

NUCLEAR FORENSICS INTERNATIONAL TECHNICAL WORKING GROUP

ITWG GUIDELINE ON HIGH-RESOLUTION GAMMA SPECTROMETRY



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ITWG Guidelines are intended as consensus-driven best-practices documents. These documents are general rather than prescriptive, and they are not intended to replace any specific laboratory operating procedures.

1. INTRODUCTION

Gamma spectrometry is a non-destructive technique used to identify and quantify gammaemitting radionuclides present in a given sample. Gamma spectrometry takes advantage of the characteristic gamma-ray energies emitted by each radionuclide and the proportionality between the intensity of the emitted radiation and the activity of the radionuclide in the sample being measured. A gamma spectrum displays the number of detected gamma photons as a function of energy. Figures 1a and 1b show typical gamma spectra of uranium and plutonium (spectra available from the Nucleonica web portal).







Fig. 1b. Gamma spectrum of plutonium with 84% ²³⁹Pu.

A typical state-of-the-art high-resolution gamma spectrometry system (Figure 2) consists of a detector crystal made of high-purity germanium (HPGe), cooled either by liquid nitrogen or by an electrical cooler, with a preamplifier, cooling system, electronics unit(s), and a computer for displaying, saving, and analysing gamma spectra. The electronics units are often integrated into one compact device. In fact, portable systems often have all components integrated into a single hand-held instrument. For specific applications in the laboratory, low-background shielding for the detectors is often used in order to isolate the detector from natural background radiation and interferences from other radioactive materials.

In order to collect a gamma spectrum, one places the item under investigation in front of the detector in a configuration specific to the detector orientation and certain sample parameters. Gamma radiation emitted from the sample enters the detector and produces electronic signals that are automatically counted and sorted into discrete channels according to their energy by the electronics and software. These data are typically displayed as a histogram of the number of detected signals (photons) in each channel. Converting the x scale (abscissa) of this histogram from channel number to photon energy yields the acquired gamma spectrum (as displayed in Figures 1a and 1b). The longer the sample is measured, the more photons are counted and summed into the histogram, which results in smaller statistical uncertainties for the measured intensity. Therefore, the longer the counting time, the more reliable the histogram of the counts (i.e. the recorded gamma spectrum).

The time needed to accumulate a reliable gamma spectrum is variable and depends on properties of the investigated item (e.g., its level of radioactivity, its density), the specifications of the detector (e.g., its efficiency), the measurement configuration, and the information we want to extract from the gamma spectrum (nuclide identification versus



Fig. 2. Components of a typical gamma-spectrometric setup.

quantification). For most nuclear forensic scenarios and sample types, the counting time required for on-site categorization (see Section 2 below, as well as the "ITWG guidelines for establishing core capabilities in nuclear forensics" [1]) is on the order of tens of minutes, while the time required to collect spectra for a detailed nuclear forensic analysis in the laboratory is on the order of hours to days.

An important figure-of-merit for gamma spectrometers is their efficiency. The efficiency of a gamma spectrometer is the ratio of the number detected gamma photons to the number of photons incident on the detector. The higher the efficiency, the shorter the time required to collect a representative spectrum of the investigated sample.

Another important figure-of-merit is the energy resolution of the spectrometer. The energy resolution measures the ability of a spectrometer to distinguish between photons of slightly different energy. It is usually specified as the "Full Width at Half Maximum" (FWHM) of a gamma peak at a predefined energy, for example the 1332 keV peak of 60Co. The smaller the FWHM, the higher the resolution and the better adjacent peaks can be resolved in the spectrum. The difference between low-resolution and high-resolution gamma spectra is illustrated in Figure 3. This Guideline is focused on high-resolution gamma spectrometry, because high-resolution is required in order to investigate the parameters relevant to nuclear forensics (see Table 1, below). It should be mentioned, however, that low-resolution gamma spectrometers can also be used, to a limited extent, to identify radioactive sources and to detect the presence of nuclear material.

Unfortunately, the resolution of a gamma spectrometer decreases with increasing efficiency.

Therefore, the choice of the appropriate gamma spectrometer is always a compromise between efficiency and resolution.



Fig. 5. Spectra of low-enriched uranium taken with a lowresolution Nal detector (red) and with a high-resolution HPGe detector (black).

2. USE FOR NUCLEAR FORENSICS

Nuclear forensic analysis is the process by which intercepted illicit nuclear and radiological materials and other associated materials, such as containers, are analyzed to provide clues to attribution. Nuclear forensic analysis typically consists of categorization of the material to address the threat posed by that material, followed by a more detailed characterization of the material to determine the exact nature of the radioactive material and associated evidence (see reference [1]). Following these analyses, interpretation of the results is carried out to assess indicators of the origin of the illicit material.

Gamma spectrometry plays a dual role in the field of nuclear forensics. In the location where illicit nuclear material is first detected, gamma spectrometry is a useful tool for the fast in-field

| Parameter | Comment | |
|--|---|--|
| $^{\rm 234}$ U, $^{\rm 235}$ U and $^{\rm 238}$ U content of U | Usually no need for low-background shielding | |
| 238 Pu, 239 Pu, 240 Pu, 241 Pu and 241 Am content of Pu | Usually no need for low-background shielding | |
| Total U and Pu content | | |
| Age of uranium | Low-background shielding recommended | |
| Age of plutonium | | |
| ²³² U content | ²³² U indicates presence of reprocessed uranium | |
| Presence of fission products | Indicates previous irradiation or contamination | |
| Type and activity of other radioactive materials | Materials in a Radiological Dispersal Device (RDD) or contamination of the sample, for example | |
| Presence of alpha- or neutron- induced gamma-ray signatures | Indicate the presence of non-radioactive construction materials in contact with or surrounding the nuclear material | |

Table 1. Parameters investigated by high-resolution gamma spectrometry relevant to nuclear forensics.

identification and categorization of radiological or nuclear material. In this application, the goal is to quickly identify the material and obtain a rough idea about its isotopic composition. For more comprehensive characterization of the material, gamma spectrometry is also used in the laboratory as a non-destructive analysis technique. The goal in this application is to more accurately determine the isotopic composition of the material. In addition, for special nuclear material, such as U or Pu, gamma spectrometry may also be used to determine the total nuclear material content, the age of the material, the presence of reprocessed uranium, and the presence of fission products. The various parameters that can be quantified with gamma spectrometry are outlined in Table 1. Both of these applications are further described in the ITWG Guidelines: "In-field applications of high-resolution gamma spectrometry for the analysis of special nuclear material" and "Laboratory applications of high-resolution gamma spectrometry for the analysis of special nuclear material" [2, 3].

3. SAMPLE REQUIREMENTS

Gamma spectrometry is a non-destructive method. Thus, the technique can be applied to a wide range of sample types and configurations. Investigated items can usually be analyzed as they are received without any sample preparation. Isotope-ratio and age determination measurements do not depend on the sample shape, physical form and chemical composition, nor do they require the use of reference samples. When quantification of radioactive material content is the goal of the analysis, measurement configuration becomes important and comparison to a reference sample similar to the investigated item or the use of modelling techniques is required.

Critical sample properties which might influence a gamma spectrometric measurement are the following:

- Activity of the sample: In the analysis of highactivity samples, if dead-time reduction cannot be achieved by appropriate shielding or by increasing the source-to-detector distance, sample splitting may be required to reduce detector dead-time (the time required for the detector and electronics to accurately count two or more photons arriving closely spaced in time).
- Sample shielding: If there is too much shielding (e.g. ~5 cm of lead) between the detector and the item to be analyzed, then only limited information can be extracted by passive gamma spectrometry.
- Presence of other radioactive materials whose gamma radiation might interfere with the radiation from the investigated material ("masking effect").

For samples of nuclear material the following additional properties are important:

- Nuclear-material content: approximately 0.5 g of nuclear material is typically needed in order to obtain meaningful results for the isotopic composition within a reasonable counting time (i.e. several hours).
- Pu isotopic composition: The Pu isotopic composition is often evaluated using commercially available codes which automatically calculate the Pu isotopic composition from the gamma spectrum. Different codes and different versions of the same code perform differently with the same spectra. In particular, high burn-up Pu, with its increased levels of ²⁴²Pu and correspondingly reduced levels of ²³⁹Pu, represents a severe challenge for these codes. Results showing a ²³⁹Pu isotopic weight fraction of less than 60% should be treated with some caution. The commercially available codes sometimes completely fail to report a result for such an isotopic composition or even report incorrect results. This is especially true for older versions of the codes and for spectra taken in sub-optimal conditions. In such cases, alternative approaches are needed. In order to obtain meaningful results, sometimes it is sufficient to increase the counting time (i.e. improve the counting statistics), but sometimes one either has to use the advanced features of the latest versions of the codes or to revert to manual calculations.
- Neutron radiation: Pu was often used in the past for the production of Pu-Be sources, which are strong neutron emitters. Neutron radiation is a concern because of three main reasons:
 - Radiation hazard to the persons performing the measurement.
 - Possible radiation damage of the HPGe detector.
 - The spectral background continuum due to gamma radiation induced by interactions

of neutrons with the HPGe crystal interferes with the evaluation of the gamma peaks from the spectra. In some cases (e.g. for Pu-Be sources made using high burn-up plutonium) the interference can be quite significant, causing the commercial codes for automatically evaluating the Pu isotopic composition to fail.

4. PRO'S AND CON'S OF THE TECHNIQUE (GENERAL)

Pro's:

- Fully non-destructive method, preserving forensic evidence.
- Item analysis without dismantling the item.
- Minimal or no sample preparation requirements.
- Portability.
- Fast results available in the field.
- Relatively inexpensive equipment compared to destructive techniques. The operating costs are also low. A high-resolution (HPGe-based) gamma spectrometry system costs typically between 60,000 – 100,000 Euro, depending on detector type and additional components.

Con's:

- For high-resolution gamma spectrometers, detector cooling is needed: liquid nitrogen or electrical cooling.
- The precision is often less than the precision of destructive techniques (the standard deviation for a given measurement result using highresolution gamma spectrometry will typically be higher).
- With present-day commercial gamma spectrometers it is not possible to directly derive information on ²⁴²Pu from the measured spectrum. Isotopic correlations have to be used to estimate the ²⁴²Pu content of the investigated

items. For Pu with high ²⁴²Pu content, this causes a large uncertainty in determining the Pu isotopic composition. Note, however, that by combining gamma spectrometry with two other non-destructive methods, calorimetry and neutron coincidence counting, it is possible to derive the ²⁴²Pu content in a fully nondestructive way.

 Due to on-site limitations (e.g. relatively short spectrum collection times) and due to the inability to fully control the measurement conditions on-site, the accuracy and precision of in-field measurements is typically lower than that in a laboratory setting.

5. FAQ – COST, DO'S AND DON'T'S, COMMON MISTAKES

(See Refs. [2] and [3].)

6. REFERENCES

- 1. ITWG Guideline: "Establishing core capabilities in nuclear forensics."
- 2. ITWG Guideline: "In-field applications of highresolution gamma spectrometry for the analysis of special nuclear material."
- 3. ITWG Guideline: "Laboratory applications of high-resolution gamma spectrometry for the analysis of special nuclear material."
- Web-sites of equipment manufacturers, e.g.: a. http://www.ortec-online.com/Library/index. aspx,

b.http://www.canberra.com/literature/,

c.and others.

 D. Reilly, N. Ensslin, H. Smith, Jr. and S. Kreiner (editors): Passive Nondestructive Assay of Nuclear Materials, Office of Nuclear Regulatory Research, U.S. Nuclear Regulatory Commission, NUREG/CR-5550, LA-UR-90-732, March 1991. Also available online at http://www.lanl.gov/orgs/n/ n1/panda/.

 The Nucleonica portal: www.nucleonica. net – see, e.g., Gamma Spectrum Generator and Nuclear Data Retrieval (free registration required).

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