DEPENDENCE OF THE MGA CODE PERFORMANCE ON DETECTOR ENERGY RESOLUTION

A. Bosko and S. Croft
Canberra Industries Inc.
800 Research Parkway, Meriden, CT, 06450, USA

ABSTRACT

Nondestructive measurements of γ-ray and X-ray emissions are often made to characterize special nuclear materials. Various computer codes are available to determine the relative isotopic composition of Pu from analysis of the spectra resulting from such measurements. MGA is one of the major isotopic codes and when possible it makes use of the low-energy region below 208 keV. A *good* analysis of the low-energy region of a spectrum requires a high-resolution germanium detector. A typical Ge detector used for these measurements would be 200 mm^2 in area by 10 to 16 mm deep. Small detectors of this type can provide an excellent resolution of about 500 eV full width half-maximum (FWHM) at 122 keV. Because of the complexity of the important 100 keV region from Pu, it is quite important that the resolution meet certain standards. Historically the suggested FWHM at 122 keV has been 525 eV or better, however in many cases, for example when using large detectors or when a short amplifier time constants are needed, this specification can not be met. The purpose of this study was to investigate MGA performance versus energy resolution of the counting system. A set of Pu standards with different burn-up was measured with several types of detectors having a wide range of energy resolution at 122 keV. The detector set included examples of – the Low-Energy Germanium Detector (LEGe), the Broad-Energy Germanium Detector (BEGe), the Reverse-Electrode Coaxial Germanium Detector (REGe), and the conventional coaxial germanium detector. The effects of a poor resolution on MGA performance were analyzed and are discussed.

INTRODUCTION

The Multiple Group Analysis (MGA) code was developed by Ray Gunnink and co-workers at the Lawrence Livermore National Laboratory, for use in the safeguards community using small volume planar Low Energy Germanium (LEGe) detectors [1]. Due to their low sensitivity and poor efficiency at high energies LEGe detectors are ineffective for some types of measurements e.g. radioactive waste characterization where a broad energy coverage is needed to include isotopes emitting high energy gamma rays. In this case different detector types (e.g. COAX, REGe, BEGe) are often used [2]. In particular the development of large volume Broad Energy Germanium (BEGe) detectors has given rise to increasing use of MGA, including the low energy analysis, with large volume detectors in the arena of nuclear waste assay. BEGe detectors have a resolution at low energies comparable to that of LEGe detectors, while at high energies the behavior is comparable to that of good quality coaxial detectors. The aspect ratio (large area) and large germanium volume make the BEGe a good choice for the efficient detection of special nuclear materials.

MGA uses the most intense peaks of a spectrum – the 94-104 keV region and the 129 and 148 keV peaks – when possible. The 100 keV region contains about 15 strongly overlapping peaks, so having the counting system with good energy resolution is considered vital for producing a reliable

results during isotopic analysis. Traditionally it was recommended when using MGA to select a detector with a full width half-maximum (FWHM) resolution at 122 keV of less than 525 eV. However in many cases when using large detectors or when using a short amplifier time constant this ideal can not be met. The purpose of this work was to experimentally examine to what extent the detector resolution impacts the results generated by MGA.

EXPERIMENTAL SETUP

Ten different detectors were used in this study, each having a different energy resolution. A summary of the detector information can be found in Table 1. A set of Pu isotopic standards (Certified Reference Materials) identified as CRM 136, 137 and 138, was measured using different counting conditions and geometries. Each standard consists of about 0.25 grams of Pu, in the form of plutonium sulfate tetrahydrate, contained in a glass microbottle housed in a sealed can [3]. The isotopic composition of the standards is given in Table 2. The ^{241}Am weight fraction is not certified but has been estimated from the approximate date of chemical separation [4].

Table 1 Detector information

Detector Type	Detector ID	Detector Model	Resolution at 122 keV, eV
BEGe	BEGe - 1	BE3825	583
BEGe	BEGe - 2	BE3830	576
BEGe	BEGe - 3	BE5030	579
LEGe	LEGe - 1	GL0510P	514
LEGe	LEGe - 2	GL0110R	583
COAX	COAX - 1	GC4020	1194
COAX	COAX - 2	GC4020	930
COAX	COAX - 3	GC2020	899
REGe	REGe - 1	GR4520	1059
REGe	REGe - 2	GR1318	1026

Table 2 Isotopic composition of the plutonium samples (^{241}Am is quoted relative to total Pu).

	Weight percent of Pu and Am-241 as of February 28, 2007					
	Pu-238	Pu-239	Pu-240	Pu-241	Pu-242	Am-241
CRM136	0.193	85.962	12.498	0.754	0.592	3.584
	±0.007	±0.015	±0.015	±0.005	±0.003	±0.0123
CRM137	0.232	78.636	19.027	0.861	1.243	3.848
	±0.005	±0.022	±0.022	±0.006	±0.004	±0.0137
CRM138	0.0086	91.913	7.955	0.090	0.0335	0.557
	±0.001	±0.010	±0.010	±0.001	±0.0003	±0.0017

Each sample was measured five times, and each time a spectrum was acquired under certain counting conditions, such as:

- the sample-to-detector distance varied for different samples, so that the dead-time was kept the same and at about 5%

- the counting time varied between 140 – 600 seconds depending on the detector type and the measured sample in order to acquire about 60,000 counts in the 100 keV region for each spectrum
- all the measurements described here were performed using detectors covered with 2 mm tin and 0.5 mm copper filters to suppress the 60 keV radiation from ^{241}Am

DATA ANALYSIS

MGA ver.9.63H was used for the analyses presented here. All spectra were analyzed and the results for each of them are presented on Figure 1 and Figure 2. We have chosen to consider the ^{239}Pu weight fraction and the $^{240}Pu_{eff}$ weight fraction as the key figure of merit for waste applications. This is because the fissile mass often used as a fiducial line from which other nuclides are derived by correlation and is also important for criticality and other reasons while the $^{240}Pu_{eff}$ abundance is used to interpret passive neutron coincidence counting measurements.

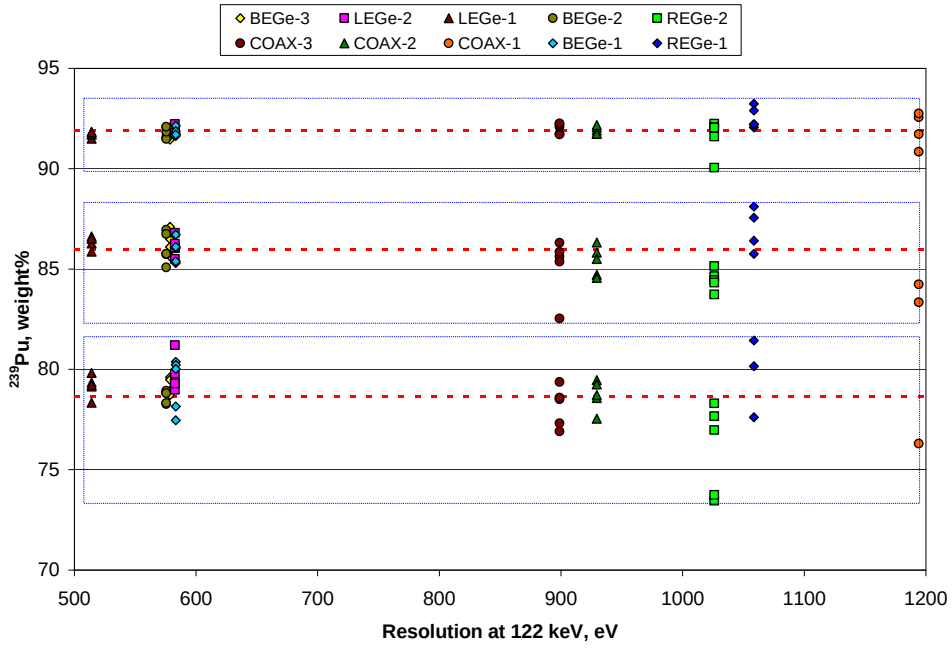

Figure 1 ^{239}Pu weight fractions as a function of FWHM. The red dotted lines represent declared values while the blue dotted boxes group the results obtained for different samples. Note the vertical scale is linear so that the relative tightness of the clusters can be readily appreciated.

It may be noted that the results tend to exhibit greater scatter around the declared value as the FWHM of the counting system increased. When the FWHM at 122 keV exceeded approximately 1050 eV, MGA began to fail to produce any results.

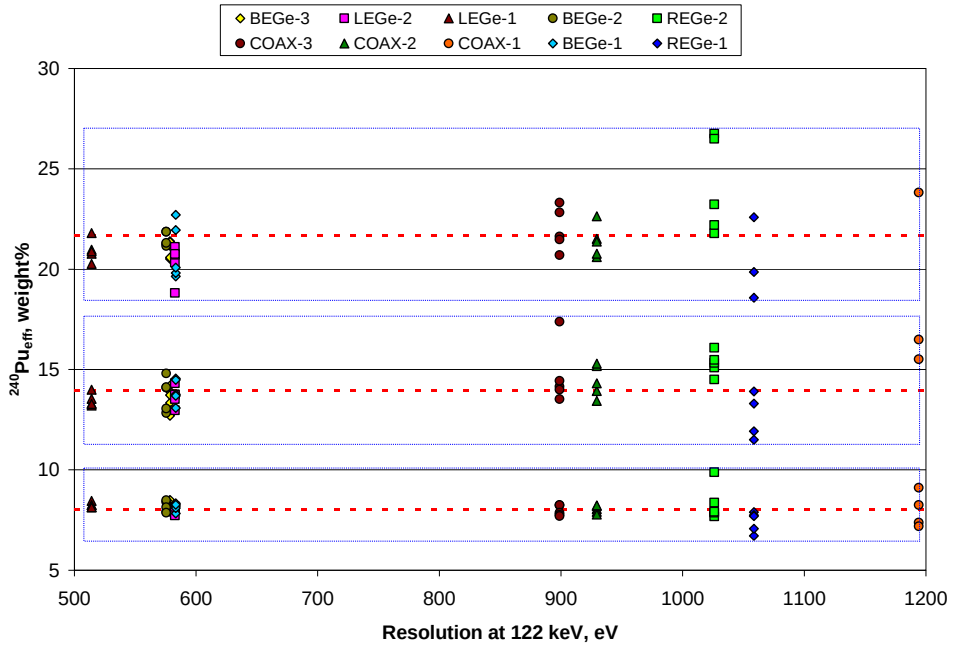

Figure 2 $^{240}Pu_{eff}$ weight fractions vs FWHM. The red dotted lines represent declared values, blue dotted boxes group the results obtained for different samples.

MGA performed least well when analyzing spectra taken with high burn-up samples. Examples of MGA fits in the 100 keV region for two different detectors with high and low energy resolution are given on Figure 3. The coaxial detector had a resolution of 1194 eV at 122 keV, while LEGe had 514 eV. It may be noted that especially with the coaxial detector the 104 keV line from ^{240}Pu, which is a key line for this isotope, cannot be reliably resolved from ^{241}Am gamma lines.

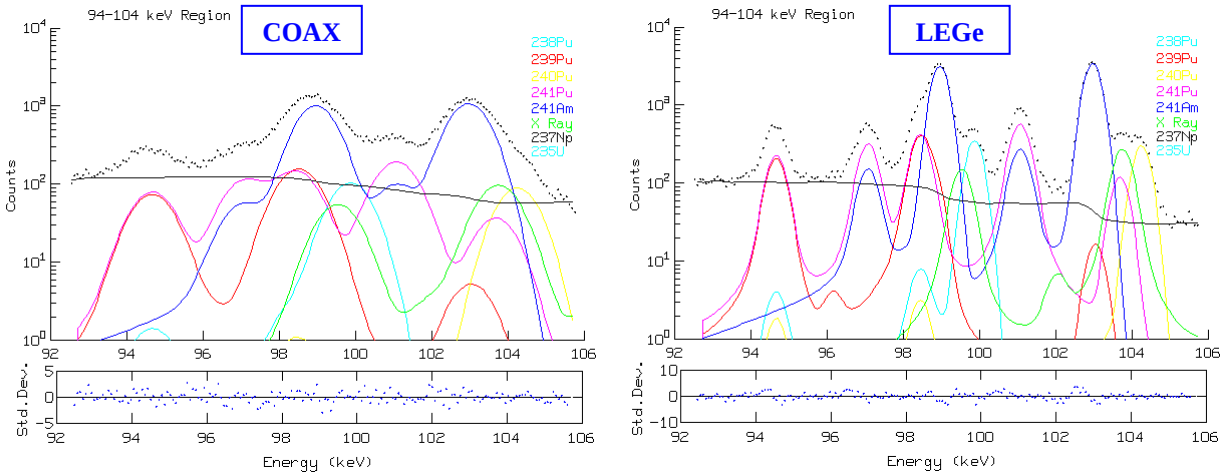

Figure 3 MGAver.9.63H fits in the 100 keV region.

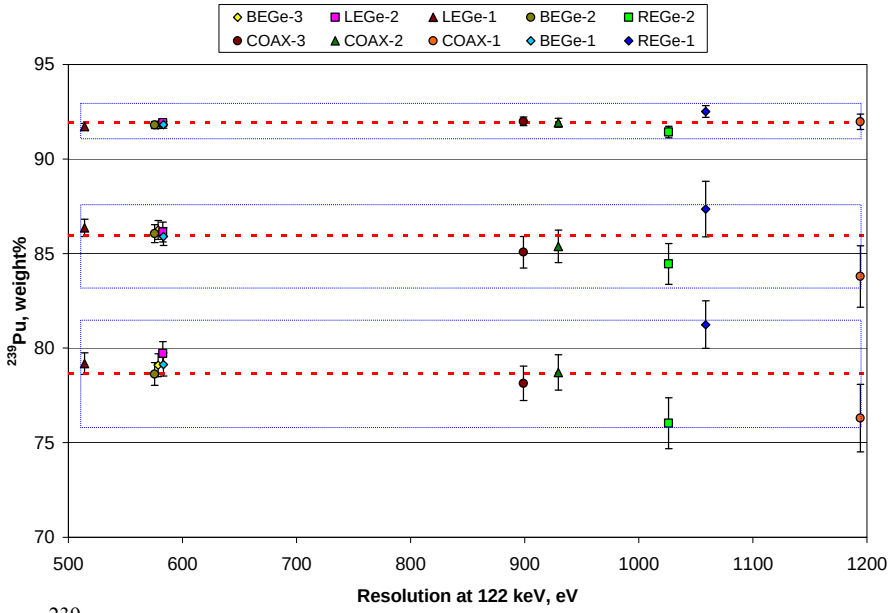

Figure 4 Average ^{239}Pu weight fractions measured by each detector plotted against resolution. The error bars represent an average of the uncertainty reported by the code for each sample.

Figures 4 and 5 show the average values reported by MGA for ^{239}Pu and ^{240}Pu$_{eff}$ for each detector. As it is shown on these plots, while all the spectra had approximately the same counting statistics the reported uncertainty for the results grows as the resolution of the system decreases. The reported uncertainty ranged from about 0.18% for LEGe detector and the low-burn-up sample, and up to about 2.4% for the coaxial detector measuring the high burn-up sample. The differences from the expected values are as expected from the quoted measurement uncertainties, so there was no particular evidence for bias.

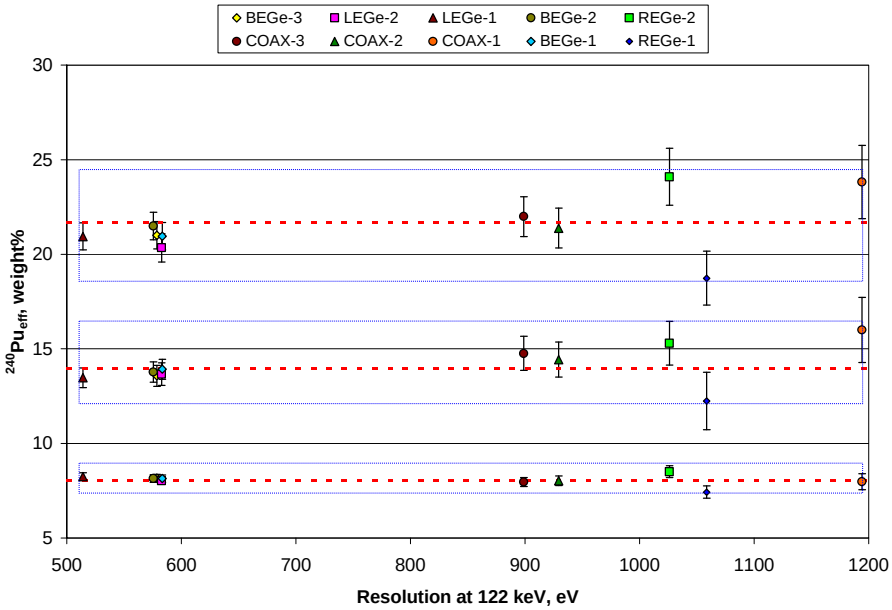

Figure 5 Average ^{240}Pu$_{eff}$ weight fractions measured by each detector (error bars represent an average uncertainty reported for each sample).

CONCLUSIONS

Three plutonium samples of different levels of burn-up have been used in a practical study to directly assess the dependence of the performance of the 9.63H version of the MGA code on detector energy resolution. The plutonium items were assayed using 10 detectors of different types, and energy resolution of those varied from 514 eV to 1194 eV at 122 keV. Although the recommended value for energy resolution is less than 525 eV at 122 keV there was no significant difference found in the results for systems with resolution up to about 600 eV. This work also showed systems with resolution of about 900 eV at 122 keV can still be used for isotopic assays although the measurement uncertainties may strongly depend on sample isotopic composition. The better results, judged against known values, are usually achieved for low burn-up samples. Experimental measurements also showed that MGA version 9.63H starts failing to produce results when the FWHM at 122 keV exceeds about 1050 eV. These occurrences would usually be subjected to subject matter expert review. Additional work remains to be done to cover the missing region in the detector energy resolution from 600 eV to 900 eV. Our study was also limited to only three compositions. We note that for waste applications the measurement objectives may be less stringent than is the case for classical safeguarding of bulk materials and the creation of balance accounts. This is was the primary driver for investigating larger volume detectors. Overall we are impressed by how robust MGA version 9.63H performed outside of the original detector envelope envisioned by the designers. We note also that MGA is a living code and that these results need to be re-verified on newer versions as they enter service and also should be extended to help in the refinement of such versions.

REFERENCES

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3. R. Venkataraman and S. Croft, “Determination of plutonium mass using gamma-ray spectrometry”, Nucl. Inst. and Meth. A505 (2003) 527-530.
4. R. Venkataraman and S. Croft, “Determination of plutonium mass using gamma ray spectrometry”, Canberra Industries Internal Report, April 2002.