MODERN TRENDS IN MINING GEOPHYSICS AND NUCLEAR BOREHOLE LOGGING METHODS FOR MINERAL EXPLORATION

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Abstract

This paper reviews recent developments in basic research and in the field practice of nuclear logging for mineral exploration, which have been published during 1974-1977, in about 200 papers from geophysical laboratories mostly in eastern Europe.

The main achievements in theory and experimentation in the following logging methods are presented: gamma-ray logging, XRF logging, Mössbauer effect, nuclear gamma resonance, neutron-neutron (resonance, epithermal and thermal) logging, spectrometric neutron-gamma logging, photon-neutron logging, die-away logging with pulsed neutron sources, and activation logging. Some problems of geostatistics applied to nuclear borehole logging also are described. For each logging method the fields of application reported during the last four years are given.

In the second part of the paper, the combined application of different nuclear logging methods for different groups of deposits is reviewed. The deposits discussed are: Iron-bauxite group – Fe, Mn, Al; Base metal group – Cu, Zn, Pb, Hg, Ba; Tin-rare metal group Sn, W, Be, Mo; Ultrabasic group: Cr, Ni; Gold group: Au, Sb, U; Sediments, evaporites and other types of deposit: S, K, B, phosphorite, apatite, fluorite, alunite. Some new possibilities in the future development of nuclear logging methods are also described.

The general conclusion is that the period 1974-1977 was characterized by a moderate development of theoretical research for different kinds of mineral nuclear logging (except for gamma and gamma-gamma methods). The practical application of XRF logging has been rather broad, the neutron methods are starting to be very promising, especially in the spectrometric version (both for radiative capture and activation). New neutron methods for uranium detection have been tried. The practical application of borehole neutron generators is still very limited, and has been confined to the search for mercury deposits. The first applications of the pulsed borehole photon generators for density logging have been reported. For each nuclear logging method the accuracy of the grade determination was at least comparable to that obtained by the usual chemical assay of cores.

Résumé

Dans cet article, l'auteur examine les derniers développements de la technologie des diagraphies nucléaires en prospection minérale (recherche fondamentale et applications sur le terrain), en se basant sur les résultats publiés de 1974 à 1977, dans près de 200 rapports provenant de laboratoires de géophysique dont la majeure partie se trouvent dans l'Europe de l'Est.

On y trouvera exposés les résultats les plus importants, en ce qui concerne la théorie et l'expérimentation, obtenus dans les méthodes de diagraphie suivantes: rayons gamma naturels, fluorescence X, effet Mössbauer, fluorescence nucléaire résonante, neutron-neutron (n. de résonance, n. épithermique et n. thermique), photoneutrons, spectrométrie du rayonnement gamma de capture, temps de relaxation neutronique (avec une source de neutrons pulsés), activation neutronique. On y aborde aussi quelques problèmes de géostatistique appliquée aux diagraphies nucléaires. Pour chaque méthode, les domaines d'application sont indiqués, tels qu'ils ressortent des travaux publiés pendant les quatre dernières années.

En deuxième partie, l'auteur donne des exemples d'application combinée des différentes diagraphies nucléaires dans différents groupes de gisements. Il s'agit des gisements suivants: le groupe fer-bauxite –Fe, Mn, Al; le groupe des métaux de base – Cu, Zn, Pb, Hg, Ba; le groupe étain-métaux rares – Sn, W, Be, Mo; le groupe ultrabasique – Cr, Ni; le groupe de l'or – Au, Sb, U; les gisements sédimentaires, d'évaporites et autres – S, K, B, phosphorite, apatite, fluorine, alunite. L'auteur décrit aussi quelques nouvelles possibilités dans le développement futur des diagraphies nucléaires.

On peut conclure en disant que la période de 1974 à 1977 se caractérise par une évolution modérée dans la recherche théorique des différents types des diagraphies nucléaires dans le domaine minéral (à l'exception des diagraphies de rayons gamma naturels et gamma-gamma). L'application pratique de la diagraphie de fluorescence X est assez répandue; les diagraphies de neutrons commencent à être assez prometteuses, surtout dans la version spectrométrique (spectrométrie du rayonnement gamma de capture et diagraphie d'activation neutronique). Pour la recherche de l'uranium, on a essayé de nouvelles diagraphies de neutrons. L'application pratique des générateurs de neutrons dans les sondages reste encore très limitée: en prospection minérale ils n'ont guère été utilisés que pour la recherche des gisements de mercure. On signale les premières applications des générateurs de photons à impulsions pour les diagraphies de densité. La détermination de la richesse d'un minerai est au moins aussi précise par les méthodes de diagraphies nucléaires que par l'analyse chimique des carottes.

INTRODUCTION

Nuclear logging methods are very sensitive to the presence of a given element in the multielemental system for this reason they are as convenient as the more common drillhole logging techniques for mineral exploration and mining. The three main groups of radiation sources used in practical applications are:

- Sources naturally existing in rocks, and those artificially introduced into drillholes:
- Gamma-ray sources,
- Neutron sources.

The gamma-ray sources due to the natural radioactivity of rocks are the most important in the first group.

The second group is composed of isotopic gamma-ray sources with a broad range of primary energies. Some accelerator-type gamma-ray sources are also included. Very soft gamma-ray energy (in the X-ray region) is used for X-ray fluorescence (XRF) logging for heavy elements and for the Mössbauer effect for tin ores. Gamma-ray energies in the region 100 to 300 keV are used in the so-called selective gamma-gamma method for detecting the presence of heavy elements (without distinguishing between them) in ores. Higher energies are used for density logging, the nuclear gamma resonance method (for Cu or Ni), and for the photon-neutron method (for Be mainly).

Among the neutron sources, the steady state and pulse operated ones have to be distinguished. The steady state neutron sources are usually of the α -Be type although the application of 252 Cf sources is becoming popular.

The pulsed neutron sources are usually those generated by the D, T reaction and are 14 MeV in energy. However, the application of these sources is not yet in common use because of the high cost of borehole neutron generators and the utilization of large diameter tools (except for the oil industry which is outside of the scope of this paper).

SUMMARY OF METHODS

Usually the development of any nuclear technique follows this sequence:

- 1. physical idea and feasibility of measurement,
- 2. laboratory experiments,
- 3. design and development of field equipment,
- 4. qualitative application,
- 5. establishing the theory for a given method,
- 6. solution of the so-called inverse problem to obtain the algorithms for quantitative interpretation,
- 7. quantitative applications.

The particular steps of this development can sometimes be changed into another sequence if new advances in apparatus design so dictate. Various nuclear methods used for mineral exploration and mining are classified at different stages in this development. Here we shall try to give a very short and simple description of the actual state-of-the-art for each method.

Steady state methods

In general the problem of quantitative interpretation and even the applicability of the method is linked with the solution of the equation:

$$R(\vec{r}) = \sqrt{\int G(\vec{r},\vec{r}^{1}) \cdot P(\vec{r}^{1}) \cdot d\vec{r}^{1}}$$
(1)

where $R(\vec{r})$ is the logging probe response at the point \vec{r} due to a given set of values of geological parameters P at the points $\vec{r}^1 \cdot G(\vec{r},\vec{r}^1)$ is the effect (for a given particular nuclear method) caused at the point \vec{r} by the presence of a given geological parameter P at the point \vec{r}^1 . The integration is over the whole space V at which the parameter P is different from zero. The goal is to find the set of values $P(\vec{r}^1)$ from a knowledge of the experimental values of R_{ex} at \vec{r} :

$$R_{ex}(\vec{r}) = R(\vec{r}) + \delta_R$$
(2)



GAMMA RAY LOG

Figure 11.1. Example of a gamma-ray log interpretation of a uranium deposit. Uncased borehole, diameter 132 mm, filled with drilling mud. Linear absorption coefficient taken for interpretation: $\mu = 0.09$ cm⁻¹, p = 4, (after Czubek and Zorski, 1976).

where $\delta_{\rm P}$ is a random error of the experimental value of the probe response R. The functions $G(r_{\rm y}r_{\rm 1})$, different for different nuclear methods, usually are not known exactly. Sometimes only a very rough approximation of their form can be postulated.

In view of these remarks the solution $P(\vec{r}^{1})$ of Equation 1, when the values $R_{ex}(\vec{r})$ are known, belongs to the so-called "improper" problems of mathematics (cf. Tikhonov and Arsenin, 1976).

Natural gamma-ray logging

The natural gamma-ray logging technique which has undergone development for many years especially for uranium and potassium ores, has the best quantitative achievements among all the nuclear logging techniques. But even for that technique, the problem of the exact theory of the method is not entirely solved. The state-of-the-art in this subject was recently reviewed by Czubek and Zorski (1976). Under some simplifying conditions about the function $G(\vec{r},\vec{r}^{1})$ in Equation 1, the depth distribution of the radioactive ore grade $q(z) \equiv P(\vec{r}^{1})$ along the borehole axis z with a given depth resolution Δh can be obtained. The simplifying condition mainly involves the nature of the build-up factor for scattered radiation. A unique, equivalent attenuation coefficient μ is postulated for the whole observed gamma-ray spectrum in the rock. The value of μ can depend upon the detectors and the gamma-ray windows used in the logging tool. Novikov et al. (1974, 1976) have found the values:

- $\mu = 0.032 \pm 0.034 \text{ cm}^2/\text{g}$ for uranium series
- $\mu = 0.028 \pm 0.032 \text{ cm}^2/\text{q}$ for thorium series
- $\mu = 0.05 \text{ cm}^2/\text{g}$ for potassium

which are essentially the same as were found earlier for the gross count of the whole scattered spectrum.

The final results of interpretation of a gamma-ray log can be presented as a step-wise function as shown in Figure 11.1. Here, for the elementary layer of thickness Δh , the gamma-ray intensity I_{∞} can be determined. This intensity, free of the influence of measuring parameters (such as logging speed, ratemeter time constant, detector dead time, borehole diameter, borehole fluid, activity of the neighbour layers, etc) is directly proportional to the grade (per unit weight of the natural wet rock) of radioactive material. This natural radioactivity can be related either to the uranium, thorium or potassium ore grade, or to the grade of some other mineral, whose concentration is sometimes correlated with the natural radioactivity of the rock. Such a correlation has been reported, for example, for phosphorites by Rudyk et al. (1974) where the correlation coefficient was about 0.95, or for alunites (Muravev and Yakubson, 1975).

The problems occurring in the quantitative interpretation of gamma-ray logs have not been fully investigated and solved. The existing systems of interpretation need to be compared in detail to define the most accurate. The problem of accuracy of interpretation was discussed by Varga (1975) for the interpretation procedure proposed earlier by Rösler. His procedure is based on the solution of an infinite set of linear equations using the matrix method. Varga found that for this method of interpretation the statistical error of the grade determination could be three times higher than the statistical error of the input data.

The gamma-ray logger, especially the spectrometric versions, is an excellent tool for potassium determination; however it also has great disadvantages when used for uranium grade determination. Uranium which is commonly in radioactive disequilibrium with its decay products, cannot be accurately determined without a good knowledge of the disequilibrium factor. This is the main reason why there has been an increased effort in recent years to find other possibilities of measuring uranium content.

Other theoretical problems related to the contribution of different radioactive series to the whole gamma-ray spectrum as recorded by the gamma-ray logger have been discussed by Arakcheev and Bondar (1975) in order to distinguish different lithologies.

Methods of using gamma-ray sources

Very broad and successful application of gamma-ray sources to exploration and mining control was recently reviewed in an excellent monograph by Ochkur (1976). The main progress in recent years has been in the practical application of XRF logging of multicomponent ores with up to four elements being simultaneously determined (Bolotova and Leman, 1976).

XRF logging

In the X-ray fluorescence assay, the measurements are performed either on outcrops or in boreholes. The principal method is the so-called "spectral ratio" with an internal self-standardization of background as described by Medvedev et al. (1973). This method avoids the influence of the XRF and primary scattered radiation of other elements on the registered energy line I(E) of the investigated element. When one takes the ratio

$$\eta = \frac{I(E)}{I(E_s)}$$
(3)

where $I(E_s)$ is the intensity of scattered radiation at energy E_s , it is possible to find a value of E_s , for which the η value for a given, fixed energy E of the XRF line is independent of the grade of the interfering elements. An example is given in Figure 11.2. Here the source used was $^{14.7}Pm$ ($E_{\gamma}=38$ keV) and the detector was a xenon proportional counter with a resolution of 16 per cent for the 22.5 keV line. The value of η for the antimony K_{α} line (E=26 keV) does not depend upon the admixtures of Fe, Ba or Pb in the alabaster when the E_s energy is chosen in the region of 37 to 38 keV.

There are a number of different XRF probe constructions. The one used by Zgardovskiy et al. (1974) is shown in Figure 11.3. Other constructions have been reported by Leman et al. (1975) and by Yanshevskiy et al. (1976a, 1976b) and were designed for variable borehole diameters. The logs can be recorded either in dry or in water-filled holes (Krasnoperov and Zvuykovskiy, 1976). Other designs have been reported by Meyer and Filippov (1974), Meyer and Rozuvanov (1974), Baldin et al. (1976), Christell et al. (1976), Landström (1976) and many others. Landström has measured Pb ore grade using the spectral ratio but without any artificial gamma-ray source. The K_{α} line of lead was excited by the natural rock radioactivity.

Some details of XRF apparatus, filters, etc. have been described by Chernyavskaya et al. (1975). The optimum parameters for XRF loggers have been discussed by Nakhabtsev (1977) and Yanshevskiy et al. (1976a, 1976b). The application of solid-state detector to this kind of logger has been presented by Baldin et al. (1975, 1976).

In order to indicate how the system with background self-standarization works for the simultaneous determination of four elements, the results of Bolotova and Leman (1976a) are presented in Figures 11.4 and 11.5. The relative accuracy of the XRF logger is not very high being within the limits of \pm 25 per cent for any particular point assay. Such a low accuracy is



Figure 11.2. Determination of the self-standardized background in XRF logging for antimony ores (in alabaster) (after Medvedev et al., 1973).



Figure 11.3. Measuring head of the XRF probe SRPD (after Zgardovskiy et al., 1974).



Figure 11.4. Separate determination of Sn, W, Cu, and As by XRF logging (after Bolotova and Leman, 1976a).



Figure 11.5. Comparison of XRF assaying with the chemical sampling results, (after Bolotova and Leman, 1976a).

due mainly to the very short range of penetration of the XRF method and variable ore grain size. The problem of the proper averaging or filtering of the raw data together with the physical principles of penetration of gamma radiation through heterogeneous rock media remain, therefore, a main research subject in many geophysical laboratories. In spite of rather low accuracy, the limit of detection of XRF methods is quite good. It depends, of course, upon the particular logging equipment being used and upon the investigated element. Bolotova and Leman (1976b), for example, give the following limits of detection: 0.05% for Sn, 0.1% for Cu, 0.1% for WO₃, 0.01% for Mo, 0.1% for As with the final accuracies being comparable with those obtained by chemical analysis.

The theoretical problems of XRF logging concerned with the cross-sections for scattered radiation have been discussed by Pshenichnyy and Meyer (1974, 1975) and Nakhabtsev (1974, 1975).

Mössbauer effect

Methodological problems similar to those of the XRF method are experienced in the practical application of the Mössbauer effect to geophysics. This effect is applied in the field and in logging practice mainly to tin ores (Ochkur, 1976; "Nuclear Geophys. Assay...", 1976; Goldanskiy et al., 1974). Three count rates are measured in this method:

I(O) – scattered radiation when the source velocity is v = O

I(∞) – scattered radiation when the source velocity is ν = ∞ in practice for tin ores, this velocity is ν = 2 mm/s,

I_b – background.

The parameter

$$\varepsilon_{s} = \frac{I(O) - I(\infty)}{I(\infty) - I_{b}}$$
(4)

depends upon the tin grade of the ore.



Figure 11.6. Decay scheme of Sn-119 m isotope, Energy in keV (after Stevens and Stevens, 1975).

The source of the resonance radiation is $^{119}{\rm mSnO}_2$ of the activity of the order of 10^8 gammas/s. The decay scheme of this isotope is given in Figure 11.6. To eliminate the 65.66 keV line, which is almost completely converted, which gives the ${\rm K}_{\rm Q}$ line of Sn of energy 25.27 keV, the source is covered by a palladium sheet of thickness of 100 to 120 mg/cm². The source is vibrated by a bimorphous piezoelement using the polarized ceramic system of PbZrTiO3. For detection purposes, a special resonance scintillation detector is utilized. Logging is performed step by step in 5 cm intervals. The range of investigation is of the order of 100 mg/cm².

Gamma-gamma methods

When a gamma-ray source is imbedded in a rock medium, the scattered gamma-ray spectrum reflects different properties of the rock surrounding the borehole. The whole spectrum is usually a function of the geometry of measurement such as borehole radius, type of shielding, presence or absence of drilling fluid, etc. In the energy region below 100 keV, the intensity of this spectrum is strongly influenced by the X-ray fluorescence of the rock and probe elements.

The measurement of scattered gamma radiation above the 100 keV energy level is the principal goal of the so-called gamma-gamma methods. They are used in geophysics in two distinct versions: for the measurement of density and in selective gamma-gamma methods. The first application is based on the measurement of the scattered gamma radiation from a relatively high energy gamma-ray source usually ¹³⁷Cs or ⁶⁰Co in the energy region above 200 or 300 keV. For this energy region the amount of scattered radiation is closely related to the bulk density of the scatterer, i.e. the rock. When ore bulk density is well correlated with ore grade, this type of measurement can be used to estimate ore grade. Sometimes ore density is used as one of the parameters in the ore grade determination by means of the multivariational correlation analysis.

The low energy region of the scattered gamma-ray spectrum, between 100 and about 300 keV, depends upon the photoelectric absorption properties of the rock. Therefore, this part of the spectrum is used in the second application of the method i.e. the selective gamma-gamma method. It is sensitive to the presence of heavy elements in the rock, or to the change in the equivalent atomic number Z_{eq} of the rock (Czubek, 1966; 1971). When the ores are of the mono-elemental type, this log may be used to determine ore grade. To negate the influence of noncorrelated density variations on the selective gamma-gamma log results, the so-called P_z technique was introduced (Czubek, 1966). P_z is simply the ratio of the high energy part to the low energy part of the scattered spectrum. The P_z parameter is a strong function of Z_{eq} and is relatively insensitive to variations in rock bulk density. In dry boreholes, the shape of the scattered spectrum near the source energy is a strong function of the borehole diameter. By taking the ratios of intensities in the energy region above 400 or 500 keV Aylmer et al., (1978); Charbucinski et al., (1977); and Eisler et al., (1976) were able to correct the P_z and density values for the influence of borehole diameter.

The density method has been used for iron ore grade determination by Szymborski (1975) with an accuracy of 3.4 per cent of iron and by Ochkur et al. (1976). This logging technique was also used by Koshelev et al. (1976) to distinguish apatite-nepheline ores from spheno-apatite ores, and P_2Os was determined in apatite-nepheline ores by Startsev et al. (1975a) by the correlation with density. In a qualitative way this logging technique has been used by Kozlov et al. (1975a, 1975c) on copper deposits. The lithological differentiation of the bauxite ores by means of density logging was reported by Shishakin et al. (1974). Density logging has been used together with other kinds of gamma-gamma logs by Eisler et al. (1976), Charbucinski et al. (1977) and Aylmer et al. (1978) to determine iron content very precisely in West Australian deposits.

The selective log was used by Gera (1974) for the localization of quartz veins in gold deposits using backscattering radiation from a 170 Tm source (52 and 84 keV) with a source-detector distance of the order of 50 mm. The same method, but using a higher energy 137 Cs source (the registered window was 130 to 150 keV) was used for lead and barium assays by Shmonin et al. (1976a) as well as for the analysis of a lead-zinc deposit by Shmonin et al. (1976b). Tungsten grades higher than 0.2 per cent were investigated with this method by Kuchurin et al. (1976). It was also possible to determine the iron content of skarns with an accuracy of 2.5 per cent (Senko and Zorin, 1975).

The majority of applications of the gamma-gamma methods were more or less purely experimental and without a deep physical knowledge of the problems from the point of view of the gamma-ray transport in heterogeneous rock media. The real need for such knowledge stimulated research work in this subject. Gulin (1975) has published a monograph on gamma-gamma methods where the most important physical features of the method are given. His results have been obtained mainly by a theoretical approach using the Monte-Carlo technique for calculation of the space-angle-energy distributions of scattered photons in actual tool-borehole-rock systems. The other problems concerned with angular distribution of scattered radiation, together with the question of how to distinguish thin layers, were considered by Popov et al. (1974), Popov and Vishnyakov (1974), Utkin et al. (1974, 1975, 1976), Lukhminskiy and Galimbekov (1975), Galimbekov et al. (1976), Galimbekov (1975), and Utkin and Ermakov (1975).

In order to characterize the ores of minerals using the gamma-gamma method, their equivalent atomic number Z_{eq} and their heterogeneity have to be known. The Z_{eq} defined by Czubek (1966) has been investigated experimentally by Artsybashev and Ivanyukovich (1974). They obtained the same results as Czubek obtained theoretically for the gamma-ray energy range from 30 to 2500 keV. By the Monte-Carlo technique, the problem of Z_{eq} was also treated by Lukhminskiy and Galimbekov (1975) with similar results.

Rock heterogeneity problems were treated usually by the Monte-Carlo technique, by Leman et al. (1975b), Lukhminskiy (1975), Umiastowski et al. (1976) and Umiastowski and Buniak (1977). For a given grade of heavy mineral, the effect of its heterogeneity has the same influence on scattered radiation as the decrease in grade for homogeneous ore. For some range of gammaray energies, the concurrent, reciprocal influence of grade and grain size on scattered radiation becomes so strong that grade determination is impossible unless the grain size, or heterogeneity, is fixed or known. These theoretical results, confirmed by experiments, were used by Charbucinski and Umiastowski (1977) to develop experimental tools for selective gamma-gamma measurements on lead-zinc ores.

In order to calculate the probe response for a given ore grade in the selective gammagamma method, Galimbekov and Soboleva (1976) have established a special computer program called MOK-22.

Simultaneously with the development of theory and experimental research, progress in the design of new tools was made. Utkin and Burdin (1975) have discussed the necessary specifications for digital recording in order to obtain a given accuracy in the density measurements. New small diameter borehole tools were designed for the applications of both selective and density versions of the method (Voskoboynikov et al., 1975a, 1975b; Utkin, 1975). The most important requirement for each tool is good spectral stability. This problem was solved using either the property of the stable shape of the scattered gamma-ray spectrum (Utkin, 1975) or by using a light emitting diode as the energy mark for the spectrometer (Bakhterev et al., 1975b).

Parallel to an improvement in the performance of the "classic" gamma-gamma tools, an effort was made to construct a borehole gamma-ray generator. This project was carried out in the All-Union Science and Research Institute of Nuclear Geophysics and Geochemistry (VNIIYaGG) in Moscow (Belkin and Kolesov, 1976; Belkin et al., 1975; Grumbkov et al., 1975, 1976). It consists of a pulsed X-ray tube with an average energy of photons equal to 250 keV and with the maximum energy between 550 and 600 keV. The total photon output (for photons of energy above 200 keV) is of the order of 10^{12} photons per second. The sensitivity of the density measurement with this generator was doubled due to the increase in the source-detector spacing compared to the usual density tools.

Nuclear gamma resonance method

The nuclear gamma resonance technique (Sowerby and Ellis, 1974; Sowerby, 1974) also belongs to this group of logging methods. As opposed to the Mössbauer effect, the energy levels of nuclei (at the level of about 1 MeV) are excited by gamma rays of the same energy as of the nuclear level itself. The energy of the excitation source is increased by increasing the thermal motion of the atoms. The difference in energy, ΔE , needed to observe this resonance

$$\Delta E = \frac{E_0^2}{M \cdot c^2}$$
(5)

where E_0 is the energy of the nuclear level, M is the atomic mass and c is the velocity of light, is obtained by irradiation of the surrounding rock with the gamma-ray source being in the vapour state. This method was tested in Australia under laboratory conditions on copper (with 5 Ci* of the 65 Zn source) and nickel (with 0.7 Ci of the 60 Co source) ores. This method needs very active gamma-ray sources of the order of few curies which have to be transformed by heating them into the gaseous state. Probably for this reason no field application has been reported as yet (Sowerby et al., 1977)

Photo-neutron method

The photo-neutron method is usually used for beryllium exploration because of its low energy threshold for this reaction (Fig. 11.7). This energy threshold is achieved using a 124 Sb gamma-ray source. The other isotopes, deuterium and 13 C are interesting for geohydrology and petroleum geology applications but are outside the scope of this paper. The possible detection of other heavy elements that have relatively low energy thresholds for this reaction (in the vicinity of 5 MeV) will be possible when borehole generators of gamma rays with sufficiently high photon energy are available.



Figure 11.7. Energy thresholds for γ , n reactions for natural isotopes.

The theoretical approach to the photo-neutron method was made by Krapivskiy and Saltsevich (1975) using analytical methods for photon and neutron transport (age, diffusion and age-diffusion approximations). Gorev et al. (1975a) have used the Monte-Carlo technique of calculation for the same purpose and obtained a much better agreement with experiment than Krapivskiy did. Berzin et al. (1975), have nvestigated the pulsed photo-neutron method on a layered model using the betatron as a source of gamma rays of energy between 5 and 30 MeV.

It is very difficult to follow all the applications of the gamma source methods in different countries. Just to give some idea of how large the application of the XRF assay technique is in the exploration and mining of minerals, we can present some figures reported by Osmonbetov et al. (1976) for the Kirgiz Republic, USSR: In 1975, more than 75 000 assays were performed for Sn, W, Sb, and since 1970 more than 400 000 were made. The cost of one XRF assay is between 2 and 4 times cheaper than a chemical assay which saves a lot of money in exploration and mining. No similar figures have been published for other methods.

Neutron methods

The main development in the neutron methods of borehole logging has been carried out by the petroleum industry. These achievements are of some use for mineral exploration, but some new effects should also be considered. The very sophisticated methods developed in the petroleum industry for the calculations of neutron flux in rock can be utilized in mineral exploration, especially when the rock medium is weakly absorbing. On the other hand, some new nuclear reactions should also be taken into account. The possible presence of strongly absorbing elements requires some different approaches to the problems of neutron transport. The heterogeneous structure of the rock media, when the absorbing elements are concentrated in some grains whereas others are free of them, presents new difficulties for theoretical considerations. If all of these problems were solved, a good knowledge of the physics of a given logging device could be obtained which would permit the optimization of the logging tools.

In the design of logging tools, it is not only a question of using different borehole diameters or borehole fluids from those used in oil fields, but very often the neutron sources, detectors, source-detector spacings, filters, etc. should be specially chosen for a given type of neutron method to be applied on a given mineral deposit. In spite of the foregoing remarks, the fundamental understanding of the physics of most of the phenomena listed above and already achieved in the petroleum industry is of great importance and help in the development of neutron methods for solid mineral exploration.

It is not possible to present a very detailed and distinct classification of neutron methods. Each element that occurs in a given type of deposit and in given borehole conditions requires its own specific discussion. In general the neutron methods can be divided into two groups: the first utilizing steady state sources, and the second utilizing pulsed neutron sources. In the first group, the measurement of a given product of neutron interaction with the rock material is observed at a given distance from the source along the borehole axis. In the second group, almost the same interaction products are observed in some time sequence related to the time sequence of the neutron source pulsing. This kind of measurement permits the measurement of some physical phenomena which are not obtainable using the steady state methods.

When the steady state source is used the following phenomena, which can be used as logging parameters, can be observed in the borehole at a distance z from the source:

- 1. Epithermal neutron flux of slowed-down neutrons.
- 2. Resonance neutron flux of slowed-down neutrons.
- 3. Thermal neutron flux from the thermalized slowed-down neutrons.
- 4. Gamma radiation from the radiative capture of resonance neutrons.
- 5. Gamma radiation from the radiative capture of thermal neutrons.
- 6. Gamma radiation from activation by fast neutrons.
- 7. Gamma radiation from activation by thermal neutrons.
- 8. Fission neutrons (prompt or delayed) when the uranium or thorium series are present.

The possibility of detection of gamma radiation from the inelastic scattering of fast neutrons or from activation by resonance neutrons is of less importance here and gives rather an increase in the background than the measurable effect. When induced gamma radiation is detected with the spectrometric tool, especially when a solid-state detector is utilized, the possibility of detection of separate elements increases considerably.

With the pulse neutron source, the phenomena accessible for observation are:

- 1. Die-away curve of epithermal neutrons.
- 2. Die-away curve of thermal neutrons.
- 3. Die-away curve of photons from radiative capture of resonance neutrons.
- 4. Die-away curve of photons from radiative capture of thermal neutrons.
- 5. Photons from inelastic scattering of fast neutrons.
- 6. Photons from activation by fast neutrons.
- 7. Photons from activation by thermal neutrons.
- 8. Fission neutrons (prompt or delayed) when the uranium or thorium series are present.

The detection of the phenomena listed above for pulsed neutron sources is always performed in the time windows which are correlated with the pulsing of the neutron beam.

Let the tool response for all phenomena listed above be R. The R value is always observed inside the borehole of a given geometry. In this case the tool response R can be presented in the form:

$$R(z) = R_{\infty}(r=z, P) \cdot \left[1 + S(r_B, z, P, P_B)\right]$$
(6)

where $\mathsf{R}_{\infty}(\mathsf{r=z},\mathsf{P})$ is the tool response when the borehole radius is $r_B=0$ at a distance r=z from the point neutron source. The rock medium is characterized by a set of geological parameters P (chemical composition of the rock, ore grade, density, etc.); $\mathsf{S}(r_B, \mathsf{z}, \mathsf{P}, \mathsf{P}_B)$ is the borehole influence function; P_B is the set of the physical and chemical parameters for the borehole. The main objective in designing a given logging tool for a given method is to minimize the function $\mathsf{S}(\ldots)$ and to get it as close as possible to zero. When this is achieved the tool response R is a strong function of the geological parameters P which are the object of investigation using a given method.

The behaviour of the function S(...) will not be discussed because of its complexity. Suffice it to say that for each method it can be minimized; attention will be focused on the principal value of the tool response, namely the $R_{\infty}(r=z,P) = R_{\infty}(r,P)$ functions.

Very sophisticated methods of description of the neutron and gamma-ray transport in the rock media are used when neutron and neutron-gamma logs are considered. For neutrons, the most common approximations of the Boltzmann transport equation used are: age and multigroup diffusion approximations, Greuling-Goertzel approximation, Monte-Carlo techniques

(Shimelevich et al., 1976; Postelnikov et al., 1975; Drozhzhinov et al., 1975), P_2 approximation (Kozachok 1975; Kozachok and Riznik 1977), whereas for gamma rays: radiative and build-up factor approximation (Shimelevich et al., 1976), and diffusion approximation (Davydov, 1975a; Davydov et al., 1976) are used.

Steady state neutron methods

The steady state neutron methods are used for two different purposes:

- 1. as lithology and porosity logging in the petroleum industry,
- 2. as a logging tool which gives information about ore grade.

In the latter case, the grade is determined either from the general decrease of the epithermal resonance or thermal neutron flux when the grade of the absorbing elements is increased, or from the increase of a given gamma-ray line from radiative capture or activation connected with the increase in grade of a given element.

When the neutron flux \emptyset is measured i.e. the $R_{\infty}(r,P)$ function in Equation 6, its general behaviour is:

where Q is the source neutron output, Σ_a is the total absorption cross-section of the rock and the $F_1(\ldots)$ function depends upon the source-detector spacing r, slowing-down (L_s) and diffusion (L) lengths for neutrons in the rock medium, which translated into geological language means that it depends upon the lithology, bulk density and porosity of the rock. These latter factors being constant, the neutron flux \emptyset becomes a function of Σ_a , which in the case of only one anomalously absorbing element "x" present in the rock is:

$$\Sigma_{a} = \rho \cdot N_{o} \cdot \left[(1-p) \cdot (\sigma_{a}/A)_{R} + p (\sigma_{a}/A)_{x} \right]$$
(8)

where ρ is the bulk density, N_p is the Avogadro number, p is the grade of a strongly absorbing element x with microscopic absorption cross-section σ_a and atomic weight A. $(\sigma_a/\Lambda)_R$ is the average σ_a/A ratio for the barren rock. Finally the relative effect of the presence of an absorbing element observed in neutron flux is:

$$\frac{\phi(\mathbf{r},\mathbf{p})}{\phi(\mathbf{r},\mathbf{p}=\mathbf{O})} \approx \frac{1}{1 + \mathbf{p}\left[\left(\sigma_{a}/A\right)_{x}/\left(\sigma_{a}/A\right)_{R}-1\right]}$$
(9)

which can be considered as the first approximation for the calibration curve. When $(\sigma_a/A)_x > (\sigma_a/A)_R$, the calibration curve very quickly reaches the value zero, even for a very low value of the grade p. For common rocks, for which the macroscopic absorption cross-section is in the range 4.10⁻³ cm⁻¹ $\leq \Sigma_a \leq 20.10^3$ cm⁻¹, the $(\sigma_a/A)_R$ value is in the limits 2 mb $\leq (\sigma_a/A)_R \leq 10$ mb (milibarn); for example for boron $(\sigma_a/A)_B = 73.47$ barns = 73.47 10³ mb. To avoid any difficulty with a lack of probe sensitivity when logging higher ore grades, registration of higher energy neutrons is recommended because for the higher neutron energies the ratio $(\sigma_a/A)_X/(\sigma_a/A)_R$ becomes lower. This was the reason for the application of the resonance neutron-neutron method for boron deposits (Vakhtin et al., 1973, 1975) where it was possible to extend the measuring range up to 16 per cent of B₂O₃, instead of 4 per cent for the thermal region. An example of such a log is given in Figure 11.8, where the detector was the sandwich-type resonance detector described by Vakhtin et al. (1972).

This method was also used in the epithermal version by Grigoryan (1975) on copper deposits to distinguish the homogeneous or heterogeneous quality of ores. Fatkhutdinov and Urmanov (1975), and Fatkhutdinov et al. (1976) used it on a Hg-Sb deposit using Cd-In filters. For the multielemental gold deposit in East Zabaykal, Kuchurin et al. (1976) have used the thermal version of this neutron-neutron logging tool. Ochkur et al. (1976) report the application of both the thermal and epithermal versions on chromite and manganese deposits. Krapivskiy (1976) has used a combination of the thermal neutron method with the gamma-neutron method for the determination of lithium in boreholes. The accuracy of this assay was comparable to that of chemical assays.

When the neutron-gamma log of radiative capture or activation is taken into account, the neutron flux given by Equation 7 at the point \vec{r}^1 interacts with the rock matter at a rate given by the value of the macroscopic cross-section Σ for that reaction (radiative capture on a given isotope or activation, etc.). The gamma photon of a given energy originates at the elementary volume $d\vec{r}^1$ around the point \vec{r}^1 and it has a probability $G_2(|\vec{r} - \vec{r}|^2)$ of reaching the point \vec{r} at



Figure 11.8. Boron determination in a magmatic limestone lithology by neutron resonance logging. Po-Be source: $4.3x10^6$ n/s. Cd-Rh-In-Tu-Ta-Ag filter (after Vakhtin et al., 1973).

which the detector is situated. This being valid for the elementary volume $d\vec{r}^1$, it can now be integrated over the whole space giving the response N_{γ} of the gamma-ray detector:

$$N_{\gamma}(\vec{r},P) = Q \cdot \frac{\Sigma_{r}}{\Sigma_{a}} \cdot F_{2}(\vec{r},L_{s},L,\mu)$$
(10)

Here again the function $F_2(\ldots)$ is a weak function of the ore grade and a rather strong function of the moisture content, bulk density and lithology. μ is the gamma-ray absorption coefficient for a given rock. For the same type of ore, the functions $F_1(r,L_g,L)$ and $F_2(r,L_g,L_s)$ are very similar in form. Therefore, when one needs to eliminate the influence of the variable porosity or bulk density on the logging data, the ratio

$$\frac{N_{\gamma}(\vec{r},P)}{\phi(\vec{r},P)} \approx \text{ const. } \Sigma_{r}$$
(11)

is recommended.

The reaction cross-section on the element "x" is given as:

$$\Sigma_{\mathbf{r}} = \mathbf{p} \cdot \mathbf{a}_{\mathbf{x}} \cdot (\sigma^{\mathbf{r}} / \mathbf{A})_{\mathbf{x}}$$
(12)

where a_x is the abundance of the isotope of element x on which the reaction with the crosssection σ^r is going on. Similarly to Equation 9, the net calibration curve i.e. when the background gamma radiation is subtracted, for the neutron-gamma methods is:

$$N_{\gamma}(\vec{r},p) \approx \text{const} \cdot \underbrace{p}_{(\sigma/A)_{R}}_{a_{\chi}} + p \cdot \{1 - \frac{(\sigma/A)_{R}}{a_{\chi}} \cdot (\sigma^{\Gamma}/A)_{\chi}} \}$$
(13)

Here again, when

the calibration curve given by Equation 13 is saturated for a low p value and the method is not convenient in this case for use in the quantitative application. The normalization given by Equation 11 does not help too much in this case, because the background is usually too high in comparison with the net effect.

Bakhterev and Senko-Bulatnyy (1975) have used the neutron-gamma spectrometric log on nickel deposits. The calibration factor in Equation 13 was influenced by the iron content, thus the second energy window for measurement of the iron line served as the correction for the calibration factor.

Chrusciel (1976) has performed some laboratory experiments with the solid state detector to determine the detection limits of W, Ti, Ni, Mn, Cu and S in the borehole geometry for radiative capture. His results were next applied by Niewodniczanski and Palka (1976) and Niewodniczanski et al. (1977) for the determination of the sulphur content in boreholes using the spectral ratio from the radiative capture of thermal neutrons and the NaI(T1) detector. Similar work was done by Blinova et al. (1974). Much more sophisticated neutron-gamma spectrometry was used by Eisler et al. (1977) for iron determination in blast holes in Australian deposits. Egorov et al. (1974), Sokolov et al. (1975), Afanasev et al. (1974) and Balakshin and Kravchenko (1976) have used neutron-gamma spectrometry in different combinations for mercury determination in boreholes with a precision between 0.02 to 0.08 per cent of mercury. To determine chromite in boreholes, Ochkur et al. (1976) have used a combination of neutron-gamma spectrometry with density and neutron (thermal) logs.

Fundamental theoretical and experimental research in the application of neutron methods to absorbing media i.e. for ores have been done by Krapivskiy and Brem (1975a, 1975b), Postelnikov et al. (1975), Drozhzhinov et al. (1975), Egorov et al. (1975), Fatkhutdinov and Urmanov (1975), Kozachok (1975), and Kozachok and Riznik (1977).

Pulsed neutron methods

Pulsed neutron methods using the borehole neutron generator have no wide application, as yet, in mineral exploration. One reason is the difficulty in the availability of small diameter tools needed for the small diameter boreholes usually drilled in mineral exploration. The other reason is the high cost of neutron generators.

The time sequence of different radiations occurring in the rock space when a fast neutron burst is injected is given in Figure 11.9. Almost instantly with the neutron burst, the photons resulting from the excited states of nuclei by the inelastic scattering of fast neutrons can be observed. This technique, used sometimes in the petroleum industry, and very sensitive, especially for heavy elements, to the presence of different elements, has been not yet reported in any mineral exploration application.

Following the neutron burst is a transition time zone, t_s , where the neutron flux from the burst is influenced by the proximity of the borehole and by the properties of the tool, and is not in equilibrium. During this period the epithermal neutron flux disappears. Following the transition time zone is the die-away time zone, where the thermal neutron flux from the slowed down neutrons becomes the most important component. During this period (of the few neutron lifetimes τ , with $\tau \gg t_s$), the thermal neutron flux is the carrier of information about the environment absorption cross-section Σ_a . The logarithmic time decrement λ of the neutron flux $\phi_{th}(t)$ is directly related to the rock absorption cross-section $\Sigma_{a,R}$:

$$\lambda = \frac{d \ln \phi_{\text{th}}(t)}{dt} \approx \sim \Sigma_{a,R}$$
(14)



Figure 11.9. Behaviour of different radiations in the pulsed neutron methods during one cycle.

This is only true, however, when the absorption cross-section $\Sigma_{a,R}$ of the rock is lower than the absorption cross-section $\Sigma_{a,B}$ of the borehole. In terms of geological practice, it means that for mineral exploration the measurement of $\Sigma_{a,R}$ is not possible when the boreholes are dry. Otherwise the S(...) function in Equation 6 becomes much higher than 1. When the borehole is highly absorbing the determination of the grade p of the absorbing element according to Equation 8 is possible, or the localization of the mineralized zone may be made cf. Kashkay et al. (1976) for mercury deposits.

During the same die-away time zone, the thermal neutrons being captured by nuclei give the radiative capture gamma rays. When one observes a given energy line or energy range, the presence and the grade of a particular element can be determined. The problem, however, is rather complicated because of the complexity of the physical phenomenon. Due to the instant emission of the capture gamma lines there is no time shift between the pulsed neutron-neutron and neutron-gamma logs.

The pulsed thermal neutron flux in the infinite medium is:

where D is the diffusion coefficient of thermal neutrons in the rock medium, and v = 2200 m/s is the velocity of thermal neutrons, Q is the fast neutron output. By similar reasoning as for the neutron-gamma method, the time distribution of the neutron capture gamma radiation is with the scattered gamma-ray background subtracted given by

$$N_{\mathbf{Y}}(\mathbf{r},t) \approx \nu \cdot \Sigma_{\mathbf{n}\mathbf{Y}} \cdot e^{-\mathbf{t} \cdot \nu \cdot \Sigma_{\mathbf{a}}} \cdot F_{\mathbf{3}}(\mathbf{r},\mathbf{L}_{\mathbf{s}},\mathbf{L},\mathbf{D},\boldsymbol{\mu},t)$$
(16)

where again the function $F_3(\ldots)$ depends upon the source-detector distance, rock lithology, its moisture content or porosity and bulk density. $\Sigma_{n\gamma}$ is the macroscopic cross-section for the emission of a given gamma-ray energy line due to the radiative capture of thermal neutrons and it is directly related to the grade of the element being investigated. For strongly absorbing elements, when $\Sigma_{n\gamma} \approx \Sigma_{a}$, a special delay should be applied to the measurement time window to obtain unequivocal correspondence between the pulsed neutron capture gamma-ray flux and ore grade. This kind of measurement has been reported, as yet, only for mercury detection (Nikulin et al., 1976).



Figure 11.10. Activation Cu-64 log from the Tash – Tau deposit (after Bakhterev et al., 1975).

The last time zone of the pulsed neutron log cycle (Fig. 11.9) belongs to gamma radiation from the radioactive decay of nuclei activated by fast and/or thermal neutrons during the earlier time zones and during the previous cycles. The decay periods $T_1/2$ of activated nuclei, being much longer than the time between two consecutive neutron bursts, provides the possibility of observing this activation effect as a build-up of consecutive cycles which would increase the sensitivity of the method. Such a mode of detection introduced first by Givens et al. (1968) in the United States is called the cyclic activation log. This is usually used for oxygen and silicon determination for lithology purposes by fast neutron activation and its application has also been considered for manganese (Muravev et al., 1974). Other applications are also possible, as for example for fluorine by the (n, α) reaction. Cyclic activation logging problems, being at first considered separately from activation logs obtained with steady state sources, can be now, due to the work of Barenbaum and Yakubson (1974), be considered as two versions of the same problem.

Activation logging

The radioactivity of rocks and ores induced by irradiation with fast and thermal neutrons can be observed from a measurement of gamma-ray emission characteristic according to a given decay scheme. When irradiation is performed step by step in the borehole followed by the detection of induced radiation after a given delay time, such stationary logging has similar advantages and disadvantages to that provided by the usual activation method used in the laboratory for the analysis of the rock and/or ore samples. The difference however is that the



Figure 11.11. Relation between the calibration factor K of the Cu-64 activation log and selective gamma-gamma intensity using a Cs-137 source (after Bakhterev et al., 1975).

calibration curve follows Equation 13, where σ^{Γ} is the activation cross-section. Such kinds of logs are used for the long-living isotopes: 64 Cu, 56 Mn, 24 Na, etc., and because of its incremental nature it is rather time consuming. In complex ores, the calibration factors derived from Equation 13 can be variable due to the variable elemental composition of the rock matrix (variable (σ/A)_P and μ). An example of such a measurement for 64 Cu is given in Figure 11.10 (Bakhterev et al., 1975a), where the copper grade obtained from an activation log and from a chemical assay are compared. Here the calibration factor varied by a factor of 10 and its values were derived from the selective log mean-gamma log. The correlation of the calibration factor (in cpm/1% Cu) with the selective log readings is given in Figure 11.11.

Some theoretical problems concerning the interpretation of stationary activation logs of layered media were investigated by Vozzhenikov and Zaramenskikh (1975). Also Vozzhenikov and Davydov (1977) have considered, both theoretically and experimentally, the influence of the water-filled borehole diameter, rock porosity and its bulk density on the activation signal from ⁶⁴Cu and ²⁸Al isotopes. Alsayed and Dumensil (1977) considered the new approach to the semitheoretical calibration of the logging tool for the stationary activation method.

A much more economic version is the continuous activation logger. Here a steady state source is moved down-hole with a constant velocity followed by the detector at a distance large enough not to be influenced by the radiative capture gamma rays. When the neutron source is pulse operated, it is sufficient to perform the measurement in the time window situated at the activation time zone (Fig. 11.9), and a large source-detector distance is not necessary. When the width of this time window is Δt and the time separation between the consecutive neutron bursts is T (T> Δt), the continuous log signal $N_{cont}(d,\nu)$ for a given source-detector spacing d and logging velocity ν is related to the cyclic activation log signal $N_{cycl}(d,\nu)$ by

$$N_{cycl}(d,v) = (1 - \Delta t/T) \cdot N_{cont}(d,v)$$
(17)

which was first derived by Barenbaum and Yakubson (1974).



Figure 11.12. Dependence of the activation logging signal N(d,v) on logging speed and source-detector distance in uniform rock medium. (Czubek and Loskiewicz, 1976).

If an infinite medium with a constant grade of activating element is considered, the detector response function, i.e. $N_{cont}(d,v)$, has the behaviour shown in Figure 11.12 (Czubek and Loskiewicz, 1976). The source-detector spacing d in this figure is given in units of the neutron migration length L. It is easy to show that for d = d₂ = const and d >> L, the maximum signal amplitude occurs at the logging speed $v = v_2$:

 $\nu_2 \approx \lambda \cdot d_2$ (18)

where λ is the radioactive decay constant. Equation 18 is valid for all borehole conditions and is marked in Figure 11.12 by the heavy line. When the gamma-ray background is taken into account, Barenbaum and Yakubson (1976) have found:

$$v_2 \approx 0.6.\lambda.d_2 \tag{18a}$$

For a given logging speed $v = v_1 = \text{const}$, the maximum activation signal is obtained for some source-detector spacing d = d₁ which is shorter than the distance d₂ and usually

 $d_1 \approx L$ (19)

Such a short source-detector distance is not feasible for steady state neutron sources (due to the radiative capture background around the source), but is quite possible using pulse operated neutron sources with cyclic detection (Fig. 11.9). Unfortunately this optimum condition is very sensitive to borehole conditions and to the rock neutron properties i.e. mostly on its porosity or hydrogen content and bulk density.

Activation logging is used for the detection of many principal or accessory elements in ores. The difficulty is that the detected radioactive isotope can be obtained in different ways from the different primary nuclei, especially when the neutron flux is mixed i.e. is fast and thermal. For example, the following nuclear reactions are possible with the Fluorine-19 isotope:

			Nuclear reaction	1:	
		¹⁹ F(n,γ) ²⁰ F	¹⁹ F(n,2n) ¹⁸ F	¹⁹ F(n,p) ¹⁹ O	¹⁹ F(n,a) ¹⁶ N
Τ _{1/2}		11.36 s	109.7 min	29.1 s	7.3 s
E _y MeV		1.631	0.51	0.197;1.36	6.134; 7.112
%	/decay	100	194	97; 59	69; 5
i	thermal	0.009	-	-	-
σ	resonance	0.24	-	-	-
(b)	fission	-	7.2×10 ⁻⁶	0.0005	0.0045
	14 MeV	0.01	0.043	0.02	0.05

On the other hand, Nitrogen-16 which is usually used for the detection of fluorine can also be produced by other reactions:

		¹⁵ N(n,γ) ¹⁶ N	¹⁶ O(n,p) ¹⁶ N
	thermal	2.4×10 ⁻⁵	·
σ	fission	-	1.95×10 ⁻⁵
(b)	14 MeV	-	0.042

The contribution of different elements in the build-up of $^{1\,6}N\,$ has to be considered using the knowledge of the physical and chemical parameters of a given deposit.

The influence of borehole diameter on the activation log readings for the ^{16}N isotope was investigated by Potopakhin et al. (1975) for borehole diameters in the range 90 to 220 mm and for tool diameters 89 and 51 mm for both stationary and continuous logs. Some other methodological aspects of the activation logs obtained using steady state and pulsed neutron sources have been studied by Muravev et al. (1974a, 1974b).

Neutron activation surveys of phosphate ores were used by Matyukhin et al. (1976) and Koshelev et al. (1975d, 1976) by detecting ${}^{16}N$ and ${}^{28}Al$. The same isotopes were used by Startsev et al. (1975a, 1975b) and Koshelev et al. (1975a) for the determination of the ore grade of apatite deposits. Fluorspars have also been investigated by the same method by Voynova et al. (1974, 1976) in Uzbekistan who obtained a linear relation between fluorine content and the borehole activation effect. Gorbachev and Petrova (1975a, 1975b) have studied some influencing effects (density, porosity) on the activation results obtained for fluorine. For fluorspars, activation logging with Po-Be and ${}^{252}Cf$ sources (for ${}^{16}N$ and ${}^{28}Al$) have been carried out by Koshelev et al. (1975a, 1975b), and some methodological properties of the activation log have been studied for these ores. Copper activation by fast neutrons using the borehole neutron generator was also reported by Wylie et al. (1976).

The problems connected with bauxite ore grade determination by activation methods were studied by Blumentsev et al. (1974a, 1974b) and by Shishakin et al. (1974). Aluminum in potassium ores has been determined with this method by Startsev et al. (1975c).

To localize gold-bearing veins, Kuchurin et al.(1976a) have used the stationary activation log of 24 Na (80 to 105 minutes per point) to estimate the specific elemental composition of goldquartz-tourmaline-sulphide deposits.

The presence of sodium in the majority of rocks permitted Bakhterev and Kharus (1975a) to use the activation borehole measurement of rock bulk density. They found that the spectral ratio for the two energy lines of 24 Na (1.38 and 2.76 MeV) isotope activated by a Po-Be source with an output of about 10⁷ n/s is well correlated with rock bulk density. An example of their results is given in Figure 11.13. The accuracy of density measurements by this method was about 0.05 g/cc.

GEOSTATISTICS OF NUCLEAR LOGGING METHODS

Each nuclear logging method has its own range of penetration which can also be considered in terms of the rock volume, say v_2 , sampled by the method. The same can be considered with regard to the chemical assay; the volume, say v_1 , of the particular sample is always defined for each type of chemical assay. Let both assays, nuclear and chemical, give an estimate of some geological parameter P e.g. ore grade, density, etc. For a given volume v of the sample from a given volume of the orebody V (V>v), the parameter P has its statistical distribution f (P,v). When the orebody is not homogeneous, the distribution function f (P,v) will be different for different



Figure 11.13. Relation between the spectral ratio g of Na-24 activation log and rock bulk density (after Bakhterev and Kharus, 1975).

values of v. For example, when v = V, f (P,V) is the Dirac delta function at P = P, where P is the average value of the ore grade within V. This situation is schematically presented in Figure 11.14. Depending upon the formation heterogeneity, the chemical assay distribution f (P,v₁) will be different from the geophysical assay distribution f (P,v₂), unless $v_1 = v_2$. Usually, when $v_1 \neq v_2$ and the formation heterogeneity is not negligible, a comparison between the logging data and the core assay is rather problematical.

The problems presented above can be solved by means of the mathematical apparatus of geostatistics. Czubek (1976) has treated them using the geostatistical approach of the French School of G. Matheron. The practical application of the reasoning presented in his paper has been reported by Czubek et al. (1977) in order to obtain the proper calibration curves for nuclear logging tools.

FUTURE DEVELOPMENT OF NUCLEAR LOGGING METHODS

Any forecast for the future development of nuclear well logging methods should be done for each element and for each method separately. This, however, could be valid for a given country only, or even for a given type of deposit. With regard to future progress in general, one can predict to some extent the further development of methods with the neutron and photon generators, or both in one tool, as proposed by Bessarabskiy et al. (1975), or even with generators of high energy protons. On the other hand the need for rather lightweight and small diameter equipment in the mineral industry does not encourage such development. For this reason more careful attention should be given to possibilities other than the application of pulsed particle generators for the time analysis.

The time analysis of induced radiation can be done either with the steady-state neutron or gamma-ray source. This solution although it cannot introduce any new nuclear reaction into logging practice can meet, the requirement for a small tool diameter and makes the equipment more portable. The idea is to treat nuclear logging methods as a stationary time stochastic process at a given borehole depth. By registering the time moments X(t) of the neutron emissions from the source and the time moments Y(t) at which the resulting photons reach the detector, the cross-covariance or even the auto-covariance functions of these two stochastic processes can



OBSERVED VALUES OF THE ORE GRADE P

Figure 11.14. Influence of sample size v on the ore grade P distribution f(P,v) within an orebody of volume V (after Czubek, 1975).

be measured. Because neutron emission from the source has a Poisson distribution, detector events are described by the shot noise process with the transmission function h(t) of the system. This function is just the pulsed neutron generator response function (Fig. 11.8). A sketch illustrating this reasoning is presented in Figure 11.15. This kind of measurement seems to be very promising. Some experimental results have already been obtained by Blankov and Kormiltsev (1972, 1974). An example of their results is given in Figure 11.16 which proves the high similarity of the pulsed and cross-correlation experiments.

The stochastic approach can also be applied to the measurement of the amplitude of the decay curve at t=0. This amplitude is directly related to the grade of mineral present. Uranium detection by the fission neutrons should also be possible using this method.

Another possibility, not yet well explored, is the time analysis of radiative capture photons, or photons from inelastically scattered fast neutrons. As an example the gamma-ray lines emitted in cascades i.e. coincident in time because of the very short lifetime of the intermediate energy level, are presented in Figure 11.17 for some iron deposits (Czubek, 1975). By the measurement of time coincidences between the photons of the two energies E_1 and E_2 , the presence of a given element can be detected without any influence of the other rock components, which have their own coincident photon pairs of energies E_1 and E_2 .

NUCLEAR ASSAYING OF THE IRON - BAUXITE GROUP

In this group iron, manganese and bauxite have been assayed by nuclear methods.

Iron ores

Several laboratories have used different nuclear methods for the assay of iron deposits. Szymborski (1975) and Christell et al. (1976) have used the usual density logging technique, whereas Senko and Zorin (1975) have utilized the selective gamma-gamma method on skarn iron deposits. Ochkur et al. (1976) have applied XRF and neutron-gamma spectrometry for the same purpose. Butyugin (1976) has proposed using the Mössbauer effect with a ⁵⁷Co source for the assay of iron. The most extensive work has been carried out by the Australian group on hematite



deposits (Aylmer et al., 1976, 1978; Charbucinski et al., 1977; Eisler et al., 1976, 1977). The correlation with natural radioactivity, as well as the spectrometry of radiative capture of thermal neutrons in the combined neutron method have been used successfully by this group. The scattered gamma-ray techniques, however, have been the most accurate. They have used the combination of density, P_z and S factor techniques fitting the iron content to the multivariable

combination of density, P_z and 5 factor techniques fitting the iron content to the multivariable formula as shown in Figure 11.18. The energy limits for each component of the formula are also given in this figure. The accuracy of iron determination in dry percussion holes of rather large and variable diameter was better than 1 per cent of iron. For water-filled boreholes, neutron-capture gamma-ray spectrometry is a preferable technique for iron assay.

Nuclear assaying of manganese ares

There have not been many papers published during the period 1974-1977 on nuclear logging methods for manganese. Apart from the laboratory research work of Muravev et al. (1974) on the activation method only Ochkur et al. (1976) reported the application of activation and neutron-neutron (thermal) logs on manganese deposits. The reason for this is, perhaps, the recent worldwide interest in marine manganese nodules instead of new minable deposits.

Nuclear assaying of bauxite ores

The main element detected by nuclear methods is aluminum by activation with thermal neutrons. Blumentsev et al. (1974a, 1974b) and Shishakin et al. (1974) have used a ²⁸Al activation logger having a Po-Be source (of activity of about 10^7 n/s) with spectrometric recording above 1.1 MeV. The source-detector spacing was 1.5 m with a 30 to 40 m/h logging speed. To determine the other ore parameters, a selective gamma-gamma logger with a ¹⁷⁰Tm source (7 cm source-detector spacing) and a spectrometric measurement capability over the range 20 to 110 keV was used together with a density logger (¹³⁷Cs source, 32 cm source-detector spacing, recording above 150 keV).

NUCLEAR ASSAYING OF THE BASE METAL GROUP

Deposits of copper, zinc, lead, mercury and barium are discussed in this Section.

Copper ores

Copper ones are usually investigated by the activation logging techniques described earlier and by the XRF method. Activation techniques have been used by Bakhterev et al. (1975a), Wylic et al. (1976) and Christell et al. (1976). The XRF logger and rock face assaying is carried



Figure 11.16. Cross-correlation and die-away curves in paraffin wax, (after Blankov and Kormiltsev, 1972).

out by measuring the spectral ratio of the ${\rm K}_{\alpha}$ copper line to the scattered photons of energy 14.5 keV (from a $^{10.9}{\rm Cd}$ source), even in multi-element ores with Pb (Ochkur et al., 1974; Grigoryan et al., 1974; Tamrazyan and Popov, 1975; Bolotova and Leman, 1976). Kozlov et al. (1975a, 1975c) have used the selective gamma-gamma method to localize and to determine the quality of the sulphide-copper ores. For the same purpose Grigoryan (1975) has used the epithermal neutron logger.

Zinc, lead and barium ores

Lead and zinc or lead and barium very often occur together in sulphide-type deposits usually in carbonate rocks. Here the XRF logger is the most popular tool. The K_{α} line of zinc and the L line of lead are used for the simultaneous determination of both elements (Kozlov et al., 1975b; Krasnoperov et al., 1976; Krasnoperov and Zvuykovskiy, 1976; Zgardovskiy et al., 1974) employing ^{119M}Sn, ¹⁰⁹Cd or H³/Zr sources. Proportional counters are usually used and the spectral ratio is recorded. For water-filled boreholes, the close collimation of the source is used and Cu-Ni filters are utilized to distinguish zinc from iron in the XRF log (Shmonin et al., 1976b). XRF logging speeds vary from 120 up to 350 m/h. When lead only is to be detected, a ⁷⁵Se source is utilized (Kobelev et al., 1974). When a high accuracy for barium and lead determination is required, the measurement often takes a longer time – up to 4 minutes (Landström, 1976), for the "natural XRF log". In order to localize lead seams with high precision,



Figure 11.17. Gamma-ray lines E_1 and E_2 emitted in cascade due to radiative capture of neutron. Example for an iron ore deposit. (Czubek, 1975).

one can use a density logger with very good results (Yasinovenko, 1975). For Ba-Pb determinations, short gamma-gamma probes (6 to 8 cm) are also used for spectrometric measurements over the range 130 to 150 keV (Shmonin et al., 1976a).

Mercury ores

Mercury is a very convenient element to detect by nuclear methods. Its high atomic number and neutron absorption cross-section permit the application of XRF and neutron methods for mercury assay. The common mercury mineral is cinnabar and the payable grades are from 0.01 per cent Hq up to 5 or 8 per cent in very rich deposits. Sometimes an increased uranium and thorium concentration is correlatable with mercury which provides an opportunity of using the gamma spectrometric survey technique (Antipov, 1975). XRF loggers were used by Mitov et al. (1975) and Balakshin and Kravchenko (1976) employing the L series lines of Hg with quite good agreement with chemical assays (±20% relative). Neutron-neutron logs (simultaneous thermal and epithermal) have been used by Fatkhutdinov (1974), Fatkhutdinov et al. (1974a, 1974b) to estimate the mercury reserves of Hq-Sb deposits. Radiative capture gamma-ray spectrometry in the region around 4 and 6 MeV using a Po-Be source has been used by Balakshin and Kravchenko (1976) and Boyarkin and Kaipov (1974). The agreement with chemical assays was varied from 0.004 up to 0.24 per cent Hq. One of their results is reproduced in Figure 11.19. The "mercury" line I_1 around 4 MeV with the background I_{1b} subtracted and the Ca-Fe line I_2 around 6 MeV with the background I2h subtracted were recalculated to the pure "mercury" intensity I13. A method of interpretation of such I13 logs in the layered media was also established.

Egorov et al. (1975) have performed some theoretical and model experiments for mercury determination using the spectral ratio 4-5 MeV/6.1-8 MeV to avoid the influence of iron. Afanasev et al. (1974) have used this method in the Kirgiz Republic and obtained a sensitivity of about 0.06 to 0.08 per cent Hg. In an attempt to avoid the influence of iron an improvement of this method was carried out by Sokolov et al. (1975).



Figure 11.18. An example of the Co-60 scattered gamma-ray spectrum in a dry blast hole used by Charbucinski et al. (1977) to assay iron content.

Pulsed borehole neutron generators have been used to measure the $\Sigma_{\rm a}$ in mercury deposits (Putkaradze et al., 1973; Kashkay et al., 1976). The values of $\Sigma_{\rm a}$ for mercury ores of different lithologies obtained by Putkaradze et al. (1973) are presented in Figure 11.20.

Another new approach was proposed by Nikulin et al. (1976) applying a spectrometric technique to the radiative capture gamma rays in the pulsed neutron method. Some results of their laboratory experiments are shown in Figures 11.21 and 11.22. The method is based on the detection of the spectral ratio

$$\eta = \frac{I(3 - 4 \text{ MeV})}{I(> 6 \text{ MeV})}$$

which permits the utilization of an optimal delay time t_d and width of the detection window $\ \Delta t$ the measurement of mercury grade in dry boreholes with a negligible influence from iron content.

NUCLEAR ASSAYING OF THE TIN - RARE METAL GROUP

In this group deposits of Sn, W, Mo, Nb, Hf, Ta, Be, Li, Rb-Cs, Cs, Bi, mica, feldspar, Ti, rare earths, Zr are included.



Figure 11.19. Spectral neutron-gamma log from a mercury deposit (after Boyarkin and Kaipov, 1974).



Figure 11.20. Calculated thermal neutron lifetime T in mercury ores (after Putkaradze et al., 1973).



Figure 11.21. Spectral ratios for different delay times t_d in the pulsed neutron-gamma method obtained using mercury ore models (after Nikulin et al., 1976).

Tin deposits

The most commonly-used logging methods are the XRF and Mössbauer effect techniques which give intercomparable results and have the same accuracy as chemical assays. The advantage of the XRF method is the possibility for the simultaneous determination of other elements which occur with tin. Sometimes the activation method is used (Gorbachev et al., 1974; 1975) when the grade of fluorine is correlatable with tin.

XRF logging or in situ ore face assaying for tin have been reported by Grigorkin and Neustroev (1974), Sachuk and Balashov (1974), Afanasev et al. (1974), Bolotova and Leman (1976), Meyer et al. (1976), Ochkur (1976), Balakshin and Kravchenko (1976), Christell and Ljunggren (1976), Ratnikov et al. (1976) Nuclear Geophys. Assay...(1976), and many others. The usual method was to employ the spectral ratio 25 keV/35 keV to determine tin grade which resulted in detection limits of the order of 0.005 to 0.15 per cent Sn depending upon the characteristics of the deposit. 147 Pm and 241 Am sources were utilized for this purpose. Some boreholes were water filled. In association with tin, the other elements detected by XRF logging were V, Cu, As, W, Pb, Zn, and Fe.

Cassiterite ores were assayed by the Mössbauer effect either in situ on the rock face or by step by step logging. This work was reported by Goldanskiy et al. (1974), Ochkur (1976), Nuclear Geophys. Assay . . . (1976), and others.

Tungsten ores

Tungsten was usually detected by the XRF method (already described) simultaneously with other heavy elements. Kuchurin et al. (1976) reported the application of a selective gamma-gamma logger to detect tungsten grades above 0.2 per cent W. For XRF logging, they have used the spectral ratio 55-65 keV/80-90 keV employing a 75 Se source. Manganese was also correlatable with tungsten in this deposit, thus by the activation method grades above 0.4 per cent W have been also determined Ochkur et al. (1974) gave some details of the logging apparatus.



Figure 11.22. Spectral ratios obtained for the different mercury content of ores by the pulsed neutron-gamma method (after Nikulin et al., 1976).

Beryllium and lithium ores

Beryllium ores are always detected by the photo-neutron method, both in surface surveys and in borehole logging. Suvorov and Molochnova (1975) discussed the influence of soil moisture and density, its absorption cross-section and the natural neutron background on the results of carborne surveys. The other measurement parameters for this survey technique were discussed by Brem et al. (1975), whereas Gorev et al. (1975b) and Gorev (1975) have presented the details of the apparatus design. The self-absorption of neutrons in this method was discussed by Kirichenko (1975). For logging methods, the sensitivity of the beryllium assay once of the order of 0.01 per cent BeO (Afanasev et al., 1974) has been improved to the 0.001 per cent level (Grigoreva et al., 1975) and is even more for a more sophisticated logging tool with a 5 mCi ¹²⁴Sb source (Shestakov, 1975). Some other logging applications are presented by Ochkur (1976) and Krapivskiy et al. (1976).

Krapivskiy (1976) has investigated the feasibility of lithium determination in pegmatite rocks by the combined gamma-neutron and neutron-neutron methods.

Molybdenum ores

Molybdenum has been determined by the XRF method usually together with other elements in boreholes and in surface surveys by Grigoryan et al. (1974) and Bolotova and Leman (1976a) with an accuracy of 0.01 per cent Mo.

NUCLEAR ASSAYING OF THE ULTRABASIC GROUP - CHROMIUM AND NICKEL ORES

Chromium ores have been detected by a combination of spectrometric neutron-capture gamma-ray logging to localize the ore zones with neutron-neutron logging to verify the homogeneity of ores. The spectrometry of the scattered gamma radiation has been used to determine Cr_{2O_3} grade. Density logging has also been used in this combined measurement (Feldman et al., 1974; Ochkur et al., 1976). Voznesenskiy (1976) has used the XRF method for chromite exploration.

For nickel ores, Bakhterev and Senko-Bulatnyy (1975) have used spectrometric neutroncapture logging in which the calibration factor was variable because of the varying iron content. This iron content was also determined by the other energy line in the same neutron-capture gamma-ray spectrum.

NUCLEAR ASSAYING OF THE GOLD GROUP

In this discussion gold, antimony and uranium ore deposits are included.

Gold and antimony ores

Gold usually occurs with antimony. Other types of deposits also contain antimony, mercury or tungsten.

Gera (1974) has used selective gamma-gamma logging with a ¹⁷⁰Tm source to localize quartz veins. The gold itself has been detected by spectrometric combined neutron-neutron, capture neutron-gamma, activation, selective gamma-gamma and spectrometric gamma logging (Kuchurin et al., 1975a).

Antimony ores have usually been detected by XRF logging with a 170 Tm source using the spectral ratio 25 keV/85 keV (Grigorkin et al., 1976; Petrukhin et al., 1976; Ivanyukovich et al., 1976; Ochkur et al., 1974; Afanasev et al., 1974). The agreement of the XRF logging results with chemical assays were always within ±10 to 20 per cent relative. Fatkhutdinov (1974a) and Fatkhutdinov et al. (1974) have used thermal and epithermal neutron logs to determine antimony in Sb-Hg deposits.

Uranium ores

Uranium ores are usually investigated by gamma-ray logging previously discussed. When gamma-ray spectrometry is used, the windows 1.05 to 1.65 MeV and 2.05 to 2.65 MeV are utilized to distinguish the uranium and thorium series. Some experimental aspects of this method have been discussed by Sinitsyn et al. (1974), Gabitov et al. (1974) and Kozynda et al. (1974, 1976). Khaykovich and Yakovlev (1976) have contributed to the problem of the accuracy of the computer interpretation of gamma-ray logs in layered media. Novikov and Ozerkov (1974) have investigated the influence of radon emanation into boreholes on the results of the gamma-ray logs. Some other measurement problems have been discussed by Novikov et al. (1974, 1977).

The frequent radioactive disequilibrium of uranium ores and/or the increasing need for additional remote sensing techniques for uranium exploration has stimulated the development of techniques other than the gamma-ray methods of uranium determination. Czubek's first paper (1972) on the pulsed neutron method gave the theoretical and experimental principles for the prompt and delayed fission neutron detection in uranium ores using a pulsed neutron generator. Next, this idea has been taken up in the United States (see Exploration for Uranium Deposits, 1976; Thibideau, 1977; Renken, 1977; Givens et al., 1976). European laboratories have published some theoretical papers on natural fission (from the spontaneous fission/neutron distribution) for different borehole-layer configurations (Davydov, 1975b) and for a point neutron source in the borehole, but without any discussion of time problems (Davydov, 1975c, 1975d). The theoretical paper of Czubek and Loskiewicz (1976) can be useful in continuous delayed fission neutron logging using the "jerk" source method (Californium Progress 1976, No. 20). It was also found there that the optimum condition for continuous delayed fission neutron 18) is v = 0.241 cm/s. Some other theoretical considerations for pulsed delayed fission neutron logging using the Monte-Carlo technique have been presented by Wormald and Clayton (1976).

Another possibility has been proposed by Kartashov and Davydov (1975) for detecting uranium in rock. A photo-neutron source with an output of 10^7 n/s can be used to irradiate uranium-bearing rock in order to generate fission neutrons which can be detected at the fast stage by threshold detectors. The stochastic process approach by the cross-covariance or auto-covariance measurement mentioned in this paper should also be taken into account for uranium measurement by the detection of fission neutrons.

NUCLEAR ASSAYING OF SEDIMENTS, EVAPORITES AND OTHER TYPES OF DEPOSITS

Sulphur deposits

Feldman et al. (1974) have used a combination of neutron and density logging together with the spectrometry of thermal neutron capture gamma rays to determine sulphur content. Neutron gamma spectrometry has been carefully investigated for carbonate-type sulphur deposits by Blinova et al. (1974). The same method has been applied by Niewodniczanski and Palka (1976) and by Chrusciel et al. (1977) on the Polish sulphur deposits. The gamma-ray lines for sulphur (around 4.4 MeV), calcium (around 5.4 MeV) and hydrogen (around 2.2 MeV) using a Po-Be source have been measured. One of their calibration curves is given in Figure 11.23.



Figure 11.23. Calibration curve for a 50 cm long probe used to determine sulphur content by the neutron gamma spectrometry (after Chrusciel et al., 1977).

Potassium ores

Spectrometric gamma-ray logging was used by Mishin (1976) and Mishin and Gavrilova (1976) to determine the potassium grade and the nonsoluble parts of ore. Other nuclear methods used in the potassium industry are presented in the short monograph by Saturin (1975).

Boron deposits

The boron series has been investigated by Vakhtin et al. (1972, 1973, 1975) by means of the resonance neutron-neutron method using a special resonance detector and some results are shown in Figure 11.8.

Phosphorite, apatite, fluorite and alunite deposits

These deposits are usually investigated by activation logging (Koshelev, 1975; Koshelev et al., 1975a, 1975b, 1975c, 1975d, 1975e, 1976; Startsev et al., 1975a, 1975b; Voynova et al., 1976; Matyukhin et al., 1976). Sometimes the correlation between the P_2O_5 content and ore density gives additional information about the quality of the apatite ores, or a correlation with uranium (Rudyk et al., 1974) is also observed. For alunites Muravev and Yakubson (1975) have observed an increased gamma-ray activity.

FINAL REMARKS

It was not possible to present in this review all applications of the nuclear logging methods to the exploration of minerals during the period 1974 to 1977. Only the most important have been presented here; most theoretical papers have been omitted, especially those concerned with neutron distribution in rock media.

The general trend observed in this branch of applied science is the growing importance of nuclear methods among the range of techniques used in exploration and mining. The most important development, however, is that the accuracy of assays performed using nuclear methods is now comparable with the so-called classical chemical methods being at the same time much cheaper and less time consuming.

On the other hand the application of nuclear methods requires a good understanding of the physical phenomena involved and of the sophisticated field equipment used to perform the detailed energy and time analysis of the recorded radiations. Without this deep knowledge of the physics of these methods, any practical application is very often unsuccessful and a waste of money.

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