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# MGA Analysis on Elevated $^{238}\text{Pu}$ Samples

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**Abstract.** *Plutonium gamma-ray data analysis, in the 100-keV region, using MGA has been improved to overcome the original maximum limit of 2%  $^{238}\text{Pu}$  relative plutonium content in a sample in order to perform an analysis. MGA analysis results of elevated  $^{238}\text{Pu}$  samples are compared to the results from mass spectrometry.*

## INTRODUCTION

The gamma-ray Multi-Group Analysis code<sup>1,2,3)</sup> (MGA) developed at Lawrence Livermore National Laboratory is widely used in non-destructive isotopic evaluation of plutonium materials, especially for plutonium verification and accountability. The MGA code can analyze gamma-ray data collected using a planar Ge detector or data collected using a coaxial Ge detector, or both. The MGA code analyzes all three important regions of plutonium gamma-rays: the 100-keV, the 300-keV and the 600-keV, depending on the availability of the data in these regions. The 100-keV region contains all the plutonium isotopic information, however, the data in this region can be perturbed (i.e., homogeneity of the sample, shielding of the samples, etc.) due to low energy nature of these gamma-rays; the 300-keV and the 600-keV regions, though less perturbable, each only contains a subset of plutonium isotopic information.

Recently, because of the accelerated aging studies being done on plutonium, there is a need for non-destructive gamma-ray evaluation of elevated (> 2%)  $^{238}\text{Pu}$  samples both in material accountability and material processing. Figure 1 shows the complicated processes<sup>4)</sup> involved in producing elevated  $^{238}\text{Pu}$  metals for the aging studies. These processes involve both chemical and mechanical treatments on the plutonium materials and associated chemical agents. During each process, constant monitoring of the plutonium material balance between the input materials and output products is required. Mass spectrometry, in general, can provide isotopic analysis of the products to high degree of accuracy. Unfortunately, how the sampling is done on the products is crucial, because mass spectrometry can only provide results that represent "microscopic" isotopic content of the products. On the contrary, the gamma-ray analysis results combined with calorimetry results, though not as accurate as mass spectrometry results, represent the "macroscopic" isotopic content of the products.

For non-destructive, gamma-ray isotopic analysis of the elevated  $^{238}\text{Pu}$  products, the original MGA code could not be used on such high concentrations of  $^{238}\text{Pu}$ . We have, therefore, modified the methodology used in the 100-keV region gamma-ray analysis of the original MGA analysis code to overcome this deficiency. In the paper, we will compare the results from this improved MGA code to the results from the mass spectrometry in a set of elevated (5 ~ 7 %  $^{238}\text{Pu}$ ) samples. We have also tested the ability of the code on the extremely high (> 40%  $^{238}\text{Pu}$ ) samples.



The MGA was developed to analyze either a low-energy gamma-ray spectrum taken using a high-resolution planar HPGe detector for energies below 300 keV, or to analyze a low-energy spectrum combined with a high-energy spectrum (up to 1 MeV) taken with a HPGe coaxial detector in what we refer to as the two-detector analysis mode. The reason for using two detectors is simple: a high-energy spectrum taken using a coaxial HPGe detector will not provide sufficient energy resolution for 100-keV plutonium isotopic analysis, while the small planar HPGe used at low energies has inadequate high-energy. The "two-detector" mode MGA analysis was developed to improve the determination of the  $^{241}\text{Pu}/^{239}\text{Pu}$  ratio in high burnup plutonium with the 300-keV regions from the high-energy spectrum. In high burnup plutonium,  $^{241}\text{Pu}$  is dominant in the 100-keV region and the precision of the  $^{241}\text{Pu}/^{239}\text{Pu}$  ratio from this region is reduced.

In general, MGA can provide accurate plutonium isotopic information for all burnups using the data collected with a high-resolution planar detector (below 300 keV). However, more 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  $^{238}\text{U}$  abundance can be obtained from the 1001 keV peak, a more accurate analysis for  $^{237}\text{Np}$  can be made, some fission products can be identified, and  $^{241}\text{Am}$  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.

Figure 3 illustrates how the relative detection efficiency for the measurement is determined for the low-energy detector. To properly use the higher energy gamma-ray information, a separate intrinsic efficiency curve must be determined, as shown in Fig. 4. 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.

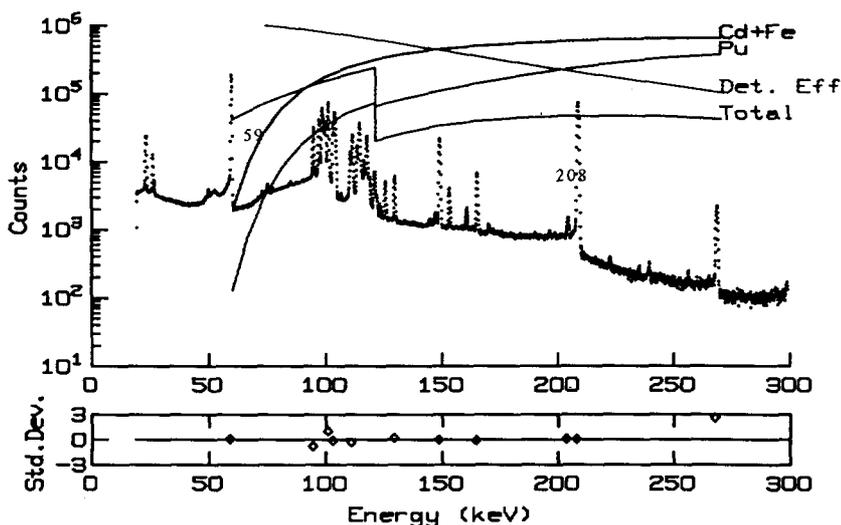


Figure 3. MGA uses physical attenuation corrections of both emission and absorption in the gamma ray interactions. This plot shows the three principal components (the detector efficiency, the absorption due to intermediate materials, and the self-absorption of plutonium) of that characterize the low-energy "intrinsic" efficiency curve.

The need for two detectors to measure high burnup plutonium is inconvenient for inspection organizations, especially with the recent introduction of the Safeguards type coaxial HPGe detectors that has adequate energy resolution for 100-keV gamma-rays and sufficient efficiency for 1-MeV gamma-rays. Therefore, MGA was upgraded to allow measurements of high burnup plutonium with a single high-resolution germanium detector<sup>3)</sup>.

The MGA can also operate in the MGAHI mode<sup>6)</sup> to obtain isotopic information of moderate shielded plutonium sample when the 100-keV region gamma-ray information cannot be obtained.

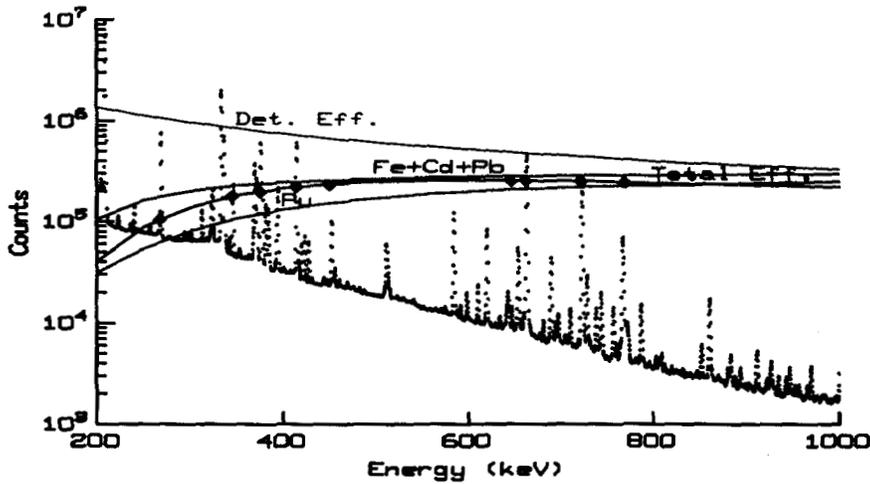


Figure 4. The same physical attenuation processes are used in analyzing the second detector data when running MGA in the two-detector mode. This plot shows the components affecting the efficiency curve. In general, there is a thin Pb absorber in front of the coaxial detector to reduce the count rate from the low-energy region of the spectrum (<200 keV).

### MGA ANALYSIS AND MASS SPECTROMETRY ANALYSIS ON ELEVATED $^{238}\text{Pu}$ SAMPLES

For elevated  $^{238}\text{Pu}$  samples, the gamma rays in both 100-keV region and 150-keV region will be dominated by the decay of  $^{238}\text{Pu}$  at 99.864 keV and 152.680 keV as shown in Figure 5. The ability of MGA analysis at the 100-keV region, which relies on the accurate energy calibration using the weighted peak heights of the 94.658-keV and 101.066-keV gamma rays, is perturbed by the "intrusion" of the 99.864-keV, 98.441-keV, and 94.658-keV  $^{238}\text{Pu}$  gamma-rays. In ordinary plutonium samples, the intensity of the 98.441-keV and 94.658-keV gamma-rays is mostly from the  $^{239}\text{Pu}$  decay.

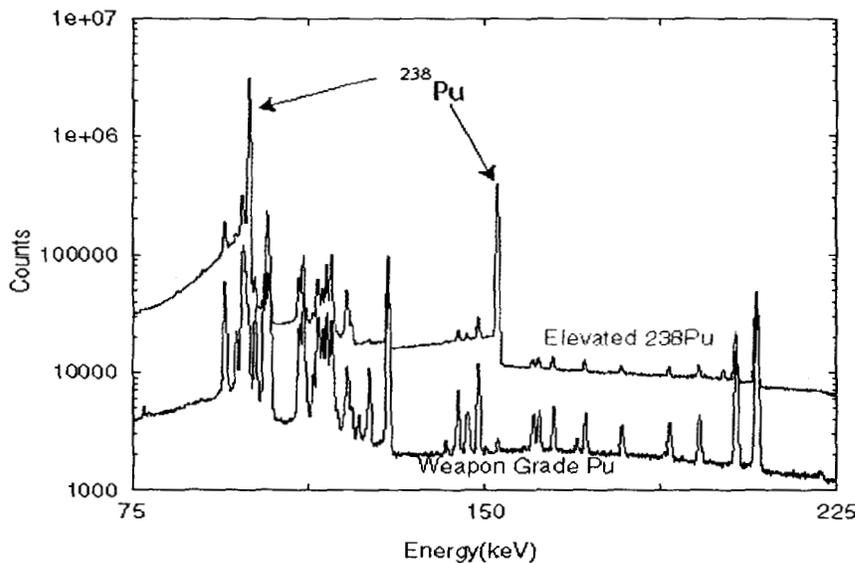


Figure 5. A comparison of the 100-keV region gamma-ray data between an elevated  $^{238}\text{Pu}$  sample and a weapon grade Pu sample.

In order to do accurate energy calibration for the 100-keV region for elevated  $^{238}\text{Pu}$  samples, we have included the weighted peak-height of the 99.864-keV into the energy calibration in the MGA code. However, this inclusion of the 99.864 peak height is meaningless for regular plutonium samples as can be seen from Figure 1 (i.e., it is impossible to obtain accurate peak-height information of this peak when the intensity is low). To be able to use this new energy calibration only for elevated  $^{238}\text{Pu}$  samples, we have added a flag that uses the peak height ratio of 152.8keV ( $^{238}\text{Pu}$ ) and 129.29 keV ( $^{239}\text{Pu}$ ) to trigger the MGA code into the elevated  $^{238}\text{Pu}$  mode. Figure 6 shows a screen dump of the modified MGA code fitting to one of the elevated  $^{238}\text{Pu}$  samples.

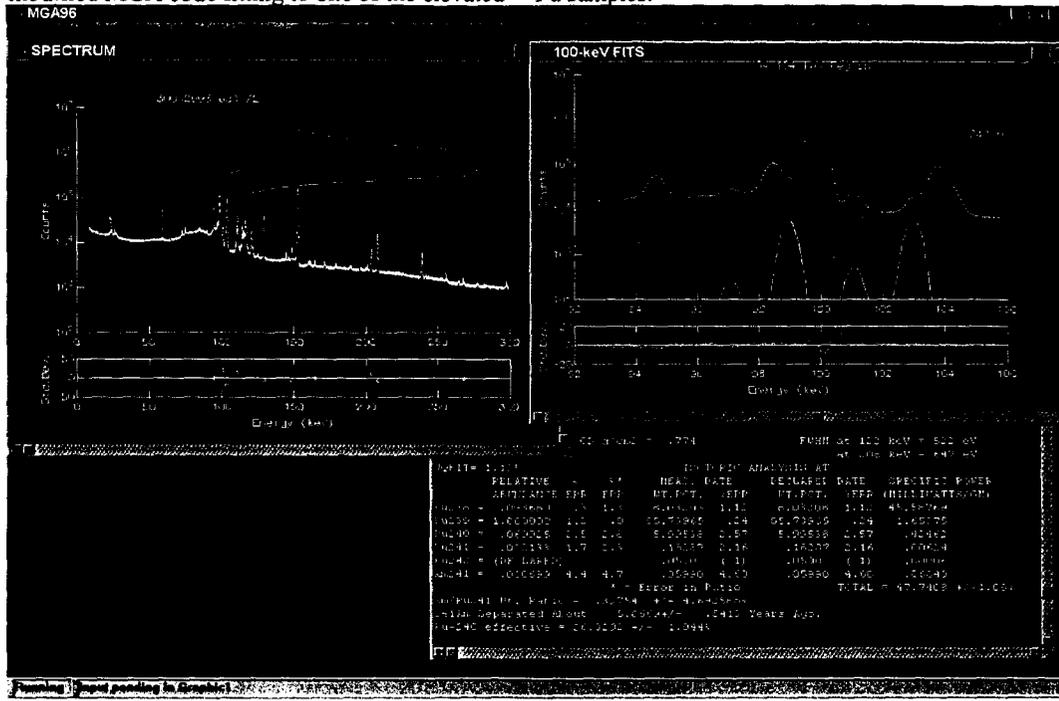


Figure 6. A screen dump of the modified MGA code while analyzing the "slug" component in the elevated  $^{238}\text{Pu}$  process. On the upper left is the physical attenuation corrections displayed in graphical form; on the upper right is the spectral deconvolution of the 100-keV region. The output is at the bottom right.

Two different mass spectrometry instruments, the Inductivity Coupled Plasma Mass Spectrometry (ICPMS) and IsoProbe, were used in providing mass spectrometry results of the elevated samples. Careful chemical separations of  $^{238}\text{U}$  from  $^{238}\text{Pu}$  were performed on the samples as well as alpha particle detection using surface barrier detectors, prior to and after the separations, were also performed to reaffirm successful chemical separations, since the mass spectrometry cannot tell the difference between these two isotopes. Long duration gamma-ray counting on these samples after the chemical separations (due to small sample size used in the mass spectrometry measurements) was performed. Table 1 shows comparisons between the mass spectrometry results and modified MGA results for a set elevated  $^{238}\text{Pu}$  samples.

Table 1. Comparisons between the Mass Spectrometry and the MGA results. All numbers are in weight per cent, the  $^{242}\text{Pu}$  weight per cent is assumed to be 0.05% for all samples. The averaged mass spectrometry results for  $^{242}\text{Pu}$  is 0.05 +/- 0.001 %.

Sampe	Analysis mode	$^{238}\text{Pu}$
#1	MGA	7.33 (1.6%)
	Mass Spec.	7.1035(0.33%)
#2	MGA	7.15 (1.1%)
	Mass Spec.	7.1593 (0.33%)
#3	MGA	7.31 (1.15%)
	Mass Spec.	7.0865 (0.34%)
#4	MGA	7.17 (1.2%)

	Mass Spec.	7.1144 (0.34%)
#5	MGA	7.25 (1.2%)
	Mass Spec	7.0607 (0.03%)
#6	MGA	4.86 (1.2%)
	Mass Spec	4.83 (0.33%)

One important thing to note is that for the modified MGA to be able to provide good results, the knowledge of  $^{242}\text{Pu}$  values for these "artificially made" elevated  $^{238}\text{Pu}$  samples is required in the input. For regular samples of different burn-up times, MGA uses empirical parameterization of the other isotopes to obtain  $^{242}\text{Pu}$  values.

For large mass (i.e., >100g) elevated  $^{238}\text{Pu}$  samples, the modified MGA analysis has encountered some problems due to the packaging of the sample. The reason is understandable: the heat output of these samples is high, and large amounts of "heat-sink" materials were placed around these samples during the packaging. The gamma-rays in the 100-keV region, which were used in the modified MGA analysis, will not be attenuated isotropically because of the irregular shape of these heat dissipating materials. One possible solution to this problem is to use higher energy gamma-rays (i.e. lesser effect due to attenuation compare to the 100-keV region gamma-rays) in the 600- and 700- keV regions. At current, we are exploring this possibility.

## CONCLUSION

The modified MGA algorithm in the 100-keV region has been tested in good agreement with the mass spectrometry results for small quantity of elevated  $^{238}\text{Pu}$  samples. For larger  $^{238}\text{Pu}$  samples, the packaging of the sample is crucial to ensure the success of the 100-keV region analysis. For some heterogeneous components produced in the process, we are using the active and passive tomography technique<sup>7)</sup> to hopefully find a solution to the problem.

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## 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. Karen Dodson, private communication, (2001)
5. T.F. Wang, K. Raschke, and W.D. Ruhter, *Two-Detector Mode MGA Analysis of Plutonium Using a Single Ge Detector*, UCRL-JC-141234, presented at the 23rd Annual ESARDA Symposium on Safeguards and Nuclear Material Management, May 2001.
6. 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)
7. T.F. Wang, H.E. Martz, G.P. Roberson, E.A. Henry, W.D. Ruhter, L.O.Hester, *Three Dimensional imaging of a Molten-Salt-Extracted Plutonium Button using Both Active and Passive Gamma-ray Tomography*, Nuclear Instrument and Method in Physics Research, **A353** (1994)