High-Resolution Spectral Gamma Logging for Characterization of Radiological Contaminant Plumes in the Deep Vadose Zone – 14538

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ABSTRACT

Broadly speaking, radiological contaminants fall into three general categories: “processed” uranium (and possibly thorium) as reactor fuel; various activation and fission products; and transuranic (TRU) radionuclides. Although some important contaminants of concern, such as tritium, technetiun-99, and strontium-90 do not emit detectable gamma rays, most radionuclides do, at least to some degree. The spectral gamma logging system (SGLS) has been developed and deployed at the DOE Hanford Site in southeastern Washington State for nearly 20 years. This system utilizes high-purity germanium (HPGe) detectors to collect high-resolution gamma energy spectra in-situ, from which specific radionclides can be identified and assayed from characteristic gamma emissions associated with decay. Minimum detectable levels (MDLs) for specific radionuclides depend on the yield (intensity) of the characteristic gamma rays and the detector count time. Under typical logging conditions, MDLs are in the range of 1 pCi/g or better for fission products, 10 to 20 pCi/g for “manmade” uranium or thorium, and 10 to 50 nCi/g for TRU.

Over the past two decades, the SGLS has been used to investigate contamination plumes in the deep vadose zone, and to establish a baseline of the nature and extent of subsurface contamination against which future log data can be compared to assess contaminant mobility.

INTRODUCTION

At the DOE Hanford Site in southeastern Washington State, plutonium production began in 1944 as part of the Manhattan Project, and continued until 1988. A total of nine reactors were operated to produce plutonium for nuclear weapons. The irradiated uranium fuel was processed in the “canyon” buildings to separate plutonium. As a result, large volumes of radioactive waste were stored in underground tanks or discharged to the subsurface in various cribs, trenches, or injection wells. As with any large industrial operation, there were also spills and “unplanned releases”. Most of the waste disposal operations occurred in the 200E and 200w areas in the central plateau portion of the Site, where depth to groundwater generally exceeds 200 ft. Geologic conditions include windblown sand and unconsolidated Ice-Age Flood deposits (Hanford unit) overlying poorly consolidated fluvial deposits (Ringold Formation) which overlie the Columbia River Basalt. Beds in the Hanford and Ringold units can be highly permeable, and groundwater flow is also affected by geologic structure and elevation of the basalt. Gephart [1] provides an excellent discussion of the Hanford Site.

At Hanford, the vadose zone acts as both a pathway to groundwater and as a reservoir for contamination. It is likely that contamination now in the deep vadose zone will continue to impact groundwater long after the waste sites and shallow contamination have been remediated.

Over the years, several thousand holes have been drilled across the Hanford Site. Geologic conditions require the use of steel casing to maintain borehole integrity. Many of the contaminants

of concern emit detectable gamma rays which can be detected through casing and high-resolution spectral gamma logging provides a means to monitor these contaminants as they move through the vadose zone.

SPECTRAL GAMMA LOGGING

Spectral gamma logging has long been used in the mineral and petroleum industry. The technique records gamma activity emitted from the surrounding formation as a function of energy level. In uncontaminated sediments, nearly all gamma activity originates from three sources:

- potassium (radioactive K-40 comprises 0.0117% of all potassium);
- uranium decay series (primarily U-238, with lesser contribution from U-235): and
- thorium decay series (Th-232)

Variations in potassium, uranium and thorium content have shown to be indicative of the nature and compositions of the surrounding formations, and the spectral gamma logs is widely used for lithology identification and correlation between boreholes. It's use as a uranium exploration tool is obvious.

In the case of the uranium and thorium decay series, most geologic media have likely lain undisturbed for periods of a million years or more, such that secular equilibrium is fully established throughout the decay series. For both uranium and thorium, most of the gammas actually originate from daughter radionuclides, such as Pb-214 and Bi-214 for uranium, and Ac-228 and Tl-208 for thorium. The result is a number of distinctive gamma emissions, mostly between 180 keV and 2615 keV. When gamma-emitting contaminants are also present, there will be additional characteristic gamma lines. When high levels of gamma-emitting contamination are present, radioactive contamination can be detected from elevated total activity alone. At lower levels, however, it is necessary to isolate specific gamma emissions for detection and assay.

Conventional Spectral Gamma Logging

Conventional spectral gamma logging equipment typically employs scintillation-type detectors, such as sodium iodide (NaI) or bismuth germanate (BGO). These are relatively sensitive and rugged, but they lack the energy resolution necessary to discriminate between characteristic gamma lines. They generally employ photo-multipliers to convert the scintillations into an electrical signal: a photo-multiplier tube can be very sensitive to magnetic effects unless properly shielded, and "open hole" spectral gamma logs may be subject to interference when run in steel casing.

Analysis of conventional spectral gamma logs is based on the assumption that all of the observed gamma activity can be attributed to either potassium, uranium, or thorium. Relatively wide spectral windows are established and potassium, uranium, and thorium are calculated from count rates in these windows, using a coefficient matrix developed during calibration [2]. Where one or two gamma-emitting contaminants are present, it is at least theoretically possible to add windows for the target radionuclides and expand the coefficient matrix. At least one additional calibration model will be required for each target radionuclide, and the coefficient matrix is expanded accordingly. There is, however, a major fallacy with this approach: it implies that the target radionucldies may be present in any measurement, and the matrix solution may well return

non-zero values even when no contamination is present.

Other methods, such as “Full-Spectrum Analysis” [3] can be used to “back out” potassium, uranium and thorium contributions from the gamma energy spectra, and any remaining activity would provide evidence of manmade radionuclides.

High-Resolution Spectral Gamma Logging

The high-resolution spectral gamma logging system (SGLS) uses high-purity germanium (HPGe) detectors to collect gamma energy spectra with resolution sufficient to detect gamma emissions characteristic of specific radionuclides. An early discussion of the high-resolution system is provided by Brodeur et al, [4].

Conventional methods for gamma spectroscopy are applied. Target radionuclides are only detected when the appropriate “lines” are present in the spectrum. Assay values are calculated from net count rates for one or more characteristic photo-peaks, using energy-dependent detector calibration and borehole correction functions.

For the high-resolution system, an inverse efficiency function is determined as a function of gamma energy from measurements made in the Hanford borehole calibration models. Curve fitting to gamma emissions from potassium, uranium and thorium is used to establish a continuous calibration function between 180 and 2615 keV. This eliminates the need for calibration models for specific radionuclides. Early calibrations are discussed by Koizumi [5, 6]. The current approach to calibration is described by McCain [7]. The models themselves are described by Stromswold [8] or McCain [9].

Correction functions are also available for steel casing and water. These are described by McCain, [10]. Gamma energy and intensity (yield) values for radionuclides can be obtained from the National Nuclear Data Center [11].

Natural Radionuclides

Characteristic emissions and yield data for naturally occurring radionuclides are listed in Table 1. These radionuclides can be assumed to be present to some degree in all geologic media.

Fission and Activation Products

Common fission products detectable with the SGLS are listed in Table 2. Activation and fission products tend to be highly radioactive, and detection limits are generally in the pCi/g range for typical borehole conditions. It may be possible to determine isotope ratios, such as. Cs-134 / Cs-137, which can help identify the specific source. For “hard” beta emitters such as Sr-90, the high-energy betas may interact with the steel casing or wall of the logging sonde. Sr-90 can be qualitatively detected as a result of low energy incoherent gammas resulting from bremsstrahluung.

Co-60 and Ag-108m deserve mention. Technically, they are activation products. They are created by neutron absorption in Co-59 and Ag-107. Both tend to be present as impurities in uranium. Much of the uranium used at Hanford was not enriched, so these elements are present.

When the fuel elements ("slugs") are processed, Co-60 and Ag-108 follow the fission products. Co-60 (half life = 1925.28 d, or 5.27 y) is widespread in high-level processing waste. [12] It decays by beta emission and emits two prominent gamma rays at 1173.23 and 1332.49 keV – both have intensities over 99%. Although generally not considered a significant contaminant, Co-60 is relatively mobile in the vadose zone and easily detectable through steel casing. It may serve as a useful surrogate for other contaminants such as Tc-99 that do not emit detectable gamma rays. Unfortunately, Co-60 has a short half live and it has decayed below detectable levels in many boreholes. Ag-108m has a more favorable half life, but it is not as common as Co-60

"Manmade" vs "Natural" Uranium

Processed or "manmade" uranium can be discriminated from "natural" uranium by the absence of secular equilibrium in the decay series. "Processed" uranium will have been chemically separated from its daughters during refining and enrichment. The major gamma emitters in the U-238 decay series are Bi-214 and Pb-214, and these will not have had time to "build in" to secular equilibrium over the time span of human activity. U-238 can be assayed on the basis of low-intensity gamma lines associated with Pa-234-m, which achieves secular equilibrium with the parent U-238 relatively quickly. Also, U-235 can be detected directly. At typical background levels, U-235 gammas are generally too weak to be detected, but they can be detected at levels indicative of uranium contamination. Detection limits for manmade U-238 are about 10-20 pCi/g (30 – 60 ppm).

Thorium and U-232

All gamma activity associated with Th-232 originates from daughters in the thorium decay series, but the situation for thorium is different. As with uranium, it is reasonable to assume that all daughters will be removed as the thorium is purified, but secular equilibrium within this decay series is re-established quickly. Th-232 must be classified as "natural" or "manmade" on the basis of activity level. For Hanford, values exceeding about 2 to 4 pCi/g would be classified as manmade.

Thorium can be irradiated to produce fissionable U-233 by neutron absorption and beta decay. U-233 emits no detectable gammas. Eventually, it can be detected by gammas emitted from it's daughters, notably Bi-213 and Fr-221, but these tend to build in to secular equilibrium very slowly. When Th-232 is irradiated, trace amounts of U-232 will also be created from the (n,2n) reaction and beta decay. U-232 follows the same decay path as Th-232, with the exception that Ac-228 is not a U-232 daughter. Hence, gamma activity from the Th-232 daughters Pb-212 (238.63 keV) and Tl-208 (2614.53 and 583.19 keV) without corresponding gamma activity from Ac-228 (911.2 keV) may indicate the presence of U-232.

Transuranic Radionuclides

TRU radionuclides are known primarily for alpha emissions, but some do emit detectable gamma, albeit at very low yields. Pu-239 is detectable at concentrations as low as 10 nCi/g. Pu-241 may also be detectable, primarily through gammas emitted by U-237. Am-241 is also detectable. The 59.53 keV gamma normally associated with Am-241 is severely attenuated by steel casing and likely will not be detectable. Am-241 does emit other characteristic gammas, but at much lower intensity. These are listed in Table 2. Note that Am-241 emits a gamma at

662.4 keV, very close to the characteristic gamma for Cs-137 at 661.62 keV. Since Cs-137 is very common, it is easy to assume that any peak at 662 keV represents Cs-137. There are no other confirming lines for Cs-137. The difference in yields is very large: The 662 keV Cs-137 gamma has a yield of 0.851 gammas per decay, while the 662 keV Am-241 gamma has a yield of 3.64 E-6 gammas per decay. If the 662 keV gamma is incorrectly attributed to Cs-137 instead of Am-241, the error can be enormous. What appears to be only 1 pCi/g of Cs-137 could represent as much as 234 nCi/g (234,000 pCi/g) of Am-241.

Henwood et al [13] discuss the use of the SGLS in a high-level waste trench at the Hanford Site where very high levels of TRU were known to exist. In addition to characteristic gammas related to decay of Pu-239, Pu-241, Am-241, and Np-237, evidence of gammas related to alpha capture reactions were also detected. Characteristic gamma emissions from Na-22 and extraordinarily high neutron activity indicate that at least some of the TRU is likely present as fluoride compounds.

CONCLUSIONS

High-resolution gamma energy spectra collected with the SGLS over the past 20 years have been used to identify and assay manmade radionuclides in the subsurface and to define a “baseline” estimate of the nature and extent of subsurface gamma-emitting contamination. Existing wells provide access to the deep vadose zone, but it is very difficult to collect samples over time. High-resolution spectral gamma logging offers an opportunity to “see” through the casing and track contaminant movement over time.

When Hanford waste sites are finally remediated and closed, it is highly unlikely that contaminants deep in the vadose zone will be removed. If closure is to be achieved with contaminants remaining in place, it will be necessary to demonstrate that they are “fixed” and unlikely to continue moving. High-resolution spectral gamma logging can help collect data to support this.

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Table 1: Naturally Occurring Radionuclides Detectable With Logging Equipment

	Primary Gamma Rays			Secondary Gamma Rays		
radionuclide	**daughter**	**E, keV**	**Y**	**daughter**	**E, keV**	**Y**
K-40[1]	Ar-40	**1460.83**	**0.1067**			
Th-232[2]	**Tl -208**	**2614.53**	**0.3534**	Ac-228	911.21	0.266
	Pb-212	238.63	0.433	Ac-228	968.97	0.1617
	Tl-208	**583.19**	**0.3011**	Ac-228	338.32	0.1125
U-238[3]	Pb-214	351.92	0.358	Pb-214	295.21	0.185
	Bi-214	**609.31**	**0.4479**	Bi-214	1120.29	0.148
	Bi-214	**1764.49**	**0.1536**	Pb-214	241.98	0.0750
				Bi-214	1238.11	0.0586
				Bi-214	2204.21	0.0486

Useful Conversions:

potassium:	1 pCi/g K-40	= 0.1221 w% total potassium	~ 0.193 ppm eU
uranium:	1 pCi/g U-238	= 2.997 ppm U	
thorium:	1 pCi/g Th-232	= 9.091 ppm Th	~ 3.32 ppm eU

1 pCi/g = 0.037 Bq/g

Environmental Dashboard for Hanford Geophysical Log Data: http://environet.hanford.gov/eda/

Radionuclide Data (National Nuclear Data Center): http://www.nndc.bnl.gov/chart/

[1] K-40 represents 0.0117% of total potassium

[2] Th-232 occurs naturally in geologic materials. At Hanford "background" values are generally in the range of 0.5 to 1 pCi/g. Th-232 will establish secular equilibrium throughout the decay series relatively quickly. Concentrations above 2 pCi/g warrant further evaluation. Excess activity at 583 and 2615 keV without accompanying Ac-228 gammas may indicate the presence of U-232 (byproduct of Th-232 irradiation for U-233)

[3] U-238 occurs naturally in geologic materials. At Hanford "background" values are generally in the range of 0.5 to 2.5 pCi/g. For "manmade" U-238, the decay series will not be in secular equilibrium, and the peaks shown above will not be elevated. Elevated Bi-214 and Pb-214 concentrations may also occur temporarily as radon (Rn-222, 3.82d) daughters.

Table 2: Man-made Gamma Emitting Radionuclides Detectable with Logging Equipment

Radionuclide	half life (years)	Primary Gamma Rays		Secondary Gamma Rays		Typical MDL, pCi/g
		E, keV	Y	E, keV	Y	
Co-60[4]	5.2714	**1332.50**	**0.9998**	**1173.24**	**0.9990**	0.15
Sr-90	28.8	no specific gammas but concentrations > 500 pCi/g generate detectable bremsstrahlung				
Ru-106	1.0238	511.86	0.2040	621.93	0.0993	
Ag-108m[5]	438	722.91 433.94 614.28	0.908 0.905 0.898			
Sb-125	2.7582	427.88	0.2960	600.60 635.95 463.37	0.1786 0.1131 0.1049	
Sn-126	2.07E5	414.52	0.977	666.16 694.83	0.999 0.959	
Cs-134[6]	2.06	604.72	0.976	795.86	0.855	
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
U-235	7.04E8	185.72	0.5720	205.31	0.0501	0.6
U-238 (Pa-234m)[7]	4.47E9	**1001.03**	**0.0084**	766.36	0.0029	10-15
Np-237 (Pa-233)	2.14E6	**311.90**	**0.385**	300.13 340.48 415.76	0.0662 0.0445 0.0173	1
Pu-239	24110	**375.05** 413.71	**1.554E-5** 1.466E-5	203.55 345.01 332.85	5.69E-6 5.86E-6 5.48E-6	20,000 (20 nCi/g)
Pu-241 (U-237)	14.3	**208.005**	**5.19E-6**[8]	332.35	2.94E-7[2]	
Am-241	432.2	208.01 335.37 **662.40**[9] **722.01**	7.91E-6 4.96E-6 **3.64E-6** **1.96E-6**	368.05 376.65 322.52 332.35	2.17E-6 1.38E-6 1.52E-6 1.49E-6	50,000 (50 nCi/g)

[4] Results from activation of Co-59 present as an impurity in unenriched uranium
[5] Results from activation of Ag-107 present as an impurity in unenriched uranium
[6] Results primarily from activation of stable Cs-133 created as a fission product, level depends on burnup
[7] Pa-234m is a short term daughter of U-238. Other gamma-emitters in the U-238 decay series not likely to be present in processed uranium.
[8] Yield corrected for branching ratio of 0.0000245
[9] May be mistaken for Cs-137 in boreholes. (59 keV characteristic gamma ray suppressed by casing)