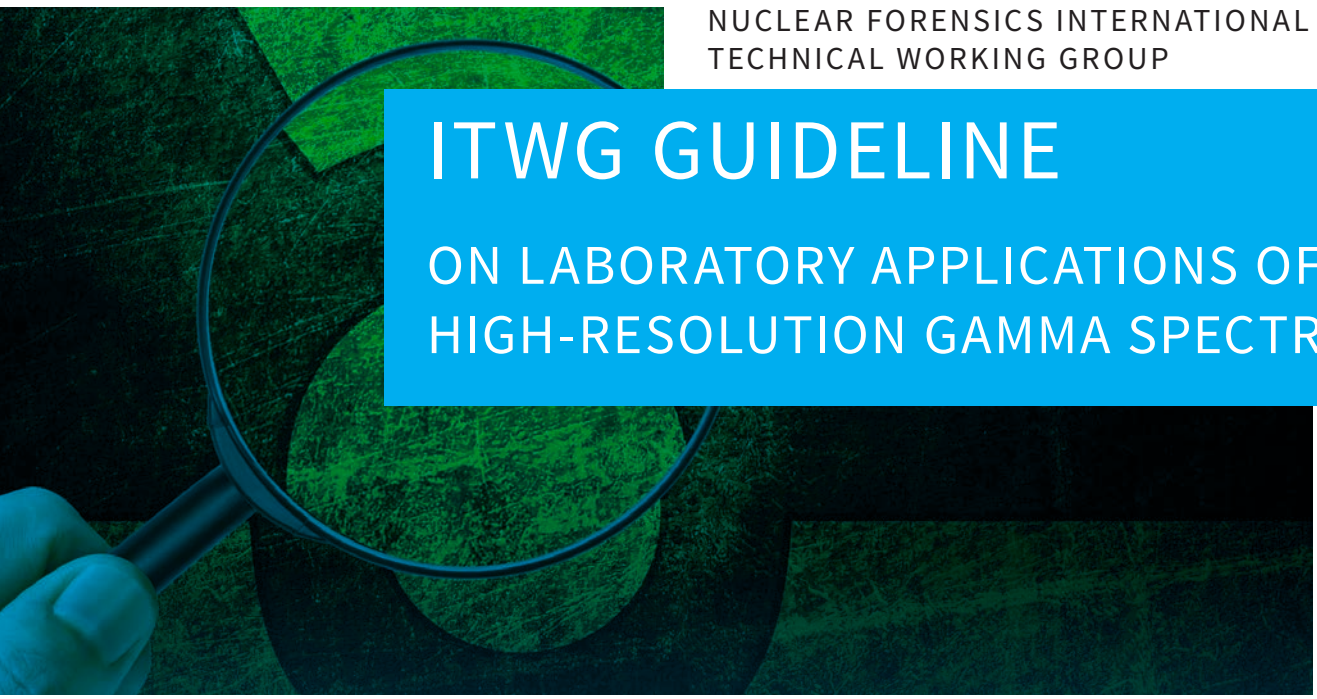
NUCLEAR FORENSICS INTERNATIONAL
TECHNICAL WORKING GROUP

ITWG GUIDELINE

ON LABORATORY APPLICATIONS OF HIGH-RESOLUTION GAMMA SPECTROMETRY



This document was designed and printed at Lawrence Livermore National Laboratory in 2016 with the permission of the Nuclear Forensics International Technical Working Group (ITWG).

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

This document describes the use of gamma spectrometry in laboratory conditions as a non-destructive tool for the characterization of nuclear material. A general description of gamma spectrometry and its use for nuclear security and nuclear forensics applications is given in the INFL Guideline, "High resolution gamma spectrometry – General overview" [1]. A document on the in-field use of gamma spectrometry is also available in the ITWG Guideline, "In-field applications of high-resolution gamma spectrometry for analysis of special nuclear material" [2].

Laboratory applications of gamma spectrometry for nuclear security and nuclear forensics purposes provide a means to accurately determine the isotopic composition of gamma-emitting nuclides in a sample and to quantify the amount of each radioisotope present. In the analysis of special nuclear material, such as uranium and plutonium, 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. This information can assist an investigation by providing indicators of the origin of the illicit material.

2. USE FOR NUCLEAR FORENSICS

Low-resolution and medium resolution detectors, based on, e.g., Cd(Zn)Te semiconductors or on scintillator crystals, such as NaI or $LaBr_3$, although satisfactory for some in-field applications, cannot provide the accuracy and precision needed for detailed laboratory investigations. High-resolution gamma spectrometers, with detectors made of high-purity germanium (HPGe), are required for laboratory use. Note, however, that even the performance of currently available, electrically cooled, compact portable detector systems is not satisfactory for detailed analyses in the laboratory. For laboratory investigations, only "classical" liquid-nitrogen cooled HPGe detectors may be used.

For each item which requires nuclear forensic characterization, two different types of gamma spectrometric measurements, using two different HPGe detectors, are performed in the laboratory. First, the isotopic composition of U and/or Pu, as well as the age of Pu, is determined by a gamma spectrometer having good energy resolution for the low energy range around 100 keV (usually "planar detectors" with resolution (FWHM) <600 eV at 122 keV). Additional information about the investigated material can be obtained from the higher energy range. Unfortunately, detectors with good energy resolution in the low-energy range have poor efficiency in the higher energy range, so high-energy spectra taken with these detectors are not suitable for analysis. Therefore, another measurement using a high-efficiency (coaxial) detector is also performed in order to obtain information about the age of the U, the presence of reprocessed U, and the presence of fission products. In addition, the high-energy spectrum can also confirm the Pu isotopic composition obtained from the low-energy spectrum.

Note that there are also commercially available "broad energy detectors" which offer relatively good resolution, combined with a relatively high efficiency, that can be used for both purposes, although with some trade-offs in performance. Such broad energy detectors can be a cost-effective solution, especially if the same detector is to be carried into the field. However, for best performance in the laboratory, two separate detectors are required, both optimized for specific purposes.

Because the effects to be measured with the high-efficiency detector in the high-energy range

are usually quite small, this detector must be shielded from natural background radiation, which might interfere with the evaluation of the data from the sample itself. For isotopic composition measurements with the low-energy detector, the natural background radiation is negligible, so there is usually no need for low-background shielding. However, in active laboratories, where many other nuclear samples are also handled, a shield for the low-energy detector may also be desirable.

3. SAMPLE REQUIREMENTS

Being a non-destructive method, gamma spectrometry can be applied to a wide range of sample types. In most cases, there are no special sample requirements for gamma spectrometry – often the investigated items can be analyzed as they are received without any sample preparation.

For isotope-ratio measurements, as well as for age determination, gamma-spectrometric methods do not depend on the sample shape, physical form, or chemical composition. The total content of nuclear material and the activity of other radioactive isotopes, however, can only be determined if the self-absorption within the specified shape can corrected, either by mathematical modelling or by calibration with reference materials similar to the investigated item.

For gamma spectrometric measurements, it is usually assumed that the samples are homogenous. However, one must keep in mind that, for very large items, the detected gamma radiation mainly derives from the outer regions of the item. Therefore, only limited information can be obtained about the interior of a large item.

4. PRO'S AND CON'S OF THE TECHNIQUE

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

How much does a laboratory gamma spectrometry system cost?

About 60,000 – 100,000 Euro, depending on detector type, additional components, and analysis software. In order to have the full range of gamma-spectrometric capabilities, two different systems, i.e. a planar and a coaxial detector, should be installed (which, in principle, could share the same set of electronics). The costs of low-background shielding and of liquid nitrogen supply are not included in the above estimate.

How long does a gamma-spectrometric analysis take?

The time required for a detailed nuclear forensic analysis in the laboratory by gamma spectrometry is about 1/2 day to 2 days.

What is the thickness of the shielding needed for low-background measurements?

Around 10 cm lead or 20 cm iron.

What is the expected measurement uncertainty associated with gamma spectrometry?

The relative uncertainty for typical items of interest for nuclear forensics (e.g., low-enriched uranium pellets) for ^{235}U enrichment is around 1% ("1 sigma") for a half-day measurement and around 5% for a 1-2 hour measurement. For low-burn-up plutonium (high ^{239}Pu and low ^{242}Pu content) the relative uncertainty of isotope ratios is close to 1% for a 1 day measurement, while for high-burn-up plutonium (low ^{239}Pu and high ^{242}Pu content) it depends on the exact isotopic composition and ranges from a few percent to few tens of percent. It should be emphasized, however, that the above uncertainties are typical values, but for specific samples the precision can be either much better or much worse.

What are typical mistakes when doing gamma spectrometry?

A typical mistake is the acceptance of the results of an off-the-shelf gamma spectrum analysis code, without getting acquainted with the details of the algorithm. For example, it is important to know how the program handles cases in which secular equilibrium has not been reached.

6. REFERENCES

1. ITWG Guideline: "High-resolution gamma spectrometry – General overview."
2. ITWG Guideline: "In-field applications of high-resolution gamma spectrometry for the analysis of special nuclear material."
3. D. Reilly, N. Ensslin, H. Smith (eds.), Passive non-destructive assay of nuclear materials, NUREG/CR-5550, LA-UR-90-732. Also available online as "Passive Nondestructive Assay Manual – PANDA": www.lanl.gov/orgs/n/n1/panda/.
4. A. N. Berlizov, R. Gunnink, J. Zsigrai, C. T. Nguyen and V.V. Tryshyn, Performance testing of the upgraded uranium isotopics multi-group analysis code MGAU. Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment, 575, 498-506.

5. Nguyen, C. T. and Zsigrai, J. (2006). Gamma-spectrometric uranium age-dating using intrinsic efficiency calibration. Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms, 243, 187-192. (Preprint also available online: http://arxiv.org/abs/nucl-ex/0506029.)

7.0 CONTRIBUTORS

This document was prepared on the basis of contributions from:

J. Zsigrai
Institute for Transuranium Elements
European Commission

C. Larsson
Defense R&D Canada
Canada

N. Cong Tam
IKI
Hungary

J. Safar
Hungarian Atomic Energy Agency
Hungary

M. Wallenius
Institute for Transuranium Elements
European Commission

DOCUMENT REVISION HISTORY

Document INFL-GSLA			
Version No.	Version Date	Description of Changes	Changes made by
1	March 2013	Initial Draft	J. Zsigrai et al. (authors)