CONF-950923--14

LA-UR- 95-3326

Title: **PC/FRAM: ALGORITHMS FOR THE GAMMA-RAY SPECTROMETRY MEASUREMENT OF PLUTONIUM ISOTOPIC COMPOSITION**

Author(s): **T. A. Kelley, T. E. Sampson, and D. M. DeLapp**

Submitted to: **American Nuclear Society "Fifth International Conference on Facility Operations—Safeguards Interface." Jackson Hole, Wyoming USA September 24-29, 1995 (FULL PAPER)**

MASTER

Los Alamos
NATIONAL LABORATORY

Form No. 836 R5

PC/FRAM: ALGORITHMS FOR THE GAMMA-RAY SPECTROMETRY MEASUREMENT OF PLUTONIUM ISOTOPIC COMPOSITION[a]

Thomas A. Kelley, Thomas E. Sampson, and Dorothea DeLapp
Los Alamos National Laboratory
Los Alamos, New Mexico 87545 USA
(505) 665-5684

ABSTRACT

The Safeguards Program at Los Alamos National Laboratory has developed versatile software for the isotopic analysis of SNM in a sample. The FRAM code has been used routinely at LANL for years. Its capability has now been greatly expanded, and it has been given a graphical user interface. Some of the details on the internal workings of the code are given in this paper.

I. INTRODUCTION

The FRAM code[1,2] was originally designed by the late George Nelson of the University of Arizona and Thomas Sampson of LANL to analyze a gamma-ray spectrum and estimate the isotopic composition of plutonium in the sample that produced that spectrum. The authors of this paper have enhanced this code and converted it to run on a PC under Windows 3.1. This paper will describe the algorithms used in the analysis and explain the software features that make this a useful tool. A companion paper at this symposium describes the new measurement capabilities of the code.[3]

II. INPUTS AND OUTPUTS

PC/FRAM requires a spectrum taken with a high-resolution gamma-ray detector along with a set of parameters that drives the analysis. PC/FRAM generates estimates of the plutonium isotopic ratios represented by that spectrum and voluminous secondary information about the analysis performed.

The spectrum may come from a multichannel analyzer or from a disk file. The code currently supports the collection of data with a Canberra S100 or an ORTEC multichannel buffer. The code can interpret the storage format for data from a disk file used either by the S100 or by the ORTEC. It can also read data stored in a text file if that data is properly structured.

The parameters that direct the analysis of a spectrum are an important part of PC/FRAM. They describe the default calibration settings, the gamma-ray peaks to search for, the regions of interest, the isotopes to consider, and a number of application-specific constants. If properly designed, these parameters can be used for a relatively wide variety of spectra. Certainly not all spectra can be accommodated by a single set of parameters, so the code's database was constructed to handle multiple parameter sets. A utility function is built into the software to allow the user to view or modify the parameter values in any given set.

The complete results of each analysis are written to temporary files on disk. A summary of those results is displayed on the screen; the summary includes the estimated isotopic fractions, the specific power of the sample, and the effective ^{240}Pu fraction.

III. INTERNAL CALIBRATION

In the first stage of the analysis, PC/FRAM performs an internal calibration. Selected peaks in the spectrum are used to provide a calibration of energy vs channel, full width at half maximum (FWHM) vs energy, and peak shape (tailing parameters) vs energy. This means that the analysis does not depend on measurements that may have been taken under different conditions such as count rate, resolution, or other electronic adjustments. The internal calibration is performed with the same unknown spectrum that is being analyzed.

A. Energy

The initial energy calibration is used to locate peaks at those energies specified in the parameter set. The raw counts in the peak channel and two channels on either side are used to determine a peak centroid. PC/FRAM constructs a table of centroid-energy

[a]This work is supported by the US Department of Energy, Office of Nonproliferation and National Security, Office of Safeguards and Security, and Stockpile Support Program.

pairs. For the rest of the analysis, interpolations in this table are made to convert between channel and energy.

B. FWHM

Before the internal calibration proceeds any further, the background underneath each peak region is determined. This guarantees that an initial set of net counts is available.

For each of the peaks in this calibration, we choosea range of channels around the centroid: those channels below the centroid whose net counts exceed 75% of the maximum and those channels above the centroid whose net counts exceed 25% of the maximum. Then a quadratic curve is fitted to the logarithm of the net counts. From this, the FWHM in channels is estimated and stored away with the peak energy E. When all the FWHM's are computed, the coefficients in the following model are determined by a linear least squares analysis.

$$(FWHM)^2 = A_1 + A_2 E + A_3 E^{-1}$$

This equation is used in the rest of the analysis to estimate the FWHM of a peak at any given energy.

C. Shape

The shape of a gamma-ray peak in the spectrum is described by a central Gaussian component with a single exponential tail on the low energy side of the peak.

$$Y(x) = H\left\{\exp\left[-\alpha(x - x_0)^2\right] + Tail(x)\right\}$$

where

$Y(x)$ is the net count in channel x,

H is the peak height at the peak centroid x_0,

$\alpha = 4\ln(2)(FWHM)^{-2}$ the peak width parameter,

$$Tail(x) = \exp\left[(T_1 + T_2 E) + (T_3 + T_4 E)(x - x_0)\right] r(x - x_0),$$

$r(x) = 1 - \exp(-0.4\alpha x^2)$ if $x \le 0$ and $r(x) = 0$ otherwise.

We have allowed both the amplitude, $\exp(T_1 + T_2 E)$, and slope, $T_3 + T_4 E$, of the tailing function to depend on the energy. In practice however we set T_4 to zero.

The peaks that are to be used to determine these tailing coefficients are specified in the parameter set. For each of these peaks, the Gaussian portion of the peak can be determined because we now have a calibration for FWHM vs energy. A range of channels extending from 1.5 FWHM's below the centroid to 0.5 FWHM's below the centroid is selected and in each one the Gaussian portion of the peak is subtracted from the net counts. All usable data is collated together across the spectrum. Then a linear least squares fit of the model is made to the data.

IV. ANALYSIS

In the second stage of the analysis, each region is examined in the order listed in the parameter set. For each region a continuum background is estimated, then the necessary response functions are created, and finally these response functions are fitted to the net counts so as to determine the peak areas. The peak areas are used to calculate a relative efficiency function across the spectrum and then to estimate the relative activities of the isotopes. This whole process is usually repeated two more times. The backgrounds are adjusted. The relative efficiencies and activities are used to better separate peaks in a multiplet. The new areas are used to produce better relative efficiencies and activities.

A. Background

For each region, 1, 2, 3, or 4 intervals of channels may be specified as background. The raw counts in these channels are used to construct a background function. Several choices of background functions, $bkg(x)$, are available to the user. In the following descriptions, a data point consists of the pair (x,y) where y is the net counts in channel x.

none	$bkg(x) = 0$
flat	The equation $bkg(x) = C$ is fitted to the data points.
linear	The equation $bkg(x) = C_1 + C_2 x$ is fitted to the data points.
quadratic	The equation $bkg(x) = C_1 + C_2 x + C_3 x^2$ is fitted to the data points.

exponential The equation $bkg(x) = \exp(C_1 + C_2 x)$ is fitted to the data points.

flat step The equation $bkg(x) = C_1 + C_2 F(x)$ is fitted to the data points where $F(x) = S(x)/S(xhigh)$. $S(x)$ is the cumulative sum of the net counts from the very first bkg channel, xlow, to channel x which can be as large as the very highest bkg channel, xhigh. Initially the raw counts are used in place of the net counts. After a background is determined, it is used in a second pass through the algorithm. The second determination of the background is used as the background for the peak region.

linear step The equation $bkg(x) = C_1 + C_2 F(x) + C_3 x$ is fitted to the data points where F(x) is defined above. This imposes a smooth step function on top of a straight line so that the step function has a definite slope at each end.

bilinear step The equation $bkg(x) = C_1 + C_2 F(x) + C_3 x + C_4 x F(x)$ is fitted to the data points where F(x) is defined above. This is a smooth step function with different slopes at either end.

B. Response Functions

In the simplest case, one response function $R(x)$ is constructed for each peak in a peak region. The function $R(x)$ has the same form as described in III. C above except that the tail portion is written as $Tail(x) = A\exp(B(x - x_0))r(x - x_0)$ and it is normalized to have a unit area. The normalization factor can be determined by analytically computing the area of the function in III. C assuming that $H=1$. It is given by the following formula.

$$Area = \sqrt{\frac{\pi}{\alpha}} + \frac{A}{B}\left[1 - \sqrt{\pi} D \exp(D^2) erfc(D)\right]$$

where $D = \dfrac{B}{\sqrt{1.6\alpha}}$

You can, however, specify that a peak in a region be fixed to another peak in that region. In this case the former peak will depend on the latter peak in the sense that its area will be forced to be a certain fraction of the peak it is fixed to. For each free peak in a region, i.e., one that is not fixed to any of the others, a response function $\sum f_i R_i(x)$ will be constructed where each $R_i(x)$ is a unit-area function describing the shape of a photo peak and f_i is the associated area factor. The area factor for the free peak is one, but if peak i is fixed to peak j, the area factor will be $f_i = (BR_i / BR_j)(RE_i / RE_j)(RA_i / RA_j)$ where BR stands for branching ratio, RE stands for relative efficiency, and RA stands for relative activity. At the start, the relative efficiencies and activities are set to one. The dependence here on quantities that are the eventual output of the analysis is the major reason why the analysis is done three times. The output from one iteration can be fed back into the analysis for the next iteration to improve the areas which will improve the output, and the process can be repeated.

C. Peak Areas

When the response function has been constructed, a least squares fit to the net counts is performed using the model

$$Y(x) = \sum_j C_j \sum_i f_i R_i(x)$$

where Y is the net count at channel x, the outer sum ranges over the free peaks in the region, and the inner sum ranges over the peaks fixed to a free peak (including the free peak itself). The area of each peak is given by the product $C_j f_i$.

D. Relative Efficiencies

For the peaks in the parameter set that are tagged to be used in determining the relative efficiencies, let N be the number of isotopes represented and M the number of efficiency functions chosen. PC/FRAM uses the following empirical model.

$$Y = C_1 + C_2 E^{-2} + C_3(\ln E) + C_4(\ln E)^2 + C_5(\ln E)^3 + C_i + C_j E^{-1}$$

In this formula Y is the logarithm of the ratio of the peak area to its branching ratio. There are N-1 terms C_i for the isotopes beyond the first one. There are M-1 terms C_j for the different efficiency functions beyond the first one. A linear least squares analysis is performed to determine the unknown coefficients which in turn defines the efficiency function used in the rest of the analysis.

E. Relative Activities

The model

$$Area = \sum_i C_i \sum_j (BR_j)(RE_j)$$

is used to calculate relative activities of the isotopes. In this formula, the outer sum ranges over the isotopes and the inner sum includes the peaks belonging to that isotope and any other peaks "summed" with them. A linear least squares analysis is performed to determine the coefficients, which are the required relative activities.

V. SUMMARY

After the last iteration, the final relative activities are converted to relative masses. These are then used to compute the absolute isotopic fractions without ^{242}Pu. The amount of ^{242}Pu is either entered by the operator at the time an analysis is requested or it is calculated by the following correlation scheme.

$$^{242}Pu = A * (^{238}Pu)^B * (^{239}Pu)^C * (^{240}Pu)^D * (^{241}Pu + ^{241}Am)^E$$

The isotopic fractions are then renormalized to account for ^{242}Pu.

Two other results are reported. The first is the specific power of the sample and the other is the effective ^{240}Pu fraction. Both of these quantities are calculated from the isotopic fractions and appropriate constants found in the parameter set.

VI. SOFTWARE TOOLS

PC/FRAM comes with a number of software tools that are very useful for the spectroscopist. They are all invoked by selecting particular options in the menu system provided by the program.

A. Setting Defaults

A number of operations performed by this program depend on information supplied by the user. This information is collected by means of "dialog boxes" on the screen. Although these may seem complicated for routine use by a novice, it is possible to edit these dialog boxes and establish default values for all the required responses. If these do not change over a period of time, one can get past them with as little as a single keystroke or mouse click.

B. Change Parameter Utility

This utility allows you to manipulate the parameter sets in the database. When you open a parameter set, you transfer it to a "worksheet" in memory. There you can choose to view or modify any of the parameter values or both. The information about the peaks, regions, and isotopes will be displayed as a spreadsheet. You may save the changes back to the database or even save the whole worksheet as a new parameter set in the database. You generate a hard copy listing of the parameter values on the system printer. You can "export" the parameters, i.e., save them in text format in a disk file. You can "import" parameter values from a previously exported text file. This allows for the transferal of information from one system to another. You can remove a parameter set from the database. You can even delete all the parameter sets from the database and start from scratch. In Fig. 1 there is an example of how the isotope information is organized and presented to the user for review or modification or both.

C. Display of Spectrum

When spectral data is read into memory either from a multichannel analyzer or from a disk file during an analysis, it stays resident in memory until overwritten later. One of the options available in the program allows you to graphically view this spectrum. The window in which the spectrum is plotted also has a number of controls which give you many of the features of an MCA emulator. You can expand in either the horizontal or the vertical direction. You can scroll the spectrum either horizontally or vertically. You can manipulate a cursor and tell what the counts are for any given channel. But in addition to all this, you can read in a parameter set and have its region information overlaid on the plot of the spectrum. Points belonging to a region interval will be plotted in red while points belonging to a background interval will be plotted in green. This is enormously useful in determining how well the chosen regions match the type of spectrum you have. In Fig. 2 there is an example of a spectral display as it would first appear in the window.

D. Display of Fits

A complete set of the results of each analysis is saved on disk. One of the options available in the program allows you to view the results of the

response function fits for each region. A plot is generated on screen for a given region with the spectral counts plotted as black points, the calculated background plotted as a green line, and the sum of all the response functions plotted as a red line. You can choose to display this for any of the regions used in the analysis. You can expand in either the horizontal or the vertical direction. You can scroll the spectrum either horizontally or vertically. You can manipulate a cursor and tell what values correspond to any given channel on any of the three plots. An example of such a plot appears in Fig. 3.

E. Display of Efficiencies

One of the options available in the program allows you to view the results of the relative efficiency calculations. You can view a plot of the relative efficiency curve that the program produced along with the data points used to generate that curve. An example of this plot appears in Fig. 4.

F. Display of Results

For the very last analysis or for any previous analysis in which the results were saved, those results can be displayed on the screen or sent to the printer in various levels of detail.

REFERENCES

1. Thomas E. Sampson, George W. Nelson, and Thomas A. Kelley, "FRAM: A Versatile Code for Analyzing the Isotopic Composition of Plutonium from Gamma-Ray Pulse Height Spectra," Los Alamos National Laboratory report LA-11720-MS (December 1989).

2. Thomas E. Sampson, George W. Nelson, and Thomas A. Kelley, "FRAM: A New, Versatile Gamma-Ray Spectrometry Code for Measuring the Isotopic Composition of Plutonium," in *Nuclear Materials Management* (Proceedings Issue) XIX, 420 (1990).

3. Thomas E. Sampson, Thomas A. Kelley, Teresa L. Cremers, Robert J. Friar, and Todd R. Konkel, "PC/FRAM: New Capabilities for the Gamma-Ray Spectrometry Measurement of Plutonium Isotopic Composition," (these proceedings).

APPENDIX

OK Cancel

	isotope	half-life	units	mass	power (mw/g)	pu240 coef.	eff. function
1	Pu239	2.4119e+004	years	239.05220	1.92880	0.0000	1
2	Pu241	1.4348e+001	years	241.05690	3.41120	0.0000	1
3	Am241	4.3380e+002	years	241.05679	114.20000	0.0000	1
4	Pu238	8.7740e+001	years	238.04961	567.57000	2.5200	1
5	Pu240	6.5640e+003	years	240.05380	7.08240	1.0000	1
6	Pu242	3.7630e+005	years	242.05874	0.11590	1.6800	1
7							
8							
9							
10							
11							
12							
13							
14							

Fig. 1. Example of the spreadsheet presentation of isotope information in a particular parameter set.

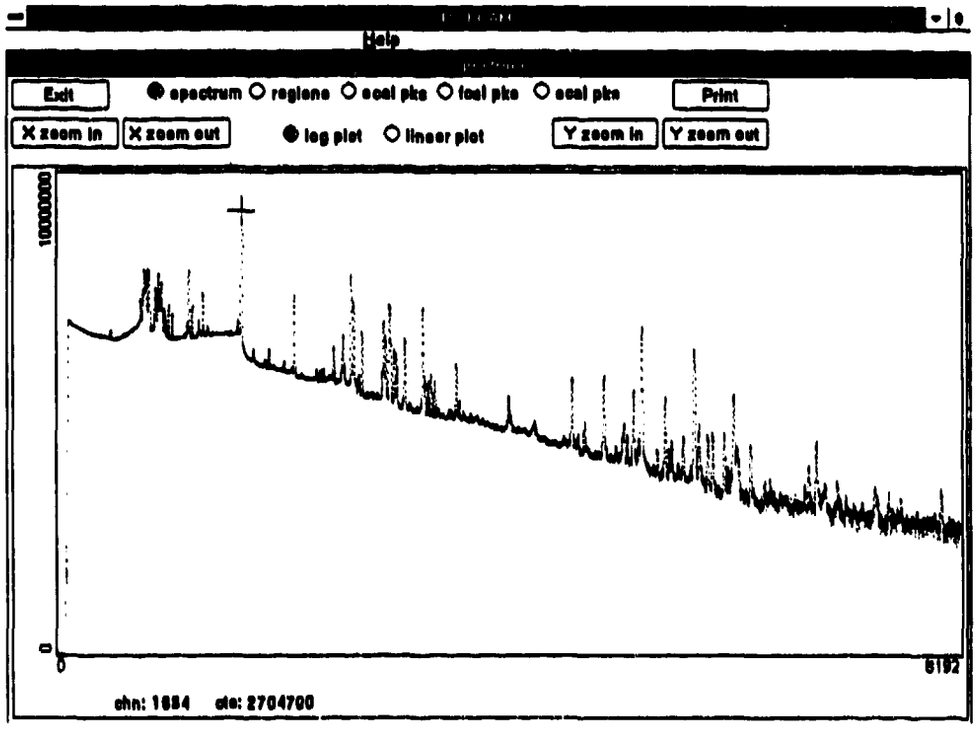

Fig. 2. Example of a spectral display.

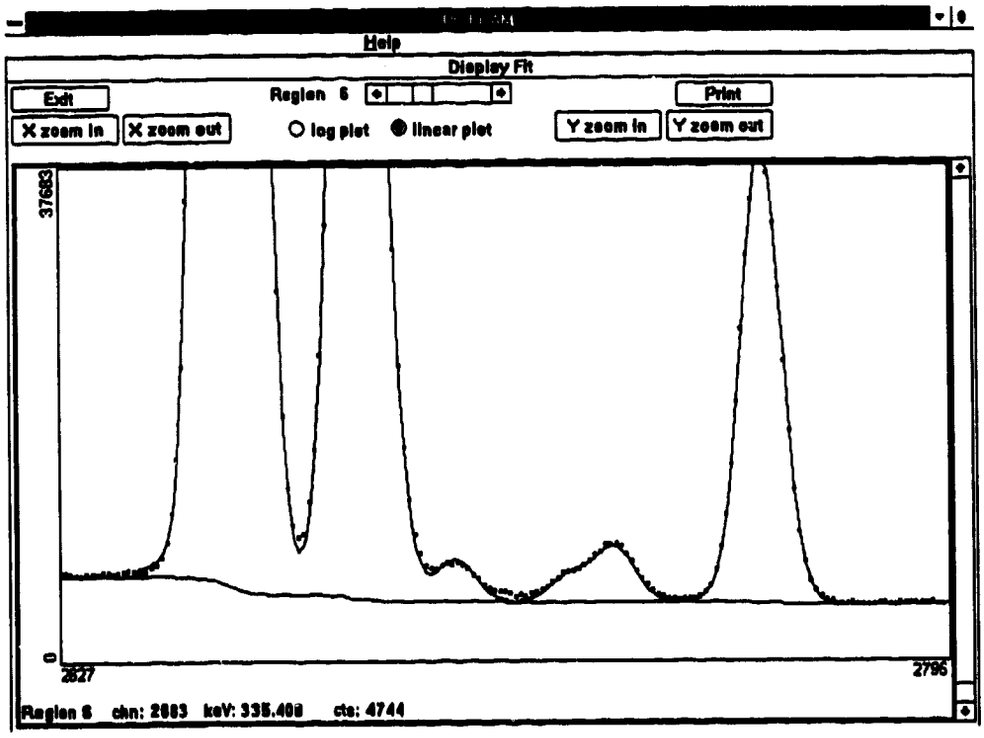

Fig. 3. Example of a display of the results of the fitting process for a particular region.

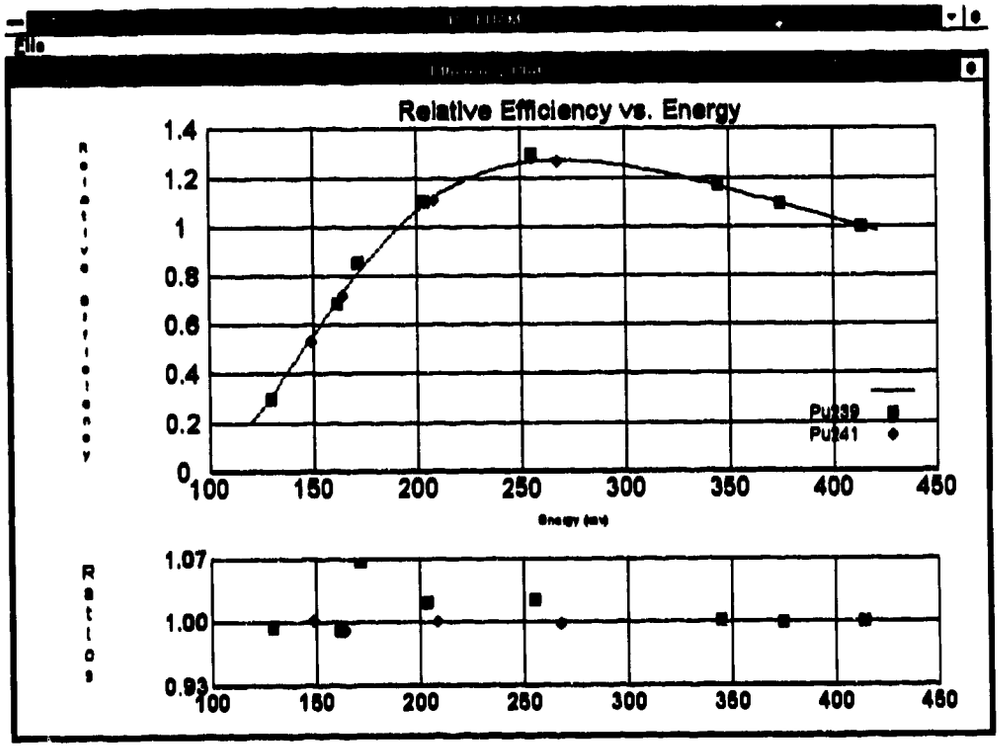

Fig. 4. Example of a display of the relative efficiency curve generated during the course of an analysis.