In-Situ Assay of Transuranic Radionuclides in the Vadose Zone using High-Resolution Spectral Gamma Logging – A Hanford Case Study - 10438

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ABSTRACT

High-resolution spectral gamma logging in steel-cased boreholes is used to detect and quantify transuranic radionuclides in the subsurface. Pu-239, Pu-241, Am-241, and Np-237 are identified based on characteristic decay gammas. Typical minimum detectable levels are on the order of 20 to 40 nCi/g. In intervals of high transuranic concentrations, gamma rays from other sources may complicate analysis and interpretation. Gamma rays detected in the borehole may originate from three sources: decay of the parent transuranic radionuclide or a daughter; alpha interactions; and interactions with neutrons resulting from either spontaneous fission or alpha particle interactions.

INTRODUCTION

Geophysical logging has been used to investigate subsurface contamination at the Hanford Site since the 1940s. Log data can be acquired cost effectively with minimal personnel exposure and used to evaluate a very complex mix of transuranic (TRU) radionuclides in the subsurface. Data from selected boreholes are evaluated to:

- Detect and quantify natural and manmade radionuclides, including transuranic (TRU) radionuclides
- Assess plutonium and americium mobility by comparing data over time and by using Am-241/Pu-239 ratios to differentiate waste streams
- Estimate approximate age of waste from Pu-241/Pu-239 ratio
- Assess chemical speciation of Pu-239 and other TRU

High levels of TRU contamination results in a complex set of reactions that include alpha and neutron interactions, in addition to ordinary radioactive decay. Gamma rays resulting from all these reactions are detected in the gamma energy spectra.

SPECTRAL GAMMA LOGGING SYSTEM

The high-resolution spectral gamma logging system (SGLS) utilizes cryogenically cooled coaxial high-purity germanium (HPGe) detectors. Gamma counts are recorded in 4096 channels, with energy resolution on the order of 2 to 4 keV. Conventional gamma spectroscopy software is used to process raw field gamma energy spectra. This involves identification of individual photopeaks and determination of net counts, counting error and minimum detectable activity. Radionuclides are identified from characteristic gamma energies, and concentrations are calculated from net count rate at specific photopeaks under the assumption of a uniform, infinite, homogeneous distribution around the borehole. In addition to net counts and detector efficiency, gamma yield (gammas per decay) is an important factor,

particularly for TRU where gamma yields are very low. Finally, corrections are applied for detector dead time and borehole factors such as casing thickness or water.

The detector calibration function and casing correction function are determined through measurements in borehole calibration models at the Hanford borehole calibration facility. The models consist of concrete cylinders containing minerals rich in potassium, uranium and thorium. This provides a wide range of gamma rays from K-40 and the uranium and thorium decay series, whose intensities are well known and stable. Most of these gammas have energy values between 185 keV and 2615 keV. The detector calibration function is determined by curve fitting to various gamma photo peaks in this range and reported as a continuous function. This allows assays to be calculated for any energy value within this range. However, extrapolation below 180 keV is not recommended. The energy range over which gammas can be reliably counted is also constrained by the steel sonde housing and steel borehole casing. Both the sonde housing and the borehole casing block alphas and betas and reduce the number of gamma rays reaching the detector. The effects of the sonde housing are accounted for by making calibration measurements with the detector inside the sonde. A casing correction function, $K_{c,}$ is provided to account for the effects of the borehole casing. K_c is a function of both gamma energy and casing thickness. Gamma attenuation through steel increases significantly for gamma energies below 180 keV. All assays of radionuclides from borehole spectral gamma log data must therefore be based on gamma rays with energies between 185 and 2615 keV. Many characteristic decay gammas for TRU fall below this range. Prominent gamma rays with lower energies may be cited for confirmation purposes, but should not be used for analysis.

PASSIVE NEUTRON LOGGING SYSTEM

The passive neutron logging system uses a He-3 detector to count epithermal neutrons originating in the surrounding media. Any detectable neutron activity can be attributed to the products of alpha interactions and/or spontaneous fission, and is thus at least a qualitative indicator of TRU. The most common approach to passive neutron logging is to simply remove the Am-Be source from the neutron moisture log. Typically, the passive neutron log is run in move-stop-acquire mode, with a count time of 60 or 100 seconds, and a depth increment of 1 foot, where the neutron moisture log is typically run with a count time of 15 seconds and a depth increment of 0.25 feet.

SOURCES OF GAMMA ACTIVITY

TRU waste streams represent the final steps in plutonium recovery and purification, so significant quantities of fission products would not be expected and the dominant radionuclides should be Pu, Am, and Np. These are primarily alpha emitters, but they do emit detectable gamma rays. Although alpha particles cannot penetrate steel casing and thus cannot be directly detected, alpha interactions in the surrounding media can generate secondary emissions such as gammas and neutrons, which can be detected.

Other than energy level, gammas counted by the HPGe detector provide no clue as to their origin. Analysis for identification and assay of specific radionuclides requires that the most probable source for each gamma energy peak be determined. Gamma rays detected in boreholes at TRU waste sites appear to be the result of several phenomena. These include radioactive decay, alpha interactions, and neutron interactions. Alpha interactions include “prompt” gammas emitted as a result of capture or inelastic scattering, and delayed gammas from decay of activation products created by alpha capture. Neutrons created as a result of alpha interactions can also generate gamma rays related to capture, inelastic scattering, and activation. The wide range of contaminant levels further complicates analysis: even gamma lines with relatively low yields may be evident in the spectra when contaminant levels are very high.

Gammas from Radioactive Decay

The primary product from Hanford was weapons-grade plutonium, which is generally defined as 94% Pu-239 by weight. It contains lesser amounts of Pu-240, Pu-241, Pu-238, Pu-232, Am-241, and Np-237, but these may be more important on an activity basis. Figure 1 illustrates the "short term" decay schemes for the various plutonium isotopes. For the time frame of Hanford operations, only those members shown need to be considered. Also shown on Figure 1 are the relative proportions (in terms of both weight and activity) of each isotope based on the nominal composition of "fresh" Hanford plutonium. On a weight basis, Pu-239 is the main component, but Pu-241 represents the primary source of radioactivity. The short half-life of Pu-241 also means that total activity will change by significant amounts over the time frame of Hanford operations (a maximum of about 65 years). During this time period, Am-241 activity will increase.

Also shown on Figure 1 are the primary gamma rays used for TRU assay in spectral gamma logging. Gamma data for Pu-238, Pu-239, Pu-240, Pu-241, and Pu-242 decay were downloaded from the National Nuclear Data Center, using the decay radiation search page at:

http://www.nndc.bnl.gov/nudat2/indx_dec.jsp

Gamma data for Am-241, Np-237, U-237 and Pa-233 were also downloaded, as these are radionuclides which may also be present, or which may grow-in to detectable levels within a time frame of one hundred years. U-237 (half-life 6.75 days) and Pa-233 (half-life 26.9 days) can be expected to reach secular equilibrium with the parent radionuclides (Pu-241 and Np-237, respectively) quickly and can be used to detect and quantify Pu-241 and Np-237.

Gammas from Alpha Interactions

Most TRU radionuclides decay by alpha emission, with alpha particle energies on the order of 5 MeV. Alpha particles have a very limited range, and cannot be detected through the steel casing and sonde housing. However, interactions between alpha particles and elements in the surrounding media may result in secondary gammas and neutrons, which can be detected. This includes gammas from inelastic scattering and capture, gammas associated with decay of radionuclides created by alpha capture and neutrons generated by alpha interactions (alpha, n). The neutrons may go on to generate additional gammas rays through scattering and capture.

"Reaction gammas" have long been recognized as a means to detect impurities in plutonium and other alpha-emitting radionuclides. One early study (McKibben 1967) used gamma spectrometry to identify light element impurities in Pu-238 intended for use in isotopic power generators for space vehicles. This paper evaluated gamma emissions related to alpha interactions by measuring gamma energy spectra from samples of Pu-238 oxide before and after addition of known quantities of various compounds. Other researchers have used alpha particles generated by accelerators to assess "prompt" gamma rays related to alpha interactions. Literature related to gamma rays associated with alpha interactions was reviewed. No consistent listing of such gammas was found; each paper listed gamma rays at various energy levels for a variety of elements.

In terms of gamma activity resulting from TRU alpha interactions in the subsurface, light elements present in the soil or in chemical combination represent the most likely targets. This would include elements such as Al, Si, O, or Ca present in the soil matrix. Plutonium in a waste stream is likely to be present as an oxide, meaning that O atoms are in very close proximity and thus the most likely alpha targets when decay occurs. Other compounds such as plutonium nitrate or plutonium fluoride are also

possible. Identification of gamma rays associated with alpha capture in O, N, or F may provide useful information regarding the chemical state of the TRU.

Alpha particle interactions with fluorine also create the possibility of activation gammas through the reaction:

F-19 (alpha, n) Na-22

Na-22 (half-life 2.6 years) decays by positron emission to Ne-22, with a characteristic gamma at 1274 keV, in addition to the annihilation gamma at 511 keV. Since the half-life is relatively short, detection of Na-22 is an indication of on-going alpha capture reactions in fluorine.

Sources of Neutron Activity

TRU radionuclides can generate detectable neutrons, either from spontaneous fission, or from alpha interactions discussed above. This effect has long been recognized in the handling of nuclear materials. Table 1 provides neutron generation rates from spontaneous fission, and from (alpha, n) reactions in plutonium oxide and plutonium fluoride for significant Pu isotopes. Plutonium nitrate is not shown: the neutron generation rate in plutonium nitrate would be roughly equivalent to plutonium oxide.

Table 1 Alpha and Neutron Generation Rates In Plutonium Isotopes

Isotope	Weapons-grade composition, wt%	Average Alpha Energy, MeV	Alpha Emission Rate, a/s/g	Spontaneous Fission, n/s-g	(alpha, n) Rates	
					oxide, n/s/g	fluoride, n/s/g
Pu-238	0.05	5.49	6.40E+11	2587	1.412E+4	2.175E+6
Pu-239	93.1	5.15	2.30E+09	0.022	40.13	5653
Pu-240	6	5.15	8.40E+09	1026	148.1	2.091E+04
Pu-241	0.8	4.89	9.40E+07	0.049	1.357	170.4
Pu-242	0.05	4.90	1.40E+08	1717	2.15	269.6
Am-241		5.48	1.3E+11	1.18	2817	4.331E+5

Using the nominal isotopic composition for weapons-grade Pu, one can calculate total neutron generation rates of about 129 n/s/g for oxide and about 8457 n/s/g for fluoride. In terms of activity, this is equivalent to about 192 n/s/Ci in the oxide and 11800 n/s/Ci in the fluoride.

The disparity in neutron generation rates between plutonium fluoride and plutonium oxide has important implications with respect to identification of waste streams. In general, Pu is handled as an oxide or as a metal. Any Pu discharged in a waste stream as a metal would quickly oxidize. However, fluorination is an important process in Pu metal production, so it is possible that some waste streams could contain fluoride compounds. A waste stream containing plutonium fluoride would produce a neutron flux approximately one to two orders of magnitude greater than a comparable waste stream containing plutonium oxide. Any correlation between passive neutron activity and TRU content must take this into account. Neutron activity in the subsurface provides a qualitative indication of the presence of TRU, but it is not recommended that passive neutron activity be used to calculate TRU concentrations.

Gammas from Neutron Interactions

Neutrons created by spontaneous fission and alpha interactions described above also have the potential to generate gamma rays through inelastic scattering, capture, and activation. Since neutrons are very

penetrating, they may interact with elements present in the surrounding media, borehole construction materials, or even within the logging sonde or the detector itself.

Gamma rays associated with neutron interactions were compiled from several sources. Average background bulk composition of Hanford soils was used to list elements likely to be present in significant quantities in the surrounding soil. These include oxygen (O), silicon (Si), aluminum (Al), iron (Fe), calcium (Ca), potassium (K), titanium (Ti), and manganese (Mn). Fe is also the dominant element in the borehole casing. Additional elements likely present include hydrogen (H), carbon (C), sodium (Na), chlorine (Cl), nitrogen (N), and magnesium (Mg). Cerium (Ce) is included because cerium was frequently used as a surrogate for plutonium. Finally, elements present in the detector and/or the logging sonde were also added. These include germanium (Ge), copper (Cu), and indium (In).

Prompt gammas from thermal neutron capture in the above elements were downloaded from the National Nuclear Data Center at:

http://www.nndc.bnl.gov/capgam/indexbyn.html

Additional gammas associated with inelastic scattering were obtained from the International Atomic Energy Agency (IAEA) publication "Handbook on Nuclear Data for Borehole Logging and Mineral Analysis" (IAEA 1993). Potential activation products were identified and evaluated using the "Universal Nuclide Chart", a JAVA-based application provided by Magill and Galy 2005.

EXAMPLES FROM HANFORD

Logs from several boreholes at the Hanford Site will be presented to illustrate how SGLS logs can detect and quantify TRU over a wide range of concentrations.

Case 1: Minimum TRU

Borehole 299-W10-69 is located near the 216-T-7 tile field, just west of the T Tank Farm in the 200 West Area. The borehole is known to be on the margin of a plutonium plume associated with the 216-T-32 cribs. The borehole was logged with the SGLS in 2006, using typical logging parameters (100 second count time and 1.0 foot depth increment). Note that the total gamma count rate associated with the contamination layer at the bottom of the tile field (41 foot depth) is not any higher than natural gamma activity detected deeper in the borehole (86 to 96 feet). The passive neutron log detected some activity, but count rates were very low (less than 0.1 cps). This suggests that conventional gamma logging, even when supplemented with passive neutron logging, would not be capable of detecting plutonium at this level.

Because plutonium was suspected, all spectra were examined for evidence of a peak at 375 keV. In addition, a repeat section was run between 39 and 46 feet, using a count time of 400 seconds and depth increment of 0.5 feet. Figure 2 shows log data from W10-69. For a 100 second count time, the minimum detectable level (MDL) for Pu-239 (at 375 keV) was about 20,000 pCi/g or less. In the repeat section, the 400 second count time achieved an MDL of less than 10,000 pCi/g. Pu-239 was detected between 29 and 42 feet with a maximum concentration of approximately 29,000 pCi/g at 42 feet. Detection and assay of Am-241 in this borehole is complicated by the presence of Cs-137 and Eu-154, which emits gammas that interfere with Am-241 decay lines at 662 and 722 keV.

Case 2: High Pu-239 and Am-241

Borehole 299-W15-08 is located in the 216-Z-9 crib, near the Plutonium Finishing Plant (PFP). This borehole had been previously logged with the radionuclide logging system (RLS) in 1992. The RLS also used an HPGe detector, but detector efficiency was lower and shorter count times were used.

W15-08 was logged with the SGLS in April 2005. Log plots for this borehole are shown in Figure 3. The log showed only moderately elevated gamma activity, with a maximum of 690 cps at 53 feet, accompanied by neutron activity of about 15 cps. Pu-239 was detected from 52 to 70 feet, with a maximum concentration of 350,000 pCi/g at 53 ft. Similar values were obtained from both the 375 and 414 keV peaks. Pu-239 values in the 1992 RLS log are slightly lower, but the data appear to be under-corrected. The 1992 RLS log reported Cs-137 with concentrations less than 1 pCi/g. The 2005 SGLS log showed a plot of Am-241 calculated from both the 662 and 722 photopeaks. Both plots showed similar values, indicating that the 662 keV gamma ray identified as Cs-137 in the 1992 log actually originates from Am-241. Because of the great difference in gamma yields (0.851 gammas per decay for Cs-137 compared to 0.00000364 gammas per decay for Am-241), what appears to be minor concentrations of fission products in the older log is actually a significant concentration of TRU.

Case 3: Complex Spectra

Figure 4 shows log plots from borehole 299-W18-159, in the 216-Z-1A tile field south of the PFP. This borehole encountered intense gamma and neutron activity beginning at 10 feet deep. Neutron count rates as high as 2600 cps were encountered. Maximum Pu-239 concentrations of about 42,000,000 pCi/g were found at a depth of 11 feet. Am-241 concentrations based on the 662 and 722 keV peaks agree over most of the borehole. Am-241 values from the 722 keV gamma line are used to estimate the count rate at 208 keV, and the "excess" gamma activity at 208 keV is used to calculate Pu-241. Pu-241 values "track" the Pu-239 curve. The Pu-241 / Pu-239 ratio is estimated to be about 1.5 to 1.8. The initial composition, based on the nominal weapons-grade composition, would suggest the plutonium is about 43 to 47 years old (1963 to 1967).

Np-237 is also detected in W18-159, through gammas emitted by its daughter Pa-233 at 312 keV. Note that the profiles for Am-241 and Np-237 are different from that of plutonium and from each other. This suggests either preferential migration or different waste streams.

The neutron activity in W18-159 is much higher than that encountered elsewhere for similar levels of TRU. Moreover, gammas are detected at 1274 keV. Although Eu-154 is a possible source for this gamma ray, its existence is not supported by confirming peaks. A more credible explanation is either prompt gammas associated with alpha capture in F-19 or decay of Na-22 created from the alpha-n reaction on F-19. This is consistent with the high neutron flux and strongly suggests at least part of the plutonium is present as a fluoride compound.

At least two Pu/Am waste types were encountered. Reclamation waste originates from solvent extraction systems used to recover plutonium from various scrap and waste. Low-salt wastes originate from multiple sources such as the metal production lines, the process development laboratory, and the analytical laboratory. There are significant differences in the "radiological signature." Reclamation waste can be characterized by relatively high Am/Pu activity ratios, which result from reprocessing aged plutonium to remove in-grown Am-241 and Np-237. Thus, the waste stream is higher in Am-241 activity than would be expected from processing "fresh" plutonium. The Am/Pu ratio in weapons-grade Pu increases steadily and approaches a value of 0.42 after about 50 years. For a "typical" plutonium waste stream, the Am-241/Pu-239 ratio should be less than 0.5. Am-241/Pu-239 ratios significantly greater than

0.5 indicate either Am-241 is present as the dominant contaminant or that preferential migration has occurred.

The low salt waste streams are characterized by Am/Pu activity ratios less than about 0.5. The low salt waste is typically accompanied by fluoride compounds. In Figure 4, the amount of Am-241 relative to Pu-239 increases with depth. This suggests separate waste streams or perhaps preferential migration of Am-241.

Finally, W18-159 exhibits evidence of another contaminant stream. High concentrations of Th-232 have been reported in logs for boreholes in this area, and the condition has been described in previous log reports as "thorium disequilibrium." Close inspection of gamma energy spectra from these intervals suggests an alternative source.

In spectral gamma logging, Th-232 is detected and assayed from the 2614.53 keV gamma emitted by Tl-208, a member of the Th-232 decay series. Within this decay series, half-lives of intermediate members are relatively short, and secular equilibrium will quickly be achieved throughout the entire series. Ac-228 is a prolific gamma emitter in the early part of the Th-232 decay series. If elevated levels of Th-232 were present, one would expect equivalent levels of Ac-228 would also be present, and that Th-232 concentrations calculated from Ac-228 gamma lines, such as the 911.21 keV peak, would be equivalent to those calculated from the 583 or 2615 keV gamma lines from Tl-208. However, Th-232 concentrations calculated from the 911 keV gamma line are consistent with background levels of natural Th-232, while concentrations calculated from the 583 keV or 2615 keV gamma lines appear to be elevated. One explanation for this is that the "excess" Tl-208 counts at 583 and 2615 keV result from decay of U-232. U-232 decays by alpha emission to Th-228, and from there the decay chain is the same as Th-232, but Ac-228 is not part of the U-232 decay series. Hence, U-232 decay would result in elevated counts for 583 and 2615 keV, but not at 911 keV or other Ac-228 gammas. U-232 (half-life 68.9 years) is known to be a byproduct of irradiation of Th-232 to produce fissile U-233.

U-232 will be at a minimum of at least 1 to 5 ppm (by weight) relative to U-233 (Kang and von Hippel 2001). U-232 quickly reaches secular equilibrium throughout the decay series, and the short half-life (68.9 years) means that specific activity (decay rate per unit mass) is high.

Relatively few gammas are emitted by members of the U-233 decay series, and secular equilibrium will not be established during the time frame of Hanford operations. U-233 (half-life 1.59E5 years) decays by alpha emission to Th-229 (half-life 7340 years). The relatively long half-life of Th-229 means that a long time will be required to achieve secular equilibrium. Below Th-229, half-lives are relatively short, and subsequent daughters will quickly reach secular equilibrium with Th-229. The primary gamma emitters in this decay series are Fr-221 (half-life 4.9 months), Bi-213 (half-life 45.5 months) and Th-229.

U-232 is determined from the 2615 keV gamma ray, after correcting the count rate for the contribution from natural thorium, determined from the 911 keV Ac-228 gamma. The results show small amounts (5 pCi/g or less) of U-232 between 10 and 51 feet. From 20 to 32 feet, there is evidence of Bi-213, based on the 440 keV decay gamma. It is estimated that U-233 concentrations are on the order of about 500 to 1000 pCi/g. The Th-229/U-232 ratio appears to be consistent with U-233 containing a few ppm U-232 and aged approximately 40 years.

Several "thoria campaigns" are known to have occurred at Hanford. The PUREX facility ran two campaigns, one in 1966, and one in 1970, to recover U-233 from the irradiation of thorium. Some U-233 recovered at PUREX was concentrated at the 231-Z Building (Fluor Hanford 2001). The site description document for the National Institute of Occupational Safety and Health dose reconstruction project (ORAU, 2007) also states, "Studies on the use of U-233 were also performed" at the 231-Z Plutonium

Isolation Facility. Therefore, U-233 (with accompanying trace amounts of U-232) is a likely component in at least some of the waste streams discharged from 231-Z.

CONCLUSIONS

Passive logging techniques such as high-resolution spectral gamma and passive neutron are effective in detecting and quantifying TRU radionuclides. Under typical logging conditions, MDLs are adequate with respect to the TRU classification criterion of 100 nCi/g (100,000 pCi/g). Passive logging methods do not require large radioactive sources or downhole neutron generators and are thus easier and less expensive to run. Previous tests of active tools using isotope neutron sources or pulsed neutrons from accelerator-based generators have encountered difficulties because of the high ambient neutron and gamma flux.

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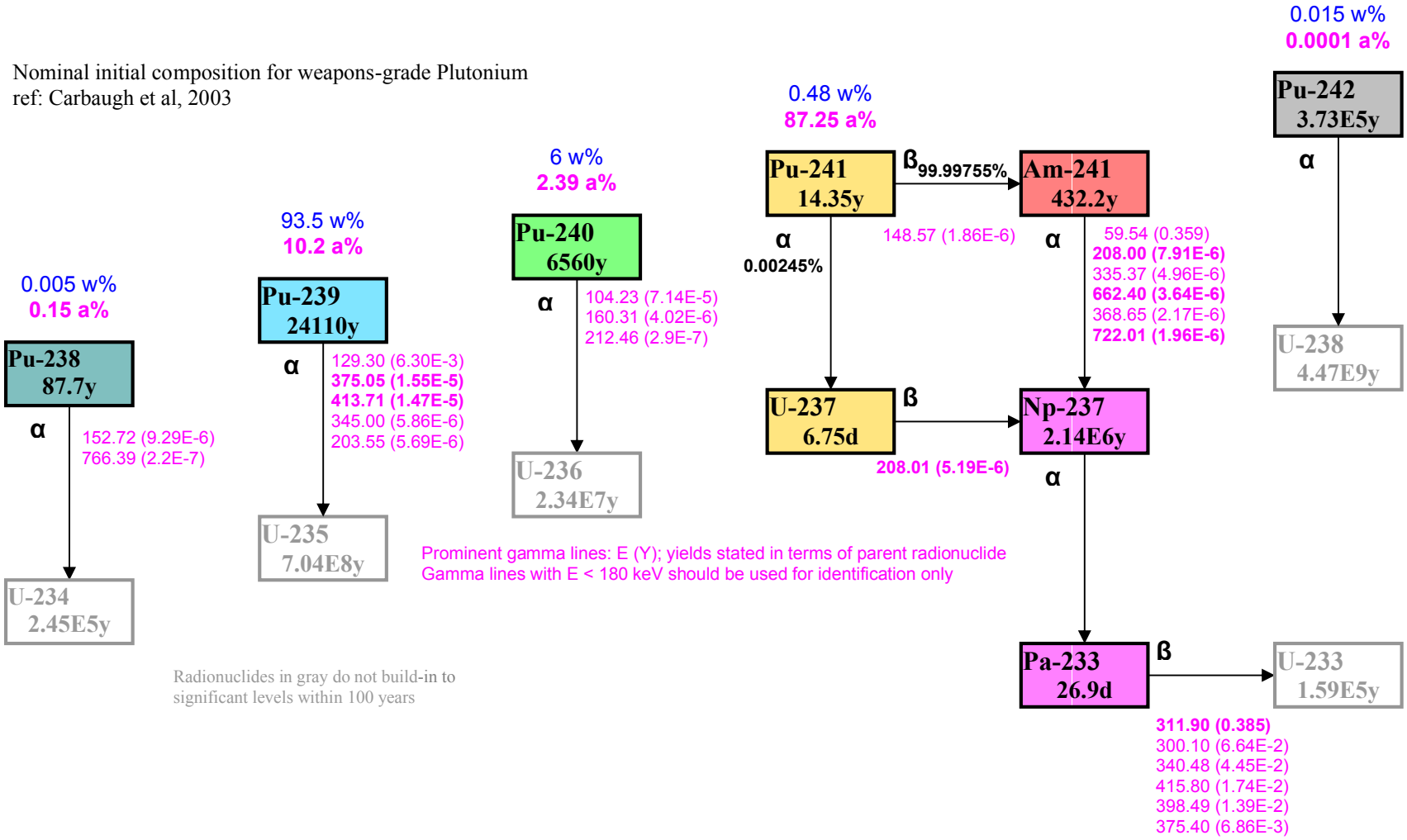

Figure 1
Simplified Decay Diagram for Plutonium and Associated Radionuclides

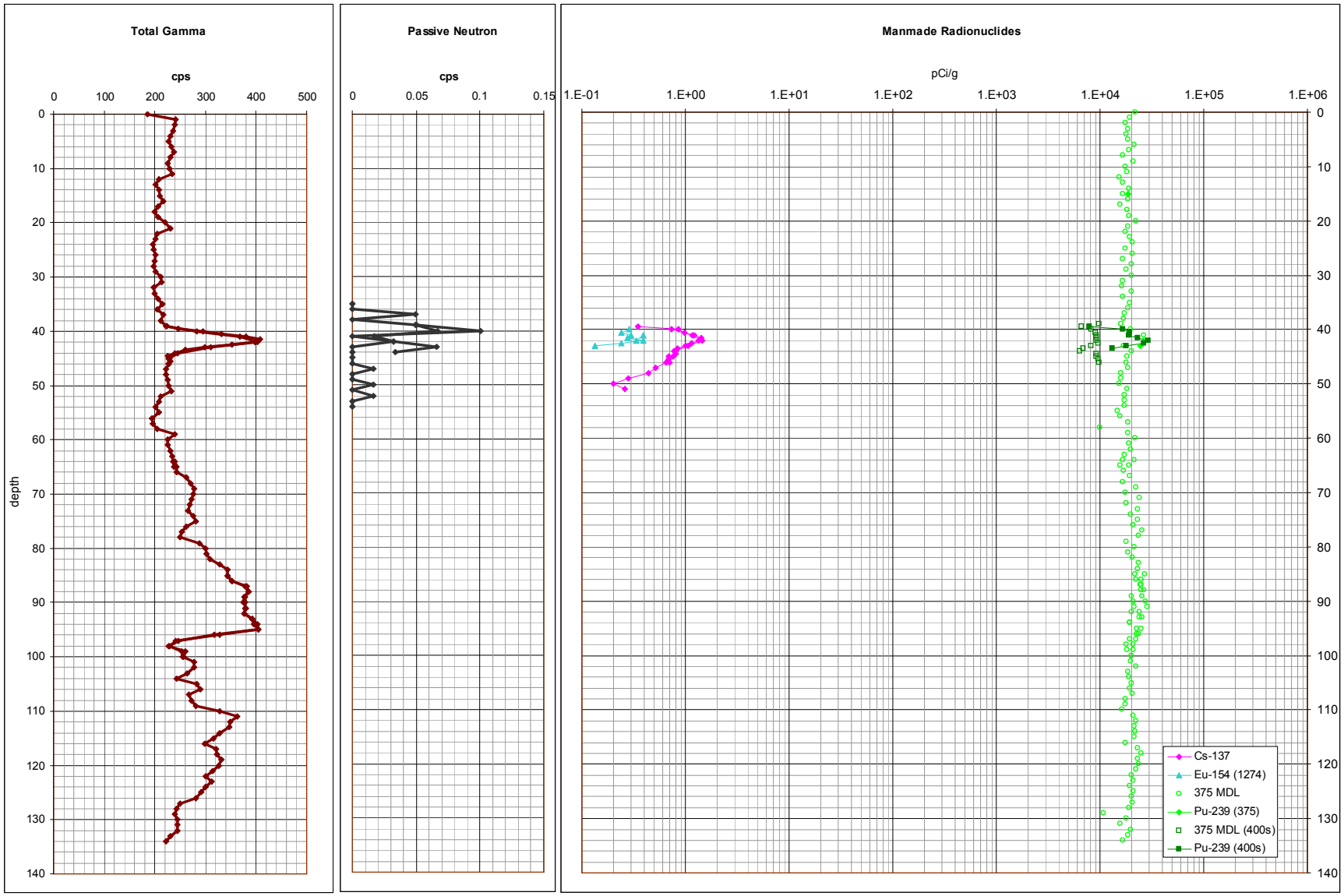

Figure 2
299-W10-69 Spectral Gamma and Passive Neutron Logs

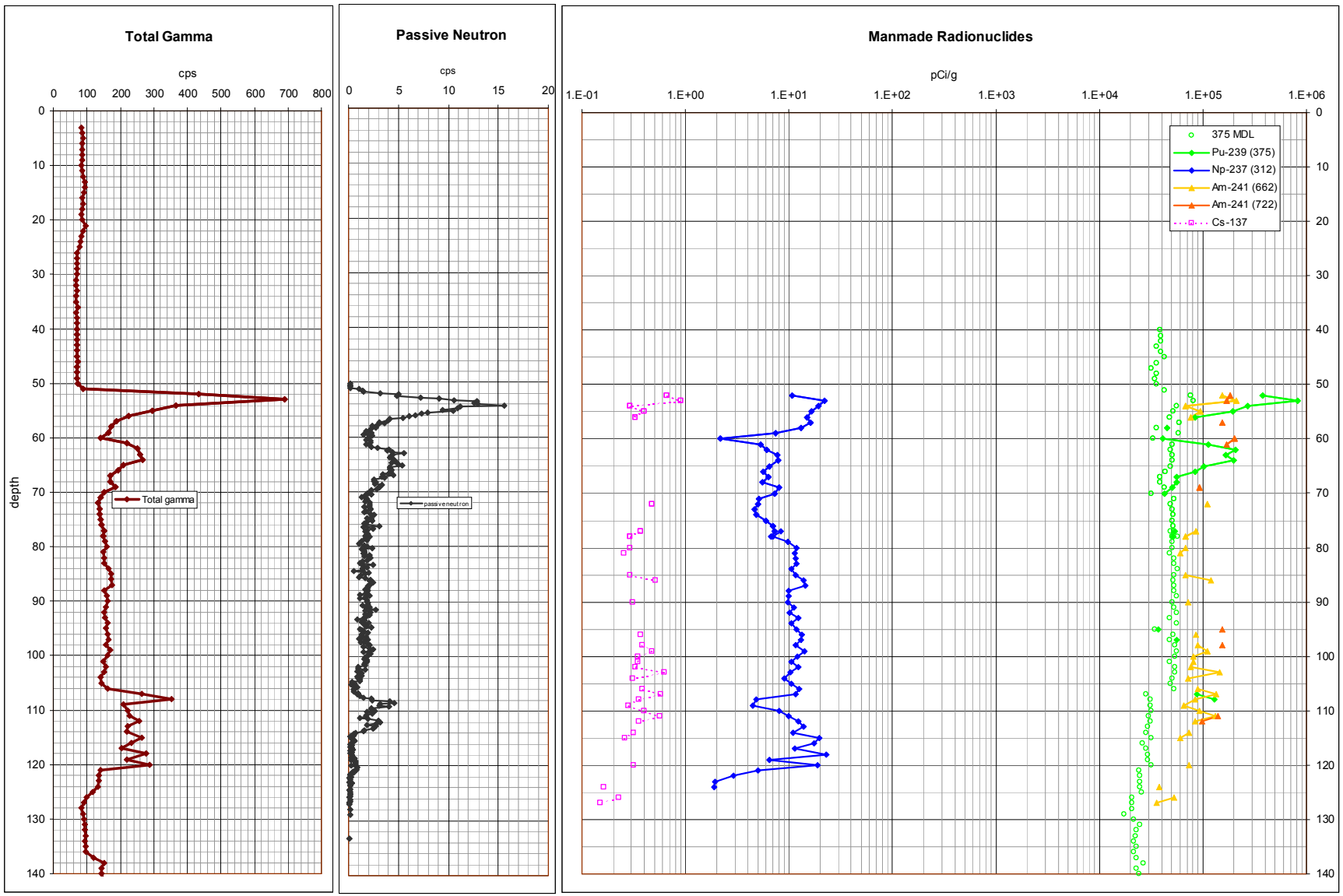

Figure 3
299-W15-08 Spectral Gamma and Passive Neutron Logs

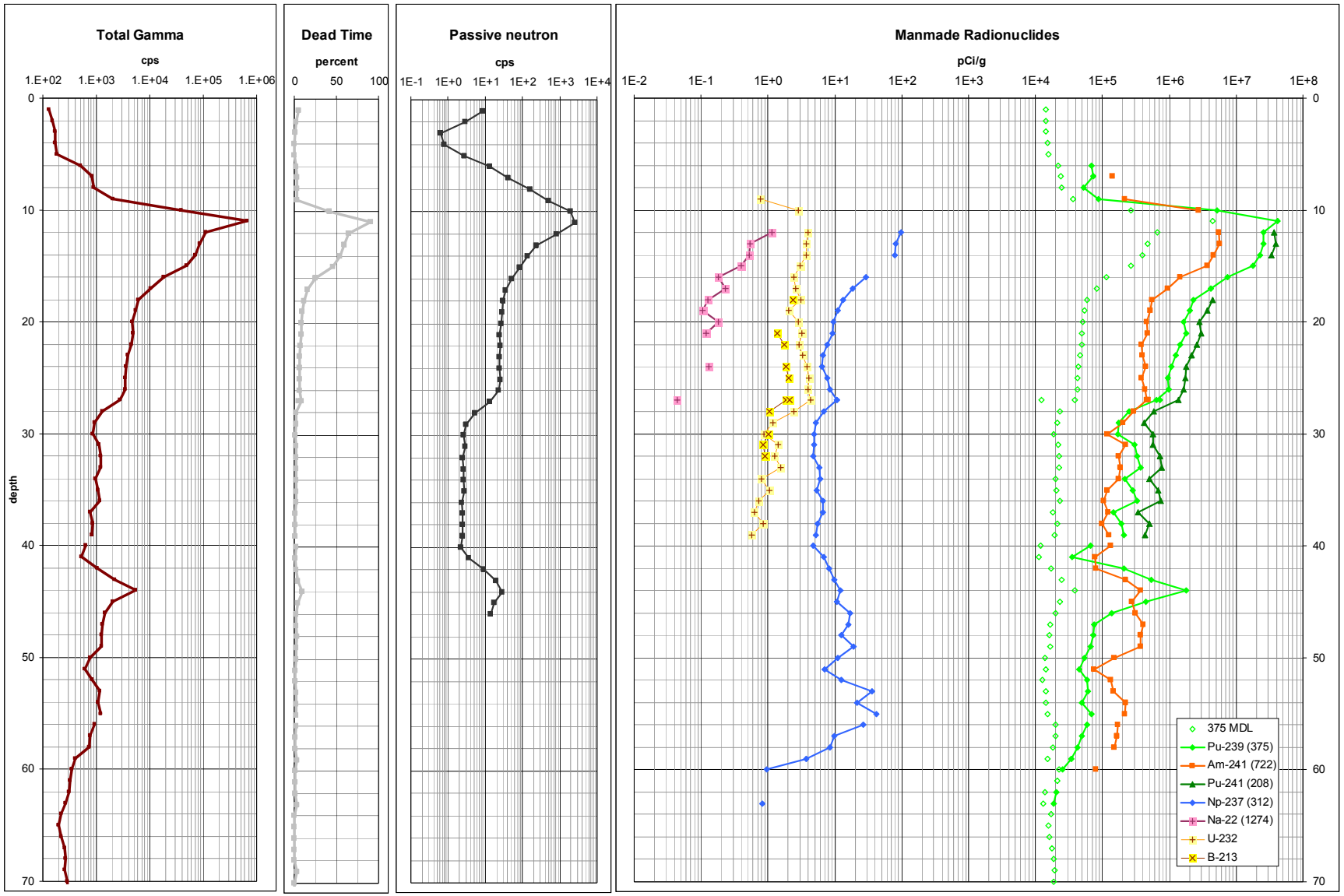

Figure 4
299-W18-159 Spectral Gamma and Passive Neutron Logs