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# Transuranic Isotopic Analysis Using Gamma Rays

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# TRANSURANIC ISOTOPIC ANALYSIS USING GAMMA RAYS

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## ABSTRACT

Transuranic waste typically emits gamma rays that are characteristic of the isotopic composition of the materials. If the area of the gamma ray photopeaks in a High Purity Ge (HPGe) spectrum can be accurately determined and if the gamma ray/x-ray branching ratios and half-lives for the radionuclides in the sample are known the relative concentration of each isotope in the waste can be determined using tomographic techniques. Methods used to accurately determine these photopeaks usually requires a computer code that does multi-peak analysis and unfolding of a given part of the gamma-ray spectrum. Computer techniques allow an accurate determination of the photopeaks and hence the isotopic composition of the waste material.

These computer techniques can be automated for different spectra within a wide range of possible isotopic compositions. To improve photopeak statistics all of the spectra taken in a tomographic survey of the sample are summed and are used in the isotopic analysis. The method, accuracy, and limitations of this type of isotopic analysis system will be discussed.

The gamma ray acquisition system is currently being upgraded with multiple HPGe detectors to improve the counting statistics obtainable in a given amount of time. The results of the DOE performance evaluations and the progress of the multiple detector upgrade will be discussed.

## INTRODUCTION

During the Active and Passive Computed Tomography (A&PCT) data acquisition the item to be assayed is translated, elevated and rotated about its vertical axis and full gamma spectra acquired segment by segment along each axis. This reduces the effects of non-uniformity in both matrix density and nuclide distribution. The gamma attenuation coefficient of each element (voxel) is calculated in the A&PCT analysis from the measurement of the transmitted intensity of an external gamma source—the active part of the A&PCT measurement. In the typical A&PCT active analysis the data is gathered from up to fourteen different positions at seventeen different elevations on the drum and at 21 different angles (projections) for each elevation.

In the typical A&PCT passive analysis the spectral data from the sample is gathered from up to six different detectors at seventeen different elevations on the drum and at ten different angles for each elevation. This passive analysis may take from five to 24 hours, depending on the activity in the sample, matrix attenuation and the number of detectors used. Multiple detectors are being purchased to reduce this time to approximately one hour. Although the statistics of each of these spectra are low they can be combined to produce a summed spectrum of good statistics for gamma-ray spectroscopic analysis. By analyzing the summed spectrum, one can extract a great deal of information about the isotopic contents of the sample.

Once the absolute intensity of one gamma is found by the A&PCT active analysis, the intensity of all other gammas of that isotope is given by their relative branching ratios. If plutonium is present in the sample, an analysis of the summed gamma-ray spectrum can determine the mass ratios of the important plutonium isotopes and their associated daughters. A multi-peak analysis of all the gammas emitted in a given energy range (also called Multi-Group Analysis, MGA[1]) finds the intensity of a given isotope's peak(s). Analysis of different regions gives data on different isotopes—see Table 1. By accurately determining the intensity of the gamma rays of each isotope and a nearby $^{239}$Pu isotope the following isotopic mass ratios can typically be determined: $^{238}$Pu/$^{239}$Pu, $^{240}$Pu/$^{239}$Pu, $^{241}$Pu/$^{239}$Pu, $^{241}$Am/$^{239}$Pu, and $^{237}$Np/$^{239}$Pu. If uranium is present $^{235}$U/$^{239}$Pu, and $^{238}$U/$^{239}$Pu and also typically be determined. In addition the ratio $^{242}$Pu/$^{239}$Pu is calculated from the results of the previous ratio determinations. The ratio analysis typically includes the isotopes important to transuranic waste characterization and also identifies other isotopes that might be of interest or that could interfere with the analysis.

The A&PCT analysis method could, in principle, determine a mass and average gamma transmission for each of these isotopes by reconstructing the sinograms of their characteristic gamma rays in the spectrum. However, that is rarely possible in any reasonable amount of time due to the varying statistics and spectral shapes exhibited by these data. Instead, the A&PCT assay only needs to determine the mass and average gamma transmission of one isotope, usually the 413.714 keV line of $^{239}$Pu. The isotopic ratio information found by the multi-peak analysis can then be used to determine the masses of the other plutonium (and daughter) isotopes present in the waste. This information can then be used to calculate the derived quantities, i.e., thermal power, total alpha activity, and fissile gram equivalent that are necessary for the characterization of the sample.

## DATA CONVERSION AND REDUCTION

The data that the multi-peak analysis code (MPAC) needs to run is in several different data formats and in several different files. Setup, calibration and data base files are typically in ASCII format. These files have to exist before each run. The spectral data are usually in a binary format specified by the multichannel analyzer manufacturer. MPAC can read several different data types. The file type of the data is specified in the setup file.

Typically the statistical variation in each individual data file for each segment or ray is too large (poor counting statistics) to get reliable results for isotopic analysis. The first step in the normal spectral analysis is to sum the individual spectra from each detector, view and elevation into a single "high" statistics gamma-ray spectrum. Obviously, when using multiple detectors, each detector plus analyzer has to be initialized as nearly identical as possible. In the A&PCT analysis, output of each data run is assigned a unique file a name representative of its detector, rotation, translation and elevation. To facilitate reading these data files, file names are identical for each tomographic run. The number of files and the range of names is specified in a A&PCT data file--*templet.sct*. Each different A&PCT data set is put into folders with different names. Each data file in a folder is then read and added into a single summed file--named after the folder's name.

After each file is read, it is analyzed for the specified peaks that have a energy of interest (EOI) and a database containing the results is accumulated. Results are output in several other files needed for analysis by the tomographic routines. The final results of the calculation and other files of interest are put in the same folder as the rest of the data. All files are saved for later reference.

## PRELIMINARY DATA ANALYSIS

After the sum file is created, one of the first steps is to identify the main gamma peaks in the summed spectrum. This is done by first finding a "universal" background for the entire spectrum. Next the counts that are above this background are analyzed for peak characteristics i.e. a statistically significant number of adjacent counts above background. All such regions in the data are flagged, the counts above background are fitted with a gaussian peak routine to find their peak energy and width and then compared to lists of known gamma energies emitted by isotopes of interest. The spectrum's gain and the zero (specified in the setup file) have to be correct for this comparison to be valid. In this first preliminary step only individual peaks are checked against lists of known peaks. No detailed analysis is performed on peak multiplets etc. The peak criteria is intentionally set "loose" so that some "peaks" that are detected are only statistical anomalies. This assures finding nearly all easily determined peaks and significant isotopes. Many "background" isotopes are often found in this step as well as isotopes of primary interest. The peaks found and there statistics are saved in the analysis output file.

From this preliminary analysis the data is flagged for further analysis if there are plutonium, americium, and/or uranium or their daughters isotope peaks detected. If these isotopic peaks are not detected the spectrum is either a calibration spectra or a spectra that requires special analysis and is flagged as such and the code terminates without further analysis. The summed file is left in a format easily plotted to facilitate further analysis.

## ISOTOPIC ANALYSIS

The characteristic energies and intensities of gamma rays emitted by radioactive isotopes are commonly used as fingerprints for isotope identification. Accurately measuring the peak intensities from different isotopes allows the sample's isotopic composition to be determined if each peak's halflife, branching ratio, detector efficiency and gamma transmission are also known[2,3,4,5,6]. By performing a multi-peak analysis on all the peaks in a given energy range, an accurate determination can be found of each peak's intensity. The other information can either be obtained from the isotopic data or accurately estimated.

To find these peak intensities in a multi-peak analysis several things are needed:

1. The full energy gamma/x-ray peak signals needs to be separated from the background (Compton continuum) signal.
2. A mathematical description of the peak shapes, as a function of energy, for gammas and x-rays is needed.
3. A fitting algorithm, which varies the peak shape parameters to get the best fit, is needed. Where the best fit is defined as the minimum least square deviation between the data points and their calculated values.
4. A data set of theoretical relative intensities (branching ratios) and energy of each gamma ray or x-ray in a region of interest for each isotope or x-ray source is needed.
5. A way to correct, at least approximately, the data set for instrumental efficiencies and photon transmission to the detector peculiar to each data set collected is needed.

## MULTI-PEAK ANALYSIS

The intensity (counts greater than background) of a given peak is related to the isotopic source strength by the equation:

$$I_1 = \lambda_1 A_1 B_1 \Omega_1 \varepsilon_1 \tau_1 \text{ (counts/sec)} \qquad [1]$$

Where:

$I_1$ = measured peak intensity of isotope 1
$\lambda_1 = .6932/T_1$ = decay constant [$T_1$ = material halflife (in seconds)]
$A_1$ = Number of atoms of isotope 1
$B_1$= branching ratio of isotope 1
$\Omega_1$ = Fractional solid angle of detector from source
$\varepsilon_1$ = Gamma counting efficiency of isotope 1
$\tau_1$ = Gamma transmission to detector of gamma 1

The following relation relates the ratio of two isotopic abundances to the observed peak intensities:

$$A_1/A_2 = I_1 \lambda_2 B_2 \, \varepsilon_2 \tau_2 \Omega_2 \, / \, I_2 \lambda_1 B_1 \, \varepsilon_1 \tau_1 \Omega_1 \qquad [2]$$

From gammas which are near the same energies the following observations can be made:

$\varepsilon_2\tau_2 / \varepsilon_1\tau_1 \approx 1$ if the two gammas are close to the same energy.

$\Omega_1 = \Omega_2$ The fractional solid angle of detector is the same for both gammas and cancels out.

$\lambda_1$ and $\lambda_2$ are known from the previously measured half lives.

$B_1$ and $B_2$ are known from the previously measured branching ratios.

$I_1$ and $I_2$ have to be determined accurately to get accurate isotopic ratios.

If the amount of one isotope, $A_{std}$, can be found from other techniques that determine the amount of a "standard" gamma ray of the same isotope the value of $A_2$ can be easily determined by:

$$A_2 = A_{std} B_2 / B_{std} \qquad [3]$$

where:

$A_{std}$ = the amount of a given isotope determined by the A&PCT analysis of a "standard" gamma ray of a given isotope.
$B_{std}$ = Branching ratio of the "standard" gamma ray.
$B_2$ = Branching ratio of the comparison gamma ray.

The accuracy of determining the isotopic ratio, $A_1/A_2$ , is largely determined by the accuracy of determining the respective peak intensities $I_2$ and $I_1$ for gammas and x-rays that are near the same energy. Accurate detector efficiencies and transmissions for all energies are no longer needed. First order detector efficiencies and gamma transmission corrections can usually be found and since they are used in a ratio their errors largely cancel. The accuracy of determining the peak intensities depends upon the individual statistics of the peaks, interference from other peaks, accuracy of the background determination and other factors. These errors are often larger than the errors in determining the ratio $\varepsilon_2\tau_2 / \varepsilon_1\tau_1$, the accuracy of the efficiency and transmission corrections. To minimize the error in determining the isotopic composition, averages of several peaks in different regions often gives the "best" answer.

From the above analysis it is seen that to optimize the accuracy of isotopic determination it is desirable to choose peaks from different isotopes that are reasonably close together in energy and have roughly the same intensity. Analyzing the data this way minimizes the corrections that have to be applied and the errors in the final results. Six different energy regions of the gamma-ray spectrum were chosen for detailed multi-peak analysis. Multiple energy regions were chosen to minimize the effects of interfering peaks, statistics etc. on the data. The low energy regions below 100 keV were excluded from the analysis because of the large attenuation these gammas typically incur in escaping from a typical waste sample. Each spectral region analyzed typically contains one or more peaks from $^{239}$Pu and one or more peaks from other isotopes of interest.

## DETERMINING THE COMPTON CONTINUUM BACKGROUND

Several energy regions containing many peaks typically compose a gamma ray spectrum. The analysis is done on several energy regions of the spectrum that contain peaks from isotopes of interest. The net (peak) signal in a give energy region is found by determining and then subtracting a background function from the raw data. This process eliminates the Compton continuum from the analysis. The background function is found by first choosing areas, on each side of the region of interest, that have no peaks and finding the average background there by averaging over several adjacent "background" channels. Choosing these background regions requires analyzing many spectra to assure that there are no "contaminating" peaks in each background region. The background counts, b(i), under the peak region are then obtained by:

$$b(i) = B_{left} - D * \Sigma C(i)/S_{tot} \quad [4]$$

where:

$B_{left}$ = average background on low energy side of the region of interest
D = difference between the left and right average backgrounds
$\Sigma C(i)$ = running sum of the counts from the left background i.e. the sum of all channels from the low energy side to the location of the channel(i).
$S_{tot}$ = total number of all counts from the left to right backgrounds.

The net signal as a function of channel is given by:

$$Y_{net}(i) = Y_{raw}(i) - b(i) \quad [5]$$

Data with backgrounds removed by this technique are found to give good representations of the net signal. The background smoothly progresses from the average background on the low energy side to the average background on the high energy side; decreasing proportionally to the amplitude of the total signal. Other background determinations are usually not nearly as accurate. The main limitation found is that this background may not well represent data in areas of the spectrum where the Compton continuum is falling i.e. the background at the lower energy is lower than the background at the higher energy. Fortunately, in the energy range where we are working the backgrounds are decreasing so this is not a serious limitation.

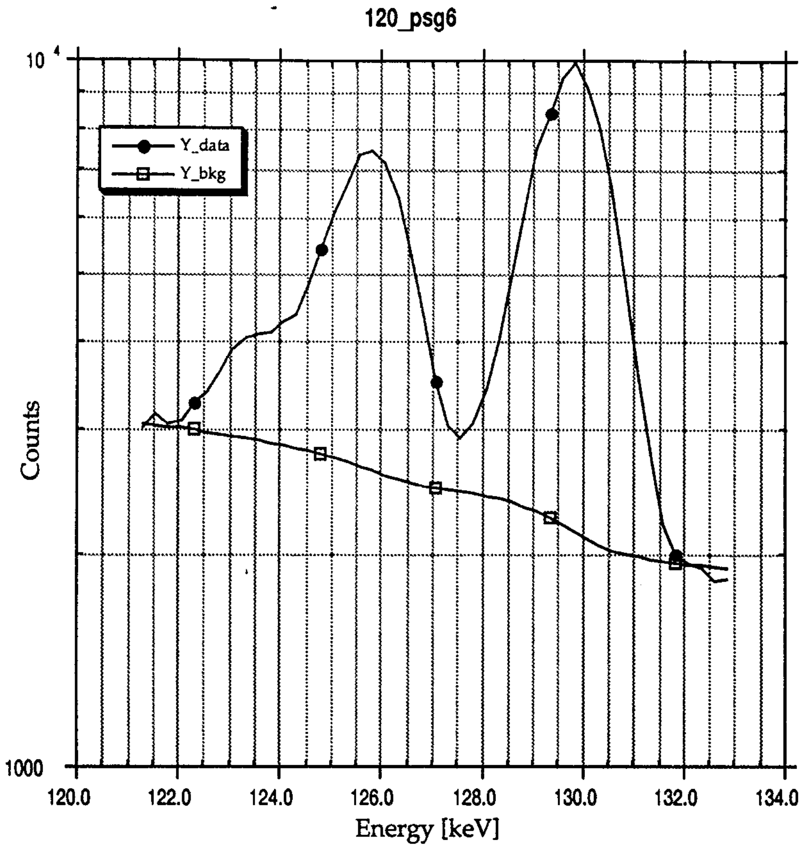

Figure 1. The raw count data and its associated background in the 120-135 keV energy region. The background function is subtracted from the raw data to find the net counts used in the multi-peak analysis.

## THE ISOTOPIC ANALYSIS PROCESS

To fit the net data, mathematical representations are needed for the peak profiles. A gaussian peak and a low energy exponential tail describe gamma peak shapes from a High Purity Germanium (HPGe) detector very well. X-ray peak shapes are described very well by a convolution of the detector response (a gaussian and low energy exponential tail) with the intrinsic x-ray energy distribution (a Lorentzian)—resulting in a so called Voigt profile. The mathematical representation of each gamma peak is calculated from:

$$Y_i = Y_0\{\exp[\alpha(x_i - x_0)^2] + T(x_i)\} \qquad [6]$$

where:

$Y_i$ = net counts in channel $x_i$

$Y_0$ = peak height at peak channel position, $x_0$,

$\alpha$ = peak width parameter [ = $-1/2\sigma^2$]

$T(x_i)$ = tailing function--given by:

$$T(x_i) = [A*\exp(B\Delta) + C*\exp(D\Delta)]*[1 - \exp(0.4\Delta^2)]\delta \quad [7]$$

where: $\Delta = (x_i - x_0)$ difference between count location and gamma peak (channels)

$A*\exp(B\Delta)$ accounts for the short term tailing; A = amplitude, B = slope

$C*\exp(D\Delta)$ accounts for the long term tailing; C = amplitude, D = slope

(for HPGe detectors C and D are set to 0)

$[1 - \exp(0.4\Delta^2)]\delta$ reduces the tailing function to zero at the center of the peak ($\delta = 1$ for $x < x_0$, $\delta = 0$ for $x > x_0$ )

To fit the data, the energy and intensity of each gamma or x-ray from the decay of a given isotope are needed as input. To get the "best" fit all gammas and x-rays in the region should be included. In the energy regions below 120 keV, it is necessary to add to the analyzed peaks all the fluorescent x-rays created by gammas/x-rays interacting in the source or collimation material.

The gamma/x-ray theoretical intensities (branching ratios) have to be corrected for detector efficiencies and transmissions vs. energy in the region of interest to give a "best" fit to the data. Calculating a "rough" detector efficiency vs. energy and a "rough" transmission vs. energy correction and then adjusting the theoretical branching ratios does this correction. Since each energy region is chosen as small as reasonable this first order correction, while not absolutely correct, does give a better fit to the data. [1]

The goal in the calculation process is to find the set of peak shape parameters and intensities that "mathematically" gives the "best" approximation to the measured net signal. The intensity (heights) of

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[1] The values of the theoretical branching ratios and energies of the gamma rays of the different isotopes are often found in the literature from different measurements with different uncertainties. To find the "best" set of values to use in a given region these measurements often have to be treated as only preliminary and adjustments made from examination of the fit of good statistical data. From the shape of the residuals--the difference between the fit and the data--adjustments to these values often have to be made to give a better fit. These adjustments are usually only minor--well within the quoted uncertainties; but can give a significant improvement in the fits.

the calculated peaks are adjusted for the different "corrected" branching ratios. This “best” fit is defined as the fit that gives the minimum reduced Chi square difference between the calculated signal and the measured data.

In the case of multiple peaks from one isotope in a given region, one peak in the region of the fit is chosen as a "standard" and a preliminary determination is made of its likely height by finding the height of the data at the peak's location. The preliminary peak amplitudes of the other peaks, of the same isotope (if any) in the fit region are found from the relative (to the "standard") corrected branching ratios for that isotope. The process is continued for all other isotopes that emit gammas or x-rays in the energy region of analysis till all peaks have a preliminary height estimate.

The fitting process adjusts the peak's parameters to minimize the difference between the equations describing the different peaks and the data. To find the adjustments that give the “best” fit, the equations describing the various peak shapes are expanded with a first order Taylor's series expansion about the trial values of the free parameters. The free parameters chosen for Taylor expansion are typically peak heights, widths and slope of the first exponential and peak energies for each isotope. The number of free parameters is kept to a "minimum" as too many "free" parameters may prevent convergence to a reasonable minimum. Adjustments to the fit are found by performing non linear least square iterations on these first order equations. [This procedure is also known as the Gauss-Seidel or Newton-Raphson method.] In each iteration the "free" parameters are adjusted to give the "best" fit. Iterations are continued till the fit converges to an acceptable limit. In the case of "poor" convergence the iterations are limited to a fixed number (usually fifteen) and the data excluded from the final analysis if the fit's Rchisq is too large. The closer the initial choices are to the “real” values the quicker the fitting process converges. A typical fit calculation takes only a few seconds with a 300+Mhz computer.

The energies of the peaks are allowed to vary only as a complete set; i.e. the energies of all peaks are all "locked" together with only one peak as the "standard" and the energies can only vary as a set. This allows small adjustments to the energy scale of the spectrum to be made to find the "best" fit. The fitting process is very sensitive to small energy variations and the gain and zero of the data (used to find the energy of the data points) are adjusted in each energy region by finding the "local" gain and zero of the data by referencing the data to known isolated strong peaks on each side of the data.

The other peak parameters needed in the fitting process are the peak's width, slope and amplitude of the exponential tail correction. The initial peak parameters, stored in the setup file, are for only two energies and are calculated for other energies from their known energy dependencies. For best results these initial peak parameters should "bracket" the energy range of interest. To assure one set of peak parameters describes the data, when using multiple detectors, the detectors should all be nearly identical. These initial peak shape parameters and the data gain and zero are usually the only required calibration steps needed to analyze the data. For a typical system these calibrations have to only be performed once and then checked periodically.

Assuming the fitting process converged to a reasonable answer, one of the outputs of the fitting process is the "best" amplitude of each isotope's peaks in fitting region. These are used to find the "best" measurement of the isotopic composition of the source. For a source with several peaks, analysis of several different sets of peaks can often help determine the "best" isotopic composition of the source. By using weighted averages of several different fitting operations the effects of imperfect counting statistics and imperfect background choices can be minimized.

Sometimes with "bad" data and/or "too many" free parameters these equations do not converge and no reasonable answer is possible. "locking" one set of peaks at a constant percent of the other set can sometimes circumvent this problem. This effectively eliminates one more free parameter and may allow the procedure to converge. Then the percentage can be varied, the data refit and the percentage that gives the minimum least squared fit found. This procedure is typically only resorted to on data with "bad" statistics.

From this analysis we determine the mass ratios found of: $^{238}$Pu/$^{239}$Pu, $^{240}$Pu/$^{239}$Pu, $^{241}$Pu/$^{239}$Pu, $^{241}$Am/$^{239}$Pu, $^{237}$Np/$^{239}$Pu, $^{235}$U/$^{239}$Pu, and $^{238}$U/$^{239}$Pu. In addition the ratio $^{242}$Pu/$^{239}$Pu is calculated ($^{242}$Pu emits no usable gamma lines) from an algorithm; given the percentage of other isotopes present. All other ratios are found by analyzing closely spaced gamma-ray lines in each energy region. The analysis seeks to find at least two or more measurements, which are averaged after weighting them with the uncertainty in their determination. Because of the statistical nature of the counting data these ratios will always have some uncertainty—sometimes large. Very weak peaks will often only be determined with large uncertainties. Normally this is adequate since weak peaks are usually symptomatic of a small component of the waste sample and contribute only slightly to the total radioactivity of the samples.

# ANALYZING WEAK SAMPLES

The code was designed to analyze "reasonable" samples that have "reasonable" counting statistics. Unfortunately this is not always the case. For weak samples the code was designed to fail "gracefully". If the code could determine that there was plutonium present but could not determine good ratios for any or all of the other isotopes typically present  its default was to assume the plutonium was "weapons" grade composition. The calculations given are then tagged with an estimate flag to indicate that the code was unable to unambiguously determine a given isotope ratio and the "weapons" grade ratios are assumed and listed.

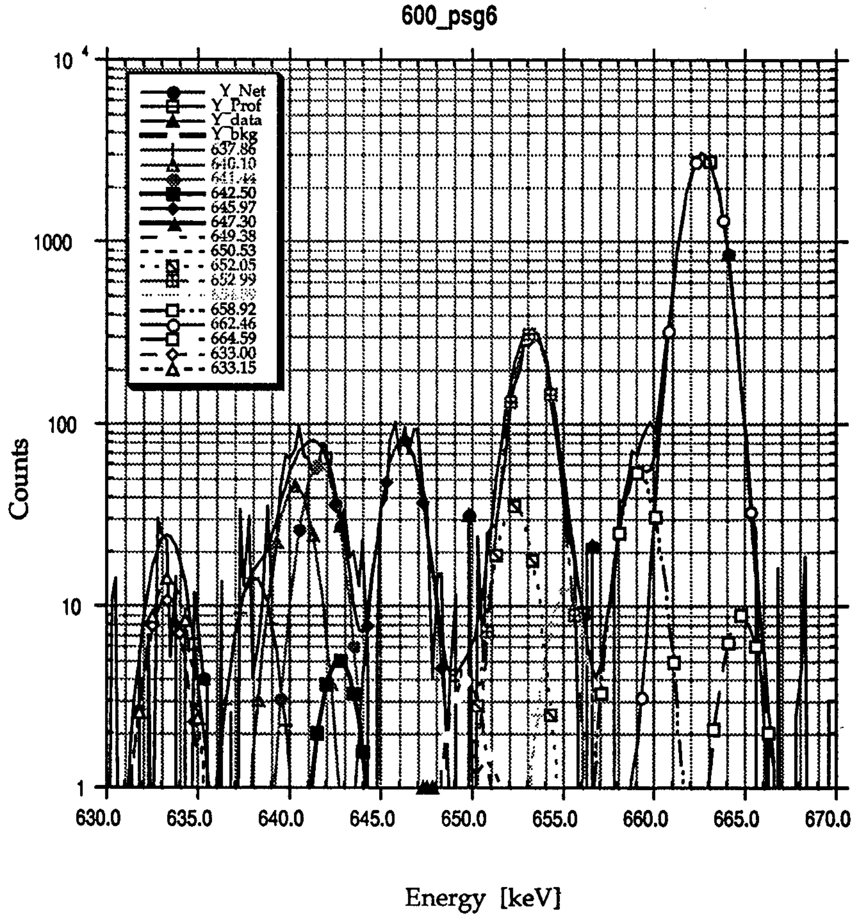

Figure 2. Multi-peak analysis fit of the fifteen peaks typically found in the 630-670 keV region. Shown are each peak independent of its isotopic composition.

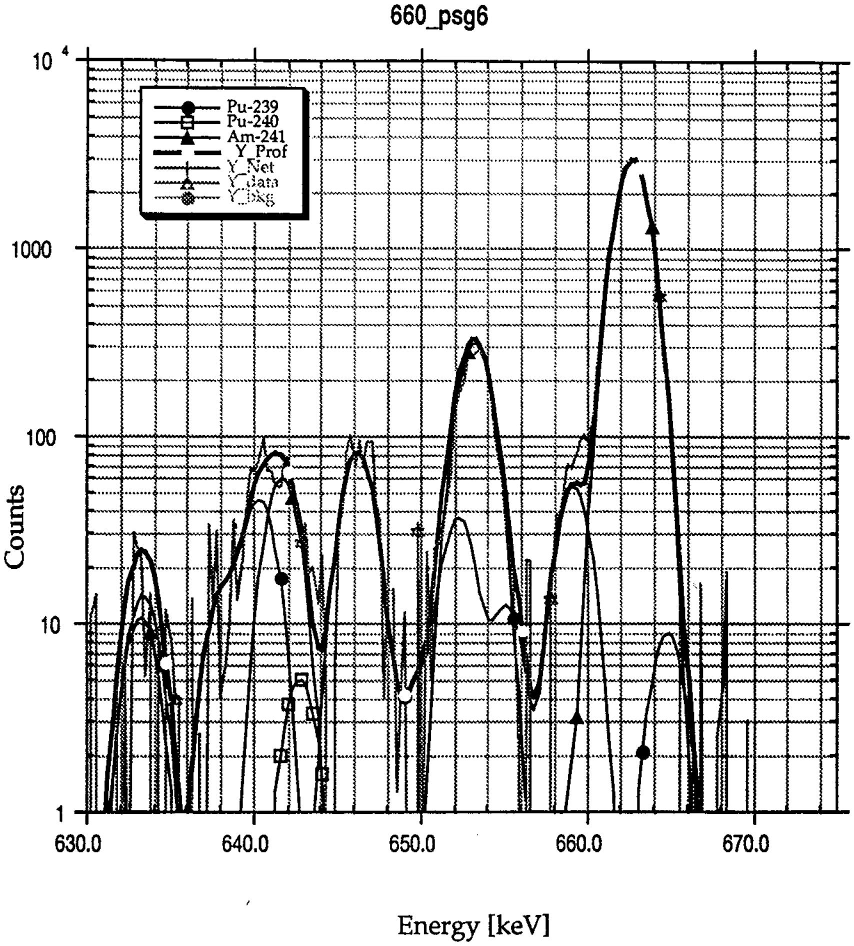

Figure 3. Multi-peak analysis fit of the fifteen peaks typically found in the 630-670 keV region. Shown are the peaks grouped by isotopic composition.

Table 1. Some of the Gamma Ray Energies and branching ratios used in determining isotopic ratios by multi-peak analysis.

| Energy [keV] | Branching Ratio [γ/decay] | Isotope | Halflife | Specific* Power [mW/g] | Notes |
|---|---|---|---|---|---|
| 125.292 | 4.080E-05 | Am-241 | 432.2 y | 114.2 | |
| 129.294 | 6.260E-05 | Pu-239 | 24110.0 y | 1.9288 | |
| 146.194 | 1.190E-06 | Pu-239 | 24110.0 y | 1.9288 | |
| 148.567 | 1.859E-06 | Pu-241 | 14.348 y | 3.412 | |
| 152.680 | 9.560E-06 | Pu-238 | 87.74 y | 567.57 | |
| 158.800 | 4.536E-06 | Pu-242 | 373300 y | 0.1159 | 1 |

| | | | | | |
|---|---|---|---|---|---|
| 160.408 | 4.020E-06 | Pu-240 | 6563 y | 7.0824 | |
| 161.450 | 1.230E-06 | Pu-239 | 24110.0 y | 1.9288 | |
| 164.490 | 6.670E-07 | Pu-241 | 14.348 y | 3.412 | 2 |
| 164.490 | 4.530E-07 | Am-241 | 432.2 y | 114.2 | 2 |
| 185.715 | 5.720E-01 | U-235 | 7.037E+08 y | 5.961E-05 | 3 |
| 203.550 | 5.680E-06 | Pu-239 | 24110.0 y | 1.9288 | |
| 205.311 | 5.010E-02 | U-235 | 7.037E+08 y | 5.961E-05 | |
| 208.000 | 7.900E-06 | Am-241 | 432.2 y | 114.2 | 4 |
| 208.000 | 5.1793E-06 | Pu-241 | 14.348 y | 3.412 | 4 |
| 258.260 | 7.300E-02 | U-238 | 4.468E+09 y | 8.510E-6 | 5 |
| 311.780 | 2.580E-7 | Pu-239 | 24110.0 y | 1.9288 | |
| 312.170 | 3.860E-01 | Np-237 | 2.140E+06 y | 0.02072 | 6 |
| 332.390 | 2.977E-07 | Pu-241 | 14.348 y | 3.412 | 7 |
| 335.440 | 2.494E-08 | Pu-241 | 14.348 y | 3.412 | 2 |
| 335.440 | 4.830E-06 | Am-241 | 432.2 y | 114.2 | 2 |
| 336.116 | 1.135E-06 | Pu-239 | 24110.0 y | 1.9288 | |
| 345.010 | 5.590E-06 | Pu-239 | 24110.0 y | 1.9288 | |
| 375.039 | 1.570E-05 | Pu-239 | 24110.0 y | 1.9288 | |
| 376.600 | 1.348E-06 | Am-241 | 432.2 y | 114.2 | |
| 413.713 | 1.466E-05 | Pu-239 | 24110.0 y | 1.9288 | 8 |
| 415.760 | 1.745E-02 | Np-237 | 2.140E+06 y | 0.02072 | 6 |
| 642.500 | 1.260E-07 | Pu-240 | 6563 y | 7.0824 | |
| 662.456 | 3.670E-06 | Am-241 | 432.2 y | 114.2 | |
| 664.587 | 1.657E-08 | Pu-239 | 24110.0 y | 1.9288 | 9 |

* The power listed is for the total power emitted by the decay of each isotope; not the power of each gamma peak

Notes:

1. The $^{242}$Pu peak is normally too weak to analyze. The $^{242}$Pu/$^{239}$Pu ratio is calculated from the other measured intensities.
2. Gammas produced in decay of $^{241}$Pu-$^{237}$U and $^{241}$Am. Total intensity is proportional to the abundances of these respective isotopes. The individual intensities can be found from the other gamma peaks around it by requiring the entire multiplets of peaks to be fit.
3. The 185.715 keV peak is the main indicator of the presence of $^{235}$U.
4. The 208.000 keV peak is produced in decay of $^{241}$Pu-$^{237}$U and $^{241}$Am; since there are no other $^{241}$Am or $^{241}$Pu peaks near by the individual intensities have to be determined from other measurements.
5. The 258.260 keV peak is from the $^{238}$U daughter $^{234}$Pa (1.17 min ) and is often quite weak.
6. The 312.17 and 415.76 keV peak are from the 26.967 d decay of its daughter $^{233}$Pa.
7. Eleven peaks are fit in the 330-350 keV region
8. The 413.760 keV peak is the main peak normally used in the A&PCT fits.
9. Eleven $^{239}$Pu peaks are fit in the 636 665 keV region.

## TECHNICAL DATA

The isotopic multi-peak analysis code described in this report was written in ANSI Fortran 77/90 and ANSI C. It was compiled using Microsoft's Powerstation compiler [version 4.0] on a Intel based computer to run under Windows NT operating systems. A typical analysis involving reading 1170 files, preparing the sum file and analyzing it takes about two minutes with a 400 Mhz Pentium II processor.

## ACKNOWLEDEMENT

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## REFERENCES:

1. Gunnink, R., *MGA: A Gamma-Ray Spectrum Analysis Code for Determining Plutonium Isotopic Abundances*, UCRL-LR-103220, Vol. 1, Lawrence Livermore National Laboratory, Livermore, CA., 1990.
2. Gunnink, R., Evans J. E., and Prindle A. L., *A reevaluation of the Gamma Ray Energies of Absolute Branching Intensities of* $^{237}U$, $^{238,239,240,241}Pu$ *and* $^{241}Am$., UCRL-52139, Lawrence Livermore National Laboratory, Livermore, CA, 1976.
3. Dragnev, T., "Intrinsic Self calibration of Non-destructive Gamma Spectrometric Measurements--Determination of U, Pu, and $^{241}$Am Isotopic Ratios." *Journal of Radioanalytical Chemistry*, Vol. 36, pp. 491-508, 1977.
4. Firestone, R. B., Shirley, V. S., *Table of Isotopes*, 8th Edition, John Wiley Interscience, New York, 1996.
5. Fazio,P. C., *Annual Book of ASTM Standards, Vol 12.01,C 1030-89, 1989*
6. McIsaac, C. V., Killian E. W., Johnson, L. O., *Description of SWEPP Gamma-Ray Spectrometer System Version 2.0 Software Calculation Methods*, INEL-96/0484 Rev. 1, Idaho National Engineering Laboratory, Lockheed Martin Idaho Technologies company, Idaho Falls, Idaho, 1996.