IPNDV Working Group 3: Technical Challenges and Solutions
Nuclear Material (2)—Technology Data Sheet

March 27, 2017

Nuclear Material (NM) Technology Name: High Resolution Gamma-Ray Spectroscopy (HRGS)

Physical Principle/Methodology of Technology:

Gamma-ray spectrometry uses the unique signature of gamma-ray quanta to identify radio nuclides and to determine their activity. A gamma-ray spectrum shows peaks for gamma-rays that completely deposited their energy in the detector. When the spectrometer is calibrated for the gamma-ray energy, the position of the peaks and corresponding energy can be used to identify the radionuclide responsible for the peak. Once radionuclides have been identified in the spectrum, their activity can be computed if information on the detection efficiency calibration is known.

In presence of complex spectra, generally High Purity Germanium (HPGe) detectors are typically used because of their very good energy resolution. One important drawback of the HPGe detectors is that they are not easily manipulated because of the required cooling of the crystal, which is often based on liquid nitrogen. Electro-cooled detectors obviate using liquid nitrogen but are still bulky instruments. When it comes to mobility of an in-situ gamma-ray set-up, small handheld detector systems operating at room temperature are to be preferred.

This technology data sheet refers to the detection of gamma lines of U and Pu radioisotopes by means of a HPGe detector. For analysis of the spectra, two approaches are considered: (1) evaluating count rate ratios of peaks of different isotopes present at similar decay energies using (simple approach), and (2) performing a full spectral analysis using Fixed-Energy, Response Function Analysis with Multiple Efficiency (FRAM)[1] or a similar code (complex approach).

A methodology based on X-rays detection and analysis e.g., with Multi-Group Analysis (MGA) was not considered because X-rays suffer more from shielding.

Potential Monitoring Use Cases (pre-dismantlement, dismantlement, post-dismantlement, storage stage):

Ideally in conditions where shielding is not an issue.

Pre-dismantlement measurements can be used to confirm attributes such as Pu presence and isotopes, combined with other measurements to establish Pu mass. Similarly, post-dismantlement measurements can confirm the same attributes. Templates based on the gamma spectra could also be used.

With appropriate analysis of certain activation lines, the gamma spectra can be used to evaluate the presence or absence of high explosives.[2]

In this technology data sheet they refer to active interrogation with a Deuterium-Tritium (DT) generator or a ^{252}Cf with an intensity of 2×10^{7} n/s. The method relies on neutron excitation of hydrogen, chlorine, oxygen, and carbon and the detection of subsequent gamma rays. Quoting Seabury et al., “Carbon and oxygen are only readily excited by inelastic neutron scattering, requiring neutron energies of 4.4 and 6.13 MeV respectively. Hydrogen is excited by thermal neutron capture, and chlorine is readily excited by both thermal neutron capture and inelastic neutron scattering.” It is arguable whether a self-interrogation with the intrinsic neutron emission of Pu is possible; the neutron emission would be mainly due to the ^{240}Pu for a total mass of 5 kg of Pu metal and 10 percent ^{240}Pu, a neutron yield of a rough estimate of the 0.5×10^{6} n/s.

For more information, refer to the technology data sheet on interrogation with fast neutrons because it focuses on the outlined principle using an external neutron source.

HRGS could be used to monitor storage or dismantlement areas, if the detailed gamma information would be of benefit compared to low-resolution measurements that would be less expensive to install and maintain.

Used to measure U, Pu, or U and Pu:

Applicability is limited to Pu, if count rate ratios of peaks at adjacent energies are evaluated.

If a full spectral analysis is performed, HRGS can be used to measure U, Pu, or U and Pu by using the code FRAM.[3]

For detection technologies, what does the method determine/measure (e.g., presence of nuclear material, isotopics, mass, etc.)?

(1) Presence of nuclear material
- (i) Activity ratio of $^{239}Pu/^{240}Pu$, if count rate ratios at adjacent energies are evaluated
- (ii) Activity ratios and masses of U, Pu or U and Pu, and time since separation/last chemical purification, if a full spectrum analysis software is used

[1] Thomas E. Sampson, "Chapter 2, Passive Nondestructive Assay of Nuclear Materials 2007 Addendum," *LA-UR-03-4403*, available at http://www.lanl.gov/orgs/n/n1/panda/2.%20FRAM%20LAUR.pdf; and T. E. Sampson, T. A. Kelley, and Duc T. Vo, *Application Guide to Gamma-Ray Isotopic Analysis Using the FRAM Software,* LA-14018 (Los Alamos National Laboratory, NM, September 2003), available at http://www.lanl.gov/orgs/n/n1/appnotes/LA-14018-M.pdf.

[2] E.H. Seabury, D.L. Chichester, C.J. Wharton, and A.J. Caffrey, "A Comparison of Neutron-Based Non-Destructive Assessment Methods for Chemical Warfare Material and High Explosives" (INL/CON-0814209, August 2008), available at https://www.researchgate.net/publication/255827301_A_Comparison_of_Neutron-Based_Non-Destructive_Assessment_Methods_for_Chemical_Warfare_Material_and_High_Explosives.

[3] Thomas E. Sampson, "Chapter 2, Passive Nondestructive Assay of Nuclear Materials 2007 Addendum," *LA-UR-03-4403*, available at http://www.lanl.gov/orgs/n/n1/panda/2.%20FRAM%20LAUR.pdf.

Physical Description of Technology (e.g., approximate size, weight):

A wide variety of HPGe detectors, both planar and coaxial, can be used. According to Sampson, a coaxial detector is the appropriate choice for the user who makes a wide range of measurements on samples contained both in thin-walled and heavy-walled or shielded containers. Specifications for a small coaxial and a planar detector are shown in Table 1 below, taken from Sampson.

Table 1. Specifications for a Small Coaxial and a Planar Detector

Coaxial Detector	
Geometry	Coaxial, P-type germanium [1]
Efficiency	At least 25% relative efficiency at 1.33 MeV in the usual definition
1000 cps FWHM (122 keV)	≤ 750 eV at ≤ 6μs shaping time [2]
1000 cps FWHM (1.33 MeV)	≤ 1.75 keV at ≤ 6μs shaping time
30 kcps SWHM (122 keV)	≤ 880 eV at ≤ 2μs shaping time
30 kcps FWHM (1.33 MeV)	≤ 2.00 keV at ≤ 2μs shaping time
Peak Shape	[FW1/50M]/FWHM ≤ 2.50 at rates ≤ 30 kcps and time constants from 2 – 6 μs
Preamplifier/Energy Rate	Resistive feedback preamplifier with Energy Rate ≥ 50,000 MeV/s
Planar Detector	
Geometry	Planar
Crystal Size	≥ 25 mm diameter, ≥ 13 mm thick
1000 cps FWHM (122 keV)	≤ 520 eV at ≤ 6μs shaping time
50 kcps FWHM (122 keV)	≤ 580 eV at ≤ 1μs shaping time
Peak Shape	[FW1/50M]/FWHM ≤ 2.50 at rates ≤ 50 kcps and time constants from 1 – 6 μs
Preamplifier/Energy Rate	Resistive feedback preamplifier with Energy Rate ≥ 10,000 MeV/s [3]

[1] We specify P-type germanium because the resolution is usually better than that of N-type for the same detector size. N-type detectors have also been used successfully with FRAM.

[2] Shaping times are specified for analog amplifiers.

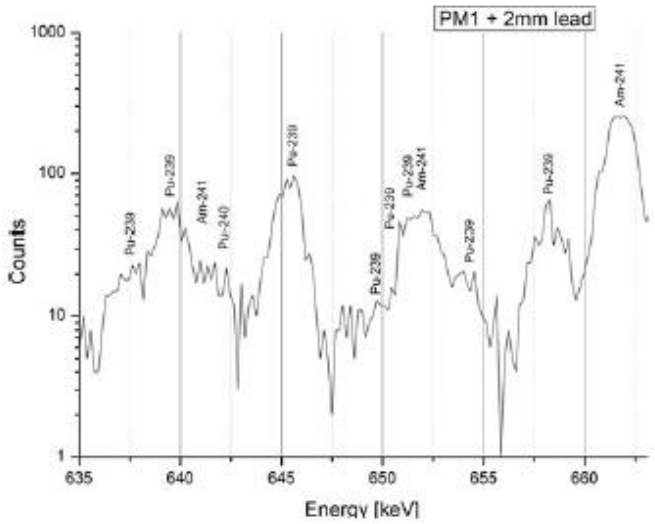
[3] The preamplifier energy rate for a planar detector is a trade off with low-rate resolution. This specification assures that the preamplifier will allow a maximum counting rate above 50 kcps for a typical plutonium spectrum. A more stringent low rate resolution specification may be achieved at the expense of the maximum preamplifier count rate.

(Table courtesy of Los Alamos National Laboratory)

Because in general no information on the presence of shielding will be available, coaxial detectors should be preferred; these show higher efficiencies at medium to high gamma-ray energies, which are less attenuated by shielding. This is illustrated by Figure 1 below, showing count rates above 600 keV at a measurement time of 2,000 seconds for a 12.5 g Pu sample shielded by 2 mm Pb.[4]

Figure 1. Count Rates and Measurement Time for Plutonium Sample

[4] M. Göttsche and G. Kirchner, "Measurement Techniques for Warhead Authentication with Attributes: Advantages and Limitations," *Science & Global Security*, 22 (2014): 83–110.



Detector efficiencies are a function of the gamma energy and depend on mass, size, density of the nuclear material, its distance from the detector, and geometry, mass, and elemental composition of any material between source and detector. Information on many of these properties will not be available.

If count rates of peaks of different isotopes at adjacent energies are used for estimating isotopic ratios of the nuclear materials, knowledge of detector efficiencies is not required. This approach, however, is limited to Pu, because U does not show adjacent gamma decay energies. For mass determination this approach needs to be combined with a second technology, e.g., passive neutron multiplicity counting.

Mass determination using gamma spectrometry requires knowledge of detector efficiencies. Software tools such as FRAM[5] calculate these by evaluating various peaks including the full energy spectrum of gamma disintegrations. They provide estimates of masses and isotopic compositions of U, Pu, or U and Pu. However, robustness, specificity, and completeness of evaluations using such complex codes still need to be assessed. Integration of complex software into an information barrier may become an issue.

According to Archer, Luke, and Parker, material age can be inferred from ^{241}Am decay produced from the decay of ^{241}Pu by using HPGe in combination with the ^{300}Pu analysis code.[6]

This is based on the detection of 332.4 and 335.4 keV gamma lines.

Age can be determined from other regions but the 300 keV region is the most promising region for transparency measurements of age, if the plutonium is stored in a thick container.

[5] Sampson, "Chapter 2, Passive Nondestructive Assay of Nuclear Materials 2007 Addendum"; and Sampson, Kelley, and Vo, *Application Guide to Gamma-Ray Isotopic Analysis Using the FRAM Software*.
[6] D.E. Archer, S. John Luke, and W. Parker, "Pu300: A Tool for Measurement of Plutonium Age for Arms Control Transparency via Gamma-Ray Spectroscopy," UCRL-JC-136626, *Fifth International Conference on Methods and Applications of Radioanalytical Chemistry—Mark V* (Kailua-Kona, Hawaii, 2000), available at https://e-reports-ext.llnl.gov/pdf/237565.pdf.

Time Constraints (e.g., measurement times including distance from object, time to install the equipment):

Maximum one hour for the setup.

Measurement time to measure 500 g of Pu (0.1 ^{239}Pu/^{240}Pu) or 500 g of ^{235}U at 1 m from the surface of the container (order of magnitude: seconds, minutes, hours, days):

For achieving adequate count rates measurement times should be not less than 30 minutes. Their actual choice depends on detector size, source-detector distance, and mass of the nuclear material.

Will this method work in the presence of shielding? If so, what is the maximum amount of shielding that will still allow the method to work?

According to Sampson as well as Göttsche and Kirchner, shielding of up to 25 mm of Pb and 25 mm of steel does not become prohibitive.[7] This has been shown also for shielding by a container used for storage of Pu.

The 186 keV line of ^{235}U is easily shielded; its intensity is attenuated by a factor 50 through 3 mm Pb. Therefore, it is not recommended to be used for HEU within the framework of IPNDV verifications.

Technology Complexity (e.g., hardware, software, and ease of use by personnel):

Both for hardware and software, personnel with experience in nuclear nondestructive assay (NDA) measurements, e.g., nuclear inspector could carry out measurements and interpret data.

Infrastructure Requirements (e.g., electrical, liquid nitrogen, etc.):

Liquid nitrogen provides the best energy resolution (performances); mechanical cooling is an option to be considered.

Power supply

Technology Limitations/Variations (e.g., detection limits for nuclear material, operational temperature range, differences in technology detector materials):

Gamma-ray spectroscopy typically relies on the assumption of samples with uniform distribution of radionuclides within the sample. It is expected that corrections are possible if the distribution of the radionuclides is known.

The deployment of Medium Resolution Gamma-Ray Spectrometry (MRGS) and associated spectral analysis is being explored by using Cadmium Zinc Telluride and Lanthanum Bromide detectors. MRGS has no issues related to cooling the detector because it operates at room temperature.

Information Collected by the Technology (used to help determine if an information barrier is required for use):

Isotopic composition, possibly information on mass

[7] Sampson, "Chapter 2, Passive Nondestructive Assay of Nuclear Materials 2007 Addendum"; and Göttsche and Kirchner, "Measurement Techniques for Warhead Authentication with Attributes: Advantages and Limitations."

Safety, Security, Deployment Concerns:

Working with high voltage, liquid nitrogen

Technology Development Stage (Technology Readiness Level, TRL):

Commonly used. TRL 8–9

Additional System Functionality (e.g., outside the monitoring use case):

Any detection of gamma emitting radioactive material

Where/How the Technology Is Currently Used (e.g., international safeguards, border protection):

International safeguards

Examples of Equipment:

BEGE, http://www.canberra.com/products/detectors/pdf/BEGe-SS-C49318.pdf

FRAM, http://www.canberra.com/products/waste_safeguard_systems/pdf/FRAM-SS-C37207.pdf.

Further References:

T. E. Sampson, T. L. Cremers, and W.J. Hansen, "Nondestructive Assay System for the ARIES Weapons Component Dismantlement System," *American Nuclear Society Third Topical Meeting on DOE Spent Fuel and Fissile Materials Management* (Charleston, SC, September 1998).

P. A. Hypes, "Plutonium Source Isotopic Analysis with up to 25 mm Pb Shielding Using the FRAM Isotopic Analysis Code," LA-UR-00-2511, *Proceedings of the Institute of Nuclear Material Management, 41st Annual Meeting* (New Orleans, Louisiana, July 16–20, 2000), available at http://permalink.lanl.gov/object/tr?what=info:lanl-repo/lareport/LA-UR-00-2511.