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**TECHNOLOGY TRANSFER OF NUCLEAR TECHNIQUES
AND NUCLEONIC CONTROL SYSTEMS
IN THE MINERAL INDUSTRY**

**REPORT OF AN ADVISORY GROUP MEETING
ORGANIZED BY THE
INTERNATIONAL ATOMIC ENERGY AGENCY
AND HELD IN BOMBAY, 15-19 JANUARY 1990**



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FOREWORD

Among the many beneficial applications of radiation and radioisotopes in industry which are now well established in advanced countries, the applications of nuclear techniques and nucleonic control systems in the mineral industry have great potential for developing Member States. The use of nucleonic on-stream analyzers in the coal industry has resulted in enormous technical and economic benefits in addition to minimization of environmental pollution. Large savings have also resulted from the use of such analyzers in the processing of other minerals. Nuclear borehole logging techniques have demonstrated great potential in oil and gas evaluation. Radiotracer investigations have led to process optimisation and trouble shooting in various stages in ore processing and metallurgy. Though the technical and economic benefits of applications of nuclear techniques in the mineral industry are well recognised, technology transfer in these areas has been hampered by a variety of factors.

In order to review the status and trends in nuclear techniques and nucleonic control systems in the mineral industry and the problems and considerations in their technology transfer to developing Member States, the IAEA convened an Advisory Group Meeting in Bombay, India, 15-19 January 1990. The present publication is based on the contributions presented at this meeting. It is expected that this publication will provide useful information to all those who are considering the introduction and applications of nuclear techniques and nucleonic controls in the mineral industry.

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1. INTRODUCTION

Background

Enormous technical and economic benefits have been conferred on the mineral industry in many advanced countries by the applications of nuclear techniques and nucleonic control systems. The major techniques which have been developed to a stage of industrial scale application include nucleonic control systems, on- and off-line nuclear analytical procedures, nuclear borehole/well logging and tracer techniques applied to the exploration and exploitation of oil, gas, coal, geothermal energy sources as well as metallic and non-metallic minerals. Nuclear techniques are important because of their enormous potential to improve the efficiency of raw materials exploration, ore extraction and processing, to effect savings in energy and materials in these processes. Their advantages include rapidity, high specificity, adaptability to multi-parameter analysis and control, relative simplicity for use, and in some cases possibility of application in hot, dusty and aggressive environments where no other instruments or methods can be used. Nucleonic controls and nuclear measurements can be made by non-contact processes through the walls of vessels. The radioisotope tracer technique is a unique tool for process optimization and provides quantitative data on distribution and flow of materials and residence time.

The last few years have witnessed many important advances in the development and applications of nucleonic control systems and on-stream analyzers in the coal industry and every year many tens of millions of tons of coal are evaluated for ash, sulphur and moisture content. The impact of these developments in environmental protection is considerable and is expected to increase with further developments in applications of nuclear technology to the coal industry. On-line analysis systems are also widely used in processing of other minerals, resulting in large savings through increased yields and reduction in wasted energy. Nuclear borehole logging techniques are extensively used for oil and gas evaluation, and to a lesser extent for evaluation of coal and iron-ore. Radiotracer techniques have been used in several countries to study ground water flow characteristics in mine areas, evaluate ventilation exchange rate in operating mines and for investigations of operational characteristics of ore processing plants.

Purpose

The purpose of this report is

- (i) to discuss the state-of-the-art and trends in radioisotope on-line analysis in the mineral and coal industry, nuclear well logging in the oil industry, and radiotracer investigations in mining and mineral processing operations,
- (ii) to evaluate the role and scope for these technologies in the mineral industry in developing member states, and
- (iii) to identify the problems and considerations in the transfer of these technologies to developing member states with a strong base in mineral industries.

2. ON-LINE ANALYSIS

Mineral and coal processing operations can be controlled more economically if materials in various process streams can be rapidly analysed on-line. The on-line analysis information provided by these systems is now widely used to improve (manual) control of mineral and coal processing operations, resulting in large savings through increased yield of product reduction in energy usage, and control and minimisation of pollutants. The trend now is to develop automatic control techniques based on the on-line analysis information, resulting in further savings.

The on-line analysis systems with proven techno-economic benefits and considered to be suitable now, or in the near future, for technology transfer to developing countries, are:

2.1. Valuable metal content of mineral slurries

The techniques used depend on radioisotope X-ray fluorescence and preferential absorption techniques. About 55 plant systems, with an average of five streams analysed per plant, are in routine use world wide in mineral concentrators. The recovery of valuable minerals is usually increased by 0.5-3%, with savings of US\$100000 to several million dollars per year depending on the size of the concentrator. The mineral industry world-wide is

well aware that recovery of valuable minerals is increased after on-stream analysis systems are installed in mineral concentrators. The trend is now for mineral companies to contract for a combined on-stream analysis and control package. The Agency's technical document IAEA-TECDOC-520 "Technical and Economic Benefits of Nuclear Techniques in Ore Processing" has summarised the current status of the technology. In his paper Ishikawa has described the benefits of application of a depth type density gauge in iron-ore treatment and Murthy et al, (in their papers) have outlined the development/applications of level control systems, radiometric density gauges and XRF systems in India.

2.2. Ash content of coal on conveyors

Coal consists of coal matter (mainly carbon with some hydrogen, oxygen and nitrogen) and mineral matter (mainly Al, and other silicates with some iron). Coal ash is the oxidised incombustible residue from the combustion of coal and is closely correlated with the mineral matter content. 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. The intrinsic heterogeneity of coal deposits and the variation in the quality of mineral 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. Continuous determination of the ash content of coal on conveyors and in slurries is required for the better control of coal washeries.

The economic benefit gained from the use of on-line ash monitors in most cases results from control of mining and plant operations closer to the required specification ash content. Further savings from use of ash monitors is likely to result from the linking of the ash monitor to a system for automatic control of the heavy medium baths and cyclones in coal preparation plants.

For continuous monitoring on conveyors the preferred technique is dual energy gamma-ray transmission. At least 80 installations of on-line ash monitors are in routine use, monitoring ash at various stages of mixing, blending, preparation and utilisation. The use of these ash monitors in Australia has increased yield of constant ash by 1-3%, the greater constancy of ash content has led to an estimated total savings of US\$125 million over a five year period.

This technology is currently being demonstrated and transferred to Asian/Pacific countries under the UNDP Industrial Project.

2.3. Solids and ash content of coal in slurries

In coal preparation plants, the fine coal fraction is usually physically beneficiated by froth flotation. Continuous measurement of both solids weight fraction and ash content of coal in slurries is required for optimisation of the flotation circuits.

The preferred technique combines neutron moderation, gamma-ray transmission, X-ray scatter and X-ray fluorescence. In Australia, there are five installations of the coal slurry analysis, with an average of three streams analysed per plant. The technology is very similar to that described in 2.1. above. Plant studies have shown that control of the flotation bank of coal preparation plants, based on the on-line analysis information, increases the yield of coal at constant ash by 10%. For a 3 million tonne per year plant, with 15% of the product coming from flotation, the increased value of product is nearly US\$3 million per year.

This technology is considered to be very suitable for transfer to developing countries which use coal flotation in coal preparation operations.

2.4. Moisture in coal

The moisture content in coal is required at various stages of coal blending, preparation and utilisation. The preferred technique is microwave attenuation and phase shift combined with gamma-ray transmission. There are now a few applications of the microwave and gamma-ray transmission technique in routine use. The applications are expected to expand rapidly in the next few years.

This technology is suitable for transfer to developing countries mining coal and using coal fired power stations.

2.5. Moisture in coke

The moisture content of coke is required so that the coke/iron ore concentrations in blast furnaces is accurately known. The preferred technique is fast neutron and gamma-ray transmission. It can be applied either directly on-line on conveyors, or to coke in hoppers. A conservative estimate of savings per year per blast furnace of US\$600000 has been reported from Japan.

This technology is suitable for transfer to developing countries which produce steel using blast furnaces.

In his paper Watt has described the status of radioisotope on-line analysis in the Australian mineral and coal industries highlighting the applications, benefits and relevant technology transfer aspects.

3. BOREHOLE LOGGING

Nuclear borehole logging techniques are a mature development, particularly for oil and gas evaluation, the major application being in the measurement of subsurface formation properties for discovering and evaluating hydrocarbons and to understand the formation geology for improved production and management of the reserves. The main interest has been the location of porous zones containing hydrocarbons and free of clay minerals whose presence would inhibit fluid flow. For coal evaluation the most important parameter to evaluate is ash content. Gamma-gamma techniques to determine average atomic number are the most appropriate techniques to use in air filled holes, and neutron capture gamma-ray measurements in water-filled holes. For iron ore evaluation, which is usually in air-filled holes, the density and average atomic number measurement is used. In addition, natural activity measurements are sometimes also included in these applications to help define the three dimensional extent of the ore body by comparing measurements from many drill holes. The more severe conditions encountered in oil and gas exploration, coupled with the wider availability of these measurements, lead to the conclusion that the most useful aspect of logging measurements in oil and gas exploration would be a firm understanding of the existing measurement techniques, and experience in analyzing data from these measurements.

From the earliest measurements of natural radioactivity generally associated with clay minerals in sedimentary formations, applications of nuclear borehole logging techniques have progressed to measurements of in-situ rock porosity, macroscopic thermal neutron absorption cross section and spectroscopic analysis of neutron capture gamma-rays.

The basic features of all nuclear logging measurements can be characterised by a gamma-ray or neutron source (except for natural activity measurements), shielding to minimize source particle transmission through the tool to the region of the detector, one or more detectors of neutrons or

gamma-rays, electronics to process the detector signals, and storage/transmission systems to buffer and transmit the data, through a cable, to surface instrumentation, typically today a computer in the logging truck. The most common sources of radiation are ^{137}Cs for gamma-gamma measurements, and AmBe, ^{252}Cf , or d,t accelerator sources for neutrons. Neutron detectors are usually presurized ^3He detectors, sometimes shielded with a thermal neutron absorber to detect only epithermal neutrons. Most gamma-ray detectors are NaI(Tl), and, occasionally, other scintillators such as CsI and BGO. Special high-temperature, ruggedized photomultipliers are used to collect the light from the scintillators and produce an electronic pulse proportional to the incident gamma-ray energy. Linear and digital electronics are included in the logging sonde to analyse the detector output, control its stability, and, when an accelerator is used, control its operation. For research purposes, a number of germanium semiconductor detector sondes have also been used.

In his paper Schweitzer has reviewed the current status of nuclear techniques in the oil industry. Recent developments in prompt gamma neutron activation analysis in borehole logging is covered in the Agency's technical document IAEA-TECDOC-537 entitled "Prompt gamma neutron activation analysis in borehole logging and industrial process control".

4. TRACER APPLICATIONS

Regular applications of tracer techniques have been made mainly in countries like Chile, Czechoslovakia, Finland, the German Democratic Republic, India and Poland and offer large potential for other countries with a strong base in mineral industries.

The following applications have been recognised as some of the more important ones:

- (a) Tracer techniques in mine hydrology to study ground water flow characteristics in mine areas to identify zones of potential hazards and evaluate the capacities of underground reservoirs.
- (b) Tracer techniques in borehole logging as a complement to other techniques established in nuclear geophysics. This is of particular importance to the oil exploration industry.

- (c) Ventilation exchange rate studies in operating mines.
- (d) Investigations of operational characteristics of ore processing plants such as residence time distribution, as well as trouble shooting and evaluation of such parameters as grinding characteristics of different types of mills.

In his paper Thyn has reviewed the state-of-the-art in radiotracer applications in the mineral industry and Murthy's paper has outlined the benefits of such applications in the Indian mineral industry.

5. ROLE AND SCOPE OF NUCLEAR TECHNIQUES IN THE MINERAL INDUSTRY IN DEVELOPING MEMBER STATES

Many developing Member States have recognised the need for modernisation, automation and expansion in their mineral exploration and exploitation programmes. This fact, with the increasing cost of raw materials and labour on the one hand and a wider appreciation of the inherent advantages of nuclear industrial on-line systems and techniques on the other, clearly indicates a strong need and good scope for these technologies in developing member states with a strong base in mineral industries.

The UNDP/IAEA/RCA Regional Industrial Project in Asia and the Pacific has helped to a great extent in increasing awareness of Nucleonic Control Systems in the Mineral Industry in this region. Through Training Courses and Seminars a number of personnel have been exposed to systems like the On-line Coal Ash Monitors and On-stream Mineral Analysers Used in the Industry. Under the project demonstration, programmes have been carried out both in Australia and in the Philippines. During the phase I of the project, an on-stream analyser was installed at the Philex Mining Corporation at Baguio, Philippines, for the analysis of process steams in copper concentrators. The Philex installation was used for conducting training courses and management seminars. In phase II, installation is now underway for coal ash monitoring system in Thailand. Executive Management Seminars, Workshops and Training Courses are planned under this programme.

The use of nuclear techniques in the Mineral Industry in many developing countries is expanding but is yet to pick up in a big way. There is a need for more On-stream Analysers which are known to improve the efficiencies of

concentrators through increased grade and recoveries. There is a large scope for nucleonic coal ash monitoring systems in the coal industry. Tracer investigations on different parameters such as residence time distribution, flow rate, flow paths, etc. should also be undertaken for optimisation of processes in the mineral processing plants. With the increased awareness of the role and scope of these techniques their introduction to the industry on a much wider scale in many developing countries is envisaged.

6. PROBLEMS AND CONSIDERATIONS IN TECHNOLOGY TRANSFER

Technology transfer of nuclear techniques to the mineral industry involves two aspects, namely (a) introduction of the technology and its regular applications in the mineral industry and (b) indigenous production of the systems and technique/services. While the former aspect is practicable and is actually under way in many developing member states, the latter aspect would be practicable only in countries with a very strong electronics and radiation/radioisotope technology base and also with a very large mineral industry. Efficient and continuing interaction between the vendor and the industrial user is essential to ensure that the technology is properly assimilated and any wrong apprehensions about the hazards of nuclear radiations are removed. The most important stages of technology transfer are customer education, plant demonstrations of the new technology leading to creation of sufficient confidence in the innovation, and establishment of the technoeconomic benefits, and finally actual installation and regular operation. Indigenous development and production of the systems and techniques require a strong base in electronics, instrumentation and radiation engineering and involve a well coordinated multi-stage programme of development, plant trials, licensing to a manufacturer and introduction to the industry. In this connection the experience of the Electronics Corporation of India and CSIRO, Australia could provide valuable guidelines.

Annex

PAPERS PRESENTED AT THE MEETING

RADIOISOTOPE ON-LINE ANALYSIS IN THE AUSTRALIAN MINERAL AND COAL INDUSTRIES: APPLICATIONS, BENEFITS AND TECHNOLOGY TRANSFER

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Abstract

The CSIRO at Lucas Heights has developed and commercialised nuclear gauges for on-line analysis in the mineral and coal industries. The most widely used are systems for the on-line determination of the ash content of coal, and for the in-stream analysis of mineral and coal slurries. More recent developments include systems for determination of the moisture content of coal, coke and other materials.

In January 1990, 96 Coalscan on-line ash gauges had been sold world-wide to a total value of US\$ 12 million. Independent consultants reported that better control of coal operations, based on the on-line ash information, resulted in a 1-3% increase in coal productivity in Australian plants, corresponding to a total savings to Australia of \$ 125 million over a five-year period. About 60 plant systems for on-stream analysis of mineral and coal slurries have been sold. The increase in recovery of metalliferous minerals is usually in the range of 0.5-3%, and for coal slurries, a 10% increase has been indicated in trials at two plants.

Technology transfer has been undertaken in Australia by plant trials, by joint development with licensees, and by licensees at the time of installation of commercial gauges. As part of the UNDP Industrial Project covering the Asia/Pacific region, Australia has undertaken training courses both in Australia and in the Philippines relating to on-stream analysis and control of mineral concentrators. This has resulted in on-stream analysis systems being installed in the regional countries, and considerable financial benefits from better control of concentrators. Australia recently commenced training courses in another sub-project in the UNDP Industrial Project relating to on-line determination of the ash content of coal and control of coal operations.

1. INTRODUCTION

Mineral and coal processing operations can be controlled more economically if materials in various process streams can be rapidly analysed on-line. The on-line analysis information provided by these systems is now widely used to improve (manual) control of mineral and coal processing operations, resulting in large savings through increased yield of product. The trend is now to develop automatic control techniques, resulting in further savings.

Developments in on-line analysis by CSIRO at Lucas Heights include systems for the determination of the ash content of coal on conveyors⁽¹⁻³⁾; the moisture content of coal^(4,5), coke⁽⁶⁾ and other materials on conveyors; the ash content and solids weight fraction of coal in slurries⁽⁷⁾; and the concentrations of metalliferous minerals in mineral slurries⁽⁸⁾. CSIRO at Port Melbourne has developed systems for the on-line determination of iron in iron ore and ash content of coal, and for the discrimination between coal and interseam sediments⁽⁹⁾.

This paper briefly describes some of the on-line analysis systems developed by CSIRO at Lucas Heights for the coal and mineral industries, and discusses the applications of these systems and economic benefits resulting from their use. Technology transfer, both to licensees and to the coal and mineral industries, is discussed.

2. ON-LINE ANALYSIS OF COAL AND COKE

On-line analysis on conveyors is required for the ash and moisture contents of coal, the specific energy of coal, and the moisture content of coke. On-stream analysis of coal slurries is required for the ash content and solids weight fraction.

2.1 Ash gauges

Two types of ash gauges are in widespread use in Australian coal operations: (a) dual energy gamma-ray transmission, in which measurements are made directly on coal on-line on conveyors; and (b) pair production, in which measurements are made on coal traversing high flow sample by-lines. Both are manufactured and marketed by Mineral Control Instrumentation Pty. Ltd. (MCI) of South Australia⁽¹⁰⁾. The former is called the Coalscan Model 3500 through-belt ash monitor, and the latter the Coalscan Model 4500 by-line ash monitor.

Dual energy gamma-ray transmission (DET)

The DET gauge^(1,2) depends on the determination of intensities of narrow beam transmission of low and high energy gamma-rays through coal on the conveyor (Figure 1). Both intensities depend on the mass per unit area of coal in the gamma-ray beam and the low energy also depends on the effective atomic number of the coal. The detected intensities are determined separately by pulse height analysis of output pulses from the scintillation detector, and then combined to give an ash content of coal which is independent of the thickness and vertical segregation of coal on the belt.

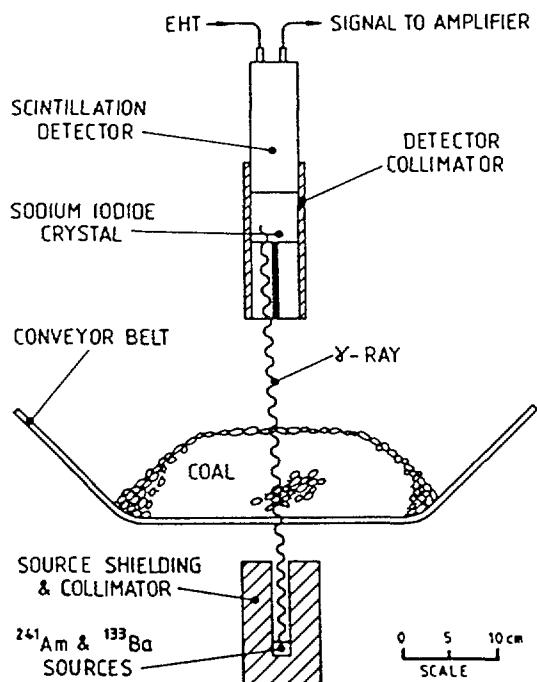


Fig. 1. Dual energy γ -ray transmission system for determination of ash in coal.

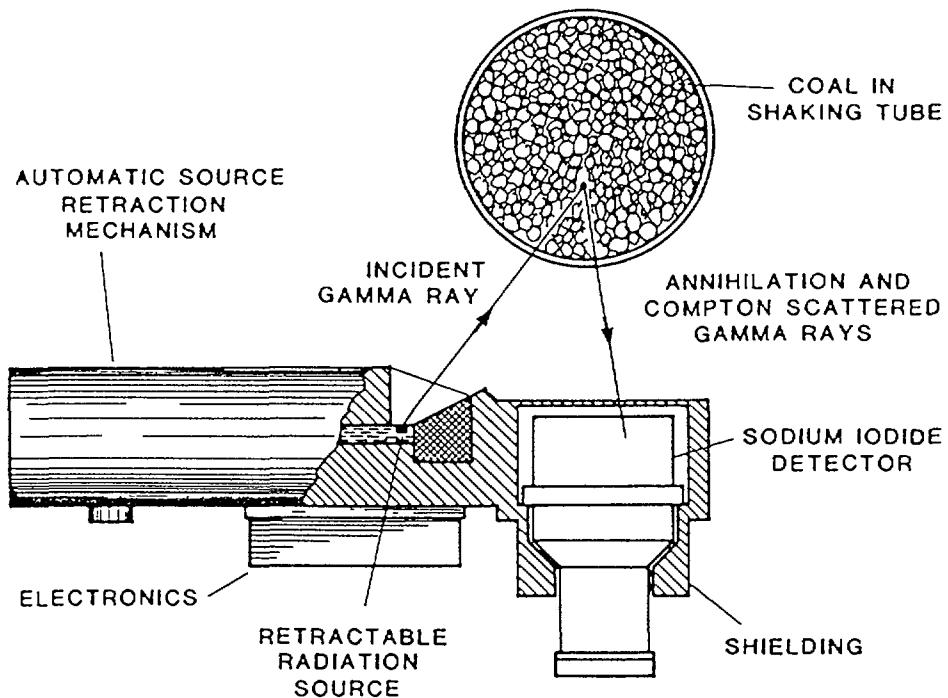


Fig. 2. Schematic diagram of the pair production gauge which determines the ash content of coal in a sample by-line.

The advantages of the Coalscan DET gauge are that coal is measured directly on the conveyor, it is more easy to install as no sampling system is required, it can handle very coarse coal, the measurement time can be made as short as a few seconds making possible rapid coal sorting operations, and its cost is only two thirds that of the Coalscan pair production gauge.

Pair production (PP)

The PP ash gauge^(1,3) depends on measurements of the intensities of gamma-rays backscattered by the coal and resulting from pair production and Compton interactions. Both interactions depend on the bulk density of the coal; the pair production also depends on the effective atomic number of the coal. The intensities of the backscattered gamma-rays resulting from the two interactions are separately determined by pulse height analysis of the output from the scintillation detector. The detected intensities are then combined to give the ash content. The main advantage of the PP gauge over the DET gauge is that it is less sensitive by about a factor of 4 to variations in ash composition.

The PP gauge views coal in a by-line stream taken from the main process line. Figure 2 shows schematically the PP gauge viewing coal passing through a cylindrical plastic tube in the by-line.

Applications of ash monitors and financial benefits

By January 1990, 96 Coalscan ash monitors had been sold world-wide (Table 1) of which 49 are in Australia, 27 in the USA and 20 in the rest of the world. The total value of these sales is about US\$ 12 million. The Coalscan Model 3500 is used in 80% of these installations.

TABLE 1. SUMMARY OF INSTALLATIONS OF COALSCAN DUAL-ENERGY GAMMA-RAY TRANSMISSION (DET) AND PAIR PRODUCTION (PP) ON-LINE ASH MONITORS (JANUARY 1990).

LOCATION	NUMBER OF INSTALLATIONS	
	DET	PP
Australia	34	15
USA	26	1
Other Countries	19	1
TOTAL	79	17

TABLE 2. SUMMARY OF APPLICATIONS OF COALSCAN DET AND PP ASH MONITORS.

APPLICATION	NUMBER OF INSTALLATIONS (JANUARY 1990)		
	AUSTRALIA	USA	REST OF WORLD
Mine grade control and raw coal monitoring	17	1	3
Preparation plant monitoring and control	24	10	5
Blending control (sorting, by-pass, stockpiles, loadout)	5	14	6
Power station feed monitoring	3	2	6
TOTAL	49	27	20

The information from Coalscan ash monitors is mainly used with manual control to improve coal mining, blending and preparation operations, as discussed in detail by Sowerby⁽¹¹⁾. Table 2 summarises the applications of Coalscan ash monitors in Australia, USA and the rest of the world.

The economic benefit gained from use of the Coalscan on-line ash monitors in most cases results from control of mining and plant operations closer to the required specification ash content. Sowerby⁽¹¹⁾ has detailed some case studies of economic benefits resulting from some Coalscan applications. CSIRO has had external consultants make an independent assessment of benefits resulting from use of Coalscan ash monitors in Australia. They found that use of the Coalscan monitors had led to a 1-3% increase in yield of coal at constant ash, the corresponding total benefits to Australia being US\$ 125 million over a five-year period⁽¹²⁾.

Further savings from use of Coalscan ash monitors is likely to result from the linking of the ash monitor to a system for automatic control of the heavy medium baths and cyclones in coal preparation plants. Romberg⁽¹³⁾ has developed advanced process control strategies which are being trialled at the MacQuarie Coal Pty. Ltd. plant near Teralba, Australia. This preparation plant produces both coking and steaming coal. The yield of the coking coal has been monitored over a total period of 110 hours, comprising 58 hours under automatic control and 52 hours under manual control. The average coking coal yield under automatic control was 43.2%, and under manual control, 38.2%. These preliminary results are most encouraging, but much further work is planned to prove the magnitude of the improvement, both at the this and other plants.

2.2 Coal slurries

In coal preparation plants, the fine coal fraction (<0.5 mm) is usually physically beneficiated by froth flotation. Continuous information of both solids weight fraction and ash content of coal in slurries is required for the better control of the flotation circuits.

The solids weight fraction W of coal in slurries is determined by combining measurements gamma-ray transmission, and neutron moderation and transmission. This determines the hydrogen (wt/wt) in the slurry which is correlated with W since water contains more hydrogen per unit weight than coal. The ash content of the slurry is determined by X-ray backscatter and iron K X-ray excitation measurements. The ash content of the coal in the slurry is determined by combining the ash content of the slurry with W.

The coal slurry analysis system⁽⁷⁾ is based on probes immersed directly into plant process streams or into high flow rate sample by-lines (Figure 3). Each contains a radioisotope source and scintillation detector. Three probes, and a temperature sensor, are required per stream.

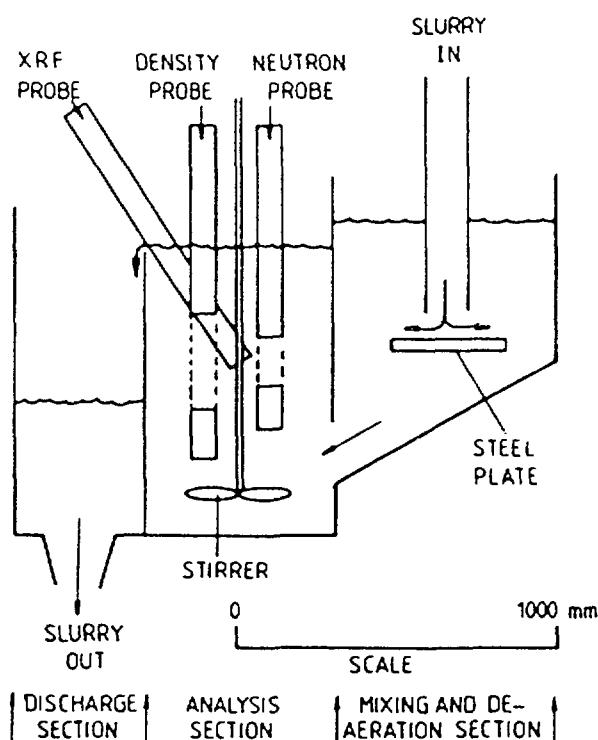


Fig. 3. Radioisotope immersion probes for analysis of coal slurries.

CSIRO has licensed the Australian Mineral Development Laboratories⁽¹⁴⁾ (Amdel) to manufacture and market this coal slurry analysis system. A three-stream system has been in routine use at the Oaky Creek Coal plant in the Bowen Basin, Queensland, since 1986^(7,15), and a five-stream system was installed at Curragh at the end of 1988. A system has recently been installed in Canada, and orders have been received from South Africa and Australia.

Two studies^(15,16) have indicated that the yield of the flotation circuits can be increased by 10% by introducing automatic control techniques based on information of on-line analysis. The savings which would result to a coal preparation plant can be calculated as follows. Assume that the plant produces 3 million tonnes of coal per year, 15% of which comes from the flotation bank, and assume that the product coal has a value of US \$ 40 per tonne. The increased value of coal is US\$ 1.8 million per year.

2.3 Moisture in coal and coke

CSIRO has developed capacitance, microwave transmission, and neutron transmission techniques for the on-line determination of moisture in coal and coke. For many on-belt moisture measurements the capacitance and microwave techniques are favoured. However the capacitance and microwave methods are not suitable for use on materials of high electrical conductivity (e.g. coke) or on many steel-cord conveyor belts. Neutron techniques are preferred in these applications.

Capacitance and gamma-ray backscatter

In this technique⁽⁴⁾, moisture is correlated with radiofrequency susceptance and conductance, determined using an under-belt capacitance sensor in which a fringing electric field interrogates a layer of coal on the conveyor belt directly above the sensor (Figure 4). Variation in the density and thickness of this layer is compensated for by using a matched under-belt gamma-ray backscatter gauge. The capacitance and gamma-ray backscatter gauges are matched so that each interrogates equivalent volumes of coal, independent of variations in thickness, density and moisture content.

A prototype gauge was trialled on the coking coal product conveyor at Stockton Borehole Colliery, Teralba, New South Wales, in September 1985 over a period of about two months. The on-line gauge determined moisture to within 0.5 wt% (1σ) over the range 7-14 wt% moisture.

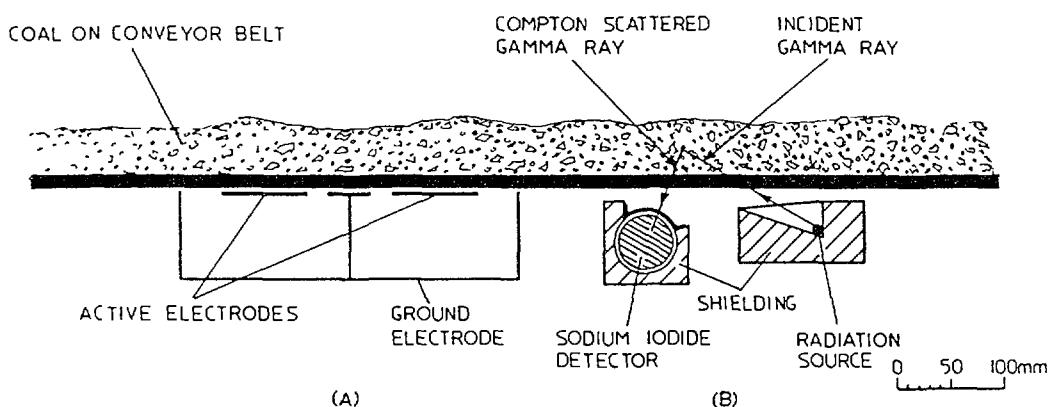


Fig. 4. Underbelt capacitance (A) and γ -ray backscatter density (B) gauges for on-conveyor determination of moisture in coal.

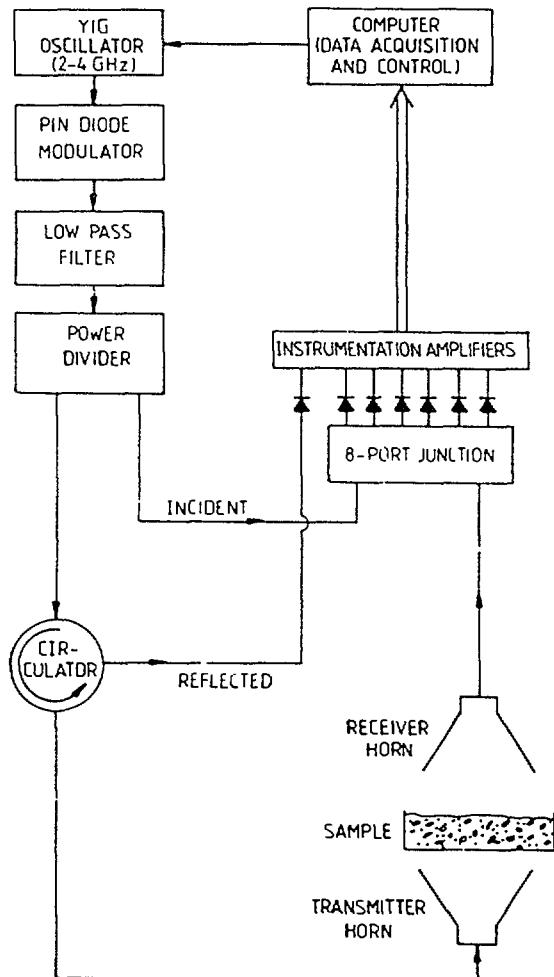


Fig. 5. Schematic of microwave transmission assembly used to determine moisture in samples.

MCI has been licensed to manufacture and market the capacitance gauges, and at January 1990 had installed 40 commercial units in industrial plants, of which approximately half were in the coal industry.

Microwave and gamma-ray transmission

Moisture measurement by microwave techniques⁽⁵⁾ is based either on the high real or imaginary part of the complex dielectric constant of water compared to the dielectric properties of the majority of dry materials. Microwave transmission techniques have the advantage of providing a bulk analysis that is not sensitive to the vertical segregation of moisture that may occur in material carried on conveyors.

A prototype microwave gauge (Figure 5) has been developed for on-belt measurement of moisture. The prototype gauge uses an yttrium-iron-garnet (YIG) oscillator that is tunable over the frequency range 2-4 GHz, and diode detectors to measure the reflected power and the power at the six output ports of the eight-port network. Transmission phase and attenuation are calculated from power measurements at the six output ports of the network and when normalised to sample mass per unit area are used to determine sample moisture content. Gamma-ray transmission was used to determine mass per unit area, giving more accurate result for moisture than use of microwaves alone.

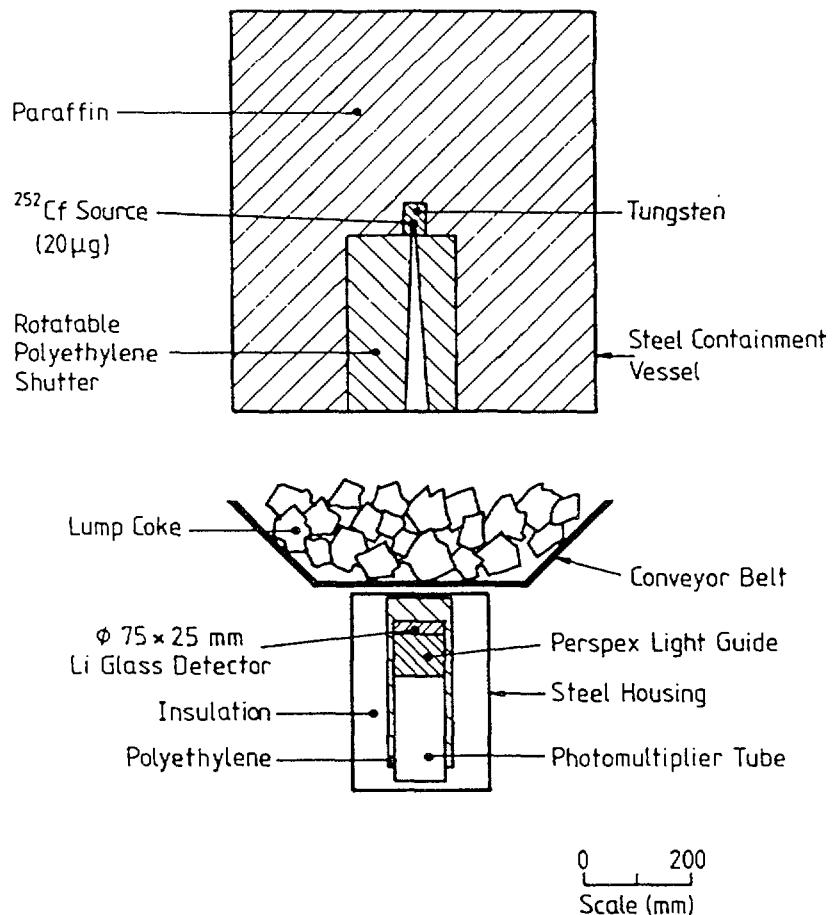


Fig. 6. Schematic of fast neutron and γ -ray transmission gauge for the determination of moisture in coke.

Laboratory measurements on a range of coals indicate that moisture can be determined to better than 0.37 wt% (1σ) over the range 0-15 wt% moisture using this gauge. An industrial prototype of the microwave moisture gauge has been trialled on the product conveyor of Howick Coal Washery, Singleton, Australia, over a three month period. Moisture in the coal was determined to 0.46 wt%.

In early 1990, licensees were being sought for this technology.

Fast neutron and gamma-ray transmission

The on-line determination of coke moisture is particularly important in the steel industry for controlling the net weights of dry coke and iron ore fed to blast furnaces. The potential benefits from improved control of the iron ore to coke ratio include stabilisation of blast furnace operation, a reduction in blast furnace hot metal temperature variability and a reduction in hot metal silicon content and its variation.

In the fast neutron and γ -ray transmission (FNGT) technique⁽⁶⁾, the transmission of fast neutrons depends predominantly on hydrogen concentration and mass per unit area, whereas γ -ray transmission depends on mass per unit area. Combining measurements of neutron and γ -ray transmission therefore permits hydrogen (and, in most cases, moisture) to be determined independently of mass per unit area.

A prototype FNGT gauge (Figure 6) for the conveyor belt determination of coke moisture has been field tested at BHP, Newcastle. The gauge has been operating on-line at BHP since June, 1988. Moisture was determined to about 0.4 wt% over the nine month period of the plant trial. MCI has been licenced to manufacture and market the FNGT moisture gauges.

3. METALLIFEROUS MINERALS

On-stream analysis of mineral slurries for valuable metal contents of the slurry solids is required for the better control of the flotation banks of mineral concentrators.

The on-stream analysis of metalliferous mineral slurries⁽⁸⁾ is achieved by use of several complementary techniques of radioisotope X-ray fluorescence and/or gamma-ray preferential absorption analysis, with the solids weight fraction being determined by gamma-ray absorption. The radioisotope sources and detectors are incorporated into probes which are immersed directly into the main plant streams or high flow rate sample by-lines. Signals from the probes are processed by electronics mounted close by in the plant, and the latter signals routed to a centrally located computer. The computer outputs the solids weight fraction and elemental concentrations in the monitored streams.

AmdeI was licensed to manufacture and market these systems in 1973. These analysis systems are now installed in about 55 mineral concentrators throughout the world. The typical plant system simultaneously monitors about 5 process streams.

Soon after installation of these analysis systems in concentrators, plant operators experiment with changing plant parameters such as reagent and air addition rates. This leads to a better understanding of control of the plant, with increase in recovery of valuable minerals (Figure 7). The value of the increased recovery depends on the ore throughput of the plant, but is usually between \$ 100,000 and a few million dollars per year.

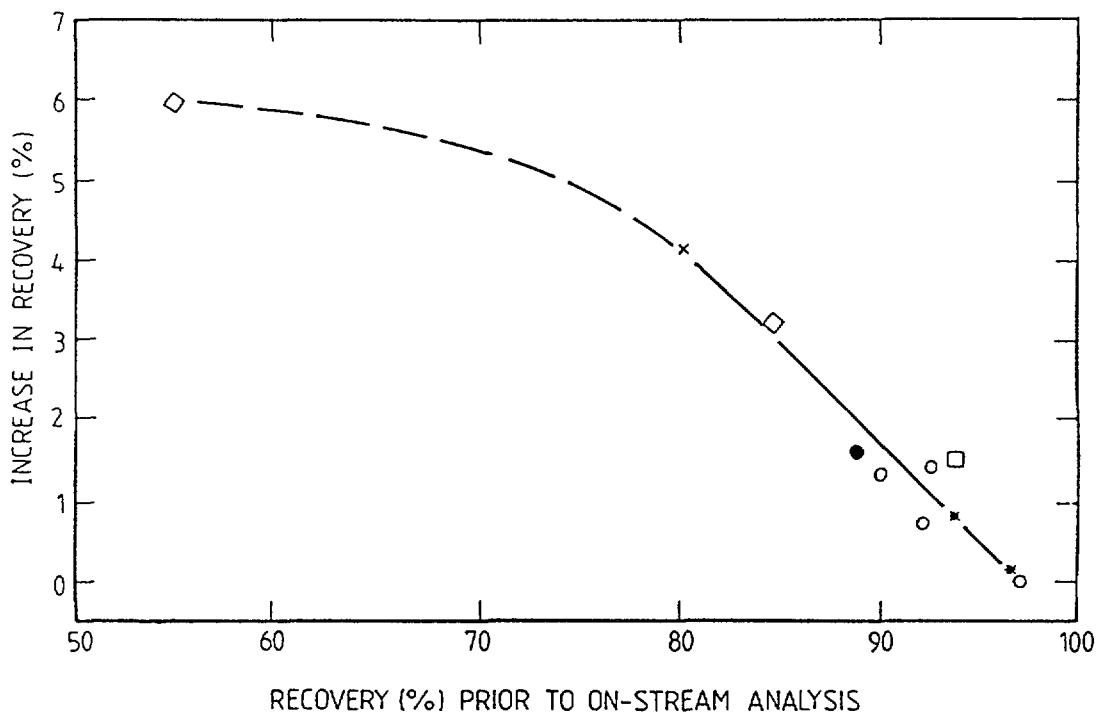


Fig. 7. Improvements in recovery of mineral concentrators after installation of on-stream analysis systems.

The mineral industry world-wide is now fully aware that recovery of valuable minerals is increased after on-stream analysis systems are installed in mineral concentrators. The trend is now for mineral companies to contract for a combined on-stream analysis and control package. A recent very successful example of this is the recent installation and control package at the Balaria Plant of Hindustan Zinc's Zawar Mines.

On-stream analysis systems have proved to be of great value in pilot plants which test alternative strategies to flotation of ore prior to the design of the main concentrator. This reduces the time required to establish the optimum strategy for the production concentrator.

4. TECHNOLOGY TRANSFER

Technology transfer in the broadest sense takes place during the whole process of research, development, plant trials, licensing to the instrumentation manufacturers, and use in industry. There is a continuing interaction between researcher, instrument manufacturer, and industrial user. Considerable research and development is undertaken after the licensing of the technique, mainly because new problems and applications arise out of experience of industrial use.

The most important stages of technology transfer are the plant trials of the new technology, and the transfer from researcher to licensee, and from licensee to industrial user.

4.1 Plant trials

All the techniques developed by CSIRO have been tested in one or more plant trials involving CSIRO scientists and industrial user prior to the system being commercialised. The system for on-stream analysis of mineral slurries was tested in six plant trials. The reasons for the large number of trials were that five different XRF and XRA techniques were involved, and industry acceptance was more difficult to obtain because this was the first on-stream analysis technique developed in Australia. The on-line analysis techniques developed later for the coal industry were accepted much more quickly because of our previous success with the mineral industry, and because commercial licensees had much more experience in designing, manufacturing and marketing the on-line equipment. Each different technique developed for the coal industry has been tested in one or two plant trials.

Plant trials are a key part of the process of technology transfer. The demonstration of the technique in the industrial plant, and the experience gained by the company metallurgists and engineers in its use, leads to staff confidence in the innovation. The researcher also gains important information on plant conditions during the trial, sometimes leading to modifications to the on-stream analysis system.

4.2 Licensees

The licensee plays a critical role in transfer of technology to industry. The licensee is the potential user's main source of expertise in on-line analysis. Hence the transfer of technology from researcher to licensee is of key importance. The licensee sometimes begins with only limited experience in the field of innovation.

The transfer of the technology for on-stream analysis of metalliferous mineral slurries to the licensee, Amdel, took much time and effort because Amdel had at that time (1970) no experience in radioisotope X-ray techniques and in industrial nucleonics. An Amdel scientist worked at the CSIRO laboratories for a year, taking part in the development of the system and in joint plant trials. With the experience gained, Amdel later developed other nuclear systems as gamma-ray density gauges, laboratory XRF mineral analysers, and solid state detector probes for on-stream analysis.

MCI was set up in 1981 by an engineer who had previously worked for Amdel, and two other Amdel scientists/engineers were soon recruited. Hence two Australian companies now had experience with nuclear on-line analysis systems. MCI were licensed by CSIRO to manufacture and market the two (Coalscan) ash gauges in 1982. MCI developed the Coalscan gauges based on CSIRO specifications, but undertaking most of the mechanical and electronics design, and all the software, themselves.

CSIRO undertook the first plant trial of the coal slurry analysis system with BHP Steel International at Newcastle. CSIRO selected Amdel as licensee in 1986, and shared with Amdel in the first installation of the commercial gauge, and in the long-term plant demonstration of the system's reliability and accuracy. This has proved to be a highly successful mode of technology transfer to Amdel and to industry, and a model for future technology transfer to industry. During this demonstration installation, technology was transferred from CSIRO to Amdel and plant staff, and CSIRO gained experience in plant installations from Amdel.

Both Amdel and MCI are well-established companies in the field of on-line analysis instrumentation, marketing the analysis systems world-wide. They undertake the technology transfer to industrial users at the time of installation of the commercial analysis system. Since their commercial systems are now used world-wide, their capabilities in transfer of the technology have been clearly demonstrated.

4.3 UNDP Industrial Project: minerals industry sub-project

The UNDP/IAEA/RCA Regional Industrial Project was set up to extend the use of isotopes and radiation technology in industries in the Asia/Pacific region. In 1982, the Australian Government agreed to play a leading role in the minerals field of this project, joining with the IAEA in the setting up of a sub-project "On-stream analysis and control of mineral concentrators". Australia provided expertise in on-stream analysis (CSIRO) and control of mineral concentrators (University of Queensland).

The original aim was to train plant metallurgists and engineers in the technologies of on-stream analysis and control of mineral concentrators. Two courses were held in 1982-83 and 1985-86, with 4-5 weeks in Australia, and 2-3 months in the Philippines. In Australia, participants were trained in nuclear techniques and control, and visits were made to Amdel and to mineral concentrators for participants to view on-stream analysis installations and to discuss control strategies with their Australian counterparts. In the Philippines, the Philippines Atomic Energy Commission gave a one week course in nuclear techniques, and this was followed by two-three months of practical training in on-stream analysis and control undertaken by Australian experts at the Philex Mining Corporation's copper concentrator near Baguio City.

An Executive Management Seminar, introduced in 1984 to establish contact with top management of important mineral companies in the Asia/Pacific region, proved to be highly successful. In hindsight, this two-day seminar should have been held in 1982 at the beginning of the project. A four-week course in control of mineral concentrators was held in Australia in 1984 for mill managers. In 1986, another Executive Management Seminar was held, and two short training courses on "Instruments for on-line and bulk analysis" were given in China.

Dr. Whiten, University of Queensland, and I visited six countries in the region late in 1986 to make an end-of-project assessment. We were told by company management of considerable savings made in their concentrators which they assessed were a direct result of the training courses^(17,18). Hence we concluded that the sub-project was successful.

Looking back now (1990), there are eleven Amdel radioisotope, and at least five X-ray tube, on-stream analysis systems installed in the region. The Amdel installations are summarised in Table 3. There were no radioisotope on-stream analysis installations in the region in 1982, when

TABLE 3. PLANT INSTALLATIONS OF AMDEL SYSTEMS FOR THE ON-STREAM ANALYSIS OF SLURRIES IN MINERAL CONCENTRATORS IN THE ASIAN AND PACIFIC REGIONS.

COUNTRY	COMPANY/MINE	LOCATION OF PLANT	ANALYSIS FOR	NUMBER OF STREAMS ANALYSED	DATE OF INSTALLATION
Burma	Number One Mining	Bawdwin	Pb,Zn,Cu	8	1987
China	Yunnan Tin Corporation	Ta Tun Mines	Sn, S	6	1985
	Hong Tou Shan	Liaoning	Cu,Zn	3	1985
	Dachang	Guanxi	Sn,Pb,Zn,As	7	1986
	Anshan Mining Co.	Anshan, Liaoning	Fe	4	1988
India	Hindustan Zinc Ltd				
	- Zawar Mine	Udaipur	Pb,Zn,Fe	5	1988
	- Rampura Agucha	Udaipur	Pb,Zn,Fe	8	1989
	Hindustan Copper Ltd.	Malanjkhand	Cu,Fe	5	1988
Malaysia	Mamut	Sabah	Cu,Fe	4	1986
Papua/New Guinea	Ok Tedi	Tabubil	Cu,Fe	11	1987
Philippines	Philex Mining Co.	Baguio	Cu	7	1983

the course was begun. Most mining companies in the region were then aware of on-stream analysis, but had not been sufficiently confident of the technology or of the potential savings to convince them to have a system installed. The Indian, Malaysian and Philippine installations were a direct result of the course. It is difficult to assess the extent of influence of the course on the other installations. Some of the installations in other countries were probably independent of the course.

My assessment of the real value of the courses was to give mineral company management and staff direct contact with Australian experience in on-stream analysis systems and control of mineral concentrators. Their very understandable doubts about the stage of development of the analysers, and whether their companies had the expertise to operate these analysers reliably, were overcome by direct hands-on experience. There is little doubt that the companies would eventually have installed analysers if the training courses had not been undertaken, but the installations would have been delayed by several years.

4.4 UNDP Industrial Project: Coal industry sub-project

The Australian Government agreed to play a leading role in transfer of technology relating to on-line determination of the ash content of coal and control of coal operations, joining with the IAEA in the setting up of the sub-project. MCI is supplying the expertise on the Coalscan model 3500 (DET) ash monitor, and the University of Queensland the expertise on control of coal operations. The first Executive Management Seminar was held in Australia in 1987, and the first training course were held in Thailand in 1989.

The Australian Government supplied the first Coalscan ash monitor, and Thailand has since ordered a further six monitors to be delivered over the period 1990-1992. Companies in India have ordered five Coalscan 3500 ash monitors for installation in 1990. This is an excellent start to the adoption of on-line ash monitors in the Asia/Pacific region.

5. CONCLUSIONS

Many of the on-line analysis systems developed for the coal and mineral industries, particularly the systems for on-line determination of the ash content of coal, and for on-stream analysis of metalliferous mineral and coal slurries, have been proved in long-term industrial use. The economic benefits resulting from use of the on-line analysis information to improve control of mining and processing operations have been proved, with payback times for the cost of the on-line analysis installation often being several months.

The technology transfer, undertaken as part of the UNDP Industrial Project, of on-stream analysis and control of mineral concentrators has been successful, with many analysis systems now installed in Asia/Pacific countries. Experience in this sub-project has shown the need for technology transfer to senior company executives, mill managers, and metallurgists and engineers working in the concentrator.

ACKNOWLEDGMENTS

The on-line analysis systems discussed above have been initiated and developed in collaboration with Australian industry, by a group working firstly with the Australian Atomic Energy Commission and then, since transfer in 1982, with CSIRO Australia. The development of the analysis systems for coal and coke have been supported, in part, under the National Energy Research, Development and Demonstration Program of the Australian Government.

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BENEFITS OF NEW APPLICATIONS AND NEW TYPE GAUGES USING NUCLEAR TECHNIQUES IN THE IRON MAKING PROCESS

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Abstract

The steel industry in Japan is a big user of radiation-applied gauges which have contributed to the mass production of high quality steel products. The steel industry has, however, still a strong requirement for more advanced instrumentation to achieve the higher stabilization of process operations. An example of new application of nuclear techniques is the introduction of a depth-type density gauge to an ore-sintering process. Another example is a new moisture gauge for coke to be supplied to a blast furnace. The gauge is based on a new measurement principle, i.e., simultaneous use of fast neutron and gamma-ray transmission and applied to a new measuring point, i.e., the exit gate of a coke hopper. The annual savings in a sintering plant is estimated at 53 million yen for 6 million tons production per year. The savings in the blast furnace due mainly to saving of coke is estimated at 57 million yen per annum.

1. INTRODUCTION

The steel industry contains coal- and iron-ore treating processes and consumes a large quantity of them for iron making. It is also a big user of radiation-applied gauges and many of the gauges have contributed to the mass production of high quality steel products. The steel industry has, however, still a strong requirement for more advanced instrumentation to achieve the higher stabilization of process operations.

An example of new application of nuclear techniques is the introduction of a depth-type density gauge to an ore-sintering process. Although the principle used is not new but conventional Compton scattering, technical and economic benefits are considered to be great, since no appropriate gauge has so far been applied to the process.

As another example, a new moisture gauge for coke to be supplied to a blast furnace is described. The gauge is based on a new measurement principle, i.e., simultaneous use of fast neutron and gamma-ray transmission and applied to a new measuring point, i.e., the exit gate of a coke hopper.

2. DENSITY GAUGE IN THE SINTERING PLANT

To blast furnaces producing molten iron, iron ore is supplied usually in the form of sintered ore. In sintering plants, raw material granules consisting of iron ore mixed with a small fraction of coke and limestone are softly loaded on a moving conveyor and heated by an ignition burner. The yield of sintered ore in the plant strongly depends on the uniformity of the loaded materials and remains still at a low level of 70–80% due to the lack of appropriate measuring means of such a material condition. The ore not sufficiently sintered must be processed repeatedly in the same plant. To improve the yield, a new sensor¹⁾ has been developed for measuring the spatial change of apparent density of the raw material loaded on the conveyor, instead of direct measurement of air flow uniformity in the materials.

2.1. Features of the Density Gauge

To measure the density gauge in the vertical direction of the raw material just loaded on the conveyor from a hopper, two probes of a specially designed gamma-scattering gauge was applied to a sintering plant of Kimitsu Works, Nippon Steel Corporation, as shown in Fig. 1. The structure of the gauge probe is schematically shown in Fig. 2 which consists of a 3.7 MBq Cs-137 gamma-source, a lead shield 25 mm Ø x 100 mmL, and a bismuth germanate gamma-detector 20 mm Ø x 100 mmL.

The main features in the functions of the gauge are summarized below:

- (1) good spatial resolution in local density measurement with a small effective measuring thickness, a little less than 50 mm, in the directions perpendicular to the axis of the probe,
- (2) minimum disturbance of the material flow by insertion of the small diameter probe along the direction of the flow,
- (3) negative slope calibration curve,

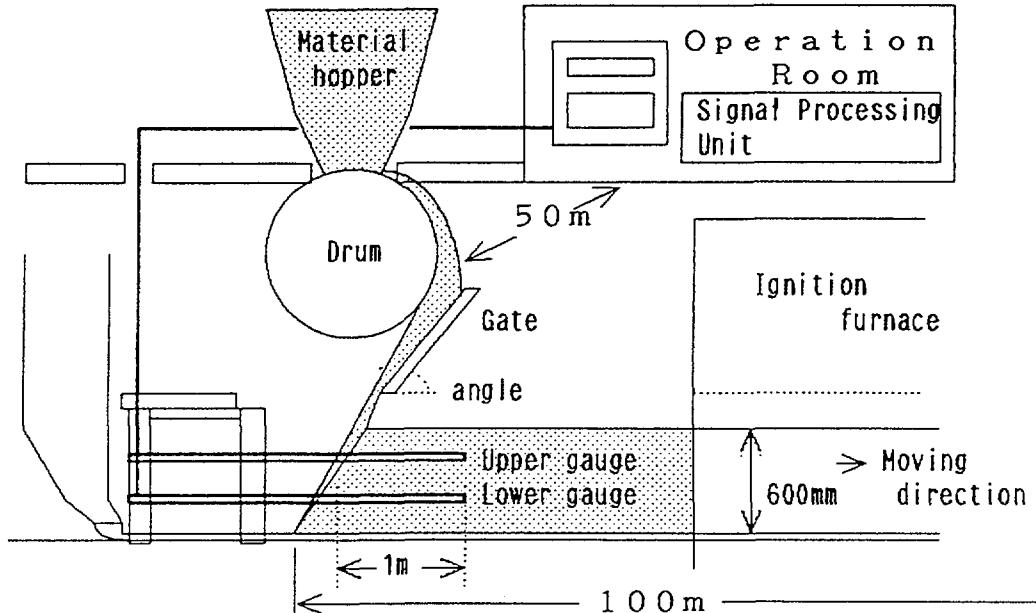


FIG. 1. On-line application of the gauge for density measurement.

- (4) sufficiently high statistical precision, $3\sigma=0.017t/m^3$ for 100s counting in a density range of $1.5-2.0t/m^3$, despite the use of a small activity source of 3.7MBq, and
- (5) high stability of the gauge with the aid of electronic feedback control.

2.2. Technical Effects in On-line Application

The on-line application of the gauge in the arrangement of Fig. 1 was started in June, 1988. Since then the gauge is working well continuously and producing data which gives valuable information on the relationship between each plant parameter and local density variation.

Figure 3 shows the effect of alteration of the gate angle. When it was set at a smaller angle, the rate of fall of the raw material granules became slow and resulted in a reduction in the density of the loaded material. The new gauge revealed such a relationship quantitatively. Figure 3 also shows the reproducibility of both the plant operation and the measuring system.

Figure 4 shows an example of local density changes observed by the upper and lower gauges. Under the normal operation condition the upper gauge indicated lower density than the lower gauge: the cross-over region in Fig. 4 means an abnormal state of the loaded material.

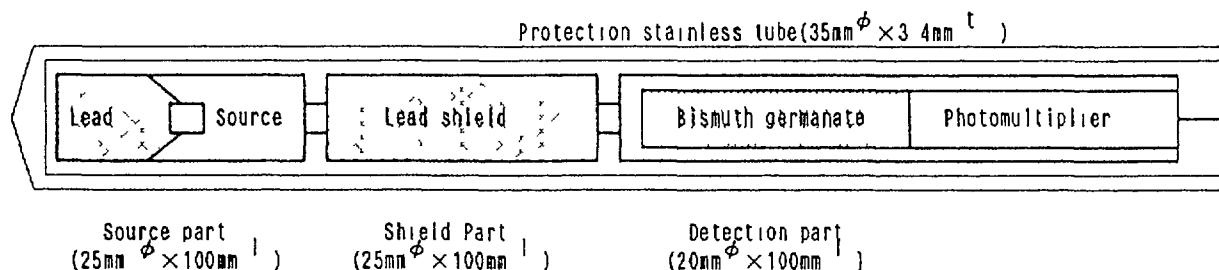


FIG 2 Cross-section of the gauge probe.

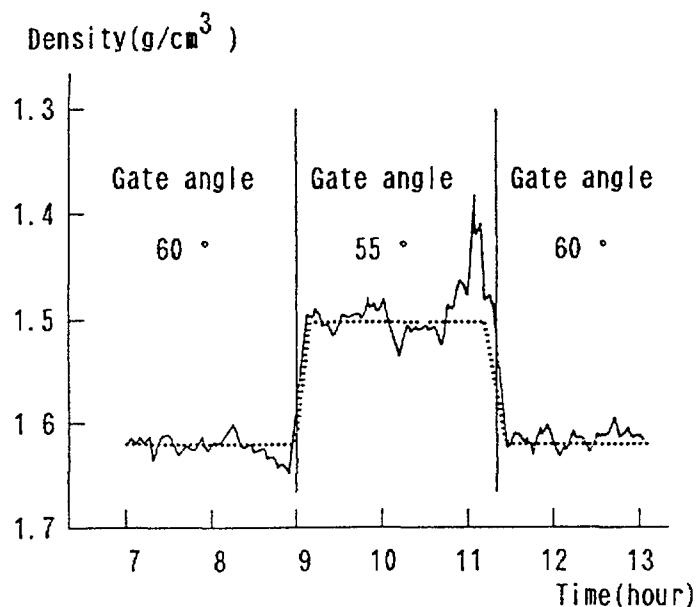


FIG 3 Observed density variation accompanied by an alternation of the gate angle

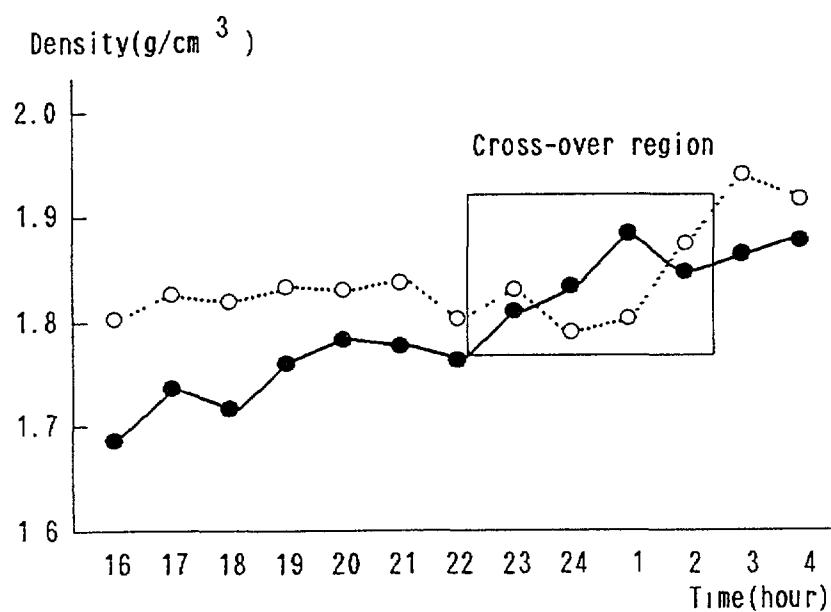


FIG 4 Example of spatial change observed in raw material density

TABLE 1. ECONOMIC BENEFITS OBTAINED BY THE INTRODUCTION OF THE DENSITY GAUGE IN A SINTERING PLANT (1988)

Sintered ore production	:	6 Million tons/year				
Yield improvement of sintered ore	:	77% - 78%				
Saving of consuming resources	:	1/77				
	Required quantity	Unit cost			Cost charge	
Coke	43.7 kg/ton	x	10 Yen/kg	x	1/77	= 5.68 Yen/ton
Coke oven gas	2.0 Nm ³ /ton	x	11.5 Yen/Nm ³	x	1/77	= 0.30 Yen/ton
Electricity	28.6 KWH/ton	x	7.5 Yen/KWH	x	1/77	= 2.79 Yen/ton
				Total		8.77 Yen/ton
Annual saving :	6×10^6 tons/year	x	8.77 Yen/ton	=	5.3×10^7	Yen/year

By the use of these data measured with the new gauge, some improvement can be expected in the plant operation.

2.3. Economic Benefits in the Routine Use

As a result of the improvement in the plant operation mentioned above, an increase of 1% (e.g. from 77% to 78%) in the yield of the sintering plant is considered to be attained comparatively easily. The yield increase reduces the consumption of resources such as coke by 1/77.

This results in a saving of 8.7 yen per unit ton of sintered ore, as shown in Table 1, and an annual saving of 53 million yen in a sintering machine of 6 million tons production per year. Since the cost of two sets of the gauge is about 10 million yen at most, the ratio of the cost to benefit is better than 1/5.

3. HIGH ACCURACY COKE MOISTURE GAUGE

The feed of coke to a blast furnace in an iron-making process must be balanced to that of iron ore on dry weight basis. Neutron moisture gauges utilizing thermalization of neutrons by hydrogen are usually used to measure the moisture content in coke. However, these conventional gauges cannot give an accurate value of the moisture content for the entire volume of a large coke hopper, due to the spatial variation of moisture and bulk density.

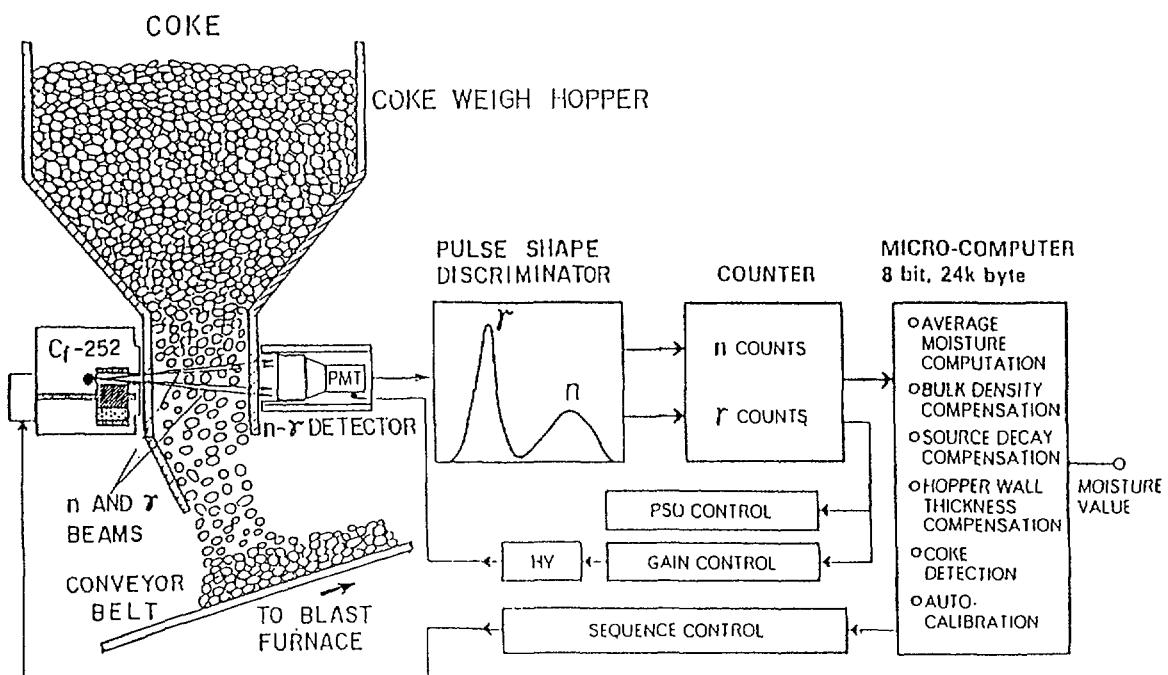


FIG. 5. High accuracy coke moisture gauge obtained by using neutron and gamma transmission.

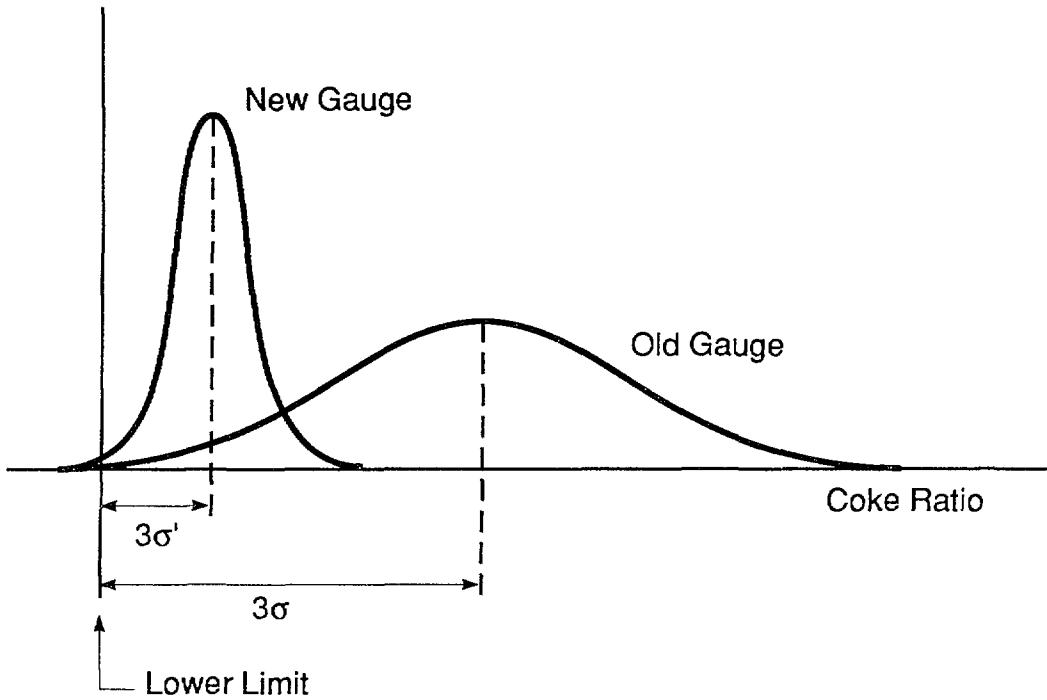
3.1. Features of the New Moisture Gauge

For saving of coke and strict control of furnace heat etc. in the iron-making process, a new type moisture gauge²⁾ has been developed by the method of simultaneous use of both fast neutrons and gamma rays from ²⁵²Cf. Measurement is performed by observing the transmission rates of both the fast neutrons and gamma rays for coke falling at the exit gate of a weight hopper, the moisture content and the bulk density being determined by solving a simultaneous equation, moment by moment (cf. Fig. 5). The average value of the moisture content for a batch of coke is given at the time when the falling is completed.

To realize the transmission measurement of neutrons and gamma rays for an identical portion of the object, a single detector of an organic scintillator, NE213, is employed which is capable of detecting with high efficiencies both fast neutrons and gamma rays separately by means of the pulse-shape discrimination (PSD), distinguishing between fast neutrons and gamma-rays based on the difference in decay time of scintillation.

An important key to develop the new gauge was the introduction of PSD into the field of industrial measurement.

The improvement in accuracy or precision of the moisture determination is considerable as compared with the conventional neutron moisture gauges.



$$3\sigma = 2.0\%, \quad 3\sigma' = 0.6\%$$

(including an error of coke weighing machine = 0.3%)

Reduction of the coke ratio :

FIG. 6. Reduction of the coke ratio as an effect of the improvement of the coke moisture measurement.

3.2. Technical Benefits of the Improvement of Measuring Precision

The improvement in the precision of the moisture determination means the improvement of measurement precision of coke weight without moisture. This makes it possible to reduce the coke ratio which is expressed as an amount of coke to be supplied for producing unit weight of molten iron, as shown in Fig. 6. Figure 6 also shows a typical calculation of the amount of coke saving. This is a direct effect of the introduction of the new moisture gauge.

TABLE 2. ECONOMIC BENEFITS OBTAINED BY THE USE OF THE NEW COKE MOISTURE GAUGE IN AN IRON MAKING PROCESS (1988)

Molten iron production	:	9,200 tons/day	
Saving of consuming resources			
	Reduction	Unit cost	Cost charge
Coke ratio	1.3 kg/ton	13.6 Yen/kg	-17.68 Yen/ton
Blast air	5.2 Nm ³ /ton	0.653 Yen/Nm ³	-3.40 Yen/ton
Blast furnace gas	4.8 Nm ³ /ton	1.776 Yen/Nm ³	+8.52 Yen/ton
HS - BFG	1.8 Nm ³ /ton	1.8 Yen/Nm ³	-3.24 Yen/ton
HS - COG	0.1 Nm ³ /ton	12.1 Yen/Nm ³	-1.21 Yen/ton
		Total	-17.01 Yen/ton
Annual saving : 9,200 ton/day × 365 days × 17.01 Yen/ton = 5.7 × 10 ⁷ Yen/year			

The direct effect results further indirectly in technical benefits as follows:

- (1) reduction of the blast furnace temperature,
- (2) reduction of the silicon content and its variation in produced molten iron,
- (3) reduction of heat radiation from the furnace, and
- (4) stabilization of operation of the furnace.

3.3. Economic Benefits of the Use of the New Gauge

The economic benefits of the use of the new gauge against old gauges can be evaluated from the following measurable effects:

- (1) saving coke,
- (2) saving of hot blast air,
- (3) saving of heating gas, namely, blast furnace gas and coke oven gas, in a hot stove, and
- (4) decrease of the production of blast furnace gas.

From Fig. 6, the economic benefit is calculated as shown in Table 2. The positive effect of the saving of coke is the largest in all the cost changes. In total, a cost reduction of 17.01 yen per unit ton of molten iron is attained. The annual saving of all the resources amounts to 57 million yen. This example shows that the introduction of a new technique results in significant economic benefits in a large scale plant.

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STATUS AND TRENDS IN NUCLEAR TECHNIQUES IN THE MINERAL INDUSTRY IN INDIA

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Abstract

India has large resources of coal, iron, manganese ores, mica, phosphate and less of non-ferrous metals and mineral ores such as corundum graphite, gypsum, sillimanite, bauxite and thorium etc. India is self-sufficient in coal, iron ore, titanium, manganese ore, mica, pyrites, feldspar, industrial clay, limestone, zircon, etc. but is deficient in sulphur, platinum, silver, nickel, cobalt, zinc, tin, mercury, molybdenum, potash fluorite and others. The most significant application of nuclear techniques in the mineral industry in India involves the use of nucleonic control systems. One of the very early nuclear applications in mineral industry in India was for controlling the level of zinc ore in an underground mine hopper. Similarly ore hoppers and bins in a copper processing complex have been provided with a number of nucleonic level control systems. Radiometric density gauge type RDG 4124 system developed and manufactured by the Electronics Corporation of India Ltd finds wide application in mineral processing industry. The instrument based on gamma ray attenuation technique is suitable for materials in liquid and slurry form. Ash monitoring in coal producing and using industry, employing nucleonic system is yet to get established in the Indian industry. A coal ash monitoring system developed indigenously by M/s Electronics Corporation of India has been installed at Central Coal Washeries Organisation, Dugdha, Bihar. The system uses X-ray back scattering. Off-line X-ray fluorescence system was adopted by a zinc ore processing plant for quite some time. Two AMDEL radioisotope laboratory analysers are used at the Hindustan Zinc, Udaipur for a fairly rapid analysis of samples from process streams of two adjacent concentrators. They have reported 1.5% increase in recovery attributable to radioisotope analysis. The two Indian oil companies, the Oil & Natural Gas Commission and the Oil India Ltd routinely use nuclear well logging techniques along with other conventional logs such as caliper, temperature, resistivity, SP, etc. Radiotracer application in mineral industry is not as wide spread as the tracer application in other industries in India. At the alumina plant of M/s Hindustan Aluminium Corporation at Renukoot, residence time studies were carried out in two bauxite digestor

units. In the oil industry, tracer techniques are extensively used for locating water channeling and to obtain water injection profiles. The Oil and Natural Gas Commission has recently started using tritiated water to trace the movement of injection fluids in secondary recovery operations.

INTRODUCTION

Nuclear Techniques have now permeated into all spheres of economic activity in the developed and in some of the developing countries. Mineral Industry is no exception. A number of nuclear techniques are used with advantages in the mineral industry also. It is often stated that the level of industrialisation in a country is determined to a great extent by its mineral resources. India has large resources of coal, iron, manganese ores, mica, phosphate and less of non-ferrous metals and mineral ores such as corundum, graphite, gypsum, sillimanite, bauxite and thorium etc. When one matches with the country's requirements India is self-sufficient in coal, iron ore, titanium, manganese ore, mica, pyrites, feldspar, industrial clay, limestone, zircon, etc. but is deficient in sulphur, platinum, silver, nickel, cobalt, zinc, tin, mercury, molybdenum, potash fluorite and others. Considering the distribution of minerals in different states we can say that State of Bihar is most abundant in mineral resources accounting to about 3%, followed by other States of West Bengal, Madhya Pradesh, Gujarat, Assam, Orissa, Andhra Pradesh, Tamil Nadu, Karnataka, Goa, Rajasthan and Maharashtra.

Coal

Coal Industry occupies the position of pre eminence. The production increased from 140 million tons in 1984 to around to an estimated 200 million tons during 1989. Most of the coal mines belong to the state owned Coal India Ltd except for the mines operated by the Tata Iron & Steel Co. Coal washing and coal beneficiation plants have been put up to upgrade local coking coals and for blending non-coking coals with coking quality. Among the 400 odd coal mines over 20 have capacity above 1/2 million tonnes per annum. India collaborates with United Kingdom, Soviet Union, France, Poland and Germany in the coal mining field.

India also have large lignite reserve and mining complex in Tamil Nadu State.

Iron

In the case of iron ore India has large reserves of haematite and magnetite iron ore mainly in the Bihar-Orissa belt and in Madhya Pradesh and Goa. Some of the mines are captive mines owned and operated by Steel Plants. Some of them are operated by National Mineral Development Corporation and State Government Undertakings for export and internal consumption. The production capacity of the mines is nearly 70 million tonnes per annum.

Manganese

Recoverable resources of manganese ore in India are estimated to be about 120 million tonnes. Orissa, Karnataka, Maharashtra, Madhya Pradesh and Goa are the biggest producers. The industry is mainly export oriented with about half the consumption within the country by Ferro Manganese plants etc.

Aluminium

Aluminium is the only non-ferrous metal for which India has adequate resources. Bauxite deposits are estimated to be over 2 billion tonnes. Most of the bauxite occurs in the east coast. Capacity utilization of many of the aluminium plants is limited by the power availability. Indian Aluminium Co, Hindustan Aluminium Co, National Aluminium Co and Bharat Aluminium Co are the main producers of Aluminium and alumina.

Copper

India's copper ore reserves are limited. The important occurrences are in Bihar, Madhya Pradesh and Rajasthan. The ore is also of very low copper content. Hindustan Copper Ltd is the main producer of the ores as well as the operator of number of concentrators and two smelter plants at Khetri and Ghatsila.

Zinc

India has fairly good reserves in zinc and is able to produce about 60% of its requirement of zinc locally. Main deposits are in Rajasthan and some deposits are also exploited in Andhra Pradesh and Orissa. The largest smelters are situated near Udaipur and in Vishakapatnam. The Debari smelter near Udaipur has a capacity of around 50,000 tons/year. One of the smelters

in Cochin depends mainly on imported concentrates. Another zinc smelter is now planned in Rajasthan.

Lead

Lead deposits are associated with the zinc deposits. Therefore the zinc smelter have down stream capacity for lead production also. An additional lead smelter is located at Tundoe in Bihar. Additional lead production capacity is expected to come up with the new zinc smelter being planned in Rajasthan.

Mica

India is the largest producer and exporter of Mica in the world. But demand for mica is coming down due to the competition from plastic films. The highest production level reached is only about 30,000 tonnes per year.

Oil

In India most of the oil production is from off-shore oil fields near Bombay. Substantial on-land production has also been going on for the last 30 years. Indigenous production amounts to about 60% of the country's requirement. Exploration results on the East Coast are promising.

NUCLEAR TECHNIQUES

The application of nuclear techniques in the mineral industry in India is not very large. Most significant applications involve the use of nucleonic control systems.

Level Control

One of the very early nuclear applications in mineral industry in India was for controlling the level of zinc ore in an underground mine hopper. In this case level control using gamma ray transmission technique was adopted. The level control was essential to make sure that optimum use of the skip carrying the ore to the surface is made. Availability of a pre measured quantity of ore in the loading hopper at the mine bottom helps in minimising the waiting time for the skip at the mine bottom.

At M/s Hindustan Zinc smelter at Udaipur zinc powder level in hopper is controlled by nucleonic level control system. Zinc powder at 1000°C is cooled while entering the hopper and the dust collected at the bottom of the hopper is discharged by a vibrator. It is essential to keep a certain level of zinc powder always in the hopper so that the zinc powder does not catch fire by the sudden entry of air. Nucleonic control systems helps to maintain this level.

Similarly ore hoppers and bins in the copper processing complex at Khetri also have been provided with a number of nucleonic level control systems.

Radiometric Density Gauge

Radiometric density gauge type RDG 4124 system developed and manufactured by the Electronics Corporation of India Ltd finds wide application in mineral processing industry. The instrument based on gamma ray attenuation technique is suitable for materials in liquid and slurry form. For ore crushing and thickening operations the instrument is specially suited. M/s Hindustan Zinc Ltd in its plant in Orissa uses such a system on its pipeline carrying crushed ore slurry.

Other instances of applications of Radiometric density gauges are (i) bauxite mud slurry measurement at Hindustan Aluminium Corporation, Renukoot, Uttar Pradesh, (ii) Magnetite water slurry measurement at West Bokaro Collieries in Bihar and (iii) Coal slurry measurement at Coal Washery of Bharat Coking Coal Ltd at Madhuban, Bihar. In the third case the system is incorporated by the supplier of the Washery equipment. All these systems are indigenously produced.

Coal Ash Monitoring

Ash monitoring in coal producing and using industry, employing nucleonic system are yet to get established in the Indian industry. Sampling type coal ash monitoring system using X-ray fluorescence technique was initially introduced in the West Bokaro Collieries of the Tata Iron & Steel Co. The system had a number of problems in the field use and is therefore not in regular use now.

A coal ash monitoring system developed indigenously by M/s Electronics Corporation of India has been installed at Central Coal Washerries Organisation, Dugdha, Bihar. The system uses X-ray back scattering. The coal from the main conveyor is tapped and crushed. The crushed coal sample (5 mm) comes on to a rotating presentation table as a continuous stream and emerges under the sensor assembly as a compact bed of coal 140 mm wide and 40 mm thick with a smooth surface with reproducible bulk density. After the measurement, the coal passes out and is fed back to the main conveyor by a secondary conveyor.

Eastern Coal Fields Ltd which is a part of M/s Coal India Ltd is now introducing an on-line coal ash monitoring device. The experience from this system may result in the introduction of more such systems in the country. The very high ash content 20-40% makes it essential to control average ash content of coal being supplied to different utilities.

Mineral Analysers

Off-line X-ray fluorescence system was adopted by a zinc ore processing plant for quite some time. Two AMDEL radioisotope laboratory analysers are used at the Hindustan Zinc, Udaipur for a fairly rapid analysis of samples from process streams of two adjacent concentrators. They have reported 1.5% increase in recovery attributable to radioisotope analysis. They are presently incorporating an AMDEL on-stream analyser in the Udaipur plant.

Oil Well Logging

Like all other major oil companies in the world, the two Indian companies, the Oil & Natural Gas Commission and the Oil India Ltd routinely use nuclear well logging techniques along with other conventional logs such as caliper, temperature, resistivity, SP, etc. The oil companies are well equipped for natural gamma, gamma-gamma, neutron-gamma, neutron-neutron (both scatter and delayed neutrons) etc. They use both isotope neutron sources and 14 MeV pulsed neutron generators. In fact, the Oil and Natural Gas Commission has a subsidiary for nuclear logging operations. The Indian companies have the devices of international companies like Schlumberger for advanced logging techniques such as high sensitivity spectral logging.

Radiotracers

Radiotracer application in mineral industry is not as wide spread as the tracer application in other industries in India. We may consider here one or

two specific applications which are useful for different mineral industries. At the alumina plant of M/s Hindustan Aluminium Corporation at Renukoot, residence time studies were carried out in two bauxite digestor units. Each of the units consisted of 3 or 4 digestors in series. The digestor volumes varies from 100-250 m³. For one unit a single tracer injection (sodium-24) was done at the first digestor inlet.

Tracer concentration time curves were obtained after each stage with suitable radiation monitoring systems. By using a multi box model, residence time distribution of each digestor was worked out. It was possible to work out dead volume for each digestor also.

RTD IN BAUXITE DIGESTOR

$$Q = 7.5 \text{ m}^3/\text{h} \quad V = 247 \text{ m}^3 \times 4 \quad t_{th} : 32.9 \text{ min}$$

Digestor No	t	Effective Vol. =	Model simulation			Dead Volume (m ³)
			n	v (m ³)	Eff.Vol. (m ³)	
			t x Q (m ³)			
1.	30.5	228.7	5	40	200.0	47.0
2.	23.7	177.7	5	35.5	177.5	69.5
3.	28.4	213.0	5	43	215.0	32.0
4.	23.9	179.3	36	5	180.0	67.0

In the oil industry, tracer techniques are extensively used for locating water channeling and to obtain water injection profiles. Down hole flow rate measurements are carried out using special tracer based probes.

The Oil and Natural Gas Commission has recently started using tritiated water to trace the movement of injection fluids in secondary recovery operations. Two injections have been carried out in two different zones in the Bombay off-shore oil field. Tracer from the first injection made in May 1988 has already yielded valuable data and the recent injection made in August 1989 is being followed up. The ONGC has plans to conduct multi-tracer studies in on-land areas also for both liquid and gas tracing. In addition to

providing information on inter-well connections useful for controlling secondary recovery operations, ONGC feels that should these reservoirs be committed to a subsequent tertiary oil recovery, the tracer information would be useful for the design and control of the recovery process.

RCA Project Activities

The UNDP/IAEA/RCA Regional Industrial Project has helped to a great extent in increasing awareness of Nucleonic Control Systems in Mineral Industry. Through Training Courses and Seminars a number of personnel have been exposed to systems like the On-line Coal Ash Monitors and On-stream Mineral Analysers used in the Industry. Under the project demonstration programmes have been carried out both in Australia as well as in the Philippines. During the phase I of the project an on-stream analyser has been installed at the Philex Mining Corporation at Baguio, Philippines, for the analysis in copper concentrators. The philex installation has been used for conducting training courses and management seminars. India has sent representatives to participate in these programmes both from the copper and zinc industry. In phase II, installation is now underway for coal ash monitoring system in the Thailand. Executive Management Seminars, Workshops and Training Courses are planned under this programme.

In conclusion it may be stated that the use of nuclear techniques in Mineral Industry in India is yet to pick up in a big way. There is need for more On-stream Analysers which are known to improve the efficiencies of concentrators through increased grade and recoveries. There is a large scope for nucleonic coal ash monitoring systems in the Indian coal industry. Tracer investigations on different parameters such as residence time distribution, flow rate, flow paths, etc. should also be undertaken for optimisation of processes in the mineral processing plants.

APPLICATION OF NUCLEONIC CONTROL SYSTEMS IN THE MINERAL INDUSTRY IN INDIA

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Abstract

Electronics Corporation of India Limited (ECIL) a Govt of India Enterprise under the Department of Atomic Energy make nucleonic systems for various mineral applications, such as nucleonic level gauges, radiometric density gauges, radiometric belt weighers and nucleonic moisture gauges. These systems are used in Steel, Cement, Thermal Power generation and Ore beneficiation plants like zinc, copper, aluminum and coal. ECIL has supplied around 1600 nucleonic level gauges, 25 radiometric density gauges, 30 belt weighers and 4 nucleonic moisture and charge control systems for the blast furnace coke applications. Brief details of these systems are discussed in this presentation.

INTRODUCTION

The ever increasing need for automation in mineral industry requires continuous measurement of various process variables. For such measurements artificially produced radioisotopes along with the radiation detector are used in the form of a measuring probe. This has become possible with the availability of radioisotopes indigenously assisted by the advancement in microprocessor based systems. Artificially produced radioisotopes have the inherent advantages of emitting radiations spontaneously require no power for operation, small in size and require no maintenance. The radiation hazards associated with radioisotopes are avoided by using properly designed and approved radiation shields and observance of safety precautions. The measurements based on nucleonic technique are generally non-contact, non-destructive and have the ability to measure continuously. Continuous measurement leads to more economic control of process and reduced labour costs. Their performance is outstanding in areas of harsh environment where the other methods fail.

Electronics Corporation of India Limited (ECIL) a Govt of India Enterprise under the Department of Atomic Energy make nucleonic system for

various mineral applications, such as nucleonic level gauges, Radiometric density gauges, radiometric belt weighers and nucleonic moisture gauges. These systems are used in Steel, Cement, Thermal Power generation and Ore beneficiation plants like zinc, copper, aluminum and coal. ECIL has supplied around 1600 nucleonic level gauges, 25 radiometric density gauges, 30 belt weighers and 4 nucleonic moisture and charge control systems for the blast furnace coke applications.

NUCLEONIC SYSTEMS

Basically nucleonic measuring devices consist of a radiation source and radiation detector which forms the measuring part and an electronic processing unit to process and display the measured parameters and give suitable outputs for the process control. The probe can be arranged in a transmission or a backscatter geometry depending on the application and variable to be measured. In the transmission geometry the radioisotope and the detector are arranged on either side of the material. The material absorbs the radiation proportional to its physical property. The residual radiation detected becomes a measure of this property. In the backscatter geometry the radioisotope and the detector are arranged on one side of the process material. The amount of residual backscatter radiation gives a measure of the physical property of the material.

LEVEL MEASUREMENT

Nucleonic Level Gauges are widely used to indicate whether the bin is over or under filled. The gauges usually provide an ON/OFF signal to indicate filling above or below a predetermined level. These gauges are based on GAMMA ray transmission technique using the radioisotopes Co-60 or Cs-137 and a Geiger Muller counter or Scintillation counter as the detector. The source holder with a source and detector probe are mounted on the outside of the process vessel diametrically opposite to each other. The source-detector combination is unaffected by temperature, pressure, velocity, corrosivity, toxicity or viscosity of material whose level is under determination. The signal from the radiation detector is measured by the electronic unit to give the alarm when the level crosses the predetermined position.

These simple and inexpensive gauges have been widely used in various industries for the past 20 years. These include CEMENT, STEEL, COAL, THERMAL POWER and CHEMICAL industries. Compared to other types of techniques, Nuclear techniques are preferred for use in hostile environmental industrial conditions.

DENSITY MEASUREMENT

Nucleonic Density Gauges are based on GAMMA ray transmission techniques using radioisotopes Cs-137 or Co-60 and ionisation chamber or a scintillation counter mounted on a pipe through which the process stream flows. Changes in density as little as 0.005 g/cc can be detected. Accuracy depends on the extent of air entrained in the slurry, distribution of solids across the pipes and variation of the constituent elements in the slurry.

The gamma source is kept in a radiation shield with a safety shutter and is mounted on one side of the pipe or tank. A measuring chamber is fitted on the other side of the pipe to be diametrically opposite to the radiation source by means of mounting saddles. The selection of the type of source and strength depends on:

- (1) application
- (2) wall thickness and diameter of the pipe
- (3) sensitivity required.

These gauges are widely used for the measurement of densities of slurries in pipes in various process industries like CHEMICAL, PAPER and PULP, SUGAR and ORE BENEFICIATION plants like copper, iron, zinc and coal. These gauges in conjunction with mass flow meters are used to measure mass flow of slurries and liquids in pipes.

WEIGHT MEASUREMENT

Nucleonic Belt Weighing System employs nuclear radiation absorption to measure belt loading. This signal is used in conjunction with a speed transducer to indicate rate of weight delivered. As the material comes in the path of source and the detector, the amount of radiation reaching the detector varies depending on the weight of the material. The output of the detector gives weight per unit length which is multiplied by the output of the speed transducer i.e. unit length/unit time to give flow rate in tons per hour.

It consists of a measuring head, speed transducer and electronic processing unit. The measuring head is assembled in a C-Frame which consists of a pair of radiation sources and an ionisation chamber with its field electronics. The top limb of the C-Frame contains the radioactive sources and the lower limb has the long ionisation chamber. The collimation of each source holder is arranged to view the entire width of the belt. The radioactive sources can be kept in OPEN or CLOSE condition by means of a

shutter mechanism operated manually. The weight and speed signals from the tacho generator are multiplied in the electronic unit to give flow-rate.

Conventional belt weighers are more widely used to determine weight per unit area by mechanical or electro-magnetic means by measuring the force from one or more idlers under the belt. Whereas, Nuclear Belt Weighers can be installed easily without disturbing the process line and can operate in windy conditions, independent of belt length, shape, tension and slope. However, Nucleonic weighers are more dependent on the profile of the material on the belt, particle size and somewhat difficult to calibrate on-line. These Nucleonic Belt Weighers find application in FERTILISERS, STEEL, CEMENT and COAL sectors, especially where mechanical systems cannot be put due to environmental conditions. Nucleonic Belt Weighters unlike conventional mechanical weighers, being non-contact, with minimum moving mechanical parts, require negligible maintenance as there is no wear and tear.

COKE MOISTURE MEASUREMENT AND CHARGE CONTROL SYSTEM

Nucleonic Moisture Gauges are used for the measurement of moisture in bulk materials like coke, sinter mix and iron ore in steel industry. The principle of moderation of slowing down of fast neutrons by Hydrogen atoms in the medium is utilised in such measurements. Fast neutrons emitted by the radioactive source move into the medium of interest and undergo elastic collisions with atomic nuclei of the medium. As a result the flight path of fast neutrons changes abruptly and they start moving in a zig-zag fashion and their energy gradually decreases. The energy decrease suffered by a fast neutron during a collision is related to the mass of the colliding nuclei. The smaller the mass of the nuclei, the greater the slowing down of neutrons. Since Hydrogen is the lightest nucleus of all the elements and water contains Hydrogen, collision with water molecules cause a large decrease in neutron energy. When the energy of the neutron is reduced to the point where they are practically in thermal equilibrium with the medium they are called thermal neutrons. A thermal neutron detector is used for their detection. The output of these detectors is proportional to the moisture content in the medium.

Coke Moisture that is fed into the Blast Furnace varies from 2 to 18% by weight as a result of inconsistent quenching at the Coke Ovens and weather conditions during transportation. Since Blast Furnace is fed on wet-weight basis, variations of 2 to 3% moisture will adversely affect the performance of the Blast Furnace. An undetected increase in Coke moisture may cause the Blast Furnace to go 'cold'. To avoid this situation the Operator may opt to feed 2 to 3% of more Coke than required.

A typical arrangement of a system consists of a pair of measuring probes mounted on the two hoppers and a microprocessor based charge control system. The probe consists of a fast neutron source of 300 mCi. (AM241-Be) and two He3 proportional counters. The source is mounted on a rotating disc for opening and closing. The electronic unit is a dust proof, floor standing unit. It contains a microprocessor, interface units, low and high voltage power supplies, 16 digit alpha-numeric display, a key board and relay compartment.

The system measures moisture content of Coke and keeps it in its memory and calculates the amount of Coke to be fed into the Blast Furnace based on the moisture measured and the weight signal from the load cells mounted in the hopper. By means of this system, the major advantages are:

- (1) coke saving of 1 to 2% on each feed,
- (2) uniform output from Furnace with less impurities like silicon and sulphur.

ON-LINE COAL AND ASH MEASUREMENT

Coal ash is the oxidised residue left after burning coal. This is an important factor in determining its calorific value. An accurate measurement of Coal ash is important in Coal industry to control cleaning and blending operations.

There are many indirect methods to measure the ash content of coal based on the measurement of specific gravity by weighing or by its absorption characteristics measured by transmission or backscatter of beta, gamma, and x-radiation or by neutron activation. The chemical analysis of ash measurement is accurate, reliable but cannot be used for on-line measurements. In the present system x-ray backscatter technique is used, accomplished by a Pu-238 radioisotope.

The operating principle is based on the concept that when a material is subjected to irradiation by x-rays, a portion of the radiation is absorbed and the remainder is reflected. The amount of absorption varies as per the average sample atomic number 'Z'. The higher the atomic number of the sample, the greater the absorption. The variation of absorption of radiation with atomic number can be directly applied to the ash measurement since ash forming elements iron, silicon, aluminium, sulphur and phosphorous have on the average a higher atomic number than coal. These elements that constitute the ash reflect fewer x-rays from a radioisotope. So if the ash content of coal is

more, then more and more radiation is absorbed and less is reflected to the detector and vice versa. The variation in the iron concentration in coal is compensated by an aluminium foil.

The system consists of a probe head, a measuring system and a mechanical feed system. The probe head consists of a pair of Pu-238 radioactive sources and an x-ray proportional counter arranged in a 'central-source geometry'. The signal from the proportional counter is taken and converted into ash : in the electronic console unit which displays ASH PERCENTAGE.

The mechanical feed system consists of a sampler for taking the sample from the main conveyor belt and a crusher to deliver a uniform sample of -5 mm to the ash measuring unit at a rate of 16 kgs per minute. After measurement of ash the sample is again fed back into the main conveyor by means of a secondary conveyor.

The Coal Ash Monitor finds application in COAL WASHERIES, STEEL PLANTS, THERMAL POWER PLANTS, etc.

CONCLUSION

The applications of nuclear techniques in the fields where conventional methods have failed or do not exist are increasing day by day. With the present trend in our Indian Industry for modernisation, automation and expansion coupled with the increasing cost of raw material, a good demand is forecasted for sophisticated nuclear industrial on-line systems.

Nuclear Industrial Systems have their own inherent advantages and limitations:

- Nuclear techniques are applicable in the most hazardous working conditions.
- They are essentially non-contact and hence do not interfere in the manufacturing process.
- They very rarely involve moving mechanical parts and hence fairly maintenance free.
- Nuclear based systems are most suited for on-line process applications.

Limitations:

The wrong impressions regarding the hazards of using radioisotopes is preventing extensive use of these techniques in Indian Industry. Extensive training and education is required to remove these misconceptions.

Trained and knowledgeable people are required to operate these systems.

DEVELOPMENT AND TECHNOLOGY TRANSFER OF LOW COST NUCLEONIC GAUGES IN INDIA

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Abstract

Electronics Division of BARC has been engaged in the development of useful nucleonic control equipment for the mineral industry in India. One such unit where the technology has been transferred to an industrial entrepreneur having a good technical base in a gamma switch. The technology has been successfully implemented in a chemical plant under expert supervision for non-contact level gauging application. Though the technology holds a good promise for future market growth, much effort is needed to remove misunderstanding about the dangers of this technology.

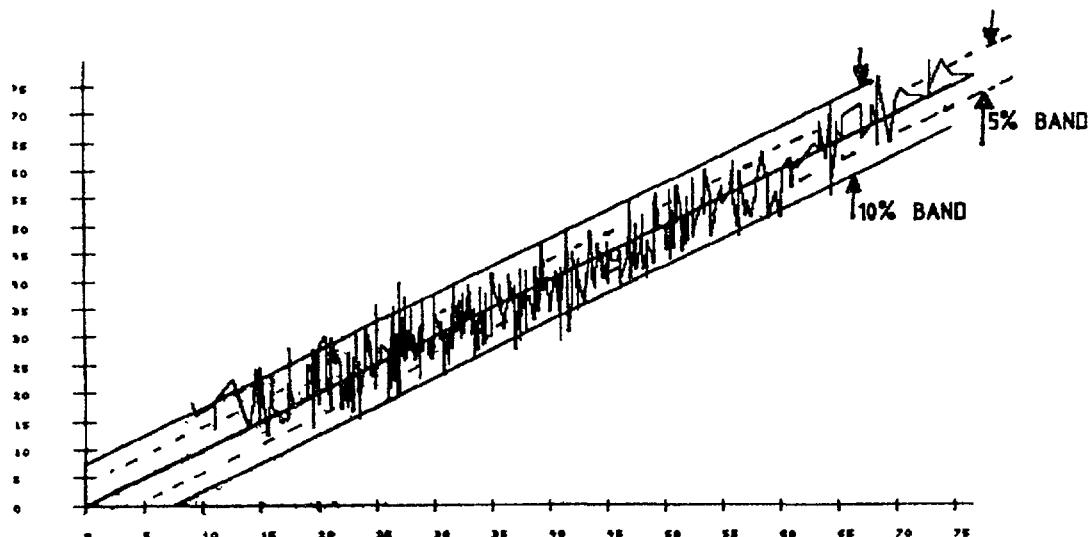
Experiences in Development of Nucleonic Gauges:

Electronics Division of BARC has played a useful role in the development of low cost nucleonic gauges for the non-contact measurement of industrial parameters. Taking advantage of the easy availability of radioactive sources in industrial grade packages, nuclear instruments were developed for the non-contact measurement of flow, density, weight and level, particularly for chemical industries. Apart from monitoring instruments, a pulsed neutron-neutron logging system was successfully developed, which was primarily aimed at investigating the water-oil and gas-liquid interfaces in the Ankaleshwar oil fields. The technique involved the generation of 14 MeV neutron bursts and detection of neutrons thermalized by the hydro-carbon formations in the borehole of the well using Helium-3 counters. The system consisted of a borehole unit with electronic control device and surface instruments for data analysis and recording. A special "on-line" data processing module was developed to determine the average life time 'T' of neutrons, and the value of 'T' for various formations of oil well as a function of depth was measured. The technique helped to determine the porosity and oil/gas saturation formation of productive strata. The experiments conducted in oil fields during the early eighties gave encouraging results.

A low cost laboratory equipment for the non-destructive monitoring of Ash Content in Coal, was fabricated and supplied to Coal Mining Research Institute in 1986. The instrument was based on the simple gamma absorption principle. It incorporated a low energy gamma source (Am 241/200 mCi) and a low noise counting system. A standardised coal sample preparation procedure was also developed by the use of a special container and vibrator for compacting the sample to uniform density. The instrument provided an accuracy of $\pm 10\%$ with a 95% confidence level for individual samples, with average values better than $\pm 3\%$ (ref. fig. 1). The error values were inclusive of variable iron content in samples. It offered the advantage of quick and easy determination of ash content as compared to conventional chemical methods.

Experiences in the development and application of industrial nucleonic control systems are summarised below:

Using a fine beam of Cesium-137 radioactive source, an equipment was developed to monitor and reject, underfilled and overfilled liquified petroleum gas containers moving on a conveyor belt at a reasonably steady



X axis : % Ash content by Chemical Analysis
Y axis : % Ash content by Gamma Absorption Technique

Points falling outside 10% error band = 31

Points falling outside 5% error band = 159

Total number of Samples = 480

FIG. 1. Performance graph of the coal ash monitor.

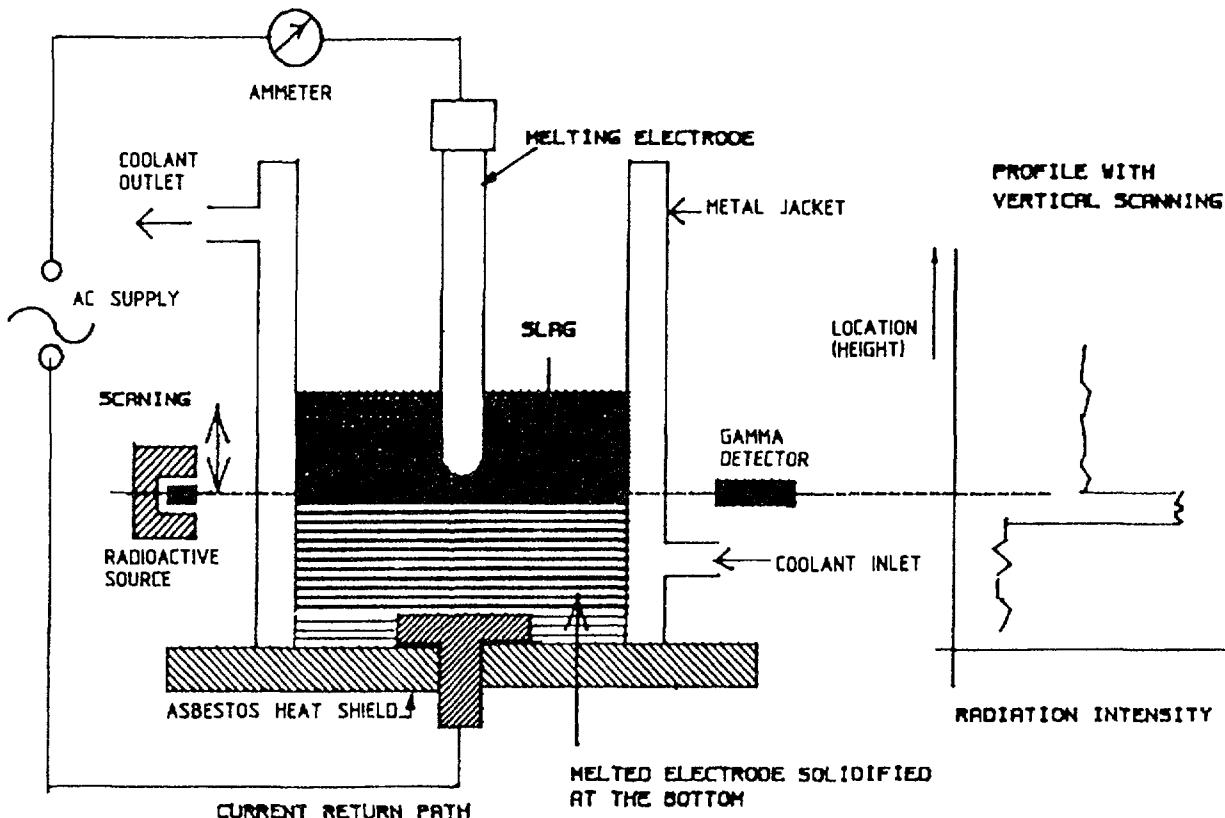


FIG. 2. Tracking nucleonic level control system.

speed. The cylinders were manufactured to very close tolerances. Hence correlation between the quantity of LPG and its level in the cylinder was precisely controlled. Under static condition, level alarm could be actuated at less than 0.5 mm liquid level change with 95% confidence. On a moving belt it gave the detection accuracy of 4 mm which corresponded to 150 gms for a 14.5 kg of liquid in the cylinder.

In another application an iron cask was to be filled with mineral powder under vacuum up to a precise location in the neck and sealed. A simple gamma absorption technique with a fine Cesium 137/5mCi beam of 3 mm diameter and a Geiger Muller (G.M.) tube, was used in this case.

A tracking nucleonic level control system was developed for a defence laboratory to maintain proper separation between molten metal and the melting electrode which were housed inside a water cooled metal casing. The Cobalt-60 radiation source and detector assembly were mechanically coupled to the electrode lifting mechanism, so that the beam passed through the slag just over the molten metal (fig. 2). As the electrode melted, the liquid level rose and the radiation path was obstructed which gave a signal for moving up the electrodes.

Technology Transfer Issues:

Based on these experiences, the development of standard low cost nucleonic control gauges was undertaken with the objective of transfer of knowhow to the industrial sector. Though self reliance is the key issue in many programmes of Government, the emphasis of industrial development is on manufacturing on mass scale, with brand name under collaboration and with good profit margin. The interaction between government, scientific research organization and industry needs to be strengthened. The fear of radioactivity and the rather high capital cost limit the demand for installation of nucleonic gauges in industrial plants. Radiation safety considerations impose stringent limits on the strength of radioactive source which is permissible in a specific application. This requires design for specific applications involving higher technical level of R&D interaction between user and supplier. Consequently it results in lower return on investment, for the industrial entrepreneur.

For the promotion of technology transfer of nuclear techniques and nucleonic control system, the following points merit attention.

1. Creating confidence in operators and public about the safe operation of nucleonic gauges in industry.
2. Highlighting of economic benefits which would accrue by the use of nucleonic gauges in process operations, by way of improved reliability, efficiency and ease of maintenance.
3. Simplification of rules for purchase and use of radioactive sources (preferably with single window clearance facility) and appropriate safety regulations.

Summary:

Electronics Division has transferred the technology of Gamma Switch with radioisotope source housing to an industrial entrepreneur having a good technical base. The technology has been successfully implemented in a chemical plant under expert supervision for non-contact level gauging application. Though the technology holds a good promise for future market growth, much effort is needed to remove misunderstanding about the dangers of this technology.

NUCLEAR TECHNIQUES IN THE OIL INDUSTRY

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Abstract

The major area of application of nuclear techniques in the oil industry is in the measurement of subsurface formation properties for discovering and evaluating hydrocarbons and to understand the formation geology for improved production and management of the reserves. Recent advances in these nuclear well logging measurements have provided a significant increase in the amount of information available for describing subsurface formations. A key element has been the routine introduction of gamma-ray spectroscopy measurements that can provide a significant number of elemental concentrations. When combined with nuclear geophysical measurements, geochemical descriptions of the subsurface environment can be obtained. On a much wider scale than could be previously considered, nuclear techniques can be applied to solving oil industry problems. The types of measurements that can be performed, their relationship to geological parameters, and the description of the equipment to perform the necessary measurements under borehole conditions are reviewed.

INTRODUCTION

Nuclear logging techniques, the use of instruments in a well to continuously measure subsurface geological formation properties, were first introduced in 1939 (*Howell and Frosch, 1939*), and, since then, they have come to play a dominant role in the analysis of subsurface formations. The development of nuclear techniques in the oil industry have been motivated by a desire to better understand the subsurface geology for the location and evaluation of oil and gas, and for better understanding geological processes (*Ellis et al., 1987*). The main interest was the location of porous zones containing hydrocarbons and free of clay minerals whose presence would inhibit fluid flow.

The earliest nuclear technique measured natural radioactivity, generally associated with clay minerals in sedimentary formations. It was immediately used to separate shaly from clean formations. Subsequent developments were related to the analysis of the fluids contained in sedimentary formations. A fundamental parameter for knowing how much fluid is present is the formation porosity. Nuclear measurements provided the first

quantitative measurements of *in situ* rock porosity. These measurements initially made use of gamma rays and neutrons from radioactive chemical sources. Gamma rays, primarily through Compton scattering, are used to determine the formation bulk density. Neutron transport, with the detection of either thermal or epithermal neutrons, has been used to determine porosity, since it is mainly influenced by hydrogen, usually associated with pore fluids. Following the development of pulsed accelerators (*Youmans et al.*, 1959; *Frentrop and Sherman*, 1960), other types of measurements became practical. The macroscopic thermal neutron absorption cross section could be measured by the time-dependence of neutron capture gamma rays following a pulse of high energy neutrons. With the advent of improved gamma-ray detectors, it became practical to perform spectroscopic analyses of the gamma-ray flux reaching a detector, initially for natural radioactivity and then for gamma rays produced by neutron interactions with nuclei in the rock. The spectroscopic techniques have been applied to bulk geophysical measurements and elemental analyses. The ability to detect specific elements, coupled with the improved understanding of the application of nuclear techniques in extended heterogeneous media, has introduced a new way of thinking about nuclear techniques. Rather than being able to make a measurement and trying to relate that measurement to a geophysical parameter, nuclear techniques have become an arsenal of measurements that can be brought to bear for the solution of complex problems, often in conjunction with other types of measurements, and not just for problems to be addressed by logging measurements.

Two basic constraints are placed on nuclear measurements by the borehole environment : geometrical and contamination. The geometrical constraints can be visualized most easily. All equipment must be contained in a cylindrical housing to withstand ambient pressures to 150 MPa (and temperatures to 150°C). The contamination of the measurements exists since wells are drilled with mud in the hole, which usually becomes part of the total measurement, and, since a pressure gradient is maintained between the hole and the surrounding rock, a mudcake is formed on the surface of the rock. In many circumstances, a steel casing is cemented into the well to insure that the borehole does not collapse.

Since sources and detectors must be contained in the pressure housing, a shielding problem exists to ensure that the detector does not significantly respond to radiation transmitted through the tool. The inclusion of large amounts of shielding is not simple, however, as detector counting rates decrease exponentially with source-detector spacing. Since measurements must typically be performed continuously at speeds of at least 5 cm/sec, maximum counting rates are needed to obtain statistically precise data. Furthermore, this geometry precludes the types of collimation typically used in the laboratory to localize the region of a reaction or to minimize multiple scattering effects. The logging measurement environment is illustrated in Figure 1 with a logging sonde having an arbitrary source, detector, and shielding in an irregular hole. The borehole contains a drilling mud. Often, a mudcake is formed on the surface of the rock.

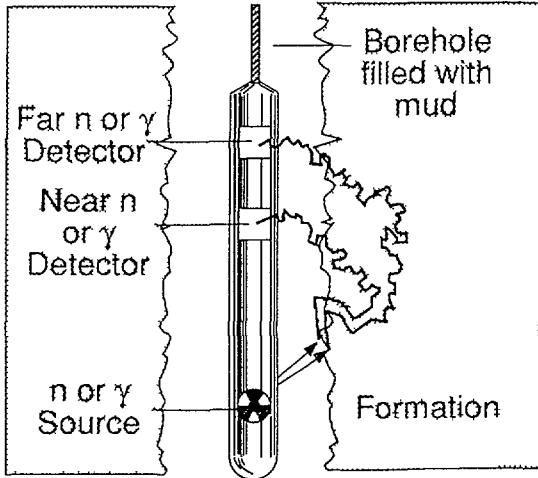


Figure 1. Schematic representation of a generic logging tool in the borehole. The borehole is typically filled with mud.

The presence of mud and mudcake introduces material into the measurement volume with significantly different properties than the rock. To obtain accurate measurements of rock or fluid properties, corrections must be made for the additional material. These corrections become so large in cased boreholes that some measurements are impractical. For tools with partial collimation, i.e. they do not have uniform circumferential sensitivity, the tool standoff from the borehole wall strongly affects the measured response.

The basic features of all nuclear logging measurements can be characterized by a gamma-ray or neutron source (except for natural activity measurements), shielding to minimize source particle transmission through the tool to the region of the detector, one or more detectors of neutrons or gamma rays, electronics to process the detector signals, and storage/transmission systems to buffer and transmit the data, through a cable, to surface instrumentation, typically today a computer in the logging truck. The most common sources of radiation are ^{137}Cs for gamma-gamma measurements, and AmBe, ^{252}Cf , or d,t accelerator (*Smith et al.*, 1988) sources for neutrons. Neutron detectors are usually pressurized ^3He detectors, sometimes shielded with a thermal neutron absorber to detect only epithermal neutrons. Most gamma-ray detectors are NaI(Tl), though, occasionally, other scintillators such as CsI and BGO have been used. Special high-temperature, ruggedized photomultipliers (*Pietras and Smith*, 1988) are used to collect the light from the scintillators and produce an electronic pulse proportional to the incident gamma ray energy. Linear and digital electronics are included in the logging sonde to analyze the detector output, control its stability, and, when an accelerator is used, control its operation. For research purposes, a number of germanium semiconductor detector sondes have also been used.

Nuclear logging techniques are used for determining geophysical and geochemical parameters. The actual quantities measured through nuclear techniques do not always correspond to the geological parameters desired, a difference that is important to understand. Geological parameters that are desired from nuclear measurements are generally: the type and amount of fluids, permeability, lithology, clay identification, and, more recently, mineralogy (*Herron*, 1986). Information on these parameters can be obtained with nuclear techniques that measure: gamma-ray transmission that depends on Compton and photoelectric cross sections, neutron transmission, dominated by elastic scattering from hydrogen, the macroscopic thermal neutron absorption cross section, and elemental analysis through gamma-ray spectroscopy of natural or neutron-induced radioactivity.

The measurements do not provide the exact information desired on the geological parameters. For example, the neutron transmission measurement, while dominated by the amount of hydrogen present, also depends on the elastic, inelastic, and reaction cross sections for all other elements present. Thus, additional information is needed to relate the measurements made with nuclear techniques to the desired geological parameters.

The difficulties in performing quantitative nuclear measurements in extended heterogeneous media required extensive calibrations under laboratory and field conditions. Recently, the use of Monte Carlo and other analytical techniques (*Ellis et al.*, 1987; *Soran*, 1988) have expanded the capabilities for performing quantitative analyses and provided insight into the physics of measurements in heterogeneous extended media. These techniques have become particularly important for quantitative elemental analysis.

Before discussing specific techniques in more detail, it is useful to appreciate some of the other nuclear techniques used in the oil industry. At the moment, some have had commercial application while others have been conceptualized or developed as a prototype for future use. An early technique was developed to monitor possible flow channels in the cement between the casing and formation rock (*Arnold and Paap*, 1979) through the use of an oxygen activation technique. While this procedure was initially instituted to solve hydrocarbon production problems, it has recently been applied to the problem of environmental monitoring to ensure a lack of contamination of surface water supplies (*Paap et al.*, 1989). Monitoring of applied radioactive tracers have been used to monitor fluid flows in pipes (*Hill et al.*, 1988) and to monitor the efficiency of hydraulic fracturing of the rock (*Lopus et al.*, 1987). Other applications have included the placement of radioactive markers in the rock to monitor subsidence from fluid production, surface systems to monitor the salt content of produced fluids (*Supernaw and Richter*, 1989), and a system that can be placed on the sea floor to monitor the flow rate of fluids produced from subsea wells (*Clayton and Spackman*, 1989). Perhaps an obvious new approach was the inclusion of nuclear measurement techniques in the area of measurements while drilling, initially natural radioactivity measurements (*Bryant and Gage*, 1988), and now including neutron and gamma-ray transport measurements (*Wright et al.*, 1989).

GEOPHYSICAL LOGGING MEASUREMENTS

Gamma-Ray Scattering

The measurement of density provides information on the amount of fluids present in the rock. The bulk density can be written as:

$$\rho_b = \phi \rho_f + (1 - \phi) \rho_{ma} \quad (1)$$

where the bulk density, ρ_b , can be related to the solid rock matrix density, ρ_{ma} , and the porosity, ϕ , which contains a fluid of density ρ_f . From equation (1), porosity can be determined from a bulk density measurement, if the matrix and fluid densities are known. These will only be known with precision if the fluid type and properties and the lithology are known. In practical terms, the density range of fluids is between 0.8 and 1.2 g/cm³ (although calcium chloride solutions may reach 1.4 g/cm³) and most rock matrix densities of interest are between 2.60 and 2.96 g/cm³.

In the energy region where the dominant mechanism for gamma-ray attenuation is Compton scattering, gamma ray transmission depends on the electron density, n_e , rather than ρ_b , the bulk density. The electron density is given by:

$$n_e = \rho_b (Z/A) N_A \quad (2)$$

where Z is the average atomic number, A is the average atomic weight, and N_A is Avogadro's number. For lower gamma-ray energies (<100 keV), the photoelectric cross section becomes significant. For geological material Z is typically < 28, so that gamma-ray energies are well above the K-absorption edge. In this energy region, the photoelectric absorption cross section per electron can be approximated by (Hubbell, 1969):

$$\tau = CZ^{3.6}/E^{3.15} \quad (3)$$

where C is a proportionality constant. The average photoelectric absorption cross section per electron for a mixture of elements is given by:

$$\tau = \sum_i \frac{Z_i m_i \tau_i}{A_i} / \sum_i \frac{Z_i m_i}{A_i} \quad (4)$$

where m_i is the mass fraction of the i th element.

A density logging sonde has a ¹³⁷Cs source and a gamma-ray detector which is well shielded from the source. The 662 keV gamma rays are below the threshold for pair-production and insensitive to significant photoelectric absorption. The gamma-ray intensity at the detector varies exponentially with the scattering material density. One explanation (Bertozzi et al., 1981) for this behavior considers the multiply-scattered gamma rays, detected far

from the source, to have undergone most of their scattering in the formation close to the detector. These multiply-scattered gamma rays are a virtual source, that consists of unscattered source-energy gamma rays which travel from the source, nearly parallel to the borehole wall, to reach the site of their last few collisions before detection. Their intensity depends on the probability of source gamma rays arriving at this site unscattered, and thus varies exponentially with formation density.

To compensate for the frequent occurrence of intervening mudcake, most devices (*Wahl et al.*, 1964; *Ellis et al.*, 1983) incorporate two detectors (generally both NaI) in a housing that shields them from direct source radiation. The device is forced up against the formation with a hydraulically-operated arm. This arm also measures the borehole diameter (along one axis). The counting rates of the detectors vary exponentially with the electron density of the formation. The formation density could be determined simply from an observed counting rate from either detector. However, when mudcake of unknown density and/or thickness is present on the borehole wall, each counting rate will be perturbed by an amount proportional to the product of the contrast in density, between the formation and the mudcake, and its thickness. This results in a discrepancy between the apparent formation density derived from each detector, which can be calibrated (*Ellis et al.*, 1983) into a correction to the apparent density measured by the farthest detector.

In modern density tools (*Ellis et al.*, 1983; *Minette et al.*, 1986), the shape of the low-energy portion of the scattered gamma-ray spectrum, which correlates with the formation photoelectric absorption parameters, is also measured. This photoelectric absorption

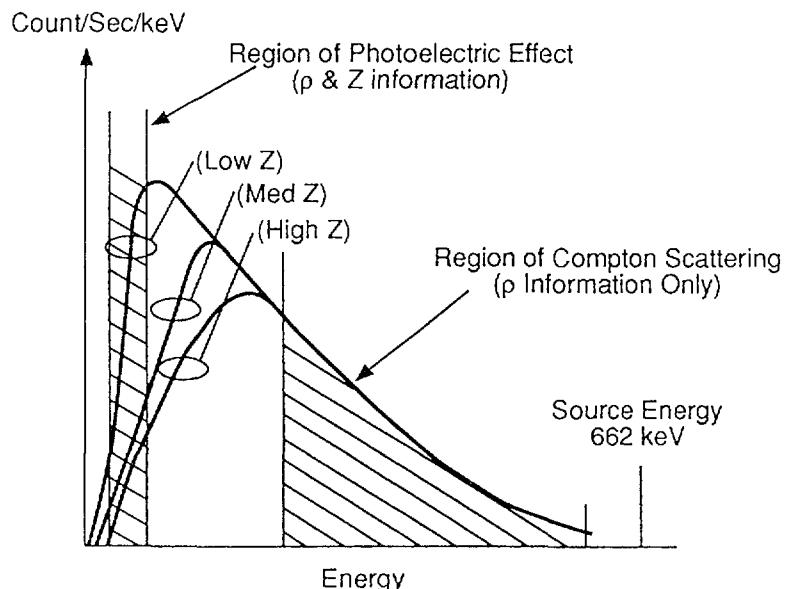


Figure 2. Schematic illustration of the behavior of the multiply scattered gamma-ray spectrum detected by a density and lithology logging tool. For illustration, the density of the formation is held constant but the average atomic number is varied. Photoelectric absorption changes the shape of the lower energy portion of the spectrum.

separates the three major types of rock due to their differing average atomic numbers. It is convenient to define a new parameter, P_e , the photoelectric index as

$$P_e = \frac{Z}{10}^{3.6}, \quad (5)$$

which is proportional to the photoelectric cross section per electron with the energy dependence suppressed. Since many geological analyses involve volume fractions, it is useful to consider the quantity U ($\propto n_e \tau$) whose scaling (*Gardner and Dumanoir, 1980; Bertozzi et al., 1981*) is analogous to equation (1), i.e.:

$$U = \phi U_f + (1 - \phi) U_{ma}. \quad (6)$$

Qualitatively, the gamma-ray spectra observed with a logging device with a window nearly transparent to low-energy gamma rays (such as Be) is shown in Figure 2. As the average atomic number (or P_e) of the formation increases, the lower-energy portion of the spectrum is progressively reduced. Thus, a measurement of this spectral shape at low gamma-ray energies can be calibrated as a function of P_e . For pure formations the values of P_e are 1.83 for sandstone, 3.1 for dolomite, and 5.1 for limestone.

Neutron Scattering

The spatial distribution of epithermal or thermal neutrons resulting from the interaction of high-energy neutrons with a formation can be related to its hydrogen content. If the hydrogen (in water or hydrocarbons) is only contained within the formation pore space, then the measurement yields porosity.

The conventional method of determining the neutron scattering properties of a formation employs a source of high energy neutrons and monitors the resultant thermal or epithermal flux. The sources generally used in such a measurement (RaBe or AmBe) are based on the α, n reaction. The neutron spectra are far from mono-energetic, but copious amounts ($\approx 10^8$ n/sec from ≈ 2000 GBq of Am) of neutrons ($\langle E \rangle \approx 4.5$ MeV) are produced. Recently, a comparable measurement has been proposed (*Gartner et al., 1986*) that uses a d,t accelerator source. Although numerous neutron detectors are available, in the hostile environment of well logging, simplicity and reliability are provided by the use of gas counters such as those based on the ${}^3\text{He}$ (n, p) reaction. To measure the epithermal neutron flux a shield of thermal-neutron absorbing material is used around the detector.

At the neutron source energy, the primary interaction mechanism is elastic scattering. In a limestone formation the mean-free-path of source neutrons is about 8 cm, practically independent of the water content of the formation. However, the effect of hydrogen is noted at lower energies. For example, at 100 keV the mean-free path ranges between 4 cm, in a pure limestone formation, and 2 cm in a 40% water-filled limestone. Thus, the

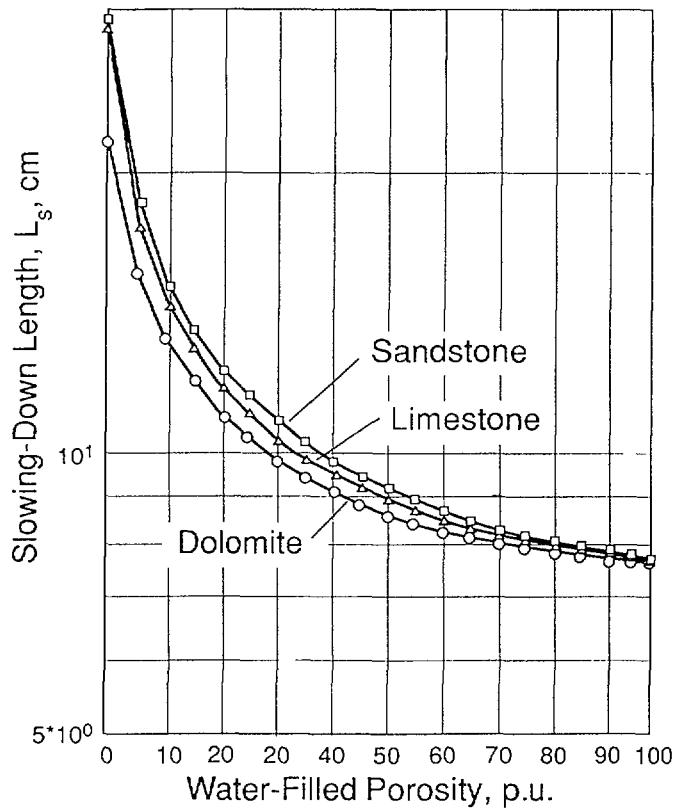


Figure 3. The slowing-down length of three common lithologies as a function of porosity.

mean-free-path depends on hydrogen content and the logging measurement can be viewed as a determination of the moderating power of the formation.

Neutron transport depends on two processes: the loss of energy from the source energy to thermal energies, and the diffusion of neutrons at thermal energies until they are captured. Allen et al. (1967) have shown that the epithermal neutron flux from a point source can be characterized by an exponential dependence on the distance from the source with a length parameter, the slowing-down length L_s . A simple model based on diffusion theory yields an epithermal neutron flux, Φ_{epi} , at a distance r from the source, of:

$$\Phi_{epi} \propto \frac{e^{-r/L_s}}{D_{epi} r} \quad (7)$$

where D_{epi} is an epithermal diffusion coefficient related to the transport mean free path. The variation of the slowing-down length as a function of water-filled porosity in three types of formations is shown in Figure 3.

For thermal neutrons, there is a comparable length parameter, the diffusion length L_d , such that the thermal neutron flux can be represented as:

$$\Phi_{th} \propto \frac{L_d^2}{D(L_s^2 - L_d^2)} \frac{e^{-r/L_s} - e^{-r/L_d}}{r} \quad (8)$$

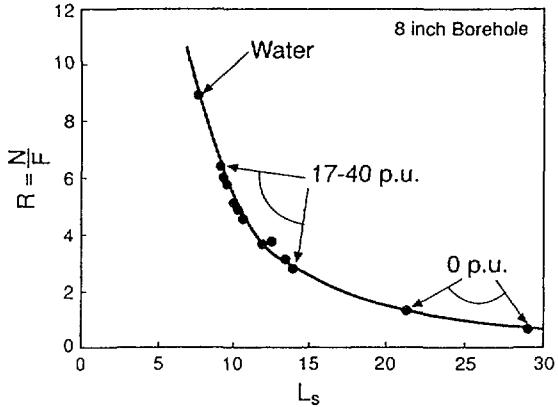


Figure 4. Laboratory data from an epithermal neutron porosity device in rock formations over a wide range of porosity. The ratio of near to far detector counting rates is shown as a function of the appropriate slowing-down length of the test formations.

where D is the thermal diffusion coefficient. D can be calculated from the thermal neutron cross sections of the elements in the material and is related to L_d by $D = L_d^2 \Sigma$ where Σ is the macroscopic thermal neutron absorption cross section for the material.

The preceding discussion indicates that a calculable parameter, L_s , will be adequate to represent the flux distribution in the formation for the case of a point source in an infinite medium. However, this is not the typical logging measurement made in a roughly cylindrical fluid-filled hole. Some of the physical parameters of the formation and borehole which affect the response of a neutron porosity device include: porosity, pore fluid type, salinity, rock type, borehole size, borehole fluid type, mudcake, and pressure. Of these, only porosity is the desired quantity. All the others change the local hydrogen density and thus influence a measurement that is sensitive to the concentration of hydrogen.

Figure 1 indicates the configuration of a typical neutron scattering device in the borehole. A first approximation to reducing the influence of the hydrogen-rich borehole on the measurement of the slowing-down properties of the formation is the use of two epithermal detectors. By comparing the counting rates of two detectors at different distances from the source, the shape of the epithermal flux distribution, which is more indicative of the slowing-down length of the formation, is obtained.

Figure 4 shows the ratio of the counting rates (near detector divided by far detector) of an experimental epithermal neutron scattering device as a function of the slowing-down length for some laboratory formations. All the formations contain a simulated drill hole of 20.3 cm diameter. There is some residual influence on the measured ratio for different borehole sizes, but this effect can be quantified by measurements in formations of different borehole sizes or by numerical simulation. These curves allow a determination of slowing-down length after correcting for the borehole diameter. Normally, the porosity

is determined from the slowing-down length in a known rock type by using charts such as Figure 3. Uncertainty in the rock type may introduce variations of several porosity units. In addition, important quantities of hydrogen may occur in formation rock, most prominently the hydroxyl radicals associated with clay minerals. Depending on the clay type, the apparent formation porosity can attain values of up to 60%, when in fact no fluid is present.

The use of epithermal neutrons is a fairly uncomplicated way of determining the formation hydrogen content. Historically, thermal neutron detection has been used due to the much higher counting rates which can be obtained from a given source activity.

Equation (8) suggests that the perturbation due to Σ may be eliminated by placing the detector at a distance $r \gg L_d$, where the spatial thermal flux distribution is determined by L_s . This has been confirmed by Allen et al. who have shown that, by taking the ratio of the counting rates of two thermal neutron detectors spaced sufficiently far from the neutron source, essentially all of the dependence on D , L_d , and source output can be eliminated. However, in practical neutron logging devices, based on Am-Be sources, this thermal flux distribution may not be generally realized due to insufficient counting rates at the distances required. In the future, neutron logging devices based on accelerator sources of neutrons may provide a sufficient neutron flux to achieve this condition. As in the case of the epithermal device, a pair of thermal neutron detectors is used to determine the gradient of the thermal flux.

To characterize the laboratory data of the thermal neutron tool, which are at some variance with the predictions of equation (8) due to the residual sensitivity to Σ , use is made of the migration length, L_m , ($L_m^2 = L_s^2 + L_d^2$), to account explicitly for the additional perturbation of neutron diffusion and absorption in the formation. To first order, representation of the tool response as a function of migration length allows the conversion of the measurements to porosity for given conditions of lithology and salinity (Ellis, 1986).

SPECTROSCOPIC MEASUREMENTS

While the previous measurements use a form of spectroscopy, they primarily are based on separating out a portion of a total energy spectrum, e.g. thermal neutrons or epithermal neutrons. However, truly spectroscopic measurements break the spectrum of interest into many individual regions to establish specific character in the total spectrum, e.g. gamma rays arriving from particular elements. The measured spectrum can be either a time spectrum of events or an energy spectrum of detected particles.

Two measurements have used the detailed time dependence of a detected signal: the measurement of the thermal neutron decay time and the measurement of the slowing down time of neutrons by the detected count rate of epithermal neutrons. Both measurements require a pulsed accelerator to provide a relatively narrow burst of 14 MeV neutrons.

Measurements of the energy spectrum of detected gamma rays were first applied to the detection of natural radioactivity from potassium and the thorium and uranium decay series. Subsequently, neutron sources were coupled with energy-sensitive detectors to observe gamma rays from neutron-induced reactions. Today, gamma-ray logging measurements are being used to detect gamma-rays from inelastic neutron scattering, prompt thermal neutron capture, and delayed activity from unstable nuclei (generally produced by thermal neutron capture reactions), in addition to natural radioactivity.

Time-Dependent Measurements

Thermal neutron decay time. The macroscopic thermal neutron absorption cross section Σ is the total capture cross section per cm^3 . For a molecule $A_\alpha B_\beta$, $\sigma_{\text{mol}} = \alpha\sigma_A + \beta\sigma_B$ where $\sigma_{A(\text{or } B)}$ is the microscopic thermal neutron absorption cross section for a nucleus A(or B). For a collection of molecules, the macroscopic cross section is given by:

$$\Sigma = \sum_i \frac{V_i \rho_i \sigma_{\text{mol}(i)} N_A}{M_i} \quad (9)$$

where M_i is the molecular weight of the i th type of molecule with volume fraction V_i . Frequently, Σ is expressed in "capture units" (cu). The capture unit is 1000 times the macroscopic cross section (expressed in cm^{-1}). In these units water has a capture cross section of 22 cu and the pure, common reservoir rocks (quartz, calcite, and dolomite) are in the range of 4-7 cu. The addition of NaCl to formation water, or the presence of impurities such as boron, leads to a rapid rise in the formation cross section, which, in practical cases, ranges from 10-80 cu. The value of Σ depends on the chemical constituents of the rock matrix and the pore fluids, whose absorption cross section is usually dominated by chlorine. Thus, this measurement can identify the formation fluid, even in wells containing metallic casings where electrical measurements are useless.

A logging tool to measure Σ requires a pulsed neutron source and a gamma-ray detector. The source of 14 MeV neutrons is pulsed for a brief period ($\approx 200 \mu\text{sec}$), forming a cloud of high energy neutrons in the borehole and formation which becomes thermalized through repeated collisions. The neutrons are captured at a rate which depends on the thermal neutron absorption properties of the formation and borehole. The decay of the capture gamma-ray counting rate reflects the decay of the neutron population.

The reaction rate for thermal neutron absorption is given by the product of Σ and the velocity of the neutron, v . The number of neutrons remaining at time t is:

$$N(t) = N(0)e^{-\Sigma vt}. \quad (10)$$

This simple analysis of the capture gamma-ray time-dependence misses an important aspect of actual measurements: the thermal neutron diffusion effect. At any observation point the local thermal neutron density decreases because the neutrons are diffusing as well as being captured. To quantify the diffusion effect on the local decay time constant requires the time-dependent diffusion equation (*Wahl et al.*, 1968). The apparent decay time of the local neutron population in an infinite medium has two components:

$$\frac{1}{\tau_a} = \frac{1}{\tau_{int}} + \frac{1}{\tau_{diff}}, \quad (11)$$

where τ_{int} is the intrinsic formation decay time (i.e. that expected from absorption alone) and τ_{diff} is the diffusion time, which depends on the distance from the source and the thermal diffusion coefficient. The apparent Σ of a formation is greater than the intrinsic value due to the diffusion rate of thermal neutrons near the detector.

As in most logging measurements, the measurement includes both borehole and formation signals. Since the borehole component is of little interest for determining formation properties, techniques which decompose the capture gamma ray time decay into formation and borehole components and correct for diffusion effects have been developed (*Schultz et al.*, 1983; *Steinman et al.*, 1986)

In the simple case of a single mineral, the measured formation Σ , after correcting for the borehole and diffusion effects, consists of two components, one from the matrix and the other from the formation fluid,

$$\Sigma = (1 - \phi)\Sigma_{ma} + \phi\Sigma_f. \quad (12)$$

To determine the water saturation, S_w , (i.e. the fraction of the pore volume ϕ containing water), the fluid component is broken further into water and hydrocarbon components:

$$\Sigma = (1 - \phi)\Sigma_{ma} + \phi S_w \Sigma_w + \phi(1 - S_w)\Sigma_h. \quad (13)$$

The Σ_h of hydrocarbon is about 20 cu, nearly the same as fresh water. The ability of this measurement to distinguish hydrocarbon from water depends on the water salinity which makes Σ_w significantly larger than Σ_h . The presence of shale, which may contain thermal absorbers such as boron, seriously affects this simple interpretation scheme, by making *a priori* knowledge of Σ_{ma} questionable, but several methods for dealing with this problem have been developed.

Pulsed neutron porosity. Previously described neutron scattering measurements have been based on a measurement of the slowing down length. Recently, a neutron scattering measurement that is based on the determination of the time it takes neutrons to slow down to epithermal energies has been proposed (*Mills et al.*, 1988). To measure the epithermal neutron die-away obviously requires a pulsed source of neutrons, achieved in this case with a standard d,t accelerator. To determine epithermal neutrons, a 0.635 cm diameter, 10 atmosphere ^3He detector is surrounded by a thin (.0152 cm) gadolinium foil to absorb the thermal neutrons before they can reach the detector.

The motivation for measuring the die-away rate of epithermal neutrons is to obtain a quantity that is more strongly correlated with the hydrogen concentration. Measurements of slowing down length depend mostly on the distance traveled during the first few collisions of high energy neutrons, where elements other than hydrogen have a significant impact. The measurement of the die-away rate is dominated by the last few neutron interactions at low energies, where the neutrons are traveling at low velocities. At these energies, the interactions are more strongly dependent on the hydrogen content.

Initial studies indicate that the die-away rate has a substantially reduced dependence on the lithology of the rock (*Mills*, 1988, Figure 3) than does the slowing down length measurement (see Figure 3). In addition, the detection of epithermal neutrons eliminates the dependence of the results on the macroscopic thermal neutron absorption cross section, as is also the case for the slowing down length measurement.

Energy-Dependent Measurements

The spectrum of detected gamma rays was first decomposed to obtain the individual components of natural activity. Subsequent spectrometers were coupled with neutron sources to determine elemental content from gamma rays produced in thermal neutron capture reactions and inelastic scattering of high energy neutrons. Typically, the results were qualitative, as important difficulties existed in converting the count rate of detected gamma rays into elemental concentrations of the elements producing the gamma rays.

Inelastic scattering has been used primarily to determine the content of carbon and oxygen (*Lock and Hoyer*, 1974; *Hertzog*, 1980; *Oliver et al.*, 1981). Most other elements are more efficiently detected through other types of reactions (*Schweitzer et al.*, 1987). Thermal neutron capture reactions have been mostly used in the oil industry to determine the major constituents of sedimentary rocks (*Flaum and Pirie*, 1981), with some recent applications in crystalline rock (*Grau et al.*, 1988). Delayed activity, produced by a ^{252}Cf source, has been routinely used to determine Al concentrations and has also been used for the measurement of V concentrations (*Schweitzer and Manente*, 1985). Current and future

efforts for more complete elemental analysis will undoubtedly use delayed activity from both isotopic sources and 14 MeV accelerator-produced sources (*Hertzog et al.*, 1986; *Hertzog et al.*, 1987b).

Most of the initial spectroscopic measurements were aimed at measuring the content of one or only a few elements, frequently in only a relative sense (*Hertzog*, 1980). A recent advance has been the ability to extend the measurements to a relatively large set of elements, and to account for the absolute unknowns in the physics of the measurements and the elements that are not measured to provide the content of the measured elements in weight percent. The combination of three gamma-ray spectrometers (*Hertzog et al.*, 1987a) provides elemental concentrations as a continuous function of depth for ten elements: Si, Ca, Fe, S, Ti, Gd, Al, Th, U, and K. This set of elements provides valuable information on the nature of the rock matrix, information which could previously be obtained only through discrete core analyses.

The spectrometers include measurements of natural activity for determining Th, U, and K concentrations, measurements of delayed activity induced by thermal neutron capture for determining Al concentrations, and measurements of prompt gamma rays following thermal neutron capture for determining Si, Ca, Fe, S, Gd, and Ti concentrations.

The quantitative determination of aluminum concentration makes use of a ^{252}Cf source to create ^{28}Al delayed activity through the $^{27}\text{Al}(\text{n},\gamma)$ reaction. To determine the absolute elemental concentration of aluminum from the measured count rate involves many geophysical parameters. Borehole and formation Σ , slowing-down length, bulk density, logging speed, and borehole size are all required to determine the correspondence between counting rate and aluminum content in the rock. In addition, it is necessary to ensure that the source strength and the efficiency of the detector have been carefully calibrated. After correcting the count rates for the speed of the logging tool during the measurement, the weight percent of aluminum, η_{Al} , is given by:

$$\eta_{\text{Al}} = \frac{N \Sigma_{\text{bh}}^{25} (\Sigma_{\text{form}} + 14.5)(L_s + 12.3)^2}{\rho (1 + 3.64e^{-\text{DIA}/13.9})} \quad (14)$$

where N is the background-subtracted, velocity-corrected counting rate and DIA is the borehole diameter in cm. Thus, a proper determination of elemental concentrations requires reliable information on geophysical parameters as well as the specific spectroscopic information related to the individual element. These factors also determine the accuracy and precision of the measurement (*Schweitzer et al.*, 1988). Because of the importance of knowing the accuracy and precision of elemental concentration measurements, recent efforts have begun (*Grau et al.*, 1990) to understand the uncertainties for elemental determinations obtained from thermal neutron capture measurements.

The goal of spectroscopic logging measurements providing elemental concentrations had been previously realized for only certain types of spectroscopy, such as natural activity measurements (*Serra et al.*, 1980), or in limited types of environments (*Senftle et al.*, 1978; *Clayton and Coleman*, 1985). A model has been developed (*Hertzog et al.*, 1987b; *Grau et al.*, 1989) for broad applicability to a diverse set of geological environments. The model, intended for general use in clastic sediments, is a combination of oxide and carbonate representations that relates the concentration of unmeasured elements to the concentration of measured elements. Only elements contained in the rock matrix, excluding those present in pore fluid or in borehole fluid, are included in the analysis. This eliminates the need to properly separate the (generally uninteresting) content of the elements in the borehole or the (possibly varying) content of the elements in the pores of the rock. The model is most applicable to open hole conditions, where there is no need to deal with the extra Fe, Ca, and Si present in casing and cement.

The model is a simple closure relation and can be expressed mathematically as:

$$F \left\{ \sum_i X_i \frac{Y_i}{S_i} \right\} + X_K W_K + X_{Al} W_{Al} = 1 \quad (15)$$

where:

- F is the calibration factor to be determined at each depth from this equation,
- Y_i the fraction of the measured prompt gamma rays that are attributed to element i,
- S_i the relative weight-fraction detection sensitivity for element i,
- X_i the ratio of the weight of the associated oxide or carbonate to the weight of element i,
- W_{Al} the weight fraction of aluminum determined from the activation measurement, and
- W_K the weight fraction of potassium determined from natural activity.

The sensitivities, S_i , are measured in the laboratory and contain most of the nuclear physics involved in the relationship between the gamma-ray yields, Y_i , and the respective elemental concentrations. The yields, Y_i , are determined (*Grau and Schweitzer*, 1987) from a least-squares fit of the measured spectra with elemental standard spectra. Thus, if the elements needed to describe 100% of the rock minerals are indeed being measured, then the only unknown, F, can be determined trivially from the above equation. The terms FY_i/S_i are then the weight percent of the elements determined through prompt gamma ray thermal neutron capture measurements.

The simplicity of this approach can best be illustrated by considering the details needed to directly calibrate an elemental concentration measurement. The calibration of the de-

layed Al activation measurement requires a laboratory formation of known Al content as an absolute standard, an on-location shop calibration procedure to account for tool-to-tool variations, and a field environmental correction algorithm, equation (14), to account for variations in neutron and gamma-ray transport introduced by the large medium of investigation. These procedures insure that gamma rays detected from $^{27}\text{Al}(\text{n},\gamma)^{28}\text{Al}$ reactions with formation Al nuclei will be properly transformed into formation Al concentrations. However, there are two other sources of ^{28}Al gamma rays which contribute unwanted background to the measurement: silicon in the rock and aluminum in the mud.

The ^{252}Cf source used for the delayed activation measurement has a relatively low mean energy of 2.35 MeV, but a significant fraction of neutrons are emitted above the 3.9-MeV threshold energy for the $^{28}\text{Si}(\text{n},\text{p})^{28}\text{Al}$ reaction. The measured Al activity will then contain some amount due to the presence of Si in the rock. A correction must be made to the measured Al count rate before converting it into a concentration of Al. If the Si measurement were directly calibrated in weight percent, this correction would be trivial. However, since the Si weight percent is determined through the closure model, which uses the weight percent of aluminum, it is first necessary to estimate the Si concentration. Fortunately, for even the most siliceous rock, only a small equivalent weight percent of Al is due to the Si. Thus, a good estimate of the Si weight percent can be obtained by assuming all of the measured Al is due to the $^{27}\text{Al}(\text{n},\gamma)^{28}\text{Al}$ reaction and then determining the weight percent of Si through the closure model. This concentration of Si yields a

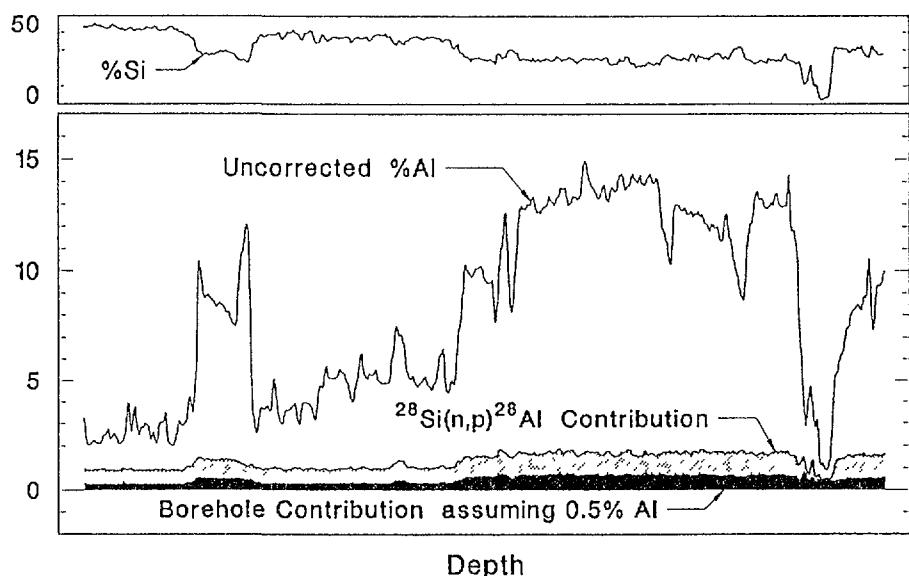


Figure 5. The total measured contribution of the delayed activity from ^{28}Al showing the contributions from Al present in the borehole and the contribution from the $^{28}\text{Si}(\text{n},\text{p})^{28}\text{Al}$ reaction produced by the neutron flux in the formation with a ^{252}Cf source and with a rock Si concentration as shown in the upper curve.

correction for the Al concentration, and this new value for Al concentration can be used in the closure model to provide the correct normalization for the elements determined from the capture spectrum. Figure 5 shows the portion (in light shading) of the apparent Al weight percent which is due to the $^{28}\text{Si}(\text{n},\text{p})^{28}\text{Al}$ reaction for a typical section of a well. In shaly sands the contribution is typically around one weight percent. It does not track the Si concentration, as well as one might first suspect, because the thermal-neutron environmental corrections, which have been applied to the raw count rates, are not appropriate for the fast neutrons which produce the (n,p) reaction, and therefore these corrections must be backed out in determining the (n,p) contribution.

The second problem for delayed aluminum activation measurements in typical boreholes is the presence of the mud, which will frequently contain Al-bearing minerals. The measurement cannot separate the Al in the mud from that in the rock, thus it is necessary to correct the measured Al concentration for the contribution from the mud. The needed parameters are the Al content of the mud, determined from the input constituents of the mud system or, preferably, from a measurement of the Al content in a sample taken from the mud circulation system, and the size of the borehole. The dark shaded area in Figure 5 is a calculation of a hypothetical borehole contribution assuming a 1.2 gm/cm^3 mud containing 0.5% Al by weight: a concentration which is perhaps on the high side, but not uncommon. While the calculated contribution for this constant borehole Al concentration varies considerably with the environment, a good rule of thumb would be that the borehole Al contribution to the apparent formation Al weight percent is nearly equal to the weight percent Al in the borehole fluid.

OTHER MEASUREMENTS

The success of logging measurements and the improved understanding of the physics of these measurements resulted in considerations of how these techniques might be applied to solving problems, other than the traditional ones of logging measurements of geophysical parameters for the discovery of hydrocarbons. After the introduction of pulsed accelerators, a technique to identify flow behind casing was instituted by activating the oxygen contained in water through the $^{16}\text{O}(\text{n},\text{p})^{16}\text{N}$ reaction (*Arnold and Paap, 1979*). If there are channels in the cement between the casing and the formation the oxygen would flow upwards through these channels and the count rate at a spaced detector, from the 7.13 sec half life, would be measured. A similar technique can be applied to measure water flow in the casing. A somewhat more general approach can be applied to the measurement of flow in a casing by using radioactive tracers (*Hill et al., 1988*). In this case the radioactive tracer is injected into the flow, in either a water-soluble or oil-soluble form, to allow the monitoring of either oil or water flow rate. This technique can be applied

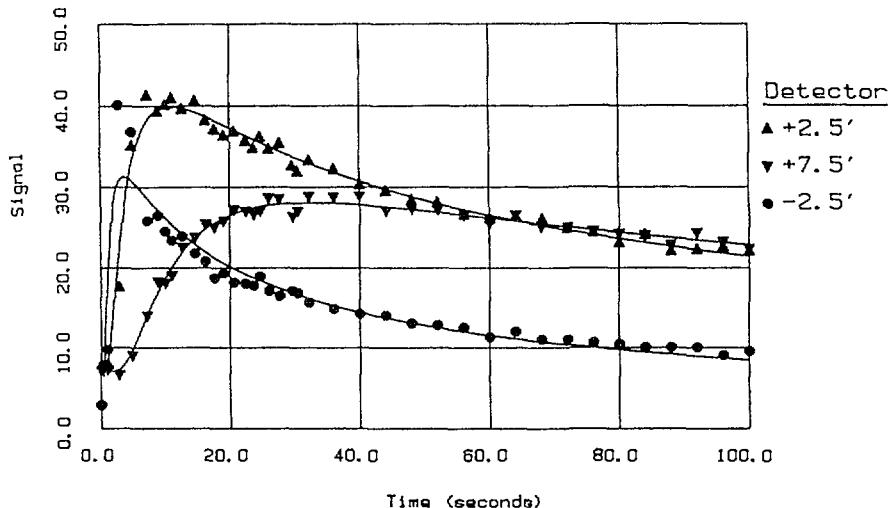


Figure 6. The individual points show the count rate at each detector from the injection of a radioactive tracer into the flow of a producing well. The solid curves are a fit to the expected time response.

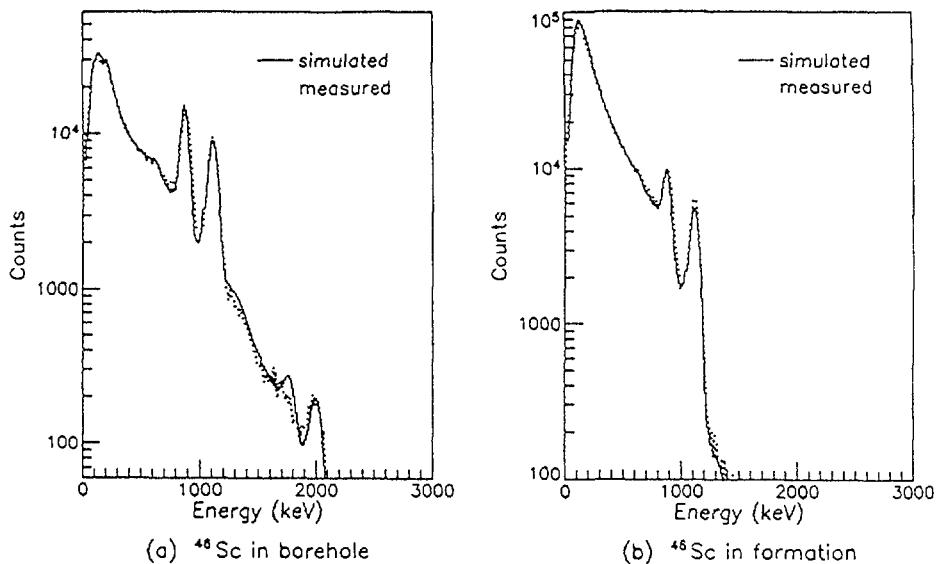


Figure 7. Comparison of measured and numerically simulated spectra of a ^{46}Sc tracer: (a) when the tracer is in the borehole fluid and (b) when the tracer is in the formation (Gartner *et al.*, 1990).

to either the problem of injecting fluid in injection wells or that of producing fluids in a production well. Following the injection of fluids, the detector count rates are monitored as a function of time, as shown in Figure 6 (Hunt, 1989). As environmental protection has grown to be of greater concern, the integrity of isolating ground water supplies from wells that penetrate the supplies has increased in importance. The same techniques for identifying flow behind casing have been applied to ensuring the integrity of ground water supplies (Paap *et al.*, 1989).

As spectrometers evolved, the practicality of using multiple radioactive tracers and spectroscopically decomposing the contribution of each component has resulted in the use of these techniques to solve more complicated problems. Thus, radioactive tracers in conjunction with spectroscopic detection have been used to determine the efficiency of hydraulic fracturing (*Lopus et al.*, 1987), to determine the efficiency of different stages of gravel packing, and monitoring the diameter and vertical extent of a cement column (*Gadeken et al.*, 1989). Of particular use in better applying these techniques was the recognition that some information on the distance the tracer is radially from the tool can be obtained by analyzing the shape of the detected spectrum (*Gadeken and Smith*, 1989) since gamma rays originating near the tool have higher photopeak to Compton ratios as they have had less opportunity to scatter in intervening material. This effect is clearly seen in Figure 7 (*Gartner et al.*, 1990). The spectra from ^{46}Sc have dramatically different peak-to-Compton ratios when the tracer is in the formation than when it is in the bore-hole adjacent to the logging tool. The solid curves in the figures are simulations of the response based on Monte Carlo calculations.

The fluid contents from producing wells can also be addressed by measurements using nuclear techniques once the fluid has reached the surface. A novel concept for measuring flow rate has been described (*Clayton and Spackman* in which flow rate measurements are desired at the sea floor for off shore producing wells. In this technique sea water is drawn into a chamber, activated with a neutron source, injected at particular time intervals into the flow stream of the producing pipe and subsequently detecting the tracer decays to determine the pipe flow velocity. Other nuclear techniques such as accurate fluid density measurements also have applications in the monitoring of fluid contents from a producing well. To minimize handling and processing problems of crude oil, it is important to minimize its salt content. A system has been developed (*Supernaw and Richter*, 1989) that monitors chlorine and sulfur content in streams of crude oil. The system can determine chlorine to about 20 ppm and sulfur to about 0.25neutron capture gamma rays.

Perhaps the most obvious modification of wireline logging techniques has been to measurements while drilling. This refers to the instrumentation of sections of the drillpipe to perform geophysical measurements while a well is being drilled. While the mechanical and power constraints for such measurements are severe (*Tanguy and Zoeller*, 1981), it was natural to include basic wireline logging measurements where practical. At present, natural radioactivity measurements (*Bryant and Gage*, 1988) and measurements of gamma-ray and neutron transport (*Wraight et al.*, 1989) have been achieved. The primary advantage of measurements while drilling is the minimal deterioration of the borehole and invasion of fluid and particles into the formation at the time of the measurement, hopefully providing a better measurement of intrinsic formation properties.

INSTRUMENTATION AND DATA ANALYSIS

Sources

Gamma-ray sources have only been used for density (and photoelectric cross section) measurements. For these measurements, the source should have an energy where the transport is dominated by Compton scattering. Thus, the source should have an energy greater than a few hundred keV and not more than an MeV. While there are a number of radioactive sources that produce gamma rays in this energy range, almost all gamma-ray scattering devices use ^{137}Cs ($E_{\gamma} = 662 \text{ keV}$). Even measurements which attempt to measure the photoelectric cross section typically use ^{137}Cs to achieve a significant counting rate that is sensitive to more than just the rock surface. Recently, an experimental gamma-ray scattering device has been demonstrated that is based on an electron linear accelerator (*King et al.*, 1987). This type of source accelerates a beam of electrons, in this case to a few MeV, that is incident on a solid target, producing a continuous energy spectrum of X-rays with the maximum energy corresponding to the electron beam energy. Since the X-rays produced are preferentially directed in the direction of the electron beam, the flux into the formation is maximized by having the target off-axis to the tool axis and bending the beam outward just before it strikes the target. Unlike a chemical source whose output is constant, a linear accelerator output varies with changes in the environment, primarily due to induced variations in electron energy and current (*Boyce et al.*, 1986), requiring a careful monitoring of the beam parameters.

Both isotopic and accelerator sources of neutrons have been commonly used. The different properties of these sources governs the source selection for a particular application. Isotopic sources, such as ^{252}Cf or $^{241}\text{Am-Be}$, have been generally used for neutron transport and thermal neutron capture reactions or delayed activation produced by (n,γ) reactions (*Senftle*, 1980). The relatively low neutron energies of these sources (2.5 - 4.5 MeV) make them impractical to use for (n,n') or (n,x) reactions. Compact pulsed accelerators that produce significant fluxes of 14 MeV neutrons from the $^2\text{H}(^3\text{H},n)^4\text{He}$ reaction allow sufficient yields to be obtained from (n,n') and (n,x) reactions. Since the 14 MeV neutrons are also moderated to thermal energies, the pulsed nature of the accelerators allow for multiple spectral measurements, gated at different times relative to the neutron burst, to heighten sensitivity to gamma rays produced by different types of reactions.

Accelerators used for logging measurements must be small, compact, sealed devices for operating in a logging sonde (*Smith et al.*, 1988). Regardless of the specific type of accelerator, there are some common features to all of them. High voltage must be generated internally to accelerate ions from an ion source to a target, usually a material such as scandium, that will successfully store a high density of the target species, e.g. deuterium and/or tritium. The gas must be stored in a source, such as a tungsten filament wrapped

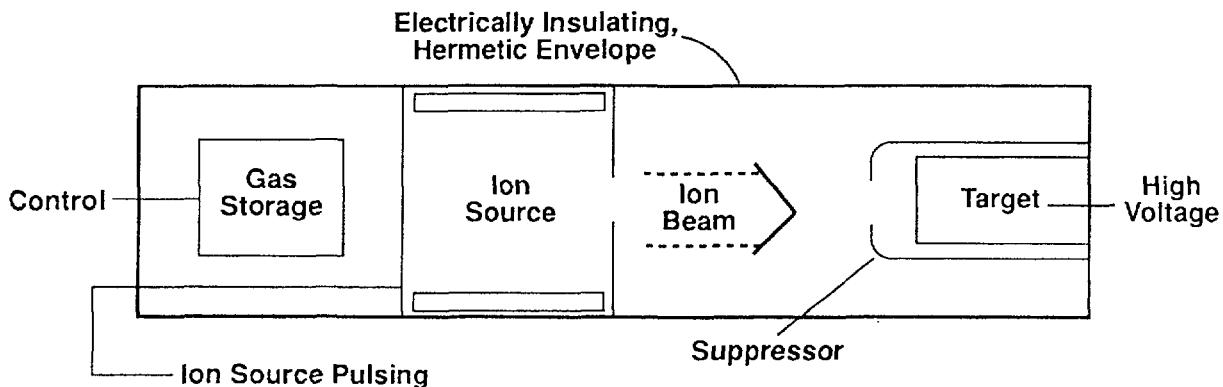


Figure 8. Schematic representation of logging accelerator for producing 14 MeV neutrons from the d,t reaction. The d and/or t gas is released from a storage medium, ionized in the ion source and accelerated to a target material containing d and/or t nuclei.

with zirconium, to maintain a good vacuum and produce the atoms to be ionized in the ion source. The beam of ions is accelerated through a suppressor, to allow for beam current monitoring, and strikes the target. These key elements of a logging accelerator are shown in Figure 8. Typical sizes for these accelerators can be as small as 2.5 cm in diameter and about 10 cm long. Under normal conditions, outputs of about 10^8 n/sec are obtained from the d,t reaction, with accelerating voltages of about 100 keV. These accelerators can be run either continuously or pulsed at rates of up to a few tens of kHz.

Detectors

Neutron detection is almost universally performed with pressurized ${}^3\text{He}$ proportional counters (*Glesius and Kniss, 1988*). When epithermal neutron detection is desired, the counters are surrounded by a thin layer of a thermal neutron absorbing material such as cadmium. The ${}^3\text{He}$ absorbs an incoming thermal neutron and produces a proton and a triton with about 765 keV of energy. These charged particles lose their energy to the counter gas by ionization. As the electrons drift to the anode wire, they encounter high electric fields, resulting in extensive ionization of neutral atoms. This multiplication process is particularly important in the logging environment, where normal shock and vibration produce microphonics in the counter. Temperature effects on the sensitivity and charge production of these counters are tied to the thermal expansion of the counter and its components. These effects are sufficiently small to maintain an accurate response over the entire range of temperatures encountered in logging.

A great deal of effort has been placed on optimizing gamma-ray detectors for spectroscopic measurements. Most spectrometers have been based on inorganic scintillators, primarily

$\text{NaI}(\text{Tl})$, though Ge detectors have also been used (Ellis et al., 1987, references 13-19 and 55), primarily for research purposes. Spectroscopic measurements based on inorganic scintillators are limited to detecting the most significant spectral contributions (*Grau and Schweitzer, 1987*) because of their inherent energy resolution. Ge detector-based spectrometers, with dramatically improved energy resolution, are capable of determining many more elements, including many that contribute only weakly to the total spectrum.

Since the primary focus of the research germanium spectrometers have been on the detection of one or a few specific elements and the spectrometers are not available for routine use, this discussion will focus on the applicability of inorganic scintillators to subsurface measurements. However, certain semiconductor detector properties must be known as a function of temperature for operation in a borehole environment (*Tanner et al., 1978*).

A gamma-ray detector in the laboratory is considered to have invariant properties. The environmental conditions of a borehole result in detector properties that continually vary (*Melcher and Schweitzer, 1988*), primarily because most detector properties vary with temperature. The most important of these temperature-dependent properties for inorganic scintillators are the light output, the decay time, and the emission spectrum. In addition, the logging environment requires maximum volumetric detection efficiency, gamma-ray detection in a neutron flux, and coupling with special photomultiplier tubes whose spectral response is narrower than for normal laboratory tubes. The light output, or scintillation efficiency, of almost all inorganic scintillators decreases with increasing

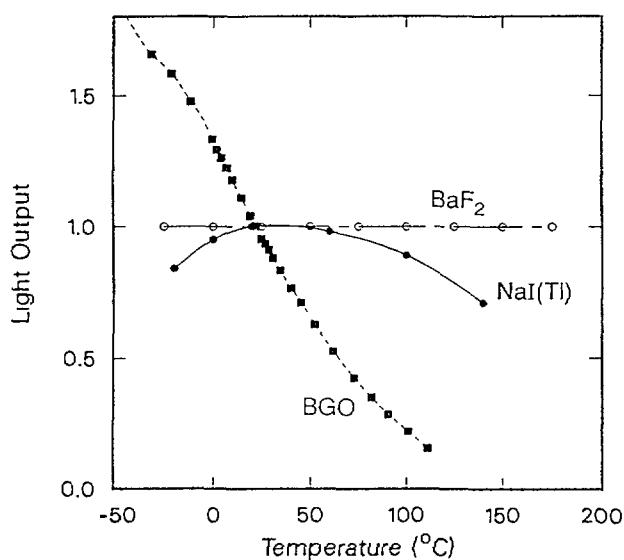


Figure 9. Temperature dependence of the scintillation emission of three scintillators. In the case of BaF_2 , the behavior of the 220 nm component is shown; the 325 nm component (not shown) is temperature dependent. The output of each scintillator is normalized to unity at 22°C.

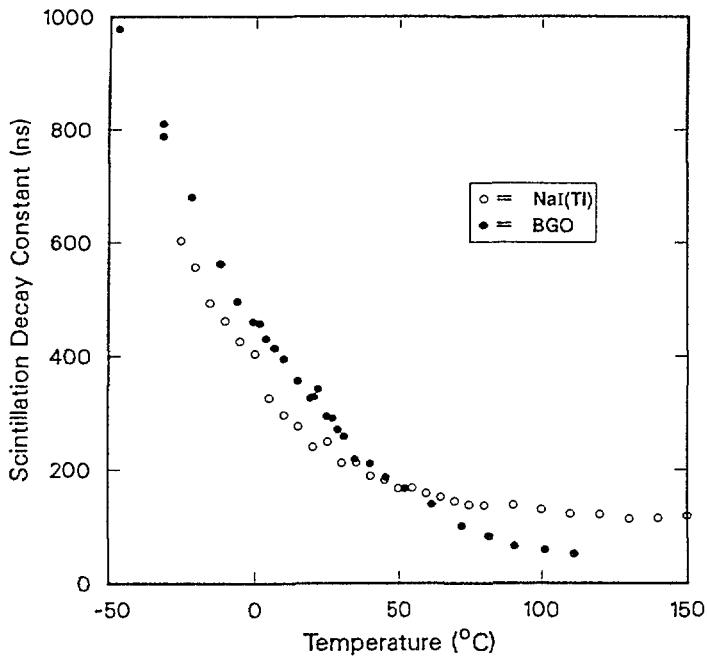


Figure 10. Scintillation decay time of two common scintillators as a function of temperature.

temperature, although the rate of this decrease varies by almost a factor of ten. The decreased scintillation output affects signal-to-noise, energy resolution, and gain control. The scintillation decay time also usually depends on temperature, thus affecting pulse shaping and counting rate. The emission spectrum may shift with temperature and, combined with the narrower response of high temperature photocathodes, requires that careful attention be paid to the compatibility of scintillator and photomultiplier. The shock and vibration encountered in the borehole is sufficient to damage detectors, and, consequently, mechanically rugged detectors and non-hygroscopic scintillators are preferred to avoid the need for special packaging that reduces detector size and performance.

The variation in the light output of the crystal as a function of its temperature will immediately govern whether the detector needs a dewar for successful operation or if it can be used without cooling. Since space for a detector is always limited, needing a dewar restricts detector size and puts added constraints on its gamma-ray detection efficiency per unit volume. A range of light output temperature responses (*Schotanus et al.*, 1985; *Harshaw*, 1985; *Melcher et al.*, 1985) is shown in Figure 9. The fast decay component of BaF₂ is independent of temperature, NaI(Tl) suffers a 30% loss by 150°C, and the light output of BGO drops by over an order of magnitude. As the light output loss of NaI(Tl) is not severe, this scintillator is often used with a high temperature photomultiplier without any cooling. A scintillator with greater light loss, such as BGO, requires a dewar to keep the detector from reaching ambient subsurface temperatures, which will often reach 150°C. The crystal and photomultiplier tube can be maintained at about 50°C, or less, permitting a wider variety of photocathode materials to be used.

For optimum spectroscopic measurements, the linear electronics should be matched to the decay time constant of the scintillator light. Amplifier shaping times are selected to minimize noise with respect to the time distribution of the light output from the crystal. As the scintillator temperature increases, the light output usually gets significantly more rapid, (*Melcher et al.*, 1985) as shown in Figure 10. The NaI(Tl) decay constant decreases by over a factor of two between room temperature and 150°C, and the BGO decay constant decreases by 7 ns/°C near room temperature. Thus the electronics must be capable of optimum analysis for the range of decay time constants to be encountered.

Since data must be obtained rapidly, scintillators with shorter decay times are always preferable to allow the highest possible count rates without severe pile up losses. Similarly, the volume available for a detector is always limited, primarily by the inside diameter of the sonde housing which can be less than 3.8 cm. The resulting relatively low detection efficiency can be increased slightly by increasing the length of the scintillator but at the cost of poorer energy resolution and reduced mechanical strength. Consequently, detectors with intrinsically high detection efficiency per unit volume are desirable. High detection efficiency per unit volume is primarily a result of high effective atomic number and, to a lesser degree, high density. The weight of the detectors is usually not of great significance.

Packaging a detector material for use in a nuclear logging measurement introduces additional constraints. Since all logging tools are subject to shock and vibration, the most rugged detector materials are preferred. Although even fragile detectors can be packaged to provide reasonable shock and vibration resistance, the large packaging volume greatly reduces the volume available for the detector itself. Thus, materials that require a minimum of protection can take better advantage of the limited space inside a sonde.

Because the energy resolution of a scintillator, as measured by a spectrometer system, depends on the total number of photoelectrons emitted from the photocathode of the photomultiplier, two other properties become particularly important. The index of refraction of the scintillator is important in determining how much of the light produced in the scintillator can be transmitted through the optical coupling and the photomultiplier front surface to the photocathode. Most photomultiplier faceplates and optical coupling materials have refractive indexes near 1.5. On the other hand, a number of common scintillators have considerably higher refractive indexes. BGO, for instance, has a refractive index of 2.15 which results in a significant fraction of the scintillation light being trapped inside the crystal due to internal reflection at the crystal-photomultiplier interface.

The emission spectra of scintillators also depend on temperature. The quantum efficiency of room temperature photocathode materials used in photomultiplier tubes is often sufficiently broad that small variations in emission spectra would not lead to a measurable difference in detected light output. However, photomultipliers that are designed for use at high temperatures generally have photocathode responses that are strongly peaked at low

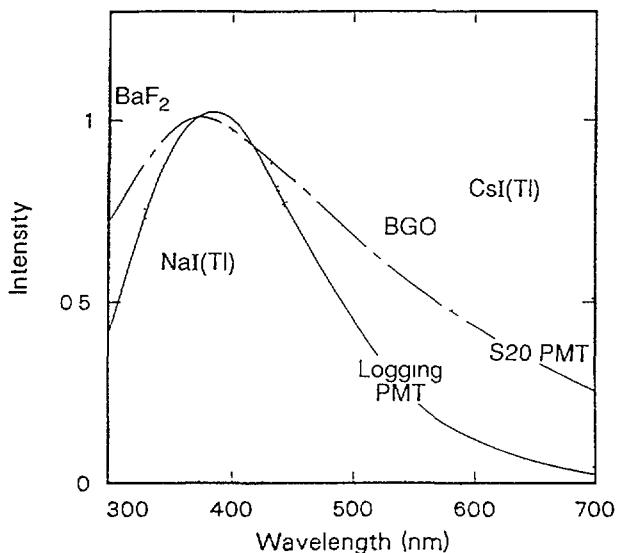


Figure 11. Quantum efficiency as a function of wavelength for a room temperature photocathode (S-20) and for a high temperature photocathode used in logging. Normalized emission curves for four common scintillators are shown for comparison.

wavelengths. For example, although the absolute shift in the emission spectrum for BGO between -20°C and 85°C is not large, the detected fraction of the light output changes by almost 10%. As mentioned above, quantum efficiency as a function of wavelength for photocathode materials that can be used at elevated temperatures is more strongly peaked than photocathodes that need only operate at room temperatures. This is illustrated in Figure 11 where the photocathode quantum efficiency is shown for a logging photomultiplier and a photomultiplier that might be used in the laboratory (S-20 photocathode). Also shown on the figure are the emission spectra for four common types of scintillators. With the S-20 photocathode, a significant portion of the light emitted from the scintillator can be detected from all of the scintillators. However, with the logging photomultiplier, only a small fraction of the light output of CsI(Tl) can be detected, and the fraction of light detected from BGO or BaF₂ is about half that obtained with the S-20 photocathode.

Many of the uses of detectors in well logging require that the detector operate in a mixed flux of neutrons and gamma rays, so that the neutron interaction properties of the detector material become important. There are two consequences of neutron interactions. The obvious problem is radiation damage to the detector, such as occurs in Ge (*Senftle, 1980*). This produces a gradual deterioration in energy resolution over time and eventually results in unacceptable performance. The second problem centers about the need to operate detectors at high gamma-ray count rates. If neutron interactions with the nuclei of the detector itself lead to detectable signals in the detector, then only a fraction of the total maximum count rate will be from the gamma rays of interest, resulting in a poorer signal-to-noise and consequently poorer statistics on the measurement.

Finally, although damage from neutron interactions are of primary importance, it is also important to note that another source of damage can be exposure to ultraviolet or visible light. Detectors in nuclear logging tools are often exposed to light during assembly or repair, and, for some materials, such as BGO, this can result in a decrease in detector performance. Clearly, a thorough understanding of this phenomenon is necessary to develop handling procedures which preserve the optimum performance of the detector.

Electronics

The details of the particular electronics incorporated in a logging sonde will of course be specific for the type of measurement. In general, the functional description of the electronics will be comparable to that employed in the laboratory for the same type of measurement. As an illustration, a schematic is shown in Figure 12 for a typical gamma-ray spectroscopy measurement system based on an accelerator source. Electronic power is sent to the logging instrument through the armored cable. Inside the logging tool, power supplies provide the necessary voltage levels for the electronics, i.e. ± 6 V, ± 12 V, and ± 24 V. An accelerator controller is shown in Figure 12 to control the gas level and operate the ion source. In addition, there will be specialized supplies to provide the accelerating voltage for the neutron generator and to provide detector or photomultiplier bias voltages. Frequently, these voltage supplies are controlled by circuitry in the tool, or by instructions from the surface computer, to maintain a particular gain for specific energy pulses from the detectors and to control the output of neutrons at a desired level.

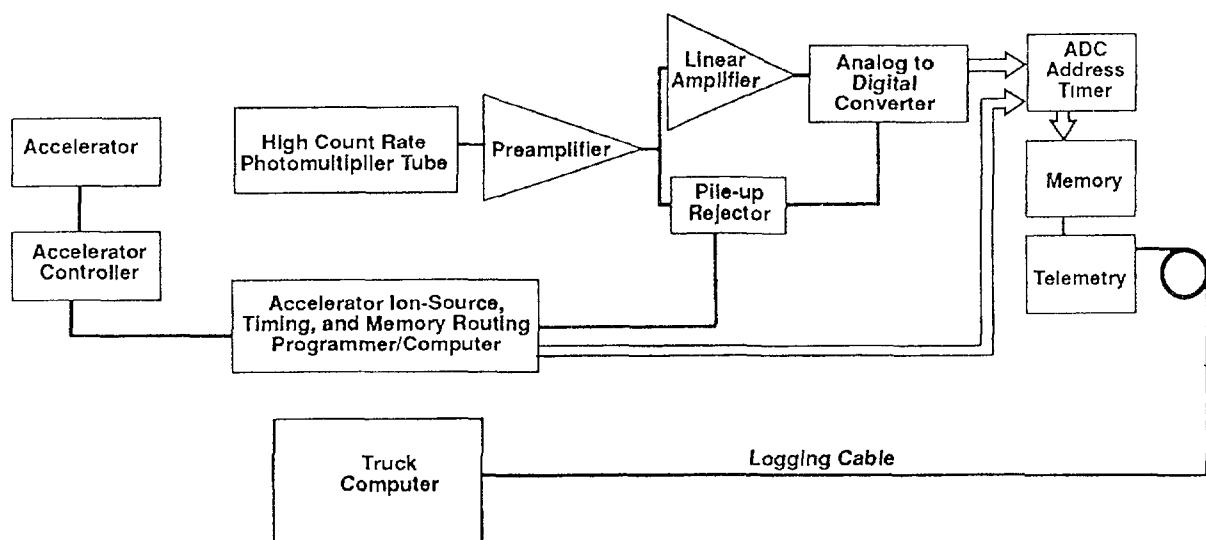


Figure 12. Schematic drawing of typical electronics needed for a pulsed neutron gamma-ray spectroscopy logging tool.

Since all electronics is contained within the logging tool housing, it is important to strongly filter all voltages to minimize noise pickup in other sections of the electronics. In addition to the preamplifier and linear amplifier for the gamma-ray energy pulses, it is often necessary to include pileup rejection circuitry to allow operation of the logging system at the highest count rates practical. Initially, linear signals were transmitted up the cable to surface instrumentation. However, it rapidly became apparent that the noise imposed on these signals severely compromised the quality of the measurement. Thus, analog to digital converters have been routinely incorporated in the tool electronics, so that only digital information needs to be transmitted to the surface. Since different spectra are often accumulated from the same logging tool, there is usually a controller which routes the ADC output into different sections of the memory. A telemetry system then reads the memory and digitally transmits the information through the logging cable to the surface computer.

The specific problems associated with developing electronics for subsurface logging measurements are generally independent of the particular type of measurement system. Commercially available electronic components are generally available that operate only to 125°C. Logging tools are expected to operate to at least 150°C, and higher temperature versions are desired to operate to temperatures as high as 250°C. In specific situations, such as geothermal wells, temperatures as high as 500°C (*Traeger and Lysne*, 1988) may be encountered. These higher temperature regimes present fundamental problems.

At the normal operating temperatures of a logging measurement system, components can be frequently selected from the normal high-temperature supply to give satisfactory performance. In such cases, the major constraint is the necessity to design the details of the circuitry in such a manner that the normal temperature drifts of individual components cancel out, leaving the circuit operating in an essentially temperature-independent mode.

In the previous section, the differences between the quantum efficiency wavelength dependence of a logging photomultiplier and that of a laboratory photomultiplier were presented. Other properties must be tailored for the logging environment (*Pietras and Smith*, 1988). The structure must survive the shocks and vibrations inherent in logging measurements. Recently, ceramic envelopes have been introduced to enhance the shock resistance of photomultipliers. In addition, the dynode structure must be inherently rugged. Special bialkali photocathodes have been developed that provide reduced photocathode dark current compared with the normal high temperature Na₂K₂Sb bialkali photocathodes and these optimized bialkali photocathodes have a reduced loss in quantum efficiency as a function of temperature. This is particularly important to avoid compounding the inherent light output loss of all scintillators with increasing temperatures.

Spectral Analysis

The initial approach to resolving individual contributions to a detected gamma-ray spectrum involved the use of relatively broad windows around individual peaks (*Rabson*, 1959; *Oliver et al.*, 1981). This approach does reasonably well for simple spectra, such as those from natural activity which have only three contributing sources (though a complex gamma-ray structure). However, for spectra with a greater number of contributions, it became clear that a more sophisticated approach was necessary (*Hertzog*, 1980). This becomes increasingly clear when one also considers the statistical quality of the spectral data desired to be analyzed. Such data cannot be well analyzed by a simple window, or peak-stripping approach. An example of a spectrum with good statistics is shown in Figure 13a. This spectrum, which was measured in the laboratory, is idealized in several respects: it contains only three elements, H, Si, and Fe; it has good statistical precision, being a one-hour accumulation; and it has optimum energy resolution, being measured

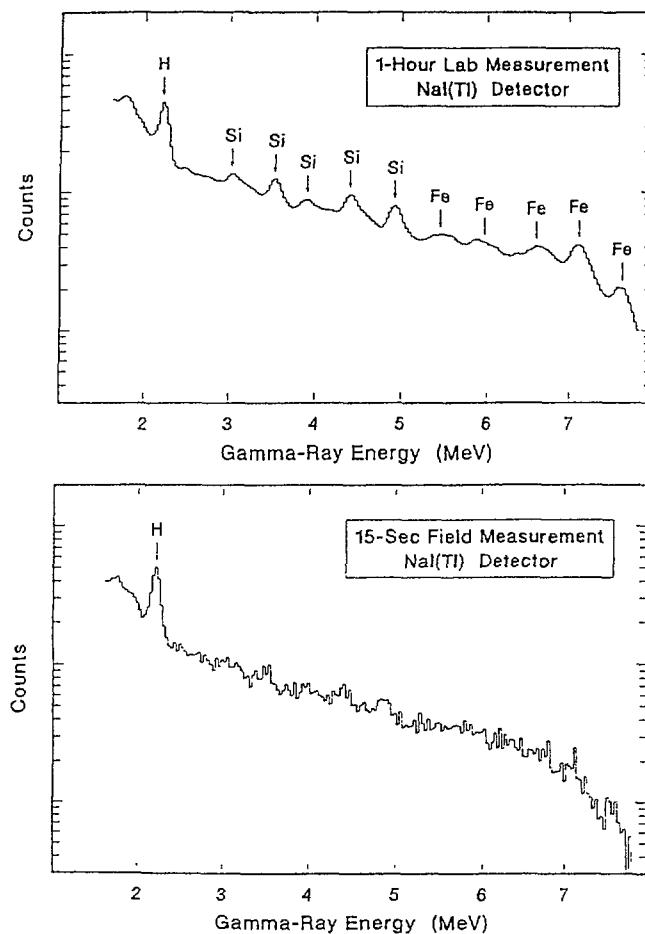


Figure 13. Typical thermal neutron capture gamma-ray spectrum in a clean sandstone: (a) a one hour accumulation showing isolated peak structure; (b) typical 15 second measurement that would be analyzed from a subsurface measurement.

at room temperature. The gamma-ray peaks from H, Si, and Fe are clearly visible, and one could think of doing a simple peak-intensity analysis of this spectrum. Figure 13b, on the other hand, shows the real world: a 15 second accumulation of data measured at a depth of 3 km in an oil well at a temperature of 100°C. Moreover, instead of 3, there are 11 elements which contribute a significant number of gamma rays to this spectrum, and only one of them, H, is visually obvious as a peak. Obviously, a more sophisticated analysis than peak stripping is needed. The analysis method usually adopted for analyzing a gamma-ray spectrum is the minimization of the weighted-least-squares error to a fit of the measured spectrum by a set of standard spectra (*Quittner*, 1972).

The spectral analysis problems are most readily illustrated by the capture spectrum obtained from a pulsed neutron spectrometer. There are many different components to the thermal neutron capture spectrum, frequently eleven different elements can make significant contributions. Before the analysis of the spectrum can begin, the data must be acquired in an appropriate manner. Gain and offset control of the spectrum during acquisition is maintained by calibration sources and/or electronic pulser peaks. The strength of calibration sources and their energy is not a trivial question. A strong enough peak is required to obtain statistically meaningful values for the calibration peak centroid. However, the more time the detector spends analyzing the calibration peak, the less time is available for acquiring the data desired. Also, the energy of the calibration source should be great enough to provide a good measure of the system gain over the energy region of the measurement, but it should not contribute to the spectral region that is being analyzed, or it will contribute background to the measurement which will affect the counting statistics per unit time. Even after good quality data is obtained, a number of adjustments must be made for an optimum analysis.

The weighted-least-squares error minimization analysis starts with a determination of the response of the spectrometer to a single element in a typical borehole environment (*Grau and Schweitzer*, 1987). Then the most probable linear combination of elemental responses that would fit the unknown composite spectrum is determined by using weighted-least-squares error minimization. With this approach, it is necessary to insure that the measured spectra have exactly the same gain and offset as the standard spectra. Furthermore, it is necessary to adjust the energy resolution of the standard spectra to be the same as that of the measured spectrum. It is assumed that the unknown spectrum U (not to be confused with the U of Equation (6)) can be represented, within the statistical uncertainties, as a linear combination of the standard spectra S :

$$U_n = \sum_i S_{ni} Y_i + \epsilon_n, \quad (16)$$

where n is each of the 256 channels in a particular spectrum. The spectral yield coefficients Y_i are then the number of events in spectrum U which can be attributed to element i .

The best estimate of the set of yields Y_i is that set which minimizes the weighted sum of squares of the errors ϵ_n over all channels n:

$$i.e. \text{ minimize } \sum_n W_n \epsilon_n^2 \text{ for changes in } Y_i. \quad (17)$$

The weight, W_n , which results in the smallest variance in the yields Y_i is the inverse of the expected variance in ϵ_n , which is assumed here to be dominated by the variance in U_n . The best estimate of the yields is then:

$$Y_i = \sum_n W_n \left(\sum_j \left(\sum_m W_m S_{mi} S_{mj} \right)^{-1} S_{nj} \right) U_n. \quad (18)$$

Historically, analyses of capture spectra in general geological environments have been focused on the cleaner sediments, with relatively few important elemental contributions to the total spectrum. Recently, the importance of understanding sediments containing a wider variety of minerals, such as clay minerals, in addition to sandstones or carbonates has lead to a reexamination of all possible spectral contributions to the measured spectra.

The extension of spectroscopic logging measurements to a wider variety of mineral assemblages originally encountered difficulties in the proper analysis of the spectra. In analyzing spectra from relatively clean formations (free of clays), the measured capture spectra could be well fit with Si, Ca, Fe, S, H, and Cl standards only. However, when other minerals, such as feldspars and clay minerals are included, as much as 70% of the measured spectrum can be due to other elements.

It became clear (*Grau and Schweitzer, 1989*) that it would be necessary to establish appropriate standards for each of the elemental contributions to the typical spectra observed in a wide variety of minerals, and include all of the required standards in the basis set used to fit each measured spectrum. In principle, this means fourteen elemental standards. However, it would be impossible to separate the measured spectrum into fourteen parts while retaining any reasonable level of statistical precision, primarily because of the energy resolution of inorganic scintillators. It has been determined that three additional standards, Gd, Ti, and K, are sufficient to describe most geological environments, though the effects of other significant contributors to the total spectrum are generally accounted for. The set of ten standards used to fit a typical capture spectrum, which includes a standard for the interaction of the NaI(Tl) detector with neutrons, is shown in Figure 14. The effect of using this expanded set of standards is shown in Figure 15. The histogram curves show a typical neutron capture gamma-ray spectrum measured in a region with significant clay minerals. The top smooth curve illustrates the poor fit to the spectrum when only the original set of seven standards (including that for the NaI detector) is used. The bottom smooth curve shows the dramatic improvement in the fit to the measured spectrum when the standards for Gd, Ti, and K are included.

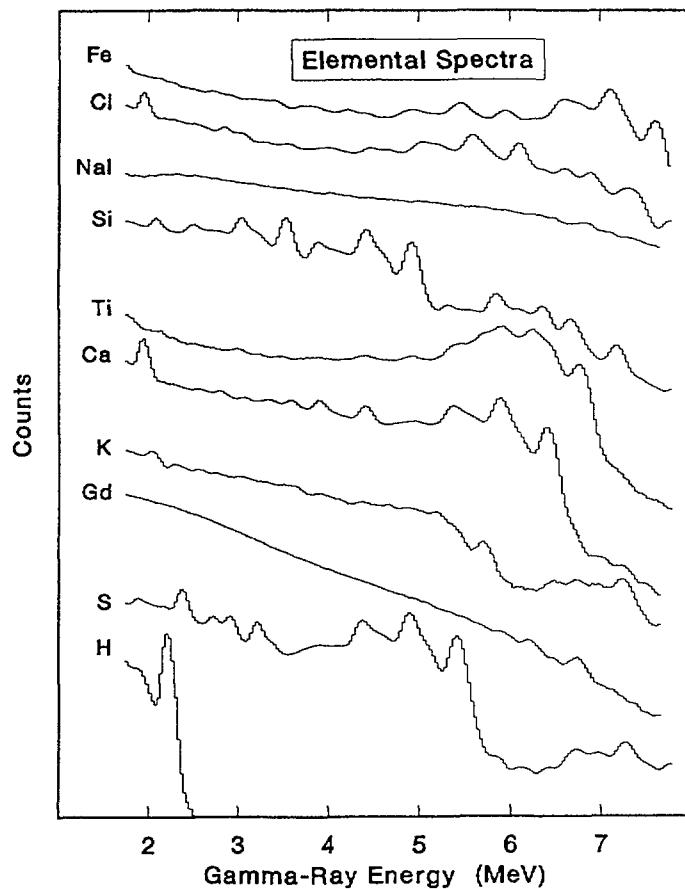


Figure 14. Set of ten standard spectra used to fit measured spectra.

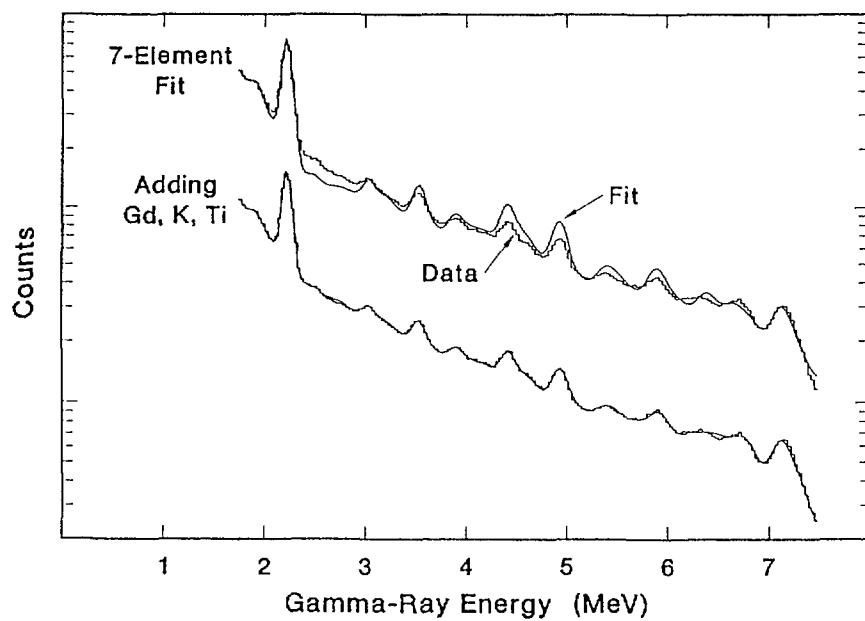


Figure 15. Comparison of the weighted-least-squares fit to a measured spectrum in a region containing significant amounts of clay minerals. The upper curve represents the fit with only the seven standards used for analyzing clean sandstones and carbonates. The lower curve illustrates the improved fit when Gd, Ti, and K standards are included.

When this set of ten elemental standards is used to fit the measured spectra, statistical uncertainties on the extracted concentrations are still large. The neutron-capture determined K concentrations are so poor statistically that it is impossible to use them for describing the K content of the rock. It is, however, necessary to include the potassium spectrum in the fit to the measured spectrum to avoid systematic errors in fitting. The highly erratic result cannot be ignored (for example, by just using the natural activity measurement) as the poor statistics are propagated into the concentrations for all the other elements in the spectrum. The solution makes use of the cross correlation between the shapes of the different elemental standards and constrains the results of the neutron-capture K concentration to agree with the K concentration from the natural activity measurement. Since the natural activity measurement is directly calibrated in weight percent and the neutron capture measurement provides a normalized spectral yield, it is necessary to use the closure normalization model (described above) to convert the concentration of K from the natural activity measurement into the expected spectral yield in the neutron capture measurement. This is accomplished by using the initial fit to the capture spectrum to obtain the normalization, F, which converts between the capture spectral yields and the elemental concentrations. This value of F is then used to convert the K concentration from the natural activity measurement into the expected correct K yield in the capture measurement. This value for the K yield is used to constrain the value in the analysis of the capture spectrum, with the difference between the new and old value of the K yield redistributed among the other yields, as described below. These new values of the spectral yields are then used to determine a new value for F to convert the capture elemental yields into elemental concentrations. This procedure could be repeated to obtain better estimates of the proper values for F, but, in practice, one iteration is usually sufficient to provide an accurate constraint on the final elemental concentrations.

The constraint for any element in the neutron capture spectrum fit can be described by the following relationship that redistributes the over- or under-estimated portion of the spectral yield for the constrained element into the spectral yields for all the other elements in the spectrum:

$$Y_i^{new} = Y_i^{old} + R_{i,j} (Y_j^{old} - Y_j^{new}) \quad (19)$$

where the Y^{old} values are the original results of fitting the measured spectrum, the Y^{new} are the values after applying the constraint, and the $R_{i,j} = -\sigma_{i,j}/\sigma_{j,j}$, the ratio of the off-diagonal to the diagonal values of the inverse matrix from the weighted-least-squares analysis. In particular, for the application of the constraint on the K yields, Y^{old} is the value from fitting the capture spectrum, and Y^{new} is the value obtained from the natural activity measurement with better statistics. The other Y^{new} will then be the redistributed yields for the other elements. The true importance of the constraint is in the proper determination of the concentrations of the other elements.

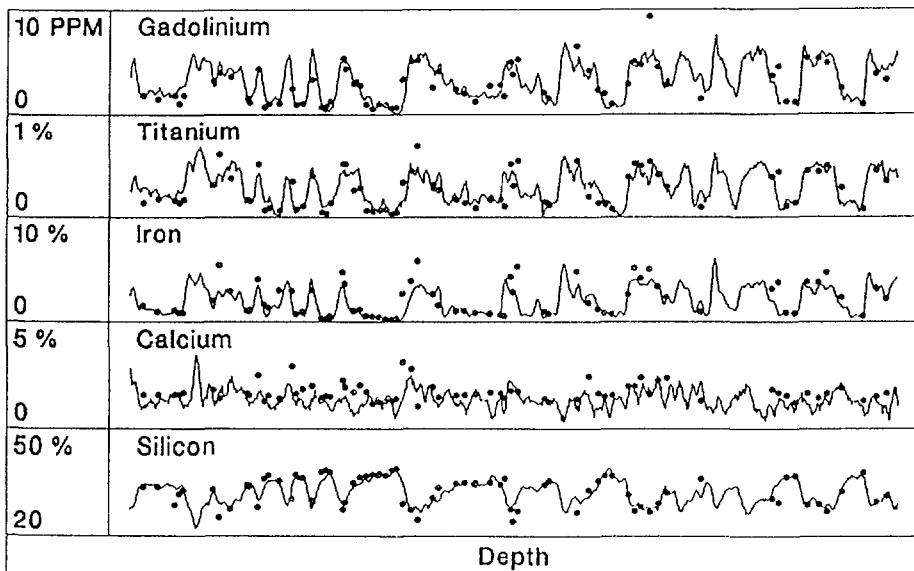


Figure 16. Typical agreement between log and core measurements for the elements determined from the thermal neutron capture gamma-ray spectra after analyzing the spectra as described here.

Aluminum can make a significant contribution to the yield in the capture gamma-ray spectrum. However, an aluminum standard is not included in the basis set of standards. The reason for this is that the shapes of the measured spectra for Al and Fe are sufficiently similar that an attempt to separate them both independently would more than double the statistical uncertainty of the Fe yield. Instead of including a standard for Al, it has been established that the Fe standard alone provides a yield that is virtually the sum of the yield from Al and Fe. Since an independent measure of Al concentration is available from the delayed activation measurement, that amount of Al, after transforming it to a spectral yield, is subtracted from the initially calculated Fe yield. In a similar way, it has been found that since the Sm spectrum is similar to the spectrum from Gd, and since the Sm/Gd ratio is reasonably constant in nature, a small correction is applied to the initial amount of Gd that is calculated to account for the contribution from Sm. In this case, the elemental standard used to fit the Gd contribution is actually a linear combination of the Gd and Sm spectra. The results of the yield from this summed spectrum are then calibrated through the sensitivity factors, S_i , into the proper content of Gd.

Two other elements, Na and Mg, have generally not been included in the fit. Because of their capture cross sections and typical abundances, they usually do not contribute a significant portion of the observed spectrum. A significant Na contribution occurs only when there is a large amount of Na feldspar present, and this is very rare. While it is regrettable that Na cannot be used as a diagnostic element from these measurements, its exclusion from the spectral analysis has virtually no adverse effect on the accuracy of the

remaining elemental concentrations. A significant spectral contribution from Mg occurs only in relatively pure dolomites. In such cases, or in some other unusual mineralogies where elements such as Na or Mg are present, their standards could be included in the fit, with an accompanying elimination of standards of other elements that are not present. Satisfactory results using this method have been obtained in some carbonate environments where the contributions from K and Ti could be safely ignored.

When the standard approach is applied to measured spectra, reliable elemental concentrations for ten elements contained in the rock can usually be obtained. A typical comparison between the results of subsurface logging measurements and laboratory analyses of core samples is shown in Figure 16 for the five significant elemental concentrations obtained from the thermal neutron capture spectrum. The two sets of data are in excellent agreement, especially after allowing for the possible errors in matching core sample depths to logging depths and in comparing measurements on a $\sim 1 \text{ cm}^3$ core with a measurement that averages over two to three orders of magnitude larger volume.

CONCLUSIONS

Subsurface nuclear logging measurements have been developed to provide a substantial amount of information that can be used to describe the properties of geological formations. In addition to measuring bulk properties, it has recently become possible to measure the concentration of a significant set of chemical elements in the rock. The increased understanding of the physics of nuclear measurements in extended media has resulted in a significant increase in the use of nuclear techniques throughout the oil industry. The equipment needed to perform these measurements is comparable to that used in the laboratory. However, the constraints on performing measurements in a borehole require sources, detectors, and electronics specifically designed for that environment. In addition, analysis techniques must be robust enough to handle the effects of the logging environment on the measured data.

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RADIOTRACER TECHNIQUES IN THE MINERAL INDUSTRY

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Abstract

Important applications of radiotracer methods in the field of mining and processes of mineral industry such as, crushing, grinding, classification, flotation, storage and drying of products are described. The important aspects which are evaluated include holdup in mills, separation coefficients, critical settling depth and efficiency of equalisation. Tracer techniques provide data which are used for control and optimisation of ore dressing systems. In the mining industry the information obtained is useful for design and control of draining systems of mines and ventilation of mines.

1. INTRODUCTION

The result of mining and mineral processing by several successive operations is separation of concentrate from gangue. If operations of similar character are assembled into one process, the treatment of ores can be schematically represented by four general processes (see Table 1).

In the first one - mining - are operations of surface or underground mining, in the second one - comminution - are operations oriented toward reduction of particle sizes (i.e. crushing, grinding, etc.). In the third process - separation - there are operations where fractions are separated (i.e. classification) which take place e.g. in hydrocyclones or in flotation or sedimentation units. Product handling - includes transport of ores or products (e.g. hydrotransport), their deposition and removal from heaps or storage tanks, bunkers and also operations which treat the ore fractions so that they are more suitable for further special treatment (e.g. drying or calcination).

Radioisotope tracer methods offer information necessary for evaluation and optimisation of individual operations and systems i.e. groups of operations and processes. It is generally assumed that the tracer material

TABLE 1. RADIOTRACER METHODS USED IN THE MINERAL INDUSTRY

Process	Offering Information by RM	RM - Application
MINING	Rate of air renewal Water menace to mines	S.R. S.R. Dating of Groundwater
COMMINUTION	Holdup of material in device Size grain distribution of holdup Size grain distribution of output	S.R. Model analysis of S.R. Model analysis of S.R. Size fraction of S.R.
SEPARATION	Separation coefficients Partition curve Cut size diameter Removal efficiencies Critical settling depth Solid flow analysis	I.M. of injection I.M. of size fraction injection S.R. - mean residence time S.R. - R.T.D. R.T.D. + settling column data Model of R.T.D.
PRODUCT HANDLING	Calibration of Magnetic Flowmeters Equilization of quality in storage Efficiencies of equalisation	Isotope velocity technique S.R. - R.T.D. S.R. - R.T.D.

RM - Radiotracer method
 S.R. - Stimulus response method
 I.M. - Integral method
 R.T.D. - Residence time distribution

has identical properties as the treated particles so that identical behaviour of tracer in the considered process is assumed. The advantage of the radiotracer method is simplicity, speed and possibility of detection of gamma radiation through the walls of the equipment. By use of radiotracer methods the dynamic characteristics of moving solid phase formed by solid particles can be determined. The method is based on measurement of the concentration change of the tracer with time (stimulus response method), or by the material balance of indicated fraction of solid phase on basis of evaluation of the total tracer quantity (integration method of the tracer injection).

For the preparation of radiotracers for investigations of mineral processing the treated ores, which mostly contain elements which on neutron irradiation produce gamma emitting tracers, are used as targets for activation.^{6,11,13,14,15,16,17,18} In studies of processes in which the physical properties of the labelled materials are not changing (e.g. at equalisation - equalisation - or sedimentation) it is possible to use also the surface labelled particles.² This method has been described in numerous studies.^{1,2}

In Table 1, details are given of individual processes which can be evaluated by use of radiotracer methods (RTM). In the final part of this

report radiotracer applications are described for evaluation of systems, i.e. whole industrial processes for their optimisation and control.

2. MINING

2.1. Mine drainage

The process of underground or surface mining of ore can be menaced by underground water streams. Information on their origin, quantity, communication with surface flows are used for design and control of pumping stations. Radiotracer data supplies information on flowrates and communication of underground water flows from important localities in the vicinity of mines e.g. from bunkers, dams or underground water sources. For evaluation of their origin, dating by determination of ^3H and ^{14}C is also used.

Dating is based on the fact that radionuclides such as ^3H and ^{14}C originate by cosmic ray irradiation and after separation from the atmosphere their content changes according to the exponential disintegration law. The range of dating corresponds to the half-time of the corresponding nuclide which is 0-30 years for tritium dating and 100-4000 years for ^{14}C dating.

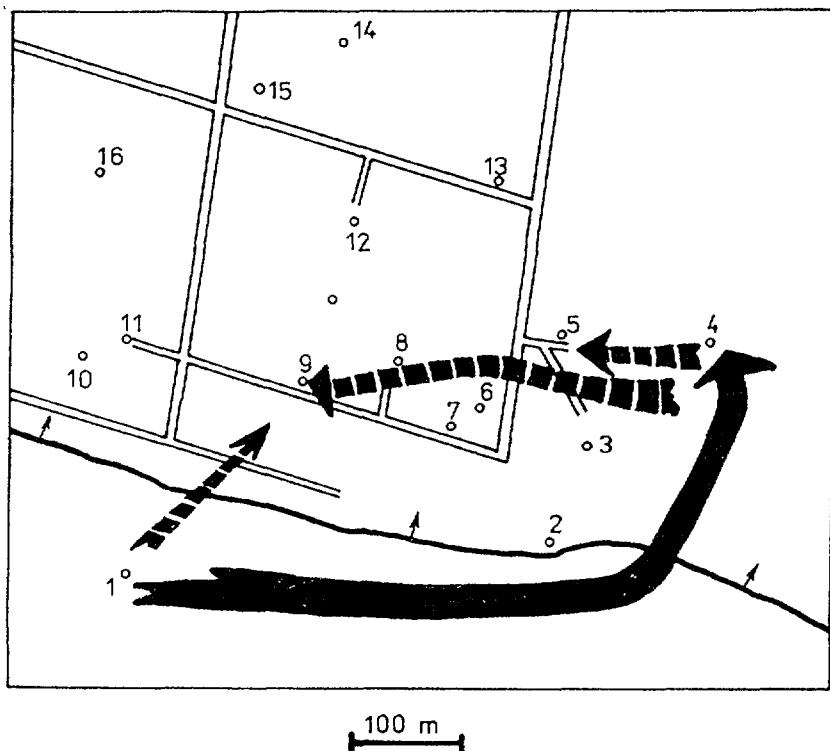


Fig. 1. Flowrates and communication of underground water.

o - 1,16 boreholes

As these are beta emitters with very low energy and low specific activity, for their detection, methods with maximum detection efficiency are used. Several publications are devoted to such measurements and evaluation of the various corrections required in the measurements.^{3,4,5}

For evaluation of the constructed draining system the integral diluting method of tracer injection¹ is used. On basis of known underground composition of ores a set of boreholes is prepared (see Fig. 1). Into one of them is injected the radiotracer (usually ⁵¹Cr as EDTA complex or ⁸²Br as solution of NH₄Br) and is flushed with water. In other boreholes the presence activity is detected. From the total amount and time of appearance of the radiotracer, the direction of motion and magnitude of flow as well as parameters of water collectors such as effective porosity, filtration coefficient etc. are determined.⁵

2.2. Mine ventilation

The radioactive gas ⁸⁵Kr is used for evaluation of the rate of air exchange -R- [m^3s^{-1}] in mine.⁶ The tracer is perfectly distributed in the measured space and by switching on the ventilator the decrease of its concentration in time -N(t)- is studied.⁸ If it is possible to assume ideal mixing in the studied space, then the relationship

$$N(t)/N(t_0) = \exp (-Rt) \quad [1a]$$

applies, where N(t) and N(t₀) are concentrations of the tracer in time t or in time t=0. The rate of air exchange -R- can be also determined from the relation

$$R = Q/V \quad [1b]$$

where Q [m^3/s] is the volumetric flow rate of air and V [m^3] the volume in the studied space. According to this relation i.e. by measurement of the volumetric flow rate of introduced air by use of ⁸⁵Kr, the efficiency of ventilation in gold mines was evaluated.⁶

⁸⁵Kr was also used for evaluation of efficiency of fire protecting walls-barriers which should prevent the inlet of air into specified space in coal mine. The wall material permeability at fixed pressure of air at ventilation at the other side was studied.⁷ Very low flow rates were considered and tracer concentration was measured by sampling of gas after adsorption in toluene by use of a spectrometer for liquid scintillators. In

this case the diluting method was used;⁸ for gas flow through the wall -q-[m³.s⁻¹], the relationship

$$q = N(Q+q)/N_0 \quad [2]$$

applies, where -N- is the tracer concentration at the vented side and -N₀- is the tracer concentration after mixing at the other side and -Q- is flow rate of air introduced [m³/s].

3. PARTICLE SIZE REDUCTION

Processing of the mined ore yields the enriched ore with small content of waste rock -gangue-. Operations such as crushing and grinding reduce the size of raw particles to smaller size from which the liberation of ore is much easier. The smaller the particles are, the easier the liberatin is. On the other hand comminution of the ore to very fine fractions represents larger energy consumption and also the danger of larger losses of ore as very fine untreatable slim particles which are discharged in tailings. For these reasons in practical cases a compromise must be made between the ease of liberation, operating costs and losses of ore.

Radiotracer methods are frequently used in this case. Without considering the type of equipment in which the reduction of particle sizes is performed the information obtained can be separated into two groups:

1. Data on hold up of the treated material;
2. Data on particle size distribution.

Both can be obtained by measurement of the impulse response.

3.1. Crushing

Reduction of particle dimensions -disintegration- is performed in several stages by crushing and repeated grinding. A typical unit used for crushing of material is the hammer mill.

Operation of the mill and measurement of the impulse response is shown in Fig. 2. For crushing of particles only a certain part of energy supplied into the mill is spent. Considerable part of total energy is spent for keeping the particles in motion. These particles form the so-called holdup.

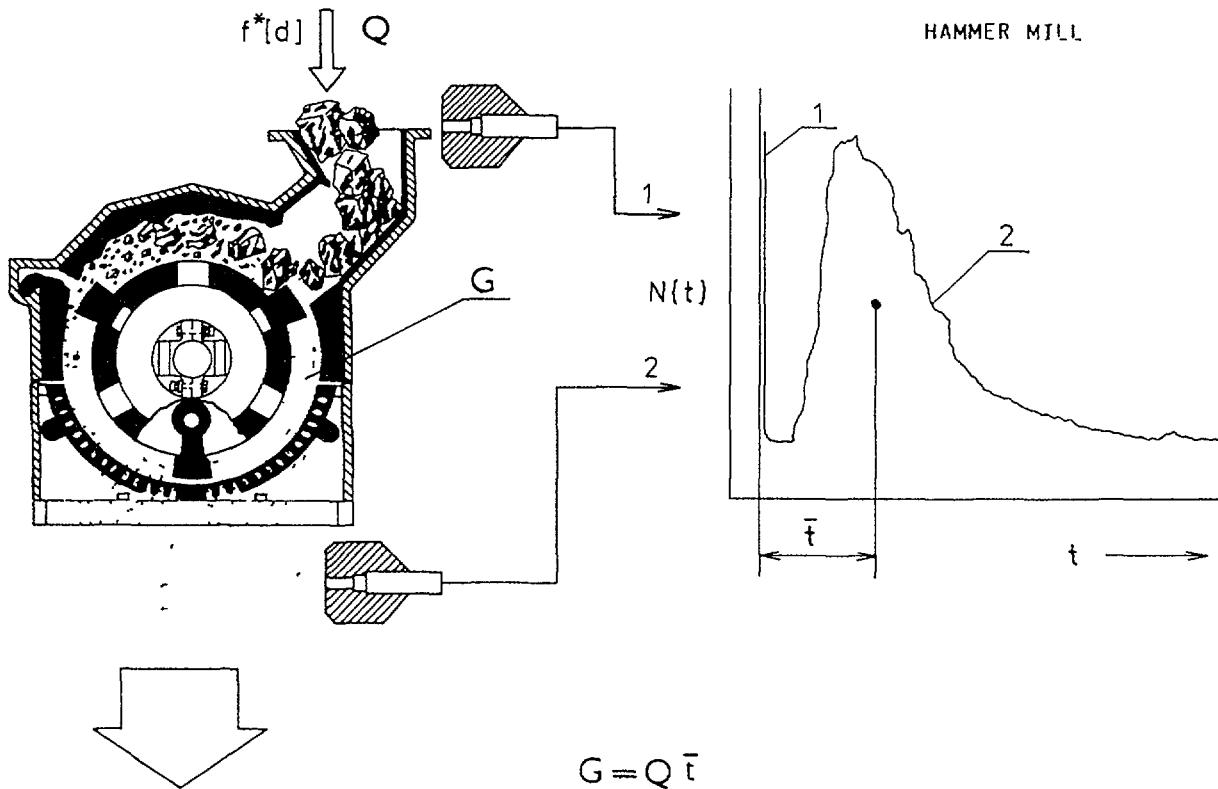


Fig. 2. Impulse response in hammer mill.

G - holdup; \bar{t} - mean residence time;

Q - material flow rate; $N(t)$ - tracer concentration

If composition of the holdup is such that the already crushed particles remain in circulation, then the energy spent for keeping the holdup in motion may be considered as lost.

The magnitude of the holdup $-G-$ is determined from the mean residence time of the material, $-\bar{t}-$. If the record of the impulse response $-N(t)-$ and the mass flow rate of material through the unit $-Q-[kg/s]$ are known, then for holdup, the relation

$$G = Q \int_0^\infty t N(t) dt / \int_0^\infty N(t) dt \quad [3]$$

applies, where t is real time and $t=0$ is the time of injection of activated particles and $N(t)$ is detected tracer concentration at the outlet from the unit. The principle of the method, i.e. the stimulus response method, is demonstrated in Fig. 2.

The composition of the holdup can be estimated from the analysis of the shape of the impulse response if the crushing operation is described by the simplified impulse response model.^{2,26}

In this derivation it is assumed that the process in the hammer mill can be divided into two phases, the first one disintegration and the second one screening. This model can be described by the next scheme

A---- disintegration --> B---- screening ---> C

where -A- is the raw material being crushed and having particle size $d_A > d_o$; -B- is crushed material with particle size $d_B > d_o$, and C material which passed through the screen with particle sizes $d_C > d_o$, where d_o is the diameter of screen mesh.

The impulse response $-f(t)-$ is then given by relation with two constants for disintegration $-k_1-$ and screening $-k_2-$:

$$f(t) = z [\exp(-k_1 t) - \exp(k_2 t)] \quad [4]$$

where $z = k_1 k_2 / (k_1 + k_2)$;

According to this model, for the mean residence time $-t-$ the relationship

$$t = (k_1 + k_2) / k_1 k_2 \text{ applies} \quad [5]$$

and for the composition of material in holdup the relation

$$B/A = k_1 / k_2 \text{ applies.} \quad [6]$$

Energy consumption for grinders with smaller material holdup will be lower. From relations [5] and [6] it is apparent that this condition is satisfied for a mill which has the impulse response with constants $k_1, k_2 > 0$. In holdup, the content of already crushed particles should be as low as possible which means that in a well designed mill, for these constants $k_2 > k_1$.

3.2. Grinding

Disintegration of crushed particles to finer fraction is performed in horizontal rotary rod or ball mills.⁹ The grinding media rods or balls prevent, to a certain degree, motion of the crushed material. This resistance depends on the type and amount of charge but also on mass flow rate i.e. amount of material which is passing through in unit time. The larger the resistance, the greater is the material holdup.

The distribution function of particle sizes leaving the mill is also affected by particle dispersion inside the unit which is characterised by the impulse response.

$$\sigma^2 = \int_0^\infty (t-t)^2 N(t) dt / \int_0^\infty N(t) dt = \int_0^\infty (t-t)^2 f(t) dt \quad [7]$$

where $f(t)$ is residence times distribution.

From these facts it is apparent that by simple evaluation of the impulse response two quantities are obtained - holdup and dispersion which are important for optimisation of the process.^{14,15,16}

The mathematic model of the grinding process enables the determination of the last important information i.e. the size distribution function obtained as the result of the grinding process. The mathematical formulation results from an analogy with conversion of chemical reaction in the flow reactor.¹⁰

If we assume that the kinetics of grinding is a linear process described by the first order differential equation, then for the mass fraction of the i -th fraction obtained by grinding run for time t the relationship

$$x_t(t) = \sum a_{ni} \exp(-k_n t) \quad \text{for } i=1,2,\dots,p; \quad [8]$$

applies, where $x_t(t)$ is the weight fraction of material belonging (as concerns the sizes) into the group i , which has formed during time t by grinding the particles of sizes $p = 1, 2, \dots, i-1$ where a_{ni} and k_n are disintegration constants.

As individual particles reside in the mill for different times characterised by the distribution of residence times $-f(t)$ - then for the fraction of particles in the flow mill the relation

$$x_t = \int_0^\infty \sum_{n=1}^i a_{nt} \exp(-k_n t) f(t) dt \quad \text{for } i=1,2,\dots,p \quad [9]$$

holds.

The residence time distribution can be obtained e.g. by continuous measurement of the impulse response of activated material.¹³ Usually it is assumed that $f(t)$ is independent of particle sizes.¹¹

If sieve analysis of the inlet and outlet materials are made, assumptions made the disintegration constants can be calculated by the iterative method from equation [9]¹¹ (see Fig. 3).

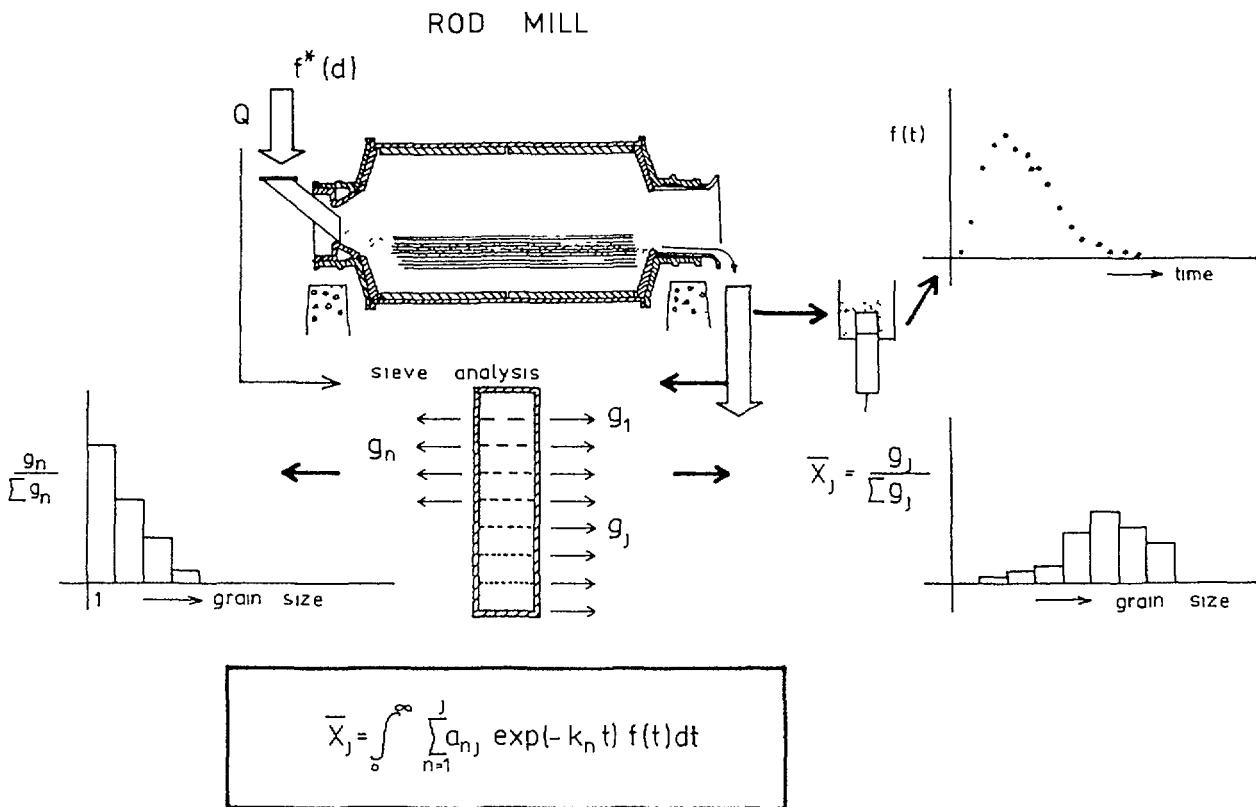


Fig. 3. Rod mill.

Q - material flow rate; $f^*(d)$ - input of tracer size distribution; $f(t)$ - RTD; q - weight fraction; \bar{X} - ratio of weight fraction; a_{nj} , k_n - disintegration parameters; n - input; j - output

In the application of radiotracers, it is possible to specifically label the selected fraction only - e.g. of the j -th fraction of the ground material. The impulse response of this fraction is determined by measurement of tracer concentration in samples withdrawn in certain time intervals. The samples must be sufficiently large so that their sieve analysis could be made together with measurement of tracer concentration in each size fraction (see Fig. 4). The obtained time dependences

$$X_t(t) = \int_0^\infty a_{ji} \exp(-kt) f(t) dt \quad [10]$$

are used in evaluation of disintegration constants by the least square method.¹¹ This method has a disadvantage in being too time consuming which can be improved e.g. by simultaneous labelling of two fractions by two radiotracers and by spectrometric measurement of their concentrations.¹³

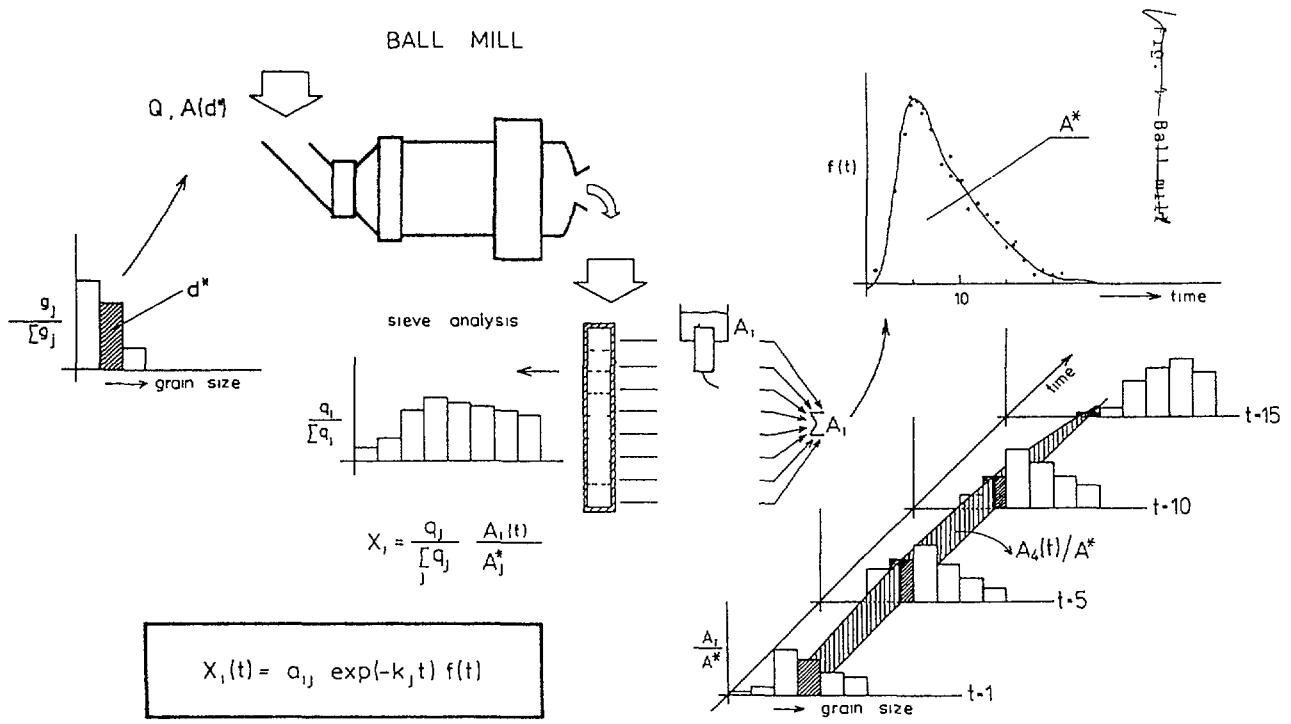


Fig. 4. Ball mill.

Q - material flow rate; $A(d^*)$ - input of tracer size fraction
 d^* - grain size of tracer; A_i - sample tracer concentration;
 A^* - tracer quantity; X_i - ratio of weight fraction;
 a_j, k_j - disintegration parameters

4. SEPARATION

The disintegrated particles differ in their size, mass and ore content. The aim of the operation is separation of concentrate from gangue. The separation processes take place in hydrocyclones and gravity concentrators or in flotation units.

Efficiency of these units is evaluated by use of the so-called separation coefficient n which can be determined by use of radiotracers by the integral method of tracer injection (see Fig. 5).^{1,2} The separation coefficient n is the ratio of the separated material (i.e. e.g. of gangue) to material entering into the unit

$$n = \frac{\Delta G}{G_{in}} = k \left[v \int_0^\infty (N(t) - N_b) dt \right] / \left[v \int (N(t) - N_b) dt \right]_1 \quad [11]$$

where v is the mean velocity in the locality j or i , and k is calibration factor which takes into account the correction for the response of detector,

geometry of detection etc. N_j , (t) is the tracer concentration (in counting rate for example) measured in the locality $-j-$ or $-i-$ in time $-t-$ after impulse tracer injection, N_b is counting rate of the background.

Similarly it is also possible to evaluate the magnitude of the recycle stream.²

For evaluation of separation efficiency the so-called partition curve which gives the ratio of mass flows of the outlet and inlet materials for all fractions entering the unit⁹ is also used (see Fig. 5). To obtain such ratios by classical methods is cumbersome and time consuming as it requires sampling, drying, sieve analysis, weighing etc. The ratio is used for determination of the so-called cut point or separation size d_{50} which corresponds to the fraction of particles which have the same probability of outlet with fine or coarse fractions (see Fig. 5). As it is possible to

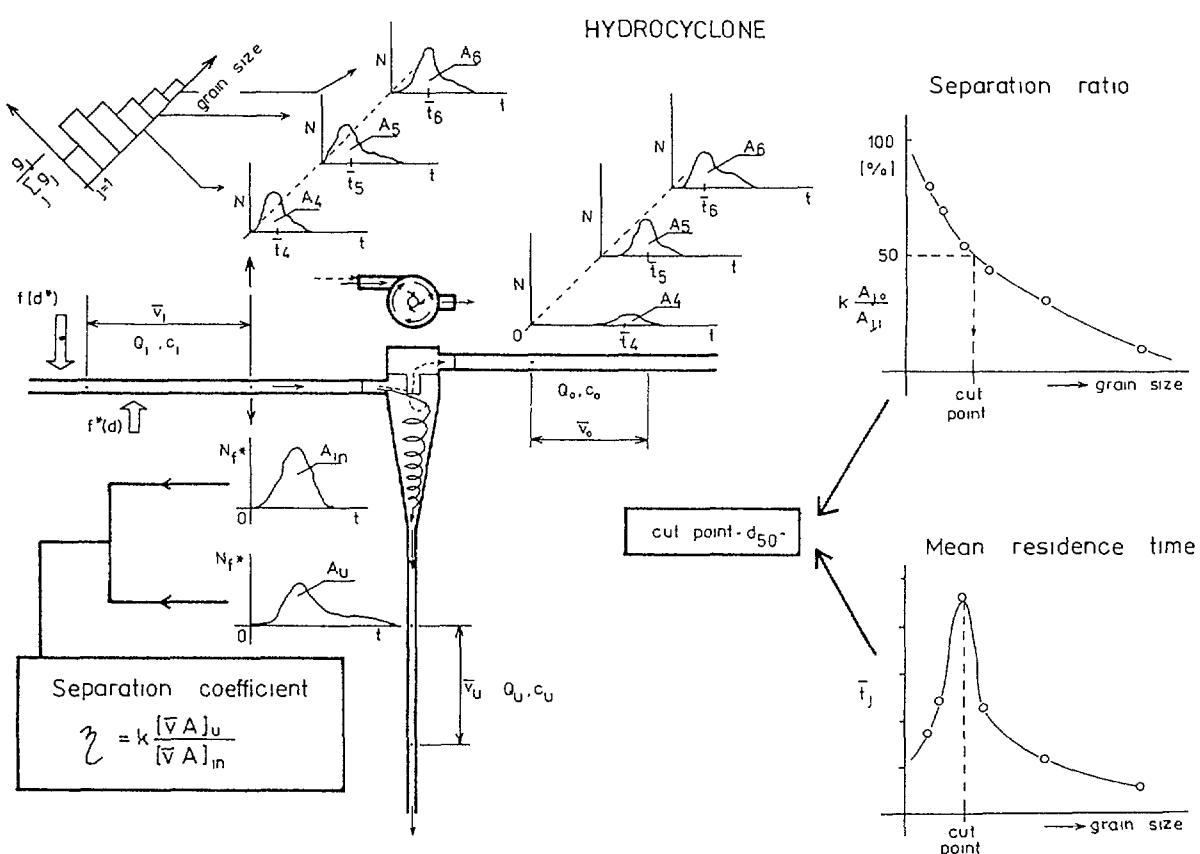


Fig. 5. Hydrocyclone.

$f(d^*)$ - input of tracer size fraction; \bar{v} - mean flow velocity; Q - liquid flow rate; $f^*(d)$ - input of tracer size distribution; c - solid particles concentration; g - weight fraction; U - underflow; O - overflow

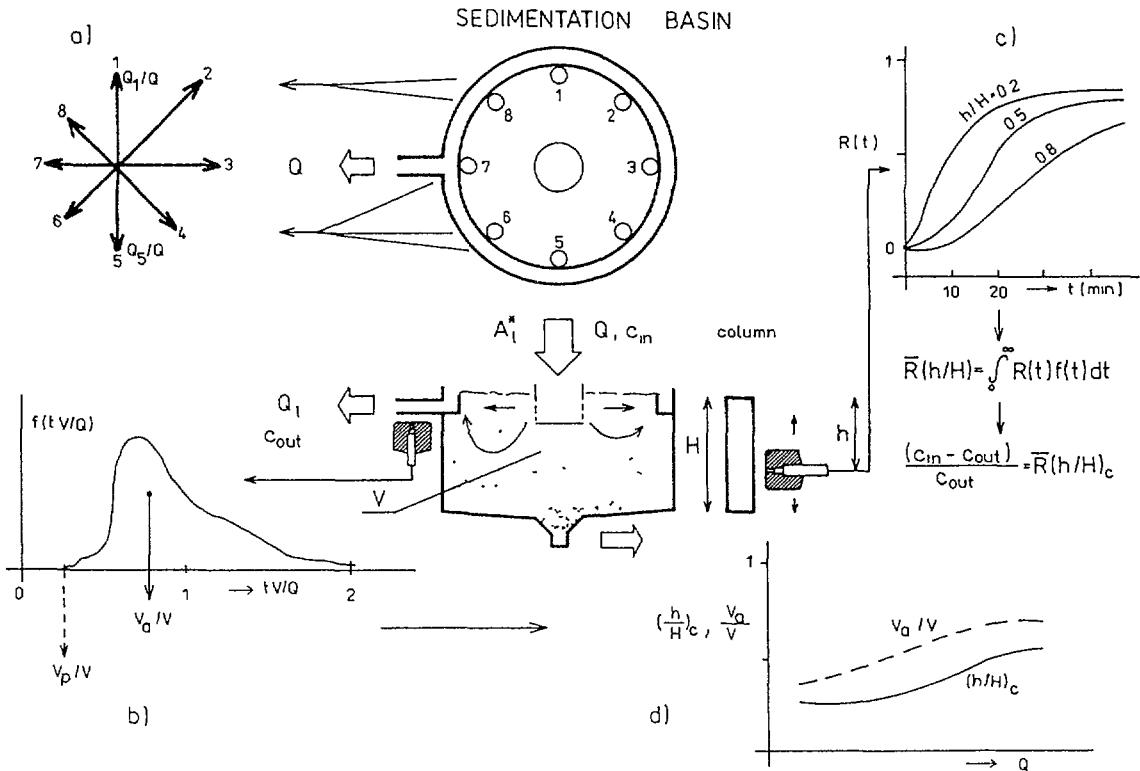


Fig. 6. Sedimentacion basin.

a) Spacial distribution of flow

Q - flow rate of liquid; α - 1-8 detectors

b) Residence Time Distribution

V_p - plug flow; V_a - active volume; V - basin volume;
 A_1^* - liquid tracer

c) Removal ratio

H - height of vessel; h - depth of measurement

d) Critical settling depth $(h/H)_c$

assume that this fraction will have the largest residence time of all fractions, the separation size d_{50} can be determined readily from measurement of the impulse response of fraction labelled by radiotracer which is measured continuously in the overflow and underflow.

As seen from the Fig. 5 (dependence of first moments on particle size), separation size d_{50} really corresponds to the largest value of the first moment of the impulse response.

4.1. Sedimentation

Even though the flotation methods have limited the use of sedimentation units, they are still an important operation at dressing of iron, wolfram and

zinc ores. Their operation is simpler, cheaper and safer than operation of flotation units from the point of view of pollution of environment.

In sedimentators radiotracer methods are used at evaluation of hydrodynamic characteristics which are obtained by measurements of impulse response of the liquid phase. They can be also used at evaluation of the separation (removal) efficiency; here, besides the impulse response, the so-called removal curves obtained by measurements with radiotracers in laboratory sedimentation columns (see Fig. 6) are also used.

Evaluation of hydrodynamics

The impulse responses are used for evaluation of deviations from ideal liquid flow at sedimentation, at evaluation of bypass, effective volumes, internal recirculation etc. Processing of the impulse response for calculation of these data is demonstrated in Fig. 6. More detailed information regarding the character of flow can be obtained by parametric analysis of model and experimental dependences of impulse response.^{1,2}

In large circular sedimentation units the method of impulse response is used also for evaluation of the spatial efficiency by measurement of concentration of indicator by detectors located at the periphery of the settler^{2,22} (see Fig. 6).

Evaluation of sedimentation in a certain depth

When it is possible to assume that sedimentation is a process of the first order then for the separation ratio in a certain depth the relation

$$R_h = \int_0^\infty (R_{h sc})_{sc} f(t) dt \quad [12] \text{ applies,}$$

where $f(t)$ is the impulse response of the liquid phase, $-(R_{h sc})_{sc}$ is the removal ratio which describes the drop in sediment concentration in a certain depth.

The removal factor $-(R_{h sc})_{sc}$ is evaluated by recording the concentration of radioactive particles in various depths $-h-$ of the statical column $-sc-$. The measuring method and obtained dependences are demonstrated in Fig. 6. From comparison of the removal factor calculated from Eq.12 with value obtained by measurement of concentration of sediment at the outlet c_{out} and inlet c_{in} i.e. $R = (c_{in} - c_{out})/c_{out}$ it is possible to calculate the so-called critical settling depth. This information together

with evaluation the active volume $-V_a-$ is used for determination of the optimum settling conditions.

4.2. Flotation

Flotation is obviously one of the most important operations used in dressing of ores. Foam flotation makes use of different physico-chemical properties of ore particles and waste rock, gangue, in liquid. After contact of particles with chemical compounds, the particles with suitable flotation properties in contact with air are wrapped up by air bubbles which carry them to the surface into the so-called foam layer. Particles with poor flotation properties remain in the suspension. The principle is depicted in Fig. 7.

Beside the above measuring methods such as impulse response (for evaluation of dynamic properties of phases) and integral method of tracer injection (for evaluation of separation coefficients and material balances of complex flows in units) parametric analysis of the RTD model of the flotation process gives important data. From the assumption that flotation is a linear

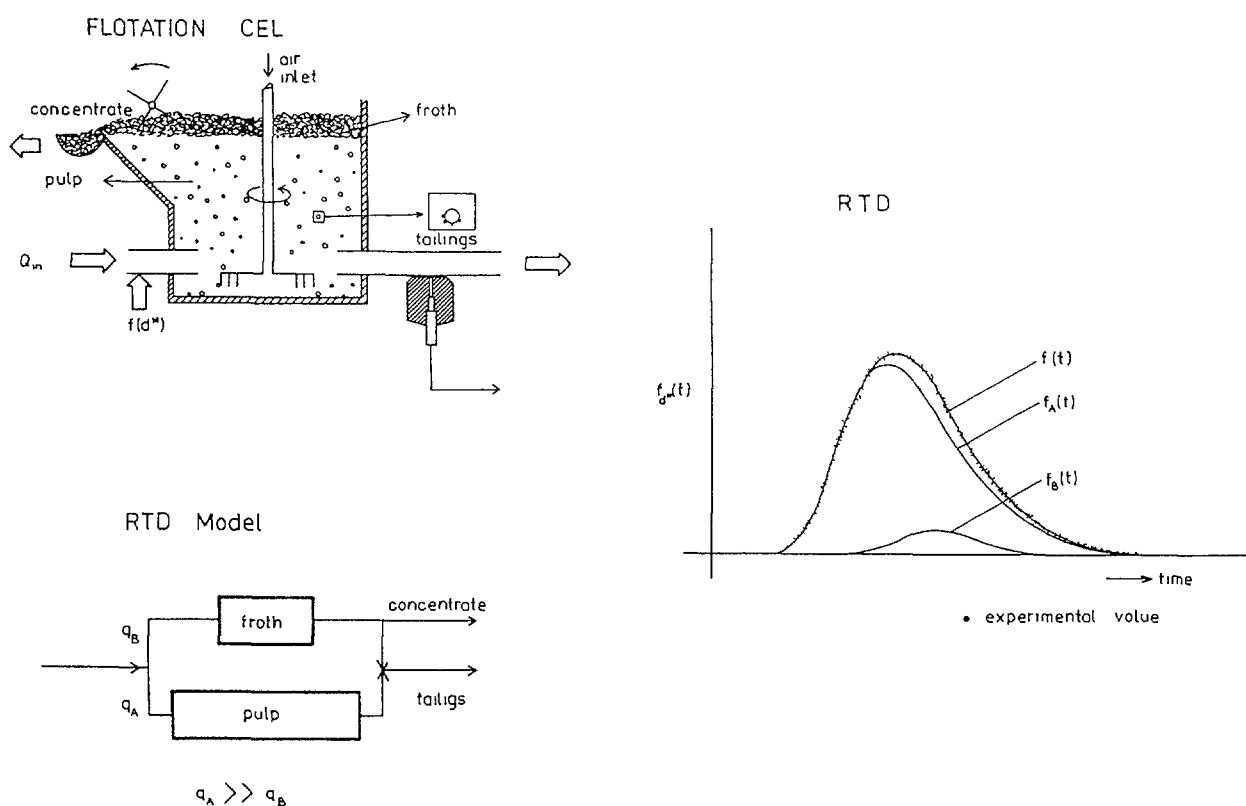


Fig. 7. Flotation.

$f(d^*)$ - input of tracer size fraction; $f_d^*(t)$ - RTD of size fraction measured in tailings

process, it is again possible to use the superposition of kinetic dependences and distribution functions of residence time i.e.

$$C_{\text{out}}(t) = \int_0^t C_0 \cdot \exp(-kt) \cdot f(t) dt \quad [13]$$

where $-C_{\text{out}}$ - is the concentration of floated particles in the flotation foam at the outlet from the unit, $-C_0$ - is the concentration at the inlet of particles into the flotation cell, $-k$ - is rate constant of flotation, and $-f(t)$ - residence time distribution.

For analysing the impulse response of ore particles measured at the outlet of tailings, use is made of the model of two parallel flows as demonstrated in Fig. 7, where the second smaller flow represents the dispersion motion of particles with partial residence in the space of the foam. Analysis of parameters of the model from the experimental dependence obtained for various fractions of material has enabled to make the correct modification of the process so that the loss of ore in the tailings is considerably reduced.

5. PRODUCT HANDLING

5.1. Hydrotransport

For successful control of mineral processes, it is important to continuously record the flow of solid particles such as raw material, ore or gangue transported between individual units in pipes in the form of aqueous suspensions. Electromagnetic flow meters and density meters (mostly based on absorption of gamma radiation) are used. These relatively expensive devices are frequently operated in industrial plants with considerable measuring errors (up to 20%).³³ Therefore it is necessary to check and calibrate them at actual operating conditions. Radiotracer methods enabling measurement of tracer concentration across the walls of the pipes in two points, measure the mean flow velocity of the continuous phase (see Fig. 8) and thus enable calibration of the flow meters. If the internal diameter of the pipe between the measured points is known, measurements can be made within the range of 1%.²

5.2. Equilizing ability of storage vessels

The chemical composition of ores in a continuous processes fluctuates considerably. This fluctuation can be suppressed by fixing a equilising vessel into the system.² The equilising ability of a vessel depends on its

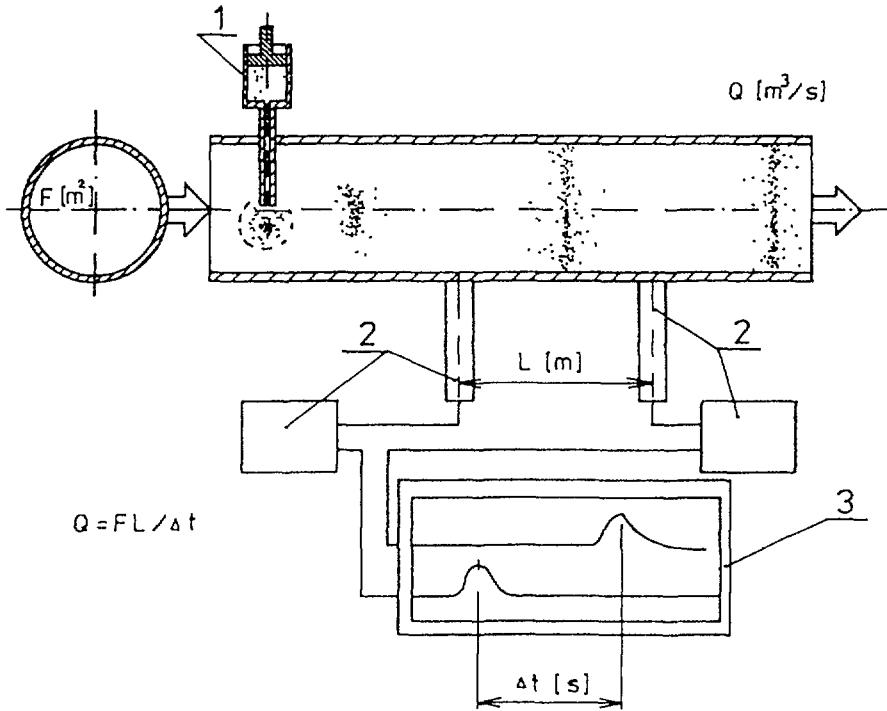


Fig. 8. Mean flow velocity of continuous phase.

1 - injektion of tracer; 2 - detectors; 3 - registration

capacity and on the pattern of flow of material through the unit. The flow pattern of material can be altered by reconstruction of the unit or e.g. by changing the operating conditions or by changing the type of the mixer, by changing the location of the inlet or outlet of material, by changing the speed of rotation of the mixer etc.

As egalisation is a linear process, the change of composition is determined by the convolution integral²

$$C_{\text{out}}(t) = \int_0^t C_{\text{in}}(t-u)f(u)du \quad [14]$$

where $-C_{\text{in}}(t)-$ is the time change of ore concentration at the inlet into the unit.

The egalisation ability of vessels depends on pattern of changes of $-C_{\text{in}}(t)-$, and on the mode of material flow which is easily determined by stimulus response method with radiotracer. Efficiency of egalisation is usually measured by use of variance of the studied fluctuated property at the outlet and inlet of the unit.²

$$n = 1 - \sigma_{\text{out}}^2 / \sigma_{\text{in}}^2 \quad [15]$$

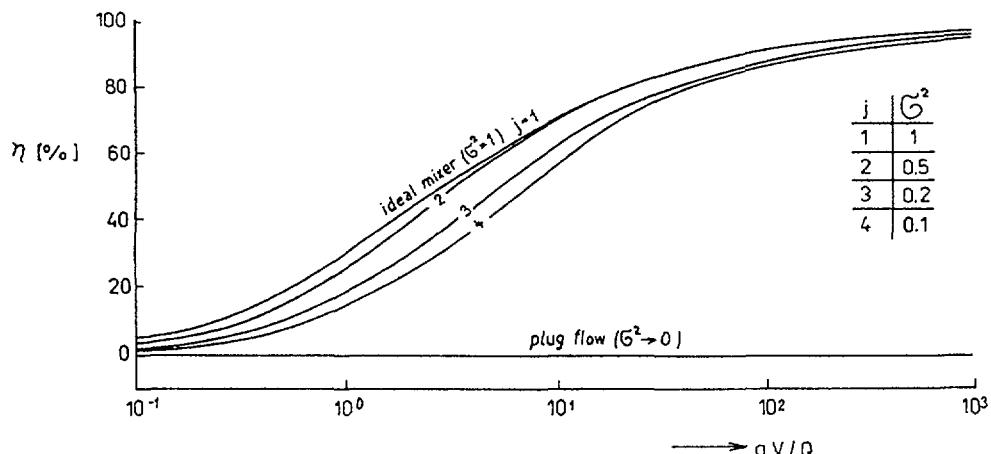
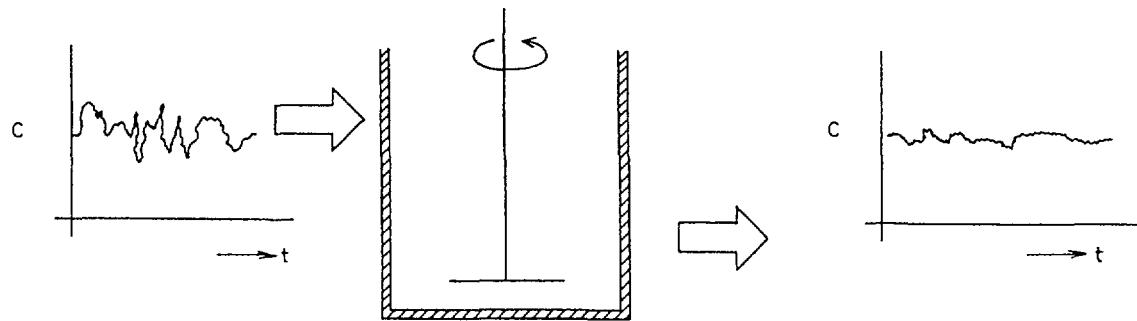


Fig. 9. Equalisation in storage vessel.

- efficiency of equalisation; V - active volume of vessel;
Q - flowrate; α - constant of autocorrelation function

For stochastic-random changes characterised by the constant of autocorrelation function α , there are (Fig. 9) demonstrated dependences of efficiency on dispersion of impulse response characterised by the reciprocal variance $-1/\sigma_f^2$ and the first moment t . As for t , the relation $t=V/Q$ holds, where V is the effective volume. It is obvious that efficiency increases with capacity of the unit and with dispersion of the impulse response.

6. APPLICATION OF DATA FOR SYSTEM ANALYSIS

The process taking place in one unit frequently cannot be studied separately as it is a part of the whole system which consists of a number of units interconnected by pipes. In some cases the data on the process are obtained only by an analysis of experimental data by use of computers. Special methods of identification are used e.g. for processes connected in series or

for flow processes which operate with recycle of the studied material. The necessary methods are presented in recent publications.^{1,2,11}

These methods enable the determination of the parameters of individual processes from measurements performed on whole systems consisting of grinding, classification and flotation operations. A typical simple system for mineral processing is a system of the rod mill in series with hydrocyclone with recirculation branch with ball mill.^{11,12,14} The optimization of the more complicated system of rod mill, spiral classifier, ball mills with recirculation, hydrocyclones and flotation cells has been also described.^{17,18}

For control of the whole process of ore dressing it is necessary to take note of the random time changes of concentration during treatment of material. This problem is frequently solved by assuming that the continuous monitoring and control of individual processes will enable control of concentrate quality. This requires expensive equipment for sampling a suitable amount of material and their automatic measurement, which is very expensive and often not very successful. The expensive continuous monitoring of certain properties can be eliminated by incorporating an equalisation vessel in the system or by suitable control of the mining process, etc.

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