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Rapid, autonomous analysis of HPGe gamma-ray spectra III: Plutonium identification and characterization

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February 25, 2014

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This work performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.

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Table of Contents

Rapid, autonomous analysis of HPGe gamma-ray spectra III: Plutonium analysis and characterization	1
Abstract	3
Introduction.....	3
Plutonium by grades and heat-source plutonium	4
Graded plutonium.....	4
Heat-source plutonium	4
Plutonium isotopes	5
Pu-236—Reactor poison and grandparent of a ^{228}Th gamma-ray signature.....	5
Pu-238— differentiation of RGPU from heat-source Pu and ^{238}Pu from ^{238}U	7
Pu-239—Fissile nuclide, fission energy source	8
<i>Mixed oxide reactor fuel</i>	10
Pu-240	10
<i>Spontaneous fission neutron source</i>	10
<i>Determining the presence of WGPU</i>	10
Pu-241—Fissile nuclide, ^{241}Am parent	11
Pu-242—Generally unobservable.....	11
Reaction gamma rays from plutonium	11
Alpha particle reactions.....	11
Gamma rays induced by fission.....	12
Conclusion	13
Acknowledgements.....	13
Appendix A: The uranium decay series.....	14
Appendix B: The thorium decay series.....	15
References	16

Abstract

RadID is a gamma-ray spectrum analysis program originally written to assist in the detection of the illicit movement of nuclear material. It is specific to the rapid analysis of HPGe gamma-ray data to reveal the radionuclide signatures of interest that may be present in the spectra. It is an autonomous, rule-based heuristic system that can identify well over 200 radioactive sources in about one second. *RadID* does not require knowledge of the detector efficiency, the source-to-detector distance, or the geometry of the inspected radiation source—including any shielding. In this third of a three-document series we discuss how RadID detects the presence of plutonium isotopes and determines a number of its characteristics, most notably: the spectral characteristics of weapons-grade plutonium, reactor-grade plutonium, and heat-source plutonium used in radioisotope thermoelectric generators.

Introduction

Every gamma-ray spectrum tells a story and, like a book, it can be read if you understand the language. A book in good condition with crisp print can be easily read. A book in poor condition, such as a tattered ancient biblical scroll with voids in the document, can only be read with imprecision. Similarly, a gamma-ray spectrum acquired in the field for a brief period of time and smeared by a detector with poor energy resolution is likely to be read with similar imprecision, sometimes resulting in nuclide misidentification [1].

Some gamma-ray spectra are collected by in the field, under suboptimal conditions, for short periods of time, and can result in sparse data that cannot reveal the finest details. Nevertheless, the superior energy resolution of HPGe spectra provides considerably greater information content than is found in commonly employed scintillation detectors, such as NaI(Tl) [1]. For this reason, HPGe excels in the analysis of complex spectra from sources of mixed radionuclides [2] such as plutonium. This is true even for spectra from high-resolution detectors where only the most intense peaks can regularly be observed with confidence.

RadID is a modified version of an earlier code written for nuclear emergency response. For this rapid nuclide identification is of the essence. Data acquisition times are short, in the neighborhood of 600 seconds, and almost always done out of doors. Program execution time also needs to be rapid. For this reason, RadID is based on an expert system [3,4]. Once feature extraction is performed, the system traverses a decision tree, with a Boolean result at each branch, that is accomplished in about one second.

The speed advantage of an expert system is balanced by a disadvantage: an expert system has expertise in a limited domain. If data from outside of that domain is introduced to the system, unpredictable results will likely be obtained. RadID was designed to analyze relatively sparse data of the 600-second kind. This is contrary to what most gamma-ray spectrometrists desire. A very long count of a complex spectrum, such as from plutonium for example, to obtain “better” statistics, may reveal peaks with low emission intensities that conflict with those that RadID is programmed to detect. This can sometimes lead to nuclide misidentification.

Plutonium by grades and heat-source plutonium

Graded plutonium

Most plutonium originates in nuclear reactors and is produced by the capture of neutrons by ^{238}U to form ^{239}U , which then undergoes a series of decays to form ^{239}Pu . Some of this plutonium gets consumed by fission before it is removed from the reactor, and some of it is transmuted to heavier isotopes of plutonium by capturing more neutrons, including ^{236}Pu , ^{238}Pu , ^{240}Pu , ^{241}Pu , and ^{242}Pu .

The isotopic composition of plutonium is greatly affected by how long it stays in the reactor. Short exposures in special reactors produce plutonium with very little ^{240}Pu and with very little plutonium being consumed by fission. This is the material desired for nuclear explosives.

The even-even isotopes, ^{236}Pu , ^{238}Pu , ^{242}Pu , and, particularly, ^{240}Pu decay in part by spontaneous fission, releasing fission neutrons. Large numbers of neutrons are detrimental to the performance of nuclear explosives. Long exposures produce high ^{240}Pu concentrations in, for example, power reactors.

In the United States, plutonium produced by this method is categorized into three grades as a function of ^{240}Pu content [5] as shown in Table 1.

Table 1. Pu graded by ^{240}Pu content

Weapons-grade Pu, WGPU	($<7\%$ ^{240}Pu)
Fuel-grade Pu, FGPU	($7\% \leq ^{240}\text{Pu} < 19\%$)
Reactor-grade Pu, RGPU	($\geq 19\%$ ^{240}Pu)

Heat-source plutonium

Heat from the alpha decay of substantial quantities of relatively short-lived ^{238}Pu (87.7 y) is used as the energy source for a variety of Radioisotope Thermoelectric Generators (RTG). RTG's have been used as power sources for satellites, remote space probes, the Curiosity planetary rover [6], unmanned remote facilities, and, in the past, for nuclear-powered cardiac pacemakers [7,8,9]. The intense heat generated by ^{238}Pu is evident in Fig. 1.

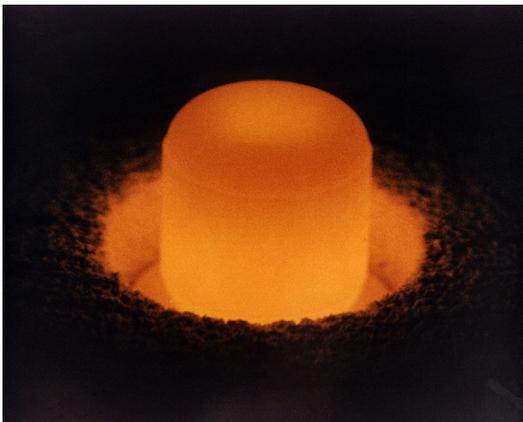


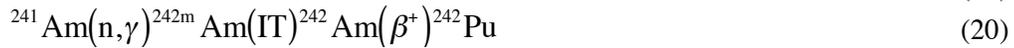
Fig. 1. A plutonium heat source glowing incandescent from ^{238}Pu decay heat. (Image from U.S. Department of Energy [10]).

Heat-source ^{238}Pu is usually produced by neutron irradiation of ^{237}Np recovered from spent nuclear fuel.

Nevertheless, the process does produce some parasitic ^{236}Pu and ^{239}Pu [11].



Production of ^{238}Pu from ^{241}Am is also possible but will also produce parasitic ^{242}Pu [11].



Plutonium isotopes

Table 2. Plutonium isotopes half-lives and significance

Pu isotope	Half-life (y)	Significance
^{236}Pu	2.9	Parent of ^{232}U and ^{238}Th , chemical yield tracer
^{238}Pu	87.74	Signature presence in RGPU and heat-source Pu
^{239}Pu	24,110	Fissile nuclide
^{240}Pu	6,563	Strong neutron emitter
^{241}Pu	14.35	Parent of ^{241}Am and ^{237}U , fissile nuclide
^{242}Pu	373,000	Trace isotope, typically not observable in a gamma-ray spectrum

Pu-236—Reactor poison and grandparent of a ^{228}Th gamma-ray signature

Tiny parasitic amounts of 2.9-y ^{236}Pu are found in all forms of plutonium. The creation of ^{236}Pu begins a decay series that is collateral to the well-known Thorium series (Appendix B). It decays by alpha decay to ^{232}U that subsequently decays by alpha emission to join the thorium decay series at ^{228}Th . The gamma- and X-ray emissions following these decays are too weak to be manifest in short-duration field measurements. Consequently the decay of ^{236}Pu exhibits a ^{228}Th spectrum. The short-lived daughters of ^{228}Th reach secular equilibrium in a matter of weeks. Its spectrum is dominated by gamma rays from beta-decay of its ^{212}Pb , ^{212}Bi , and ^{208}Tl daughters, Fig. 2.

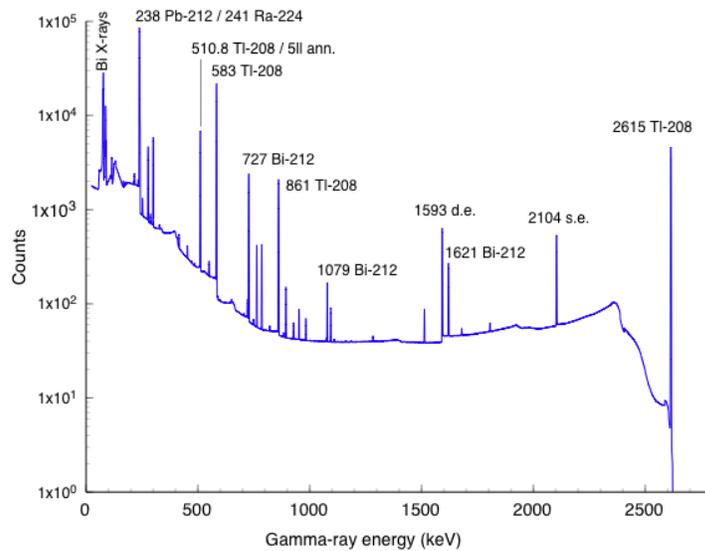


Fig. 2. Illustration of the ^{228}Th signature with a computer-simulated HPGe spectrum from a year-old ^{228}Th point source

This signature, while strongly recognizable, is fiercely ambiguous and, therefore, often of dubious value. Its ambiguity and ubiquity are illustrated by the sources that produce it, shown in Table 3.

Table 3. Source types exhibiting a ^{228}Th signature

Source Type	^{228}Th parent nuclide
Background radiation	^{232}Th
TENORM ¹ in consumer products	^{232}Th
HEU and WGU	^{232}U
^{233}U	^{232}U
Heat-source Pu	^{236}Pu
Weapons-grade Pu (<7% ^{240}Pu)	^{236}Pu
Fuel-grade Pu (7% \geq ^{240}Pu <19%)	^{236}Pu
Reactor-grade Pu (19% ^{240}Pu and greater)	^{236}Pu

The thorium series decay passes through ^{228}Ac before reaching ^{228}Th and emits a large number of gamma rays, with particularly strong emissions at 968.97- and 911.20-keV. Because ^{232}Th is a major contributor to background radiation, these gamma rays appear in background spectra that can interfere with the detection of a ^{232}U source.

In purified thorium, because ^{232}Th decays initially to 5.75-y ^{228}Ra , the following ^{228}Ac gamma rays require some time after purification before they become manifest. This has been observed in imported gas lantern mantles that still use thorium as a catalyst to provide white light.

Because ^{232}U appears in much of the world's highly enriched uranium (HEU), there is a faint hope that it can be used as a signal to detect the presence of shielded HEU. There's also the more

¹ TENORM, Technologically Enhanced Naturally Occurring Radioactive Material [12]

likely hope that it can aid in the detection of ^{233}U . Both of these hopes are subject to the ambiguities and ubiquity of the ^{228}Th signature that must be taken into account.

Pu-238— differentiation of RGPU from heat-source Pu and ^{238}Pu from ^{238}U

In heat-source Pu the isotopic fraction of ^{238}Pu is purposely dominant over other Pu isotopes. As already mentioned, short-lived ^{238}Pu (87.7 y) is used as the energy source for Radioisotope Thermoelectric Generators (RTG).

Long stays of ^{238}U in a reactor produce RGPU. The fraction of ^{238}Pu and other isotopes grow with multiple neutron capture as the fraction of ^{239}Pu diminishes through burn-up. A consequence of this is that a high concentration of ^{238}Pu is a signature for the presence of either RGPU or heat-source Pu. Examples of some isotopic vectors of RGPU and heat-source Pu are presented in Table 4. A vector of weapons-grade Pu is also included for comparison.

Table 4. Some examples of isotopic vectors of plutonium by wt% [13,14]

Plutonium type	^{236}Pu	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu
Weapons-grade Pu	Some	0.0008	93.73	6.03	0.21
RGPu (22% ^{240}Pu)	Some	.089	73.81	22.83	2.26
Heat source Pu	Some	81.2	15.8	2.5	0.1

^{238}Pu emits a number of gamma rays. Most with extremely low emission intensities, with the highest at 23.6- and 43.5-keV being too low in energy to be reliably detected in field spectra, particularly when some source shielding is likely to be present. ^{238}Pu decays to ^{234}U and populates many of some the excited states as the decay of $^{234\text{m}}\text{Pa}$, the surrogate nuclide for detection of the presence of ^{238}U . When encountered, both RGPU and heat-source Pu can be expected to be in substantial quantity—many grams. For this reason, higher energy gamma rays with emission intensities in the range of $10^{-5}\%$ and $10^{-6}\%$ will be manifest in their spectra. Good candidates for ^{238}Pu detection are the gamma rays at 766.38- and 1001.03-keV. To date, we have found that the ratio of the counts in the 1001 peak to the 766 peak fall between 0.01 and 0.08 for ^{238}Pu .

We distinguish between RGPU and heat source Pu the count ratio of the 208.00-keV gamma ray from the ^{237}U daughter of ^{241}Pu that is strongly present in RGPU, to the 766.38-keV ^{238}Pu gamma ray. This ratio can be expected to exceed unity for RGPU. Fig. 3 shows examples of RGPU and heat-source Pu HPGe spectra.

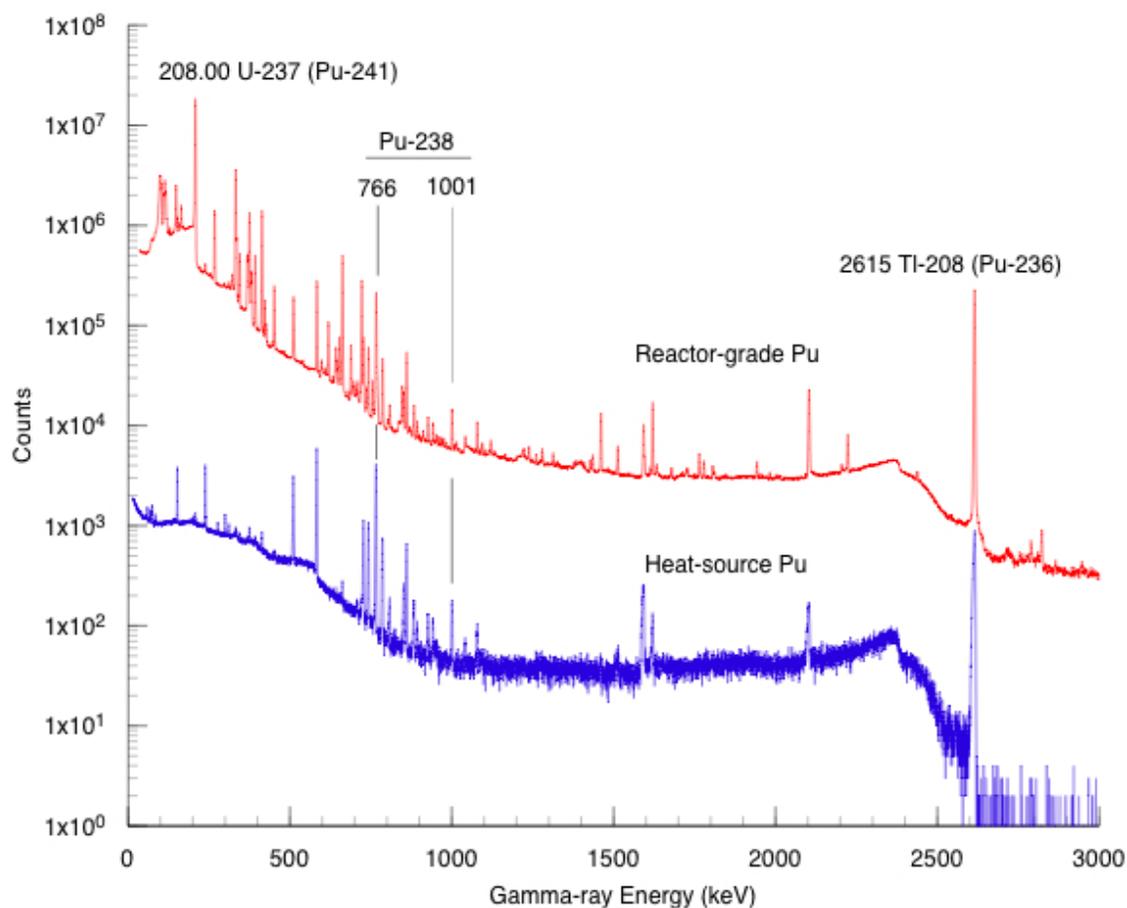


Fig. 3. HPGe spectra from reactor-grade (30% ^{240}Pu) and heat-source Pu with the 766- and 1001-keV peaks called out that could lead to misidentification of ^{238}U . Strong presence of the 208-keV gamma ray from ^{241}Pu daughter, ^{237}U , differentiates the RGPU spectrum from the heat-source Pu spectrum. The presence of parasitic ^{236}Pu is also strongly evident from the ^{228}Th signature in both spectra.

Pu-239—Fissile nuclide, fission energy source

Plutonium-239 is fissile and can sustain a chain reaction, making it useful for nuclear explosives as well as power reactors.

Use in nuclear explosives

Plutonium-239 is bred for nuclear explosives by neutron irradiation of ^{238}U in special plutonium production reactors and removed after relatively short irradiation times when ^{239}Pu is the largely dominant Pu isotope and ^{240}Pu is present at less than 7-wt% of the WGPU isotopic vector (Table 4). It is easily identifiable in a WGPU gamma-ray spectrum by gamma rays at 375.04-, 392.92-, 413.71-, and 451.48-keV. For weak sources the lines at 375 and 414 suffice for identification. If the WGPU is heavily shielded with high-Z material, and the 414-keV gamma ray is the only one observable, it has to suffice for identification. An example of the HPGe gamma-ray spectrum of WGPU, spanning 0–3 MeV, is illustrated in Fig. 4.

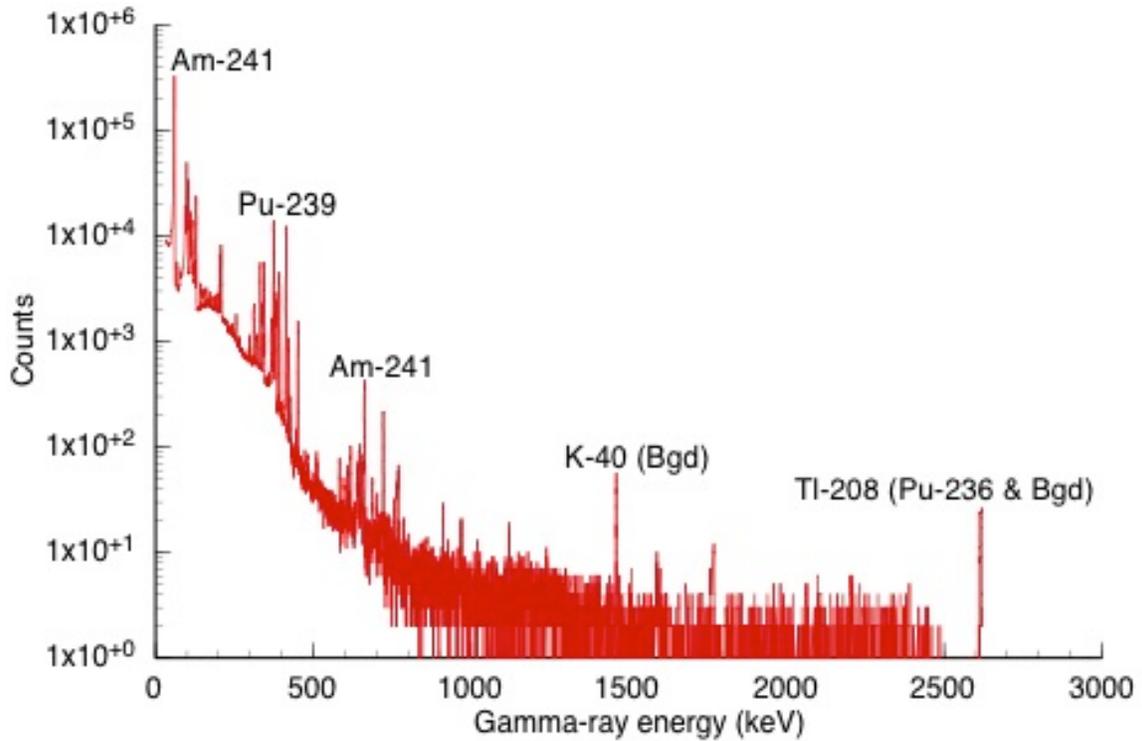


Fig. 4. The high-resolution gamma-ray spectrum of ²³⁹Pu, showing its dominant features.

Fig. 5 zooms in on a portion of n Fig. 4 to illustrate the key lines used for identification of ²³⁹Pu.

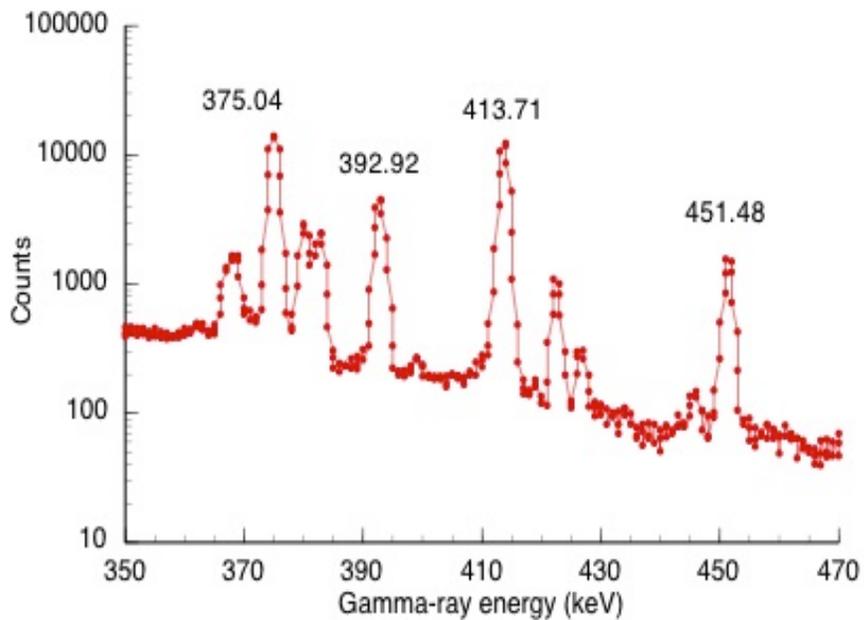


Fig. 5. The four key ²³⁹Pu gamma rays used to identify the presence of ²³⁹Pu.

Mixed oxide reactor fuel

Mixed oxide fuel, commonly referred to as MOX-fuel, is nuclear fuel that contains more than one oxide of fissile material, usually consisting of plutonium blended with natural uranium, reprocessed uranium, or depleted uranium. MOX fuel provides about 2% of the new nuclear fuel used today [15]. RadID is not currently configured to address MOX.

Pu-240

Spontaneous fission neutron source

All of the isotopes of graded plutonium exhibit spontaneous fission as a decay mode, with neutron emission from the fission fragments occurring following scission and neutron emission [16]. In weapons-grade plutonium, ^{240}Pu is the most prolific neutron emitter, accounting for its isotopic fraction alone as the criterion for plutonium grade determination. The specific neutron emission rate of ^{240}Pu is 920 n/s/g [17].

Determining the presence of WGPU

Isotopic analysis of plutonium can be done by mass spectrometry or by high-resolution gamma-ray spectrometry. In the latter case, dense multiplets in a number of selected regions of the spectrum are unfolded to yield the relative isotopic abundances.

Multiplet resolution, however, is beyond the scope of RadID. But because the plutonium items that are of interest are likely to be shielded, we are interested in the penetrating gamma rays in the 632- to 670-keV region for RadID. This region contains peaks from ^{239}Pu , ^{240}Pu and ^{241}Am , a ^{241}Pu daughter. This can be seen in Fig. 6, an illustration of the analytical results of multiplet resolution from the Pu-600 code [18], a derivative of MGA [19], developed for an arms control intuitive [20].

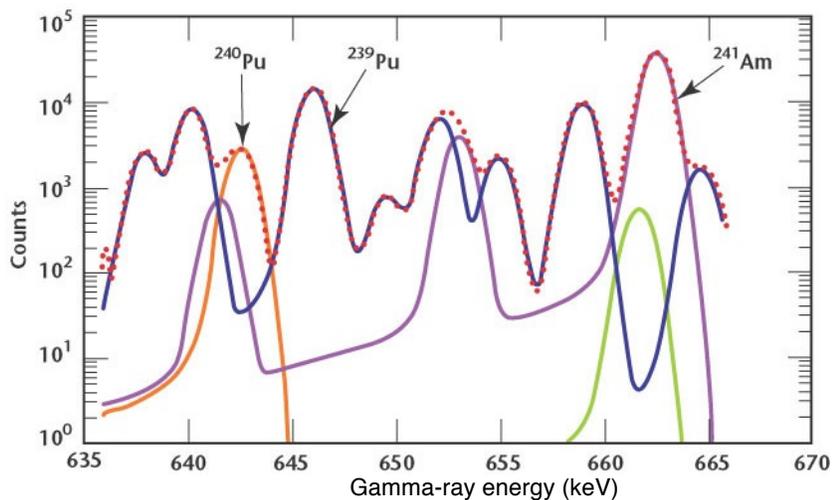


Fig. 6. Peaks in a HPGe weapons-grade plutonium spectrum (6% ^{240}Pu) are shown resolved by the Pu-600 code. The peak shown in green is from a spurious ^{137}Cs source, also resolved by Pu-600.

When the peaks in the 600-keV region are resolved, as in Fig. 6, the count ratio of the 642.35-keV ^{240}Pu peak to the counts in the 639.97-keV ^{239}Pu peak can determine an approximate

value for the ^{240}Pu isotopic abundance. To increase execution speed and reduce code size, multiplet resolution is not included in RadID. Instead we integrate the counts in two regions, 636–642 keV and 642–643.7, take their ratio and, if it exceeds a threshold value of 5, RadID declares “Pu-240 content appears low, implies WG.”

Pu-241—Fissile nuclide, ^{241}Am parent

In light water, uranium-fueled reactors, most of the fuel mass is ^{238}U . Successive neutron capture, beginning on ^{238}U , produces fissile ^{239}Pu and ^{241}Pu as well as other transuranic nuclides. Normally, the reactor’s fuel is changed every three years or so and about half of the ^{239}Pu created is “burned” in the reactor, providing about one third of the total energy. Typically about one percent of the used fuel discharged from a reactor is plutonium, and some two thirds of this is fissile (c. 50% ^{239}Pu , 15% ^{241}Pu) [15].

The fission cross section of ^{241}Pu is 1011 b, exceeding the fission cross section of ^{239}Pu at 748 b [21], making it a valuable fissile contributor to MOX fuel. However ^{241}Pu has a relatively short half-life of 14.35 years, reducing its presence by 5% each year. It decays by alpha, beta, and spontaneous fission. The alpha decay branch has only a probability of 0.00245% and goes to ^{237}Np , the principal source for breeding heat-source Pu. The beta decay branch has a probability of 99.998% and goes to nonfissile ^{241}Am with a half-life of 432.2 years. Somewhat unusually for an alpha emitter ^{241}Am produces a 59.5-keV gamma ray in 35.9% [22] of its decays. This gamma ray is the most pronounced contributor to the WGPu gamma-ray spectrum (Fig. 4). Unsurprisingly ^{241}Am is also a major contributor to the radioactivity of nuclear waste.

Pu-242—Generally unobservable

Plutonium-242 is produced by successive neutron capture, beginning with ^{238}U . Its half-life is 3.733×10^5 y [22]. Because it is not observable in most samples by primary gamma-ray emission, RadID does not address it.

Reaction gamma rays from plutonium

Alpha particle reactions

Compared to uranium, the plutonium isotopes have a high alpha particle specific emission rate and a higher alpha particle energies in the 5 MeV range. With these higher energies and emission rates, these alpha particles can induce (α, n) reactions in low-Z nuclides. Because of the very short range of alpha particles, the reacting nuclides must be intimately mixed with the plutonium or in close contact at the plutonium surface.

Below about 50 MeV, the incident alpha particle penetrates the low coulomb barrier of the low-Z nucleus and forms an excited, loosely-bound compound nucleus in about 20^{-21} s. Following a considerably longer period of time, 10^{-19} to 10^{-15} s, enough energy is concentrated on a single neutron to allow it to escape. Charged particles rarely boil off because of the coulomb barrier [23]. Following neutron emission the residual nucleus can further de-excite by gamma-ray emission. If this occurs while the recoiling nucleus is still in motion, it’s line in the gamma-ray spectrum will be Doppler broadened, as illustrated in Fig. 6 for the $^9\text{Be}(\alpha, n)^{12}\text{C}$ reaction.

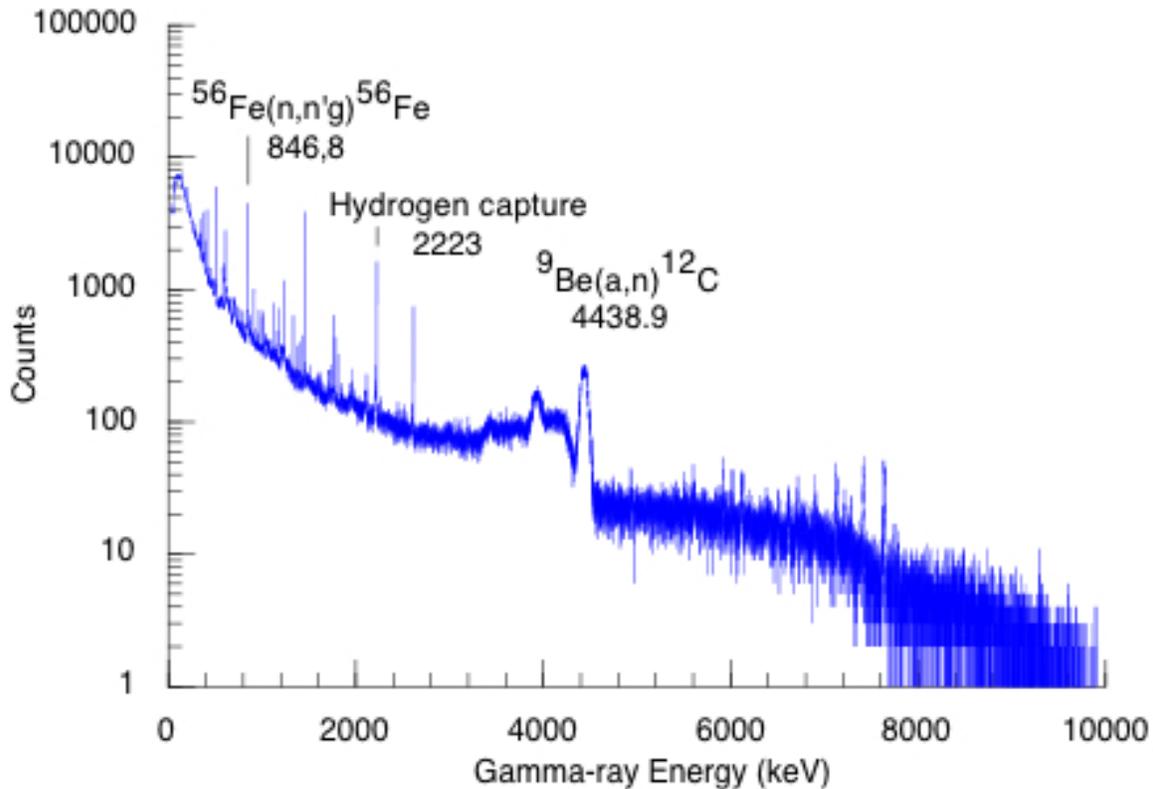


Fig. 6. HPGe spectrum of a plutonium-beryllium source illustrating the Doppler broadened 4438.9-keV $^9\text{Be}(\alpha,n)^{12}\text{C}$ peak and the hydrogen-capture peak at 2223-keV. RadID also identified by RadID is a peak from the $^{56}\text{Fe}(\alpha,n)^{56}\text{Fe}$ inelastic scattering reaction. There are other reaction lines visible on close inspection such as the iron (n,γ) reaction line pair at about 7500-keV that appears in the figure as a single broadened line.

Gamma rays induced by fission

A number of gamma rays from two processes are observable in the plutonium gamma-ray spectrum following spontaneous fission of ^{240}Pu and other Pu isotopes. Once separated, the fission fragments undergo neutron emission, followed by gamma-ray emission. In the first process, fission neutrons interact with the plutonium assembly itself and with surrounding materials in the environment, producing a manifest signatures in the spectrum. An example is the very common hydrogen capture peak at 2223-keV peak in Fig. 6.

Secondly, the fission gamma rays, both prompt and delayed, produce a broad spectrum usually most notable above 3 MeV. The shape of this “grassy” spectrum is clearly observable above 3-MeV in Fig. 6 and is better defined in in Fig. 7.

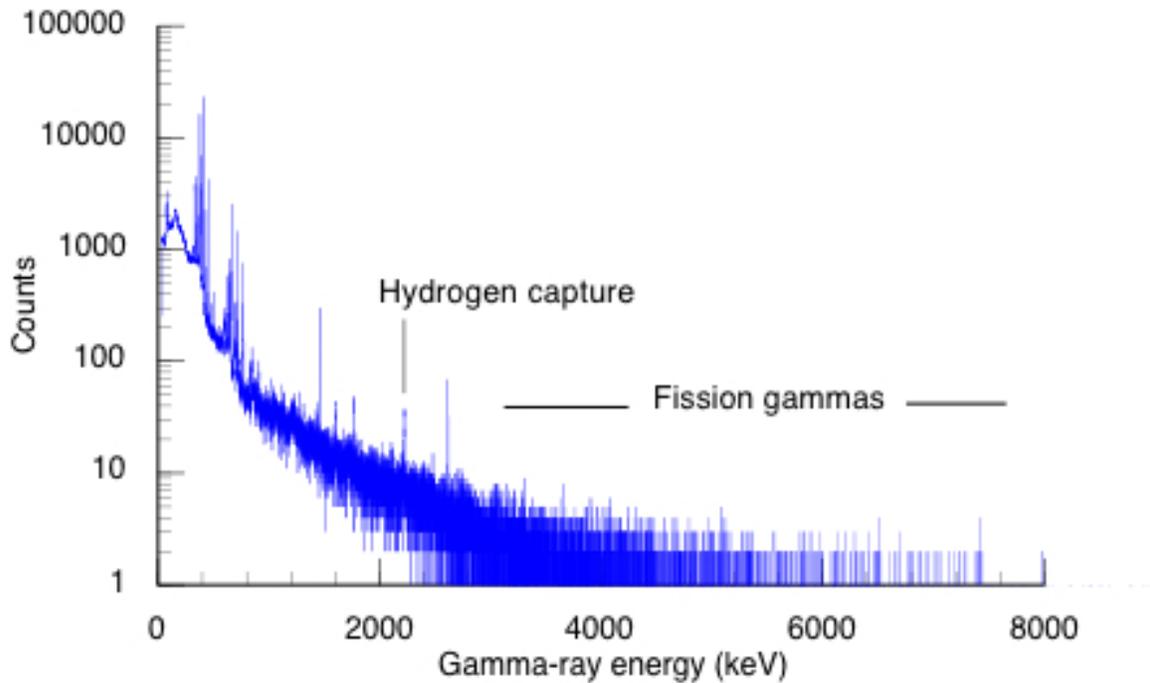


Fig. 7. A WGPU spectrum with the neutron induced hydrogen capture line and the broad distribution of fission gamma rays, both were identified by RadID.

Conclusion

Plutonium as a heat source and in three grades presents a challenge to rapidly, autonomously, and unambiguously identify its presence and its attributes from field measurements. We began this paper with a sketch of the behavior of our expert system that exploits heuristics to produce these results for uranium and similar, but less complicated results for more than 200 other radioactive sources. Following that description we discussed plutonium and plutonium grades. This was again followed by more detailed discussions of the individual plutonium isotopes and their characteristics and importance.

Future plutonium work will focus on the radiation signature of MOX fuel.

Acknowledgements

RadID is the descendent of a simple radionuclide identification code for HPGe detectors that was the brainchild of Mark S. Rowland and was implemented in software by James L. Wong.

Appendix A: The uranium decay series

The radiation signatures of natural and processed uranium in gamma-ray spectra can be understood and distinguished by the decay of ^{238}U and, importantly, by its radioactive daughters. Uranium-238 is the progenitor of a long radioactive decay series. A diagram with the main features of the uranium series is shown in Fig. A-1.

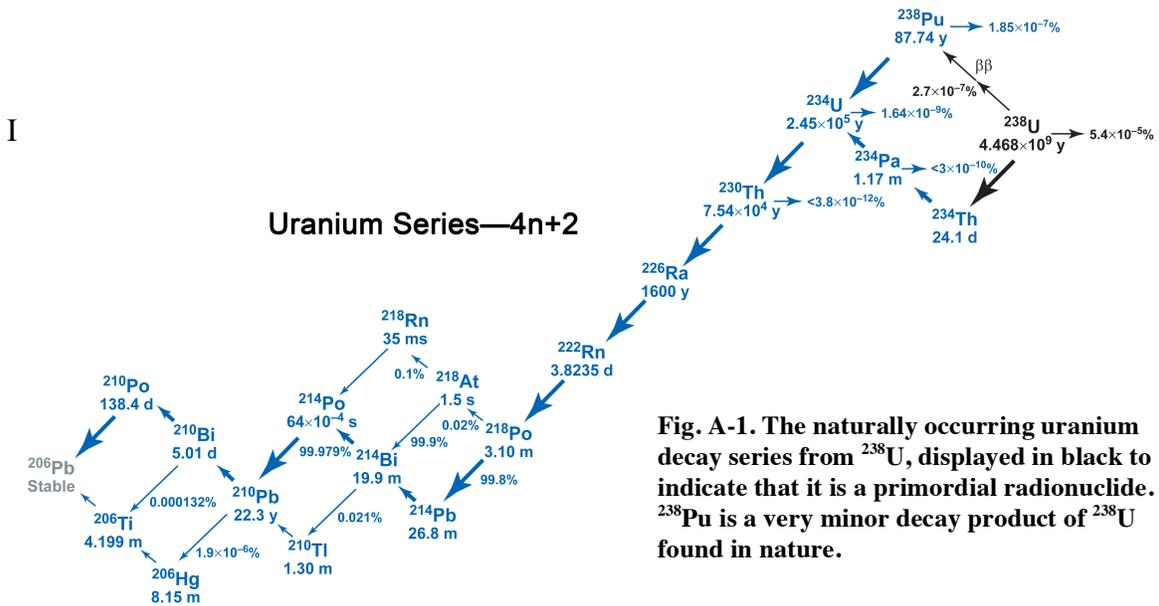


Fig. A-1. The naturally occurring uranium decay series from ^{238}U , displayed in black to indicate that it is a primordial radionuclide. ^{238}Pu is a very minor decay product of ^{238}U found in nature.

Appendix B: The thorium decay series

The gamma-ray signature of ^{228}Th is discussed throughout this paper. To see how this signature arises through radioactive decay to arrive at the signatures discussed, a graphic illustration of the thorium decay series and the collateral series beginning with ^{236}Pu shown in Fig. B-1.

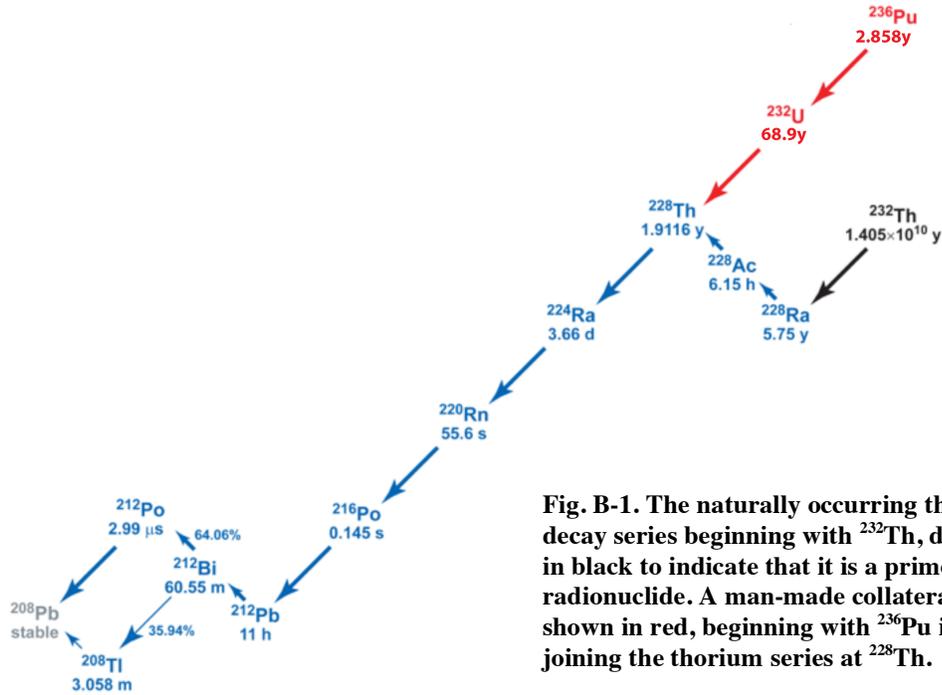


Fig. B-1. The naturally occurring thorium decay series beginning with ^{232}Th , displayed in black to indicate that it is a primordial radionuclide. A man-made collateral series, shown in red, beginning with ^{236}Pu is shown joining the thorium series at ^{228}Th .

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