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MGA++ Studies of U/Pu Gamma-ray Standards with two Falcon 5000 Mechanical cooled HPGe Detection Systems

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Abstract

With recent advancements in the mechanical cooled HPGe detection system, it is possible to perform plutonium and uranium gamma-ray analysis with isotopic analysis codes such as MGA++. In this study, we used two Falcon 5000 mechanical cooled HPGe systems (with 120-keV energy resolution of ~820eV and ~ 920eV, respectively) as well as a safeguards type liquid nitrogen cooled HPGe (energy resolution ~ 650eV) detection system. A wide range of uranium and plutonium gamma-ray standards were used. The goal of this study is to understand the limitations/biases of MGA++ when applying directly to the new detection systems that have energy resolution slightly outside the recommended energy resolution (< 600 eV) specified by the MGA++.

Introduction

Uranium and plutonium singles gamma-ray non-destructive assay has been proven to provide a measure of an accurate broad range uranium/plutonium isotopic content for safeguards. Codes such as MGA++ [1] and FRAM [2] have been developed and are widely used in the safeguards community using an HPGe gamma-ray detector system for this sole purpose.
The MGA++ code developed at Lawrence Livermore National Laboratory typically uses the low energy part (<300-keV) of the gamma-ray information from both uranium and plutonium that can be obtained with a planer detector with good energy resolution (<600eV @ 120-keV), so that robust unfolding of complicated 100-keV region for both uranium and plutonium gamma-ray spectra is possible. The unfolding mechanisms employed in the code for plutonium and uranium isotopic contents are shown in Fig, 1(a) and 1(b), respectively. The dominant gamma-ray groups, for example, used in the unfolding of plutonium gamma-rays are listed in Table 1. Since the uranium analysis relies on the daughter gamma-rays of U-238, either secular equilibrium between the U-238 parent and Pa-234 daughters or known separation time is required. In general, for pure and homogeneous samples, MGA++ analysis provides good results.

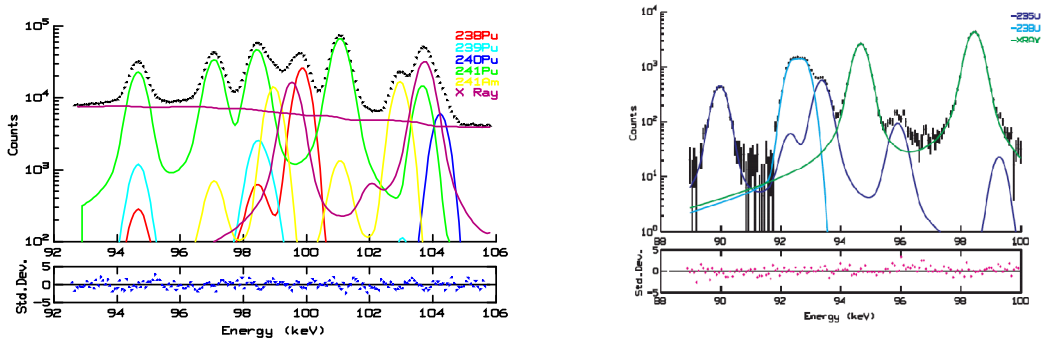

Fig. 1 (a, left) MGA++ unfolding of the complicated 100-keV plutonium gamma-ray region, and (b, right) unfolding of 100-keV uranium gamm-ray region.

Energy(keV)	Pu-239	Pu-240	Pu-241	Am-241	X-ray	Np-237
86.5	0	0	0	0	0	0.14
94.66	4.20E-05	6.80E-07	2.98E-06	0	0	0.0931
95.858	0	0	0	0	0	0.029
96.16	3.68E-07	0	0	0	0	0
97.071	0	0	3.80E-06	1.20E-05	0	0
98.443	6.82E-05	1.00E-06	4.84E-06	0	0	0.14
98.79	1.22E-05	0	0	0	0	0
98.956	0	0	0	2.00E-04	0	0
99.533	0	0	0	0	4.14E-05	0
99.864	0	0	0	0	0	0
101.063	0	0	6.12E-06	1.94E-05	0	0
102.963	0	0	0	1.93E-04	0	0
103.035	2.12E-06	0	0	0	0	0
103.679	0	0	9.85E-07	0	0	0
103.742	0	0	0	0	6.64E-05	0
103.86	0	0	0	0	0	0.0083
104.242	0	6.97E-05	0	0	0	0

Table 1 Gamma-ray branching ratios used in MGA++ for 100-keV plutonium unfolding.

In this study, we used two CANBERRA [3] Falcon 5000 (Fig. 2(a))mechanical cooled HPGe systems (120-keV energy resolution of ~820eV and ~ 920eV, respectively) and a safeguards type (ORTEC[4] SGD-GEM-5030P4, Fig. 2(b)) liquid nitrogen cooled HPGe (energy resolution ~ 650eV @ 120-keV) detection system. The goal of this study is to understand the limitations/biases of MGA++ when applying directly to the detection systems that have energy resolution slightly outside (50 – 350 eV) the recommended (< 600 eV) energy resolution specified by the MGA++.

Fig. (2a, left) One of the CANBERRA Falcon 5000 mechanical cooled HPGe detection systems used in this study, (2b, right) ORTEC safeguards type liquid nitrogen cooled HPGe detection system

Experimental Setup and Results

A set of twelve one-gram uranium gamma-ray standards (ranging from depleted uranium (DU) to highly enriched uranium (HEU)) from National Bureau of Standards (NBS) was used in this study along with six plutonium gamma-ray standards from the plutonium inter-comparison exercise (PIDIE) [4]. Uranium sources were placed 1" in from of the detection systems with a 1200s dumping sequence. For the Falcon systems, associated internal electronics were used. For the safeguards HPGE system an integrated DSPECPro electronics was used. Cadmium absorbers of thicknesses ranging from 40 – 70 milli-inch were placed in between the six PIDIE standards (PIDIE #2 - #7) and the detection systems to attenuated the strong 59-keV gamma-rays from the decay of Am-241. The same 1200s dumping sequence was used as in plutonium data collection and the source-detector distance was 2-inch. A total of 1200 spectra were collected and more than 400 were analyzed. The complete analysis results and spectra (with spectra conversions from a CANBERRA proprietary format to a standard format) will be released as an internal [6] report.

Fig. 3 shows a summary of the MGA++ analysis of the three detection system with reported U-235 enrichment values deviated from the reference values. Seven of the 1200 sec data for each NBS uranium reference were analyzed that the averaged values are used. As expected from the algorithms used in the MGA++ analysis, the better resolution results in better agreement with the reference values.

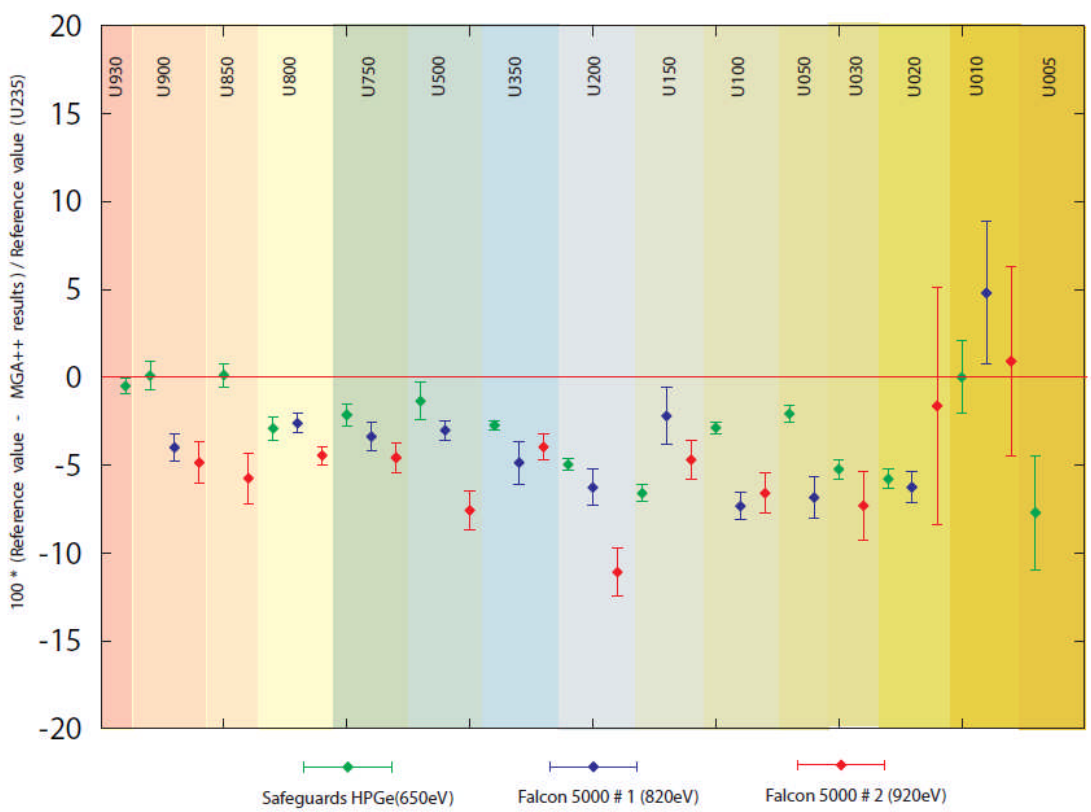

Fig. 3 Percent deviation from the reference values for the three systems with respect to various U-235 enrichments

Seven 1200 s plutonium spectra were analyzed for each PIDIE standard and the averaged Pu-239 results from MGA++ are shown in Fig. 4. For low burn-up plutonium standards, the three detection systems do not show significant departure from the reference values (and in good agreements among themselves). The higher the burn-ups, the more difficult it is to extract Pu-239 information when Pu-241 content is high (as expected from the MGA++ 100-keV unfolding) and this effect will get amplified with poorer energy resolution of the detection systems. It is interesting to note that for the plutonium study, the two Falcon systems show almost identical Pu-239 values.

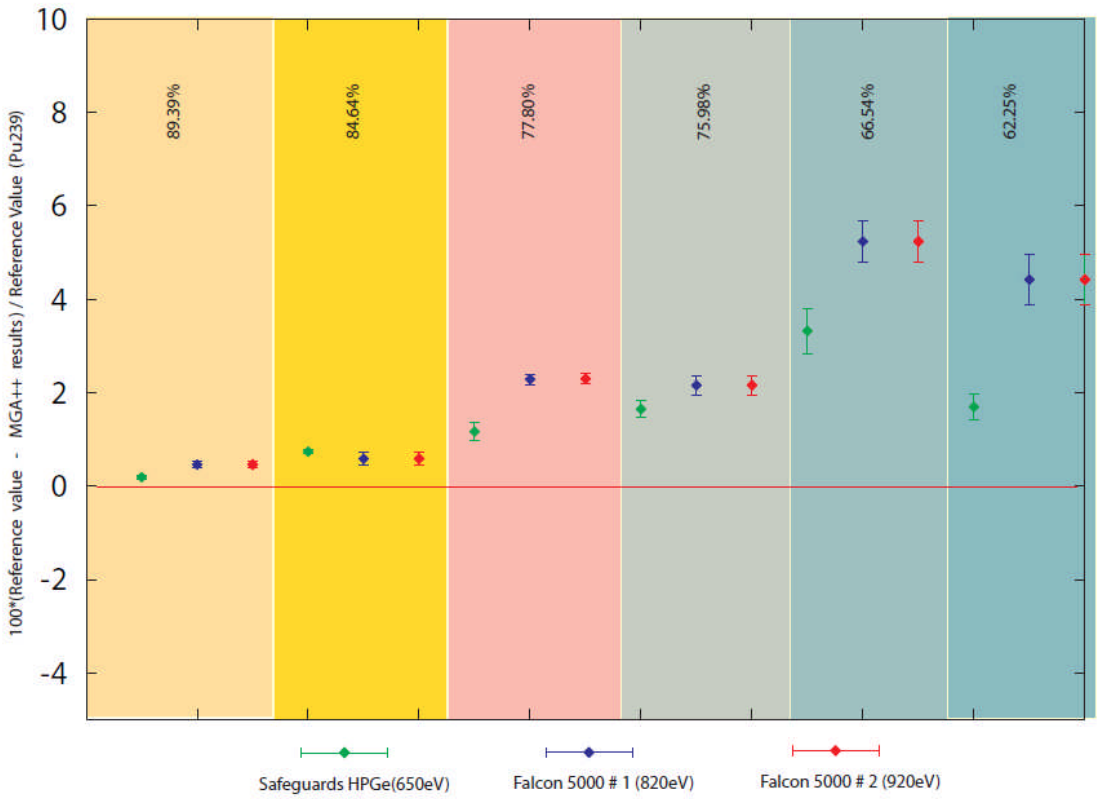

Fig. 4 Deviations to the Pu-239 reference values from MGA++ analysis for the three systems

Conclusions:

From this study without absorbers, slight departing (50 – 350 eV) from the required MGA++ resolutions at 120-keV resulted in 5-10% deviations in U-235 for some of uranium samples, and 2-6% deviation in Pu-239 for higher burn-up plutonium samples. Since all these new detection systems have quite good detection efficiency (15% - 20% @ 1.33 MeV) for high energy uranium and plutonium gamma-rays, we are currently working on using higher energy gamma-ray information to compensate deficiencies in the 100-keV MGA++ unfolding algorithms. We are also working on using the recently modernized PC-GAMANAL [7] as potential isotopic analysis software to use with mechanical cooled HPGe detection systems for field measurements.

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