Using FRAM to determine enrichment of shielded uranium by portable electrically cooled HPGe detectors

J. Zsigrai[1], A. Frigerio[1], J. Bagi[1], A. Mühleisen[1], A. Berlizov[2]

[1]European Commission, Joint Research Centre, Directorate G, Karlsruhe, Germany
[2]International Atomic Energy Agency, Vienna, Austria

Abstract:

The capability of the FRAM software to accurately determine the enrichment of shielded uranium by portable electrically cooled HPGe detectors was studied. This can have applications in the future, e.g., for the verification of aged uranium-bearing products, scrap, and waste materials, especially during short-notice random or unannounced inspections, when detector cooling with liquid nitrogen is not feasible. More than 7000 high-resolution gamma spectra of certified reference materials were taken by the ORTEC "Detective" detector under well-defined measurement conditions. Up to 16 mm of steel was used for shielding. The ^{235}U enrichment of the reference materials varied from 0.31% to 4.46%. The settings of an existing FRAM parameter set were optimized and all the collected spectra were analysed using the default and the optimized parameter sets. The results obtained with these parameter sets are compared in this paper.

Keywords: FRAM, gamma-ray spectrometry, shielded uranium, electrically cooled detector

1. Introduction

The purpose of this work was to study and possibly improve the capability of the FRAM software to determine the enrichment of shielded uranium by portable electrically cooled HPGe detectors. This task was carried out at the JRC Karlsruhe site, within the support programme to the International Atomic Energy Agency (IAEA). In particular, new customized FRAM parameter sets were developed and can be used to get more accurate results for ^{235}U enrichment than with the default parameter sets.

An advantage of electrically cooled high-resolution gamma spectrometers (ECGS) for in-field use by safeguards inspectors is that they do not require liquid nitrogen for cooling. This makes them suitable for short notice random or unannounced inspections for the verification of aged uranium-bearing products, scrap, and waste materials.

FRAM is software that calculates uranium and plutonium isotopic composition from the gamma spectra of these materials [1], [2]. It has been developed at Los Alamos National Laboratory (USA) and it has been commercialized by ORTEC and Canberra. The version used in this study was 5.1 [3].

The so called parameter sets determine what FRAM exactly does. They define the type of material (U, Pu, MOX) and the type of detector. They also contain information about the isotopes and gamma peaks to be analyzed, peak fitting parameters, energy calibration, relative efficiency constraints, etc. FRAM contains a number of default parameter sets built into the software, which cover a large number of typical measurement configurations. However, users can also prepare modified or new parameter sets to suit their specific measurement configuration. In this work we focused on parameters sets for uranium. An analogous study with plutonium parameter sets is in progress.

More than 7000 high-resolution gamma spectra of various certified reference materials were taken by the ORTEC "Detective" detector under well-defined measurement conditions with different steel shielding. These spectra were used to develop a parameter set suitable for determining the isotopic

composition of shielded uranium. Using this parameter set, the difference between the ^{235}U enrichment determined by FRAM and the certified value (FRAM's bias) can be reduced to below 2%.

2. Method and equipment

The ORTEC Detective electrically cooled spectrometer was used to record the gamma spectra. It has a high-purity germanium (HPGe) crystal of 50 mm diameter and 30 mm depth (length). Its warranted resolution is ≤ 2.0 keV at 1332 keV and ≤1.0 keV at 122 keV, while its efficiency relative to a standard 2x2 inch NaI is 10%. The conversion gain of its amplifier is set in the factory so that it can take spectra up to 3 MeV. The electronics settings cannot be changed by the user.

Spectra of all 5 items from the certified reference material set EC NRM-171 (also known as the "CBNM uranium set") [4] were recorded in 8 different geometries (5x8=40 configurations):

- At 2 cm from the detector, with 0 mm Fe shielding
- At 5 cm from the detector, with 0, 2, 4, 8, and 16 mm Fe shielding
- At 10 cm from the detector, with 0 mm Fe shielding
- At 15 cm from the detector, with 0 mm Fe shielding

For each configuration 192 spectra were recorded with 5 minute real time (16 hours total measurement time), i.e., 40x192 = 7680 spectra were recorded. Each item of the reference material set contained 200 g of UO_2 in container with 2 mm Al window. The certified ^{235}U enrichments are shown in Table 1.

Table 1. Certified ^{235}U enrichment of the reference samples [4]

Sample name	Certified enrichment ± Uncertainty (2s) [mass %]
CBNM U031	0.3166 ± 0.0002
CBNM U071	0.7119 ± 0.0005
CBNM U194	1.9420 ± 0.0014
CBNM U295	2.9492 ± 0.0021
CBNM U446	4.4623 ± 0.0032

To investigate the effect of counting statistics on the results of FRAM the 5-minute spectra were added up to make spectra with various real times which are multiples of 5 minutes. A script was written in the Python 3.5 programming language for adding the spectra. To automatically analyse the large number of spectra an Excel macro was used, which interacts with the command-line mode of FRAM v5.1 and puts the results into an Excel sheet.

To measure the performance of FRAM and different parameter sets, two quantities were used: the average relative bias and the mean absolute value of the relative deviation (MARD) of the ^{235}U results. These two quantities were calculated for each configuration (defined by enrichment, distance and shield thickness) as

$$Average\ relative\ bias = \frac{\sum_{i=1}^{n} \frac{x_i - x_{Ref}}{x_{Ref}}}{n}, \qquad MARD = \frac{\sum_{i=1}^{n} \frac{|x_i - x_{Ref}|}{x_{Ref}}}{n},$$

where n is the number of spectra analysed (e.g. n=192 for the 5-minute spectra), x_i is the ^{235}U enrichment calculated by FRAM and x_{Ref} is the certified reference value for the ^{235}U enrichment. The average relative bias can be either positive or negative. It describes the expected accuracy of many (n) measurements. The MARD is always positive and it describes expected accuracy of a single measurement.

To see how the results from FRAM can be improved all spectra were first analysed using a "default" parameter set for the Detective supplied by ORTEC on the installation CD of FRAM 5.1. This is not one of the built-in parameter sets of FRAM, though it was made by the FRAM developers [5]. After each modification of the parameter set, the entire set of spectra was reanalysed and the average relative bias and the MARD recalculated for each configuration.

3. Results

3.1. Results with the "default" parameter set

To take a snapshot of the performance of FRAM using the default Detective parameter set [5], the 5-minute and the 80-minute spectra were analysed. The average relative bias and the MARD for these analyses are shown in Figure 1. The spectra were taken at a 5 cm source-to-detector distance. This distance was an acceptable guess for having a compromise between sufficient count rate and coincidence summing effects.

For the configuration with the depleted uranium sample, CBNM U031, and 16 mm Fe shielding the average bias is much higher (more negative than -14 %) than for the other configurations and is outside of the scale of the graphs. This large bias is probably due to two reasons. First, the heavy shielding extremely reduces the already low number of counts in the peaks of ^{235}U. E.g. the number of counts in the 186 keV peak with 16 mm shielding is about 8 times lower than with no shielding while the 143 keV peak is not even visible for the 5 minute spectra with 16 mm shielding. Even for 80 minutes measurement time the number of counts for the ^{235}U peaks stays low with 16 mm shielding. Second, the Compton scattering in the heavy shielding greatly increases the background below the ^{235}U peaks. Therefore, the ^{235}U peaks in the spectra of heavily shielded depleted uranium are very difficult to fit.

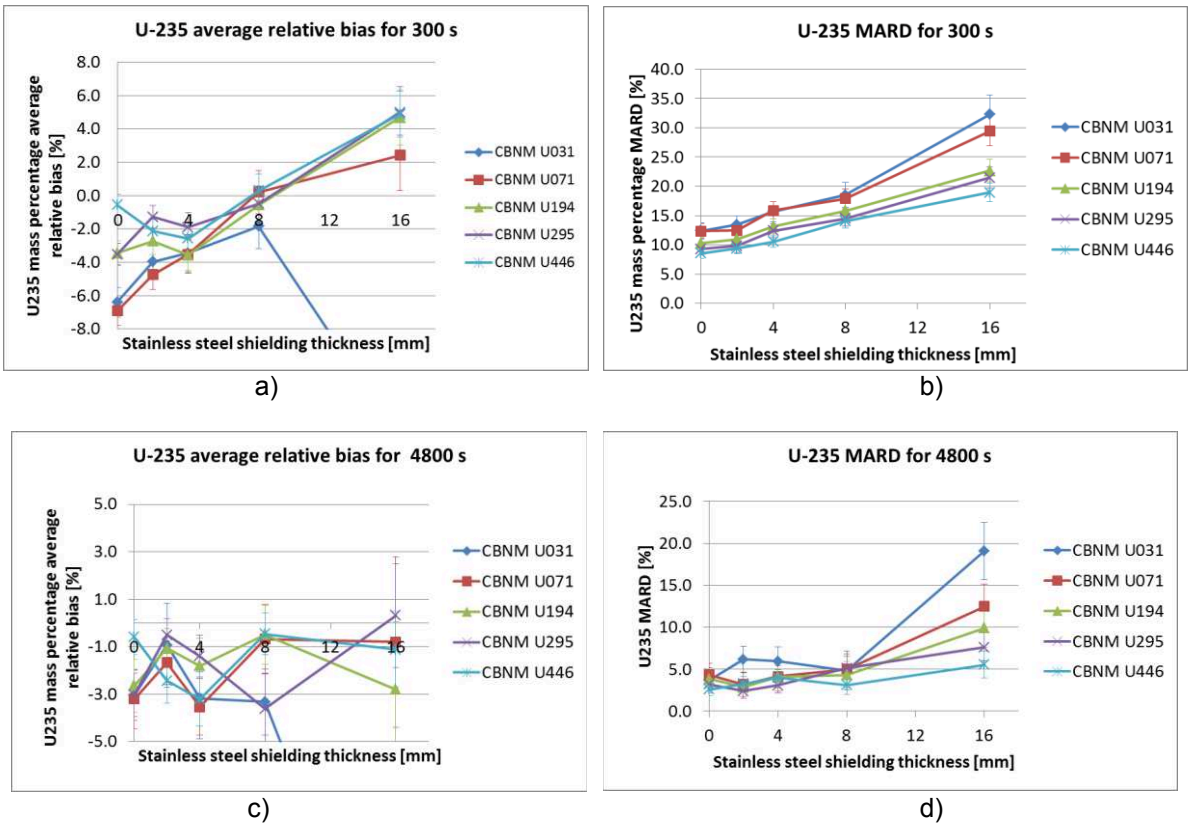
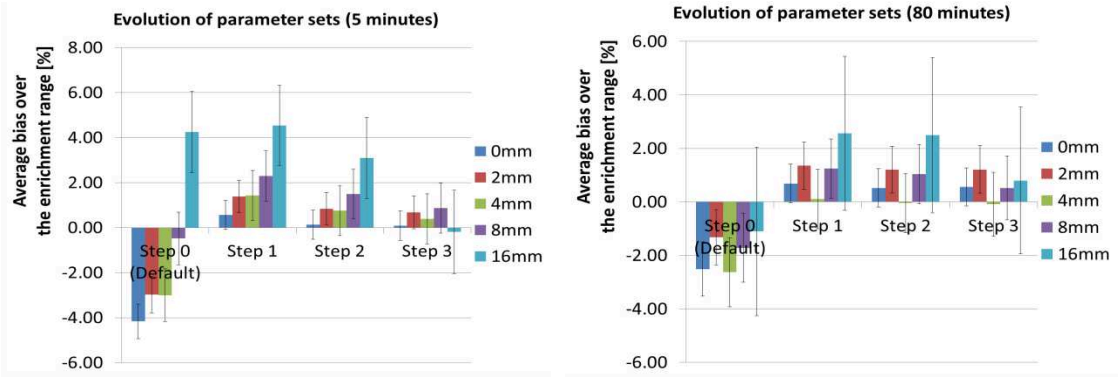


Figure 1. "Default" Detective parameter set: Average relative bias and MARD as a function of Fe shielding thickness for measurements at 5 cm from the detector: a) and b) for 5-minute spectra; c) and d) for 80-minute spectra. The error bars for the average bias are the corresponding MARDs divided by the square root of the number of measurements, while the error bars for the MARD are the corresponding average uncertainties reported by FRAM divided by the square root of the number of measurements.

3.2. Evolution of the parameter set

The parameter set was modified step-by-step, changing only one type of parameter at a time, to see the influence of each parameter on the bias and MARD. For Figure 2 the average biases for the various configurations were averaged over all samples and plotted for each step during the evolution of the parameter set and for each shield thickness. The outlier corresponding to the configuration for depleted uranium, CBNM U031, with 16 mm shielding was not included in this average.



a) b)

Figure 2. Steps in improving the parameter set: The average (over different enrichments) of average biases as a function of shield thickness is shown for each step for different Fe shield thicknesses. Error bars are the corresponding average MARDs divided by the square root of the number of spectra. a) for the 5-minute spectra, b) for the 80-minute spectra.

After a few trial-and-errors, the correct steps in modifying the parameter set were the following:

1. Turning off the coincidence summing correction. This dramatically improves the average bias of the results obtained from the measurements at 5 cm source-to-detector distance. This means that at this distance the coincidence summing effects are relatively small and FRAM overestimates the correction due to coincidence summing.
2. Fine-tuning the peak fitting parameters (energy calibration and peak-shape parameters), to improve the accuracy of the peak areas determined by FRAM. This has a minor impact on the results
3. Modifying the boundaries and types of absorber materials, to account for the effects of shielding. This involved removing Cd and adding Al as absorber (0-50 mm), and modifying the boundary values for effective Fe thickness (0-50 mm). This step has a major impact for the spectra taken with heavy shielding.

3.3. Results with the "optimal" parameter set

The optimal parameter set is Step 3 in Figure 2. The detailed results for the average relative bias and MARD obtained using this parameter set are shown in Figure 3. Just as for the default parameter set, the average bias for the configuration with the depleted uranium sample, CBNM U031, and 16 mm Fe shielding is outside of the scale of the graphs (it is more negative than -14 %).

The improvement of the average relative bias is evident by comparing Figure 3 to Figure 1. However, the modifications of the parameter set have hardly any impact on the MARD. This is because the MARD is mainly determined by the counting statistics, and is not much influenced by the bias.

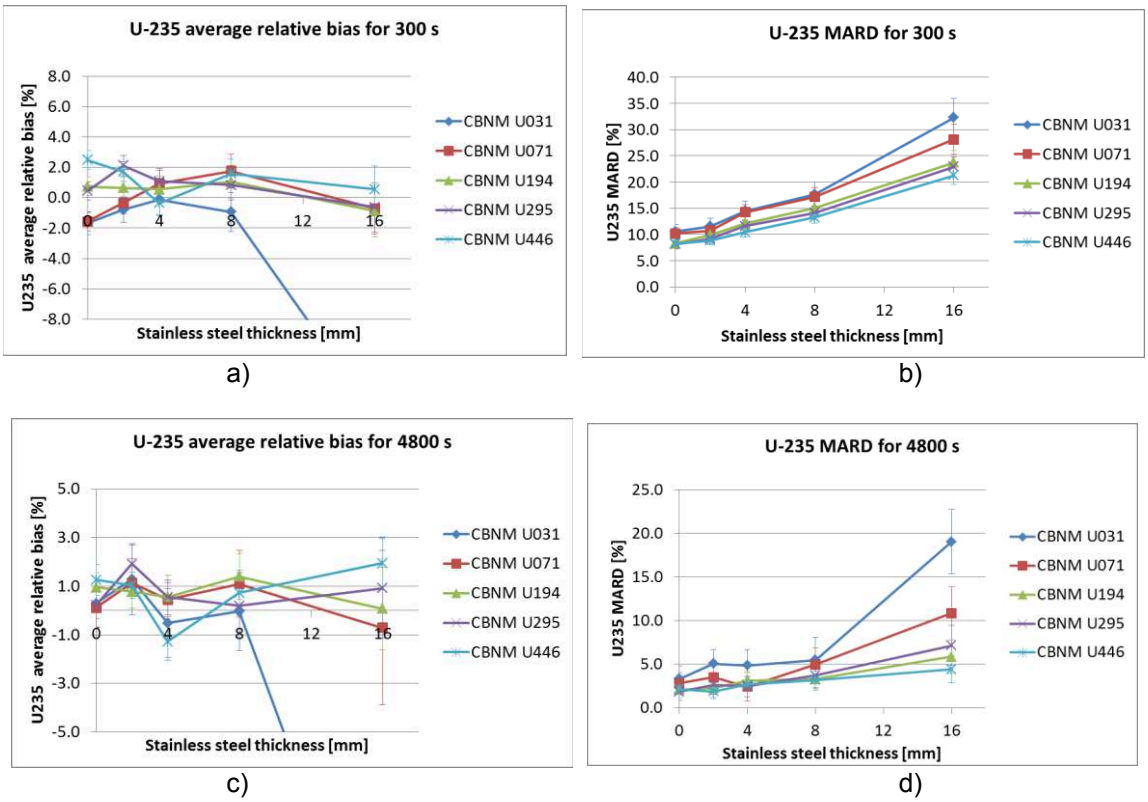


Figure 3. "Optimal" parameter set: Average relative bias and MARD as a function of Fe shielding thickness for measurements at 5 cm from the detector. a) and b) for 5-minute spectra; c) and d) for 80-minute spectra. Error bars are as in Figure 1.

3.4. Influence of sample-to-detector distance

As mentioned above, turning off the coincidence summing correction in the parameter set improves the results obtained from the spectra recorded at 5 cm source-to-detector distance. To further investigate the performance of the coincidence summing correction algorithm of FRAM, the spectra taken at different distances from the detector with no shielding were analysed by the default and the modified parameter set, as shown in Figure 4.

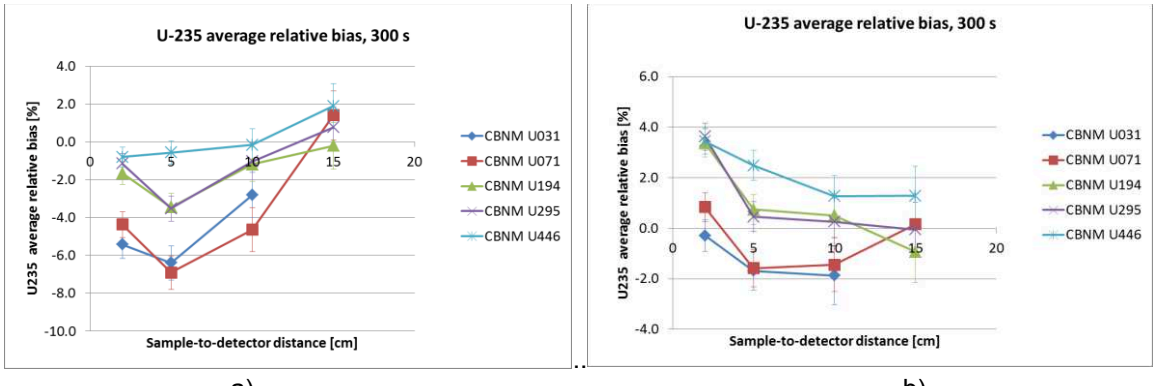


Figure 4. Average relative bias for unshielded samples as a function of source-to-detector distance with 5-minute measurement time. a) With default parameter set. b) With the modified parameter set, "Step 3". Error bars are as in Figure 1.

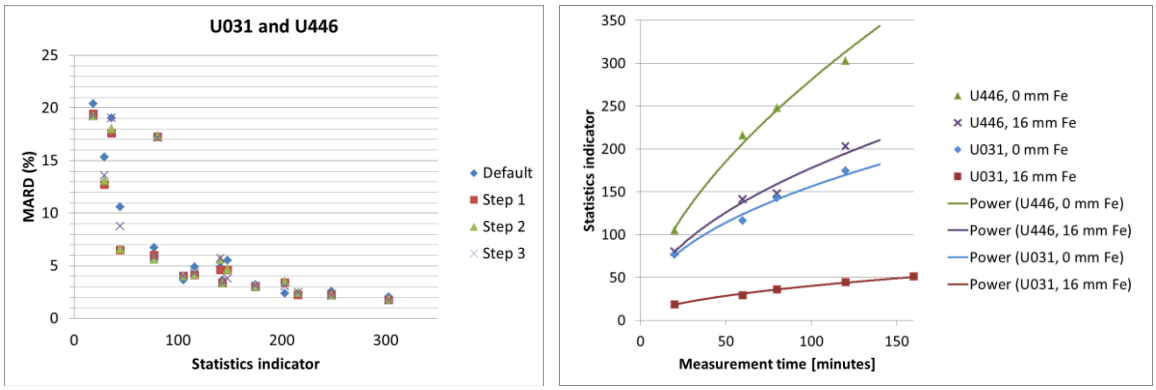
It can be seen in Figure 4 that the newly developed parameter set provides better results for most configurations. However, at 2 cm from the detector the effects of coincidence summing seem to be significant, and the correction algorithm would probably have to be turned on.

3.5. Influence of counting statistics

As observed by comparing Figure 1 and Figure 3, the MARD does not improve much by modifying the parameter set. This is because the MARD is mainly determined by the counting statistics, and the improvements in the relative bias are much smaller than the MARD. To investigate the influence of the counting statistics and measurement time on the MARD, an indicator has to be constructed which describes the statistical quality of the spectra. In this work we used a number constructed as the reciprocal value of the combined relative uncertainties of the 186 keV peak of ^{235}U and of the 1001 keV peak of ^{238}U:

$$Statistics\ indicator = \frac{1}{\sqrt{\left(\frac{\Delta S_{186}}{S_{186}}\right)^2 + \left(\frac{\Delta S_{1001}}{S_{1001}}\right)^2}},$$

where S_{186}, ΔS_{186}, S_{1001} and ΔS_{1001} denote the peak area and its absolute uncertainty of the 186 keV and 1001 keV peaks, respectively.



a) b)

Figure 5. a) Dependence of the MARD on the statistics indicator for the samples U031 and U446 measured at 5 cm with 0 mm and 16 mm of Fe shielding, for different parameter sets. b) Dependence of statistics indicator on the measurement time, for the sample U031 and U446 measured at 5 cm with 0 mm and 16 mm Fe shielding.

It can be seen from Figure 5a) that the MARD becomes smaller than 2 % if the statistics indicator is higher than about 200, almost independently of the sample (enrichment), shield thickness and parameter set used for obtaining the MARD. Furthermore, Figure 5b) shows that the statistics indicator does not go above 200 for most of the 5-minute and 80-minute spectra evaluated in this paper. That means that the statistics of most of the spectra used in this work is not enough for getting better precision (MARD) of the ^{235}U result calculated from a single spectrum, regardless of the improvements of the parameter set. Nevertheless, the analysis of the large number of spectra shows that the accuracy (bias) of the results is reduced by using the improved parameter set.

4. Conclusion

A new FRAM parameter set has been developed for analysing shielded LEU spectra taken with the ORTEC Detective. The performance of the default and the newly developed set was evaluated in detail for the ^{235}U enrichment range from 0.31% to 4.46 % and for Fe shield thicknesses up to 16 mm. It was shown that the new parameter set performs better, especially for heavily shielded samples. This parameter set can be equally used for shielded or unshielded samples. The parameter set will be made available to the IAEA within EC support programme.

References

[1] T. E. Sampson and T. A. Kelley, “PC/FRAM: A code for the non-destructive measurement of the isotopic composition of actinides for safeguards applications, LA-UR-96-3543,” 1996.

[2] T. E. Sampson, T. A. Kelley, and D. T. Vo, “Application Guide to Gamma-Ray Isotopic Analysis Using the FRAM Software, LA-14018,” 2003.

[3] D. T. Vo, “Operation and Performance of FRAM Version 5.1 Proceeding of the 52nd Annual Meeting of The Institute of Nuclear Materials Management, Palm Desert, CA, USA, July 17-21, 2011, LA-UR-11-03016,” 2011.

[4] “EC Certified Nuclear Reference Material 171, Certificate of Analysis, Commission of the European Communities, Joint Research Centre, Central Bureau for Nuclear Measurements, Geel,” 1985.

[5] T. E. Sampson, G. W. Buttler, D. T. Vo, T. Wenz, and S. C. Myers, “The use of FRAM with a portable, HPGe-based nuclide identifier to measure isotopic composition of plutonium and uranium, LANL report.”