Neutron and Gamma Ray Scattering Measurements for Subsurface Geochemistry

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Developed for the oil industry, well logging instrumentation based on electrical, acoustic, and nuclear measurements has been providing information about the localization and evaluation of hydrocarbon-bearing strata for petroleum geologists and engineers since 1927. This method of exploring properties of the earth's crust without taking physical samples is attracting a growing audience of geologists and geophysicists because of recent developments that permit nondestructive measurements of subsurface geochemistry. A combination of nuclear measurement techniques, which use gamma ray and neutron sources, can provide detailed information on rock composition of interest to both industry and academia.

THE INDUSTRY THAT HAS DEVELOPED TO FEED OUR APPEtite for hydrocarbons uses a curious mixture of high and low technology in a complicated chain of events that begins with the selection of a site for drilling a well and ends with the production of oil or gas from the reservoir. A reservoir is a layer of subsurface porous rocks that covers a large areal extent from which hydrocarbons can be extracted. The academic definition of a reservoir is made more precise by economic factors that dictate minimum values of thickness, areal extent, porosity, hydrocarbon content, and extraction possibility for the reservoir to be profitable. Well logging is concerned with the assessment of rock formation properties that allow determination of reservoir quality.

The selection of the drilling site may be based on regional geology, the proximity to other producing fields, surface seismic measurements, or a combination of all three. In the most favorable case, the well site is chosen above a geological structure that may be capable of accumulating hydrocarbons. The drilling of the well then proceeds to the approximate depth of the target rock formations. The next and most important phase is the evaluation of the rock formations traversed by the well to predict their potential for becoming a successful reservoir. The results of these analyses will be used to decide whether the well will be abandoned or additional expenditures will be made to extract the hydrocarbon.

Well logging (1-4) was developed as an alternative to the expensive and time-consuming operation of coring. The basic components of coring are extraction of cylindrical sections of rock and subsequent analyses of porosity, permeability, and hydrocarbon content. Well logging consists of lowering by cable combinations of

specially designed instruments or tools into a well that has been drilled through target rock formations. As the instruments are withdrawn, at 200 to 1200 m/hour, continuous measurements are made of the rock strata traversed by the wellbore. Measurements are conventionally sampled every 15 cm, although sampling with finer resolution (3 cm) is becoming common. Some imaging devices (5) provide measurements every 0.2 cm.

In well logging, the instrumentation package is suspended from the surface by an armored cable (Fig. 1) that conveys power to the measurement package; data are relayed to a surface computer in the instrumentation truck by telemetry. Many measurement devices have extendable arms with sensors that must contact the borehole wall to measure rock properties (Fig. 1). Other devices require centering in the borehole. Still other instruments use springs or hydraulically activated arms to force the sensors to drag against one side of the hole as the tool is withdrawn from the well while continuous measurements are made.

The surface logging unit provides the downhole electrical power and has a winch for the lowering and withdrawal of the measurement package. It also has computers for analysis and presentation of the logs—the continuous records of measured formation properties versus depth. Although Fig. 1 shows the operation taking place on land, it is also performed from offshore platforms and from shipboard.

The types of measurements made include electrical conductivity, shear and compressional acoustic velocity, bulk density, H concentration, and elemental geochemical analyses. These measurements are grouped into three families: nuclear, acoustic, and electrical. The relationship of these types of measurements to the evaluation of reservoir rocks can be illustrated by considering just two necessary properties of reservoir rocks-porosity and presence of hydrocarbons-and the qualitative relationship of these properties to measurable physical parameters. A porous rock is less dense than a nonporous rock, and its density can be measured with gamma ray scattering. Additionally, a porous rock, either water-filled or hydrocarbon-filled, will have a significant H content, which can be measured with neutron scattering. Acoustic travel times may also be used to determine porosity because the compressional velocity of rock decreases with increasing porosity. The electrical conductivity of a rock depends on both the porosity of the rock and the salinity of the water filling the pore space. Electrical measurements can be used to infer the presence of hydrocarbons in the porous rock since a hydrocarbon-filled rock is less conductive than one filled with a brine.

This article focuses on nuclear logging instrumentation in order to describe the recent developments that allow remote measurements of subsurface geochemistry. All sets of logging instrumentation have the same general appearance—they are long pieces of pipe

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Fig. 1. The major components of a logging operation displayed from a perspective close to the borehole. The wellbore traverses a number of porous strata to be evaluated for the presence of hydrocarbons.



Fig. 2. Outrasonic profile of a borehole drilled in granite showing selective enlargement. The cross section of a logging tool is shown applied in front of a breakout.

with an outer diameter of about 10 cm. These pipes are pressure housings, necessary to protect the instrumentation from the hostile borehole environment in which they must operate.

The Borehole Environment

The measurements undertaken in the borehole would be difficult even in a controlled laboratory environment. Wells in which logging measurements are routinely made vary in depth from roughly 1000 to 7000 m. Wellbore diameters range from 15 to 40 cm. During drilling and the subsequent logging operation, the borehole is filled with fluid that is graphically known as mud. The mud system is an important part of the drilling process. Pumps on the surface are used to circulate mud down through the drill pipe, out the bottom where a drill bit is attached, and up the annulus between drill pipe and wellbore. The circulating mud cools the drill bit and transports to the surface rock fragments generated as drilling proceeds. The mud system also prevents blowouts by providing a column in which the pressure always exceeds the pressure of the formation fluids. This condition is accomplished by the addition of weighting materials to the mud system that increase its density. Barite (BaSO₄) or iron oxide additives are required for densities as large as 2 g/cm³. Because of the fluid density and well depth, the instrumentation packages must withstand pressures up to 136 MPa. The geothermal gradient results in wellbore temperatures that can exceed 175°C; such temperatures impose stringent conditions on the downhole electrical and mechanical components of logging tools.

Two other borehole features sometimes complicate measurement of rock properties. One complication results from deformation of the borehole wall. Although in Fig. 1 the borehole wall appears as the surface of a smooth cylinder, this is often far from reality. Variations in the elastic properties of the rocks cause some layers to be stronger than others. Clay minerals in some layers are altered by contact with the drilling mud and can swell and be washed away from the borehole wall. These strength variations among layers can result in differential erosion either from contact with the rotating drill pipe or from the flow of abrasive mud. The result is a corrugated borehole profile in which diameter changes of up to several centimeters occur over similar length scales. Irregularity of this type is known as rugosity. Another type of borehole wall deformation, known as "breakout" (6), results from stress relief after drilling where large tectonic stress anisotropies are present. In a breakout, material fractures and falls from the borehole wall, creating a gap as deep as several centimeters. Shown in Fig. 2 is a cross section of a borehole drilled in granite. The instrument (7) used to measure the cross section records the travel time of an ultrasonic pulse emitted from a tool centered in the hole, thus providing an estimate of the borehole radius. The nominally 16.5-cm-diameter hole is seen to have two prominent extensions where breakouts have occurred. A logging device positioned over these gaps, which can persist vertically for tens of feet, will have a difficult time measuring rock properties. This is especially true for most nuclear logging tools since many of them have shallow depths of investigation, extending no farther than 15 to 25 cm beyond the borehole wall.

A second complication for the measurement of rock properties occurs in porous and permeable formations where a portion of the mud system fluid may filter into the rock formation. Filtration leaves a residue, known as "mudcake," on the borehole wall which eventually forms a pressure seal. The mud displaces the original formation fluids and creates a layer of material of unknown thickness and density, through which measurements must be made.

The scenario for logging measurements, then, is a deep, hot borehole with rather large hydrostatic pressures and a possibly rough and irregular wall that might have large vertical cracks and might be covered with mudcake, or both. All three families of logging instruments have been engineered to operate in this environment so that the borehole can be used as a window on the earth's interior. Borehole logging may not offer the panoramic view obtained from surface seismic methods, but it does allow direct measurement on otherwise inaccessible rock formations. Continuous measurement of the in situ concentration of important rock elements is provided through the use of nuclear logging methods.

Gamma Ray Scattering for Density and Photoelectric Absorption

Gamma rays are frequently used in nondestructive testing of materials. Generally, a material is irradiated with a source of gamma rays and the transmission of the gamma rays provides information on the material's density. The technique has been used, for example, to check the quality of welds or for the presence of broken bones. In the borehole, however, such a straightforward transmission measurement is not possible. Both the radioactive source of gamma rays and the detector must be contained within the logging tool, making it impossible to place the rock sample between them for an attenuation measurement.

The system (8) that has evolved for well logging, shown in Fig. 3, consists of a source of gamma rays (usually ¹³⁷Cs emitting roughly 5×10^{10} gamma rays per second at 662 keV) and two detectors at different spacings (on the order of tens of centimeters). The gamma ray detectors are typically scintillation crystals coupled to photomultipliers. Stabilization systems incorporating small radioactive reference sources are used to control the overall gain of the detectors, which could otherwise change dramatically with the large range of downhole temperatures. High-density material in the measurement package shields detectors from the direct-source gamma rays. Because of the rather short mean free path of the gamma rays (10 to 15 cm), the instrument is pressed against the borehole wall with a hydraulically activated arm (not illustrated).

The gamma rays from the source that reach the rock around the borehole are scattered, and only a small fraction $(<1/10^6)$ of them reach the detectors. Although the scattered gamma ray spectrum



Fig. 3. A dual-detector density logging tool applied to a formation with intervening mudcake. [From (8)], Ellis *et al.*, with permission of SPE.

Fig. 4. Density logs from two passes in a well drilled in crystalline rock. On the first pass, the density tool was oriented in front of a persistent breakout as illustrated in Fig. 2. The error compensation $(\Delta \rho)$ is seen in the middle graph.



measured by the detector is rather featureless, with the exception of a broad peak near 100 keV, spectroscopic analysis is possible. The high-energy gamma rays, those least affected by photoelectric absorption, are used to obtain a counting rate that is found experimentally (8) to be exponentially related to the bulk density of the scattering rock material, as would be expected in a transmission measurement.

Both detectors can be calibrated to yield a bulk density as a function of their counting rates and to produce identical values if the instrument is in direct contact with the rock formation. However, if the measurement device is not in contact with the borehole surface, because of rugosity or thick mudcake, for example, the apparent densities derived from the two detectors will be different. This difference is used to compensate the apparent density measurement of the long spacing detector, which is more sensitive to formation density and less affected by the mudcake than is the short spacing detector, thus providing a true formation density for most situations encountered.

The precision of the density measurement is ~0.01 g/cm³. This precision can be achieved even in the presence of mudcakes of thickness up to 2 cm. For the petrophysicist, an immediate concern is not the density of the rock formation but its porosity. The relation between the rock bulk density, ρ_b , measured by the logging tool and the volume fraction porosity, ϕ , is

$$\mathbf{p}_{b} = (1 - \mathbf{\phi})\mathbf{\rho}_{ma} + \mathbf{\phi}\mathbf{\rho}_{f}$$

where ρ_{ma} is the density of the rock grain (matrix) and ρ_{f} is the density of the fluid filling the pore space. Fluid density is generally 1.0 to 1.2 g/cm³ because the pore fluid is commonly a brine. However, gas densities may be as low as 0.4 g/cm³. The grain density of common sedimentary rock ranges from 2.65 to 2.96 g/cm^3 . In order to solve for porosity with a precision on the order of 1% by volume, one must know the bulk density to a precision of 0.1 g/cm^3 , and also the rock type, in order to use the appropriate value of ρ_{ma} . Although there are a number of methods for estimating rock type, including core analysis and analysis of the rock cuttings brought to the surface by the drilling mud, a by-product of the gamma ray scattering measurement provides a convenient method. This method relies on the fortuitous relation between the bulk densities of common sedimentary minerals and their average atomic numbers. For these minerals, a measurably distinct value of average atomic number is associated with each value of grain density. Thus, in simple rocks, measurement of the average atomic number can be used to infer grain density.

The measurement of the average atomic number is made from an analysis of the low-energy portion of the scattered gamma ray energy spectrum (9). Because the photoelectric cross section varies

roughly as the fourth power of the atomic number, Z, at energies below 100 keV it is possible to deduce the average atomic number, or more conventionally, a parameter called the photoelectric factor, P_e . This parameter, which is proportional to the density-normalized photoelectric cross section at 40 keV, varies numerically between 1.8 and about 5 for three common sedimentary minerals: sandstone (SiO₂), dolomite [CaMg(CO₃)₂], and limestone (CaCO₃). The P_e for clay minerals can also be calculated. Since they are silicates one expects their P_e values to be close to that of sand (1.8), but the presence of large Z elements such as Fe or K results in much larger values. Frequently, the values seen on logs in shale zones containing large concentrations of clay minerals are around that of dolomite (3.1).

A 30-m section of log from a density logging device is shown in Fig. 4. Two sets of measurements are included from the same well, which was logged twice. The results from the first set of measurements are indicated for all of the logs. On the left, in the graph labeled "caliper," is a measurement of the borehole diameter acquired by the density logging tool, which uses a spring-loaded arm that pushes it against the borehole wall. Although the borehole had a nominal diameter of 28 cm, it is seen to be enlarged above 2060 m (at A) and exceeds 50 cm in places. In this well, which was drilled in crystalline rock, breakouts of the kind illustrated in Fig. 2 have occurred. For the second set of measurements, the instrument assembly was mechanically rotated about 90° to a smooth side of the borehole. On the second logging operation, the caliper measurement indicates a rather smooth borehole of the anticipated diameter. Because of the limited penetration of gamma rays into the formation rock surrounding the borehole, the measurement of density was impossible on the first run, despite the dual-detector error compensation system. After the detectors were reoriented to the smooth side of the borehole, the error compensation was generally zero, indicating that the density estimates from the two detectors are the same and representative of the bulk density of the rock (2.6 g/cm^3) .

In the graph on the right, the density values obtained by

measurement on the smooth side of the hole are seen to be relatively constant at about 2.6 g/cm³ and P_e , in the middle graph, is nearly constant at 2.5. The smaller values of the $P_e \log$ from the first set of measurements are due to the presence of drilling mud between the detector and the formation. The drilling mud is primarily water with a few clay additives with a P_e value near that of water (~0.4). The effect of a such a layer in front of a rock with a much higher value of $P_{\rm e}$ is to depress it. This is because the depth of investigation of this portion of the measurement is related to the mean free path of lowenergy gamma rays (40 to 80 keV), which is only a few centimeters. Thus, a layer of only a few centimeters looks like an infinite sample. Increasing the thickness of the mud layer will decrease the estimate of the formation P_{e} . The difference between the values on the two runs can be used to estimate (10) the gap between the tool and the rock face, which often exceeded several centimeters in this wellsignificantly beyond the compensation capabilities of the density instrument.

Neutron Scattering for Hydrogen Content

Neutron scattering was an early application of nuclear techniques for the determination of subsurface formation porosity. First patented in the early 1940s, the idea behind it is straightforward: neutron moderation by H is quite efficient because H atoms have roughly the same mass as neutrons. Thus, porous rock filled with either water or hydrocarbons moderates neutrons more or less strongly depending on the H content. This seemingly direct method of porosity determination is somewhat more complicated than the measurement of density with gamma ray scattering, because of the nature of neutron interactions with matter (11). For neutron porosity estimation, the most important phenomenon is elastic scattering of neutrons by the rock formation nuclei. In this process, the kinetic energy of each neutron is reduced at each scattering; the amount of reduction depends strongly on the mass of the target nucleus. As the target mass approaches the neutron mass, collisions reduce the neutron energy more efficiently. For this reason, H is the most efficient moderator of neutrons in geologic formations. A measurement of the spatial distribution of multiply scattered neutrons is sufficient to characterize the moderating efficiency of a formation, which is related to the H content.

Neutron porosity logging tools (12) generally consist of a neutron source and two detectors at different spacings from the source (on the order of tens of centimeters). The instrument records the ratio of the two counting rates, which is a measure of the spatial distribution of the scattered neutrons and thus the moderating properties of the formation. In addition, the pair of detectors provides a rough compensation for borehole size and fluid variations. The source of high-energy neutrons is a mixture of Am-Be or Pu-Be and furnishes roughly 10^{7} to 10^{8} neutrons per second with an average energy of 4 MeV. The detectors are gaseous, and the detection material is composed of nuclei with large thermal neutron-absorption cross sections, such as ³He. In some devices (known as epithermal), these detectors are covered with an absorbing foil of Cd to prevent detection of the lowest energy or thermal neutrons, and thus more accurately determine the moderating properties, rather than the absorption properties of the formation.

The rock property that is most closely related to the ratio measurement of an epithermal neutron porosity tool is the slowingdown length. The slowing-down length of a formation is roughly the average distance a neutron will travel before its energy is reduced to an appropriate level for detection. For the epithermal logging tool this is roughly 0.2 eV. Because the neutron moderation is a random walk, the slowing-down length is proportional to the square

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Fig. 5. Neutron and density log estimates of porosity (ϕ_{th} , thermal neutrons; ϕ_{epi} , epithermal neutrons; ϕ_d , density). Agreement between the three is seen in a sandstone layer, which contains no clay minerals. In the shale zones above and below, the neutron estimates exceed the density estimate because of the high H concentration associated with the clay minerals. [Adapted from (11)]



root of the number of collisions required to thermalize the neutrons. In water, the number of collisions necessary is about 16, whereas in a limestone with no H present about 132 collisions are needed. Slowing-down lengths for these two extremes are 8 and 30 cm, respectively.

Neutron porosity logging tools that detect thermal neutrons have an advantage over epithermal devices because they generally record much higher counting rates, reducing statistical error. However, the measurement is complicated by the fact that, at thermal energies, absorption competes with scattering, reducing the number of neutrons available for detection. A common absorber with a large cross section is Cl, which is associated with the brine that fills most rock pores. Thus, the thermal neutron porosity measurement depends not only on the slowing-down length but also on the formation absorption cross section for thermal neutrons.

The rock formation slowing-down length measured by the epithermal neutron tool can be independently calculated from the elemental composition of the rock and the energy-dependent cross sections for neutron interaction that are associated with these elements (13). More concisely, the slowing-down length depends on the formation bulk density, ρ_b , and the weight fraction of H and several abundant low-mass constituents such as C, O, Si, and Ca (14). In an ideal formation, where H is present only as fluid in the pore space, the response of the epithermal neutron instrument can be directly translated into fractional porosity. Most sedimentary formations, however, contain additional H in the form of hydroxyls associated with clays and other minerals.

Figure 5 shows a log of about 30 m in a well where two neutron measurements of porosity, along with a density estimate of porosity, were made. In the zones of porous, water-filled sandstone, free of clay minerals or shale, the three porosity estimates agree. In shale zones, where structural hydroxyl is associated with the clay minerals, both neutron measurements show a porosity much greater than that estimated from the density measurement. The measurement associated with thermal neutrons usually shows an even higher apparent porosity than does the epithermal neutron measurement because of the presence of thermal-neutron absorbers associated with the clay minerals. Separation between the two neutron porosity estimates can be used to estimate the absorption cross section of the formation.

In the clean (clay-free) sandstone zone indicated in Fig. 5, the density porosity estimate agrees with the neutron porosity estimates. This porous formation may be filled with either water, liquid hydrocarbon, or a mixture of the two. Electrical conductivity measurements would typically be used at this point to quantify the presence of hydrocarbons. However, in a gas-bearing zone the density porosity and neutron porosity estimates may be indicative of the presence of hydrocarbons by themselves. In contrast to the



behavior in shale zones where neutron porosity exceeds density porosity, in gas-bearing zones (not shown in Fig. 5) the apparent neutron porosity may be very low while the density porosity estimate may remain high. This behavior of the two porosity estimates is the result of reduced H density in a gas-filled zone which increases the slowing-down length of the formation. The lower density gas decreases the bulk density, thus increasing the density porosity estimate. The effect is to produce a characteristic crossover of the two porosity estimate curves, a clear signal of the presence of gas. This characteristic response explains the popularity of neutron and density measurement combination in the petroleum industry.

Neutron-Induced Gamma Ray Spectroscopy

Specific measurements of the geochemical properties of formation rock can be estimated by spectroscopy of neutron-induced gamma rays as is done in neutron activation analysis. The technique relies on the prompt emission of one or more characteristic gamma rays that accompanies the capture of thermal neutrons by most of the formation nuclei. Spectroscopic detection of these gamma rays allows the identification of the nuclei and a quantification of their abundances. Some important distinctions can be made between activation analysis done in boreholes versus in the laboratory. In the laboratory, a stable and intense source of neutrons irradiates a small sample and elemental references that are surrounded by an efficient detection system. In the borehole, a relatively weak and sometimes unstable source is used to irradiate a semi-infinite sample whose properties alter the activation flux. The resulting gamma rays are measured by a gamma ray detector, which is, by necessity, geometrically inefficient. Despite these obstacles, accurate elemental concentrations are determined.

A logging system (15) designed to make such geochemical measurements is shown in Fig. 6. Its length is approximately 20 m, and it incorporates four nuclear logging tools. The top measurement section consists of a large scintillator crystal and photomultiplier used to measure the natural gamma ray activity caused by the presence of U, Th, and K. Multichannel analysis of the measured low-level gamma ray spectrum allows determination of the concentrations of these three elements (16) generally associated with clay minerals. A more direct measurement of the presence of aluminosilicates is made by neutron-induced aluminum activation (17). In order to activate Al, a source of low-energy neutrons (252 Cf) is contained in the second instrument, along with an array of neutron detectors used for the measurement of slowing-down length. The

characteristic 1.78-MeV gamma rays resulting from the capture of thermal neutrons by ²⁷Al are detected by the third stage, which is nearly identical to the topmost instrument. The gamma ray spectra from these two instruments are subtracted to obtain the net activated Al counting rate. Corrections for the absorption properties of the borehole fluid and the formation must be made in order to convert this counting rate to Al weight fraction.

The bottommost instrument of the geochemical logging system consists of a miniaturized 14-MeV neutron generator that cyclically produces a burst of neutrons and a gamma ray detector with its associated timing and energy analysis circuits (18). In the interval between pulses, prompt gamma rays produced from capture of thermal neutrons in the rock formation and borehole are recorded. The thermal-neutron absorption cross section of the formation is estimated from the overall rate of decay of the gamma ray spectra. This decay rate is frequently dominated by the presence of Cl in the brine, and in the petroleum industry, this value is used as a method of distinguishing the relative fraction of brine and hydrocarbon in the pore space. However, interpretation of the measurement requires an independent knowledge of the water salinity. Another logging method for determining the fraction of hydrocarbon and brine relies on a measurement of gamma rays from the inelastic excitation of C and O nuclei (19, 20).

Of great interest is information contained in the spectrum of capture gamma rays. Laboratory calibration of the bottommost instrument is used to establish a catalog of standard spectra for individual elements. Using this catalog, one can perform a weighted least-squares fit to the measured gamma ray spectrum to determine each element's contribution. Focusing only on those elements associated with the rock matrix, rather than the fluid, allows measurement of the concentrations of Si, Ca, Fe, Ti, and Gd (21).



Fig. 7. A geochemical log showing sandstone, shale, and limestone sequences. [Adapted from (15)]

Detectability of elements in the rock depends on their concentrations and their cross sections for the production of gamma rays. An abnormally large capture cross section accounts for the appearance of Gd in the preceding list, even though its concentration in crustal rocks is on the order of tens of parts per million.

A log from such a device presents the elemental concentration of nine elements as a function of depth (Fig. 7). The solid circles are the elemental concentrations derived from x-ray fluorescence analysis of core samples from this well, and the two data sets show excellent agreement, despite the difficult measurement conditions for the gamma ray spectra. Indicated on the figure are the positions of two thick shale zones separated by a thin section of limestone in the lower half of the log. In the upper zone there are two sandstone sections separated by a thin shale layer. The top sand is "cleaner" than the lower sand.

Elemental concentration logs are of great interest in themselves and have been used in a number of scientific drilling projects (22, 23). The logs have proved useful in the continuous identification of lithostratigraphy in wells with poor core recovery. In the petroleum industry, a great deal of work has been done to convert the geochemical measurements to mineralogical abundances (24), so that an accurate lithological description can be provided. Such data can result in a greatly improved porosity evaluation from the density measurement, because a continuous log of grain density can be computed from the mineral log. Also, considerable success has been achieved in identifying and quantifying a number of commonly occurring clay minerals that usually have deleterious effects on the measurements and interpretation of electrical conductivity measurements (25).

Borehole geophysics, the study of crustal rock properties through measurements in wells, is still in its infancy and is dominated by determinations of physical rock properties that can be directly related to seismic measurements. Efforts such as the Deep Sea Drilling Project have expanded the scope of borehole geophysics by routinely including well logging measurements. In particular, nuclear measurement techniques based on neutron and gamma ray scattering techniques provide a rich variety of additional lithological information. Improvements in instrumentation should make geochemical measurements routine and an integral part of every well logging survey, allowing geologists and geochemists to supplement their traditional observational methods.

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