Preprint
UCRL-JC-137275

Full Range MGA Plutonium Isotopic Analysis Using Single Ge Detector

W.M. Buckley, T.F. Wang, A. Friensehner, S.A. Kreek, R.G. Lanier, W.E.Parker, W.D. Ruhter, T. Twomey, D. Martinez, R. Keyser, P. Sangsingkeow

This article was submitted to
41st Annual Meeting of the Institute of Nuclear Materials Management
New Orleans, LA, July 16-20, 2000

June 26, 2000

U.S. Department of Energy

Lawrence Livermore National Laboratory

Full Range MGA Plutonium Isotopic Analysis Using Single Ge Detector

W.M Buckley, T.F. Wang, A. Friensehner, S.A. Kreek, R.G. Lanier, W.E. Parker, and W.D. Ruhter
Lawrence Livermore National Laboratory, Livermore, CA 94550
and
T.Twomey, D. Martinez, R Keyser and P. Sangsingkeow
PerkinElmer Instruments, ORTEC Division, Oak Ridge TN 37830

Abstract

The gamma-ray multi-group analysis code MGA developed at Lawrence Livermore National Laboratory has been widely used in the area of gamma-ray non-destructive plutonium assay. This plutonium isotopic analysis code de-convolutes the complicated, 100-keV x ray and gamma ray region to obtain the ratio of Pu isotopes. Calibration of the detector efficiency is not required, but is determined intrinsically from the measured spectra. The code can either analyze low-energy gamma-ray spectrum taken using a high-resolution HPGe detector for energies below 300 keV, or analyze the low-energy spectrum combined with a high-energy spectrum (up to 1 MeV) in the "two-detector" analysis mode. In the latter case, the use of two detectors has been mandated by the conflicting requirements: excellent resolution at low energies (characteristic of small planar detectors) with good high-energy efficiency (characteristic of coaxial detectors). Usually, a high-energy spectrum taken using a coaxial Ge detector will not provide sufficient energy resolution for 100-keV plutonium isotopic analysis, while the small planar used at low energies has inadequate high-energy efficiency.

An optimized-geometry ORTEC HPGe detector has been developed which combines good energy resolution at 100 keV combined with acceptable high-energy (~1MeV) efficiency in a single detector. It has been used to gather spectra of both low- and high-energy regions of plutonium spectra simultaneously, for analysis by MGA in the "two-detector" mode. Five Pu gamma-ray calibration standard sources were used in this study of this special detector.

Introduction

The isotopic composition of a plutonium sample is used to calculate Pu-240 effective or specific-power that are used in conjunction with passive neutron coincidence or calorimetry measurements respectively to obtain absolute plutonium mass. MGA[1,2,3] can determine plutonium isotopic abundance with accuracy better than 1% using a high-resolution planar germanium detector in a few minutes of counting time.

A less known feature in MGA is the ability to analyze gamma-ray spectra collected using both a high resolution planar Ge detector and a high efficiency coaxial Ge detector. In general, MGA obtains the isotopic information using the data collected with the high-resolution planar detector (below 300 keV). A spectrum de-convolution plot of this region is shown in Figure 1. However, refined isotopic results and additional isotopic information can be obtained from using higher-energy gamma rays (above 300 keV)

collected with a high efficiency coaxial detector. For example, the ^{238}U abundance can be obtained from the 1001 keV peak, a more accurate analysis for ^{237}Np can be made, some fission products can be identified, and ^{241}Am inhomogeneities in the sample can also be determined. The two-detector mode of MGA is the only way to provide homogeneity information for a sample. It is worth noting that these two sets of data do not have to be collected at the same time under the same geometry. To properly use the higher energy gamma-ray information, a separate intrinsic efficiency curve must be determined, as shown in Fig. 2. Like the low-energy curve in Fig. 3, the components of the efficiency are based on the physical processes involved in attenuating and detecting the gamma rays. In general, there is a thin Pb absorber in front of the coaxial detector to reduce the count rate due to the strong 100-keV region gamma rays.

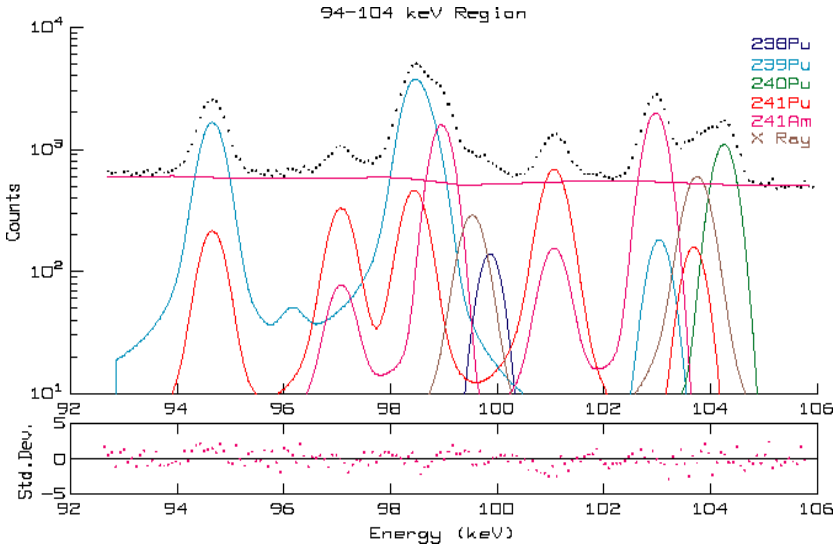

Figure 1. MGA analyzes the complicated 100-keV energy region to obtain plutonium-^{241}Am isotopic information

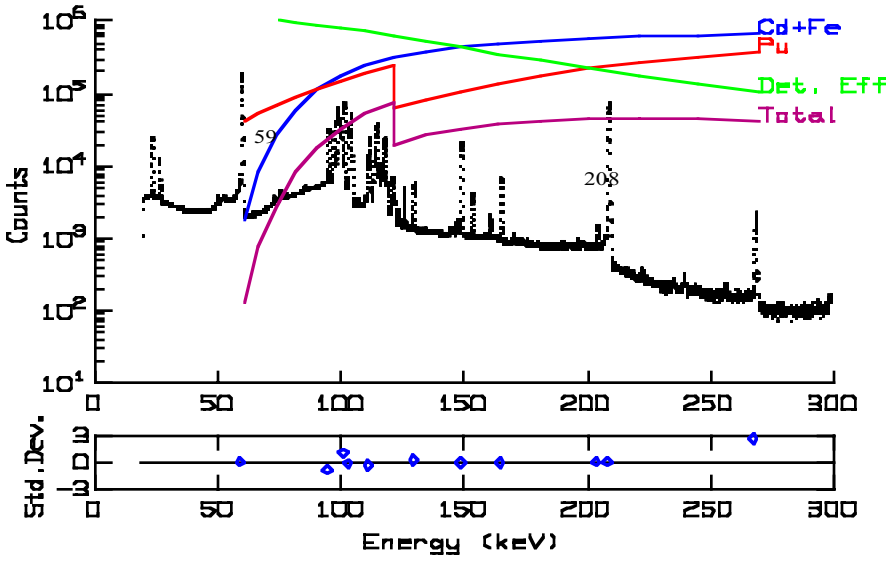

Figure 2. MGA uses physical attenuation corrections of both emission and absorption in the gamma ray interactions. This plot shows the three principal processes that characterize the low-energy "intrinsic" efficiency curve.

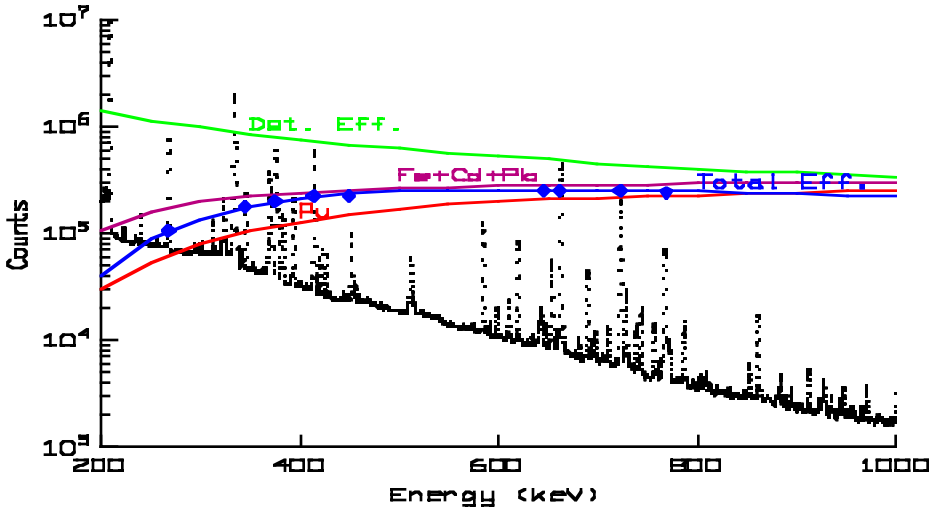

Figure 3. The same physical attenuation processes are used in analyzing the second detector data when running MGA in the two-detector mode. This plot shows the components affecting the efficiency curve.

Prior to the introduction of the new ORTEC HPGe detector, the two-detector mode of the MGA could be used only with two separate Ge detectors having appropriate characteristics. They could be two separate Ge detectors or two Ge crystals mounted in a tandem form within a single enclosure. The reasons are simple: MGA in the two-detector

mode requires high resolution for the low-energy gamma rays and high efficiency for the high-energy gamma rays. The planar Ge will not provide sufficient efficiency for high energy, the coaxial Ge normally can not provide sufficient resolution at low energy.

The Detector

Recently, three new ORTEC coaxial safeguard detectors were introduced; their specifications are listed in Table I. These detectors are optimized to have adequate resolution for low-energy gamma rays and good efficiency for high-energy gamma rays. Because of both resolution and dead-time considerations in plutonium/MGA applications, we chose the SGD-GEM-15170 for our test. This detector has the best low energy resolution and possesses reasonable dead time for Pu measurements in a high radiation environment. The detector is nominally 50mm diameter by 30 mm deep. The resolution at 122 keV is about 725eV for a 2μs shaping time at a count rate of 30 kcps. Applying the usual coaxial efficiency definition, the relative efficiency at 1.33MeV is approximately 15% which is not too far away from the MGA suggested 20% coaxial detector efficiency. Figure 4 shows a plutonium gamma-ray spectrum taken with this 15% detector compared to a regular planar LEPS detector.

Table I. Specifications of the three new ORTEC SGD Safeguards Coaxial Detectors

SGD GEM Coaxial Safeguards Detectors						
Model number	Nominal	Nominal	Energy	Warranted Resolution		
	Active Diameter	Thickness (mm)		@1kcps (6μS)	@1kcps (2μS)	@30kcps (2μS)
	(mm)					
SGD-GEM-15170	50	30	122 keV	625eV	675eV	725eV
			1.33 MeV	1.70keV	1.85keV	2.05keV
SGD-GEM-25175	50	50	122keV	750eV	870eV	880eV
			1.33 MeV	1.75keV	1.95keV	2.0keV
SGD-GEM-50180	65	65	122keV	800eV	925eV	950eV
			1.33 MeV	1.80keV	2.05keV	2.15keV
First two digits in model number (e.g., the "25" in SGD-GEM25175) define the nominal relative efficiency, according to the usual IEE standard at 1.33 MeV						
Peak shape at all count rates <= 30KHZ and time constants from 2-6μS, FW0.2M/FWHM <= 2.75 made with ORTEC 672 or DSPEC/DSPEC Plus						

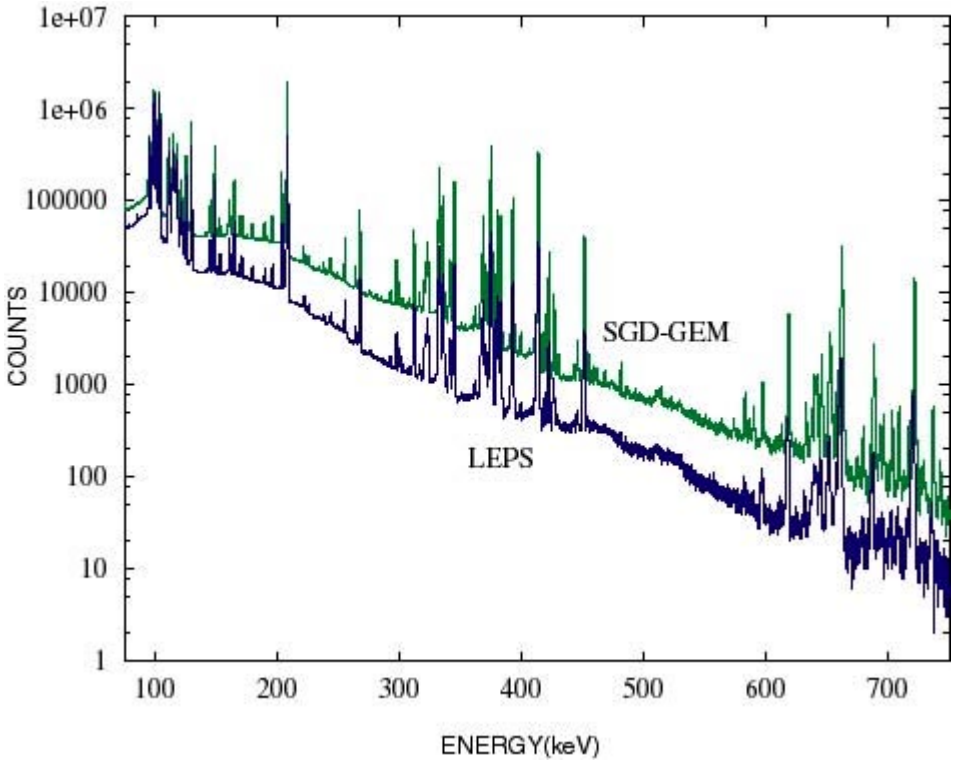

Figure 4. A comparison of plutonium gamma-ray spectra collected using the SGD-GEM detector (TOP) and a planar LEPS detector (Bottom). At 100-keV region, the LEPS has slightly better energy resolution, however, for higher energy (>100 keV) gamma rays, the SGD-GEM has significant higher efficiency. The source was placed at the same distance from both detectors and collected with the same live time.

Experimental Setup

Five Plutonium Isotopic Determination Intercomparison Exercise[4)] (PIDIE) sources were used in this study, these sources have ^{240}Pu enrichment ranging from 6% to 21% and weight about 450mg. Cadmium absorbers ranging from 30 mils to 50 mils in thickness were put between the source and the detector to reduce the count rate due to low energy Pu and 59-keV ^{241}Am gamma rays. Signals were collected using either ORTEC DSPEC or DSPEC^{plus} spectrum analyzers at either 0.1 keV/channel for 16k channels or 0.075 keV/channel for 16k channels. Data were collected for 1 hour, 12 hour, and 1 day. For some tests the sources were place as closed as possible to the detector to simulated high radiation environments as well as to evaluate the performance of DSPEC/DSPEC^{plus} analyzers. Because our sources are in a fixed configuration for a given measurement, there is no effect in the relative isotopic compositions when comparing the spectrum taken with the DSPEC^{plus} operated in the zero-time-corrected mode vs. non-zero-time-corrected mode. The DSPEC and DSPEC^{plus} performance in this relative plutonium isotopic study were found to be almost identical. The result is expected, since the DSPEC^{plus} goes to shorter then 2μS shaping times with finer steps than DSPEC, which, in particular, will

allow to optimize the detector for the highest possible throughput. However, we have to balance throughput and resolution in this study, therefore a 2μS shaping time was used.

MGA analysis and results

In the one detector mode, MGA uses 100-keV region to obtain the Pu isotopic information as well as ^{241}Am/Pu information. The 100-keV region also provides the U/Pu ratio using fluorescence x-rays, the ^{235}U/Pu ratio obtained by using the intensity of the 185 keV gamma rays. In the two-detector mode, the MGA goes even further: the additional ^{241}Am/^{239}Pu ratios can be obtained by analyzing the gamma rays at 300-keV and 600-keV regions; the ^{238}Pu/^{238}U ratio can be obtained using the 766 keV and the 1001 keV gamma rays; the ^{237}Np can be obtained using the 312 keV peak from the decay of the ^{233}Pa (the daughter product of ^{237}Np.) This additional information provides a set of over determined linear equations and is then solved using the least-squares method to obtain the final results. A further check on the presence of uranium is performed using both the information of the uranium fluorescence x-rays and the results from the least square fit. It is also worth noting that the MGA analysis in the two-detector mode can analyze MOX samples with U/Pu ratios ranging from 2 to 150. The first 4k channel of SGD-GEM data were used as the planar detector data for the one-detector mode MGA analysis or as the first detector (i.e., the "LEPS") data in the two-detector mode MGA analysis. The same data (up to 16k channels) were binned to 0.3keV/channl as the second detector (i.e., the "COAX" detector) data for MGA analysis in the two-detector mode. Table 3 shows one of the PIDIE sources results using the SGD-GEM detector in one detector mode, two-detector mode, compare to the mass-spectrometry numbers and a standard LEPS, the resolution at 100 keV for SGD-GEM is about 660 eV and 510 eV for LEPS. Table 3 shows one of the PIDIE results when the source was placed as close to the detector as possible to simulate a high count-rate environment, the 100-keV resolution is about 760 eV.

Table 2. MGA analysis results of PIDIE(#4) when using SGD-GEM data in one-detector modes, two-detector mode. The isotopic composition of the plutonium in PIDIE #4 is listed along with the ^{241}Am/Pu and ^{237}Np/Pu ratios. Low level ^{237}Np has been detected only in the two-detector mode.

	SGD-GEM	SGD-GEM	LEPS	Mass Spectrometry
	One-detector	Two-detector		
Pu238	0.0010871(.59)	0.0010803(.60)	0.0010830(1.5)	0.00108985(1.0)
Pu239	0.7739951(.19)	0.7753632(.19)	0.7765888(.20)	0.77798559(.017)
Pu240	0.1996378(.34)	0.1984970(.32)	0.1973699(.70)	0.19704951(.048)
Pu241	0.0185695(.31)	0.0184373(.25)	0.0183379(.55)	0.01826701(.029)

Pu242	0.00671(10)	0.006622(10)	0.006620(10)	0.5604(.57)
Am241	0.0158217(.47)	0.0157076(.39)	0.0157020(.73)	0.01573(2.0)
Np237		0.000572(1.34)		

Table 3. The MGA analysis results of PIDIE(#2) source using the SGD-GEM detector and a DSPEC^{plus}.The dead time was about 40% during the measurements.

	SGD-GEM	SGD-GEM	Mass Spectrometry
	One-detector	Two-detector	
Pu238	0.002563(2.06)	0.0002536(2.24)	0.000227(3.1)
Pu239	0.889218(.06)	0.8898289(.06)	0.89463(.01)
Pu240	0.1057880(.49)	0.1052120(.52)	0.10071(.088)
Pu241	0.0045840(.31)	0.0045991(.32)	0.004428(.52)
Pu242	0.000154(10)	0.000146(10)	0.000932(2.8)
Am241	0.0027215(.69)	0.0026964(.66)	0.002633(1.2)
Np237		0.000126(1.99)	

Conclusions

Because of its adequate energy resolution at 100-keV region and good efficiency at 1 MeV (~15%), the SGD-GEM detector performs as well as a LEPS detector when using MGA in the one-detector mode for data analysis. The gamma-ray spectrum collected with the SGD-GEM detector contains far better isotopic information (e.g., ^{237}Np, ^{238}U/^{235}U, etc.) than the spectrum collected with a LEPS detector when using MGA in the two-detector mode for data analysis. We have shown that MGA can analyze the data collected with such a detector and a DSPEC type analyzer in either one detector or two-detector mode. We have also shown with such combinations, even for a simulated high radiation environment, it is possible for the SGD-GEM detector to obtain meaningful results. Thus ORTEC SGD-GEM type detector can be a powerful tool when combining the two-detector mode MGA for analyzing U, Pu mixtures, MOX samples, and Pu[5] "PUCKS". The detector can also be used with MGAU/LLNL[6], MGAHI[7] isotopic analysis software for uranium, shielded-plutonium data analysis, respectively.

Acknowledgements

This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under contract No. W-7405-Eng-48. This work is supported by the Office of Safeguards and Security.

References

1) W. E. Parker, T.F. Wang, D. Clark, W. M. Buckley, W. Romine and W. D. Ruhter, *Plutonium and Uranium Isotopic Analysis: Recent Developments of the MGA++ Code Suite,* Proceedings of the Sixth International Meeting on Facilities Operations - Safeguards Interface, pp. 192 - 197, American Nuclear Society, Jackson Hole, Wyoming, September 1999.
2) R. Keyser, T. Twomey, S. Haywood, W. E. Parker, T.F. Wang, D. Clark, K. Raschke, W. Romine, W. Buckley and W. Ruhter, *Recent Developments in the MGA++ Codes,* ESARDA Conference, Seville, Spain, May 1999.
3) R. Gunnink, W.D. Ruhter, *MGA: A Gamma-Ray Spectrum Analysis Code for Determining Plutonium Isotopic Abundances*, Volume I and II, Lawrence Livermore National Laboratory, Livermore, CA., UCRL-LR-103220, (1990)
4) Jean Morel, Bruno Chauvenet, *Intercomparaison des Measures de Composition Isotopique du Plutonium Par Spectrometrie X et Gamma Resultats de L'action "PIDIE" Rapport Final,* Rapport CEA-R-5582, Centre d'Etudes Nucleaires de Saclay, (1992).
5) M. Mitchell, T.F. Wang, D. Pugh, *Nondestructive Evaluation and Assay for the Plutonium Ceramification Test Facility*, American Nuclear Society, San Diego, June 2000.
6) T. F. Wang, W. D. Ruhter, R. G. Lanier, *Re-Measured Uranium Branching Ratios and thier Impact on Removing Biases from MGAU Analyses,* Proceedings of the Sixth International Meeting on Facilities Operations - Safeguards Interface, pp. 220 - 223, American Nuclear Society, Jackson Hole, Wyoming, September 1999.
7) T.F. Wang, K.E. Raschke, W.D. Ruhter, S.A. Kreek, *MGAHI: A Plutonium Gamma-Ray Isotopic Analysis Code for Nondestructive Evaluations*, ANS Transactions, **81**, 234 (1999)