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PC/FRAM: A Code for the Nondestructive Measurement of the Isotopic Composition of Actinides for Safeguards Applications

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1. ABSTRACT

The Nuclear Safeguards Program at the Los Alamos National Laboratory has developed and fielded techniques for the gamma-ray spectrometry measurement of the isotopic composition of plutonium and other actinides for over 20 years, ever since Parker and Reilly first proposed a practical method for the measurement of the arbitrary sample. Their procedures, incorporating internal (to the measured gamma-ray spectrum) or "intrinsic" self-determination of the relative efficiency function of the sample-detector measurement system, are widely applied today.

The PC/FRAM code is the most recent and most highly-developed Los Alamos code developed specifically for the nuclear safeguards applications of this technique. We will describe the measurement principles that allow accurate measurements to be taken on samples of arbitrary size, shape, and measurement geometry—and of arbitrary physical and chemical composition—through the use of known nuclear decay data (half-lives and branching intensities). Subsequently, we will describe the analysis methodology, which is driven by an easily edited parameter file that frees the user from dependence on a dedicated programmer for analyses of special cases. This methodology relies on internal gamma-ray peaks from the spectrum under analysis to self-calibrate the unknown spectrum for energy and peak shape (energy dependence of full width at half maximum (FWHM) and tailing parameters). The program uses these parameters to calculate response functions that are fit to the analysis peaks requested in the parameter file.

The structure of the code and its Windows 3.1 user interface allows use with equal ease by the experienced spectroscopist or operator-level personnel in a working facility. The code has been applied primarily to the analysis of gamma-ray spectra from plutonium with the isotopic results used to calculate the parameters $^{240}\text{Pu}_{\text{eff}}$ for interpretation of neutron coincidence counting assays and P_{eff} (W/gPu) for the interpretation of calorimetry measurements. These measurement combinations form the backbone of nondestructive assay measurements for materials control and accountability both domestically and internationally. We will present examples of the results of measurements of these parameters with PC/FRAM and discuss the wide range of materials measured, including uranium samples and samples with lead shielding.

2. INTRODUCTION

The analysis of gamma-ray spectra from plutonium to yield the isotopic fractions of ^{238}Pu - ^{242}Pu , ^{241}Am , and other constituents is, perhaps, the most widely used nondestructive measurement method in the field of nuclear safeguards. Many bulk

nondestructive assay methods quantify only a single isotope or measure a quantity that is not a direct function of the mass of the measured item. These measurement methods must rely on knowledge of the isotopic composition of the plutonium and other constituents of the measured item to interpret the bulk measurement in units of elemental plutonium mass. The two most common applications of the interpretation of a bulk measurement using knowledge of the isotopic distribution are the interpretation of neutron coincidence counting and calorimetry measurements of bulk samples.

Neutron coincidence counting measures only the even isotopes of plutonium and yields a result for the effective ^{240}Pu mass as:

$$^{240}\text{Pu}_{\text{eff}}(g) = 2.52 * \text{grams}^{238}\text{Pu} + 1.00 * \text{grams}^{240}\text{Pu} + 1.68 * \text{grams}^{242}\text{Pu} \quad (1)$$

where $^{240}\text{Pu}_{\text{eff}}$ is the mass of ^{240}Pu that gives the same coincidence response as that obtained from the actual sample. If one defines the effective ^{240}Pu fraction in the same manner and determines this fraction by measurement of the isotopic composition, we then obtain the elemental plutonium mass by

$$\text{grams Plutonium} = \text{grams}^{240}\text{Pu}_{\text{eff}} / \text{fraction}^{240}\text{Pu}_{\text{eff}} \quad (2)$$

In a similar fashion calorimetry measures the total sample power in watts. The measured isotopic fractions are combined with the known specific powers (watts/gram isotope) for each heat-producing isotope in the sample to produce the sample's effective specific power P_{eff} in watts/g Pu. The elemental plutonium mass is given by

$$\text{grams Plutonium} = \text{watts (from calorimeter)} / \text{watts/g Pu (from isotopics)} \quad (3)$$

These two measurement combinations are used worldwide for the nondestructive assay of plutonium. The common element is that both require an accurate knowledge of the isotopic composition of the plutonium in the measured item. These isotopic distributions may be conveniently and accurately measured by gamma-ray spectrometry. A review of the characteristics of the gamma-ray spectrum of the isotopes of plutonium and the general features of isotopic measurements may be found in Ref. 1.

3. HISTORY

Work on the isotopic analysis of plutonium in samples began in Los Alamos over 20 years ago when Parker and Reilly² were the first to propose a practical method for measuring the arbitrary sample. Their same principles, incorporating internal or "intrinsic" self-determination of relative efficiency, are still applied today and form the physical basis for the PC/FRAM code described in this paper.

These measurements are performed on samples of arbitrary size, geometry, and physical and chemical composition, and the results are obtained without calibration using only fundamental tabulated nuclear constants. The fundamental measurement is one of isotopic ratios given by

$$N^i/N^k = C(E_j^i)/C(E_j^k) * T_{1/2}^i/T_{1/2}^k * BR_j^k/BR_j^i * RE(E_j^i)/RE(E_j^k) \quad (4)$$

where

N^i	=	Number of atoms of isotope i
$C(E_j^i)$	=	Photopeak area of gamma-ray j with energy E_j emitted from isotope i
$T_{1/2}^i$	=	Half-life of isotope i
BR_j^i	=	Branching ratio (gamma rays/disintegration) of gamma-ray j from isotope i
$RE(E_j)$	=	Relative detection efficiency of gamma ray with energy E_j . Includes detector efficiency, measurement geometry, sample self-absorption, and attenuation in materials between the sample and detector.

The relative efficiency is determined from each measured spectrum by fitting the energy variation of the quotient of C/BR for a series of gamma rays from a single isotope.

The PC/FRAM code is a third-generation isotopic analysis code developed at Los Alamos. References 3-6 describe the first generation code which used simple region-of-interest summation techniques to obtain peak areas. The second generation code, FRAM, ran on a VAX system and is described in References 7-10. The PC/FRAM code is also discussed in References 11-12.

4. PC/FRAM CHARACTERISTICS

4.1 Single Detector System

Like all previous Los Alamos isotopic analysis codes, PC/FRAM uses only a single detector to acquire data. Single detector systems use a minimum of facility space and are inherently

- More versatile
- More reliable
- Easier to use
- Less expensive

4.2 Planar or Coaxial Detector

PC/FRAM is the first isotopic analysis system that can obtain a complete isotopic analysis using either a single planar or a single coaxial detector. When using the traditional single planar detector, PC/FRAM has most often been used to collect and analyze data in the 120-420 keV range although it is not limited to this range. The most widely used mode of operation with a single coaxial detector is to acquire a single spectrum in the range from 0-1024 keV. Various analysis modes can then be used with this wide data range. If the widely used region between 120 and 200 keV is available, PC/FRAM will work best analyzing in an energy range from 120-460 keV. When analysis below 200 keV is

precluded (sample shielding or thick-walled sample container), PC/FRAM can still obtain a complete isotopic analysis using only gamma rays above 200 keV from a single coaxial detector spectrum.

The optimum choice of planar or coaxial detectors is made only after all applications are considered. The planar detector is usually the detector of choice if all measured items are unshielded or contained in "thin" containers. If shielded containers, thick-walled containers, or a mixture of thin and thick/shielded containers are encountered, then a single coaxial detector system is optimum.

4.3 *Shielded Sample*

Some isotopic analysis codes (including previous versions of FRAM) require the presence of spectral peaks in the region below 200 keV, regardless of whether they acquire data from one or two detectors. When this region is not available to the spectroscopist, perhaps because the sample is shielded to lower radiation exposure or because the sample is inside a very heavy-walled container, other isotopic analysis codes will not function. The PC/FRAM code will function in these cases.

Detector	Analysis Range (keV)	Gain (keV/ch)
Planar	120-420	0.100
Planar	120-307	0.075
Coaxial	120-460	0.125
Coaxial	120-460	0.250
Coaxial	200-800	0.125
Coaxial	300-800	0.125
Coaxial	120-1200	0.125
Coaxial	120-1200	0.156

4.4 *Flexible Data Acquisition and Analysis*

The structure of the PC/FRAM code allows data acquisition and analysis for nearly all measurement situations without costly, time-consuming reprogramming. The user is not limited by hard-wired, fixed data acquisition conditions. Some examples of how PC/FRAM has been used are given in Table I below.

4.5 *User-Editable Parameter Database*

The flexibility in acquisition and analysis is made possible by placing all the parameters that govern data acquisition and analysis in a user-editable database. The user has complete control over the setup of acquisition parameters, analysis parameters, diagnostic parameters, data storage formats, and default and global settings. All parameters may be changed from within a password-protected (three levels of protection) Change Parameter option that has the look and feel of a standard spreadsheet

4.5.1 Acquisition Parameters. The user may set parameters governing the choice of the multichannel analyzer (MCA) for data acquisition. Currently PC/FRAM can control

data acquisition from two commercially available MCAs. Compatibility with other MCAs is being added as needed. Defaults for other standard acquisition features such as counting time and live or true time may be preset.

4.5.2 Analysis Parameters. The user may control the setup and values of all the parameters that govern the data analysis. These include

- Analysis region boundaries
- Location and size of background regions (up to four for each analysis region)
- Background continuum shape (choice of seven functions)
- Peaks for analysis, peak energy, branching ratio
- Isotopes for analysis, half lives
- Peaks for internal self-calibration
 - Energy vs channel
 - FWHM vs energy
 - Tailing parameters

4.5.3 Diagnostic Parameters. Diagnostic tests can be tailored to match every measurement situation. These tests record

- Energy resolution
- Peak position (energy calibration)
- Peak tailing (detector deterioration)
- Interference peaks (unanticipated isotopes)
- Pu-241/U-237 decay equilibrium
- Am/Pu physical heterogeneity

4.5.4 Data Formats. Data formats for two popular commercial MCAs are supported in addition to a versatile ASCII format. Data may be converted among the available formats. New data formats are being added as required.

4.5.5 Default Parameters. Default settings may be used for often-entered values governing data acquisition, analysis, and storage. Default settings also govern starting values for the energy calibration and the detector peak shape parameters. These default settings make the code very easy to use in an in-plant environment because only minimal data entry is required for most measurements. A measurement (data collection followed by data analysis) may be performed with the entry of only two parameters—sample identification and operator identification.

4.5.6 Global Default Parameters. Several system-wide global defaults can also be adapted to the users' needs.

- Select date format for all input and output (accommodates US and international conventions)
- Select warning level for low disk space
- Select printer parameters for laser or dot matrix printers
- Select maximum number of channels to be analyzed
- Select MCA type
- Choose default path for saving spectral data and analysis results
- Identify name of data collection/data analysis system

4.5.7 Data and Results Archiving. Spectral data may be stored on disk in any supported format. Analysis results, including all intermediate calculations as well as complete channel-by-channel peak fitting results, may also be archived including the complete parameter set. This provides a complete record of the measurement for QA/QC purposes. A choice of three levels of analysis results may be printed or recalled to the screen for viewing.

4.6 Parameter Set

A complete group of parameters in the parameter database is called a Parameter Set. Parameter Sets may be exported to or imported from an ASCII text file, allowing transfer between systems.

Parameter sets exist for all common acquisition and analysis modes so the user is freed from the initial responsibility of setting up the system. The structure of the parameter database and its presentation to the user makes "on-the-fly" changes to the analysis a simple task. For example, a user may need only a minute or two to add a new peak to the list of peaks to be analyzed after finding an unexpected interference in the spectrum. All peak regions and analysis are set up on an energy basis. Therefore, spectra taken at different gains may be analyzed with the same parameter set by changing only two default energy calibration parameters. A user working with the parameters in the parameter database should be an experienced spectroscopist but is not required to be a programmer.

This flexible and versatile parameter database structure allows the user to analyze the widest possible variety of items without any reprogramming or software changes. Table II displays this flexibility.

Table II. Material Categories Analyzed with FRAM and PC/FRAM

• 2– 90% ^{240}Pu	• 80% ^{238}Pu
• 0.01–50% ^{241}Am	• Lead-shielded samples
• Interferences from	• Heterogeneous Am/Pu
^{243}Am - ^{239}Np	• ^{235}U : ^{241}Am : Pu = 24:1:1
^{237}Np	• 80– 95% ^{242}Pu
^{244}Cm	
• Nonequilibrium ^{241}Pu - ^{237}U	
• MOX: ^{235}U /Pu from	
0.005–35	
• ^{234}U , ^{235}U , ^{238}U in uranium (only), no Pu	

5. A PC/FRAM MEASUREMENT

A measurement consists of spectral data acquisition followed by spectral data analysis. The spectral data may come from a disk file or may be acquired on-line with PC/FRAM control of a supported MCA.

5.1 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, 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.

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

5.1.2 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 FWHMs 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.

5.1.3 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. 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.

5.2 Analysis

In the second stage of the analysis, each region is examined in the order listed in the parameter set. A continuum background is estimated for each region, then the necessary response functions are created, and finally these response functions are fitted to the net counts 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.

5.2.1 Background. For each region, one, two, three, or four 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.

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

6. APPLICATION OF PC/FRAM

Table III shows measurement results for the plutonium isotopes and the important parameter P_{eff} , the effective specific power used with calorimetry measurements. The bias of the effective ^{240}Pu fraction $^{240}\text{Pu}_{\text{eff}}$ used with neutron coincidence counting measurements is similar to that of ^{240}Pu . The isotopic contents of the samples in Table III ranged from 3.5% to 26% ^{240}Pu while the sample masses varied from 2 g to 2 kg. Over this wide range of samples, the average measurement bias is seen to be typically less than 1%.

The measurement biases for the individual samples in Table III are not always as good as the biases averaged over all samples. The distribution of the individual sample measurement biases about the mean can be thought of as an individual sample bias. This parameter is shown in Table IV where the values given are the percentage relative standard deviation (% RSD) of the distribution of the individual sample means (typically from 10–15 measurements) about the mean for all samples. These values are a more realistic estimate of the bias for a single sample.

Table II. PC/FRAM Measurement Bias Ratio: Measured/Accepted Values

Isotope	Planar Detector	Coaxial Detector	"Shielded" Coax
	120-420 keV	120-460 keV	Detector 200-800 keV
^{238}Pu	0.9929	1.0009	1.0095
^{239}Pu	0.9999	0.9997	0.9991
^{240}Pu	1.0005	1.0009	1.0009
^{241}Pu	0.9969	1.0014	0.9970
^{241}Am	1.0028	1.0026	1.0046
P_{eff}	1.0001	1.0007	1.0021
# of Samples	28	22	29
# of Measurements	420	220	301

Table III. Distribution of Individual Bias Values (% RSD)

Isotope	Planar Detector	Coaxial Detector	"Shielded" Coax
	120-420 keV	120-460 keV	Detector 200-800 keV
^{238}Pu	4.42	1.72	4.06
^{239}Pu	0.12	0.12	0.23
^{240}Pu	0.59	0.71	1.71
^{241}Pu	0.62	0.34	2.08
^{241}Am	1.94	1.88	1.14
P_{eff}	0.30	0.20	0.40
# of Samples	28	22	29

6.1 Uranium Measurements

The use of PC/FRAM to measure the isotopic composition of uranium items was first described in Ref. 8. We have enhanced this measurement capability by adding measurements for ^{234}U and by making other improvements for more accurate measurements at the extremes of the ^{235}U enrichment range. This current measurement capability as tested on a suite of Los Alamos standards is shown in Fig. 1

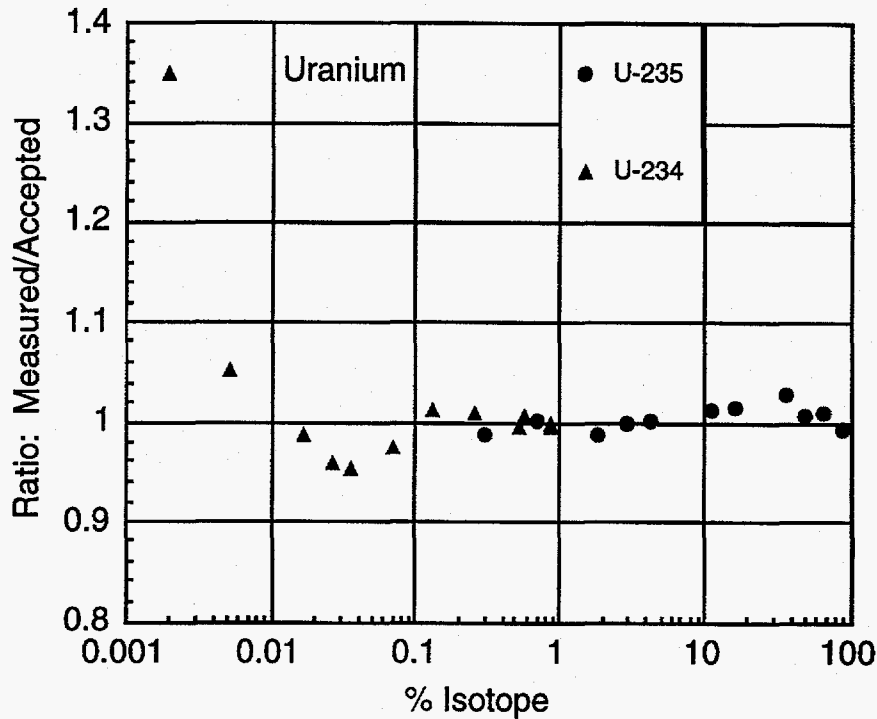


Fig 1. Measurement results for ^{235}U and ^{234}U with the PC/FRAM code.

7. CONCLUSIONS

We have described the features and noted the performance of the PC/FRAM code for the isotopic analysis of plutonium and other actinides from the measured gamma-ray spectrum from an arbitrary sample.

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 unpack 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.

This versatile, user friendly code is currently being used worldwide for safeguards and materials protection, control, and accountability applications.

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