

TABLE 6b – Plutonium Isotopic Abundances (NDA)

Method/measurement system (including software): HRGS / XMCG / FRAM-MGA

Application: Measurement of Plutonium Isotopic Composition in Pure Plutonium Samples (Low-energy analysis)

Short Description of the method/measurement system and typical measurement conditions (shielding, measurement time, etc.)
The method determines plutonium isotopic composition by analyzing high resolution gamma spectra acquired with a planar or small semi-planar High-Purity Germanium (HPGe) detector having the energy resolution FWHM < 750 eV @ 122 keV. The method employs the low-energy region (59-230 keV) analysis of the isotopic analysis software codes FRAM v.6.1 (Fixed-Energy Response-Function with Multiple Efficiency [1]) and MGA v.10.0 (Multi Group Analysis Code for Plutonium [2]). The method can be used to measure plutonium items of arbitrary size, shape, and composition. It is applicable to any type of plutonium (aged, fresh, high- and low-burnup samples) and is useful for in-situ measurements of bulk materials with net weight of tens of mg up to several kg. A typical measurement system includes a collimated HPGe detector, a Multi-Channel Analyzer (MCA), and an instrumentation laptop. The MCA is typically set to acquire spectra with 4096 channels and detector gain adjusted to ~0.075 keV/channel. A 1-2 mm cadmium or tin filter is normally used to suppress the 59.5 keV gamma line of ²⁴¹Am to the extent when its amplitude is at about the same level as the intense X-ray peaks in the 100 keV region. Typical measurement time is 300-1800 s, although the presented data were normalized to a live time (LT) of 600 s. The LT of 600 s allows for ~50,000 counts in the reference 129 keV peak of ²³⁹Pu to be acquired with a high-burnup plutonium sample at output count rate (OCR) of ~22,000 cps.
Factors affecting the method performance
High count rate applications, i.e. with DT > 40% (or OCR > 22,000 cps), may broaden the peak energy resolution, alter the peak shapes, and also lead to pulse pileup. This will affect the accuracy and precision of the measurement results. The method generally provides good results when measuring plutonium in a thin wall container (< 3 mm SS (or equivalent)), which does not significantly attenuate the intense X-ray region around 100 keV. Moderately thick and/or heavy absorber (> 10 mm SS (or equivalent)) will significantly suppress the X-ray peaks in the 100-keV region, which can effectively render the technique useless. The ITVs were derived assuming homogeneous samples. The material inhomogeneity, as well as heavy shielding, high ambient radiation background (including presence of fission product emissions, e.g., at nuclear power plants), presence of high-Z elements in material matrix with X-ray emissions close to the Pu X-rays (e.g., such as Th), would worsen the performance of the method.

Source of bottom-up value (only for prospective ITV not based on historical data)

Current evaluations are based on the evaluation of a number of plutonium spectra available at Los Alamos National Laboratory (LANL) and IAEA's Safeguards Analytical Laboratory (IAEA SAL).

LANL analyzed its plutonium data using FRAM v.6.1 with the parameter set GePInr_Pu_060-230. The spectra were measured in lab environments with counting time from 900 to 5400 s and the results were normalized to a counting time of 600 s. The input count rates for the data varied from about several thousand cps up to 50,000 cps. The filters were 0.8-1.6mm cadmium. 67 data sets with 1117 spectra were used in the uncertainty evaluation.

IAEA analyzed its plutonium data using FRAM v.6.1 with the parameter set GePInr_Pu_060-230, and MGA v.10.0. This data included the collection of 111 high-burnup plutonium samples measured over several years in a laboratory environment at IAEA SAL, and 29 good statistics spectra of low-burnup plutonium extracted from the international database. All data were normalized to a LT=600s.

Method references

- [1] D. Vo, "Improvements of FRAM version 6.1," International Workshop on U/Pu Isotopic Analysis by Nondestructive Assay Techniques for Nuclear Safeguards, IAEA, February 2021, Journal of Nuclear Materials Management 2021 Volume XLIX No. 3: 19-30.
- [2] R. Gunnink, MGA: A Gamma-Ray Spectrum Analysis Code for Determining Plutonium Isotopic Abundances, Vol. I, LLNL, Ca. UCRL-LR-103220, 1990.

Prepared by: Duc Vo & Andrey Bosko

Date: 2022-02-16

Method/measurement system (including software): HRGS / XMCG / FRAM-MGA

Application: Measurement of Plutonium Isotopic Abundances in MOX (Low-Energy Analysis Option)

Short Description of the method/measurement system and typical measurement conditions (shielding, measurement time, etc.)
The approach is based on the peak-ratio methods implemented in such isotopic analysis codes as FRAM v.6.1 (Fixed-Energy Response-Function with Multiple Efficiency [1]) and MGA v.10.0 (Multi Group Analysis Code for Plutonium [2]). The codes employ intrinsic energy, peak shape and detection efficiency calibrations to derive isotopic abundances from the efficiency corrected full energy peak ratios extracted from a single high-resolution gamma-ray spectrum. A typical measurement system includes a collimated HPGe detector of a planar type with energy resolution FWHM ≤ 750 eV @ 122 keV, a digital MCA, and an instrumentation laptop. A 1-2 mm cadmium or tin filter is used to effectively suppress the intense 59.5 keV gamma-rays of ^{241}Am. The measurements are normally performed at high count rates (5-20 kcps), to allow sufficient statistics of peak counts within practically feasible measurement time. For a standard IAEA XMCG setup involving a collimated HPGe Model BE1015 (Mirion Technologies) with crystal dimensions - $\varnothing 36 \times 15$ mm and collimator opening - $\varnothing 35 \times 15$ mm, the typical 30,000 counts in the reference 129 keV peak of ^{239}Pu from a lightly shielded material can be normally acquired at OCR=10 kcps within the measurement live time of LT=600 s. To allow good spectrum quality, the dead times are typically kept below 40%.
Factors affecting the method performance
High count rate applications, i.e. with DT > 40% (or OCR > 22,000 cps), may broaden the peak energy resolution, alter the peak shapes, and also lead to pulse pileup. This will affect the accuracy and precision of the measurement results. The method generally provides good results when measuring plutonium in a thin wall container (< 3 mm SS (or equivalent)), which does not significantly attenuate the intense X-ray region around 100 keV. Moderately thick and/or heavy absorber (> 10 mm SS (or equivalent)) will significantly suppress the X-ray peaks in the 100-keV region, which can effectively render the technique useless. The ITVs were derived assuming homogeneous samples. The material inhomogeneity, as well as heavy shielding, high ambient radiation background (including presence of fission product emissions, e.g., at nuclear power plants), presence of high-Z elements in material matrix with X-ray emissions close to the Pu X-rays (e.g., such as Th), would worsen the performance of the method.
Source of bottom-up value (<u>only for prospective ITV not based on historical data</u>)
Current evaluations are based on limited scope inspector vs. operator paired dataset consisting of ~50 field MOX measurements, obtained with FRAM 6.1 and parameter set "Pu_Plnr_060-230". The evaluations included MOX materials made of naturally enriched Uranium and high-burnup Plutonium in the range of the U/Pu ratios from 10 to 100.

Method references

- [1] T.E. Sampson, T.A. Kelley, PC/FRAM: a Code for the Nondestructive Measurement of the Isotopic Composition of Actinides for Safeguards Applications, Appl. Radiat. Isot. Vol. 48, No. 10-12 (1997) 1543-1548.
- [2] R. Gunnink, MGA: A Gamma-Ray Spectrum Analysis Code for Determining Plutonium Isotopic Abundances, Vol. I, LLNL, Ca. UCRL-LR-103220, 1990.

Prepared by: A. Berlizov & V. Nizhnik (IAEA SGTVT)

Date: 2022-02-07

Method/measurement system (including software): HRGS / XMCG / FRAM

Application: Measurement of Plutonium Isotopic Compositions (High-energy analysis)

Short Description of the method/measurement system and typical measurement conditions (shielding, measurement time, etc.)

The method determines plutonium isotopic composition by analyzing spectra acquired by a coaxial or large semi-planar HPGe detector with the energy resolution FWHM < 1 keV @ 122 keV and < 2 keV @ 1332 keV. The method employs the high-energy region (180-1001-keV) analysis of the isotopic analysis software code FRAM v.6.1 (Fixed-Energy Response-Function with Multiple Efficiency [1]). The method can be used to measure items of arbitrary size, shape and composition. It is applicable to any type of plutonium (aged, fresh, isotopic homogeneous/heterogeneous, wide range isotopic compositions) and is useful for in-situ measurements of bulk materials with net weight of a fraction of a gram up to several kg. A typical measurement system includes a collimated HPGe detector, an MCA, and an instrumentation laptop.

The spectra are normally measured at 8K channels and with energy calibration of 0.125 keV/ch. Typical measurement time is 600-1800 s. A 1-2 mm cadmium or tin filter is used to suppress the 60-keV gamma rays of ²⁴¹Am. The method can analyze spectra of bare plutonium or plutonium that is shielded by up to 0.5 cm lead or 5 cm steel. For plutonium shielded by even thicker absorber up to 2.5 cm lead, the peaks below 300 keV may not be visible and the analysis should be modified to analyze the 300-1001-keV region.

Factors affecting the method performance

The random sum peaks of a source with significant Np-237 and measured at high count rate can interfere with the peak fitting of the 640-keV region. This will affect the accuracy of the measured area of the 642-keV peak of ²⁴⁰Pu. The inaccurate ²⁴⁰Pu fraction would affect the accuracy of all other isotopes.

The ITVs were derived assuming homogeneous samples. The material inhomogeneity, as well as heavy shielding, high ambient radiation background (including presence of fission product emissions, e.g., at nuclear power plants) would worsen the performance of the method.

Source of bottom-up value (only for prospective ITV not based on historical data)

The ITV values are from the analysis of lab data using FRAM in the high-energy mode. The spectra were measured with counting times from 900-7200 s and the ITV results were normalized to a counting time of 600 s. The counting rates for the data varied from about several kHz up to 50 kHz. In addition to 1-2mm cadmium filter, up to 1.6 mm lead were also used for some data sets. Sixty-eight data sets with 1075 spectra were used in the analysis.

Method references

[1] D. Vo, "Improvements of FRAM version 6.1," International Workshop on U/Pu Isotopic Analysis by Nondestructive Assay Techniques for Nuclear Safeguards, IAEA, February 2021, Journal of Nuclear Materials Management 2021 Volume XLIX No. 3: 19-30.

Prepared by: Duc Vo
Date: 2022-02-16

Method/measurement system (including software): HRGS / XMCG / FRAM

Application: Measurement of Plutonium Isotopic Abundances in MOX (High-Energy Analysis Option)

Short Description of the method/measurement system and typical measurement conditions (shielding, measurement time, etc.)

Typical measurement system includes a portable gamma-ray spectrometer with electrically cooled coaxial HPGe crystal and a digital MCA. The spectrometer's energy resolution is $\text{FWHM} \leq 1.5 \text{ eV @ } 122 \text{ keV}$. The IAEA instrument (ECGS - Electrically Cooled Gamma Spectrometer) is represented by the ORTEC Micro-UF6™ system [1], a variant of the standard ORTEC Micro-trans-SPEC portable gamma-spectrometer. The system includes a coaxial HPGe crystal with $\text{Ø}50 \text{ mm} \times 30 \text{ mm}$ nominal dimensions, furnished with the 8 mm thick side shielding and a $\text{Ø}50 \text{ mm} \times 8 \text{ mm}$ front-end tungsten collimator. For materials containing high-burnup Pu, a 1-2 mm Cd or Sn filter is used to suppress the intense 59.5 keV gamma-rays of ^{241}Am .

The spectrometer is set up to acquire an 8k-channel spectrum with the 0.125 keV/chn gain. Measurements are performed at $\leq 20 \text{ kcps}$ count rates and dead times $\leq 30\%$, to allow good statistics of counts and sufficient spectrum quality within practically achievable measurement times ($\text{LT} \leq 1000 \text{ s}$). The spectrum evaluation approach is based on the peak-ratio method implemented in FRAM 6.1 (Fixed-Energy Response-Function with Multiple Efficiency [2]) isotopic analysis code. The code employs the intrinsic energy, peak shape and detection efficiency calibrations, to derive isotopic abundances from the efficiency corrected full energy peak ratios, which are extracted from a single high-resolution gamma-ray spectrum.

Factors affecting the method performance

The ITVs were derived assuming isotopically homogeneous samples. The material inhomogeneity, heavy shielding, and high ambient radiation background (including presence of fission product emissions) would worsen performance of the method. The Detective (family) detectors use a special shaping model, about 3 times wider than normal, to process the pulses from the detector crystal. As a result, the dead time can become significantly large for moderate count rate (50% dead time for 20 kHz input rate).

The normally large dead time even at low or moderate input rate can greatly increase the sum peaks, broaden the peaks, alter the peak shape, and increase the biases of the results. If the energy resolution of the detector does not meet the required resolution then the bias is likely increased.

Source of bottom-up value (only for prospective ITV not based on historical data)

The ITVs were evaluated based on the top-down approach applied to an inspector vs. operator paired dataset consisting of ~ 25 field verification measurements of MOX fuel assemblies using ECGS. The gamma-ray spectra were acquired for 1000 s live time. They typically contained $\sim 5,000 \text{ cnts}$ in the 129 keV peak of ^{239}Pu and $\sim 700,000 \text{ cnts}$ in the 208 keV peak of ^{241}Pu . The measured MOX fuel was made of depleted uranium and high-burnup plutonium ($^{239}\text{Pu} \leq 55 \text{ wt\%}$, 2-3 years after chemical separation) with U/Pu ratio ≈ 10 . The spectrum processing was performed with FRAM 6.1 using a modified version of the FRAM parameter set "det_coax_120_800_1". The modified parameter set included the interfering background peaks of Cs-137 (662 keV) and Ag-110m (658 keV) that impacted evaluations of the ^{240}Pu and ^{241}Am contents.

Method references

- [1] ORTEC Micro-UF6™ Portable UF6 Cylinder Verification System, Software v.2.3 — User's Manual, ORTEC Part No. 932500, 177 p.
- [2] D. Vo, Improvements of FRAM v.6.1, Journal of Nuclear Material Management, 49 (3) (2021) 19-30.

Prepared by: A. Berlizov (IAEA SGTVT)

Date: 2022-02-08

Method/measurement system (including software): HRGS / XMCG / FRAM

Application: Measurement of Plutonium Isotopic Compositions using the Ortec Detective Family (medium- and high-energy analysis)

Short Description of the method/measurement system and typical measurement conditions (shielding, measurement time, etc.)

The method determines plutonium isotopic composition by analyzing spectra acquired by a very large coaxial or the electrical cooled HPGe detector made by Ortec (Detective family) with the energy resolution FWHM < 1.5 keV @ 122 keV and < 2.3 keV @ 1332 keV. The method employs the mid- and high-energy regions (120-460-keV and 180-1001-keV) analysis of the isotopic analysis software code FRAM v.6.1 (Fixed-Energy Response-Function with Multiple Efficiency [1]). The method can be used to measure items of arbitrary size, shape and composition. It is applicable to any type of plutonium (aged, fresh, isotopic homogeneous/heterogeneous, wide range isotopic compositions) and is useful for in-situ measurements of bulk materials with net weight of a fraction of a gram up to several kg. A typical measurement system includes an instrumentation laptop together with the Detective/Transpec or an LN2 cooled HPGe detector connected to an MCA.

Depending on the detector system, the spectra are measured at either 8K channels with 0.125 keV/ch energy calibration (Transpec and LN2 cooled HPGe system), 8K channels with 0.366 keV/ch energy calibration (Detective), or 16K channels with 0.183 keV/ch energy calibration (Detective X). A 1-2 mm cadmium or tin filter is used to suppress the 60-keV gamma rays of ²⁴¹Am. The method can analyze spectra of bare plutonium or plutonium that is shielded by up to 0.5 cm lead or 5 cm steel. For plutonium shielded by even thicker absorber up to 2.5 cm lead, the peaks below 300 keV may not be visible and the analysis can be modified to analyze the 300-1001-keV region.

Factors affecting the method performance

The random sum peaks of a source with significant Np-237 and measured at high count rate can interfere with the peak fitting of the 640-keV region. This will affect the accuracy of the measured area of the 642-keV peak of ²⁴⁰Pu. The inaccurate ²⁴⁰Pu fraction would affect the accuracy of all other isotopes. The Detective (family) detectors use a special shaping model, about 3 times wider than normal, to process the pulses from the detector crystal. As a result, the dead time can become significantly large for moderate count rate (50% dead time for 20 kHz input rate). The normally large dead time even at low or moderate input rate can greatly increase the sum peaks, broaden the peaks, alter the peak shape, and increase the biases of the results. If the energy resolution of the detector does not meet the required resolution then the bias is likely increased.

Source of bottom-up value (only for prospective ITV not based on historical data)

The LANL spectra were lab data and measured with real counting times of 900 s and the ITV results were normalized to a live time of 600 s. The counting rates for the data varied from about 2kHz up to 20 kHz. In addition to 1-2mm cadmium filter, up to 8 mm steel were also used for some data sets. Twenty-eight data sets with 539 spectra were used in the analysis.

The JRC spectra were lab data measured using two sets of samples. Combinations of cadmium filters of up to 2 mm, steel filters of up to 16 mm, and a lead filter of 4 mm thickness were used (the lead filter was used without any other filter). There was a total of 1440 spectra, grouped into 40 categories for the ANOVA analysis. For the first set of samples, the count rate varied between about 2 kHz and 16 kHz, and spectra of 900 s true time were used for the analysis. For the second set, the count rate varied between about 0.2 kHz and 4kHz, and spectra of 7200 s true time were used. In this way, the total number of counts in the spectra was approximately between 1e6 and 1e7 counts for all samples. The full JRC dataset is available for download at <https://data.jrc.ec.europa.eu/dataset/d4df018b-7ad6-476c-bd6c-781c24bdc836>

Method references

- [1] D. Vo, "Improvements of FRAM version 6.1," International Workshop on U/Pu Isotopic Analysis by Nondestructive Assay Techniques for Nuclear Safeguards, IAEA, February 2021, Journal of Nuclear Materials Management 2021 Volume XLIX No. 3: 19-30.

Prepared by: Jozsef Zsigrai and Duc Vo

Date: 2022-02-17

Method/measurement system (including software): MRGS / XMCC-L / FRAM-PAT

Application: Determination of ^{239}Pu and ^{240}Pu Abundances Using Medium Resolution Gamma-Ray Spectrometers

Short Description of the method/measurement system and typical measurement conditions (shielding, measurement time, etc.)

Utilization of the MRGS for Plutonium isotopic measurements is an emerging application area in nuclear safeguards. It has been promoted recently as a result of the multilateral effort on U/Pu Isotopic Analysis Inter-Comparison Exercise by MRGS [1], which was coordinated by the IAEA. Two technologies identified as representing viable spectrum analysis options were subsequently implemented in the isotopic analysis codes – FRAM 6.1 (Fixed-Energy Response-Function with Multiple Efficiency [2]) and PAT (Plutonium Attribute Test [3]). Both codes contributed to the current evaluations of the prospective (bottom -up) ITVs for CZT and LaBr_3 based gamma-ray spectrometers. Both codes are based on the peak ratio method with application of iterative intrinsic calibration approaches for energy, peak shape and detection efficiency.

For technique 3 category, the FRAM code determines the ^{239}Pu , ^{240}Pu , ^{241}Pu , and ^{241}Am abundances from the spectrum analysis, while isotopic correlations are used to calculate the abundances of minor Pu isotopes – ^{238}Pu and ^{242}Pu . For technique 4 category, the FRAM code at first determines the $(^{241}\text{Pu} + ^{241}\text{Am})/^{239}\text{Pu}$ ratio from spectrum analysis. It then calculates the $\text{Pu240}/^{239}\text{Pu}$ ratio using its correlation with the $(^{241}\text{Pu} + ^{241}\text{Am})/^{239}\text{Pu}$ ratio and then the $^{238}\text{Pu}/^{239}\text{Pu}$ ratio using its correlation with the just determined $\text{Pu240}/^{239}\text{Pu}$ ratio. Finally, it calculates the ^{242}Pu fraction using its correlations with other Pu isotopes, either measured or calculated from correlations.

To determine the ^{239}Pu and ^{240}Pu abundances, the PAT applies correlations of these isotopes vs. $^{239}\text{Pu}/(^{241}\text{Pu} + ^{241}\text{Am})$ ratio, which is derived from the fitting of the 230-470 keV spectral region.

Typical measurement systems applied by the IAEA are composed of a shielded CZT ($\geq 1.5 \text{ cm}^3$) or LaBr_3 ($\varnothing 2'' \geq 0.5''$) detector, a digital MCA, and an instrumentation laptop. A 1-2 mm cadmium or tin filter is used to effectively suppress the intense 59.5 keV gamma-rays of ^{241}Am . Using these spectrometers at $\text{OCR}=10 \text{ kcps}$ and measurement live time $\text{LT}=600 \text{ s}$, one can normally acquire 2,000/20,000 counts (CZT) and 10,000/50,000 counts (LaBr_3) in the reference 413 keV peak of ^{239}Pu from a lightly shielded High/Low burnup Pu material. The quality of respective spectra will be sufficient to warrant the performance of the isotopic analysis by FRAM and PAT at the level of the stated ITVs.

Due to the large uncertainty estimates found in the bottom up approach these methods should be limited to plutonium attribute at this time.

Factors affecting the method performance

Material inhomogeneity, heavy shielding, high ambient radiation background, spectral interferences (e.g., with emissions of fission products). Time since the spent fuel is removed from the reactor and the Americium removed from the plutonium can significantly affect the results from the ratio method (technique 4 category).

Source of bottom-up value (only for prospective ITV not based on historical data)

The FRAM code evaluations are based on measurements with a variety of materials with known (certified) isotopic compositions under laboratory conditions. 299 CZT spectra in 9 sets and 530 LaBr₃ spectra in 21 sets were used in the analysis. The PAT code was evaluated against sets of spectra collected within the framework of the inter-comparison exercise [1]. The ITVs represent approximate average performance values obtained by the both codes.

Method references

- [1] A. Berlizov, M. Koskelo, B. McGinnis, J. Carbonaro, "The Inter-Comparison Exercise on Uranium and Plutonium Isotopic Analysis with Medium Resolution Gamma-Ray Spectrometers," International Workshop on U/Pu Isotopic Analysis by Nondestructive Assay Techniques for Nuclear Safeguards, IAEA, February 2021, Journal of Nuclear Materials Management 2021 Volume XLIX No. 3: 94-107.
- [2] D. Vo, "Improvements of FRAM version 6.1," International Workshop on U/Pu Isotopic Analysis by Nondestructive Assay Techniques for Nuclear Safeguards, IAEA, February 2021, Journal of Nuclear Materials Management 2021 Volume XLIX No. 3: 19-30. [3] R. Gunnink, A. Berlizov, PAT, a Program for Determining the Category of Plutonium Samples, International Workshop on U/Pu Isotopic Analysis by Nondestructive Assay Techniques for Nuclear Safeguards, IAEA, February 2021, Journal of Nuclear Materials Management 2021 Volume XLIX No. 3: 84-93.

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