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Study on the performance of different uranium isotopic codes used in nuclear safeguards activities

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Abstract

Accurate characterization of Nuclear Materials (NMs) for nuclear safeguards purposes requires studying the optimal conditions over a wide range of experimental conditions. Gamma spectroscopy is commonly used to measure the ^{235}U enrichment in NMs bearing samples. Several commercial Codes such as the Multi Group Analysis (MGA⁺⁺), the Multi Group Analysis for Uranium (MGAU) and the “Fixed energy, Response function Analysis with Multiple efficiencies” (PC/FRAM) are used to measure ^{235}U enrichment. Many factors, such as sample enrichment, density, homogeneity, geometry, and container type may affect the results of measurement. This work studies the effect of these factors on the performance of each of the three codes to estimate uranium enrichment. The results are compared to determine the optimum conditions and best performance among the three codes for achieving the most accurate estimation of ^{235}U enrichment in the measured samples.

Keyword: Nuclear physics

1. Introduction

The main target of safeguards inspections is to verify that the data declared by the operators are correct and complete. Presently, several inspection devices are available that utilize different types of measurement technologies. During safeguards inspections, there is often a need for a quick estimate of the isotopic contents and mass in uranium (U) samples. The nuclear material (NM) should be accurately accounted for at all times, which requires a measurement of the isotopic abundances as well as the elemental weight or concentration (Dias and Grund, 2007).

One of the most common techniques used in safeguards applications is gamma ray (γ -ray) spectroscopy for assaying NMs. In the last couple of years, many Non-Destructive Assay (NDA) techniques have been used for U enrichment measurements (Yucel, 2007). MGAU, MGA++, and PC/FRAM are common programs used for safeguards activities to measure plutonium (Pu) and/or U enrichment. These codes do not require the use of any NM standards or calibration standards, but they determine these intrinsically from the measured spectral data and can be used to measure virtually any size and type of U or Pu sample (Abousahl et al., 1996). MGA methodology, including detailed descriptions of peak shapes, efficiencies, geometry considerations, and background subtraction, is described in detail in several references (Gunnink et al., 1990; Clark 1996, 1998). For Pu isotope measurements, MGA was reported to analyze the low-energy γ -ray spectrum, along with higher-energy γ -rays (Gunnink and Ruhter 1990). While the original MGA code relies on the analysis of the 100-keV region, the U235 U isotopic analysis code uses γ -rays less than 300 keV. MGA++ is a suite of three software programs (MGA, U235, and MGAHI, a Pu isotopic analysis code that uses the 200 keV–1 MeV energy region) for the analysis of actinide spectra acquired by germanium (Ge) detectors.

The MGAU code analyzes the complicated 80–130 keV region of the spectrum taken with a high-resolution planar Ge detector for U measurement (Gunnink et al., 1994). PC/FRAM, on the other hand, can use the full energy range and does not have strict requirements for high detector resolution (Solodov et al., 2006). However, planar detectors have most often been used to collect and analyze data in the 120–420 keV range, although they are not limited to this range (Sampson and Kelley, 1996).

In this work, the performance of MGAU, MGA++, and PC/FRAM for nuclear safeguards applications will be discussed, along with some parameters that may affect these programs' performance, such as containers (materials and densities), time of measurement, and count rates.

2. Theory

The usual method of determining relative isotopic abundances from γ -ray spectra that contain peaks from two or more isotopes is to measure and interpret the peak intensities of the neighboring γ -ray peaks associated with the different isotopes. This method becomes slightly more complicated when analyzing peaks that significantly overlap with each other; then, the peak intensities must be determined by using peak-fitting techniques. To overcome this difficulty, Gunnink et al. (1994) developed a "response function" method that allows many peak parameters to be fixed relative to each other, thus greatly reducing the degrees of freedom in the fitting process. The MGAU method uses X- and γ -rays in the 80–130 keV region of the γ -ray spectrum of a U sample. The 89- and 93-keV X-ray peaks are from the decay of ^{235}U and the 92.367- and 92.792-keV γ -ray from ^{238}U - ^{234}Th . This makes the region suitable for determining the relative abundances of ^{235}U and ^{238}U . The U isotopic ratio is determined, basically, by measuring the intensity of these γ -rays of very close energies that arise from different isotopes. Since the γ -ray emission probabilities and half-lives are known, the isotopic ratios of two different atoms can be calculated if the relative detection efficiencies for the peaks of interest can be estimated (Yücel and Dikmen, 2009).

The PC/FRAM code supports the analysis of the more energetic but less abundant γ -rays in the energy region between 125 and 660 keV. Resorting to the higher-energy γ -rays for isotopic analysis provides some advantages in cases of stronger sample shielding. The peak-ratio algorithms used by PC/FRAM do not require special energy or enrichment calibrations before the measurements can be started. Using a spectrum in memory, the software routines perform energy and full-width at half maximum (FWHM) calibrations, subtract the background, and then calculate the U enrichment levels based on the ratios of the peaks that are specific to the isotopes of interest (Solodov et al., 2006). This concept is applicable to any samples regardless their geometries and composition. No calibration or standard materials are required.

Sampson and Kelley, 1996 defined the fundamental measurement in Eq. (1) as one of isotopic ratios that are given by expressing photo peak counts for γ -ray l from isotope k and combining it with the expression for γ -ray j from isotope i ; therefore, the expression for the atom ratio of isotope i to isotope k from the measurement of a γ -ray with energy E_j from isotope i and E_l from isotope k can be given as:

$$N^i / N^k = C(E_j^i) / C(E_l^k) * T_{1/2}^i / T_{1/2}^k * BR^k / BR^i * RE(E_i) / RE(E_j) \quad (1)$$

where N^i = Number of atoms of isotope i

$C(E_j^i)$ = Photopeak area of γ -ray j with energy E_j emitted from isotope i

$T_{1/2}^i$ = Half-life of isotope i

BR_j^i = Branching ratio (γ -rays/disintegration) of γ -ray j from isotope i

$RE(E_j)$ = Relative detection efficiency of γ -ray with energy E_j . It includes detector efficiency, measurement geometry, sample self-absorption, and attenuation in materials between the sample and detector. The relative efficiency is determined from each measured spectrum by fitting the energy variation of the quotient of C/BR for a series of γ -rays from a single isotope.

3. Experimental

3.1. Material and equipment

In this study, a set of U standard reference materials consisting of nominal abundances 0.31%, 0.71%, 2.96%, and 4.46% of ^{235}U were used for the measurements. Each of the samples contained 200.1 ± 0.2 g of U_3O_8 powder encased in aluminum (Al) cylindrical containers (Carpenter et al., 1986). The dimensions of the Al cans were 70 mm in height and 20.8 mm in diameter. Table 1 shows the characteristics of the used samples.

To study the performance of the MGAU, MGA++, and PC/FRAM codes, a portable γ -ray spectrometer (Canberra-U-Pu inspector 2000), based on a Digital Multi-channel Analyzer (MCA) with a planar HPGe detector (Canberra GL0515R), was used. As provided by the manufacturer, the detector has a Ge crystal with a 49-mm active diameter and 14.5-mm thickness and a 0.5-mm-thick Al window. The detector has a measured resolution of 650 eV at 122 keV (^{57}Co) gamma energy (Darweesh et al., 2014). The output signal of the detector is processed through a Canberra preamplifier (model 2002CP). The data acquisition was carried out using gamma spectroscopy software based on Canberra Genie 2000. Three sets of software analysis were used to analyze the output spectra:

Table 1. Description of SNM-969 samples used in the measurement.

Sample ID	Full Hight (cm)	Denisty g/cm^3	Certified/Decleared values of Uranium				
			U_3O_8	^{235}U	Refernec relative abundance		
					Wieght(g)	Wieght(g)	^{235}U (atom%)
031	2.08 ± 0.05	2.50 ± 0.06	200.1 ± 0.2	0.5260	0.3206 ± 0.0002	99.6627 ± 0.0004	0.0020 ± 0.0002
071	2.08 ± 0.05	2.50 ± 0.06	200.1 ± 0.2	1.2047	0.7209 ± 0.0005	99.2738 ± 0.0004	0.0053 ± 0.0002
194	2.08 ± 0.05	2.50 ± 0.06	200.1 ± 0.2	3.2918	1.9664 ± 0.0014	98.0159 ± 0.0159	0.0174 ± 0.0002
295	2.08 ± 0.05	2.50 ± 0.06	200.1 ± 0.2	5.0056	2.9857 ± 0.0021	96.9826 ± 0.0029	0.0280 ± 0.0002
446	1.58 ± 0.05	3.291 ± 0.1	200.1 ± 0.2	7.5678	4.5168 ± 0.0032	95.4398 ± 0.0032	0.0365 ± 0.0004

- 1 The first set is “U-Pu Inspector,” dedicated to the direct measurement of the isotopic composition of U with the code MGAU (version 4.1 provided by Canberra).
- 2 The second set is “MGA++,” dedicated to the direct measurement of the isotopic composition of U using the code U235view (version 1.06 provided by ORTEC).
- 3 The third set is “PC/FRAM,” dedicated to the direct measurement of the isotopic composition of U using the code PC/FRAM (version 4.2 provided by LANL).

For the PC/FRAM analysis, the low-energy range was implemented using the parameter ULEU_Plnr_060–250 with a gain of 0.075 KeV/channel. To calculate the counting statistics, average, and RSD, each sample was measured three times.

3.2. Measurement

The following parameters were chosen in order to fulfill — as accurately as possible — on-site measurement configurations. To avoid the sum peaks phenomenon, the source was located 10 cm from the end cap of the detector.

The effect of container type and thickness on the performance of the different codes used for ^{235}U enrichment estimation was investigated at different thicknesses, from 0.1 up to 0.9 cm.

To check the performance of the codes with different container materials and thicknesses, the standard SRM 969 was measured at a fixed time of 30 min. Samples were placed at a fixed distance (10 cm) from the surface of the detector’s cap. The geometry was fixed for all measurements of each U isotopic composition. In order to study the effect of measuring time, three sets of measurements were conducted at different time durations (15, 30, 45, or 60 min) and individual averages were calculated.

4. Results and discussion

In a previous study, we presented the effect of detector resolution on the performance of the three codes, expressed as biases of normalized measured/certified values under standard experimental conditions. These biases ranged from about 0.01 to 2.1 % (Darweesh et al., 2017). Performance values obtained during other work indicated that the use of MGAU for ^{235}U enrichment measurements under laboratory conditions, associated with an improved high-resolution γ -ray spectroscopy system and a robust statistical treatment of data, is capable of achieving a final standard uncertainty of 0.5% (Grund and Dias 2009). The performance of different evaluation codes with attenuators between the detector and the measured sample simulating different container thickness and material types is presented in Figs. 1(a-e), 2(a-e) and 3(a-e). The effects of using stainless steel (density 8000 kg m^{-3}), aluminum

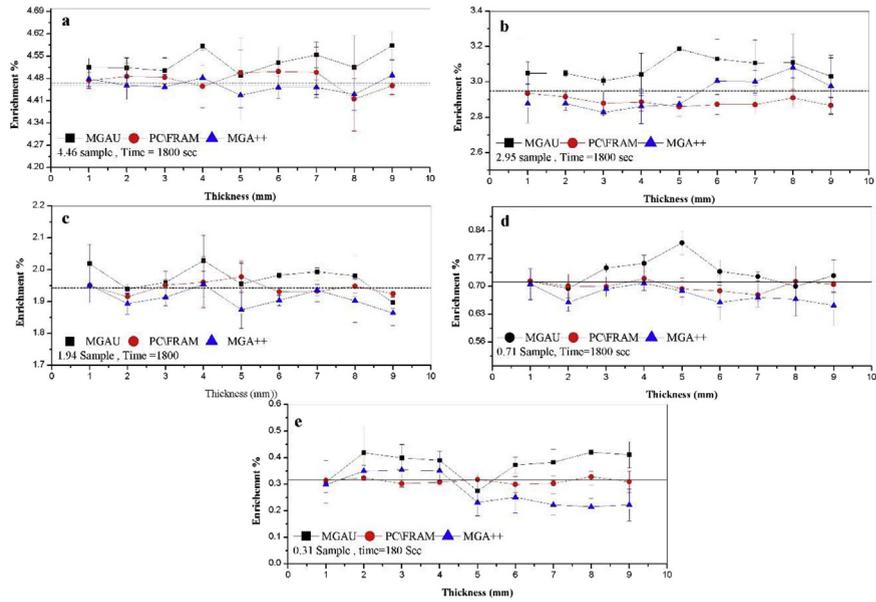


Fig. 1. Measured enrichment at different Aluminum thickness for samples (a) 4.46 (b) 2.95 (c) 1.94 (d) 0.71 and (e) 0.31.

(density 2700 kg m^{-3}), and iron (density 7870 kg m^{-3}), which are common container material types, are also described in these Figures.

In order to test the influence of measuring time on the performance of the different evaluation codes, the certified standard reference materials were measured for

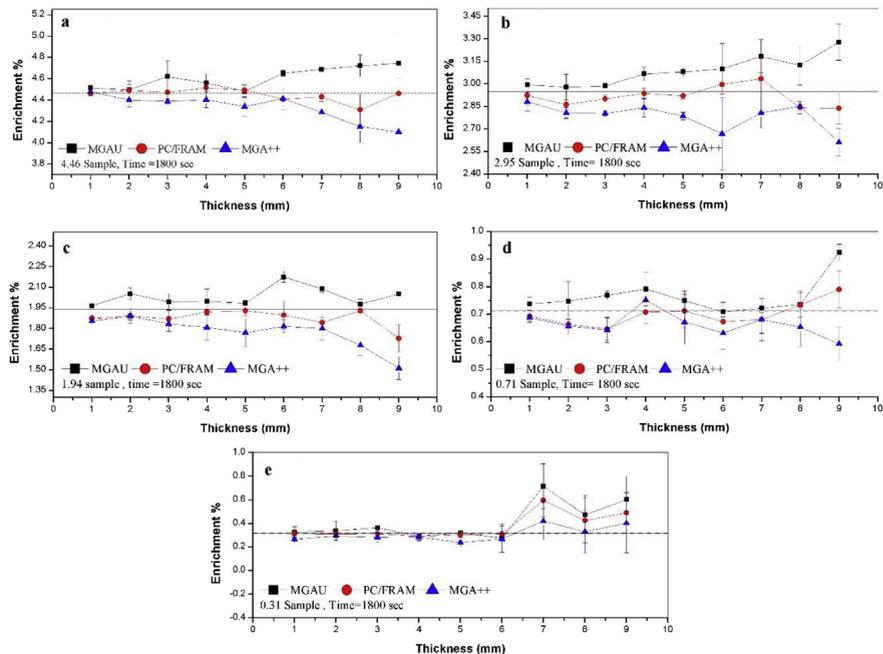


Fig. 2. Measured enrichment at different Iron thickness for samples (a) 4.46 (b) 2.95 (c) 1.94 (d) 0.71 and (e) 0.31.

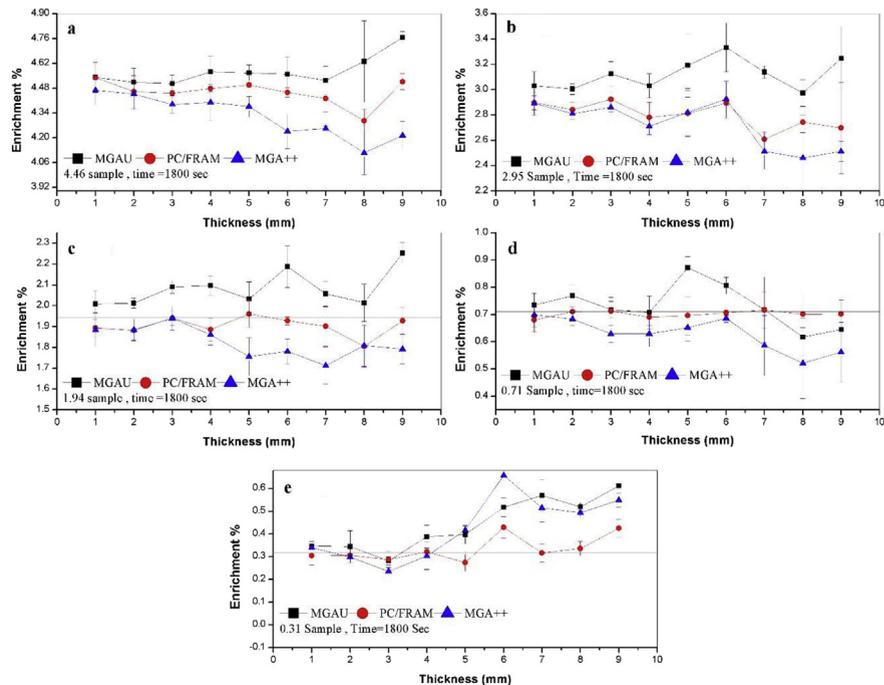


Fig. 3. Measured enrichment at different Steel thickness for samples (a) 4.46 (b) 2.95 (c) 1.94 (d) 0.71 and (e) 0.31.

different time periods, starting from 15 to 60 min for each sample. Fig. 4(a-e) shows the effect of measuring time on the obtained results of ^{235}U enrichment in comparison with the declared values. Tables 2, 3, and 4 show the estimated ^{235}U values using MGAU, PCFRAM and MGA++ codes for different enrichment samples at different measurement times in comparison with the declared values. The estimated values for the different enrichment samples show better statistical accuracy as the measurement time increases.

For most of the investigated cases, the estimated ^{235}U enrichment values using MGAU were positively biased, whereas those for MGA++ (using the U235, U isotopic analysis code) were negatively biased, which is in agreement with our previous findings (Darweesh et al., 2017). This could be attributed to two things: the differences in the algorithms of the two codes or the combination of the detector type and code. While MGA++ (using the U235, U isotopic analysis code) uses γ -rays smaller than 300 keV, the MGAU method relies on X- and γ -rays in the 80–130 keV region of the γ -ray spectrum of U, which are more sensitive to container thickness and the type of material or attenuator. The relative efficiencies are based on comparing the intensities of the close energies X-ray peaks of U. The principal limitation of the method, besides requiring a secular equilibrium for the ^{235}U , ^{238}U daughter nuclides, arises from the fact that the γ - and X-ray signatures from ^{231}Th and ^{234}Th become strongly unbalanced in intensity at very low (<1%) and

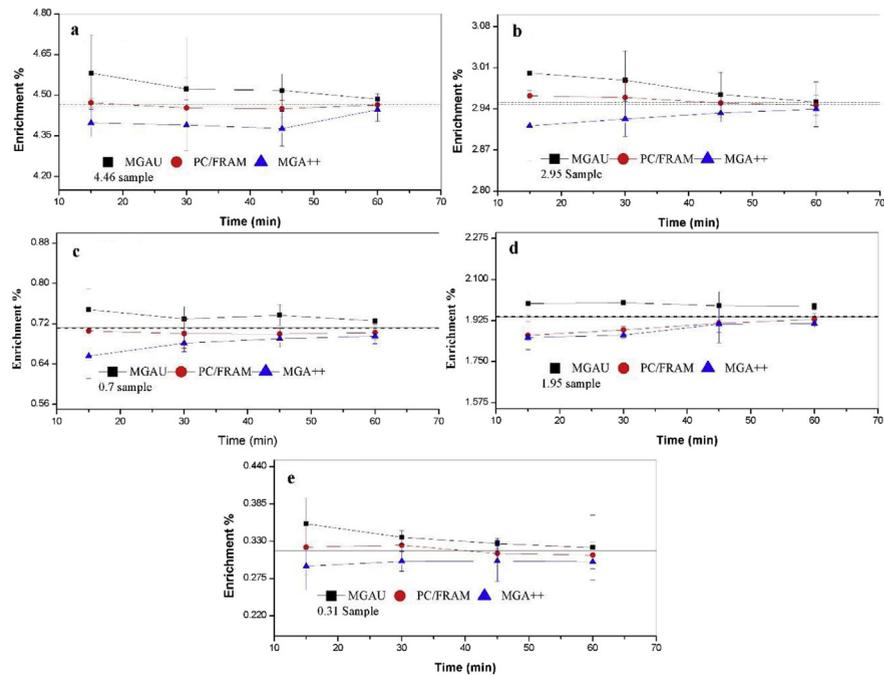


Fig. 4. Measured enrichment in comparison with the certified values at different measuring time durations for samples (a) 4.46 (b) 2.95 (c) 1.94 (d) 0.71 and (e) 0.31.

very high (>90 %) enrichments (Abousahl et al., 1996). It is also hard to determine the branching ratios for gamma transitions in the range of the 100-keV energy region because of X-ray tails and uncertainties in applying the background subtraction algorithm to remove their effect from the spectrum in this energy range. This explains the less precise results obtained at low enrichment grades. RSDs of 29.8, 93.4, and 90.0% at 0.31% enrichment were obtained using 9-mm sheets of aluminum, stainless steel, and iron, respectively. This is in agreement with reported findings that a UF₆ cylinder with 16-mm-thick walls attenuates the 90- to 100-keV radiations by about a factor of 250 (Gunnink, R. et al., 1994). The precision of the results is obviously reduced when the signals are so highly attenuated. As the ²³⁵U ratio in samples increased, the difference between the estimated value and declared values decreased

Table 2. Estimated ²⁵³U values using MGAU code for different enrichment samples at different measurement time in comparison with the declared values.

Sample ID	Declared Value	RSD%	Relative Bias %
031	0.3166	1.5	1.3 to 12.4
071	0.7119	0.9	1.95 to 5.009
194	1.9420	0.78	2.2 to 3.03
2.95	2.9492	2.23	0.098 to 1.75
4.46	4.4623	4.08	0.51 to 2.67

Table 3. Estimated ^{253}U values using PCFRAM code for different enrichment samples at different measurement time in comparison with the declared values.

Sample ID	Declared Value	RSD%	Relative Bias %
031	0.3166	2.00	-2.20 to 2.30
071	0.7119	0.27	-1.76 to -0.88
194	1.9420	3.04	-4.16 to -0.62
2.95	2.9492	0.75	-0.101 to 0.43
4.46	4.4623	1.01	-0.28 to 0.21

Table 4. Estimated ^{253}U values using MGA++ code for different enrichment samples at different measurement time in comparison with the declared values.

Sample ID	Declared Value	RSD%	Relative Bias %
031	0.3166	0.3	-7.29 to -4.8
071	0.7119	1.7	-7.8 to -2.33
194	1.9420	3.09	-4.6 to -1.56
2.95	2.9492	1.24	-1.28 to -0.37
4.46	4.4623	3.07	-1.92 to -0.34

due to the better count rate. As the time of the measurement is constant, the change in the count rate is mainly due to the content of ^{235}U in the measured sample and the proportionality between the count rate and activity.

The estimated errors in the MGA++ results for the 0.31% depleted sample were -29.84, 73.41, and 26.94 % for 9-mm sheets of aluminum, stainless steel, and iron, respectively. For the natural sample with the 0.7119% ^{235}U abundance, the errors were -8.4, -21.04, and -16.84 % for 9-mm sheets of aluminum, stainless steel, and iron, respectively. For the enriched sample of 4.4632% ^{235}U , as estimated using MGA++ software, the errors were 0.634, -5.591, and -8.101 % for 9-mm sheets of aluminum, steel and iron, respectively.

The estimated errors for the PC/FRAM results for the 0.31% depleted sample with the use of different attenuators were 2.4, 34.5, and 54% for 9-mm sheets of aluminum, steel, and iron, respectively. For the natural sample with the 0.7119% ^{235}U abundance, the highest errors were -0.955, 1.25, and 11.01 % for 9-mm sheets of aluminum, steel, and iron, respectively. For the enriched sample with 4.463% ^{235}U , the estimated values using PC/FRAM software were more precise; the associated errors were -0.12, 1.22, and 0.0045 % for 9-mm sheets of aluminum, steel, and iron, respectively. These results could be attributed to two reasons. First, the use of low-energy range analysis mode for PC/FRAM (060–250 keV) which challenged the performance of the algorithm for shielded samples. Second, the use of planar detector instead of coaxial detector. These findings are in agreement with Sampson,

T.E. 1996 where precluding the analysis below 200 keV, (sample shielding or thick-walled sample container) PC/FRAM can still obtain complete the isotopic analysis. using only γ -rays above 200 keV from a single coaxial detector spectrum. The authors also stated that the planar detector is usually not the detector of choice when the measured items are shielded or in thick containers. The combination of PC/FRAM, shielding, and a planar detector is challenging; the spectra are weak because of the small detector and the low number of channels/keV, resulting in very narrow peaks at low channel numbers for the high-resolution planar detector. The precision of coaxial detector U measurements is about a factor of two better than that of the planar detector.

5. Conclusion

In this study, the performance and applicability of MGAU, PC/FRAM, and MGA++ enrichment codes for shielded U using a planar detector was investigated. With increasing wall thickness at low enrichment grades, the performance of the codes, expressed in terms of precision, was in the order PC/FRAM, MGA++, and MGAU. Although measurements of U using planar detectors are difficult for FRAM to analyze, such measurements were performed successfully under the chosen experimental conditions despite the relatively high statistical uncertainties. Therefore, in heavily shielded conditions, it is recommended to use PC/FRAM for γ -rays above 200 keV using the coaxial detector spectrum. At low enrichment grades and the large wall thickness of stainless steel, MGAU failed because of the attenuation. The optimum choice of evaluation code and detector type (planar or coaxial detectors) is strongly dependent on the measuring conditions.

Declarations

Author contribution statement

Moustafa Darweesh: Conceived and designed the experiments; Performed the experiments; Analyzed and interpreted the data; Wrote the paper.

Sahar Shawky: Analyzed and interpreted the data; Wrote the paper.

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Competing interest statement

The authors declare no conflict of interest.

Additional information

No additional information is available for this paper.

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