Discrimination of Radionuclides in High-Resolution Spectral Gamma Logging

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ABSTRACT

Since 1995, the Hanford Geophysical Logging Project has been performing high-resolution spectral gamma logging in steel-cased boreholes at the Hanford Site in Richland, Washington. The project has established a baseline of contamination conditions in the vicinity of the single-shell tank farms and recently began collecting data in boreholes associated with liquid waste disposal sites. Detection and quantification of gamma-emitting radionuclides in the subsurface is based on specific gamma energy lines between 185 and 2615 keV.

Liquid waste sites in the vicinity of the Plutonium Finishing Plant are known to contain various transuranic radionuclides, primarily plutonium isotopes, Np-237, and Am-241. These are generally characterized as alpha emitters, but they do emit detectable gamma rays within the energy range of the spectral gamma logging system. The co-existence of several transuranic radionuclides presents analysis challenges because many gamma rays have similar energies (i.e., within 3 keV). Neutron activity may also be observed as a result of (α, n) interactions. Assessment of neutron activity and gamma energy spectra can provide information on the presence and concentration of transuranic radionuclides and may provide clues as to the chemical state of the waste. The presence of Am-241 presents a special problem in that shielding effects of the relatively thick steel borehole casing preclude detection of the characteristic gamma ray at 59.6 keV. A secondary gamma ray at 662.40 keV may be mistaken for Cs-137 (661.62 keV), which is commonly detected as a fission product or as an indicator of fallout from atmospheric weapons tests. Because of the large difference in gamma yields, this can lead to significant errors in contamination assessment. Strategies and lessons learned for the accurate identification and quantification of man-made radionuclides are presented.

INTRODUCTION

Spectral gamma logging has long been recognized as an effective lithologic indicator. In the natural environment, three radionuclides and their associated decay products account for the bulk of the detectable radioactivity. K-40 (half-life = 1.25 E+09 years) occurs as a trace component of all potassium (0.0117%). With decay, it emits a distinctive gamma ray at 1460.83 keV. U-238 (half-life = 4.49 E+09 years) accounts for more than 99% of all uranium. It decays to Pb-206 through a complex chain of intermediate daughters. "Natural" thorium consists entirely of Th-232 (half-life = 13.9 E+09 years). It decays to Pb-208 with a number of intermediate daughters. Table I lists various natural radionuclides with associated gamma emissions. Most geologic media have generally lain undisturbed for millions of years, and it is likely that secular equilibrium will have been established throughout the U-238 and Th-232 decay chains such that

the specific activity of any daughter is equal to the specific activity of the parent. Geologic interpretation of spectral gamma logs assumes that decay equilibrium has been established.

Table I. Naturally Occurring Radionuclides

Radionuclide	Primary Gamma Rays			Secondary Gamma Rays		
	daughter	E (keV)	Y	daughter	E (keV)	Y
K-40		1460.83	0.1067			
Th-232	Tl-208 Pb-212 Tl-208	2614.53 238.63 583.19	0.3534 0.433 0.3011	Ac-228 Ac-228 Ac-228 Tl-208	911.21 968.97 338.32 510.77	0.266 0.1617 0.1125 0.0806
U-238	Pb-214 Bi-214 Bi-214	351.92 609.31 1764.49	0.358 0.4479 0.1536	Pb-214 Bi-214 Pb-214 Bi-214 Bi-214 Bi-214	295.21 1120.29 241.98 1238.11 2204.21 2447.86	0.185 0.148 0.0750 0.0586 0.0486 0.0150

For example the "thorium" gamma ray emission at 2614.53 keV actually originates from Tl-208, and the "uranium" gamma ray emission at 1764.49 keV originates from Bi-214.

This paper presents a discussion of spectral gamma logging in steel casing for detection and quantification of man-made radionuclides associated with nuclear weapons production and offers a summary of lessons learned during more than 10 years of experience at the Hanford Site. It is not our intent to show specific logs or contaminant plumes in this discussion, but rather to discuss how contamination can be recognized and evaluated and to provide useful tips and warnings for other practitioners.

CONVENTIONAL SPECTRAL GAMMA LOGGING

Conventional spectral gamma logging is based on two assumptions: (1) that only three natural sources account for detectable activity and (2) that secular equilibrium has been established so that the specific activity of any daughter is equivalent to that of the parent. These assumptions greatly simplify the task of spectral gamma analysis and allow the use of relatively simple and rugged instruments such as NaI detectors. These detectors have relatively poor energy resolution, and it is difficult to resolve individual gamma energy peaks. Analysis and interpretation generally involves count rates in relatively broad energy "windows," and calibration involves measurement in various mixtures of sources to establish a matrix equation from which the specific activities of the three constituents can be determined.[1] This approach works well for most geologic applications, but it implicitly assumes that all detected gamma radiation can be accounted for by a mixture of three components and that all three components are present to some degree.

Gamma rays at energies greater than 180 keV can easily be detected through steel, and spectral gamma ray logging is a logical choice for detecting man-made gamma emitting radionuclides from inside steel-cased boreholes or drive tubes. Where man-made radionuclides are present, the situation becomes much more complex. Some common man-made radionuclides, such as Cs-137, emit a few distinctive gamma rays at characteristic energies. In the matrix-based approach previously described, these radionuclides would most likely be recognized by anomalous activity levels and/or apparent "interference" with one or more of the natural radionuclides. In theory, at least, it should be possible to add energy windows and expand the matrix to account for a few additional components. However, this process would require more complicated calibration measurements because it will be necessary to account for more inter-element effects. This approach implicitly assumes that all components are likely to be present to some degree, and the possibility of false positives is high. Also, it is likely that a number of man-made radionuclides may be present, and the dimensions of the calibration matrix will increase accordingly. Finally, many common man-made radionuclides, such as Eu-154, emit gamma rays at multiple energies, each with relatively poor yield, and these are not likely to be detectable under a conventional approach.

HIGH RESOLUTION SPECTRAL GAMMA LOGGING

Gamma activity associated with man-made radionuclides can occur at levels that are obviously anomalous, and simple total gamma logs based on Geiger-Müller or NaI detectors can be used to delineate contaminated intervals, even if the source of the gamma activity cannot be identified. NaI detectors with spectral capability allow some degree of identification. However, unambiguous identification and quantification of many radionuclides require the ability to isolate individual gamma energy peaks. The high-resolution spectral gamma logging system was developed specifically to detect and quantify a wide range of radionuclides. The system consists of a cryogenically cooled high-purity germanium (HPGe) detector with specialized cable and draw works. Logging is generally performed in move-stop-acquire mode where the detector is moved to the measurement depth and then held stationary for a relatively long count time. This method provides high-quality gamma energy spectra with energy resolution on the order of 2 to 3 keV and detection limits in the picocurie per gram range for most radionuclides.

Individual radionuclides are identified from one or more characteristic gamma energy peaks. For example, gamma energy peaks at 1173 and 1332 keV indicate the presence of Co-60, while activity at 662 keV (usually) indicates the presence of Cs-137.

Once identified, the apparent concentration is determined from an individual gamma energy peak by:

$$C_a = \frac{27.027}{Y_E} \times I(E) \times N_E \times K_C(T,E) \qquad \text{(Eq. 1)}$$

Where:

C_a	= "apparent" concentration (actually specific activity in pCi/g),
E	= gamma energy level (keV),
Y_E	= gamma yield, gammas per decay,

$I(E)$ = detector calibration function (inverse efficiency),
N_E = net (peak – background) count rate at energy E, and
$K_C(T,E)$ = casing correction function.

C_a is designated the "apparent" concentration because the analysis is based on the assumption that the gamma-emitting radionuclide is uniformly distributed in the media outside the casing, with unlimited horizontal and vertical extents, a condition that is seldom met. For example, a thin layer or lens of contamination would likely have an actual concentration significantly higher than the apparent concentration.

E is the energy of the gamma ray of interest, and Y_E is the gamma yield in terms of gammas emitted per decay. For any radionuclide, characteristic energy and yield values can be obtained from a number of sources, including Firestone and Shirley [2] and on the Internet from the National Nuclear Data Center (http://www.nndc.bnl.gov/). For some radionuclides where gamma emissions from a daughter are being used to quantify the parent, it may be necessary to adjust the yield value to account for branching ratios. Table II lists gamma energy values and yields for various man-made radionuclides. "Primary" gamma rays are those that are most useful in quantification, while "secondary" gamma rays help provide confirmation.

$I(E)$ is the "inverse efficiency function" that describes the detector response as a function of gamma energy.[3] $I(E)$ is determined from measurement in a series of calibration standards consisting of cylindrical blocks of concrete with particular concentrations of orthoclase feldspar, uraninite, and monazite to provide sources of K-40, U-238/U-235, and Th-232 in geologic media, such that secular equilibrium is established throughout the decay chain. This provides a large number of distinct gamma energy lines in the range of 185 to 2615 keV that can be used to determine $I(E)$ as a continuous function of energy by curve-fitting techniques.

Conventional gamma spectroscopy software is used to analyze gamma energy spectra, detect and identify peaks and determine net count rate, N_E, for each peak. The apparent concentration must be corrected to account for the fact that gamma rays originating in the surrounding formation are attenuated as they pass through the steel casing. The casing correction factor, K_C, is a function of casing thickness and gamma energy. Other correction factors may be necessary to account for the effects of water in the borehole or detector shielding.

Table II. Man-Made Gamma-Emitting Radionuclides

Radionuclide	Half-Life (years)	Primary Gamma Rays		Secondary Gamma Rays		Typical Minimum Detection Limit (pCi/g)
		E (keV)	Y	E (keV)	Y	
Co-60	5.2714	1332.50 1173.24	0.9998 0.9990			0.15
Ru-106	1.0238	511.86	0.2040	621.93	0.0993	
Sb-125	2.7582	427.88	0.2960	600.60 635.95 463.37	0.1786 0.1131 0.1049	
Sn-126	1 E+05	414.50	0.86	666.10 694.80	0.86 0.8256	
Cs-134	2.062	604.70	0.9756	795.85	0.8544	
Cs-137	30.07	661.66	0.851			0.2
Eu-152	13.542	1408.01	0.2087	344.28 964.13 1112.12 778.90	0.2658 0.1434 0.1354 0.1296	
Eu-154	8.593	1274.44	0.3519	723.31 1004.73 873.19	0.2022 0.1801 0.1227	0.2
Th-232	1.39 E+09	2614.53 238.63 583.19 911.21	0.3534 0.433 0.3011 0.266	Th-232 is listed in Table I as a natural radionuclide, but it is a source material for [233]U and may be present as man-made contamination associated with U-233 production. It is distinguished from U-233 by gamma activity associated with Ac-228 (see Table I).		
U-232	68.9	2614.53 238.63 583.19	0.3534 0.433 0.3011	U-232 most likely occurs as an impurity in U-233. It is distinguished from Th-232 by the absence of activity associated with Ac-228 (see above).		
Th-233/U-233	7340 (Th-229) 1.59 E+05 (U-233)	440.46 218.12 193.51 210.85 1567.09 465.13	0.2555 0.1140 0.0441 0.0280 0.0209 0.0203	The U-233 decay chain will *not* be in secular equilibrium. Concentration values should be reported in terms of Th-229. U-233 can be estimated from decay calculations if the approximate age of the contamination is known. For 10y < t < 100y: U-233/Th-229 ≈ exp [9.254 – 0.999ln(t)]		
U-235	7.04 E+08	185.72	0.5720	205.31	0.0501	0.6
Pa-234 (U-238)	4.47 E+09	1001.03	0.0084	766.36	0.0029	10-15
Np-237	2.14 E+06	312.17	0.386	300.34 340.81 415.76 398.49 375.40	0.0662 0.0447 0.01745 0.01391 0.00679	1
Pu-239	24110	375.05 413.71	1.554 E-05 1.466 E-05	203.55 345.01 332.85	5.69 E-06 5.56 E-06 4.94 E-06	13000
Am-241	432.2	208.01 662.40 722.01	7.91E-6 3.64E-6 1.96E-6	335.37 368.05 376.65 322.52 332.35	4.96 E-06 2.17 E-06 1.38 E-06 1.52 E-06 1.49 E-06	50000

H-1	N/A	2223.3	1.0	Prompt capture gamma ray. Secondary indication of neutron activity that may indicate alpha-emitting radionuclides.
Na-22	2.6	511 (annihilation) 1274.53	1.81 0.9994	Indicates probable presence of alpha-emitting radionuclides as fluoride compounds.

A typical log represents a very large number of gamma energy spectra. With a depth increment of 0.5 to 1.0 ft, and depths to 200 to 300 ft, hundreds of spectra must be analyzed to produce a single log. The sheer number of spectra makes it necessary to process data in batch mode. In general, the spectral analysis should be more or less automatic, with detailed examination of individual spectra limited to special cases.

Spectral analysis programs have provisions for user-defined libraries for radionuclides. Energies associated with specific radionuclides are included on the basis of the highest yields and the expectation for detection. Careful consideration must be given to gamma energy lines with similar energies. Most identification routines will "match" a peak with the closest energy in the library, but slight variations in system gain may result in "shifts" between peaks if closely spaced energies are included in the library. Therefore, individual gamma energies in the library should be no closer than about 5 to 10 keV. Automated processing and peak identification must be followed up by operator evaluation of the data. For example, Table II indicates two primary gamma rays at 375.05 and 413.71 keV associated with Pu-239. Np-237 has two secondary gamma rays at 375.45 and 415.76 keV. A typical library may be set up to identify the Np-237 using the highest yielding 312.17-keV energy peak and to identify the peaks at approximately 375 and 414 keV as Pu-239. This could result in an erroneous detection of Pu-239 at concentrations in the nanocurie range (i.e., definition of transuranic) where the detection actually reflects Np-233 concentrations of 3 orders of magnitude less.

There are numerous examples where nearly coincident gamma energy peaks can be attributed to two or more radionuclides. It is important for the spectral analysis to include examination of individual spectra after the batch is run so that these types of discrepancies can be resolved. Equally important, principal investigators must be aware of possible misidentifications reported by spectral gamma logging and from laboratory analysis of soil samples. If there is a radionuclide reported that does not "fit" with the presumed waste stream, the investigator should ask for additional analysis. Fortunately, each measurement point is related to some degree to adjacent points: an anomaly or discrepancy observed at one point will more than likely be repeated in adjacent points within a depth interval, which allows segments of a borehole to be addressed in a common approach.

FISSION AND ACTIVATION PRODUCTS

Detection and quantification of fission and activation products is relatively straightforward. Most are intensely radioactive with relatively high gamma yields. The major challenges are identification of diagnostic and supporting gamma energy lines, recognition of sum peaks and escape peaks, and deconvolution of overlapping peaks. Ideally, a gamma line can be identified with relatively few interferences.

One potential trap is the misidentification of Am-241 as Cs-137. Ordinarily, Am-241 is recognized in gamma energy spectra by the relatively intense gamma emission at 59.6 keV. This gamma ray has a yield of 0.36 gammas per decay. However, it is too weak to penetrate a significant thickness of steel and is seldom seen in borehole logging. Am-241 also emits a gamma ray at 662.4 keV, which is practically indistinguishable from the 661.66-keV gamma ray associated with Cs-237. There are no other significant gamma rays for Cs-137, and the presence of gamma activity at 662 keV is almost always assumed to be an indication of Cs-137, which is widespread as a component of fallout from weapons tests as well as one of the most common fission products. However, if transuranics are present, it is possible that the observed gamma line at 662 keV is actually associated with Am-241. The consequences of a mistake in identification can be serious: Cs-137 has a yield of 0.851 gammas per decay at 661.66 keV, while the 662.4-keV gamma ray from Am-241 has a yield of 3.64 E-06 gammas per decay. Thus, what appears to be 1 pCi/g of Cs-137 could actually be 234,000 pCi/g of Am-241.

The best way to resolve this issue is to consider the environment. If other fission/activation products such as Eu-154/Eu-152, Sb-125,or Sn-126 are present, then the activity at 662 keV is most likely Cs-137. If transuranics are suspected, then Am-241 may be present. For Am-241, other gamma lines may provide confirmation, particularly at 722 keV (1.96 E-06 gammas per decay). If the 722-keV line is also present and calculated Am-241 concentrations from both 722 keV and 662 keV are approximately equal, then Am-241 is the most likely choice. If the 662-keV gamma ray originates from Cs-137, then either the 722-keV line will not be present or Am-241 concentrations calculated from 722 and 662 keV line intensities will be very different. Note that Eu-154 also emits a gamma ray at 723.31 keV, and a mixture of Cs-137 and Eu-154 is not uncommon in fission products.

The range of activity that may be present presents problems in log analysis. At Hanford, a typical minimum detectable level for Cs-137 is generally a few tenths of a picocurie per gram. Concentrations as high as 1 E+09 pCi/g have been measured in boreholes, which represents a range of 10 orders of magnitude. At such high levels, pileup, peak broadening, and dead time effects become significant. The high-resolution spectral gamma detector is capable of measurement from background to about 5,000 to 10,000 pCi/g (Cs-137). Higher activity levels are measured with a specially configured high-rate counting system. Cs-137 tends to be the dominant radionuclide in high-activity intervals, and gamma activity from other radionuclides that may be present tends to be "masked" by very high background levels. In very high activity intervals, it is possible to identify only the dominant radionuclide. Any other gamma emitters present are likely to go undetected, although they may be seen in the margins of the zone where activity level and background are lower.

Although not considered highly mobile, Cs-137 is intensely radioactive (about 88 Ci/g). When a borehole is advanced through a contaminated zone, small amounts of Cs-137 may adhere to the outside of the casing. This may be expressed as intermittent or low-level contamination extending some distance below the actual contaminated interval. High levels of Cs-137 in a nearby pipeline or other underground structure may also be detected. Cs-137 dust from surface contamination or fallout may find its way into a borehole, where it will show up as detectable activity at the bottom of the hole. Sampling and drilling operations may spread contaminated material on the inside of the casing and may be especially noticeable at casing joints, where

specks of Cs-137 may adhere to the pipe dope used in the threads. Situations have also been observed where contaminants may have been deposited on the inside of the casing from contaminated groundwater. In many areas of the Hanford Site, cessation of liquid discharge in the past 10 years has led to declining water levels. In some older wells, it is not uncommon to see a "bathtub ring" consisting of low levels of Co-60 and/or Cs-137 near the former groundwater level.

When contamination exists on the inside of the casing, it is also possible that the detector sonde may become contaminated. The best defenses against sonde contamination are an alert operator and pre-run and post-run verification measurements. These measurements provide energy and resolution calibration for the log spectra and allow the analyst to assess any changes in detector performance that may have occurred. It is not unusual to see trace amounts of gamma activity associated with Cs-137 in both pre-run and post-run verification spectra. This activity should be relatively unchanged over the day. However, it is important to note that detector efficiency may decline by 5 to 10 percent between the pre-run verification and the post-run verification. Cs-137 activity that appears to be relatively constant can be an indication of sonde contamination, particularly where other peaks show a detectable decrease. The log engineer should collect the pre-run and post-run spectra at the same location to minimize any variability because of location. Ratios between the Cs-137 peak at 662 keV and other peaks, such as K-40 at 1461 keV may help in detecting sonde contamination.

TRANSURANIC RADIONUCLIDES

Transuranic radionuclides include various isotopes of americium, neptunium, and plutonium. These radionuclides decay primarily by alpha emission. Although gamma rays are emitted during decay, yields tend to be relatively poor, and peaks tend to occur at similar energies. The problem with misidentification of Am-241 as Cs-137 was previously discussed. Table II lists specific gamma-ray energies for common transuranics. Analysis generally involves evaluation of multiple peaks for each radionuclide to resolve interferences. Detection limits for most transuranics tend to be in the nanocurie per gram range (1000 pCi/g) because of the poor gamma yields.

The alpha activity associated with transuranics may result in neutrons generated by (α,n) reactions on light elements, notably O-18. These neutrons can be detected by a passive neutron log as a qualitative indicator of the presence of alpha activity, but it is not possible to identify specific radionuclides. Where hydrogen is present (in water or perhaps in hydrides or hydroxides) even in relatively small amounts, the hydrogen may participate in neutron capture reactions and emit capture gamma rays at 2222 keV. These gammas are easily detectable. Because there are few interferences at this energy range, the presence of gamma activity at 2222 keV strongly suggests that alpha emitters are also present.

In some cases, relatively high neutron activity has been detected, along with Na-22, as indicated by the presence of a characteristic gamma ray at 1274 keV, accompanied by the positron annihilation peak at 511 keV. This is interpreted as evidence that the transuranics are present as fluoride compounds. Na-22 has a relatively short half-life (2.6 years) and would decay away quickly if not replenished. F-19 is converted to Na-22 by the (α,n) reaction for which it has a much higher cross section than oxygen. This phenomenon is recognized in uranium and

plutonium processing, and fluoride compounds are common in the separation process. Note that the 1274.44-keV Eu-154 energy peak can be mistaken for the 1274.53-keV Na-22 peak.

Uranium-233/Uranium-232

Th-232 may be irradiated to produce U-233, which is also a fissile isotope. U-233 processing operations will generate a thorium-rich waste stream, as well as secondary waste streams contaminated with U-233. In the Th-232 decay chain, secular equilibrium is quickly established, and the resulting thorium waste would be indistinguishable from natural thorium. Detection of such waste would depend on identification of anomalous levels of Th-232. U-233 decays by alpha emission to Th-229 (half-life 7340 years). Within the U-233 decay chain, the primary gamma emitters are Bi-213, Fr-221, Th-229, and Tl-209. These radionuclides will most likely be in equilibrium with Th-229, but equilibrium between U-233 and Th-229 is established slowly so that gamma activity builds in over time. Within the timescale of nuclear processing operations (approximately the last 50 years), observable gamma activity associated with U-233 will be about 3 orders of magnitude less than the actual activity. During the thorium irradiation process, U-232 is also created, primarily from the Th-232 (n, 2n) Th-231 reaction. This creates Pa-231 (half-life 32760 years) by beta decay. Capture of an additional neutron will yield U-232 (half-life 68.9 years), which occurs as a contaminant in U-233. U-232 decays to Th-232 and then follows the Th-232 decay series. Although it constitutes only a few percent of the U-233, U-232 contributes a significant fraction of the total gamma activity. However, it is easily mistaken for Th-232 because the primary "thorium" gamma rays originate from Tl-208, which is common to both decay chains.

Significant quantities of U-233 may at first glance appear to be only a relatively minor anomaly in "natural" thorium content, based on elevated gamma activity associated with Tl-208 from U-232. Th-232 can be differentiated from U-232 by gamma activity associated with Ac-228, which is not part of theU-232 decay chain. Unusual activity at 2615 and 583 keV without accompanying activity at 911 keV may signal the presence of U-232, likely accompanied by U-233. U-233 is detected by gamma activity at 440 keV (Bi-213) and 1567 keV (Tl-209). However, activities for these radionuclides do not reflect the parent U-233 because secular equilibrium has not been established. Results should be expressed in terms of Th-229. U-233 can be estimated from the decay curve if the approximate age of the waste is known. Where the age of contamination (t) is between 10 and 100 years, U-233 concentration can be estimated by

$$C_{U-233} = C_{Th-229} \times \exp(9.254 - 0.999 \times \ln(t)) \qquad \text{(Eq. 2)}$$

NATURAL AND MANMADE URANIUM

At the Hanford Site, U-238 in reactor fuel was irradiated by neutrons from U-235 fission to produce Pu-239, which was then separated for nuclear weapons production. The remaining U-238 is, therefore, a significant waste product. But conventional spectral gamma log practice assumes that secular equilibrium is established throughout the decay chain. When uranium is mined and processed, the daughter products are removed, and secular equilibrium is disrupted. "Pure" U-238 emits relatively few detectable gamma rays, and approximately a million years are required for the Bi-214 to "build in" to levels equivalent to the parent. Unless the U-235 fraction

has been significantly enriched, "pure" or "manmade" uranium will have little detectable gamma activity. Pa-234 is a short-term daughter that emits gammas at 1001 and 766 keV. It quickly reaches equilibrium with the parent U-238, but yields are relatively poor (0.008 and 0.005 gammas per decay). Although present, these gamma rays are seldom detected at typical background levels. Therefore, the presence of gamma activity at 1001 and 766 keV without corresponding high activity at other lines, notably 609 and 1764 keV, is an indication of processed or manmade U-238. Because of the low yield for these lines, the minimum detectable limit for Pa-234 (manmade U-238) is on the order of 10 to 20 pCi/g, while the minimum detection limit (MDL) for natural U-238 is generally less than 0.5 pCi/g.

Strontium-90

Sr-90 is a common fission product and a "pure" gamma emitter with no characteristic gamma emissions. Because the beta particles will not penetrate either the steel casing or the sonde housing, it is impossible to directly detect Sr-90 from spectral gamma logging. However, Y-90, the immediate daughter, also decays by beta emission with a relatively high-energy beta at 2.28 MeV. When this beta particle interacts with the steel casing, bremsstrahlung, or secondary gamma radiation associated with the deceleration of the beta particle is generated. This is expressed in the gamma energy spectra as low-energy, incoherent gamma energy. Brodzinsky and Nielsen [4] suggested that this phenomenon could be used to qualitatively detect Sr-90. Wilson et al. [5] showed that the overall shape of the energy spectra is affected by bremsstrahlung, and that shape factor analysis could be used to detect Sr-90. McCain and Koizumi [6] evaluated gamma energy spectra from a borehole that encountered significant Sr-90 in soil samples and showed that a modified shape factor could be used to detect Sr-90 in excess of 500 to 1000 pCi/g. SF2* is defined as the ratio of total counts in the 60- to 350-keV range divided by total counts in the 350- to 650-keV range. In "clean" soil subject only to background activity, SF2* typically has a value of 3 to 4. With increasing Sr-90, SF2* increases to values between 6 to 8. Additional work will be required to assess the effect of casing thickness on bremsstrahlung and to understand how the presence of other radionuclides such as Cs-137 affect the SF2* response.

Radon

Rn-222 is a decay product of U-238. As an inert gas, it is highly mobile and can migrate through the subsurface, accumulating in boreholes and other enclosed spaces. Radon has a relatively short half-life (3.6 days). When radon decays, the daughters Po-218, Pb-214, and Bi-214 tend to "plate out" on adjacent surfaces. These radionuclides all have short half-lives and establish secular equilibrium with radon in a few hours. Because Bi-214 is the source of the natural uranium gamma rays at 609 and 1764 keV, the presence of radon daughters on the inside of the casing or the logging sonde will result in an apparent increase in natural U-238. Careful analysis can detect the presence of radon because U concentrations calculated from the 609- and 1764-keV gamma lines will diverge. This occurs because the gamma rays originating from Bi-214 deposited as a radon daughter are not passing through the casing. The casing correction factor is dependent on both casing thickness and gamma energy. For a given casing thickness, the magnitude of the casing correction decreases with increasing gamma energy. When radon is present, uranium concentrations from both the 609- and 1764-keV gamma lines are over

corrected, but the effect is more pronounced for the 609-keV gamma line. Thus, the U-238 concentration based on the 609-keV line will be greater than that calculated from the 1764-keV gamma line. When the radon is vented from the hole, this effect will dissipate in a few hours. Radon accumulations in old boreholes frequently result in discrepancies between U-238 concentrations in repeat intervals. When the interval is originally logged, radon is present and the daughters are in equilibrium. The motion of the sonde through the borehole may act to vent the radon, with the result that uranium concentrations will appear to be lower when the repeat log is run near the end of the day.

SHAPE FACTOR ANALYSIS

When man-made contamination is detected, the question arises as to whether the contamination is distributed in the formation, deposited on the inside or outside of the casing, or perhaps as a remote concentration, such as waste in a nearby pipe or other structure. Wilson et al. [5] describe how spectral shape factor analysis can be used to assess the probable nature of the observed contamination. Shape factor analysis involves ratios between peak counts and various energy ranges. This analytical method has proven useful in some circumstances, but application is generally limited to Cs-137 or Co-60 for a relatively limited contamination range. Additional study is required to assess effects related to borehole diameter and casing thickness before the method can be widely applied.

CONCLUSIONS

The presence of man-made radionuclides introduces complications in spectral gamma logging that may be confusing and occasionally misleading to log analysts familiar with conventional spectral gamma logging. The volume of data that must be reduced to produce a log argues strongly for automated data processing, but many of the factors previously described may trap an unwary user. Careful planning and evaluation of supporting data can help improve interpretation; the analyst must be able to evaluate data in the context of the borehole and waste site environment to ensure that the most reasonable interpretation is provided.

ACKNOWLEDGMENT

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