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*Title:* **THE LOS ALAMOS PC/FRAM CODE FOR THE NONDESTRUCTIVE ANALYSIS OF THE ISOTOPIC COMPOSITION OF PLUTONIUM AND OTHER ACTINIDES**



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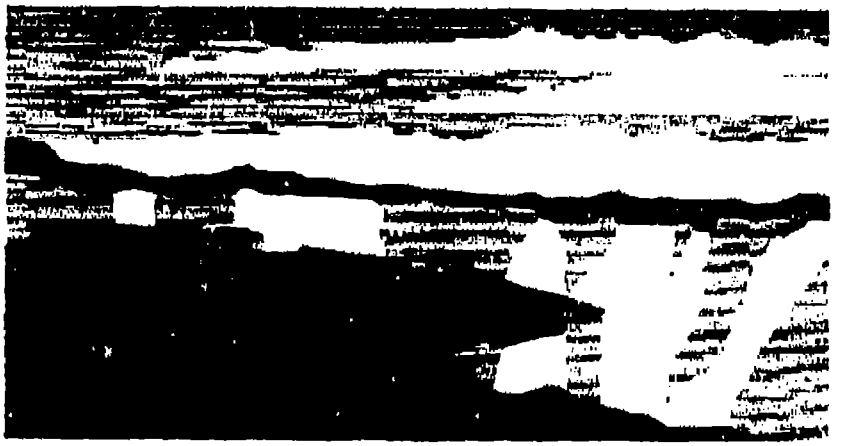

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## DISCLAIMER

# THE LOS ALAMOS PC/FRAM CODE FOR THE NONDESTRUCTIVE ANALYSIS OF THE ISOTOPIC COMPOSITION OF PLUTONIUM AND OTHER ACTINIDES

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## ABSTRACT

The Safeguards Program at Los Alamos National Laboratory has developed versatile software for the isotopic analysis of plutonium and other actinides from the gamma ray spectrum of an arbitrary sample. These developments began over 20 years ago and have been used routinely at Los Alamos for 15 years. We will present details of the PC/FRAM code as well as discussing its application to a wide variety of measurement problems.

## OVERVIEW

PC/FRAM requires a spectrum taken with an HpGe 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 memory or from a disk file. The code currently supports the collection and analysis of data from a Canberra S100, an ORTEC multichannel buffer, and the Los Alamos M3CA. It can also read data stored in a properly-structured text file.

The parameters that direct the analysis of a spectrum describe the default calibration settings, the gamma-ray peaks to be analyzed, the regions of interest, the isotopes to consider, and a number of application-specific constants that govern customized diagnostic tests, parameters for the $^{241}$Pu correlation, and other functions.

A properly-designed set of parameters can be used for a relatively wide variety of spectra. Because not all spectra can be accommodated by a single set of parameters, the code's database was constructed to handle multiple parameter sets. A utility function built into the software allows the user to view or modify the parameter values without reprogramming the software.

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## 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, detector resolution, or other electronic adjustments. The internal calibration is performed with the same unknown spectrum that is being analyzed. All the peaks used in the internal calibrations are specified in the parameter set selected for analysis. A separate list of calibration peaks may be specified for each parameter. The calibration parameters may be set to default values for analysis of spectra with weak peaks or poor counting statistics.

### Energy

The energy calibration is a piecewise linear calibration between the pairs of peaks specified in the parameter set.

### FWHM

The FWHM of the designated internal calibration peaks is obtained from a quadratic curve fit to the logarithm of the net counts. 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. The

third term in the model accommodates the observation that these curves do not always follow the expected linear relationship at low energies.

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 $x_0$,
$\alpha = 4\ln(2)(FWHM)_i^{-2}$ the peak width parameter,
$Tail(x) = \exp\left[(T_1 + T_2E) + (T_3 + T_4E)(x - x_0)\right]$
$r(x - x_0)$,
$r(x) = 1 - \exp(-0.4\alpha x^2)$ if $x \le 0$ and $r(x) = 0$
otherwise.

We have allowed both the amplitude, $\exp(T_1 + T_2E)$, and slope, $T_3 + T_4E$, 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 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.

## 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.

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 are available to the user.

| | |
|---|---|
| *none* | no background subtraction |
| *flat* | a straight line with zero slope |
| *linear* | a sloping straight line |
| *quadratic* | a parabolic function |
| *exponential* | gives a background with mild curvature |
| *flat step* | a smoothed step function with zero slope at the ends |
| *linear step* | a smoothed step function superimposed on a sloping straight line |
| *bilinear step* | a smoothed step function with different slopes at either end. |

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 for the gamma ray peak shape 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 peak shape function 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, also, specify that a peak in a region be fixed to another peak in that region. 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 unity. The dependence here on quantities that are the eventual output of the analysis is the major reason why the analysis is repeated three times.

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 $C_j f_i$.

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 for the relative efficiency.

$$Y = C_1 + C_2E^{-2} + C_3(\ln E) + C_4(\ln E)^2 + C_5(\ln E)^3 + C_i + C_jE^{-1}$$

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. The capability for multiple relative efficiency curves improves the analysis for isotopically heterogeneous materials.

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.

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.

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

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

## APPLICATION

PC/FRAM can be used with either a single planar or a single coaxial detector. With the single planar detector, the traditional mode of operation, the analysis range is typically from 120–420 keV.

The current emphasis has been to acquire one spectrum with a single coaxial detector spanning the energy range from 0 to 1024 keV in 8192 channels. We use a coaxial high-purity germanium (HpGe) detector of approximately 25% relative efficiency with nominal low-rate, long-time-constant resolution of <1.75 keV at 1332 keV and <750 eV at 122 keV. With this single detector and single set of data acquisition conditions, we are able to acquire and analyze data from both "normal" and "shielded" items without changing any measurement conditions. If the sample is "normal," meaning gamma rays in the 120 to 200-keV range are present with meaningful intensities, we usually obtain the best results by analyzing data in the energy range from 120-450 keV. If these lower-energy gamma rays are absent, usually because the item is packaged in a heavy-walled or shielded container, we analyze the spectrum remaining above 200 keV, obtaining $^{240}Pu$ at 642.5 keV and $^{239}Pu$ at 766.4 keV.

The user-editable parameter file structure permits the code to easily be adapted to many measurement problems without reprogramming. Some of the types of items that have been measured are shown in Table I. We particularly note that this single code can also measure the isotopic distributions in uranium by choosing the appropriate parameter set. Figure 1 displays typical spectra from "shielded" and unshielded spectra

Table I

| Material Categories Analyzed with the PC/FRAM Code | |
|---|---|
| • 2 - 38% $^{240}Pu$ | • 0.01 - 50% $^{241}Am$ |
| • Interferences from other actinides (Np, Cm) | • 80% $^{238}Pu$ |
| • Lead-shielded samples | • Heterogeneous Am/Pu |
| • Non-equilibrium $^{241}Pu$-$^{237}U$ | • MOX: $^{235}U$/Pu from 0.005 - 35 |
| • $^{235}U/^{238}U$ in uranium (only), no Pu | • $^{235}U$:$^{241}Am$:Pu = 24:1:1 |

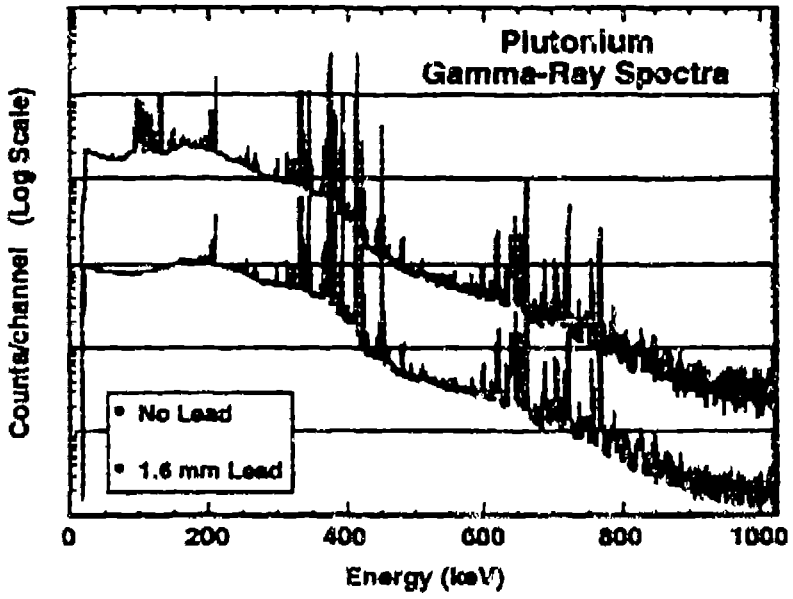



*Fig. 1. Coaxial-detector gamma-ray spectra from shielded and unshielded low-burnup plutonium. A small amount of shielding removes all useful gamma rays below 200 keV. The PC/FRAM code allows a complete analysis using only gamma rays above 200 keV and has demonstrated measurements through as much as 6mm of lead. This shielding thickness may be further extended by starting the analysis at 300 keV.*

The most significant new application permitted by the PC/FRAM code is the complete isotopic analysis of spectra acquired with a single coaxial detector from items in shielded or heavy-walled containers. Facility operators will not have to unpackage items in shielded containers before performing an isotopic measurement. This avoids the additional radiation exposure that this extra handling produces. This new application also permits safeguards inspectorates to verify or measure items in heavy-walled storage containers without opening the container or handling the items: actions that might compromise sensitive or classified information. In Fig. 2 we show measurement results for the two most important parameters extracted from plutonium isotopic measurements, the effective specific power $P_{eff}$ used with calorimetry measurements and the effective $^{240}Pu$ fraction $^{240}Pu_{eff}$ used with neutron coincidence counting measurements.

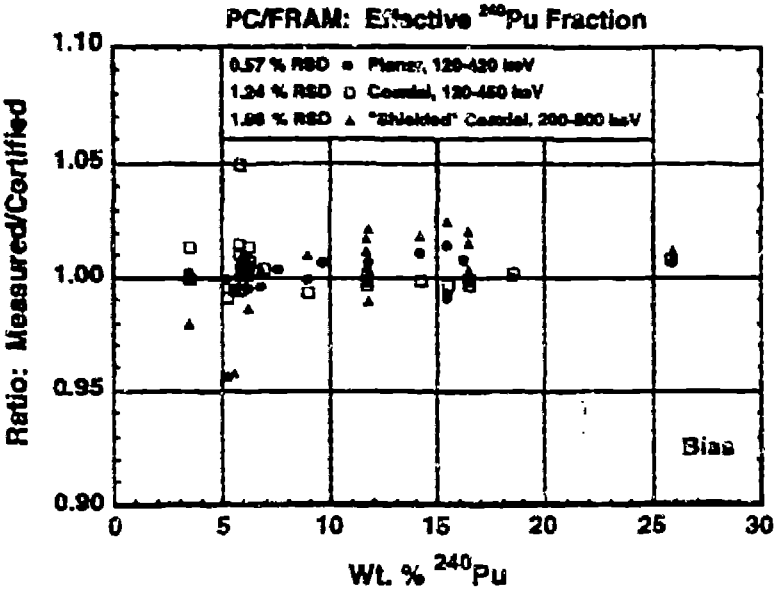
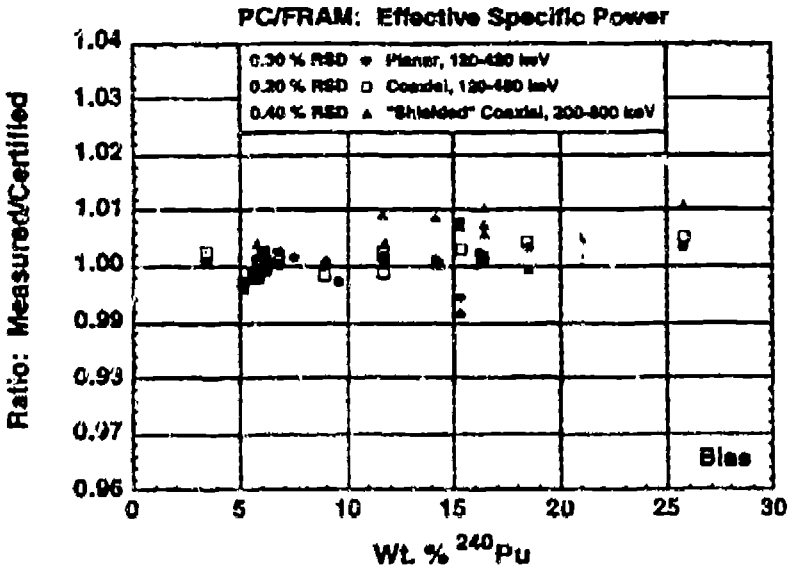



*Fig 2. Typical measurement bias for widely-used analysis results. (both figures)*




Under good conditions most isotopes can be measured with an accuracy of ~ 1%. Counting times may be as low as 5-10 min but are usually recommended to be 30 min to one hour. Measurement precision or repeatability at the 1 RSD level for $^{240}$Pu for 30 min to one hour measurements is typically 1-2% for low energy measurements (120-420 keV) with planar or coaxial detectors and 2-3% for "shielded" samples (> 100 gPu) measured from 200-800 keV with a coaxial detector. For low burnup Pu the precision for $P_{eff}$ is typically a factor of five smaller than that for $^{240}$Pu.

## CONCLUSIONS

We can accurately and precisely measure the plutonium isotopic composition of arbitrary samples contained in heavy-walled or shielded containers, packaging configurations heretofore unmeasurable. We carry out these measurements with a single coaxial HpGe detector which, without changing instrument settings, can acquire and analyze data from 120 keV to above 800 keV. The flexibility of the new PC/FRAM code with its user-editable parameter file allows these measurements without software reprogramming.

This enhanced capability now allows operators to measure shielded containers without handling and unpacking them before measurement. This significantly reduces the radiation dose to plant personnel and increases safety margins through less handling of plutonium-bearing containers.

The measurement accuracy and precision for $P_{eff}$ and $^{240}Pu_{eff}$ for coaxial detectors are entirely adequate for most measurements and in some cases meet or exceed the quality of measurements from traditional planar detectors.

This capability is currently being installed in numerous facilities in Russia under the auspices of the US-Russia Laboratory-to-Laboratory Program.