

Effect of Detector Resolution on the Measurement of Nuclear Material Enrichment

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Abstract

Nuclear material verification for safeguards purposes is an activity carried out to confirm that the amount of nuclear material (NM) present at a given time within a certain place is in agreement with the operator declarations. Nuclear materials are usually measured using gamma-ray spectrometer in order to quantify certain isotopes. Different codes have been used to quantify the isotopic abundance in nuclear material samples. This study is investigating the performance of the Multi Group Analysis (MGA), the Multi Group Analysis for Uranium (MGAU) and the Full Range Analysis (PC/FRAM) at different energy resolution of the counting systems and different uranium isotopic compositions. The normalized measured/certified values (M/C) were used to monitor the performance of each code. The performance of the three codes showed proportional relation to measured enrichments. PC/FRAM analysis provided the best consistency along the studied resolution and enrichment ranges with normalized measured/certified values ranging from 0 to 7%. MGAU and MGA showed more sensitivity towards low resolution detectors especially at lower enrichments with normalized measured/certified values ranged from 0 to 30 % and 0 to 20 % respectively.

Keywords: Nuclear Safeguards; MGAU; PC/FRAM; U235; Uranium Enrichment.

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1. Introduction

Resident materials at nuclear facilities or being in shipment must be categorized for domestic and international materials control and accountability. One of the key elements in nuclear material accounting system is the measurement system, which is used to verify and characterize the NM.

Several destructive analysis (DA) and non-destructive assay (NDA) techniques are available for elemental and isotopic determinations [1-3]. The non-destructive assay is very common as it is a measurement of the nuclear material content, element or isotopic concentration of an item without producing significant physical or chemical changes in this item [4,5].

Gamma spectrometry is the most applicable, non-destructive technique used for uranium isotopic composition analysis. The detector has to be chosen appropriately for every specific type of measurement. MGAU, MGA and PC/FRAM are common programs used for uranium isotopic measurements in safeguards activities. The MGA is a widely used method in the isotopic abundance determination of nuclear materials. The MGAU [6] code analyzes the complex region in uranium spectra around 100 keV which requires the use of a High-r-Resolution Germanium (HpGe) detector in the acquisition of the spectrum. The general recommendation for MGAU analysis is to have the detector energy calibration gain set at approximately 0.075 keV/channel. In order to obtain those gain settings, it is usually set 185.71 keV on 2480 channels [7]. The large volume Broad Energy Germanium (BEGe) or Coaxial (COAX) detectors are generally preferred for their high efficiency and better sensitivity at high energies. The disadvantage of using large volume detectors is that they have significantly poorer energy resolution compared with the small planar detectors. Although BEGe detectors may have an energy resolution of less than 600 eV at 122 keV, which is still comparable with Low Energy resolution Germanium LEGe detectors [8], MGAU can be used to analyse such spectra and the code retrieves the initial energy calibration directly from the spectrum file or uses the user input. On the other hand, PC/FRAM, uses the full energy range, and it does not have strict requirements for high detector resolution [9]. In this work, the influence of detector resolution on the estimation of uranium isotopic using MGAU, MGA and PC/FRAM has been studied.

2. Material and Equipment

In this study, counting systems with different detector types (LEGe, BEGe or COAX) and consequently, energy resolution was used. Table 1 shows summary of the system information.

Table 1: Characteristic of the used gamma spectroscopy systems

System No.	Detector Type	Manufacturer	Resolution at 122 (eV)
Detector (1)	Planer	ORTEC	550
Detector (2)	Planer	Canberra	650
Detector (3)	Coaxial	Canberra	939
Detector (4)	Coaxial	ORTEC	1450

In this study, a set of Uranium Standard Reference Materials (SRM 969) of nominal abundances 0.31%, 0.71%, 2.96% and 4.46% of ²³⁵U were used for measurements. Each sample contains 200.1±0.2 gram of U₃O₈ powder, encased in aluminum cylindrical containers [10]. The dimension of Al cans is 70 mm height and 20.8 mm diameter. Table 2 shows the characteristic of the used samples.

Table 2: Description of SNM-969 samples used in the measurement [10].

Sample ID	Full High (cm)	Density g/cm ³	Certified /Declared values of Uranium				
			U ₃ O ₈		Reference relative abundance		
			Weight (g)	²³⁵ U Weight (g)	²³⁵ U (atom%)	²³⁸ U (atom%)	²³⁴ 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

3. Measurements

Each sample was measured three times at 10 cm distance from the end cap of the detector for measuring time of duration one hour with each detector. The obtained uranium spectra were analyzed using MGAU version 4.2, PC/FRAM version 5.2 and U235 version 1.06. Figure 1 shows the configuration of the experimental set up.

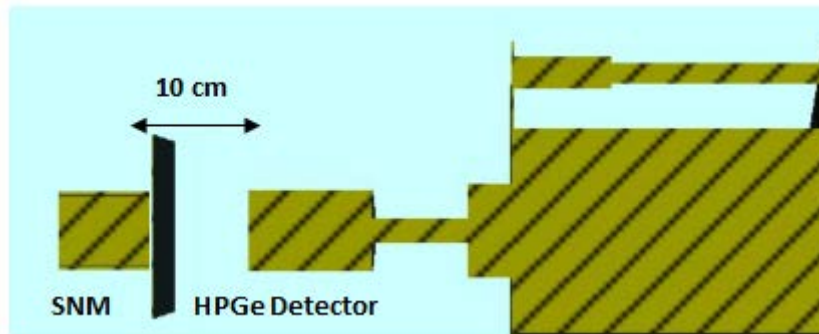


Figure 1: Experimental setup configuration.

4. Results and Discussion

The normalized measured/certified values (M/C) were used to monitor the performance of each code. Figure 2 shows the effect on accuracies of calculated enrichment using MGAU, PC/FRAM and MGA codes for systems with resolutions below and above 750 eV at 122 keV. For the three used codes, the accuracy of calculated enrichments was much better using detectors with resolution below 750 eV than with resolutions above 750 eV. Although one of the features of MGAU code is the ability to analyze gamma-ray spectra collected using both

high resolution planar detector and high efficiency coaxial detector, still accuracies obtained at resolution 1450 eV were affected more than those obtained by MGAU and PC/FRAM code. At 1450 eV resolution, for the four measured enrichments in ascending order, the (M/C) calculated percentages based on MGAU, MGA and PC/FRAM were [36, 18, 4, 2, 0.4%], [28, 12, 8, 4, 3%] and [6, 7, 4, 5, 0.9 %] respectively

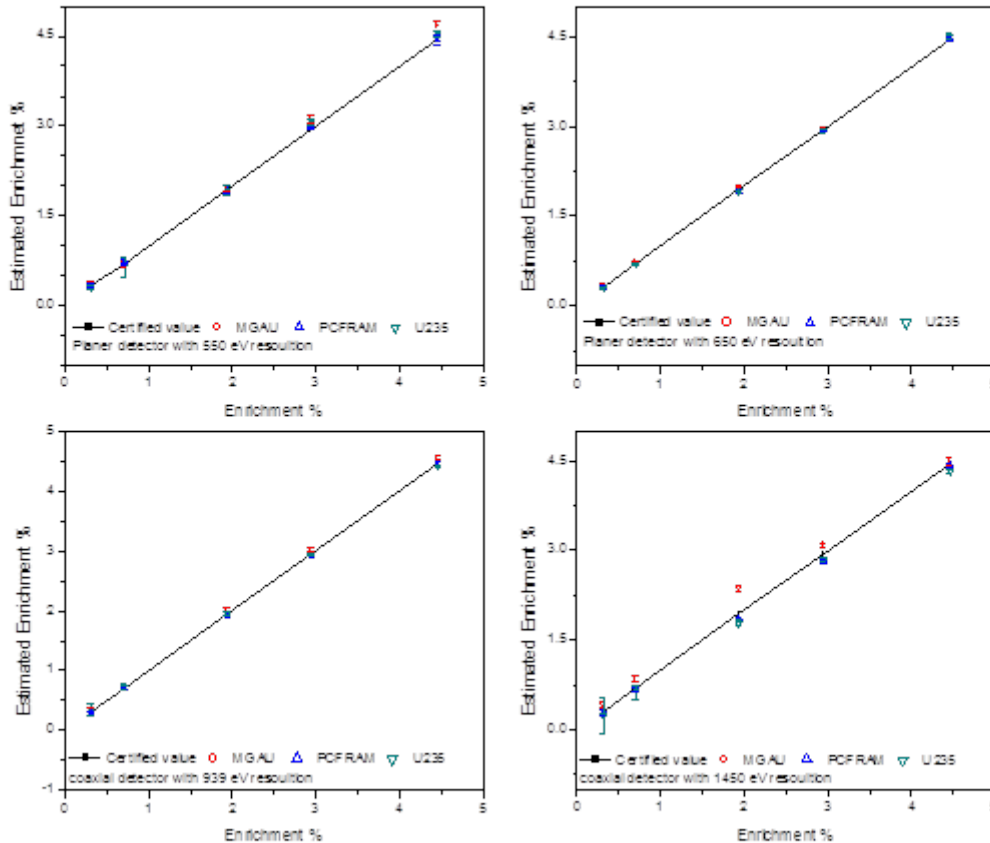


Figure 2: Calculated enrichment using MGAU, PC/FRAM and MGA codes using different detectors with different resolutions.

As seen in Figure 3, enrichment of measured samples had a pronounced impact on the obtained accuracies. The performance at each resolution, improved as the enrichment increased. At low enrichments, the (M/C) percentages obtained by MGAU, MGA and PC/FRAM were 30 %, 28 % and 6 % respectively. On average the most consistent performance expressed in (M/C) values over different resolutions and range of enrichments was obtained by PC/FRAM. At enrichment 4.46%, the calculated (M/C) percentages at 550 eV, 650 eV, 939 eV and 1450 eV were 0.4, 0, 0 and 0.9.

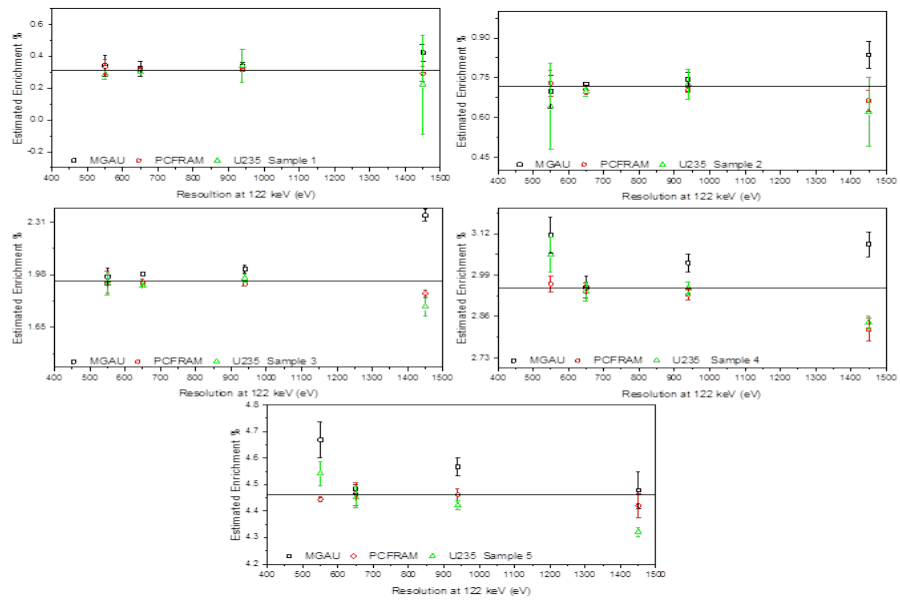


Figure 3: Calculated biases at different enrichment using MGAU, PC/FRAM and MGA code

One of the features of MGAU is the ability to analyze gamma-ray spectra collected using both a high resolution planar Ge detector and a high efficiency coaxial Ge detector. Figure 4 shows a comparison of enrichment calculated using data collected with high efficiency coaxial detector below and above 300 keV. Analysis using spectrum above 300 keV showed slight deviation from certified values at 3% enrichment while spectrum below 300 keV showed deviation at 0.71%.

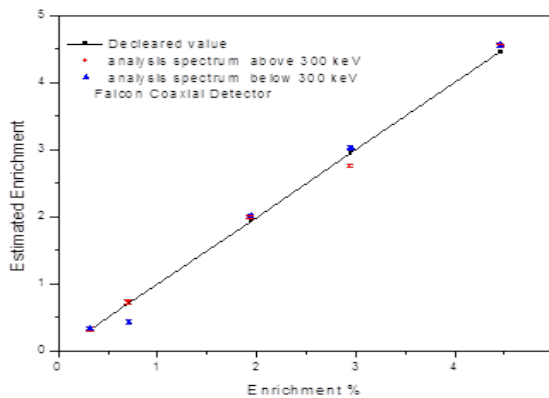


Figure 4: Obtained enrichment using MGAU and two energy segment spectral analysis

5. Conclusion

It is recommended to use MGAU, PC/FRAM and MGA codes with systems with resolutions below 750 eV at 122 keV. For measuring systems with resolution above 1450 eV, up to 30% bias may be expected. The performance of the three codes showed proportional relation to measured enrichments. PC/FRAM analysis provided the best consistency along the studied resolution and enrichment ranges with biases ranging from 0 to 7%. MGAU and MGA showed more sensitivity towards low resolution detectors especially at lower enrichments.

References

- [1] Morel, J., Etcheverry, M., Riazuele, G., 1988. Uranium enrichment measurement by X- and g-ray spectrometry with the URADOS process. *Appl. Radiat. Isot.* 49 (9–11), 1251.
- [2] Morel, J., Hill, C., Bickel, M., Alonso-Munoz, A., Napier, S., Tharel, B., 2000. Results from the international evaluation exercise for uranium enrichment measurements. *Appl. Radiat. Isot.* 52, 509.
- [3] Marcos S. Grund and Fábio C Dias, APPLICATION OF THE MGAU CODE FOR MEASURING 235U ENRICHMENT AT THE BRAZILIAN SAFEGUARDS LABORATORY, 2009 International Nuclear Atlantic Conference - INAC 2009 Rio de Janeiro, RJ, Brazil, September 27 to October 2, 2009
- [4] Reilly D, Ensslin N, Smith H, *Passive Nondestructive Assay of Nuclear Materials*, USA, LANL, 1991, 218–234.
- [5] P McClelland, V Lewis. *Radiometric Non-destructive Assay*. UKAEA, Dounreay Vic Lewis National Physical Laboratory, 2003, 23–45.
- [6] A. Bosko, S. Croft, S. Philips, R. Gunnink, Impact of the detector resolution on the performance of MGA and MGAU isotopic codes, *J Radioanal Nucl Chem*, Vol 282, No3 (2009) 282:855–860
- [7] Multi-Group Analysis for Uranium manual, MGAU-SS-C39051
- [8] A. Bosko Æ S. Croft Æ S. Philips Æ R. Gunnink, Impact of the detector energy resolution on the performance of MGA and MGAU isotopic codes, *J Radioanal Nucl Chem* (2009) 282:855–860
- [9] A. A. Solodov, S. E. Smith and J. S. Bogard, 2006, Uranium Isotopic and Quantitative Analysis Using a Mechanically-Cooled HPGe Detector, ORNL/TM-2006/150
- [10] B.S. Carpenter, J.W. Gramlich, R.R. Greenberg, L.A. Machlan, P. DeBievre, H.L. Eschbach, H. Meyer, J. Van Audenhove, V.E. Connolly, N.M. Trahey, A.C. Zook, Uranium-235 Isotope Abundance Standard Reference Materials for Gamma Spectrometry Measurements, NBS SPECIAL PUBLICATION 260-96, Issued September 1986.