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**LA-UR** -90-2204

CONF-9007106--37

AUG 0 6 1990

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LA-UR--90-2204

DE90 015106

TITLE FRAM: A NEW, VERSATILE GAMMA-RAY SPECTROMETRY CODE FOR MEASURING THE ISOTOPIC COMPOSITION OF PLUTONIUM

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SUBMITTED TO Institute of Nuclear Materials Management (INMM)
INMM Annual Meeting
Los Angeles, California
July 15-18, 1990
(FULL PAPER)

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# FRAM: A NEW, VERSATILE GAMMA-RAY SPECTROMETRY CODE FOR MEASURING THE ISOTOPIC COMPOSITION OF PLUTONIUM

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## ABSTRACT

We describe the characteristics and features and demonstrate the performance of a new code for determining the isotopic composition of plutonium using gamma-ray spectroscopy. This versatile code can measure an extremely wide range of isotopic compositions and is extremely easy to tailor to specialized measurement conditions. Measurement precision, accuracy, and throughput are significantly improved over previous Los Alamos National Laboratory (LANL) codes.

## I. INTRODUCTION

Software used to determine the isotopic composition of plutonium contained in arbitrary plutonium-bearing samples has been in use in the United States for over a decade. Descriptions of many of the methods in use, their principles, and their performance may be found in Ref. 1.

Several years ago the LANL Plutonium Facility recognized the need for improved software that would enable them to increase throughput and also analyze materials with heterogeneous Am/Pu distributions. This difficult sample category arises in the residues from pyrochemical purification processes. The existing MUDPI[2] software at LANL could not handle this case. At that time some improved analysis codes were available, namely the MGA code[3,4] and the GRPAUT[5,6,7] code. However, it is difficult to implement such complex codes in a production facility with the developer/experts not being readily available for consultation. Also, the MGA code was not in wide use and had not been widely tested on a variety of samples at that time. Therefore, it was decided to develop a code at LANL so that implementation in LANL facilities would be easier. We also used the best features of the codes available at that time and made improvements where warranted.

This report will discuss the approach taken in the code development, the features of the analysis and use of the code, and will also present and discuss measurement results. Reference 8 discusses the FRAM (Fixed Energy, Response Function Analysis with Multiple Efficiency) code in more detail.

## II. GENERAL APPROACH

### A. Energy Region Analyzed

For arbitrary samples, the 120- to 450-keV region is the most versatile region for use with a single detector. It has been shown to provide sufficient information to analyze heterogeneous (Am/Pu) samples.[7] Because we desired a versatile analysis system, we have chosen to use the 120- to 450-keV energy region in our analysis. Nothing in the software precludes analysis in other energy regions although the formalism to fit x-ray line shapes is not currently built into the code.

### B. Number of Detectors

Previous LANL systems for arbitrary samples have used only a single detector and analyzed data in the 120-450 keV region. A single detector system, being easier to develop, field, operate, and maintain, offers the best overall compromise for production facilities. The space savings of a single detector over a two-detector system is also important in production facilities where space is limited.

### C. Peak Area Extraction

The region-of-interest (ROI) summation technique used in previous LANL software cannot easily handle unexpected interferences. At least two other proven approaches are available to remedy this weakness, peak fitting by nonlinear least squares techniques as used in the GRPAUT isotopic code[6] and the response function method implemented in several Livermore codes.[9,10] The general approach taken in the response function method would seem to make it more robust with regard to unrecognized interferences and poor counting statistics. We chose the response function method for these reasons.

### D. Relative Efficiency Curve

The determination of the relative efficiency curve is a fundamental part of the analysis in nearly all methods of measuring plutonium isotopic composition. Three general methods have been used. In existing LANL codes, the efficiency is interpolated or extrapolated with simple linear or quadratic methods between a small number of relative efficiency points. Fleissner[6] uses the proven method of fitting a polynomial function in logE to the relative efficiency data. Gunnink[4] takes this one step further by using knowledge of the physical processes involved in the relative efficiency curve to specifically account for detector efficiency, cadmium absorbers, and plutonium self-absorption. We have chosen the method used by Fleissner because it is versatile and easy to implement.

### E. Isotopic Ratios

Previous LANL codes as well as GRPAUT use the proven method of peak pair ratios from neighboring peaks. Gunnink uses the general technique of finding a least squares solution to a set of linear equations involving peak areas, relative efficiency, and isotopic ratios as unknowns. We use this same general approach because it uses more of the available data, can provide results from more peaks from each isotope to check for consistency, and can provide ratios for peaks with no near neighbors.

### F. Summary of Chosen Approach

- Single detector
- 120-450 keV region
- Response function analysis for peak areas
- Least squares fitting of polynomial in logE for relative efficiency
- Least squares solution of simultaneous equations for mass ratios

## III. IMPLEMENTATION FEATURES

The implementation of the chosen approach can best be described by discussing the user selectable parameters that are accessible through an extremely versatile parameter file structure. Literally every constant that governs the analysis physics can be easily accessed, set, changed, or updated. Analysis parameter files are tailored to broad ranges of sample types as far as is feasible. The program contains a structure accommodating four categories of parameters. We call these

- Analysis parameters
- Default parameters
- Diagnostic parameters
- Program development/User authorization parameters

### A. Analysis *Parameters*

1. **Peak Information.** Each peak that is analyzed can be assigned an energy in keV, a branching ratio in gammas/disintegration (not required), and an isotope name (not required). We can designate if the area is to be fixed by branching and efficiency ratios to that of another peak or if the area is to be summed with another peak before finding a least squares solution for ratios (used for coenergetic $^{241}$Am, $^{237}$U peaks for example). We also can select whether the peak is to be used to define the relative efficiency curve and/or be used in the solution for activity ratios. Peaks can easily be added or deleted anywhere in the peak list.

2. **Fitting Region Information.** Information on fitting-region boundaries is given in units of energy to be independent of the energy calibration. We define the boundaries of each fitting region and the code searches the peak list and automatically includes all peaks defined in the region. The starting energy of up to four regions designated as background can be defined for each fitting region. These background regions can be inside or outside the boundaries of the fitting region. The number of data channels in each background region is defined. A very useful feature of this code is the versatility that arises from being able to choose the background function for each region from a selection of five functions. This allows the user to tailor the background to the exact nature of the spectrum in each region. The five possible background functions are:

- Zero slope straight line
- Sloping straight line
- Sloping straight line with smoothed step function
- Quadratic background with smoothed step function
- Smoothed step function on zero slope straight line

All the above parameters can be easily set or changed for each fitting region.

3. **Isotope List.** This section of the analysis parameters includes information on all isotopes to be analyzed. Any isotope in the list will be quantified as a ratio to total plutonium in the sample. Information included is the half-life, atomic mass, and the number of the relative efficiency function. For materials with heterogeneous components, for example, americium in a different matrix than plutonium, the americium component may have a different relative efficiency function than the plutonium isotopes. More than one heterogeneous component is allowed.

4. **Relative Efficiency Peaks.** This option presents the user with the list of peaks that are to be used to define the relative efficiency curve. Every analysis parameter file can have its own selection of relative efficiency peaks to fit the specific analysis conditions. The only requirement is that there be two or more peaks from each isotope included in the relative efficiency list.

5. **Energy Calibration Peaks.** The code accepts a list of peaks, by energy, to use in a piecewise linear energy calibration between each pair of peaks in the list. This calibration is typically done with strong single peaks from each spectrum.

6. **FWHM Calibration Peaks.** These peaks are used to parameterize the FWHM vs energy relationship for the Gaussian portion of each spectrum analyzed. The fitted function is then used to define the FWHM as a function of peak position for any peak in the spectrum. These peaks are often the same peaks that are used in the energy calibration and the shape calibration.

7. **Shape or Tailing Calibration Peaks.** The shape of each peak is defined for each measurement by the FWHM parameters and the tailing parameters. The peak shape is defined as a Gaussian with an exponential term on the low-energy side to describe the tailing. The peaks in the Shape Peaks list are used to define the tailing parameters in each spectrum. They can be easily changed for different materials.

8. **Initial Values for Energy, FWHM, and Shape Constants.** The algorithms that determine these parameters for each spectrum analyzed are iterative and require some starting values. While the particular values are not critical, it may be desirable to modify them for gross

changes in detector resolution. These initial values can also be fixed or free in the computation. As an example, it may be desirable to fix the parameters when attempting to analyze spectra with very poor counting statistics. The usual procedure is to keep the parameters free and thus determine FWHM, energy calibration, and shape parameters for each spectrum analyzed.

**9. $^{242}$Pu Correlation.** The analysis parameter file allows for two parameters to define the correlation governing $^{242}$Pu. Currently a single parameter is used to define the correlation

$$242 = K*240*241/[239^2]$$

with americium being added back to the $^{241}$Pu (addition not done for heterogeneous samples) before calculating the correlation. The user is given the option of using this correlation or entering his own value for $^{242}$Pu at every measurement. Empirically one typically finds a correlation constant K different from that suggested by studies[11] that examine the correlation at reactor discharge time.

### B. Default Parameters

These parameters are used to govern features of the user customizable dialog and printout to simplify operation. These parameters allow the user to select one of three lengths of output ranging from an isotopic results summary to detailed region-by-region-fitting residual information. These parameters also govern whether any of three different questions are presented to the operator. The first question that can be presented or suppressed governs the $^{242}$Pu correlation. If the question is suppressed, a correlation is used. Presenting the question to the operator allows the operator to either use the correlation or enter the $^{242}$Pu percentage. A second parameter allows the operator to respond to a request to enter the date and power from a calorimeter measurement. If this information is entered, the code will calculate the total plutonium mass in the sample on the calorimeter date. If the question is suppressed, no total plutonium information is available; only a result for the specific power, $P_{eff}$, is given. A third flag in this set of default parameters allows a question to be presented to the operator on spectral data storage. If this question is not presented, no data are stored. These parameters govern the operator dialog and program output so that only the minimum input/information needed for the measurement program at hand is required. All of the flags governing the default parameters can be changed at any time, independent of the analysis parameters in use.

### C. Diagnostic Parameters

These parameters control a series of diagnostic tests performed on every spectrum to check for proper spectrometer operation and to give the user some assurance that the data is of sufficient quality for correct analysis. These parameters go along with the analysis parameters so they can be specific to a sample category. The FWHM diagnostic tests the FWHM of a selectable number of peaks against a variable upper limit. Failure of this test may indicate a poor quality detector or too high count rate. The peak centroid diagnostic tests the centroid of a selectable number of peaks against a ± limit, checking correct stabilizer operation and overall system stability. The tail area check tests the fraction of the total peak area under the tail against a selectable upper limit. This checks for detector degradation from excessive neutron exposure. The interference peaks test is important; it is easily customized for each set of analysis parameters. It checks for the presence of possible interferences arising from peaks or isotopes not included in the analysis parameter file's peak list. Analysis of "normal" plutonium samples may typically require an interference check for peaks from $^{235}$U, $^{239}$Np, and $^{237}$Np. Such peaks would not be included in the peak list if it were unlikely for them to be present because the presence of unneeded peaks tends to reduce the robustness of the fitting process. The interference peaks test for a MOX sample might only request checks for $^{239}$Np and $^{237}$Np because $^{235}$U would already be included in the analysis file's peak list.

A second type of diagnostic parameter governs what we call a sample type test. The test determines the mass ratio from any two selectable peaks and tests the ratio of the mass ratios against an upper limit. The uses of this test are only constrained by the user's imagination. So far it has been applied in two ways. The mass ratio of the 148-keV and 164-keV $^{241}$Pu peaks should be unity for equilibrium samples. Deviation from unity may indicate a nonequilibrium sample. The americium mass ratio at 125-keV ratioed to that at a higher energy, say the 336- or 370-keV americium peak, will likely deviate from unity for a sample whose Am/Pu ratio is not homogeneous throughout.

### D. Program Development/User Authorization Parameters

These parameters govern many facets of the software development of the code. They also allow the system manager to set user authorizations and passwords. They are accessed by an off-line program.

## IV. HARDWARE/OPERATIONAL CHARACTERISTICS

The systems built to date have been constructed around a Canberra Series 90 multichannel analyzer (MCA) and a Digital Equipment Corporation MicroVax II computer operating under the VMS operating system. Such systems can operate up to four detectors and count four samples simultaneously. Filtering is typically accomplished with 0.080 in. of cadmium and ~0.015 in. of copper.

We use a side-looking detector to be compatible with a manually controlled scan table that can rotate and vertically translate the sample, a requirement for heterogeneous Am/Pu materials. Scan height is easily set at the scan table so that only the height of the can is scanned. The scan table is shielded with 0.25 in. of lead to reduce the photon radiation dose to the operator. The operator varies the detector count rate by moving the rail-mounted detector to vary the sample-detector distance. The operator can monitor the detector count rate from a counter timer in a NIM bin on top of the scan-table housing. This arrangement allows the scan table to be remote from the data acquisition electronics/MCA. Data are acquired at input rates up to 50 kHz at a shaping time of 1 µs. Analysis time on the MicroVax II is typically less than 30 s. Analysis is typically done on-line with off-line analysis capability also available.

## V. CALIBRATION

Isotopic methods, such as this one, that use fundamental constants and intrinsic relative efficiency curves do not require calibration in the usual sense. However, one must verify each analysis parameter file on appropriate samples. Often adjustments are necessary. With so many parameters available, user experience becomes valuable. Good documentation and knowledgeable users are needed to fine tune the analysis. Common user adjustments are the following: (1) branching ratio adjustment; (2) peak energy adjustment for interferences or new peaks; (3) fixing of peaks to other peaks; and (4) background ROI positions, number of channels, and selection of the background function. Most of these have been studied for commonly used analysis parameter files and additional changes are not needed.

Branching ratios may not be the same from one analysis parameter file to another. While they do not physically change, adjusting them may be the only way to make small adjustments for relative efficiency curves defined by different sets of peaks or difficult background fitting over large fitting regions. Typically such adjustments correct biases of a few percent or less.

## VI. PERFORMANCE

The initial testing of the FRAM system has been accomplished with perhaps the widest range of material types ever presented to a new isotopic code. We will present results for the major categories tested.

### A. Equilibrium, Homogeneous Am/Pu Materials

This class of materials covers most of the "usual" samples presented to isotopic systems. Here $^{237}$U is in secular equilibrium with its $^{241}$Pu parent and americium is distributed uniformly throughout all the plutonium in the sample. We have measured well-documented samples with $^{240}$Pu fractions ranging from 5% to 18% and $^{241}$Am concentrations of 200 µg/g Pu to 30 000 µg/g Pu. Table I presents the accepted isotopic values for the samples in this

**TABLE I. HOMOGENEOUS (Am/Pu) SAMPLES**

| | Accepted Values (wt%) | | | | | | |
|---|---|---|---|---|---|---|---|
| Sample ID | 238 | 239 | 240 | 241 | 242 | $^{241}$Am (µg/g Pu) | $P_{eff}$ (mW/g Pu) |
| STD-151 | 0.0022 | 97.971 | 2.009 | 0.0148 | 0.0030 | 80 | 2.0545 |
| A1-92 | 0.0087* | 94.606 | 5.262 | 0.1099 | 0.0142 | 1 760 | 2.4513* |
| A1-86 | 0.0104* | 94.228 | 5.605 | 0.1385 | 0.0183 | 1 869* | 2.4916* |
| STDR3 | 0.0103 | 94.041 | 5.766 | 0.1623 | 0.0208 | 1 885 | 2.5017 |
| CALEX | 0.0095* | 93.860 | 5.860 | 0.2412 | 0.0209 | 1 354 | 2.4424* |
| SRPSTDPUEU7 | 0.0144 | 93.782 | 5.862 | 0.2762 | 0.0659 | 234* | 2.3419* |
| HSUESOL | 0.0153* | 93.792 | 5.865 | 0.2825 | 0.0452 | 110* | 2.3334* |
| PUTIBSPC1 | 0.0108 | 93.806 | 5.881 | 0.2604 | 0.0420 | 745* | 2.3813* |
| J00132501 | 0.0112 | 93.876 | 5.903 | 0.1816 | 0.0280 | 1 214 | 2.4374 |
| STD117 | 0.0149* | 93.579 | 6.154 | 0.2134 | 0.0391 | 1 209 | 2.4707* |
| STD8 | 0.0099 | 93.476 | 6.328 | 0.1615 | 0.0254 | 1 344 | 2.4663 |
| STD6 | 0.0099 | 93.476 | 6.328 | 0.1614 | 0.0254 | 1 344 | 2.4663 |
| STD3 | 0.0227 | 91.930 | 7.615 | 0.3545 | 0.0772 | 3 113 | 2.8090 |
| STD118 | 0.0261 | 90.385 | 9.000 | 0.4848 | 0.1043 | 2 731 | 2.8574 |
| PEO382C | 0.0264* | 89.597 | 9.693 | 0.4785 | 0.1123 | 4 225 | 3.0649* |
| STD40 | 0.0651 | 87.139 | 11.768 | 0.8282 | 0.2000 | 4 334 | 3.4068 |
| STD119 | 0.0373 | 87.262 | 11.784 | 0.7486 | 0.1678 | 4 231 | 3.2385 |
| NBS946 | 0.2229 | 84.974 | 12.374 | 1.8431 | 0.5857 | 25 511 | 6.7574 |
| STD116 | 0.3640 | 79.804 | 15.455 | 3.3274 | 1.0496 | 18 459 | 6.9228 |
| STD120 | 0.3640 | 79.806 | 15.455 | 3.3257 | 1.0497 | 25 743 | 7.7545 |
| LAO256C10 | 0.0580 | 82.289 | 16.268 | 1.0258 | 0.3400 | 3 528 | 3.5083 |
| LAO225BS | 0.0604 | 81.986 | 16.490 | 1.1102 | 0.3530 | 4 844 | 3.6834 |
| STD121 | 0.0603 | 81.990 | 16.491 | 1.1057 | 0.3531 | 4 889 | 3.6883 |
| NBS947 | 0.2674 | 77.608 | 18.802 | 2.0990 | 1.2240 | 27 961 | 7.6121 |

* Denotes accepted value thought to be incorrect and not used in averages.

category of materials. A few of the individual isotopic values are thought to be in error and are denoted with an asterisk. The isotopic errors automatically translate into errors in the specific power, $P_{eff}$, which are also noted. Values thought to be in error are usually flagged when there is a lack of agreement between the measurements and the accepted values, but good agreement on the same isotope for several similar samples.

In Table II we show the sample mass, measurement conditions, and measurement precision for these samples. The measurement conditions consist of a count rate in kHz and a count time in hours. Both represent practical conditions for these samples. We conclude that count times of 30 minutes to 1 hour are realistic and acceptable for most samples containing enough plutonium for calorimetry. The measurement precision for $^{240}Pu$ and $P_{eff}$ presented in the last two columns is calculated from the spread of the data from the number of repeated runs in the 5th column. These realistic measurement conditions yield measurement precisions for $P_{eff}$ that are typically under 0.3% [1 relative standard deviation (RSD)]. This precision is very well matched to that observed from calorimeter measurements of total wattage.

The measurement precision for $P_{eff}$ and $^{240}Pu$ in Table II is typically a factor of 1.5-2.0 better than that of the previous LANL systems[2] while simultaneously improving the throughput by a factor of 2-3. This improvement arises from using more data in the analysis and acquiring data at a higher rate with fewer counting losses.

In Table III we list the ratio of the average measured value to the accepted value for all the samples and data acquisition conditions in Tables I and II. At the bottom of Table III we show the average of the ratios for each isotope and sample. The row denoted "Average" represents the average bias for the particular parameter over the wide range of sample types and compositions represented by the 23 listed samples. Note that it is well under 1% (under 0.1% for most) for all isotopes. The standard deviation or %RSD of this average can be interpreted as the spread in the measurements likely to be encountered in the measurement of any single sample and can be viewed as a typical, expected measurement accuracy.

The data in Table III would appear to indicate a possible bias for $^{241}Am$ at concentrations below 1000 ppm. We have extensive data on additional samples

TABLE II. SAMPLE MASS, MEASUREMENT CONDITIONS, AND MEASUREMENT PRECISION

| Sample ID | Pu Mass (g) | Ct Rate (kHz) | Single Meas. ct.time (hr) | No. of Repeated Runs | Single Meas. Precision from Repeated Runs $^{240}Pu$ (%RSD) | Single Meas. Precision from Repeated Runs $P_{eff}$ (%RSD) |
|---|---|---|---|---|---|---|
| STD-151 | 2 | 3.3 | 2 | 9 | 3.68 | 0.24 |
| A1-92 | 10 | 18.5 | 1 | 20 | 0.68 | 0.08 |
| A1-86 | 10 | 22 | 1 | 20 | 0.99 | 0.14 |
| STDR3 | 21 | 7.2 | 1 | 6 | 1.18 | 0.19 |
| CALEX | 400 | 23 | 0.5 | 15 | 1.64 | 0.33 |
| SRPSTDPUEU7 | 1747 | 41 | 1 | 20 | 1.15 | 0.13 |
| HSUESOL | 5 | 7.1 | 1 | 20 | 1.36 | 0.19 |
| PUTIBSPC1 | 1736 | 40 | 1 | 6 | 1.65 | 0.29 |
| J00132501 | 500 | 22 | 1 | 15 | 1.43 | 0.27 |
| STD117 | 1.7 | 4 | 1 | 15 | 2.2 | 0.33 |
| STD8 | 240 | 32 | 1 | 5 | 1.56 | 0.21 |
| STD6 | 120 | 27 | 1 | 15 | 0.81 | 0.12 |
| STD3 | 60 | 21 | 1 | 5 | 0.92 | 0.16 |
| STD118 | 1.6 | 3.4 | 1 | 6 | 0.81 | 0.31 |
| PEO382C | 150 | 31 | 0.5 | 15 | 1.24 | 0.2 |
| STD40 | 875 | 40 | 0.5 | 15 | 1.13 | 0.26 |
| STD119 | 1.7 | 5.2 | 1 | 8 | 1.38 | 0.29 |
| NBS946 | 0.8 | 4 | 2 | 10 | 1.5 | 0.13 |
| STD116 | 1.7 | 12 | 1 | 15 | 2.13 | 0.21 |
| STD120 | 1.8 | 18.7 | 1 | 15 | 1.27 | 0.18 |
| LAO256C10 | 876 | 42 | 0.5 | 15 | 0.96 | 0.22 |
| LAO225BS | 875 | 40 | 0.5 | 15 | 0.82 | 0.23 |
| STD121 | 3 | 7.4 | 1 | 15 | 1.13 | 0.28 |
| NBS947 | 0.7 |  | 2 | 10 | 1.01 | 0.12 |

TABLE III. RATIO OF MEASURED/ACCEPTED VALUES

| | Measured/Accepted | | | | | |
|---|---|---|---|---|---|---|
| Sample ID | 238 | 239 | 240 | 241 | $^{241}$Am | Peff |
| STD-151 | 1.05250* | 1.00083* | 0.95962* | 0.95412* | 1.21006* | 0.99924* |
| A1-92 | 0.87419* | 1.00017 | 0.99727 | 0.99714 | 0.99592 | 0.99685* |
| A1-86 | 0.85894* | 1.00047 | 0.99233 | 0.99922 | 1.03610* | 0.99888* |
| STDR3 | 0.97921 | 1.00049 | 0.99206 | 0.99996 | 1.00525 | 0.99900 |
| CALEX | 1.06842* | 1.00007 | 1.00007 | 1.00229 | 0.99821 | 1.00147* |
| SRPSTDPUEU7 | 1.00177 | 1.00007 | 0.99894 | 0.99940 | 1.11558* | 1.00124* |
| HSUESOL | 0.88949* | 1.00007 | 0.99914 | 1.00210 | 1.13652* | 0.99654* |
| PUTIBSPC1 | 1.03558 | 0.99964 | 1.00588 | 0.99448 | 1.10067* | 1.00524* |
| J00132501 | 1.01354 | 0.99972 | 1.00455 | 0.99855 | 0.99376 | 1.00056 |
| STD117 | 0.91994* | 0.99985 | 1.00255 | 0.99586 | 1.00415 | 0.99782* |
| STD8 | 1.00869 | 1.00046 | 0.99328 | 0.99415 | 0.99631 | 0.99908 |
| STD6 | 0.99461 | 1.00006 | 0.99930 | 0.99298 | 0.99870 | 0.99971 |
| STD3 | 1.00565 | 1.00013 | 0.99827 | 1.00528 | 1.00805 | 1.00105 |
| STD118 | 0.98097 | 0.99999 | 1.00057 | 0.99166 | 1.00922 | 1.00010 |
| PEO38C2 | 0.94287* | 0.99917 | 1.00687 | 1.01979 | 0.99553 | 0.99768* |
| STD40 | 0.99931 | 0.99953 | 1.00334 | 1.00230 | 1.00454 | 1.00119 |
| STD119 | 1.01534 | 0.99934 | 1.00517 | 0.99438 | 0.99966 | 1.00190 |
| NBS946 | 0.99791 | 1.00135 | 0.99122 | 0.99708 | 1.00591 | 1.00132 |
| STD116 | 0.99702 | 1.00121 | 0.99388 | 0.99960 | 0.99522 | 0.99703 |
| STD120 | 1.0004[illegible] | 1.00137 | 0.99263 | 0.99964 | 0.98929 | 0.99531 |
| LA0256C10 | 0.97853 | 0.99880 | 1.00583 | 1.00490 | 1.01819 | 1.00150 |
| LA0225BS | 1.00253 | 0.99819 | 1.00880 | 1.00281 | 1.00148 | 1.00250 |
| STD121 | 1.00900 | 1.00131 | 0.99894 | 1.00035 | 1.00502 | 1.00134 |
| NBS947 | 1.00596 | 1.00044 | 0.99816 | 0.99957 | 0.99396 | 0.99842 |
| Average | 1.00153 | 1.00008 | 0.99952 | 0.99972 | 1.00097 | 1.00000 |
| Std Dev. | 0.01409 | 0.00080 | 0.00525 | 0.00571 | 0.00691 | 0.00196 |
| %RSD | 1.40686 | 0.07959 | 0.52541 | 0.57108 | 0.69005 | 0.19586 |

*Values not used in average.
Note: STD151 not used in average.

characterized by three laboratories that indicate that FRAM is accurate to about 1% for $^{241}$Am concentrations as low as 300 μg/g Pu. We believe that the bias for low americium concentrations in Table III arises from the sample characterization and not the FRAM measurement.

### B. Nonequilibrium, Homogeneous Am/Pu Materials

The same data discussed above was also analyzed assuming no $^{241}$Pu-$^{237}$U equilibrium. This may not prove that the analysis is correct for actual nonequilibrium samples but it is a necessary condition for that to be true. These results (not shown) generally show the same results as the equilibrium analyses in Table III.

### C. Heterogeneous Am/Pu Materials

One of the main purposes of this code was to be able to analyze process residues at the LANL Plutonium Facility that had heterogeneous Am/Pu distributions. This situation arises in pyrochemical processing residues where the americium is present in a low-Z salt matrix in which plutonium metal residues are imbedded. In this situation plutonium gamma rays suffer attenuation that may be dominated by self-absorption in plutonium whereas americium gamma rays are primarily absorbed by the salt matrix. This gives rise to different absorption characteristics depending on the element emitting the photon. Thus relative efficiency curves are different for plutonium gamma rays and americium gamma rays.

Fleissner[7] has developed a method for analyzing these material types and it has been compared with analytical chemistry in a single comparison on a small number of samples.[11] These comparisons are extremely difficult, expensive, and time consuming because of the necessity for total chemical dissolution of large quantities of highly radioactive residues. Nevertheless, the limited comparison with chemistry showed an average bias of only 1.5%. This bias may seem large compared to results on other material types shown above in this report, but it must be put in proper context. If conventional analysis methods had been used, biases could easily have been 50%-100%.

Fleissner's method essentially assumes that the samples are two-component mixtures that can be represented by two relative efficiency curves, one for plutonium and one for americium. This same method was adopted for use in FRAM. Because the Fleissner approach in GRPAUT and FRAM are similar, we should expect similar, but not necessarily the same, results when analyzing identical samples. We would not necessarily expect identical results because the two codes use different peak fitting methods, different analysis regions, and different branching ratios.

Because only a single comparison with chemistry was available, an additional comparison program was established at LANL, not only to test FRAM, but also to test other NDA techniques for these difficult materials. FRAM measured five samples that were subsequently crushed, blended, sampled, and analyzed by the LANL analytical chemistry group, CLS-1. Some of these same samples were also measured by Fleissner at Rocky Flats prior to the chemical analysis. These samples were all nominal 6% $^{240}$Pu and contained from 3.5%-5.3% $^{241}$Am (two samples were lower). In Table IV the measurement results for the specific power, $P_{eff}$, are compared to the accepted values from chemical assay. For these samples with high americium, the comparison is influenced mainly by the ability to measure the correct Am/Pu ratio because the majority of the sample power comes from americium. The XBLP samples arise at LANL whereas the MSE residues were produced at Rocky Flats from a different process and have different packaging. It is not surprising that one type of sample shows a bias while another does not. The average bias of 2.4% from this very limited sample set is comparable to the bias found in Fleissner's original study.[11]

## D. Mixed Uranium-Plutonium Materials

FRAM was tested on a wide range of mixed uranium-plutonium oxide samples. We made measurements on two different sample sets, one with 6% $^{240}$Pu and <1% $^{235}$U/U, and the other with 12% $^{240}$Pu and high concentrations of 93% enriched uranium. The $^{235}$U/Pu ratio varied over a range from 0.005 to 35, a factor of 7500.

TABLE IV. PLUTONIUM ISOTOPICS MEASUREMENTS COMPARED TO CHEMISTRY

| Sample ID | $P_{eff}$: ratio of FRAM/CLS-1 | $P_{eff}$: ratio of FLEISSNER/CLS-1 |
|---|---|---|
| MSE-1 | 0.9806 | 0.9612 |
| MSE-2 | 0.9451 | 0.9338 |
| MSE-3 | no FRAM data | 0.9929 |
| MSE-4 | 0.9523 | 0.9472 |
| MSE-5 | no FRAM data | 0.9934 |
| ARF876642 | no FRAM data | 0.9866 |
| XBLP121 | 1.0049 | |
| XBLP278 | 0.9905 | |

The samples with the largest $^{235}$U/Pu ratios (>15) were extremely difficult to measure. Even with long count times, measurement precision was still poor. These probably represent a practical limit to the method. $P_{eff}$ was accurate, on the average, to 0.1% over this enormous range of uranium concentrations as shown in Fig 1.

## E. Special Materials

The versatility of the FRAM system has been demonstrated by its ability to analyze materials with extreme isotopic distributions and/or interferences. All that is necessary is to modify a parameter file to account for the specific material characteristics. Typically this will require about a half day.

1. **$^{238}$Pu.** We have demonstrated that FRAM can measure the isotopic composition of $^{238}$Pu heat-source-grade material containing ~80% $^{238}$Pu. The measurement is difficult because of the weak gammas from the low concentrations of $^{239,240}$Pu. Limited measurements have been done but the analysis proceeded routinely.

2. **$^{242}$Pu.** We have also measured samples with high enrichments (80%-95%) of $^{242}$Pu. While we still cannot directly measure $^{242}$Pu, the collected spectra can be analyzed with no code modifications. The complete isotopic distribution can be determined if the correct $^{242}$Pu percentage is entered. If this is not known, the user still has access to ratios of the other isotopes.

3. **$^{239}$Np.** This isotope arises in samples as the decay product of $^{243}$Am and is sometimes seen in high-burnup materials. While its strongest gamma rays are at 228 keV and 277 keV, it is the gamma rays at 209 keV and 334 keV that interfere with normal analyses. It is straightforward to include these gamma rays in the analysis parameters and we have successfully analyzed samples with

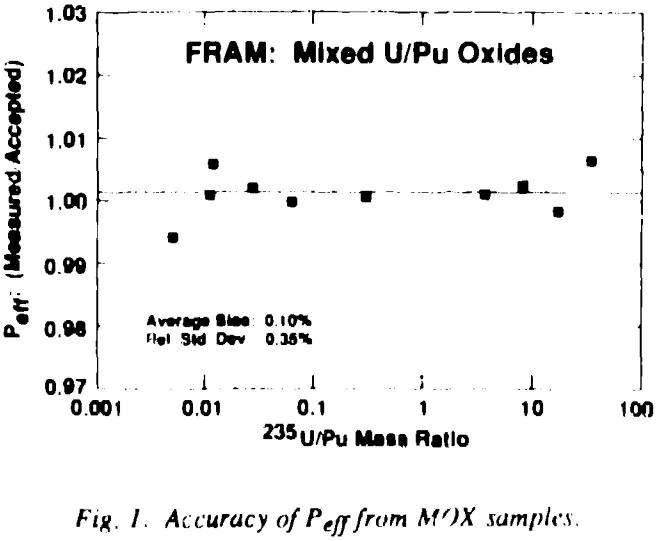

*Fig. 1. Accuracy of $P_{eff}$ from MOX samples.*

>500 ppm $^{243}$Am. At this level the 209-keV $^{239}$Np peak is over 5 times as intense as the 208-keV peak and the 334-keV $^{239}$Np peak is about 10 times as large as its 332-keV neighbor. Not only has this analysis been done but we have also analyzed materials in which the $^{243}$Am-$^{239}$Np is heterogeneous with respect to the plutonium in the sample.

4. **Uranium (only).** One of the most interesting characteristics of the FRAM code is its ability to measure, *without any modifications to the code*, the $^{238}$U/$^{235}$U ratio in materials that contain only uranium. This measurement, using intrinsic relative efficiency curves, has been discussed for nearly 15 years. (See Ref. 13 for a recent discussion and references to earlier work in this area.) However, until the development of the FRAM code, no practical method has been available for in-plant implementation, let alone using the same unmodified code that can also perform a wide range of plutonium isotopic composition measurements. The ability of FRAM to measure the $^{238}$U/$^{235}$U ratio in uranium was demonstrated with measurements taken with a coaxial detector spanning the energy range up to 1200 keV in 8192 channels. Figure 2 displays the result of measurements of samples with $^{235}$U enrichments ranging from 0.7% to 66%. While a small enrichment dependent bias is present, the measurements are accurate to 1% to 2% over this entire range. For the first time there is now a tool that can be used within the uranium enrichment community to perform measurements of $^{238}$U/$^{235}$U in samples of arbitrary physical and chemical composition, geometry, and mass.

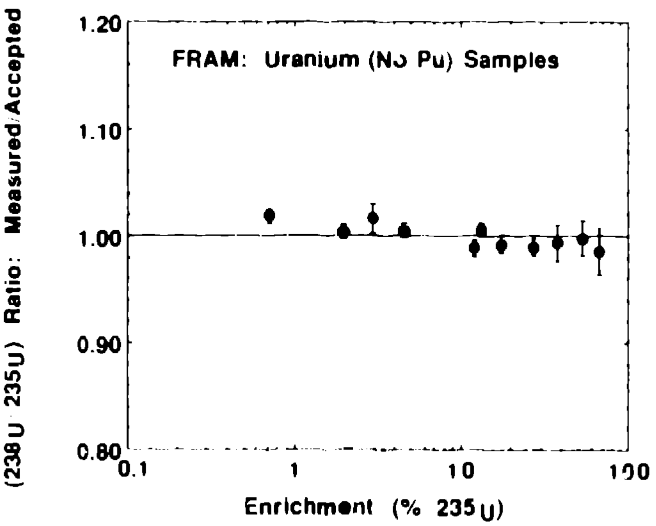

*Fig. 2. Accuracy of $^{238}$U/$^{235}$U ratio measurements.*

## VII. CONCLUSION

The FRAM code has demonstrated its ability to measure the widest possible range of material types and compositions. The key to this versatility is an analysis parameter file that can be easily modified for the specific measurement conditions without any time consuming, labor intensive, main code changes. The performance of this new data acquisition and analysis system has been improved significantly in accuracy, precision, and throughput over previous LANL codes.

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