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Improvements in the MGA Code Provide Flexibility and Better Error Analysis*

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The Multi-Group Analysis (MGA) code is widely used to determine nondestructively the relative isotopic abundances of plutonium by gamma-ray spectrometry. MGA users have expressed concern about the lack of flexibility and transparency in the code. Users often have to ask the code developers for modifications to the code to accommodate new measurement situations, such as additional peaks being present in the plutonium spectrum or expected peaks being absent. We are testing several new improvements to a prototype, general gamma-ray isotopic analysis tool with the intent of either revising or replacing the MGA code. These improvements will give the user the ability to modify, add, or delete the gamma- and x-ray energies and branching intensities used by the code in determining a more precise gain and in the determination of the relative detection efficiency. We have also fully integrated the determination of the relative isotopic abundances with the determination of the relative detection efficiency to provide a more accurate determination of the errors in the relative isotopic abundances. We provide details in this paper on these improvements and a comparison of results obtained with current versions of the MGA code.

1. INTRODUCTION

The world-wide use of the MGA code[1] has resulted in users encountering plutonium gamma-ray measurement situations where the code cannot meet the user's needs, because of unexpected additional peaks in the spectrum from other isotopes or where expected gamma-ray peaks are absent due to new plutonium conditions. A recent example was freshly processed high-burnup plutonium where the 59.5-keV gamma ray from ^{241}Am and the 129.3-keV gamma ray from ^{239}Pu were absent from the measured gamma-ray spectrum. MGA uses these peaks to determine an accurate gain and zero for the measured spectrum and as a result, the MGA failed to analyze the spectrum.

The gamma- and x-ray peaks used by the MGA code are hardwired into the code and in situations like the one described above, the MGA code had to be modified for it to perform the analysis. There have been many modifications made to the MGA code as a result of situations like this, because of the lack of a capability in the code for the user to add or remove gamma- and x-ray peaks from the analysis.

In a recent paper,[2] we described a general approach to gamma-ray isotopic analysis that gave the user the capability to specify the isotopes and the gamma and x rays to be used in the desired analysis.

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In this paper, we describe how we have expanded that general approach to include the analysis of complex regions of gamma-ray spectra such as the 100-keV region of plutonium

gamma-ray spectra. A unique feature of the MGA code is its analysis of the 100-keV region of the plutonium gamma-ray spectrum. As a result, measurement times can be as short as a few minutes; measurements are frequently accurate to within 1%. This region is very intense and permits MGA to obtain significantly better statistical precision than by using other regions of the plutonium spectrum. It also contains peaks from all of the isotopes of interest. However, as shown in Figure 1, the more than 15 x-rays and gamma rays in this region are significantly overlapped and thus require a sophisticated method of analysis. We have added a capability to our general approach to gamma-ray isotopic analysis to include the analysis of complex gamma-ray regions such as the 100-keV region.

2. DESCRIPTION OF THE ANALYSIS SOFTWARE

Our approach to the development of the new gamma-ray isotopic analysis tool is based on the fact that the observed intensity of any gamma-ray (or x-ray) emitted from a sample is a function of the quantity of the isotope present in the sample, the emission probability for the gamma-ray being measured, the self attenuation of the emitted photons in the sample, the attenuation of the emitted photons between the sample and the detector and the detector efficiency. This functional dependence can be described by a mathematical relationship shown in the expression below:

$$I_j \equiv \sum_{k=1}^{N} \left(p_{j,k} \cdot X_k \right) \left[\frac{e^{\left(-\mu_{con}(E_j) t_{con}\right)} \left(1 - e^{\left(-\mu_{smp}(E_j) t_{smp}\right)}\right)}{\left(\mu_{smp}(E_j) \cdot t_{smp}\right)} \right] \left[\varepsilon_j^0 \left(1 + bE_j + cE_j^2\right) \right] \qquad (1)$$

In the above expression I_j represents the intensities of the selected peaks in the gamma-ray pulse height distribution from the isotopes of interest and X_k represent the unknown disintegration rates of the selected isotopes. The emission probabilities are given by the terms $p_{j,k}$ for each peak *j* belonging to isotope *k*. The absorption coefficients, μ_{con} and μ_{smp}, are for the given energy of the j^{th} peak for the sample container and the sample, respectively. The container wall thickness t_{con} and the sample thickness t_{smp} are treated as unknown parameters that need to be fit. The final term in the expression above, ε_j^0, represents the estimated relative efficiency of the detector for peak j. The polynomial seeks to adjust the relative efficiency for small variations. The base value for ε_j^0 may be calculated from the physical geometry of the detector.

This equation is very nonlinear in form and therefore the variables must be solved by an iterative least-squares method. Note that in addition to determining the relative detection efficiency for the measurement, the solution to the developed equations provides the relative amounts of the isotopes involved, when corrected for half-lives.

Our gamma-ray isotopic analysis tool provides the capability for the user to specify which gamma-ray regions and which peak intensities in these regions to use in solving Equation 1 as was described in our previous paper.[2] The user provides this information in a text file, which can be created and edited with a simple text editor. In this text file, the user also specifies which peaks are to be used to determine the energy per channel and the zero channel

energy for the spectrum. This addresses the recent analysis problem with freshly processed, high-burnup plutonium, which was described above.

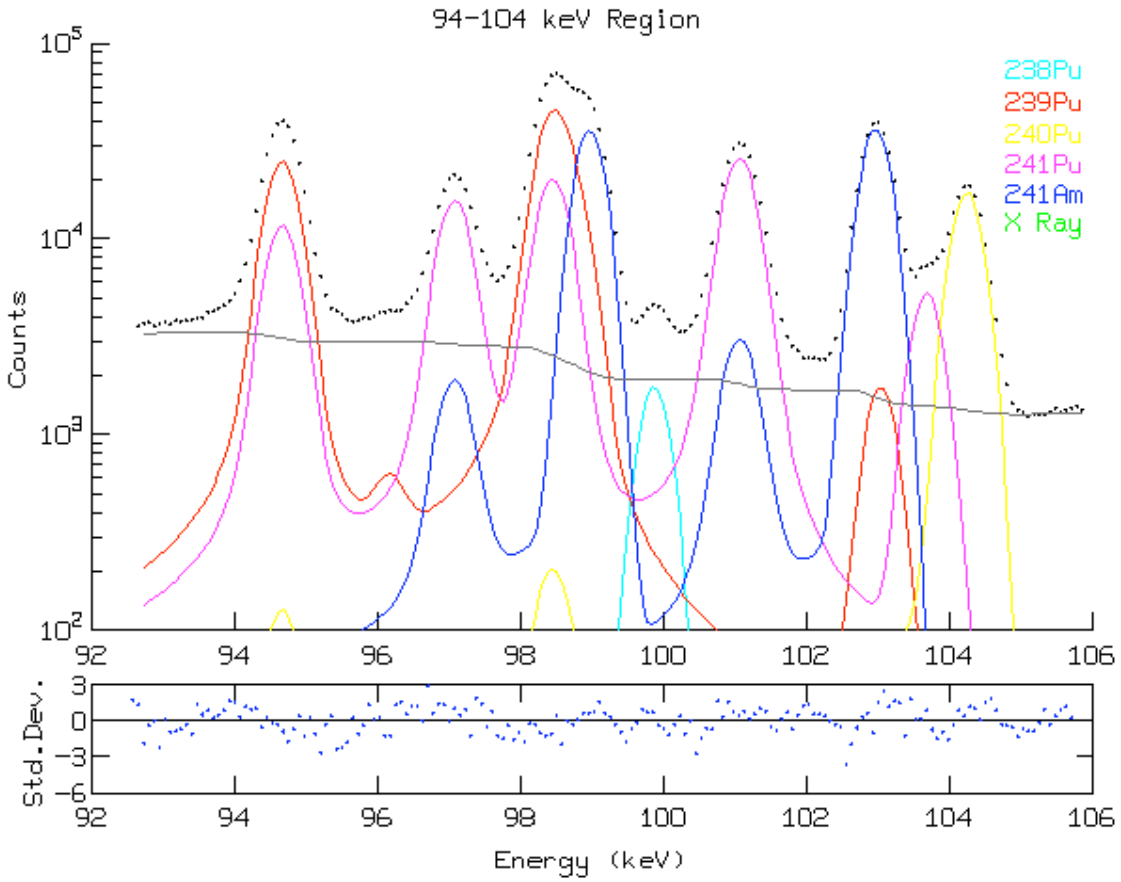

Figure 1. The spectral response of each plutonium and americium isotopic component in the 94- to 104-keV region can be calculated by using known and measurable parameters. The relative plutonium isotopic abundances are then determined by fitting these responses to the data in this energy region using Equation 2.

For those peak regions with more than one peak, a peak-fitting routine similar to GRPANL[3] is used that fits a Gaussian peak shape with an exponential low-energy tail to each specified peak. The fitting determines the net peak area for each of the specified peaks along with an error that is based on counting statistics and the quality of the fit.

For the 100-keV region in plutonium gamma-ray spectra shown in Figure 1, this peak fitting approach will not work. Because of the severe overlap of the peaks, individual peak intensities cannot be determined. To fit the net channel-by-channel data in the 100-keV region of plutonium gamma-ray spectra, we use the same approach used in MGA by generating gamma-and x-ray response profiles for each isotope contributing to the 100-keV region.

$$y_i \equiv \sum_{k=1}^{N} \left(p_{j,k} \cdot X_k\right) \left[\frac{e^{\left(-\mu_{con}(E_j)t_{con}\right)}\left(1 - e^{\left(-\mu_{smp}(E_j)t_{smp}\right)}\right)}{\left(\mu_{smp}(E_j) \cdot t_{smp}\right)} \right] \left[\varepsilon_j^0 \left(1 + bE_j + cE_j^2\right) \right] * e^{\alpha(x_i - x_0)^2} + \mathrm{T(x_i)} \quad (2)$$

Equation 2 is used to fit the net channel-by-channel data, y_i, at x_i in the 100-keV region. The reader should note that Equation 2 is similar to Equation 1 except for the additional Gaussian factor which disperses the peak intensity about the peak's centroid, x_0, and a low-energy tailing factor, $T(x_i)$, which describes the low-energy tailing usually observed in spectra measured with a germanium detector. The results of a fit to the 100-keV region with our gamma-ray analysis tool are shown in Figure 1.

In MGA, the corrections to the relative efficiency, variables b and c in Equation 2, are determined by fitting equations like Equation 1 to the intensities of 11 gamma-ray peaks in the spectrum. These eleven peaks are specified in the MGA code and cannot be changed by the user. In our general gamma-ray isotopic analysis tool, the user specifies which peaks to use in solving Equation 1. Once MGA has determined the values of b and c from solving these 11 equations, it uses these values in Equation 2 to fit the 100-keV region and assumes that there are no errors in b and c. In our gamma-ray analysis tool, equations like Equation 1 are developed for individual peak intensities and solved simultaneously with the equations like Equation 2, developed to the fit the net channel-by-channel data in the 100-keV region. By integrating the solutions to Equations 1 and 2 and weighting the equations by the variance of the data they represent, we are able to determine the relative isotopic abundances along with a better determination of their uncertainties than in MGA.

To solve the set of equations created from the selected peak intensities and channel-by-channel data, the user also specifies the composition and estimated mass thickness of the sample, as well as the compositions and mass thickness for up to three absorbers. The thickness of at least two of the three absorbers must be known. Finally, the user specifies the coefficients from a log-log fit to the detector's relative efficiency normalized to 1.0 at a selected energy. The three coefficients give the intercept, slope, and curvature of the relative efficiency curve.

All of this information is used to determine the relative disintegration rates of the isotopes contributing to the selected peak intensities. The equations are nonlinear in several unknowns and thus require iterative, nonlinear least squares to solve. We have developed software to monitor and control the iterative process to insure that the mass-thickness values for the absorber and sample stay within reasonable ranges. Initial estimates of the relative disintegration rates for the isotopes involved are made from a selected peak area for each isotope. The iterative process begins with freed values for the isotope disintegration rates and the absorber thickness. After four iterations the sample thickness is also freed. After seven iterations the absorber thickness is fixed, unless convergence is reached first. Once convergence is reached in the sample thickness, the efficiency slope is freed. When convergence is reached in the efficiency slope, the efficiency curvature is freed for two iterations. If convergence is not reached in twenty iterations for any of the thickness or efficiency curve values, the process stops and reports that convergence was not reached. When convergence is reached in the thickness and efficiency curve values, the relative amounts of the isotopes involved are calculated from the relative disintegration rates determined from the iterative process and the half-lives given by the user.

3.0 ANALYSIS RESULTS

We have tested our gamma-ray analysis tool on plutonium gamma-ray spectral data that were taken with a high-resolution germanium detector with an energy resolution of nominally 550

eV at 122 keV. These data were taken with a gain of 75 eV/ channel for analysis by the MGA code. In Table 1, we show the plutonium isotopic abundances and their relative errors determined by MGA in comparison with results from our general gamma-ray isotopic analysis tool.

Sample/Analysis Code	Pu-238	Pu-239	Pu-240	Pu-241
CBNM61				
MGA	1.127+/- 1.06%	64.533+/- 0.57%	25.268+/- 1.17%	4.880+/-0.98%
Gamma Tool	1.193+/-0.75%	62.977+/-0.41%	26.430+/-0.77%	5.208+/-0.67%
CBNM70				
MGA	0.789+/-1.12%	75.194+/-0.38%	18.004+/-1.38%	3.937+/-0.96%
Gamma Tool	0.831+/-0.28%	73.846+/-0.91%	19.048+/-0.91%	4.198+/-0.70%
CBNM84				
MGA	0.0656+/-1.00%	84.671+/-0.07%	14.152+/-0.42%	0.753+/-0.34%
Gamma Tool	0.0652+/-1.12%	84.539+/-0.06%	14.274+/-0.36%	0.765+/-0.34%
CBNM93				
MGA	.0099+/-2.77%	93.462+/-0.03%	6.340+/-0.39%	0.149+/-0.33%
Gamma Tool	.0094+/-3.01%	93.555+/-.02%	6.247+/-0.35%	0.150+/-0.28%

Table 1. This table shows a comparison between plutonium isotopic abundance results and their calculated errors from MGA and our general gamma-ray isotopic analysis tool.

4.0 SUMMARY

Our gamma-ray isotopic analysis tool utilizes a unique and accurate methodology for determining the relative detection efficiency that enables us to relate the intensities of widely separated gamma-ray peaks to determine relative abundances of specified isotopes. We have generalized the application of this methodology in software, so that the users can easily modify and adapt it to their analysis needs. The user may specify which peak intensities to use and how to fit peaks in a given energy region with input from a text file. We have fully integrated the determination of the relative detection efficiency using isolated peak intensities with the fitting of channel-by-channel net data from the complex, 100-keV region of the plutonium gamma-ray spectrum. The inclusion of the 100-keV region data gives more precise plutonium isotopic abundances from the measured data. Integrating the fit to the 100-keV data with the determination of the relative detection efficiency gives a more accurate approach to the determination of the errors in the relative isotopic abundances. The analysis software is very transparent with it using a set of equations that represent the physical processes that determine the measured gamma-ray intensities and physical parameters that are specified by the user. The relative plutonium isotopic abundance results and the calculated errors from our gamma-ray isotopic analysis tool are in generally good agreement with the results obtained using MGA on the same spectral data. Extensive testing of our new gamma-ray isotopic analysis tool still needs to be done a broad range of plutonium gamma-ray spectra taken under varying measurement conditions and plutonium burnups and ages.

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