>As mined</td>
<td>6–15</td>
<td>No results yet available</td>
</tr>
</tbody>
</table>
FIG. 4. The Coalscan through-belt ash monitor (a LET gauge) installed about the mine output conveyor of the Hebden Mining Co. near Liddell, Australia.

belt by an automatic sampling system. The gauge ash has been averaged over the period corresponding to the time taken to collect sufficient samples to obtain a representative sample from the conveyor.

The plant measurements (Table II) for low-ash coals include a significant error due to sampling and chemical assay, particularly for the trial at Gregory. The result for the high-ash coal at Hebden is about the same as that obtained in laboratory measurements on Liddell coal samples (Table I) which were from the Ravensworth mine located close to Hebden.

2.6. Summary of accuracy of ash determination

The laboratory and plant RMS differences agree well for both low- and high-ash coals. The RMS difference for low-ash coals is in the range 0.3–0.45 wt% ash (1σ), and contains a significant error due to sampling and conventional assay. The RMS error for intermediate- and high-ash coals varied in the range 0.36 to 2.24 wt% ash. The lowest error corresponded to intermediate ash content coal (12–21 wt% ash) with little variation of Fe₂O₃ in the ash. The highest error corresponded to coal with large variations in Fe₂O₃ in the ash (4.3 to 27 wt%). For high-ash coals, the RMS error in ash determination is normally expected to be 0.7–1.5 wt% ash.

2.7. Comparison of the two Coalscan ash gauges

The Coalscan through-belt (LET) and sample by-line (pair production [6]) ash monitors are complementary in application. The through-belt ash monitor has the advantage of being able to measure coal directly on the conveyor, a short
measurement time (tens of seconds), and lower cost. Both ash monitors accurately determine the ash content of low-ash coals. The sample by-line monitor is considerably less sensitive to variations in composition of ash and hence more accurate for high-ash coals.

3. COAL SLURRIES

The SIROASH coal slurry gauge determines solids weight fraction (W) and ash content of coal in slurries independently of variable voidage [7–9]. The technique depends on the correlation between W and hydrogen concentration (wt/wt) of the slurry. This concentration is determined by combining measurements of neutron moderation and γ-ray transmission. The solids weight fraction and ash content of coal in the slurry are then determined by combining the hydrogen concentration (wt/wt) with ash-sensitive measurements, e.g. backscatter of X-rays coupled with iron K X-ray excitation, or transmission of low- and high-energy γ-rays [2, 4].

3.1. Technique

The solids weight fraction of a coal slurry can be expressed in terms of hydrogen concentration H (wt/wt) of water (ω), slurry (sl) and coal matter (cm) by

\[ W = \frac{(H_\omega - H_{sl})}{(H_\omega - H_{cm}[1 - C_{mm}])} \]

where \( C_{mm} \) is the concentration of mineral matter in the coal and \( C_{cm} + C_{mm} = 1 \). Ash and mineral matter concentrations are closely correlated. \( H_\omega \) is constant and \( H_{cm} \) can be assumed to be constant for coal from the same seam.

Over a limited range of ash or mineral matter concentrations, the solids weight fraction can be determined from Eq. (2) with sufficient accuracy simply by determining \( H_{sl} \) [8]; this can be done by combining measurements of neutron moderation and γ-ray transmission. Over a wider range of ash concentrations, the solids weight fraction can be determined accurately only if an ash-sensitive measurement is also made.

The detected intensities measured using the probes (Fig. 5) and associated electronics are:

(a) \( I_{Am} \) and \( I_{Ba} \), respectively, of \( ^{241}Am \) (60 keV) and \( ^{133}Ba \) (~356 keV) γ-rays transmitted through the coal slurry
(b) \( I_\gamma \) and \( I_{Fe} \), respectively, of scattered \( ^{238}Pu \) X-rays (~17 keV) and X-rays in the iron K X-ray channel, and
FIG. 5. Immersion probes used in experimental measurements on coal slurries:

(a) $\gamma$-ray transmission

(b) X-ray backscatter and iron K X-ray excitation

(c) Neutron moderation.

(c) $I_n$ of slow neutrons originating as fast neutrons in the $^{241}\text{Am}/\text{Be}$ neutron source and slowed down to thermal and near-thermal energies, mainly by hydrogen atoms in the slurry.

The hydrogen content (wt/wt) of the slurry is given by [10]

$$H_{sl} \approx \frac{a_1 I_n + a_2 I_n^2 + a_3}{\log (I_0/I)_{Ba}} + a_4 \quad (3)$$

For slurries with variable voidage, the neutron measurement compensates the $\gamma$-ray transmission measurement so that $H_{sl}$ is determined independently of voidage. The subscripted values of $a$, $b$, $c$ and $d$ in this and Eqs (4-6) are constants.

Solids weight fraction can be determined directly from $H_{sl}$ when variations of ash in the coal are small. When ash varies considerably, solids weight fraction is given by

$$W \approx b_1 H_{sl} + b_2 A + b_3 \quad (4)$$

where $A$ is the ash-sensitive measurement. For low- ($^{241}\text{Am}$) and high- ($^{133}\text{Ba}$) energy $\gamma$-ray transmission,
FIG. 6. Two of the radioisotope immersion probes mounted on a steel framework, the inner part of which is immersed into the 200 L drum of coal slurry.
When $^{238}$Pu backscattered X-rays and X-rays in the iron K X-ray channel are used,

$$A = c_1 \log(I_0/I)_{Am}/\log(I_0/I)_{Ba} + c_2$$  \hspace{1cm} (5)

or can be determined directly from the measured count rates. A similar equation but with different values of constants is used to determine the ash content of coal in the slurry.

### 3.2. Experiments

The probes (Figs 5 and 6) were immersed directly into coal slurry in a 200 L drum. Slurries were made up of four steaming coal samples taken from input conveyors at the Liddell and Vales Point power stations. The samples were ground to ~1 mm, and made up into slurries having solids weight fractions between 5 and 22 wt%. Voidage of the slurry was varied in steps between 0 and 4 vol.% by injection of air from a compressed air line through a flexible tube coiled inside the base of the drum and pierced with fine holes.

The neutron count rate is slightly affected by the temperature of the slurry and a correction was made to compensate for this.

### 3.3. Results

Figure 7 shows results of the determination of hydrogen concentration (wt/wt) using neutron moderation and $^{133}$Ba $\gamma$-ray transmission measurements. The hydrogen concentration was determined to 0.3% relative for slurries without voidage and to 0.4% relative for slurries with voidage in the range 0 to 4 vol.%.

Figure 8 shows $W$ determined from the combined probe measurements plotted against $W$ determined by sampling and weighing. The RMS difference is 0.54 wt% solids, compared to 2.78 wt% using $^{133}$Ba $\gamma$-ray transmission alone, and 0.4 wt% from the combined probe signals for slurries without voidage.

Figure 9 shows the ash content of coal determined by combined probe measurements plotted against ash content determined by chemical assay. The RMS difference is 0.78 wt% over the ash range of 20.7 to 30 wt%.

Plant trials of the above technique are currently (December 1984) being undertaken at the BHP Steelworks washery at Newcastle, Australia.

### 4. CONCLUSIONS

On-line conveyor ash gauges based on low-energy $\gamma$-ray transmission techniques are in routine use in Australian industry. Root mean square differences
FIG. 7. Determination of hydrogen concentration (wt/wt) of coal slurries based on measurement of neutron and $^{133}$Ba γ-ray count rates. Voidage was varied between 0 and 4 vol.%. A single point is indicated by *, and if more than one point is superimposed, the number (e.g. 3) of superimposed points is given.

FIG. 8. Solids weight fraction determined by nuclear technique and by conventional assay of sampled slurry, for slurries of 0 to 4 vol.% voidage.
between gauge ash and chemical assay ash are 0.30 to 0.45 wt% ash (1σ) for low-ash coals, and usually 0.7 to 1.5 wt% ash for high-ash coals.

Techniques for determining the solids weight fraction and ash content of coal in slurries of variable voidage have been proved in laboratory trials and plant trials are now in progress.

ACKNOWLEDGEMENTS

The results relating to the Stockton Borehole washery (Fig. 2 and Table II) were obtained in a joint project between CSIRO and the BHP Steelworks, Newcastle, Australia. All work was supported, in part, by a grant from the National Energy Research Development and Demonstration Council of the Government of Australia.

REFERENCES


DISCUSSION

D. PAGE: You showed that by comparing the gauge results with the conventional sampling results over a long time, you were getting essentially the same pattern of variation, which indicates that what the gauge was doing was adequately representing, or at least to the same degree representing, the quality of the total coal as the conventional sampling does. What, if anything, do you do to ensure that you don't have any problems of segregation transversely on the belt?

J.S. WATT: That is a very good question and one of the things that, when we are developing the gauges, we are quite worried about. The obvious way, and our first field trial, used two gauges: one which was kept at the centre and the other which could be varied across the belt. In that case we were pleasantly surprised to find there was not significant segregation across the belt. The fact that the chemical assay of sampled coal agrees well with the gauge reading suggests that segregation across the belt is not such a problem. If it is a problem then you just make measurements at more than one place across.

S. CIERPIEZ: What is your estimation of errors of the classical methods of measuring ash content by combustion, taking into account errors due to sampling and errors of the method itself? Secondly, your monitor also measures ash content in coking coals. Variations of ash content in coking concentrates are very small, I think around 1%, and the accuracy of the radiation method is about 0.4 to 0.5%. Is it really possible to control the washing process with such accuracy?

J.S. WATT: In relation to errors in conventional assay techniques it depends very much on whether you have a well-established laboratory making the measurements. In all our cases errors are based on ash measurements made at the plant.
itself and there is no doubt the accuracy there is much worse than you would get in a good standards laboratory. We have had errors of the order of 0.2–0.25 wt% ash. That is only the chemical side. The sampling is open to even more problems. The long-term installation at Liddell power station was giving 1.3 wt% ash error but there were serious doubts about the sampling validity. We have been basing our techniques on measurements made at the mine, and certainly these aren’t so good, but I would have thought that 0.25, 0.3 and even 0.4 could be possible.

The second question is whether an RMS deviation of say 0.3 wt% is sufficient: it is certainly sufficient to make measurements about the plant. Say if you want to correlate an ash gauge with the density of a heavy medium, you can certainly quite accurately calculate the effect of a change in density of the heavy medium on ash content. That is probably the most important parameter that you want to know in a plant.

S. CIERPISZ: Have you any experience with a closed loop control system?

J.S. WATT: Yes, there is one closed loop system built in at the Ulan mines. After a lot of very heady celebration because it seemed to be working, long-term trials showed that the operators can control the system better than the closed loop, which has now been taken out of operation. There is a lot of interest in closing the loop and I don’t think there is much more work needing to be done to achieve this.

T. GOZANI: We have looked carefully at the accuracy of the traditional chemical measurement, because all our instruments are calibrated against these methods. To our surprise, when you compare several laboratories over a wide range of coals in a controlled experiment, we have seen biases and more surprising systematic error above the bias of the order of 6% relative. This is of a range of ash values 4 to 30%. If you look at one laboratory, they are much more consistent. Concerning the comparison between the pair production and the transmission gauge, one should realize that the cost in going to pair production is less sensitivity to ash. If you reduce the Z dependence you reduce the sensitivity to ash. In the extreme if you have no sensitivity to Z you have no sensitivity to iron, no sensitivity to ash altogether. So one has to weight that also. When you are looking at low-level ash you need all the Z dependence you can get in order to have sensitivity to ash. When you have high ash you can afford to lose the Z dependence. I am sure Sowerby will answer that fully in his paper.

J.S. WATT: With modern electronic equipment you can measure stably quite small changes and certainly over periods of at least months the determination to 0.3 wt% at 15 wt% has been quite possible. So this is down to the errors of chemical assay and sampling. Therefore I don’t think that you can say the sensitivity is insufficient.

A.S. PRASAD: Regarding the gamma transmission system, I feel that is the right thing for coal industries where the ash content of coal is very high. In India, when the ash content is very high the gamma ray transmission system is useful. But when the material is on the belt is it the screened material or the directly
mined material? Because the material must be in a uniform size to give the correct reading.

J.S. WATT: Obviously if you could collimate perfectly and use a very short time period for measurement, it wouldn’t matter at all what the particle size was. In practice so far we haven’t had enough error due to particle size to cause us worry in the measurement. And really we are working at fairly low count rates at present; we could go to shorter times and higher count rates by at least a factor of 3, by putting in good, fast electronics. So far it hasn’t been a problem.

A.S. PRASAD: We have found that when coal is crushed to -5 mm size, or produced as fines in a mechanical mining system, this -5 mm coal size has ash content higher than the bulk size. It is entirely different from the general picture of world coal where the finer size should have less ash content. So if more fines are produced naturally we will get a wrong reading for the ash content because fines have to go for washing, whereas the screen fraction up to +10 mm can be directly shipped to the coking plant site.

J.S. WATT: It really depends a lot on how you make your measurement. If you take a very collimated beam, as we do, and take very small measurement times, I don’t really think it is a problem.

P.L. SURMAN: I think a good touchstone here, and one that we’ve used in assessing these devices, is that an on-line device should perform as well as the conventional systems of sampling and analysis. British Standards quote for ash a precision for estimation of a single consignment of 1% — that is at the 2σ level. So at your RMS level that would be the same as 0.5%. So I think the precision of estimation of a single variety does meet our needs. Perhaps where we would have difficulty on a power station, where we receive a whole mixture of coals, is that then precision will deteriorate. Perhaps you will be talking of 1.5% RMS, or 3% on a 2σ basis. So it does depend on the application. For some process control applications on plant, where for instance if we wish to reject abnormal deliveries, then that might be quite adequate.

J.S. WATT: If you are taking coal from a lot of different places as you do in a power station, that is an ideal place for the pair production gauge, which is less sensitive to error, by a factor of 4, as regards chemical compositional changes than the LET technique. So really this is part of the complementary nature of the two gauges.

T. GOZANI: Did you ever try to use a conductivity gauge to measure solid fraction?

J.S. WATT: No. I was very interested in your measurements of this. I take it the conductivity would change with voidage just the same?

T. GOZANI: Oh yes. Voidage has to be taken care of by other means. Voidage is like adding another variable into your set of equations. If conductivity works then it is a direct measurement. You don’t make any assumptions about the constancy of the hydrogen-coal ratio or others of that kind. But it is a problem; it’s not trivial.
DETERMINATION OF ASH, MOISTURE AND SPECIFIC ENERGY OF COAL

B.D. SOWERBY
Commonwealth Scientific and Industrial Research Organization,
Division of Mineral Physics,
Lucas Heights Research Laboratories,
Sutherland, New South Wales,
Australia

Abstract

DETERMINATION OF ASH, MOISTURE AND SPECIFIC ENERGY OF COAL.

On-line ash gauges based on the pair production (PP) technique are in routine use on high-throughput sample by-lines in the Australian coal industry. Root mean square differences between PP gauge ash and chemical assay ash are in the range 0.21 to 0.34 wt% ash for on-line measurements on coal of up to 20 wt% ash. The PP technique is about a factor of 4 less sensitive to variations in ash composition than the alternative lower-energy $\gamma$-ray techniques. A number of techniques are being investigated by the Commonwealth Scientific and Industrial Research Organization for the on-line determination of moisture in coal. These include capacitance, microwave, neutron transmission and scattering, nuclear magnetic resonance and infra-red reflectance. One promising development is the non-contacting under-belt capacitance gauge for on-line conveyor belt determination of coal moisture. Laboratory experiments have shown that neutron techniques can be used to determine accurately the carbon and specific energy content of coal. Nuclear techniques have been used to determine coal mass flow to an industrial reverberatory furnace.

1. INTRODUCTION

There are large potential savings from the use of on-line analysis techniques in the coal industry, particularly in the control of coal washeries and in the more efficient operation of coal-fired power stations. Three important parameters for which on-line analysis is required are ash, moisture and specific energy.

The Commonwealth Scientific and Industrial Research Organization (CSIRO) Division of Mineral Physics has developed two techniques for the on-line determination of ash in coal, namely the pair production (PP) and low-energy $\gamma$-ray transmission (LET) techniques. The present paper reviews the application of the PP technique to the on-line determination of the ash content of coal and compares it to the LET technique. Recent Australian work on the development of techniques for the on-line determination of moisture and specific energy in coal is also reviewed.
2. ON-LINE DETERMINATION OF ASH USING PAIR PRODUCTION

2.1. Technique

In the PP technique [1, 2] coal is irradiated with high-energy \( \gamma \)-rays which interact in the coal to produce positron-electron pairs. The positron rapidly loses most of its energy by collisions in the sample and subsequently annihilates by colliding with an electron. Both the positron and electron disappear with the appearance of two oppositely directed annihilation \( \gamma \)-rays of energy 0.51 MeV. Ash is determined by combining the intensities of annihilation \( \gamma \)-rays and Compton-scattered \( \gamma \)-rays from the coal. The intensity of Compton-scattered \( \gamma \)-rays depends only on the bulk density of the coal whereas that of annihilation \( \gamma \)-rays depends on bulk density and atomic number (Z). The pair production probability per unit weight is proportional to \( \sum w_j Z_j^2/A_j \), where \( w_j \), \( Z_j \) and \( A_j \) are the weight fraction, atomic number and atomic weight of the jth element. The determination of ash content by the PP technique is based on this \( Z^2/A \) dependence since ash has a higher \( Z^2/A \) than coal matter.
TABLE I. CALCULATED ERRORS IN ASH CAUSED BY A CHANGE IN COMPOSITION FOR COAL CONTAINING 20 wt% ASH

<table>
<thead>
<tr>
<th>Change</th>
<th>Error in ash a (wt%)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>PP</td>
</tr>
<tr>
<td>+ 1 wt% CaO in ash</td>
<td>0.22</td>
</tr>
<tr>
<td>+ 1 wt% Fe2O3 in ash</td>
<td>0.32</td>
</tr>
<tr>
<td>+ 0.1 wt% Sr in ash</td>
<td>0.08</td>
</tr>
<tr>
<td>+ 0.1 wt% Ba in ash</td>
<td>0.13</td>
</tr>
</tbody>
</table>

- PP errors calculated from $\sum \frac{Z_j^2}{A_j}$ and LET errors from the ratio of mass absorption coefficients at 60 and 356 keV.
- The errors caused by ash composition changes are directly proportional to the ash content.

TABLE II. MEASURED AND CALCULATED RMS DIFFERENCES BETWEEN CHEMICAL LABORATORY ASH AND PAIR PRODUCTION GAUGE ASH FOR STATIC BULK COAL SAMPLES

<table>
<thead>
<tr>
<th>Bulk samples from a</th>
<th>No. of samples</th>
<th>Range of ash (wt%)</th>
<th>RMS error in ash determination (wt%)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>Calculated b</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Experimental</td>
</tr>
<tr>
<td>Vales Point, NSW</td>
<td>22</td>
<td>16–23</td>
<td>0.46</td>
</tr>
<tr>
<td>Liddell, NSW</td>
<td>18</td>
<td>22–33</td>
<td>0.54</td>
</tr>
<tr>
<td>Leigh Creek, S. Aust.</td>
<td>24</td>
<td>16–50</td>
<td>1.17</td>
</tr>
<tr>
<td>Blackwater, Qld</td>
<td>24</td>
<td>5–29</td>
<td>1.60</td>
</tr>
<tr>
<td>Blackwater, Qld c</td>
<td>12</td>
<td>5–17</td>
<td>0.32</td>
</tr>
<tr>
<td>South Africa</td>
<td>15</td>
<td>22–53</td>
<td>0.43</td>
</tr>
</tbody>
</table>

- For details on the composition of these samples see Refs [2, 3]. Sample mass approximately 70 kg except those from South Africa which were 4.3 kg.
- Based on calculated intensities of annihilation and Compton $\gamma$-rays using measured chemical laboratory ash compositions.
- Low-ash subset of the above 24 Blackwater samples.
The calculated errors in PP gauge ash caused by increase of 1 wt% CaO, 1 wt% Fe$_2$O$_3$, 0.1 wt% Sr and 0.1 wt% Ba in the ash (all replacing SiO$_2$) are given in Fig. 1 and Table I. These errors are calculated for a standard ash composition of 29.1 wt% Al$_2$O$_3$, 63.3 wt% SiO$_2$, 2.5 wt% CaO and 5.1 wt% Fe$_2$O$_3$, derived from the mean composition of 112 Australian black coals [3]. Strontium and barium are included in Table I, as several coals listed in Table II (from Blackwater and South Africa) contain up to 0.3 wt% Sr and 0.9 wt% Ba in the ash.

The main advantage of the PP technique over the LET technique [3, 4] is its reduced sensitivity to changes in ash composition (Table I). The LET gauge errors in Table I are more dependent on the magnitude of the various ash components than the PP gauge errors [3]. Both techniques are relatively insensitive to changes in moisture and coal matter composition [3].

2.2. Equipment and calibration

Until now, the PP gauge has been used mainly in three backscatter configurations, namely, a sample box geometry for static samples [1, 2], a cylindrical tube geometry for continuous by-line analysis shown in Fig. 2 [3, 5] and an on-line conveyor belt geometry [6]. In each of these geometries, the annihilation and Compton-backscatter $\gamma$-rays were detected in a large NaI(Tl) detector and a typical pulse height spectrum is shown in Fig. 3. As both annihilation and Compton-scattered $\gamma$-rays originate in essentially the same sample volume, errors due to changes in density and particle size are very small.

Ash is calculated using an equation of the form

$$\text{Ash} = f(P + gC) + h$$

where $P$ and $C$ are the measured count rates of annihilation and Compton-scattered $\gamma$-rays, respectively, and the constants $f$, $g$ and $h$ are determined by least squares fitting the chemical laboratory ash and measured values of $P$ and $C$.

Calibration of the PP gauge can be carried out in two steps to overcome problems which can arise because $P$ and $C$ are not independent variables. The first step is to select the value of $g$ in Eq. (1) which makes the measured ash independent of the separation of the gauge and sample. The second step is to determine constants $h$ and $f$ from measured $P$ and $C$ on a range of samples of known ash.

Counting statistical errors in the Compton count rate are very small, so the overall counting statistical error $E$ (wt% ash) is essentially due to pair production alone and is given by

$$E = \sqrt{P/t}$$

where $t$ is the counting time (s). Shown in Fig. 4 is a typical curve of counting statistical error versus counting time for the PP gauge in Fig. 2.
FIG. 2. Cross-section of the pair production (PP) gauge used for the determination of the ash content of coal moving through a vertical shaking tube.

FIG. 3. Typical pulse height spectrum obtained using the PP gauge in Fig. 2 with a coal sample containing about 20 wt% ash.
FIG. 4. Counting statistical error versus counting time for the PP gauge in Fig. 2 using a 3.9 GBq $^{226}$Ra source.

FIG. 5. Comparison of chemical laboratory and PP gauge ash assays for 18 bulk coal samples from Liddell, New South Wales. Shown are the mean and standard deviations of a series of measurements on 50 kg subsamples of each bulk coal sample.
2.3. Laboratory trials

The PP gauge was tested in the static box geometry on 40 kg subsamples of 88 samples of about 150 kg each from four different coal mining areas of Australia (Vales Point, Liddell, Leigh Creek and Blackwater) [2, 3]. Most of these were as-mined coal samples. A comparison of PP gauge and chemical laboratory ash for the high ash steaming coals from Liddell, New South Wales is shown in Fig. 5. The RMS differences between chemical laboratory ash and ash calculated from the measured values of P and C are in the range 0.46 to 1.3 wt% (Table II). These compare with RMS differences from 1.0 to 2.2 wt% for the LET gauge on the same samples [3, 4].

The experimental errors in Table II are due to a combination of such factors as variations in chemical composition of the coal and mineral matter, counting statistical errors, errors in sampling and errors in chemical laboratory ash assays. The calculated errors due to variations in chemical composition of the mineral matter are shown in Table II.

Laboratory measurements have shown that the PP gauge can be used to measure ash accurately in small coal samples of constant mass. For example, an RMS deviation of 0.98 wt% ash was obtained for 15 high-ash South African samples (Table II) of constant mass 4.3 kg. For these samples the mean and standard deviation of CaO, Fe₂O₃, Sr and Ba in the ash were 5.80 ± 2.56, 3.94 ± 1.77, 0.132 ± 0.087 and 0.152 ± 0.080 wt% respectively. The measured LET gauge accuracy on these samples was 3.2 wt% ash.

Calculations and experiments have shown that errors due to ash composition changes can be reduced by combining PP and LET measurements [7]. The method is most effective when only one high-Z element in the ash varies.

2.4. Installations

Mineral Control Instrumentation Pty Ltd (MCI)¹ is licensed to undertake the commercial development of the SIROASH (an acronym for CSIRO ash) PP gauge under the Coalscan trademark. MCI markets the SIROASH PP gauge in conjunction with a coal sample presentation system which has been licensed to MCI by the University of Queensland. The sample presentation system is based on the shaking tube principle in which an angular vibration is applied about the vertical axis of the tube [5]. The Coalscan PP gauge system uses a 2.4 m by 30 cm diameter tube constructed from filament-wound Kevlar fibre set in polyester resin with a wear-resistant polyurethane lining bonded to the inside surface of the tube. Coal flow rate is regulated by a vibrating feeder at the base of the tube. The feeder is controlled by coal sensors at the top of the tube and on the feed

---

¹ Mineral Control Instrumentation Pty Ltd, P.O. Box 64, Unley, South Australia 5061.
TABLE III. SUMMARY OF INSTALLED AND ORDERED COALSCAN PP GAUGES IN THE COAL INDUSTRY. THE FIRST FIVE GAUGES WERE INSTALLED BEFORE FEBRUARY 1985

<table>
<thead>
<tr>
<th>Location</th>
<th>Coal type</th>
<th>Gauge location</th>
<th>Ash (wt%)</th>
<th>RMS deviation (wt% ash)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ulan Coal Mines Ltd, NSW</td>
<td>Steaming</td>
<td>Washery product</td>
<td>12-20</td>
<td>0.33</td>
</tr>
<tr>
<td>Collinsville Coal Mining Co., Qld</td>
<td>Coking</td>
<td>Washery product</td>
<td>7-10</td>
<td>0.34</td>
</tr>
<tr>
<td>Newlands Coal Ltd, Qld</td>
<td>Steaming</td>
<td>Washed and unwashed product</td>
<td>10-16</td>
<td></td>
</tr>
<tr>
<td>Blair Athol Coal Ltd, Qld</td>
<td>Steaming</td>
<td>Raw coal</td>
<td>8-11</td>
<td>0.35</td>
</tr>
<tr>
<td>Oaky Creek, Qld</td>
<td>Coking</td>
<td>Washery product</td>
<td>7-10</td>
<td>0.21</td>
</tr>
<tr>
<td>Escom South Africa</td>
<td>Steaming</td>
<td>Washery</td>
<td>35-40</td>
<td></td>
</tr>
<tr>
<td>State Electricity Commission of Western Australia</td>
<td>Steaming</td>
<td>Raw coal</td>
<td>4-8</td>
<td></td>
</tr>
<tr>
<td>Mount Thorley, NSW</td>
<td>Coking</td>
<td>Washery product</td>
<td>7-9</td>
<td></td>
</tr>
<tr>
<td>Mount Thorley, NSW</td>
<td>Steaming</td>
<td>Washery product</td>
<td>12-20</td>
<td></td>
</tr>
</tbody>
</table>

a No results yet available.

hopper so as to keep the tube full of coal. The preferred flow rate is about 4 tonnes/hour, however flow rates of 1 to 10 tonnes/hour may be handled. No handling problems are experienced with coal of up to 50 mm top size.

The PP gauge in the Coalscan analyser (Figs 2 and 6) is essentially the same as that used for static bulk analysis except that it is fitted with an automatic source retraction device. The radiation beam from the 4 GBq $^{226}$Ra source is intercepted by a 150 mm thick shield located on the opposite side of the sample tube to the gauge. This shield reduces dose rates around the gauge to less than 10 $\mu$Sv/h.

This system also incorporates a moisture monitor which uses the capacitance technique of moisture measurement, with conducting plates built into the tube wall. Moisture is determined by simultaneously measuring capacitance, conductance, bulk density (from the PP gauge) and temperature.
FIG. 6. The first commercial Coalscan PP gauge, which has been operating at Ulan Mines Limited, New South Wales, Australia, since February 1983. The gauge continuously measures the ash content of coal passing down the sample presentation tube.

So far, commercial Coalscan PP gauge systems have been installed in five coal mines in Australia and orders have been received for a further four systems as listed in Table III.

The first Coalscan system was installed at Ulan Coal Mines, New South Wales (Fig. 6). This system has been operating on the product line of the 1200 tonnes/hour washplant since February, 1983. The mean ash content of the Ulan product coal is about 17.5 wt%. Two studies have been made on the accuracy of the Ulan gauge [8]. In the first, a series of 51 twelve-hour tonnage weighted averages of Coalscan ash measurements were compared with results from an automatic sampling
system. These results showed an RMS deviation of 0.34 wt% ash between the Coalscan and chemical laboratory ash analyses. This deviation includes errors due to sampling and chemical laboratory assay. In the second test, Coalscan ash readings were compared with laboratory assays of coal samples from the automatic sampling system over a total of about 60 eight-hour periods for half a million tonnes of coal. In this case the RMS deviation was 0.33 wt% ash. The PP gauge is being used to control the density of heavy medium in the baths and cyclones in the Ulan washery.

The Collinsville gauge monitors ash in the washery product line and is presently used for manual control of the washery. A comparison of Coalscan ash readings and chemical laboratory assay of ash over eight-hour shifts indicated an RMS deviation of 0.34 wt% ash [8]. The Newlands installation is on a washery by-pass line and the Blair Athol gauge (Fig. 7) monitors raw coal ash and is used for mine grade control. For the Blair Athol gauge, the RMS deviation was 0.35 wt% ash for samples taken over 16 half-hour sampling periods.

The Oaky Creek PP gauge was installed in January 1985 to monitor the ash content of coking coal product. The accuracy of the gauge was measured by comparing Coalscan ash readings with chemical laboratory assays of samples taken from the discharge chute of the gauge. Sixteen 4 kg samples were taken each half-hour sampling period. The RMS deviation between gauge and chemical laboratory ash was 0.21 wt% for 29 half-hour periods as shown in Fig. 8 [8].
The materials-handling aspects of the Coalscan system have also been thoroughly tested over the period of installation. The system is unaffected by changes in moisture or sizing, and there has been no observable wear of the shaking tube lining.

2.5. On-line conveyor belt plant trial

A plant test to assess the PP gauge for direct on-line conveyor belt analysis was undertaken in late 1982 on the recirculating coal facility at the pilot plant coal washery at the BHP Steel Works, Newcastle, New South Wales [6]. Seven Hunter Valley coals with ash in the range 7.5 to 33 wt% were circulated on the conveyor loop, and scanned by both PP and LET gauges. The PP gauge was corrected for variable mass per unit area of coal on the belt by measuring the transmission of $^{226}$Ra γ-rays. Samples were measured on-belt as a function of sample depth, compaction, moisture and particle size. The mass per unit area of coal on the belt was varied in the range 40 to 210 kg·m$^{-2}$. The RMS deviation between PP gauge ash and chemical laboratory ash was 1.07 wt% ash for 370 individual 300 s on-belt measurements on coal of mass per unit area greater than 60 kg·m$^{-2}$ and 0.45 wt% ash for the mean ash of the seven samples. However, calibration of the on-belt PP gauge is more difficult than for the by-line gauge and inaccuracies can arise, because of non-overlapping volumes of backscatter and transmission measurements, particularly at low mass per unit areas (<100 kg·m$^{-2}$).

For samples of mass per unit area >250 kg·m$^{-2}$, no correction for the thickness variations is required for a backscatter PP gauge as the sample is infinitely
thick with respect to backscattered radiation. One example of the application of a PP gauge to direct on-line conveyor belt analysis in which thickness correction is not required is the development and field testing of a PP gauge for the analysis of iron in iron ores [9].

2.6. Comparison of Coalscan PP and LET gauges

The main advantage of the PP gauge over the LET gauge is its reduced sensitivity to variations in ash composition, particularly for high Z elements such as Fe, Ca and Ba. Calculations using sets of coal analysis data from 17 mines and washeries showed that PP gauge errors due to composition changes are a factor of 3.6 ± 0.6 less than those for the LET gauge. This factor is independent of ash content. As errors due to composition changes are proportional to ash content, this advantage of the PP gauge is greater for higher ash coals. The LET gauge has the advantages of measuring coal directly on the conveyor belt and having a shorter measurement time (tens of seconds compared to hundreds of seconds for the PP gauge). However, the good sample presentation of the PP gauge compared to the LET gauge means that in practice better accuracy is obtained.

3. DETERMINATION OF MOISTURE IN COAL

In recent years there has been a growing interest in measuring and controlling the moisture content of coking and steaming coals. Moisture in these coals has increased mainly because of the increased proportion of fine coal in preparation plants as a result of modern mining methods and because greater emphasis is placed on fine coal recovery for economic reasons. Lower product coal moisture results in lower transport costs, energy savings when the coal is utilized and reduced handling difficulties. Two specific areas where the on-line determination of moisture content is important are in the control of fine coal drying processes and in the determination of specific energy of coal (when combined with an on-line ash measurement). Generally, in these applications, determination of moisture to within about 0.5 wt% is required in an analysis time of less than a few minutes.

A number of techniques are being investigated and compared by CSIRO for the on-line determination of moisture in coal. These include capacitance, microwave attenuation, neutron transmission and scattering, infra-red reflectance and nuclear magnetic resonance.

3.1. Capacitance methods

(a) Cylindrical geometry

Capacitance techniques have been investigated at CSIRO by Cutmore and Rocznioik [10]. Two parallel plate arrangements have been investigated in which
the capacitor plates are located on diametrically opposite faces of chutes of diameter 10 and 30 cm. The latter chute is identical to the tube used in the commercial Coalscan on-line analysis system.

Four coals (two from Blackwater, Queensland, one each from Vales Point and Liddell, New South Wales) have been measured in both tubes, both crushed (−3 mm) and uncrushed (−30 mm). Radiofrequency (100 kHz −10 MHz) susceptance (C) and conductance (G) were measured and, together with density, correlated with oven-dried moisture using various linear equations. The large and small diameter tubes gave very similar results.

An acceptable sensitivity was obtained at measurement frequencies from 2−10 MHz and the RMS deviations between gauge and oven-dried moisture were about 0.5 wt% for a single coal type over the moisture range 0−20 wt%. This RMS deviation is in agreement with that obtained using the commercial Coalscan analyser at Ulan [5].

If all four coals are included in the one correlation, the RMS deviation increased to about 1 wt% for fine, wet coal (−0.5 mm, 15−30 wt% moisture) and 2 wt% for crushed and uncrushed coal with a moisture content of 0−20 wt%.

(b) Under-belt geometry

Cutmore [11] has developed a non-contacting under-belt capacitor gauge suitable for the on-line determination of moisture in coal. In one application, the capacitance sensor comprises a parallel strip capacitor arranged so that the fringing electric field interrogates a volume of coal on the conveyor belt directly above the sensor.

The results for five samples of crushed coal summarized in Table IV show that the technique is capable of determining moisture to better than 0.4 wt% in laboratory conditions. Plant tests of the technique are planned for 1985.

3.2. Microwave methods

Moisture measurements by microwave techniques usually involve measurement of attenuation or phase shift at a frequency in the range 2−10 GHz. A non-contacting microwave moisture gauge, in which both the attenuation and phase shift of a transmitted beam are determined, is being developed for the on-belt measurement of moisture in coal. The design parameters for this gauge have been established using measurements of transmission phase and attenuation where coal samples were packed in waveguide sections and conventional network analyser techniques were used [11]. These laboratory measurements show that a measurement accuracy of less than 0.5 wt% moisture is typical for one coal type. Additional errors of 0.5 − 1 wt%, caused by large variations in either the coal type or average particle size of the coal, can be significantly reduced by additional measurements (e.g. at a second frequency) and/or data processing (e.g. ratio methods).
TABLE IV. SUMMARY OF RMS DEVIATIONS BETWEEN CAPACITANCE GAUGE MOISTURE AND OVEN-DRIED MOISTURE FOR THE UNDERBELT CAPACITANCE GAUGE

<table>
<thead>
<tr>
<th>Sample</th>
<th>RMS deviationb (wt% H₂O)</th>
</tr>
</thead>
<tbody>
<tr>
<td>UDC washery producta</td>
<td>0.30</td>
</tr>
<tr>
<td>BHP coke oven feed blend</td>
<td></td>
</tr>
<tr>
<td>Component A</td>
<td>0.37</td>
</tr>
<tr>
<td>Component B</td>
<td>0.27</td>
</tr>
<tr>
<td>Component C</td>
<td>0.23</td>
</tr>
<tr>
<td>Component D</td>
<td>0.40</td>
</tr>
</tbody>
</table>

All samples were of particle size ~3±0 mm, thickness 5.0 to 7.5 cm and had moisture in the range 0—19 wt%.

a UDC = Utah Development Company.
b Correlation equation involving C, G, W and T where W = mass of sample and T = thickness.

3.3. Neutron moisture gauges

A number of neutron transmission and scatter methods are being evaluated for direct on-line conveyor belt determination of moisture in coal. Neutron techniques have the advantage of being independent of particle size and neutron transmission techniques are well suited to direct on-belt analysis. However neutron techniques measure the total hydrogen in the coal plus hydrogen in the moisture and therefore rely on there being only small variations in the coal hydrogen. For a neutron moisture gauge error of less than 0.5 wt%, the hydrogen content in the coal matter must be constant to within 0.04—0.07 wt% (dry ash free basis) and ash known to within 0.7 to 1.0 wt%. Data on hydrogen concentration in coals from particular seams or mines show standard deviations of the order of 0.1 to 0.15 wt%, which is approximately equal to the accuracy of the chemical laboratory assay. More accurate methods for the determination of hydrogen in dry coal are being investigated.

Preliminary results obtained with a fast neutron and γ-ray transmission gauge using ²⁵²Cf and ¹³⁷Cs sources have indicated that moisture can be determined to within 0.6 wt% on samples of thickness about 7 cm containing from 0—15 wt% moisture.

Previous work has shown that moisture can be determined to within 0.2 wt% in a single bulk coal sample containing from 0 to 25 wt% moisture [12]. The method is based on measurements of the intensities of 4.43 and 2.23 MeV γ-rays
from carbon and hydrogen, respectively, together with γ-ray backscatter. If the γ-ray backscatter measurement is not used, the RMS error rises to 0.36 wt%.

3.4. Nuclear magnetic resonance

Nuclear magnetic resonance (NMR) is potentially a rapid and accurate method for the determination of moisture in coal and, even though the technique is more complex than many of the alternatives, it has the unique advantage of being able to measure simultaneously both organic and water hydrogen in coal samples. Pulsed NMR has been investigated for moisture determination in fine product coal from various Australian coal washeries [13]. Measurements were made on samples of diameter 8 and 12 mm and length 10 and 120 mm at frequencies from 6.5 to 60 MHz. The ratio of intensities of the water and coal components in the free-induction decay can be used to determine moisture to within about 0.4 to 0.7 wt% over the range 0 to 26 wt% water, independent of sample density. This accuracy is independent of particle size (up to 1 mm) and little affected by coal type, sample length or NMR frequency.

3.5. Infra-red reflectance

A commercial moisture meter based on the measurement of infra-red reflectance has been evaluated for on-belt measurement of moisture in fine (−2+0 mm) coal. The moisture content was determined to better than 1 wt% for a single coal type. However, a coal type error of about 4 wt% was typical when comparing different samples. Also, changes in particle size from −2+1 mm to −1+0.5 mm caused errors of about 3 wt%. These, and other problems associated with instrument calibration, make the technique an unlikely candidate for on-belt analysis of moisture in coal [10].

4. SPECIFIC ENERGY DETERMINATION

The method of determining specific energy of coal from measurements of ash and moisture assumes that the material left after removing ash and moisture has constant specific energy. A more widely applicable method involves determination of the specific energy from an accurate carbon analysis. Specific energy and carbon are correlated to within about 1% relative for a wide range of coals [12].

The accurate determination of carbon in coal is based on the measurement of 4.43 MeV carbon inelastic scatter γ-rays combined with a separate γ-ray scattering measurement on essentially the same sample volume. Laboratory measurements have shown that the method can be used to determine the carbon and specific energy contents of coal to within 1.4% relative for a wide range of
coal compositions and to within 0.8% relative for coals with ash content less than 18 wt% [12].

Determination of the total energy of pulverized coal blown into furnaces and boilers requires a knowledge of the coal mass flow. A gauge for the determination of coal mass flow has recently been developed and installed by Cunningham et al. on feed to a copper reverberatory furnace at Mount Isa Mines Ltd, Queensland [14]. The gauge determines bulk density by forward scattering $\beta$-radiation from a $^{90}$Sr source and air density is calculated from pressure and temperature. Flow speed is determined by cross-correlation of triboelectric charge. The gauge has been used to determine coal mass flow to a precision of about 2% relative, and its use with automatic control has reduced coal consumption in the reverberatory furnace by about 10%.

5. SUMMARY

On-line ash gauges based on the pair production technique are in routine use in the Australian coal industry. To date, five commercial PP gauges have been installed and a further four have been ordered. Root mean square differences between PP gauge and chemical assay are in the range 0.21 to 0.34 wt% ash for the on-line analysis of coal up to 20 wt% ash.

A number of nuclear and non-nuclear techniques of moisture determination are being investigated. Plant trials of an under-belt capacitor are planned for 1985.

ACKNOWLEDGEMENTS

The author wishes to thank Dr. W.J. Howarth and Mr. K.G. Smith of Mineral Control Instrumentation Pty Ltd for supplying details on the operation and performance of the commercial Coalscan PP gauges. The author also wishes to thank Dr. N.G. Cutmore for providing results on the determination of moisture in coal using capacitance, microwave and infra-red techniques.

The CSIRO projects were supported, in part, by grants from the Australian National Energy Research, Development and Demonstration Council.

REFERENCES


DISCUSSION

S. CIERPISZ: Do you apply a microwave method for monitoring moisture on an industrial scale?

B.D. SOWERBY: No. The project investigating moisture in coal has been under way in our laboratory for only about 18 months. With the microwave system, we have only been doing laboratory measurements to date.

P.L. SURMAN: I don't think you gave us the dimensions of your shaking tube. Would it handle sticky, wet British coals?

B.D. SOWERBY: The diameter of the tube is 30 cm. We've had some experience with sticky, wet coal in one of the Queensland applications. The only problem this gave was that in the chute that sits above the shaking tube, the coal was sticking in the side of the chute and the device used for monitoring the level of the coal indicated it was full all the time because of this. It was necessary in that case to change to a microwave system for determining the level of the coal. But in the shaker tube itself, the wet coal has gone through without any trouble at all.
EXPERIENCES WITH COAL QUALITY MONITORING SYSTEMS IN POLAND

S. CIERPISZ
Research and Production Centre for Mining, Electrical Engineering and Automation EMAG, Katowice, Poland

Abstract
EXPERIENCES WITH COAL QUALITY MONITORING SYSTEMS IN POLAND.
The paper presents the results of research in Poland on coal quality monitoring systems. The methods used and technical data on radiometric ash monitors, microwave moisture meters, calorific-value monitoring systems and radiometric sulphur meters are discussed. The accuracy of the monitoring systems is illustrated by calibration curves for several industrial installations.

1. INTRODUCTION

Devices and systems which allow continuous measurement of parameters in coal preparation processing are most essential to achieve automation [1]. No less important are devices permitting rapid and discrete measurement on coal samples. The basic parameters of the coal preparation process needing to be measured are ash and moisture content and mass of coal, but sulphur content and calorific value should be considered as well. In this paper Polish achievements in this domain are presented.

2. MEASUREMENT OF ASH CONTENT IN COAL

Ash content in a quantity of coal is in practice determined by testing, a sample only, whose volume, \( V_1 \), is less than the volume of the whole, \( V \). So the content determined, \( A_{V_1} \), differs from the real ash content \( A_V \) and is a random quantity. The error of measurement can be determined by the mean square deviation \( \sigma_{A_{V_1}} \). When a volume of \( nV_1 \) samples is measured, the mean square deviation decreases according to the following formula:

\[
\sigma_n = \frac{\sigma_{A_{V_1}}}{\sqrt{n}}
\]

To obtain a representative figure for ash content, an adequate proportion of the volume of coal should be tested. A mass of coal being tested can be
FIG. 1. Radiometric ash monitor - G-type.
accepted as a characteristic parameter of an ash monitor as regards its usability in monitoring and control systems. The other parameters which determine the usability of an ash monitor are: accuracy of measurement, time delay and the method of coal sample preparation for testing. It is difficult to design an ash monitor having all the optimal parameters, hence all present designs represent a compromise between these four parameters.

Several methods for ash content measurement have been developed:
- a measuring method based on the relationship between the bulk density of coal and ash content
- radiometric methods in which the relation between penetrating or scattered radiation and ash content is used
- neutron activation techniques.

Most ash meters use radiometric measuring methods. Those which make continuous measurement of ash content possible are worthy of notice.

The Research and Production Centre EMAG designed the G-type radiometric ash meter in which special attention was paid to the representativeness of measurement results and to the possibility of application to all sizes of fine coals.

*FIG. 2. Ash monitor calibration curve, Wujek mine.*
The G-type radiometric ash meter is designed for continuous measurement of ash content in raw coals and washed ones of 0—30 mm size performed directly on a belt conveyor. The measuring method is based on the relationship between the effect of backscattered gamma radiation and ash content in coal. The ash meter consists of the following sub-assemblies:

- a measuring head (with a $^{241}$Am radiation source and NaI scintillation counter as a detector)
- a system for forming a coal stream on a belt conveyor
- an electronic block
- detectors for control of ash meter operation (sensors for conveyor movement and thickness of the coal layer).

The measuring head is placed above the conveyor and is fixed to the system which forms the coal stream (Fig. 1) which in turn is connected to the conveyor-supporting structure. A task of the system for forming the coal stream is to produce a level coal layer on the conveyor and to keep the measuring head at a constant distance from the layer irrespective of its thickness, which has to be at least 150 mm.

This ash meter permits measurement of ash content ranging from 3—40%. The calibration of the meter is performed with coal batches displaced under a head. The ash content in coal batches is measured by a conventional method.
The equation of the calibration curve is introduced into a converter, so that the meter readings may be expressed as the percentage of ash.

Based on results of tests of over 20 industrial installations of G-type ash meters, the mean square deviation of measurement was determined for different ranges of ash content:
- in the range 3—10% it amounts to 1.0% of ash content
- in the range 10—20% it amounts to 1.5% of ash content
- in the range 20—40% it amounts to 2.0% of ash content.

The above results are due to the error of the radiometric measuring method as well as to the error of the conventional measurement of ash content. The accuracy of the radiometric method depends mainly on ash content, changes in chemical constitution and homogeneity of the coal layer's cross-section. The error of the conventional method comprises the errors associated with sampling, treatment of a sample and combustion method as well. This error depends on the homogeneity of the coal batch, ash content, representativeness of sampling and further treatment of sample. Therefore, the mean square deviation between radiometric and conventional measurements does not provide a straightforward evaluation of G-type ash meter operation. For this purpose one should take the error of the radiometric ash content measurement.

FIG. 4. Ash monitor calibration curve, Powstańców Śląskich mine.
The mean square deviation $S_R$ expressed by the following formula can be accepted as an estimation of this error:

$$S_R = \sqrt{S^2 - S_K^2}$$

where $S$ is the mean square deviation between the results of radiometric and conventional methods, and $S_K$ is the mean square deviation of the conventional method.

Taking into account the values $S$ for different ranges of ash content and values $S_K$ based on operating data from various laboratories (~0.5%), the mean square deviation $S_R$ is as follows:

<table>
<thead>
<tr>
<th>Range of ash content</th>
<th>Value of mean square deviation, $S_R$</th>
</tr>
</thead>
<tbody>
<tr>
<td>3—10%</td>
<td>approx. 0.5% of ash content</td>
</tr>
<tr>
<td>10—20%</td>
<td>approx. 1.0% of ash content</td>
</tr>
<tr>
<td>20—40%</td>
<td>approx. 1.5% of ash content</td>
</tr>
</tbody>
</table>

It should be noted that $S_R$ values concern measurements of ash content in coal of 0—30 mm size, performed by the G-type ash meter directly on a belt conveyor under various industrial conditions.

Calibration curves of G-type ash meters in several coal preparation plants are shown in Figs 2—4.

At present in Poland the G-type ash meters are mostly used for measurements of ash content in fine coals being loaded into wagons. The result is obtained immediately after loading each wagon, which permits forming drafts of wagons according to customers' requirements. The G-type ash meter can be also used for automatic monitoring of the calorific value of power coal. This is the subject of a later section of this paper.

### 3. MEASUREMENT OF MOISTURE CONTENT IN COAL

In the latest version of the microwave moisture meter WILMAG the design has been upgraded in the light of results of industrial tests on several meters.

The WILMAG meter uses the phenomenon of backscattered microwave radiation from the levelled surface of fine coal. This is a substantial novelty in comparison with other existing meters which to date have used capacitance methods based on changes of the dielectric properties of wet coal (SRCW-4 moisture meter, Poland; BAK-4, Soviet Union; RV-21, Czechoslovakia, etc.).

The main disadvantage of these methods is the necessity of contact between the sensor and the coal, which causes rapid wear of the sensor and makes uncontrolled errors of measurement difficult to eliminate. The sensor has to be
replaced frequently and calibrated. The meters using capacitance sensors placed under the belt are very liable to errors resulting from the heterogeneity of the belt and the degree of its wear.

The WILMAG microwave moisture meter designed by the EMAG Centre in co-operation with ZAM WILMER, enables the above-mentioned difficulties to be avoided to a large extent.

The measuring method permits simple forming of the coal stream, involving only levelling its surface. The sensor, protected against dust and shocks, is placed above the coal surface. The measurement is performed automatically on a running conveyor and with a proper coal layer thickness. The moisture content data are displayed in % H₂O as instantaneous and mean values for a given period of time, e.g. for the loading time of a single wagon.
Technical data:

Measuring range: 3—30% H₂O
Material tested: hard coal
Maximum coal size: 30 mm
Minimum thickness of coal layer: 100 mm
Minimum width of coal layer: 250 mm
Nominal belt conveyor speed: 2 m/s
Error of measurement: 1% H₂O

Calibration curves of the WILMAG moisture meter are shown in Figs 5—7.

4.  MONITORING OF THE CALORIFIC VALUE OF COAL

Calorific value is a parameter in general use for the evaluation of the power properties of coal in industrial combustion and heat-exchange processes. It also affects, together with the ash content, the price of steam coal as a market product.

![Moisture meter calibration curve, Wujek mine.](image)
FIG. 7. Moisture meter calibration curve, Kościuszko coking plant.

TABLE 1. INDUSTRIAL INSTALLATIONS OF COAL QUALITY MONITORING SYSTEMS IN POLAND (1984)

<table>
<thead>
<tr>
<th>Monitoring system</th>
<th>Coal preparation plants</th>
<th>Power stations</th>
<th>Coking plants</th>
</tr>
</thead>
<tbody>
<tr>
<td>G-type ash meter</td>
<td>25</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>WILMAG moisture meter</td>
<td>8</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>ALFA calorific value monitoring systems</td>
<td>7</td>
<td>-</td>
<td>-</td>
</tr>
</tbody>
</table>
To satisfy the industrial requirement of rapid and continuous determination of calorific value the EMAG Centre has developed the ALFA system.

In this system calorific value is determined on the basis of the results of ash and moisture content measurements, these being obtained from meters placed above the belt conveyor. The calorific value $Q$ is calculated in a microprocessor by empirically relating the calorific value and ash and moisture content:

$$ Q = Q_0 - k_1 \cdot A - k_2 \cdot W $$

where $Q_0$ is the calorific value of ash- and moisture-free coal, $k_1$, $k_2$ are coefficients determined experimentally, and $A$ and $W$ are the ash and moisture content in the coal.

The ALFA system comprises five basic units:
- radiometric G-type ash meter
- WILMAG microwave moisture meter
- microcomputer
- control unit
- detectors of layer thickness and conveyor belt movement. The ALFA system has been applied in coal preparation plants for monitoring the calorific value of fine coal being loaded into wagons. The number of industrial installations of has, moisture and calorific value monitors is shown in Table I.

5. MEASUREMENT OF SULPHUR CONTENT IN COAL

Two types of laboratory radiometric sulphur meters have been developed by the EMAG Centre.

The MSC total sulphur meter uses the absorption measuring method. Sulphur content is determined by attenuation of soft X-rays which penetrate a coal sample inserted into a measuring container. The measuring head into which a container with a coal sample is introduced comprises a $^{241}$Am/Sn X-ray source and a scintillation counter as a detector. The MSC meter is designed for total sulphur measurement within the range of 0–6% on samples of 15 g mass and 0–0.2 mm coal size. The error of measurement is 0.2% of sulphur content.

The MSP sulphur meter is designed for pyrite sulphur content measurement. The measuring method is based on recoilless resonance absorption of gamma radiation (Mössbauer effect). The measuring head, into which a coal sample is introduced, comprises a radiation source fixed to the vibrator, a radiation detector (proportional counter) and a holder for the coal sample. The electronic unit includes a control system for detecting movement of the radiation source, a pulse converter and a power supply unit.

The MSP meter permits measurement of pyritic sulphur content within a range of 0–10% in coal samples of 5 g mass with the size of coal below 0.2 mm. The error of measurement is 0.1% of pyritic sulphur.
REFERENCES


DISCUSSION

J.S. WATT: First, I am very impressed by the number of installations in Poland. Since you have a raft on your coal, does that limit the maximum speed of coal?

S. CIERPISZ: Yes, to two metres per second.

J.S. WATT: Have you experienced any effects of vertical segregation of coal, say high ash on top or bottom?

S. CIERPISZ: Yes, but the influence of this is within the accuracy which I reported.

J.S. WATT: For the installations which are in coal preparation plants, where is the ash gauge put on the product line? Also, could you give some indication of how you are using the information to give better control.

S. CIERPISZ: Originally, when we were in the process of developing the ash monitor, we intended to use it to control the process of washing. But it turned out that all of our installations monitor ash content in untreated smalls. So usually we install our ash monitors, as others do, in loading stations. In Poland we do not usually wash small, fine steam coal, so there is no process control. I don’t think we will apply ash monitors to control the washing process, because it seems to us that an ash monitor will not provide a real benefit.

C.G. CLAYTON: Have you any idea what the energy spectrum is at the detector?

S. CIERPISZ: We monitor the whole spectrum, we don’t have any window. We count all reflected energies above a certain level.

C.G. CLAYTON: Is it partly a transmission gauge and partly a backscatter gauge?

S. CIERPISZ: That is right.

C.G. CLAYTON: It has a high proportion of single scattered radiation?

S. CIERPISZ: Yes.

C.G. CLAYTON: I wondered how you got such high sensitivity; now I can see.

G. FAUTH: We have also tried to utilize this plough technique, but we did not succeed in a practical industrial application because there were disturbances from foreign materials, wood and wires, and these built up at the plough. Do you have similar problems in your plants?

S. CIERPISZ: I think this is the most reliable part of the ash monitor in our case. We have problems associated with ensuring that we have enough coal on the belt. Firstly we apply ash monitors in loading stations having high
capacity, about 2000 tonnes per hour. In these cases we have no problems with the amount of coal transported on the belt. In other cases there are problems with the amount of coal, but I think that with a little effort they can be overcome. Usually we have a bunker, in which the coal is stored, and it is possible to apply a simple control to the speed of the conveyor to keep a more or less constant level of coal on the belt.

G. FAUTH: You are measuring both the moisture content and the ash content in a very narrow strip of the belt. Do you have problems with heterogeneity on the belt?

S. CIERPISZ: It is not a very narrow strip of coal. The width of the measuring surface is about 25 cm.

G. FAUTH: For both ash and moisture?

S. CIERPISZ: Yes. So, roughly speaking, we monitor about 60% of the coal transported on the belt.

P.L. SURMAN: This reminds me of a very serious problem we have at power stations. This is the appearance of 'tramp metal' on the conveyor belts which can get into the pulverizers and damage them. You say that you use these monitors at power stations?

S. CIERPISZ: At one.

P.L. SURMAN: Has there been something which would detect 'tramp metal'?

S. CIERPISZ: So far we do not have problems with 'tramp metal' as regards the operation of the ash meter. There is a serious problem with applying this type of ash monitor at power stations, because of the variety of coals. In this particular case we have only two types of coal which are combusted, so we can manage the problem. When we have various kinds of coals, ash monitors should be recalibrated and this is actually the most important problem for us.

J.S. WATT: On the calibration aspect, I think this is very simple to do with low-energy transmission measurement, because you can just swing the gauge off the belt and calibrate against a standard of any sample you want.
PHYSICS OF RECENT APPLICATIONS OF PGNAA FOR ON-LINE ANALYSIS OF BULK MINERALS*

T. GOZANI
Science Applications International Corporation,
Sunnyvale, California,
United States of America

Abstract

PHYSICS OF RECENT APPLICATIONS OF PGNAA FOR ON-LINE ANALYSIS OF BULK MINERALS.

In the past few years the coal, cement and other mineral industries have been slowly but steadily coming to realize the potential economic, technological and environmental benefits from using nuclear and especially prompt gamma neutron activation analysis (PGNAA) techniques. This realization, along with the great efforts and the technological developments of the last decade, culminated in successful production and operation of a family of on-line analysers under the name 'Nucoalyzer'. The main technical factors that contributed to this success are: (a) the development of very stable high count rate gamma spectroscopy for high (Ge) and medium (NaI or BGO) resolution detectors; (b) the emergence of reliable large radiation-hardened Ge detectors; (c) careful study and understanding of the transport processes of neutron and the associated gamma rays in various bulk media; (d) the incorporation, from the outset, of realistic industrial constraints along with those determined by the physics and electronics; (e) the availability of small but reliable and powerful multichannel analysers and microprocessors; (f) incorporation of non-nuclear techniques, such as microwave transmission, to supplement the nuclear techniques. The physics and engineering principles of the Nucoalyzer are reviewed. The successful system optimization that led to a major reduction in the influence of extraneous effects, such as bulk density, moisture variation, geometrical instabilities, radiation damage, etc., are discussed and some highlights of results are presented.

* The full text of this paper is to be found in the Proceedings of the 5th International Symposium on Capture Gamma Ray Spectroscopy and Related Topics, Knoxville, Tennessee, 10-14 September 1984, American Institute of Physics Publication.
J.L. MIKESELL: What techniques do you use for spectral stripping, and also, in a system with data analysis so heavily automated, how do you ensure proper gamma ray spectra calibration?

T. GOZANI: It would take hours to answer fully. Firstly, we know where the gamma lines are, so there is no isotope search. For germanium detectors we don’t do very much stripping, since we know where the gamma lines are; we do some proper background fitting which is tailored to the particular application. We have several levels of stabilization. We have electronic stabilization, digital stabilization and computer stabilization. And every time we find some new instabilities we try to correct, but now from our experience things are pretty stable. Stabilization for sodium iodide is much more serious because there the resolution is much poorer, so if you try to do any mathematical manipulation it is much more dependent on gain shifts.

J.S. WATT: A comment on the RASH gauge. You mentioned once you were using multiple gammas, another time two gammas. Do you have more than two gamma energies involved in the gauge?

T. GOZANI: Not in the standard commercial model. We have looked at the possibility but it appears not very promising for many reasons. But it is something which maybe for some applications would work.

B.D. SOWERBY: In the CONAC system, in the sample handling system, have you had any problems with wet coals? Are there problems with segregation on that belt?

T. GOZANI: We don’t have problems with the coal transport system within CONAC until it becomes a slurry. It is not designed for slurry. As for segregation, this is a good question. We require that there is very little free fall for the coal going into the hopper. This is why we want it to operate at 80%—so there is only 1 foot free fall—otherwise you start to get segregation. There were some cases when there was a lot of pyrite and there was a little segregation. So it can happen but we minimize it by design of the input hopper and by the way we operate it.

B.D. SOWERBY: Regarding your microwave system on belt, I would like to know what frequency you are measuring. What in fact are you measuring? Are you just measuring amplitude? Also, is it being used together with the RASH gauges in a commercial unit?

T. GOZANI: No, it is not installed yet. We have done laboratory measurements.

B.D. SOWERBY: But you are looking at a single frequency and just measuring attenuation?

T. GOZANI: It has the capability for double frequencies, and it has the capability of looking at phases, but our basic unit, with a minimum cost, is just transmission.
J. CHARBUCINSKI: What were the values of the standard deviations for the data you showed with compressed correlations for hydrogen and carbon?

T. GOZANI: On hydrogen it is 0.05, on carbon 1.4 wt.%, 200 Btu/pound if I don’t correct for bias, 100 Btu/pound if I do correct. This appears in Volume 10 of our EPRI report.

J. CHARBUCINSKI: Regarding the sulphur measurements, in your new coal analyser did you use only sodium iodide detectors?

T. GOZANI: Yes, sodium iodide alone.

J. CHARBUCINSKI: What sort of source, size of sodium iodide crystal and time of measurement was used? And why sodium iodide and not BGO which could decrease the size of the source by ten times?

T. GOZANI: I’m surprised that you say ten times. We use BGO when it is appropriate. The system I showed was designed in 1979 and BGO was not available. The resolution of BGO is poor. There is no reason to use it here and I can assure you the source would not be down a factor of ten. The measurement time is 100–200 seconds, with accuracy of 0.03. You know it doesn’t always make sense to use the best technology in the wrong place. One has to optimize, and also cost is very important. BGO costs 3–4 times more than sodium iodide. We are talking about a large detector, 6 × 6. The size of the source was 100 μg.

C.G. CLAYTON: You mentioned tailoring your equipment specifications to the requirement on a number of occasions. I wasn’t quite sure whether that included tailoring the CONAC system. I wonder how many parameters or how many large variations in what number of parameters the CONAC system can deal with. Or to put it another way, how constant is your coal compared for example with British coal, or the receipt of British coals at power stations coming from a number of different mines?

T. GOZANI: Firstly, when I speak of tailoring I mean what system should a utility use. Should they use CONAC everywhere when they only need ash, and when they need ash to the precision of 1%? I say don’t use CONAC, it’s a waste of money and time. When you are looking for sulphur, maybe with some information on ash, maybe some information on some other elements, use a simple sodium iodide system. The CONAC is intended to be used when you need a complete elemental analysis and when you want as much as possible coal-independent results. That is the function of the CONAC, being the most sophisticated instrument. Coal independence is one important aim of the CONAC. It has been achieved, maybe not to the ideal extent we would wish. However, most of the problems are instrumental. Basically deterioration of resolution (which was improved considerably once the n-type detector became available), radiation damage (also reduced by n-type detectors), and things of that kind.

C.G. CLAYTON: In the development of any technique one runs the system to the extreme. So one way in which you might do that would be deliberately to spike your coals with a number of elements at high concentration. Then you would really run up against some difficulties. There must be a limit to the
variations in the number of elements and the concentrations which you can cope with.

T. GOZANI: Chlorine in the British coal is a natural spike of the highest degree and we have trouble with this.

C.G. CLAYTON: The real point is this. When you begin to encounter a wider range of elements, with a wide range in concentrations, then I would guess that you then need to use the high-resolution detector more frequently than you do if there are less variations. Therefore you could run into more problems in terms of the instrumental limitations of high-resolution detectors. You have mentioned radiation damage, which is clearly better with n-type than p-type detectors. In our experience it is not as good as was originally said. That is, 20 times was claimed but this is an exaggeration.

T. GOZANI: Absolutely right. However, I think operationally if you have to take the detector every half year and send it for annealing, this is an acceptable operational procedure. Concerning the variations of elements, we know coal to a large extent, we have measured as large a range of coal as we can. If there comes a case with some unusually high level of some elements in a specific coal, we may get a wrong result for that case. But trace elements do not disturb it.

C.G. CLAYTON: Under those circumstances where you are expecting larger variations than you might be measuring in your current installations, would you then expect to use your high-resolution detector more frequently in order to get better spectral analysis?

T. GOZANI: What is more frequently? Less than 20 minutes?

C.G. CLAYTON: Yes, with a larger source activity, for example.

T. GOZANI: Larger source activity will shorten the life of the detector. There is a point of diminishing returns. So with shielding. Shielding represents cost, a larger detector represents more cost. So there is a compromise. I think the instrument, as complex as it is now, is probably responding to this situation. As I say, I can always defeat the CONAC. Nature will defeat it much less because normally you do not have, say, 10 000 ppm of boron or 5% chlorine.
EXPERIENCE WITH NUCLEAR INSTRUMENTATION TECHNIQUES IN THE PREPARATION OF HARD COAL

G. FAUTH, D. LEININGER, H. LÜDKE
Bergbau-Forschung GmbH,
Essen, Federal Republic of Germany

Abstract

EXPERIENCE WITH NUCLEAR INSTRUMENTATION TECHNIQUES IN THE PREPARATION OF HARD COAL.

The application of nuclear techniques to the monitoring and control instrumentation in Federal German coal preparation plants is reviewed. Most instruments installed to date are used for determining the quantitative properties of flowing and static products, such as bunker levels, mass flow on conveyor belts and pipes, density and solids concentration in slurries. Provided regular maintenance and checks are carried out, these instruments have proven adequate in most monitoring and control applications. Many more problems are involved with the on-stream analysis for ash, moisture and sulphur. About 35 gamma-ray backscatter instruments are in operation for determining the ash content of metallurgical and steam coal, requiring primary cutting. Transmission techniques suitable for bulk materials on conveyor belts and for slurries are under investigation. No nuclear technique is practicable for the determination of moisture content. Extensive comparative on-plant measurements have proved the superiority as regards accuracy of microwave techniques developed at Bergbau-Forschung.

1. INTRODUCTION

As in other countries, nuclear instrumentation is widely used in the Federal German coal industry. One of the main fields of application is the preparation, the cleaning of coal. The emphasis of this paper is directed to experiences with such nuclear instrumentation, more than on the presentation of new developments.

In the Federal Republic of Germany (FRG) about 160 million tonnes of hard coal are produced per year underground and prepared to 85 million tonnes usable output. In the preparation plants today the hourly throughput is sometimes more than 1000 tonnes, which represents a net product of about DM 120 000 per hour. High standards are demanded of preparation plants with regard to operational reliability and serviceability, since unexpected stoppages and yield losses give rise to unjustifiable costs or losses of profit.

By way of introduction, Fig. 1 shows a simplified flow diagram for hard coal preparation as commonly practised in the Ruhr area. Also indicated are some of the important measuring points for on-line analysis of product quality which are required for process control, as well as instrumentation for quantity properties.
From the hoisting shaft the run-of-mine coal is conveyed to the preparation plant via a homogenization pile. In the first stage of the process, the prescreening, the raw coal is screened at about 10 to 14 mm. The coarse coal is washed in jigs or heavy media separators. The smalls first are de-dusted pneumatically. The cut-point here is at 0.5 to 0.75 mm. The fine product, the dust, is a coal component exhibiting ash contents from 12 to 30% and will be blended to the final product, metallurgical or steam coal. The product from 10 to 0.5 mm is cleaned in fine coal jigs and dewatered in centrifuges. The slurry produced during wet preparation is collected in a thickener and fed to the froth flotation process. The concentrate, the clean coal, and tailings are dewatered using filters and solid bowl centrifuges, or chamber filter presses and belt screen presses respectively. The constituent products — coarse clean coal, fine clean coal, flotation concentrate, dust and middlings — are mixed together to form the saleable final products, metallurgical or steam coal.

For the purpose of monitoring and controlling individual preparation devices and process steps, extensive instrumentation and control equipment is installed in the preparation plants. Figure 2 indicates the type and frequency of instruments used in 26 of the 48 preparation plants in the Federal Republic of Germany. The absolute figures do not hold for the totality of all the preparation plants, but the relation between the different types of instruments is representative for all the plants. It clearly can be seen that the instruments used to date have mainly been those for determining the quantitative properties of flowing and static products.
For monitoring and control these instruments have proven adequate in most cases, provided that regular maintenance and checks are carried out. The determination of qualities at present is restricted almost entirely to the ash content of the final products.

For both tasks, some instrumentation is used which is based on nuclear techniques. In the following the type of instruments, the working location and operational experience will be treated in greater detail.

2. DETERMINATION OF QUANTITIES USING NUCLEAR TECHNIQUES

2.1. Bunker level monitoring

In the FRG there are a number of companies offering level meters. Measurement generally is based on the well-known transmission attenuation of gamma rays emitted from $^{60}$Co sources. The source strength used is between 5 and 100 mCi. Instruments of this type are installed both in bunkers underground and in those on the surface. They mostly are used to monitor an upper or lower limit level, less for continuous determination of the level.

These instruments have proven to be very reliable and to require little maintenance. Their main advantage is that they work on a non-contact basis, so there is no risk of destruction, for instance, by the collapse of product overhang in the bunker.

![Number of Instruments (nuclear instruments)](image)

FIG. 2. Type and frequency of instruments for measuring quantity and quality properties in 26 of 48 coal preparation plants in the FRG (1980).
At one mine in the Ruhr area, for example, there are about 75 nuclear bunker level meters in use on the surface and underground.

2.2. Mass flow determination on conveyor belts

As an alternative to mechanical belt scales, nuclear belt scales are available for determining the feed rate. A rod-shaped source with a nuclear activity distribution corresponding to the loading profile on the belt or a punctiform gamma-ray source is mounted above or below the conveyor. For applications in hard coal preparation the radiation source which can be considered in most cases is $^{137}$Cs and in extreme cases $^{60}$Co.

Such a weighing system is characterized by simple installation and short overall length and can be used also in chain conveyors. The main advantage, however, is the non-contact technique, avoiding any mechanical problems connected with the transmission of forces.

Although nuclear belt scales have been on the market for some time, to date they have not established themselves widely in the Federal German coal mining industry. The reason for this may be, apart from the understandable caution exercised as regards the use of yet untested devices, that on the basis of present knowledge, no cost benefit and no increase in accuracy in comparison with mechanical scales is to be expected. The application of nuclear scales requires a constant loading profile on the belt. Reliable statements can only be given after a test period planned for the near future.

2.3. Determination of density and solids content in slurries

As shown in Fig. 2, measuring instruments for determining the density and solids content in slurries are very frequently used in preparation plants. In most cases the technique applied is the transmission method using gamma rays with $^{137}$Cs as radiation source.

These instruments are used primarily during three processes for monitoring or closed-loop control:

- heavy media washing of coarse coal
- froth flotation of slurries
- thickening of suspensions.

When coal is subjected to heavy media cleaning, it is separated into the three products clean coal, middlings, and tailings by using mostly magnetite as solid heavy medium. In order to avoid loss of heavy medium solids, the suspension is circulated and set to a certain density by the addition of medium solids or diluting water. The accuracy of the density control as determined by the density meter should be 0.01 g/cm$^3$. 
With froth flotation the quantity of suspension and the concentration of solids are measured and in a number of preparation plants controlled. The appropriate quantity of flotation agent is added according to the solids rate calculated from the two readings of mass flow and concentration.

With the thickening of suspensions nuclear instruments are used for monitoring and in some cases for controlling the discharge of the underflow of the thickener.

With heavy media densities above about 1.4 g/cm\(^3\) and with solids concentrations above about 100 g/L, the instruments are working satisfactorily. Sometimes, however, it is difficult under operational conditions to convey the slurry free of air. Even small air inclusions can result in substantial measuring errors, especially with low solids concentrations. Similarly, variations in mineral composition can produce deviations which necessitate recalibration.

3. DETERMINATION OF COAL QUALITY PROPERTIES

3.1. Determination of ash content of bulk materials using gamma-ray backscatter techniques

Twenty years ago Hardt [1] at Bergbau-Forschung developed the backscatter method using \(^{241}\)Am as nuclear source for on-line determination of ash content in fine coal. With the nuclear, electronic and computational means available at that time the problem of varying bulk density and mass flow on belt conveyors could be solved best by this backscatter method. Since then this method has found general acceptance in the FRG's hard coal mining industry and about 35 instruments are in operation now.

Coal is sampled from the main product stream, fed to the monitoring device and pressed through a plastic tube by a conveyor screw. In Fig. 3 the arrangement of nuclear source, scintillation detector and sample stream is shown. Great importance is attributed to representative sampling reliable under industrial conditions.

At present, two types of sampling devices are used in the plants, according to the structural conditions.

The first device has a hydraulically operated swivelchute as the actual sampler by which about 5 to 6 t/h are withdrawn from the main stream.

With the second device a channel sampler is connected with an open screw conveyor feeding the sample to the pressure screw upstream of the measuring device. With this device it has been found that the sample material is well mixed in the worm conveyor, so that a mixer can be dispensed with upstream of the monitoring device.

Apart from frequent problems in introducing a cutter at the desired point and the high costs of such cutters, choking by wet material and foreign matter
creates the need for additional automatic scrapers and high maintenance expenditures.

In contrast to the transmission method, an additional measurement of bulk density or, better, mass per unit area is not necessary with the backscatter method. On the one hand a certain levelling of bulk density is achieved by pressing the sample through the tube by the conveyor screw, on the other hand an optimal distance between the coal surface and the measurement frame can be found where the effect of varying bulk density is minimized, as can be seen in Fig. 4.

**FIG. 3.** Arrangement of nuclear source, detector and coal sample stream for on-line ash analysis by the backscatter method.

**FIG. 4.** Backscattered radiation intensity versus distance between coal surface and ash gauge measurement frame for different bulk densities.
Theoretical and experimental investigations by Bachmann [2], however, showed that this so-called density-independent distance is a function of the ash content. So the geometry of the monitoring device must be adjusted to the type of final product, i.e. metallurgical or low-grade coal.

The ash monitors are used with the following coal grades:

- metallurgical and high-grade steam coals with a top size of 20 mm and ash contents in the range of 5 to 12%,
- low-grade steam coals with a top size of 25 mm and ash contents in the range of 15 to 35%.

The output is used to control the ash content in the final product, metallurgical or steam coal, according to a preset value. This is achieved by a dosed addition of a partial product which deviates substantially in ash content from the preset value, for instance dust, in a closed-loop control. In this way it was possible to improve the observance of the preset ash content as well as to reduce the range of variation, as compared to uncontrolled or manual operation. Extensive investigations and current comprehensive monitoring of the results revealed an accuracy as expressed in a double standard deviation of the nuclear determination from laboratory assay of about 0.3 wt% for high-grade coal and about 1.5 wt% for low-grade steam coal.

FIG. 5. Calculated backscattered radiation intensity versus ash content for coals of different moisture content.
The variability of the ash content with time as expressed in the double standard deviation of hourly mean values from the monthly mean value for example at the Schlägel & Eisen Mine could be reduced from 1.1 wt% to 0.3 wt% by applying the ash monitor aided control of the blending facility.

The main disturbing variables affecting the accuracy of the instrument aside from the above-mentioned bulk density variations are moisture and iron content. Bachmann [2] has shown that fortunately the influence of moisture content is minimal at ash contents of about 8%, which are characteristic for the common metallurgical coals. At lower ash contents as well as at higher ash contents the backscatter count rate depends on the moisture content, as can be seen in Fig. 5. At an ash content of 30 wt%, which is usual for high-ash steam coal, a deviation in the moisture content of 1 wt% results in a shift of the ash meter reading by 0.4 wt%.

It therefore is possible to improve the performance of such backscatter ash meters by simultaneous measurement of bulk density and moisture content, though under industrial operation this may be of questionable benefit. Some results are presented here for illustration: at one mine 32 samples of metallurgical coal were taken at the outlet of the ash gauge tube and analysed by standard methods. The correlation between the results of this laboratory assay and the ash meter readings yielded a determination coefficient $R = 0.85$ and a standard deviation $s = 0.52$ wt%. Simultaneous on-stream measurements of bulk density and moisture content as reported by Fauth et al. [3] and the inclusion of these results in regression calculations led to a determination coefficient $R = 0.95$ and a standard deviation $\sigma = 0.34$ wt%.

The influence of iron content is also dependent on the ash content. Calculations for the backscatter method resulted in a shift of about 8 wt% ash content when varying the iron content in a coal of 25% ash content by 1 wt%. Recent experimental investigations by Bachmann [2] exhibited a shift of 6.2 wt% ash content, applying the transmission method and a coal of slightly different ash composition. Because of the low variation in iron content in the final products, especially in high-grade coal, no problems have occurred during operation originating from varying iron content when the ash monitors are regularly calibrated, as the high accuracies obtained in practical operation have shown.

3.2. Determination of ash content of bulk materials using gamma-ray transmission techniques

Apart from the industrial instruments just described, laboratory-scale instruments are being developed by Bergbau-Forschung for measuring the ash content directly on conveyor belts without any sampling or belt contact using transmission techniques. Such instrumentation is required because the sampling equipment involves high investment and maintenance costs and it often is not possible to ensure trouble-free operation.
The interfering influence of density variations obtained with the backscatter method is compensated for in the transmission method by combining the measurement of attenuation of high- and low-energetic radiation transmission.

Successful tests have been conducted in this respect with a $^{241}$Am/$^{137}$Cs source, a scintillation counter and evaluation electronics comprising a desk-top computer.

With the transmission method care must be taken that the mass per unit area of the sample to be irradiated does not exceed about 20 g/cm². Otherwise no usable Am signal reaches the detector.

The influence of moisture content is very similar in both backscatter and transmission techniques. Whereas the results for metallurgical coals with an ash content of approximately 7% ash are almost unaffected by variations in moisture content, this is not the case for low-grade coal with an ash content of approximately 20 to 35%. Here the falsification of the ash reading amounts to 0.3 wt% for a change in moisture content of 1 wt%.

The iron content affects both the backscatter and the transmission technique to the same extent because backscattered radiation and attenuated transmitted radiation both are determined by the absorbing photo-effect which is governed by the effective atomic number.
Measurements performed at a test rig for ash determination with transmission techniques revealed an overall accuracy of 0.9 wt% (2s) including sampling and analysis errors for samples covering ash contents from 5 to 40%, provided different calibrations are used for different types of coal products. The results are displayed in Fig. 6.

These results mean that the accuracies to be expected for transmission measurements with products of low ash content, such as high-grade coals, will be similar to those for backscatter measurements. With high-ash products, such as low-grade coals, transmission measurement should be superior to backscatter techniques.

3.3. Determination of ash content in slurries using gamma-ray transmission techniques

For monitoring and control of the flotation process, which is becoming increasingly important in coal preparation, the measurement of ash content is an absolute prerequisite. One possible technique is nuclear determination using transmission techniques as first applied by Lyman and Cheshire [4] in Australia.

If the slurry is idealized as a mixture of the three components pure coal, ash-bearing minerals, and water, the linear attenuation coefficient for gamma rays penetrating the slurry is a function of the six variables: the three mass attenuation coefficients of the three components, the density of the slurry, the solids concentration in the slurry, and the ash content of the solid particles. Assuming a correlation between ash content and density of the solids, the variable slurry density may be calculated from the solids density and solids concentration and is not an independent variable. Assuming moreover that the mass attenuation coefficients are constant, the number of independent variables reduces to two.

![Graphs showing calibration plots for nuclear determination of solids concentration and ash content in coal slurries.](image-url)
At least for coals from the Ruhr area a significant correlation between ash content and solids density holds [5]. Deviations from this correlation as well as from the assumed attenuation coefficients, which may occur with variations in coal composition, must be compensated for by calibrations, if the determination of solids concentration and ash content in slurries is restricted to the measurement of the only two variables. This may be achieved by detecting the transmitted intensity at two different gamma-ray energies similar to the ash content determination of bulk materials.

The low-energy radiation source used at Bergbau-Forschung is $^{241}$Am, and the high-energy source is $^{137}$Cs. In a semitechnical installation, very similar to the Australian arrangement, the measuring technique together with data logging by a small process computer was tested and disturbance variables such as temperature, pressure and mass flow were investigated. Calibrations for several products were established. In these investigations an accuracy as expressed in double standard deviation of 2 g/L was found for the determination of solids content in the range up to 200 g/L and 0.8 wt% for the determination of the ash content for samples containing ash contents in the range from 4 to 70%. These results are presented in Fig. 7 and hold for measuring times of 200 seconds.

4. USE OF NUCLEAR TECHNIQUES FOR THE DETERMINATION OF MOISTURE AND SULPHUR CONTENT IN HARD COAL

As in many other countries, in the FRG extensive investigations into the applicability of the various available moisture measurement methods on coal have been carried out [6, 7]. The only nuclear technique using neutron transmission fails because of varying hydrogen contents in coal. Pulsed nuclear magnetic resonance techniques exhibit a suitable performance [6], but the restriction that the sample must pass through a tube of a very small diameter at present prevents an industrial application. Most promising are electromagnetic techniques, but no technique has yet been found to meet all the requirements of a preparation plant, e.g. economic efficiency, low maintenance costs, precision, and accuracy. The result of comparative on-plant measurements with conductance, capacitance, infrared reflectance, and microwave techniques and instruments has proved the superiority as regards accuracy of the microwave technique [7].

With on-stream moisture determination using microwave transmission techniques it is necessary to determine simultaneously the mass per unit area. Special attenuation and phase shift measurement methods as developed by Klein [8] are applied to determine the transmission properties in the microwave range, whereas use is made of a 15 mCi $^{137}$Cs source for measuring the mass per unit area. A combined arrangement with suitably adapted laboratory instrumentation in initial tests under industrial conditions and with several types of products exhibited an accuracy of $s = 0.27$ wt% and a determination coefficient of
FIG. 8. Microwave moisture content determination of Ewald metallurgical coal as measured at the tube of an ash meter.

FIG. 9. Microwave moisture content determination of General Blumenthal metallurgical coal as measured at the loading bunker feed belt.

B = 0.69 (due to the limited range of moisture variations from 8.5 to 11 wt%) as measured at the tube of an above mentioned backscatter ash meter, and s = 0.38 wt%, and B = 0.90, as measured at a running conveyor belt. Typical calibration results are presented in Figs 8 and 9.

The tests with other moisture monitoring methods at the same industrial sites also have shown that in certain cases additional consideration of the mass per unit area results in an improvement of the accuracy.

For the on-stream determination of elements in hard coal, primarily sulphur, prompt gamma neutron activation analysis (PGNAA) is a promising technique. Instruments are offered commercially by several companies in the USA. The PGNAA method also has been investigated by Herzog and Wilde [9] with the
State Material Testing Institute in Dortmund for coal and recently for cement on a laboratory scale, but no instrument is commercially available at present for industrial application.

With the high accuracy of about 0.02 wt% sulphur content as required in the FRG, and taking into account the relatively small variation at low levels in the saleable products as well as the high costs of the equipment itself and the necessary adaption of the production process technology when introducing such instrumentation, the use of the PGNAA technique at present would not appear to be viable.

5. CONCLUSION

In the coal mining industry at the present time a number of measuring instruments are in use for the nuclear on-line determination of quantities and qualities. With the determination of quantities these instruments display a high level of reliability, although the environment and potential disturbance factors such as air inclusions or variations in the material composition should be taken into account when the solids concentration is being determined. This is even more true with regard to the determination of qualities such as, for instance, the ash content.

Operational experience has shown that, even with electronics, improvements are necessary and, on the basis of current technical knowledge, possible.

Special problems exist in the determination of accuracy. With feed rates up to 1000 t/h, partially with unknown property distributions within the product stream, extensive and sophisticated sampling according to statistical principles is necessary. This sampling must also cover the determination of sampling error and analysis error, when the mean values over lengthy periods are compared with individual or averaged readings from continuous-measuring instruments.

At the present stage of development and in accordance with the requirements of the market and environmental protection regulations, it can be anticipated that in future much wider use will be made of instrumentation based on nuclear technology. The greatest emphasis will be directed to the on-stream determination of moisture and ash content in the various products present in coal preparation plants.

Investigations and developments conducted recently have shown that progress can be expected, especially in operational applications, even if not as rapidly as might be desirable. The accuracy of this instrumentation will be sufficient for process control of the preparation process, whereas its application for accounting of delivered shipments appears to be still a long way off.
REFERENCES


DISCUSSION

J.S. WATT: I was interested to see your results on measuring solids concentration which showed something like a 2% error in solids, which is extremely good. We cannot get even a uniform slurry to the accuracy you obtained.

G. FAUTH: To get such high accuracy entails the problem of getting a correct sample. We initially had a very large problem with sampling in our test loop and we had very large variations in the reference samples. We remove entrained air and must use high pressure. We must measure the pressure and compensate for it. We also investigated several sampling points and it turned out the best method to get representative sampling was to take all the stream from one of these points.

D. PAGE: You commented, when talking about your ash monitor system, on the effect of iron variability and the size of this effect. We have very recently done some experimental backscatter work in which we get a value of about 6% ash for a 1% iron deviation, which ties in with what you said. But you say you don't really have any problems because the variation of the iron in your product
is quite low. Do you have any actual figures of what these variabilities are? We find they are quite high in some cases and iron becomes a problem.

G. FAUTH: We have made no detailed investigation of this but we have obtained a surprisingly high accuracy of the ash monitor itself. So we think that the iron content can’t be a problem.

D. PAGE: I think partly, in terms of standard deviation of iron in the coal, you may get a high variability. But you may also find there is a good correlation between iron and ash. So of course that disappears in the calibration. I would be interested to know for German coals, between what kinds of limits do you expect the iron content to vary?

D. LEININGER: That depends on the ash content. About 8% of the ash content is iron. You need to add the pyrite concentration also. Then you will have a variation between 6 and 8%, I think, of iron in ash.

D. PAGE: That’s not a very wide variation in my experience.

D. LEININGER: No. Therefore we need no compensation of the iron content. We worked on it but it wasn’t necessary.
NUCLEAR TECHNIQUES FOR THE ON-LINE ANALYSIS OF COAL IN ELECTRICITY GENERATING STATIONS

P.L. SURMAN
Central Electricity Generating Board,
Scientific Services Department,
North Eastern Region, Harrogate,
United Kingdom

Abstract

NUCLEAR TECHNIQUES FOR THE ON-LINE ANALYSIS OF COAL IN ELECTRICITY GENERATING STATIONS.

A modern 2000 MW power station burns about 5 million tonnes of coal annually. The advent of on-line coal analysis will allow the operator to manage the fuel supply within the station instead of having to wait for up to two weeks for information on coal quality, by which time the coal has already been burnt. A number of specific applications for on-line coal analysis have been identified and the paper discusses these and operational requirements such as commercial price negotiation, policing of fuel supply, identification of abnormal deliveries, management of fuel stocking and blending, allocation of fuel to units and bunkers, feed forward combustion control, reduction of slagging, and control of emissions.

1. OPERATIONAL REQUIREMENTS

A large proportion of coal is delivered to British power stations by rail. Typically it is supplied in 30-tonne wagons in 1000-tonne consignments. Coal is discharged from the wagon bottoms into a track hopper and is then conveyed either to the bunkers or to stock. A typical 2000 MW power station may receive 20 consignments a day which may come from as many as 12 collieries. This pattern of fuel supply can result in quite large fluctuations in the quality of fuel being presented to the boilers. The bar chart (Fig. 1) illustrates a typical variation in ash levels in successive 1000-tonne consignments delivered by rail and corresponds to standard deviations of 3% absolute for ash. A number of operational advantages would arise from the employment of on-line coal analysis at the point of delivery.

1.1. Coal stocking and blending

Deliveries usually take place on Monday to Friday and therefore a proportion of deliveries must be diverted to stock for weekend use. Stocking policy is
usually determined by operational requirements and only occasionally are particular brands diverted to stock for the purpose of blending.

Present design and operational practice does not give much scope for fine control of coal quality by blending. In practice there would be considerable advantage in the detection of coal outside specification so that the consignment could be rejected before discharge or at least diverted to be spread out on the stock. In order to reflect this an on-line analyser would have to respond rapidly (within a few minutes) but to a precision of about ±10% relative for ash.

One potentially attractive option would be to use information on coal analysis to allocate specific ranges of quality to specific bunkers or units. This, of course, would be subject to the overriding operational constraints of securely fuelling the station. The required precision and response time would be as above.

1.2. Boiler combustion control

Whilst coal is purchased against a specification for a particular power station, the coal supplied to the boilers is inevitably variable in quality (cf. Fig. 1) and in practice, operators optimize their combustion conditions to the average product. One potential advantage of on-line analysis would be to feed forward information of coal quality so that combustion conditions could be ‘fine-tuned’ with con-
sequent gains in boiler efficiency. It is estimated that up to $\frac{1}{2}\%$ increase in boiler efficiency could be gained from such control.

Ideally a response time of less than five minutes would be required at the mill feeders. Alternatively, detection of a step change in coal quality in the supply to a bunker would be of value and in this case a response time of up to 15 minutes would be acceptable. A precision of $\pm 5\%$ (relative) for ash and moisture would be required for this purpose.

Analysis of the fuel before the bunkers would require the development of a flow model from which predictions of output could be made from the input data. Alternatively, simple ash analyses might be effective at the bunker output as long as they are ‘calibrated’ from the complete analyser.

1.3. Slagging and fouling control

As facilities for blending are generally not available at British power stations, the detection of more slagging coal can be used as a warning. For example, the operator might be able to change mill combinations or adjust combustion conditions. Advantages of on-line analysis are more likely to come from an increased understanding of the mechanisms of slagging which would arise from detailed knowledge of the coals being burnt. A response time of 5—15 minutes and a precision of about $\pm 10\%$ (relative) for ash elements and sulphur would be required for this.

If a bunker allocation scheme was in operation then it might be possible to direct slagging coals to a unit on less than full load or a particular burner set which had been adjusted to minimize slag formation.

1.4. Control of emissions

With the increasing concern over the control of power station emissions, knowledge, on-line, of coal quality is becoming of greater potential value.

Knowledge of the sulphur content could be used to optimize precipitator efficiency and to help provide a mass balance for sulphur through the system. It could also be used to allocate higher sulphur coals to a unit designed to operate with reduced emissions.

Information on nitrogen content could conceivably be used to adjust combustion conditions to minimize the formation of nitrogen oxides. Further reduction in nitrogen oxide emission may be obtained from the use of specially designed burners. These perform best with a consistent coal quality and the use of on-line analysis to control quality or to feed information to the burner controls is becoming an interesting possibility.

A precision of 10\% relative for sulphur and nitrogen on a time-scale of five minutes would be needed for these applications.
2. COMMERCIAL EVALUATION

The coal received at power stations is regularly sampled and analysed in order to check the supplier’s evaluation and arrive at an agreed analysis for the commercial accounting period. The basis for these evaluations are British Standards 1016 and 1017.

Analyses required for the evaluation of power station coals are:
- Gross as-received calorific value, which provides the base price.
- Ash and sulphur content, from which secondary adjustments are calculated.

An acceptable level of accuracy would be that which is currently achieved by British Standard methods of sampling and analysis. For a sample from a single consignment the following overall accuracy can be obtained:

- Ash (<10%) ±0.5% absolute (dry basis)
- Ash (10—20%) ±5% of true value (dry basis)
- Ash (>20%) ±1% absolute
- Total moisture (<10%) ±0.5% absolute
- Total moisture (10—20%) ±5% of true value
- Total moisture (>20%) ±1% absolute
- Sulphur ±0.15% absolute
- Gross calorific value ±400 kJ/kg

An on-line analyser would be required to have a similar performance.

Over a three month accounting period, the precision of the mean estimate of quality will improve. For example, if a supply over three months is represented by about sixty samples, then for a coal of about 15% ash and moisture the overall precision of estimation of the mean would be about:

- Ash ±0.1% absolute
- Moisture ±0.1% absolute
- Sulphur ±0.02%
- Gross calorific value ±50 kJ/kg

This, in effect, represents the long term reproducibility which would be acceptable in an on-line analyser.

In order to reflect adequately individual consignments which may pass the analyser in less than 45 minutes, it is considered that the above precision would require a reporting frequency or time constant of less than 10 minutes.

The checking of consignments on receipt at power stations raises problems, particularly for the larger proportion of coal delivered by rail. This is because it is usually not possible to empty the track hoppers between deliveries, and obtaining a representative sample of each consignment is difficult.
One approach employed is to use a separate track sample hopper. One wagon from a train is selected at random and discharged separately into the sample hopper to provide, after further sampling and reduction, samples for laboratory analysis.

In the absence of a track hopper sampling system, information about individual coal qualities over a period can be obtained from mixed samples by the application of multiple regression analysis to data obtained from representative samples of all coals taken from the conveyor belt and the supplier’s declared analysis for the fixed period of sampling. Over a period, a pattern becomes established which allows the identification, with reasonable confidence, of any colliery where the quality of supply has become suspect. Whilst never reaching the precision of track hopper sampling for individual brands, this approach is economical in its requirements for capital investment and labour and is seen as a valuable method for the ‘policing’ of coal supplies. It also provides an accurate overall estimate of heat received in a given accounting period.

The imminent availability of on-line analysis of coal provides an opportunity to refine the approach towards the sampling of rail-borne coal. Conceptually, at least, the use of on-line elemental analysis could be used to convert the above method from a ‘policing’ device into a more rigorous sampling and analysis method. For example, rapid analytical reports of some elements could be applied to reconstruct, by computer, the quality of the separate consignments from the overlapping series of products sampled on the conveyor belt (unscrambling). On-line analysis could thus improve the precision of check analyses made at power stations for evaluation purposes.

3. DISCUSSION

A number of applications for on-line coal analysis have been identified in the previous sections. Whilst any individual application might not justify an expensive installation, the total of applications looks attractive. The advent of on-line coal analysis opens up the prospect of a coal management scheme for power stations. The information flows involved are shown in Fig. 2. The overall operational objective in nearly all applications is to present a more uniform fuel for combustion.

3.1. Available instrumentation

There are a number of available instruments on the market and many of these are listed in Table I. Only the analysers using prompt gamma are likely to meet most power station requirements because the ash analysers are affected by ash composition and would need separate calibrations for each variety of coal delivered. It is conceivable, however, that ash meters could be utilized in
<table>
<thead>
<tr>
<th>Instrument</th>
<th>Country of manufacture</th>
<th>Principle</th>
<th>Analysis</th>
<th>Approximate cost (£)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Wultex</td>
<td>UK/Poland</td>
<td>Gamma backscatter</td>
<td>Ash</td>
<td>50 000</td>
</tr>
<tr>
<td>Coalscan TB</td>
<td>Australia</td>
<td>Gamma transmission</td>
<td>Ash</td>
<td>90 000</td>
</tr>
<tr>
<td>Gunson Sortex</td>
<td>UK</td>
<td>X-ray backscatter</td>
<td>Ash</td>
<td>100 000</td>
</tr>
<tr>
<td>Coalscan</td>
<td>Australia</td>
<td>Gamma pair production</td>
<td>Ash</td>
<td>150 000</td>
</tr>
<tr>
<td>RASH</td>
<td>USA (SAI)</td>
<td>Dual beam gamma transmission and microwave</td>
<td>Ash</td>
<td>180 000</td>
</tr>
<tr>
<td>Sulphur meter</td>
<td>USA (SAI)</td>
<td>Prompt gamma (NaI)</td>
<td>Sulphur</td>
<td>about 300 000</td>
</tr>
<tr>
<td>ELAN</td>
<td>USA (MDH)</td>
<td>Prompt gamma (NaI)</td>
<td>Ash</td>
<td>400 000</td>
</tr>
<tr>
<td>Gamma Metrics</td>
<td>USA</td>
<td>Prompt gamma (NaI)</td>
<td>Ash</td>
<td>400 000</td>
</tr>
<tr>
<td>CONAC</td>
<td>USA (SAI)</td>
<td>Prompt gamma (NaI; Ge, Ne)</td>
<td>Ash</td>
<td>400 000</td>
</tr>
</tbody>
</table>
conjunction with an elemental analyser which could provide continual calibration data. The particular attraction of some ash meters such as the Wultex, Coalscan TB and RASH is that they can be installed directly on existing conveyor belts with a minimum of engineering.

3.2. Suitable instrumentation

The three available prompt gamma elemental analysers have roughly similar specifications and all seem to meet the stated operational requirements. Obviously a careful test programme would need to be mounted to ensure that the specifications were met for particular coal varieties.

As an example, Table II compares the published performance figures on CONAC with the required performance. It can be seen that in general the performance is adequate for operational purposes although some development work is still desirable.

3.3. Accuracy for commercial evaluation

Whilst the elemental analysers can probably meet the required accuracy for a single consignment, it is unlikely that they will meet the quoted precision of estimation of the mean of sixty samples. This is because the analysis is based
### Operational Requirements for On-Line Coal Analysis

<table>
<thead>
<tr>
<th>Function</th>
<th>Required precision</th>
<th>CONAC precision (relative)</th>
<th>Required response time</th>
<th>CONAC response time</th>
<th>Comment</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Commercial pricing</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Ash, moisture, sulphur</td>
<td>5%, 5%, 5%</td>
<td>4%, 3%, 5%</td>
<td>10 min</td>
<td>5 min</td>
<td>Development work needed to establish high accuracy required for commercial negotiation</td>
</tr>
<tr>
<td>Calorific value</td>
<td>1.5%</td>
<td>1.8%</td>
<td>10 min</td>
<td>20 min</td>
<td></td>
</tr>
<tr>
<td><strong>Blending, stocking, bunker allocation</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Ash</td>
<td>10%</td>
<td>4%</td>
<td>5 min</td>
<td>5 min</td>
<td>CV on a shorter time-scale could be obtained</td>
</tr>
<tr>
<td>Calorific value</td>
<td>2.0%</td>
<td>1.8%</td>
<td>5 min</td>
<td>20 min</td>
<td></td>
</tr>
<tr>
<td><strong>Combustion control</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Ash</td>
<td>5%</td>
<td>4%</td>
<td>5 min</td>
<td>5 min</td>
<td></td>
</tr>
<tr>
<td>Moisture</td>
<td>5%</td>
<td>3%</td>
<td>5 min</td>
<td>20 s</td>
<td></td>
</tr>
<tr>
<td><strong>Slagging and fouling control</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Ash elements</td>
<td>10%</td>
<td>10–20%</td>
<td>5–15 min</td>
<td>20 min</td>
<td>Some information on ash elements could be extracted on shorter time-scale</td>
</tr>
<tr>
<td>Sulphur</td>
<td>10%</td>
<td>5%</td>
<td>5 min</td>
<td>5 min</td>
<td></td>
</tr>
</tbody>
</table>
upon different principles and because sufficiently accurate calibration standards are not currently available.

The empirical solution to this problem is for the analyser to be equipped with separate calibrations for each coal brand or alternatively for secondary correction factors to be established. It is doubtful that this issue will be completely resolved in the laboratory and it may require prolonged field trials where bias tests between conventional sampling and analysis and the elemental analyser are conducted.

4. CONCLUSIONS

A number of potential benefits have been identified for on-line coal analysis at power stations. Currently available instrumentation is adequate to achieve many of the operational requirements. Experience in the field will confirm the ultimate value of such devices.

DISCUSSION

J.S. WATT: One thing which becomes obvious from those train shipment figures, which to me are very dramatic, is that one needs not to have something at the power plant but have something at the coal mines. I know the problems become financial ones between two organizations. But it seems that is very important. You need a simple analyser back at the colliery.

P.L. SURMAN: It is not so much individual collieries, it is that a big power station needs to be serviced from a number of small collieries. So, unless they ship their coal to a common point for blending we are bound to end up with this situation. The National Coal Board have responded to this in England, and they have put some very large washeries in Yorkshire. That has improved things, but the standard deviation in ash we get now from the washeries is 3%. So it is ±6% as the range of composition. It is only through money that we can require that things change. Perhaps our pricing structure ought to reflect the real cost of variations to us. But, working with the present structure, there are things we can do ourselves and that is what I was trying to explain.

A.S. PRASAD: What is the carbon content of the fly ash and what do you do with the fly ash?

P.L. SURMAN: I think the carbon content is 3 or 4% in the fly ash. We make artificial hills with the fly ash which are then reclaimed. We have special ash disposal sites on the Vale of York and the whole scheme is that eventually the land will be raised 40 feet with a southern slope. It is inland disposal. We are finding it difficult to get further planning consent for these things.
J. ŁOSKIEWICZ: What is the time necessary to take samples of an 80 wagon train?

P.L. SURMAN: A single train unloads in about 25 minutes. That is one of the things that determines our response time.

J. ŁOSKIEWICZ: The question is, how much time do you need to get the samples from the train?

P.L. SURMAN: We have to work on an incremental time-scale of less than 25 minutes in order to get several increments of coal from that particular consignment. This is why we are talking of 5—10 minute time-scales. Are you thinking more of how quickly do we need advance information in order to take action? Five minutes. We want to act within a few minutes, otherwise most of the coal has already been burnt before we have time to do anything about it.

J. ŁOSKIEWICZ: This I understand, but this means you can get only one or two samples and this cannot represent the total.

P.L. SURMAN: No, it may not. But the variation within a train is much less than the variation between trains. It is about a third of the variation between trains, in our experience.

C.G. CLAYTON: Dr. Gozani was making a strong point on what it was that is required. This is clearly one answer, but it is one answer from a largish organization. It must be a fact that the producers will do whatever the users require in order to meet those specifications. So it is the market research amongst the users that one is really looking for, worldwide I guess. It is difficult to imagine a supplier ever doing anything to improve the quality of his coal unless there were a user demand.

T. GOZANI: Obviously this should be the direction and there is no reason it couldn’t have happened four or five years ago. The fact that the Central Electricity Generating Board had a little bit more foresight is probably due to its people, and the fact that it is a national entity and can have a little longer view compared to American companies, who are looking to the next quarter to make a profit and to satisfy their shareholders. Another aspect is that you need a large utility. The Thames Valley Authority is an example: it has the ability to say “OK, we think that with a knowledge of fouling and slagging or some other properties of the coal we can improve; let’s go ahead and demonstrate it”. Everybody else says “show me”. So there is a time factor for the introduction of these technologies. Compared to a conventional technology we are not slow. Conventional technology takes 25 years to introduce. Electronic technology, microchips, takes 2 years.

C.G. CLAYTON: Yes, I take all those points. I’m not sure it is all due to the CEGB, with all due respect. They obviously have very highly talented personnel. But the fact that there is a high variability in British coals also means that the opportunities for measurement and control are that much greater.
CORRELATION METHODS IN CALORIFIC VALUE MEASUREMENTS OF COAL

T. CYWICKA-JAKIEL, J. ŁOSKIEWICZ
Institute of Nuclear Physics,
Cracow, Poland

Abstract

CORRELATION METHODS IN CALORIFIC VALUE MEASUREMENTS OF COAL.

The relative merits of combustion heat calculation by ash measurement and by $^{12}$C measurement are discussed. The ability of correlation measurements to withstand a high background is shown not only on a theoretical basis but also on an experimental one. The results of previously published correlation measurements of calorific value of coal are shown. The elements of a new more efficient probe are presented and the possibility to shorten the measurement time to about 300 s is discussed.

1. INTRODUCTION

The share of coal in electricity production will probably grow during the coming years. It is therefore an important energy source.

The most important parameter of coal burned for electricity production is its combustion heat. The other parameters of interest are: the content of ash, of moisture, of sulphur, and of elements forming compounds melting at low temperature. If the concentrations of the main coal-forming elements are known, the combustion heat $Q$ can be calculated from formulas as for example that of Grummel and Davies [1]

$$Q = (3.635 \, H + 235.9) \frac{C}{3} + H - \frac{O - S}{3} \text{ kcal}$$ (1)

or by Dulong [2]

$$Q = 81.37 \, C + 345 \left( H - \frac{O + N - 1}{8} \right) + 22.2 \, S \text{ kcal}$$ (2)

Here $C$, $H$, $O$, $N$, $S$ mean weight percentages of carbon, hydrogen, oxygen, nitrogen and sulphur.

It is now possible to measure all elements of interest. We can use neutron radiation to induce gamma-ray emission and to measure with a semiconductor detector the intensity of different gamma-ray lines. In an approximate approach
we may correlate the combustion heat with, say, only carbon content, neglecting other elements.

In Fig. 1 we show the experimental dependence of combustion heat on carbon concentration. The data come from the literature and represent a mixture of Silesian and American coals. The rise of $Q$ with carbon content flattens above 70% C: this effect is probably due to the decrease of the quantity of hydrocarbons at higher carbon contents, as may be seen in Fig. 2 for American coal data. We should note that in Fig. 1 the value of $m = 0.443 \text{ MJ/kg}%C$ ($105.8 \text{ kcal/kg}%C$) is significantly different from the coefficient in Eq. (2) by Dulong. The reason is that there is a negative correlation between ash content
and combustion heat, as shown in Fig. 3. The same is true for moisture and carbon content (see Fig. 4).

The first impression we get from Fig. 3 is that the spread of experimental points is large, much larger than in Fig. 1. The correlation coefficient is much lower, amounting only to −0.25. Different parts of the plot are populated by different coal types.

The data in Fig. 3 come from very different coal basins and even continents. If we take a single coalfield, as is the case in Fig. 5, the data become much more consistent. They fall well on a single straight line with quite a small spread.

It seems therefore that for mine control purposes this type of measurement method is fully appropriate. The situation is different in a power station which receives coal from different coalfields. There the ash content is not the best indicator for assessing the coal quality.

This discussion stresses the merits of using the carbon content as the indicator of the combustion heat value of a coal.

**FIG. 2. The dependence of hydrogen content on carbon concentration for different US coals. In the figure, Δ₁ is the 95% confidence interval, calculated from linear regression.**

FIG. 4. The dependence of moisture content for different US coals on carbon concentration.
2. THE CORRELATION TECHNIQUE FOR CARBON EVALUATION

The simplest method to measure the carbon content of coal is to use the \((n, n' \gamma)\) inelastic scattering reaction on carbon. The reaction threshold on \(^{12}\text{C}\) is about 4.8 MeV. The gamma ray emitted by an excited \(^{12}\text{C}\) nucleus has 4.43 MeV energy. The range of this method is about 20 cm, therefore the sample from which the information is gathered has a volume of 30 litres. If we assign the measured values to a wagon load of 50 tonnes of coal, then the extrapolation error (extension error as it is called in geostatistics) is much smaller than in the case of probes with a sample volume of a few cubic centimetres (XRF analysis).

We would like to use an isotopic neutron source, e.g. \(^{241}\text{Am-Be}\), a source which is much cheaper than a neutron borehole generator and also has a much longer lifetime; the use of an isotopic neutron source means, however, that we have background problems because of an activation. One possible solution is to use a correlation technique which is quite insensitive to accidental background.

A few words on the correlation method seem to be necessary here. If we have the following situation we can use the correlation technique of measurement: The primary radiation from a source, say \(^{241}\text{Am-Be}\), excites in the material a certain nucleus, say \(^{12}\text{C}\). This daughter product emits a secondary radiation, e.g. a gamma ray of energy, which can be registered. The primary radiation is measured by, say, detector X and secondary radiation by
detector Y. Now we divide the time-scale into subintervals of, say, 50 μs long (sampling interval) and we count the number of pulses in channel X, \( x_i \), and in channel Y, \( y_i \), during the \( i \)-th sampling interval. We can calculate the correlation function

\[
\Phi_{xy} = \frac{\sum_{i=1}^{N} x_i y_i}{N} - \left( \frac{\sum_{i=1}^{N} x_i}{N} \right) \left( \frac{\sum_{i=1}^{N} y_i}{N} \right)
\]

(3)

Here \( N \) is the number of measurement cycles. To calculate this expression in real time we need an electronic correlator. As an example of the ability of the correlation technique to work in a high-background environment, the measurement of angular correlation between two gamma rays from \( ^{22}\text{Na} \) may be cited, one in a clean environment, the other with a disturbing \( ^{137}\text{Cs} \) source.

In Fig. 6 are shown the gamma-ray spectra, observed during the tuning stage of our experiment, using the \( ^{22}\text{Na} \) source and a disturbing source, \( ^{137}\text{Cs} \). As can be seen from the figure, the \( ^{22}\text{Na} \) peak is barely visible in the X channel. As is well known, \( ^{22}\text{Na} \) emits two gamma rays in opposite directions, therefore we have the maximum value of the correlation function at the angle of 180° between the X and Y detectors.

The correlation function \( \Phi \) measured with and without the disturbing \( ^{137}\text{Cs} \) source is shown in Fig. 7. The curves without and with the disturbing source show only a minor loss of angular resolution. In such a way the good properties of correlation detection are experimentally demonstrated.

The idea of using the correlation or covariance function in order to measure the lifetime of excited states and also the concentration of atoms is due to Goldanski and Podgorecki [3]. A practical presentation may be found in papers by Cywicka-Jakiel et al. [4] and Schwenk [5]. Czubek has suggested [6, 7] the use of a stochastic process approach for theoretical description of the measurement process.

Here we would only write down the asymptotic (for very long measurement time) equation for the dependence of covariance function \( \Phi \) on the number of excited atoms \( m \) in the medium.

\[
\Phi \text{ as } = \epsilon_x \epsilon_y \ m
\]

(4)

where \( \epsilon_x \) and \( \epsilon_y \) are overall registration probabilities in channels X and Y, whereas \( m \) is the average number of decays of the excited state during the time interval \( \tau \).

In \( \alpha\text{-Be} \) sources the emission of a neutron is accompanied in 85% of the cases by a 4.43 MeV gamma ray. It can be used to signal the neutron emission moment. Thus it may feed the X detector. The neutron travels through the matter and in inelastic scattering reaction excites the \( ^{12}\text{C} \) nucleus, which during
de-excitation emits also a 4.43 MeV gamma ray, which in turn may be registered by our Y detector.

An attempt to measure the carbon content of carbon-ash mixtures according to these gamma lines was made at the Institute of Nuclear Physics in Cracow. The results were subsequently published [4].
FIG. 7. $^{22}$Na angular correlation in the absence and presence of the disturbing $^{137}$Cs source.

FIG. 8. The experimental set-up. Dimensions are in mm.
3. INSTRUMENTATION

The experimental set-up is shown in Fig. 8. The block diagram of computerized electronics is shown in Fig. 9. The single-channel variable-interval correlator was built in our laboratory. Since then a more powerful 16-channel correlator working in real time has been built and used.

The time sequence was obtained from a 10 MHz crystal clock and timer where an appropriate sampling time could be chosen.

4. RESULTS

The results of correlator functioning were

\[ \sum_{i} x_i y_i, \sum_{i} x_i', \sum_{i} y_i \]

and number of sampling intervals N. These data were fed into a PDP11 computer which calculates the covariance function. In a preliminary measurement we used a Pu-Be source with the yield of $1.02 \times 10^6$ n/s. Measurement times of the order $(7-10) \times 10^3$ were used for $1 \mu s$ intervals.
The samples were each measured five times, in order to get an experimental estimate of the standard deviation for each sample. The results are shown in Fig. 10. A clear, almost linear increase of $\Phi/\tau$ with carbon concentration is visible. The standard deviations for particular samples ranged from 1.3 to 4.4% relative. The density of samples ranged from 0.67 to 0.82 kg/dm$^3$. The density and moisture dependence have not been studied.

5. CONCLUSIONS

The measurement errors and/or the measurement time could be substantially reduced by increasing the registration probability of the X detector, i.e. the size of sodium iodide crystal. In the previously reported measurements the NaI Tl crystal in the X detector was only $\phi 40 \times 40$ mm in size.

A new source assembly containing a larger $\phi 51 \times 51$ mm sodium iodide crystal was developed for another experiment. We also used a more powerful 3 Ci Am-Be source producing $6.6 \times 10^6$ n/s. The sodium iodide crystal was put nearer to the source (at 2 cm), thus increasing the detector's solid angle.
FIG. 11. Scintillation crystal shielding for the second version of the source assembly.

A special shielding was developed (shown in Fig. 11) in order to diminish the thermal neutron activation of the NaI Tl scintillator and to diminish the flux of low-energy (<60 keV) gamma rays from the Am-Be source. The detection efficiency increased 5 times and the counting rate 40 times, which put a strain on the photomultiplier divider and pulse-forming circuits. At the moment we are counting a total of 600 000 pulses/s, of which 30 000 pulses/s fall in the 4.43 MeV energy window.

Use of this source assembly would cut the measurement time by a factor of only fifteen because the background also grows.

Further pursuing the attempt to shorten the measurement time we can easily increase the Y detector size to $\phi 4$ in $\times 4$ in instead of $\phi 3$ in $\times 3$ in crystal. This would result in an increase of the measured rate by a factor of 2.7. The background will also increase but it seems to us that we could count on a net gain of two, which would bring the measurement time down to 300 s. It would therefore be nearing industrially acceptable conditions.

ACKNOWLEDGEMENTS

It is a pleasure to thank the Pergamon Press for permission to reproduce here three figures from Ref. [4].

REFERENCES

DISCUSSION

T. GOZANI: You could measure carbon directly by the neutron interaction plus indirect scattering in the coal, or in the carbon itself. What are the advantages of doing the coincidence measurement?

J. ŁOSKIEWICZ: The advantage is to have smaller errors. If the registration efficiency is sufficient, we get smaller errors, because we are using the correlation. We are only taking those gamma rays for which we know that a neutron has been produced.

T. GOZANI: The only gamma rays that will be produced are the 4.4 in the carbon, and the Compton. So with your system you suppress the Compton.

J. ŁOSKIEWICZ: This is a method of suppressing the Compton, yes.

T. GOZANI: The Compton in the coal is suppressed but not the Compton in the detector. This is usually a very large part of it. Therefore the gain would be outweighed by the disadvantage that now you are using coincidence, which usually means much lower efficiency, because you get the product of the efficiencies of the two detectors.

J. ŁOSKIEWICZ: Yes, but this is the reason why the efficiency of the detector that is near the source should be of the order of 30 per cent. Then the loss is small.

B.D. SOWERBY: We did work on carbon measurement using the direct method that you are talking about and we were able to measure carbon contents to within about 1% relative in five minutes or so counting time. So I can’t quite understand the advantage of using the coincidence method either.

T. GOZANI: Did you measure down to very low carbon concentration? As in a fly ash?

J. ŁOSKIEWICZ: Yes, to 20%.

T. GOZANI: The curves showed from zero to 60 so I assumed you measured in the 0 to 4% region as you have in fly ash.

J. ŁOSKIEWICZ: No. 20% is the region where some limits are reached.

C.G. CLAYTON: Coming back to carbon in fly ash, at the 6% level ±0.5%, by the direct measurement. We take about 20 minutes to do it, but that is three times as fast as is required. Obviously we can do it faster if we have a larger source but using a smaller source keeps the radiation problems down.
J.L. MIKESELL: Is the signal that you are correlating with the 4.4 MeV an X-ray from carbon?

J. ŁOSKIEWICZ: If you have an Am/Be source, the source emits at the same time a neutron and a gamma ray of 4.43 MeV, the same energy as that from carbon, in 85% of cases.

C.G. CLAYTON: The alpha from americium goes to the beryllium, which becomes carbon and gives the same 4.43 line from the source as you get from the interaction of neutrons with the carbon in the material.

T. GOZANI: There is no X-ray involved.
SUMMARY AND CONCLUSIONS

1. PREAMBLE

Increases in the cost of coal and the growing pressures to use coal with minimal impact upon the environment have highlighted the requirement for better measurement and control of the total process from mining through beneficiation to utilization. Commercial pressure and modern automated mining procedures have also led to the utilization of lower-grade coals for some applications than would previously have been used, imposing further requirements for control of the quality and of the process operations.

Conventional on-line process control instrumentation, employing electro-mechanical principles, has not always withstood the arduous service conditions required in the coal industry. Off-line systems for quality evaluation are so slow that often operational decisions must be made long before results are available. The representation of thousands of tonnes of coal by grammes of samples has also always been a less than satisfactory procedure for such a highly heterogeneous material as coal. The availability of on-line nuclear techniques can do much to improve this difficult situation because large volumes of coal can be interrogated almost in real time and the methods are usually non-intrusive and robust and do not require contact with the coal.

Clearly no coal producer will go to the trouble and expense of measuring and controlling the quality of his coal unless he is required to do so by his customers, the coal users. And neither will consider using nuclear techniques for this purpose unless they are made aware of the capabilities and advantages of these techniques. It is necessary then to define the requirements of the coal industry, to establish the capabilities of nuclear techniques and to make this information widely available in the industry.

2. SPECIFIC REQUIREMENTS OF THE COAL INDUSTRY

2.1. Coal mining

Techniques are needed to assist in seam delineation and guidance of cutting machines to avoid the undesirable inclusion of adjacent stone strata in the product. In some mining operations both coal and development stone are sent to the
surface sequentially on the same conveyor belt. A fast response detector of stone would provide valuable information to avoid inappropriate material being sent to the beneficiation plant.

2.2. Beneficiation plant

In addition to the diversion of stone, mentioned above, the plant operator needs to assess, on-line, the suitability of coal for beneficiation, blending or stocking. The principle parameter for such assessment is ash (precision ± 5% relative ($\sigma$); time, a few minutes). Measurements of moisture and particle size at this stage would also be of value in some circumstances. Further application of simple ash monitors in each stream within the plant has been identified as a new application which could useful for the early detection of plant malfunction.

Measurements of ash, moisture, sulphur, calorific value and volatile matter are required for commercial evaluation of the end-product and also for assessment and optimization of plant performance. In this case the measurements need to approach the precision and accuracy of current standards such as ASTM, BS, and ISO for coal quality.

2.3. Power generation

In addition to the same requirements as set out in Section 2.2 for commercial evaluation, the operator requires on-line data for the management of fuel stocking, blending and routing; for the control of combustion and boiler slagging; and for the management and control of emissions. This requires the production of data for ash, moisture, calorific value, volatile matter, sulphur, nitrogen and ash elements to a precision of about ±10% relative in a time of about five minutes.

2.4. Iron and steel industries

In addition to measurements of ash, sulphur and moisture to about 10% relative, more rapid assessment of coking properties would be an advantage.

2.5. Exploration and resource delineation

This is discussed in Section 4.

3. INSTRUMENTATION AND CAPABILITIES OF NUCLEAR TECHNIQUES

Many nuclear techniques have been developed over the last decade to allow on-line, near real time analysis of coal, and these have been described in these
SUMMARY AND CONCLUSIONS

Proceedings. A main advantage of such on-line analysers is that they measure a large proportion of the total throughput and avoid mechanical sampling and the errors inherent in taking small samples from a highly variable material. To this end there is a need for development of on-belt analysers with full belt interrogation to avoid the bias which can be introduced in narrow beam instruments. In any case, whenever partial or full coal streams are measured, some care should always be taken to ensure reasonable uniformity of the material distribution across the stream.

Currently there are several commercially available systems based on those techniques which allow satisfactory — namely accurate, precise and timely — analysis of coal for ash, moisture and calorific value through to a full elemental composition of coal. The main requirement at this time is more and fuller implementation of the available systems in the coal fuel cycle — namely, mining, transportation, beneficiation, combustion and disposal; more data on system performance and its beneficial use are highly desirable. However, further improvement of some technical aspects of the techniques as described below is definitely desirable. This could lead to an even more efficient use of the technology for better utilization of our fossil energy resources.

3.1. Gamma densitometry

The most widely used nucleonic device within the beneficiation plant is the density gauge. When installed and maintained correctly this device has proved valuable for optimization of control of such processes as flotation. By use of new techniques which make fuller use of the information available from this device, wider applications are now foreseen.

3.2. Mining control

The control of cutting horizons by the detection of natural radioactivity in the seams has proved beneficial in some countries and is now being more widely deployed. This is not possible in circumstances where the level of natural radioactivity is low and further development of an alternative technique is required. Gamma backscatter sensors have been developed but they are generally less successful than natural gamma-based systems. It would be useful to obtain more information on the extent and success of application of such techniques.

3.3. Nuclear belt weighers

Common nuclear weighers are often not as accurate (typically 1–2.5% relative) in field applications as conventional electromechanical devices. However, the latter are high-maintenance items. Work is required to improve accuracy to
the level of better than 0.5%, with the goal of 0.1%, through design improvements and compensation for compositional effects (mainly H) and material profile.

3.4 Ash

Considerable research and development has been carried out on nuclear methods for the determination of ash in coal. For solid coal there are now several commercially available instruments with satisfactory performance. It should be noted however that ash meters using X-rays or gamma rays cannot usually be calibrated for coals having a very wide range of composition.

Two nuclear techniques are currently in routine industrial use for direct determination of the ash content of coal on conveyors. The first depends on the combination of low- and high-energy gamma-ray transmission through the coal. The second depends on backscatter of low-energy gamma rays from the coal. The accuracy of ash determination is about 0.3 wt% for product coal in the range 6 to 14 wt%, and usually 1 to 1.5 wt% for run-of-the-mine coals of ash in the range 20 to 35 wt%. This accuracy is sufficient for some but not all applications of high-ash coal.

Although, in general, direct on-line analysis is strongly recommended, accuracy in ash determination is not always sufficient for the application because of excessive variations in the coal mineral composition. In this case it can be necessary to resort to measurement of a part of the main coal stream on a sample by-line. Two techniques are available which are less sensitive to variations in coal mineral composition: (a) The low-energy X-ray backscatter technique with compensation for variations of iron in the ash. With this technique mass flow on the by-line is limited to one to two tonnes per hour and particle size must be less than 5 mm. (b) The pair production technique, based on detecting positron annihilation gamma rays, which can cope with flow rates of up to 30 tonnes per hour. As well, the more complex prompt gamma ray neutron activation analysis techniques can be used to determine ash on a sample by-line.

It is felt that generally sufficient research has been carried out on these techniques for solid coals although further development is required to obtain better accuracy, particularly for high-ash coals, and on specific aspects such as the effect of hydrogen on scattering, residual compositional effects and non-linearity effects in the various gauges. It would be of significant value to potential users of these nuclear techniques to prepare a report giving a detailed comparison of the techniques and their limitations with respect to:

- sampling volume
- on-belt or by-line use
- sensitivity to compositional changes
- calibration
- analysis times and field performance record.
Ash content of coal in slurries is also an important parameter for control of flotation circuits in coal preparation plants. Nuclear techniques are being developed for this purpose but none have yet been fully tested or proved in industrial conditions and a high priority should be given to further research and development in this area. The techniques being developed include:

(a) combination of neutron moderation, gamma-ray transmission and X-ray backscatter, which makes the determinations independent of the variable voidage of the slurry;
(b) combination of X-ray backscatter and gamma-ray transmission which can be applied to the flotation tailings stream where variable voidage is less of a problem;
(c) combination of high- and low-energy gamma-ray transmission in highly pressurized sample by-lines having voidage reduced to negligible amounts.

3.5. Moisture

At present the most favourable technique for determining moisture in coal is the non-nuclear technique based on microwave attenuation, sometimes combined with phase measurement. Moisture determination is also possible using radio-frequency capacitance measurements and by neutron transmission measurement. For moisture measurement the nuclear method has the disadvantage of responding to total hydrogen content (including hydrocarbons). However, where neutron methods are already in use for elemental analysis in coal, and providing coal-fixed hydrogen content does not vary widely, it may also be possible to determine simultaneously moisture content in the coal by the neutron technique. Further development of both nuclear and non-nuclear methods should be encouraged.

3.6. Calorific value

Three methods for the estimation of calorific value of coal, in order of increasing complexity and cost, are:

(a) Measurement of moisture and ash content and assuming that the material left after removing ash and moisture has constant calorific value. This method is very useful but is usually applicable to a limited range of coal types because of this assumption. The measurement errors of moisture and ash should be consistent with the errors introduced by this assumption. By using gamma rays to measure ash and a non-nuclear technique for moisture this method is the most simple and inexpensive, and is therefore preferred unless neutron techniques are already in use for other reasons.
(b) Measurement of carbon content using neutron inelastic scattering. The accuracy of this method depends on the degree of correlation between
total carbon content and calorific value. The relative errors of carbon content measurement need to be lower than the desired precision in calorific value.

(c) Carbon, hydrogen and moisture determination. This method involves fewer assumptions but is more complex than the preceding methods. It involves carbon measurement by neutron capture, and measurement of hydrogen in coal by the difference in total hydrogen and moisture (by microwave).

Further research, development and field testing is desired particularly to assess the simpler methods above.

3.7. Elemental analyses

The development of neutron interaction techniques during the last few years now presents the opportunity of measuring the concentrations of all the principal elements in coal. Although further development is still required to meet all potential applications, equipment based on neutron techniques is now being introduced on-line. Measurement of all the principal elements leads to a technique for overcoming perturbations in neutron transport. It also allows determination of gross parameters of coal, such as ash content and specific energy, as well as the identification of the concentration of such elements as sulphur and nitrogen which are important for pollution control. On-line determination of S, N, Fe, Cl, and ash is available using a system incorporating a medium-resolution (NaI) detector and using some kind of spectral unfolding to achieve reasonable coal independence. All these determinations require great electronic stability.

Equipment designed to derive a total elemental analysis is essentially complex and includes much state-of-the-art technology. For this reason it is expensive and its main potential is therefore seen to be in combustion control where very substantial savings can be made. The most accurate method for the quantitative determination of all elements is prompt gamma-neutron activation analysis (PGNAA) based on the germanium detector because of its high-energy resolution as compared to the NaI detector. The advent of the N-type hyper-pure Ge detector has considerably alleviated the problem of fast neutron radiation damage. This has allowed the attainment of much higher signal count rate with reasonable service life before the detector is sent for annealing of radiation damage — estimated today to be about 4 to 6 months for 24 h/day operation for one commercial system. The current commercial full coal elemental analyser is achieving accuracies equal to or exceeding those of the conventional standards set by ASTM, ISO or similar organizations, for a wide variety of coals from all over the world. This accuracy is being achieved in measurement times of 20 minutes using the Ge detector and in a few minutes when a NaI detector is used in parallel, compared with the one or two days required by the conventional methods.

A continuous exchange of information is required in this developing area and the IAEA is strongly recommended to encourage research and information exchange on this topic, especially in mathematical methods of data analysis, spectral
analysis and correcting for second order neutron-gamma transport effects in coal. It is also desirable to develop stable absolute standards for calibration of PGNAA on-line systems. Further development is also required for direct on-line measurements of oxygen content, if possible within the framework of existing thermal capture PGNAA systems.

3.8. Other parameters

Coal users are often interested in other properties of the coal such as coking properties and volatile matter. The quality of coal for coke-making depends primarily on the petrology of the coal substance and other physical properties which are presently measured by non-nuclear laboratory techniques. There is potential for the use of such data on-line at coking plants but the current measurement techniques cannot be applied. Volatile matter is an important parameter which can affect the safe operation of boilers. Again no conventional on-line technique exists. There are indications of correlations between these properties and elemental composition (particularly carbon, hydrogen and oxygen) but data so far are scarce. Work on the development of these correlations needs to be expanded before an assessment can be made of the potential for the on-line determination of these properties via nuclear techniques of elemental analysis.

3.9. Calibration and standardization

While the nuclear methods of analysis of coal have potential accuracies equal to or better than the traditional standard methods based on chemical assay of individual samples, the actual accuracy achieved will depend on the reliability of the calibration. Calibration standards have proved difficult to produce with any accuracy because of the high heterogeneity of coal. For similar reasons it is difficult to calibrate an on-line nuclear instrument against analysed samples from the actual process line because in general the sampling error will be much larger than the analytical error of either technique. Perhaps synthetic standards will be required. Nonetheless routine calibration of nuclear instruments to compensate for excessive drift can be accomplished by use of built-in relative standards. This is usually the main requirement for process control purposes. One on-line ash meter already has this facility and this approach is urged on other manufacturers. Stable, absolute standards are required for PGNAA on-line systems.

Because of the potential advantages in avoiding sampling errors, there is scope for applying nuclear instrumentation for reference techniques for commercial arrangements between the coal producers and users. To achieve this objective it is
necessary to have the nuclear methods accepted as standard methods and for this purpose it will be essential to address the problems of absolute standardization and of compensating for coal composition dependence.

4. BOREHOLE LOGGING FOR COAL

4.1. Current use

Three nuclear techniques are in common usage today in borehole logging for coal. These are:

(a) natural gamma ray logging
(b) gamma-gamma (density) logging
(c) neutron-neutron (porosity) logging.

These logs are for identification of coal seams, delineation of the seam boundaries, and for determination of ash content as well as for lithological correlation from hole to hole. It is suggested that the following improvements to these techniques would greatly increase their utility:

(a) Natural gamma ray logs. The present gamma-ray tools are capable of measuring only the total natural radioactivity of rocks and hence their use is limited to the identification of coal and providing information on seam thickness. A modification of this technique could provide additional information. The use of three single-channel analysers for energy discrimination makes it possible to discriminate between the natural sources of radioactivity, potassium, uranium and thorium. This spectral gamma-ray information used in the form of radioelement ratios can provide characteristic signatures of the rocks adjacent to the coal seams, permitting positive correlation from hole to hole of the lithology encountered in complex coal deposits. Also, by utilizing scintillation detectors with higher spectral efficiency than sodium iodide, the logging speed for gamma ray logging can be significantly increased.

(b) Gamma-gamma density logs. The present density logging tools and techniques give adequate information regarding variation in bulk density of rocks, and in most cases coal bed boundary definition (bed resolution). However, coal ash calculations based on density/ash relationships are subject to error which could be eliminated by the addition of spectral logging capability to the gamma-gamma tool. By recording the ratios of two energy regions of the gamma-ray spectrum the effective atomic number of the rocks can be determined and hence the ash content can be accurately computed.

It is recognized that the necessary hardware and methods to carry out the above suggested improvements already exist. However, to make them more widely
accepted and to stimulate their usage a number of demonstrations of the value of
the techniques must be conducted. There is a need for a body such as the IAEA
to organize such demonstrations in conjunction with the approximately ten
organizations currently having the technical capability to be involved.

(c) **Calibration facilities.** It is recognized that improvements in quantitative
logging measurements are only possible with proper calibration facilities. Such
facilities need to be established in coalmining areas for the use of logging
service companies. To facilitate this it is necessary to compile a manual on the
characteristics of borehole calibration models for a number of physical properties,
and to include expert recommendations for construction of such models. This is
an appropriate activity for the IAEA to sponsor.

4.2. The state of the art

The use of scintillation detectors and of solid-state detectors in performing
elemental analysis of coal in a borehole has already been demonstrated. The
scintillation detector technology is ready for transfer to the logging industry.
After acceptance of the scintillation detector technology, the solid-state detector
technology, which can provide even more information but at the cost of higher
complexity, will be ready for transfer.

Of the available neutron sources this group recognized the superiority of
californium-252 sources whenever the higher neutron energy of americium-
beryllium sources is not required, and it is recommended that the latter be
replaced by $^{252}$Cf wherever possible, for example in the commonly used neutron-
neutron logging tools. A wider acceptance and use of neutron generators is also
foreseen when more rugged and reliable hardware is developed. Pulsed neutron
techniques will be introduced in gamma-ray spectrometry and the measurement
of carbon and oxygen in coal will be facilitated by the use of 14 MeV neutrons
from the neutron generators.

5. IMPLICATIONS FOR INTERNATIONAL CO-OPERATION

(a) **Information exchange.** Although the more widespread utilization of nuclear
techniques would have advantages for the efficient utilization of coal, there has
been a general reluctance to implement the new technology due to generally
inadequate recognition of the available instrumentation and its range of possible
application. The development of the techniques themselves has also been somewhat
hindered by lack of knowledge of the range of variability of coals and of the
precise needs of coal users. The type of information which could be assembled
by international co-operation includes:

- elemental composition, ash content and calorific value, and the variability
  of these properties, for a wide range of coal types: this information would
SUMMARY AND CONCLUSIONS

help to establish correlations to simplify analytical procedures and improve accuracy of nuclear methods by reducing compositional dependence; (some information of this type is collected by the World Coal Resources and Reserves Data Bank Service in Washington D.C., USA);

- surveys of commercially available equipment with advantages and limitations assessed;
- technology updates as provided by meetings such as the current one in which dialogue between researchers, coal producers, coal users and instrument manufacturers is encouraged and the results publicized.

(b) **Technology transfer.** The development of equipment for borehole logging for ash and for direct on-line measurement of ash is now well established and can be used profitably in those countries which are working to establish or to exploit coal reserves. The best route to technology transfer is believed to be by initial training at a central laboratory which has the required expertise, followed by ‘hands-on’ experience in field trials or demonstration plants in the appropriate country under the guidance of experts from the technology holder.

(c) **Research.** There is a need for further international collaboration in research aimed at refining and extending these techniques in the directions indicated in the preceding section, and widely to disseminate the results of this work.

6. **CONCLUSIONS**

The time has come for nuclear techniques to assume a very widespread and valuable role in optimizing the exploitation of coal around the world. Nuclear techniques should play a prominent role in all aspects of the coal industry, from exploration through mine development to testing of coal quality during production. The advent of on-line nuclear techniques allows continuous, near real time appraisal of coal quality free from the errors inherent in conventional techniques based on discrete samples. This means that for the first time the plant operator can receive information on coal quality before it is processed or consumed in his plant.

The prospect of coal management systems which allow the utilization of coal more efficiently is most attractive. This can now be achieved from the coal beneficiation plant to the power plant or steel works, with savings in energy and the prospect for better control of emissions. This exciting new technology is recommended to all with an interest in the coal industry.
LIST OF PARTICIPANTS

Bull, P.  
(Scientific Secretary)  
Division of Research and Laboratories,  
International Atomic Energy Agency,  
P.O. Box 100, A-1400 Vienna, Austria

Charbucinski, J.  
Division of Mineral Physics, CSIRO,  
P.O. Box 124, Port Melbourne, Victoria 3207, Australia

Cierpizs, S.  
EMAG,  
Ul. A. Czerwonej 83a, Katowice, Poland

Clayton, C.G.  
(Chairman)  
Nuclear Geophysics Group,  
Building 7.21, AERE Harwell, Oxfordshire OX11 0RA,  
United Kingdom

Fauth, G.  
Bergbau-Forschung GmbH,  
Postfach 130 140, D-4300 Essen 13,  
Federal Republic of Germany

Gozani, T.  
Science Applications International Corporation  
1257 Tasman Drive, Sunnyvale, CA 94089,  
United States of America

Guizerix, J.  
Division of Research and Laboratories,  
International Atomic Energy Agency,  
P.O. Box 100, A-1400 Vienna, Austria

Killeen, P.G.  
Geological Survey of Canada,  
601 Booth Street, Ottawa, Ontario K1A 0E8, Canada

Leininger, D.  
Bergbau-Forschung GmbH,  
Postfach 130 140, D-4300 Essen 13,  
Federal Republic of Germany

Łoskiewicz, J.  
Institute of Nuclear Physics,  
Ul. Radzikowskiego 152, 31–342 Cracow, Poland

Mikesell, J.L.  
US Geological Survey M.S.—990,  
Reston, Virginia, VA 22092, United States of America

Page, D.  
National Coal Board,  
Coal House, Lyon Road, Harrow, Middlesex,  
United Kingdom

215
<table>
<thead>
<tr>
<th>Name</th>
<th>Institution</th>
</tr>
</thead>
<tbody>
<tr>
<td>Prasad, A.S.</td>
<td>TISCO, Jamshedpur 831007, India</td>
</tr>
<tr>
<td>Sowerby, B.D.</td>
<td>Division of Mineral Physics, CSIRO, Lucas Heights, Private Mail Bag 7, Sutherland NSW 2232, Australia</td>
</tr>
<tr>
<td>Surman, P.L.</td>
<td>Central Electricity Generating Board, Scientific Services Department (North East), Beckwithe Knowle, Otley Road, Harrogate HG 1PR, United Kingdom</td>
</tr>
<tr>
<td>Van den Berg, E.A.</td>
<td>Shell Research B.V., P.O. Box 3003, NL-1003 AA Amsterdam, Netherlands</td>
</tr>
<tr>
<td>Vovk, I.F.</td>
<td>Division of Research and Laboratories, International Atomic Energy Agency, P.O. Box 100, A-1400 Vienna, Austria</td>
</tr>
<tr>
<td>Watt, J.S.</td>
<td>Division of Mineral Physics, CSIRO, Lucas Heights, Private Mail Bag 7, Sutherland NSW 2232, Australia</td>
</tr>
</tbody>
</table>
It would greatly assist the International Atomic Energy Agency in its current review of its publications programme if you could kindly fill in one of the attached postcards and return it to the address shown. Your co-operation is greatly appreciated.

| 1. Title of book: | ............................................................................................................ |
| 2. Did you purchase the book? [ ] Did you borrow it from a library? [ ] |
| 3. By what means did you learn of its existence? A book notice [ ]; a book review [ ]; the IAEA publications catalogue [ ]; IAEA meetings [ ]; IAEA newsletters [ ]; a professional colleague [ ]; scientific literature [ ]; other means (please specify) [ ]: ............................................................................................................ |
| 4. How do you rate the usefulness of the content? Very useful, not found elsewhere [ ]; useful as a survey [ ]; useful for reference [ ]; useful because of its international character [ ]; useful for training or study purposes [ ]; not very useful [ ]. |
| 5. How do you normally purchase IAEA publications? Through booksellers [ ]; through direct purchase [ ]; through your national Atomic Energy Commission or similar body [ ]. |
| 6. Would you like to have a free subscription to the IAEA publications catalogue? Yes [ ] No [ ] |
International Atomic Energy Agency
Sales and Promotion Unit
P.O. Box 100
Wagramerstrasse 5
A-1400 Vienna
Austria
It would greatly assist the International Atomic Energy Agency in its current review of its publications programme if you could kindly fill in one of the attached postcards and return it to the address shown. Your co-operation is greatly appreciated.

1. Title of book: ........................................................................................................................................................................

2. Did you purchase the book? [ ]
   Did you borrow it from a library? [ ]

3. By what means did you learn of its existence?
   A book notice [ ]; a book review [ ]; the IAEA publications catalogue [ ];
   IAEA meetings [ ]; IAEA newsletters [ ]; a professional colleague [ ]; scientific
   literature [ ]; other means (please specify) [ ]: ...........................................................

4. How do you rate the usefulness of the content?
   Very useful, not found elsewhere [ ]; useful as a survey [ ]; useful for reference [ ];
   useful because of its international character [ ]; useful for training or study
   purposes [ ]; not very useful [ ].

5. How do you normally purchase IAEA publications?
   Through booksellers [ ]; through direct purchase [ ]; through your national
   Atomic Energy Commission or similar body [ ].

6. Would you like to have a free subscription to the IAEA publications catalogue?
   Yes [ ] No [ ]
An exclusive sales agent for IAEA publications, to whom all orders and inquiries should be addressed, has been appointed in the following country:

UNITED STATES OF AMERICA  UNIPUB, P.O. Box 433, Murray Hill Station, New York, NY 10157

In the following countries IAEA publications may be purchased from the sales agents or booksellers listed or through your major local booksellers. Payment can be made in local currency or with UNESCO coupons.

ARGENTINA  Comisión Nacional de Energía Atómica, Avenida del Libertador 8250, RA-1429 Buenos Aires

AUSTRALIA  Hunter Publications, 58 A Gipps Street, Collingwood, Victoria 3066

BELGIUM  Service Courrier UNESCO, 202, Avenue du Roi, B-1060 Brussels

CHILE  Comisión Chilena de Energía Nuclear, Venta de Publicaciones, Amunategui 95, Casilla 188-O, Santiago

CHINA  IAEA Publications in Chinese: China Nuclear Energy Industry Corporation, Translation Section, P.O. Box 2103, Beijing

CZECHOSLOVAKIA  S.N.T.L., Mikulandska 4, CS-116 86 Prague 1

FRANCE  Office International de Documentation et Librairie, 48, rue Gay-Lussac, F-75240 Paris Cedex 05

HUNGARY  Kultura, Hungarian Foreign Trading Company, P.O. Box 149, H-1389 Budapest 62

INDIA  Oxford Book and Stationery Co., 17, Park Street, Calcutta 700 016

ISRAEL  Helliger and Co., Ltd, Scientific and Medical Books, 3, Nathan Strauss Street, Jerusalem 94227

ITALY  Libreria Scientifica, Dott. Lucio di Biasio "seiou", Via Meravigli 16, I-20123 Milan

NETHERLANDS  Martinus Nijhoff B.V., Booksellers, Lange Voorhout 9-11, P.O. Box 269, NL-2500 The Hague

PAKISTAN  Mirza Book Agency, 65, Shahrah Quaid-e-Azam, P.O. Box 729, Lahore 3

POLAND  Ars Polona-Ruch, Centra!a Handlu Zagranicznego, Krakowskie Przedmiescie 7, PL-00-068 Warsaw

ROMANIA  Illexim, P.O. Box 136-137, Bucharest

SOUTH AFRICA  Van Schaik Bookstore (Pty) Ltd, P.O. Box 724, Pretoria 0001

SPAIN  Díaz de Santos, Lagasca 95, E-28006 Madrid

SWEDEN  AB Fritzes Kunsl, Hovbokhandel, Fredsgatan 2, P.O. Box 16356, S-103 27 Stockholm

UNITED KINGDOM  Her Majesty's Stationery Office, Publications Centre, Agency Section, 51 Nine Elms Lane, London SW8 5DR

USSR  Mezhdunarodnaya Kniga, Smolenskaya-Sennaya 32-34, Moscow G-200

YUGOSLAVIA  Jugoslovenska Knjiga, Terasije 27, P.O. Box 36, YU-11001 Belgrade

Orders from countries where sales agents have not yet been appointed and requests for information should be addressed directly to:

Division of Publications
International Atomic Energy Agency
Wagramerstrasse 5, P.O. Box 100, A-1400 Vienna, Austria