

*FRAM: A Versatile Code for Analyzing
the Isotopic Composition of Plutonium
from Gamma-Ray Pulse Height Spectra*

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FRAM: A VERSATILE CODE FOR ANALYZING THE ISOTOPIC COMPOSITION OF PLUTONIUM FROM GAMMA-RAY PULSE HEIGHT SPECTRA

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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 a 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 codes.

I. INTRODUCTION

Software for determining 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. Los Alamos-developed codes, LAPIS and MUDPI² have been in routine production use since 1981 at the Los Alamos Plutonium Facility and at the Savannah River Plant. Adaptations of these Los Alamos codes are also in use at the Atomic Weapons Establishment, Aldermaston, and British Nuclear Fuels plc, Sellafield in the United Kingdom.

Several years ago the Los Alamos Plutonium Facility recognized the need for improved software that would enable them to increase throughput and analyze materials with heterogeneous Am/Pu distributions. This difficult sample category arises in the residues from pyrochemical purification processes. The existing MUDPI software at Los Alamos 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 well known that implementing such complex codes in a production facility with the developer/experts not being readily available for consultation presents difficult problems. 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 Los Alamos that would be easier for the user to implement in Los Alamos facilities. 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 on a wide variety of materials. The origin of the name FRAM for the code is discussed in Appendix C of this report.

II. GENERAL APPROACH

Designing a new code of this type requires that decisions be made in many areas that govern the ultimate nature of the isotopic system. Some of these decisions will now be discussed.

A. Energy Region Analyzed

Reference 1 describes the available energy regions in detail. For arbitrary samples, only three regions have been used. The 100-keV region is best for rapid analysis, but has not been used for analysis of heterogeneous (Am/Pu) samples. It is also the most complex region to analyze. If one desires analysis in this region, it would be best to make use of the available MGA software and avoid a new development effort. The 600-keV region is used in systems that use a second detector. The 120- to 450-keV region is the most versatile region for use with a single detector. It provides sufficient information to analyze heterogeneous (Am/Pu) samples.⁷ Because we desired a versatile analysis system, we chose to use the 120- to 450-keV energy region in our analysis. Nothing in the software precludes analysis at higher energies, including 600 keV, if the user desires.

B. Number of Detectors

Previous Los Alamos systems have used a single detector. Other systems⁵ use two detectors. Single detector systems usually use a planar germanium detector up to energies of 400 keV while a two detector system usually adds a more efficient coaxial detector to acquire data in the 300-700-keV range. This provides additional information for ²⁴⁰Pu and ²⁴¹Am. The ²⁴¹Am data at 662 keV yield improved precision over that from single detectors that do not use the 100-keV region. The ²⁴⁰Pu data do not add significantly to the precision already available at 160 keV for typical single detectors. We have not changed our philosophy that a single detector system, being easier to develop, field, operate, and maintain offers the best overall compromise for production facilities.

C. Peak Area Extraction

The Region of Interest (ROI) summation technique used in previous Los Alamos 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 is used in the GRPAUT isotopic code.⁶ With modern computers, analysis time is not the problem it once was with this method. This method is potentially susceptible to unrecognized interferences if the full width at half

maximum (FWHM) parameter is left free in the fitting. The response function method is a simpler variant of peak fitting that requires only linear least squares fitting. It is well understood and has been implemented in several Livermore codes.^{8,9} The general approach taken would seem to make it more robust with regard to unrecognized interferences and poor counting statistics. Reference 1 describes the three methods, as applied to plutonium isotopic data, in more detail.

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 widely 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. The peaks used are those from ²³⁹Pu and ²⁴¹Pu and are fixed in the code. Fleissner⁶ uses the well-proven method of fitting a polynomial function in logE to the relative efficiency data. Gunnink⁴ 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 and can provide results from more peaks from each isotope to check for consistency.

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

The analysis parameters govern the physics of the spectral data analysis, that is, peak areas, relative efficiency, isotopic ratios, etc.

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, used in the solution for activity ratios, or both. 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 and therefore is 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 fitting region information also includes the definition of the background function to be used in each 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. The number of data channels must be the same for each of the four designated background regions for any fitting region but may be different for each fitting region. 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 gives unprecedented versatility to allow 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. Isotopes can be added or deleted from the isotope list.

4. Relative Efficiency Peaks. This list 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 real requirement is that there be two or more peaks from each isotope included in the relative efficiency list. The relative efficiency function has the form:

$$\text{Log}(\epsilon) = C(1) + C(2)/E^2 + C(3)\text{Log}(E) + C(4)[\text{Log}(E)]^2 + C(5)[\text{Log}(E)]^3 \\ + C(5+N) + C(5+N+M)$$

where

ϵ = relative efficiency

E = gamma-ray energy

N = number of isotopes (in excess of 1) defining the relative efficiency curve

M = number of different relative efficiency functions for heterogeneous isotopes in addition to the principal relative efficiency curve

The relative efficiency curve is defined first by designated peaks from the first isotope in the isotope list. The coefficient $C(5+N)$ normalizes peaks from the N th additional isotope to the curve. The coefficient $C(5+N+M)$ is the "beta factor," which governs the shape and magnitude of additional relative efficiency curves for "heterogeneous" isotopes, isotopes that have a different spatial distribution in the sample compared to isotope #1. These additional relative efficiency curves have a shape that differs from the primary curve by a factor $\exp(\beta/E)$.

An example of a need for a second relative efficiency curve is in pyrochemical residues in which the americium is a salt, typically a chloride, and the plutonium is a metal dispersed in the salt. Thus

gamma rays from americium and plutonium at the same energy will be attenuated differently as they escape the sample.

5. Energy Calibration Peaks. The code accepts a list of peaks, and their energies, 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. The list can be tailored to various sample types by adding or deleting peaks from the list.

6. FWHM Calibration Peaks. These peaks are used to parameterize the FWHM vs energy relationship for the Gaussian portion of each photopeak analyzed. The FWHM for each peak in the list is found and fitted to a function of the channel number of the form:

$$\text{FWHM}(\text{Ch}) = \sqrt{F1 + F2*\text{Ch} + F3/\text{Ch}}$$

The F1 and F2 terms govern the expected relationship from physics principles. The F3 term attempts to account for the observed flattening of typical curves at lower energies. The fitted function is then used to define the FWHM as a function of peak position for any peak in the spectrum. Strong single peaks are used to define this relationship. Often they are 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 basic peak shape is defined as a Gaussian with added terms describing the tailing. The tailing terms have the form given below in which the empirically known energy dependence of some of the shape parameters is explicitly included.

$$\text{Tail}(\text{Ch}) = \text{HT}*\exp\{\text{T1} + \text{T2}*E + \text{T3}*(\text{Ch} - \text{X0})\} * [1 - \exp(\text{T4}*(\text{Ch} - \text{X0})^2)]$$

- Ch = channel at which tail contribution is calculated
- HT = height of Gaussian portion of peak
- X0 = centroid of Gaussian portion of peak

The three parameters T1, T2, and T3 are determined from least-squares fits to tail data after stripping out the background and the Gaussian portion of the designated shape peaks. T4 is a fixed function of the FWHM. References 1, 6, and 9 give more information on this tailing shape

function. The peaks in the Shape Peaks list are used to define the tailing parameters in each spectrum. They can be easily changed for different material categories.

8. Initial Values for Energy, FWHM, and Shape Constants. The algorithms that determine these parameters for each spectrum analyzed are iterative in nature and require some starting values. The particular starting values are not critical when the parameters are free to be determined from the spectrum. These parameters can also be fixed with the starting values. As an example, it may be desirable to fix the parameters when attempting to analyze spectra with very poor counting statistics. For spectra with good statistics and some clean, strong peaks the usual procedure is to keep the parameters free and thus determine FWHM, energy calibration, and shape parameters for each spectrum.

9. Plutonium-242 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 among the masses or isotopic fractions of ^{242}Pu , ^{240}Pu , ^{241}Pu , and ^{239}Pu .

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

with americium being added back to the ^{241}Pu before calculating the correlation. The user is given the option of using this correlation or entering his own value for ^{242}Pu for every measurement. Empirically one typically finds a correlation constant K different from that suggested by studies¹⁰ that examine the correlation at reactor discharge time.

B. Default Parameters

These parameters control features of the user customizable dialog and printout. They allow the user to select one of three lengths of output ranging from an isotopic results summary to a printout of 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 choose the correlation or enter the ^{242}Pu percentage. A second parameter presents the operator with a question requesting the entry of 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 will be given, only a result for the specific power, P_{eff} . A third question concerns spectral data storage. If this question is not presented, no data storage takes place. These parameters govern the operator dialog and program output so that the amount of

input/information needed for the specific measurement program at hand is a minimum. All of the flags governing the default parameters can be changed at any time, independent of the analysis parameters in use.

C. Diagnostic Parameters

A series of diagnostic tests is performed on every spectrum to check for proper spectrometer operation and to give the user some assurance that the data are of sufficient quality for correct analysis. A second series of tests is performed to verify that the sample type is appropriate for the selected analysis parameter file. The parameters governing these diagnostic tests are part of 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 a count rate that is too high. The peak centroid diagnostic tests the centroid of a selectable number of peaks against a \pm limit, checking for 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 important interference peaks test 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 peak list of the analysis parameter file. Analysis of "normal" plutonium samples may typically request 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 in the fitting process tends to reduce its robustness. However, the interference peaks test for a MOX sample might request only checks for ^{239}Np and ^{237}Np , because ^{235}U peaks would already be included in the peak list of the analysis file.

A second type of diagnostic parameter is used to check the sample type. The test determines the mass ratio from any two peaks and tests the ratio of the mass ratios against an upper limit. The uses of this test are only constrained by the operator'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 for a higher energy, say 336 keV or 370 keV, americium peak will likely deviate from unity for a sample whose Am/Pu ratio is not homogeneous throughout the sample.

D. Program Development/User Authorization Parameters

These parameters govern many facets of the software development for 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. Table I shows the major components in a typical system. While the analog to digital converter (ADC) and stabilizer must be Canberra units for compatibility, the linear electronics and other items such as the amplifier, high voltage bias supply, count rate monitors, detector, and oscilloscope can be makes/models other than listed here.

The above system can operate up to four detectors and count four samples simultaneously. Filtering is typically accomplished with 0.080 in. cadmium and ~ 0.015 in. copper.

We use a side-looking detector to be compatible with the scan table that can rotate and vertically translate the sample, a requirement for heterogeneous Am/Pu materials. The scan table is controlled manually. 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 detector with a 2-in.-thick lead shield is mounted on rails; the operator varies the detector count rate by moving the detector to change the sample-detector distance. The operator can monitor the detector count rate from a counter/timer in a nuclear instrumentation module (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 μ s.

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 according to appropriate samples and standards. 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: (1) branching ratio adjustment, (2) peak energy adjustment for interferences or new peaks, (3) fixing of peaks to other peaks, and (4) background ROI position, number of channels, and selection of the background function. Most of these have been studied for the range of plutonium and Am/Pu mixtures that may be encountered and changes are not needed.

TABLE I
MAJOR COMPONENTS IN A TYPICAL SYSTEM

Detector:	Canberra Planar, 16 mm diam by 13 mm deep. Al end window.
Resolution:	<510 eV at 122 keV, Shaping Time $\leq 6 \mu\text{s}$, 1 kHz <560 eV at 122 keV, Shaping Time = $1 \mu\text{s}$, 50 kHz
Max Count Rate:	> 10 000 MeV/s (typically > 150 kHz for ^{57}Co source)
MCA:	Canberra Series 90, 16-k channels
Digital Stabilizer:	Canberra 8232
ADC:	Canberra 8077
NIM Bin:	Built in to Series 90
Bias Supply:	Canberra 3105
Amplifier:	Tennelec TC244
Count Rate Monitor:	Canberra 2071AS
Oscilloscope:	Tektronix 2225
Computer:	DEC MicroVax II, VMS vers. 4.4 5 MB memory, RD53-71 MB hard disk, TK50 Cartridge Tape, RX50 800-KB Floppy Disk Drive
Terminal:	DEC VT220
Printer	DEC LA75
Power Controller:	DEC 861C
Line Filter and Reg.:	Topaz Line 2, Model 70306
Scan Table:	Custom LANL Design
Scan Controller:	Custom LANL Design
Scan Table NIM Bin:	Tennelec MB-2/TC910
Count Rate Monitor:	ORTEC 871

Branching ratios may not be the same from one analysis parameter file to another. While they have not physically changed, adjusting them may be the only way to accommodate 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 samples with ^{240}Pu fractions ranging from 2% to 26% and ^{241}Am concentrations from 80 $\mu\text{g/g Pu}$ to 30 000 $\mu\text{g/g Pu}$. Table II presents the accepted isotopic values for a subset of the samples in this category of materials. Data from other samples are similar. While some radiochemical results are "accepted" for ^{238}Pu and ^{241}Am and a few of the ^{238}Pu values have been assigned from previous gamma-ray spectroscopy measurements, the majority of the accepted values come from mass spectrometry measurements. A few of the individual "accepted" 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 coupled with good agreement on the same isotope for several similar samples. The americium value for sample A1-86 was not available. The

TABLE II
HOMOGENEOUS (Am/Pu) SAMPLES

Sample ID	Accepted Values (wt%)					^{241}Am ($\mu\text{g/g Pu}$)	P_{eff} (mW/g Pu)
	238	239	240	241	242		
STD-151	0.0023	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*	89.690	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	16.288	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, not used in averages.

value in the table was obtained by assuming the americium concentration to be zero on the mass spectrometry date. This is clearly not correct so the value is flagged even though FRAM measurements show that the table value is not far off.

In Table III we show the 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. This is a factor of up to 3 less than the 1.5 hour count time currently used with the LANL MUDPI system. 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. The number of repeated runs ranges from 5 to 20 for these samples. These realistic measurement conditions yield measurement precisions for P_{eff} that are typically under 0.3% (1 relative standard deviation). This precision is very well matched to that observed from calorimeter measurements of total wattage.

TABLE III
SAMPLE MASS, MEASUREMENT CONDITIONS,
AND MEASUREMENT PRECISION

Sample ID	Pu Mass (g)	Ct Rate (kHz)	Single Meas. ct.time (hr)	No. of Rep. 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	5	2	10	1.01	0.12

The measurement precision for P_{eff} and ^{240}Pu in Table III is typically a factor of 1.5-2.0 better than that of the previous LANL systems² while simultaneously improving the throughput by a factor of 2-3. Table IV displays the improvement for all isotopes over the LANL MUDPI code for two samples. The first portion of the comparison shows the results of analyzing the same data with the two codes. The improvement here results from better analysis algorithms that use more of the data. The second portion of the comparison shows additional improvement arising from the ability of FRAM to acquire quality data at higher counting rates; the improvement is demonstrated with a different data set characteristic of actual FRAM measurement conditions. The 1 kg PuO_2 sample is sample STD40 listed in Tables II and III. The 500 g Pu metal sample is JOO132501 in the same tables.

In Table V we list the ratio of the average measured value to the accepted value for all the samples and data acquisition conditions in Tables II and III. At the bottom of Table V 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 24 listed samples. Note that it is under 1% 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 V would appear to indicate a possible bias for ^{241}Am at a concentration below 1000 ppm. We have extensive data on additional samples characterized by three laboratories that indicate that FRAM is accurate to about 1% for ^{241}Am concentrations as low as 300 $\mu\text{g/g}$ Pu.

TABLE IV

COMPARISON OF MEASUREMENT PRECISION: FRAM and MUDPI

Measurement Conditions		%RSD From Repeated Measurements					P_{eff}	Comment
		^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{241}Am		
1 kg PuO_2 , 12% ^{240}Pu , 13 kHz, 1 h, 30 meas.	MUDPI	3.4	0.35	2.6	0.71	6.0	0.52	same data, both codes
	FRAM	2.2	0.25	1.9	0.38	3.6	0.42	
1 kg PuO_2 , 12% ^{240}Pu , 40 kHz, 0.5 h, 15 meas.	FRAM	1.6	0.15	1.1	0.33	0.4	0.26	different meas., same sample
500 g Pu metal, 6% ^{240}Pu , 13 kHz, 2 h, 30 meas.	MUDPI	8.1	0.22	3.5	0.65	8.2	0.46	same data, both codes
	FRAM	6.2	0.11	1.8	0.33	4.1	0.26	
500 g Pu metal, 6% ^{240}Pu , 22 kHz, 1 h, 15 meas.	FRAM	7.2	0.08	1.3	0.27	1.5	0.24	different meas., same sample

TABLE V
RATIO OF MEASURED/ACCEPTED VALUES

Sample ID	Measured/Accepted					
	238	239	240	241	²⁴¹ Am	P _{eff}
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	1.00044	1.00137	0.99263	0.99964	0.98979	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.

We conclude that the bias for low americium concentrations in Table V arises from the sample characterization and not the FRAM measurement.

The table values for each sample for P_{eff} are plotted vs ²⁴⁰Pu content in Fig. 1. Values for samples with "accepted" values thought to be in error are not included (points with an asterisk in Table V). The average bias for P_{eff} for this data set is zero. This is better than the previous LANL software.² Figure 1 would be little changed if all samples were included, indicating that the suspected errors in the accepted isotopics have little effect, on the average, on P_{eff}.

B. Nonequilibrium, Homogeneous Am/Pu Materials

The same data discussed above were also analyzed with a parameter file assuming no ²⁴¹Pu-²³⁷U equilibrium. This may not prove that the analysis is correct for actual nonequilibrium

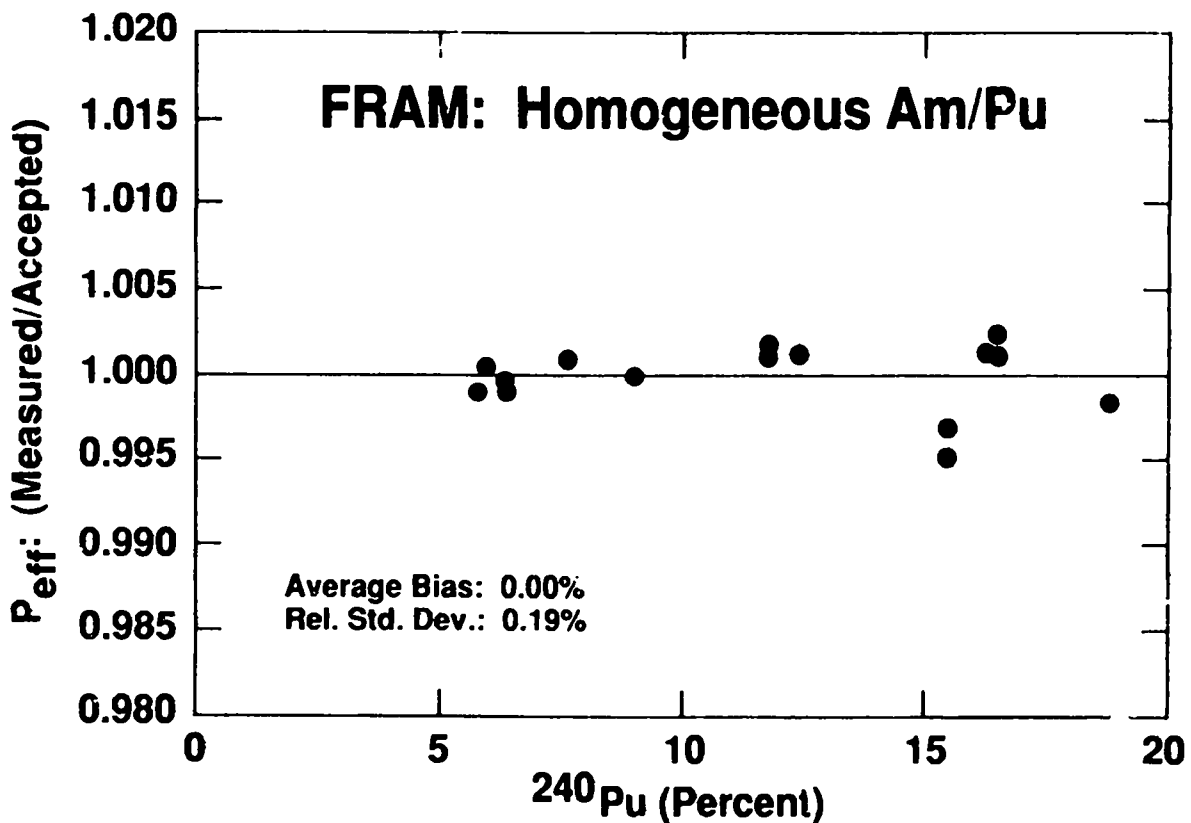


Fig. 1. Agreement with accepted values for analysis of samples with homogeneous Am/Pu.

samples, but it is a necessary condition for that to be true. The parameter file used only ^{237}U and ^{239}Pu peaks for relative efficiency and only the 148-keV peak for determining the ^{241}Pu content. The 148-keV ^{241}Pu peak was removed from the relative efficiency curve. Also, the 159-keV ^{241}Pu peak area was fixed to the 148-keV peak instead of the 164-keV peak. These results are shown in Table VI and generally show results that are comparable to the equilibrium analyses in Table V.

C. Heterogeneous Am/Pu Materials

One of the main motivations for developing this code was to analyze process residues at the Los Alamos 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 high-density plutonium metal residues are imbedded. In this situation the plutonium gamma rays are more attenuated than americium gamma rays because the plutonium gamma-rays are primarily self-absorbed by the plutonium whereas americium gamma rays are primarily absorbed by the salt matrix. Therefore absorption characteristics depend on the element emitting the photon. Thus the

TABLE VI
ANALYSIS WITH NONEQUILIBRIUM PARAMETERS

Sample ID	Measured/Accepted					
	238	239	240	241	²⁴¹ Am	P _{eff}
A1-92	0.86753*	1.00007	0.99888	0.99661	0.99349	0.99678*
A1-86	0.85857*	1.00011	0.99830	1.00238	1.03059*	0.99900*
STDR3	0.97945	1.00024	0.99602	1.00029	1.00321	0.99923
CALEX	1.07102*	0.99983	1.00387	1.00576	0.99573	1.00180*
SRPSTDPUEU7	1.00285	0.99980	1.00306	1.00422	1.09380	1.00156
PUTIBSPC1	1.03688	0.99934	1.01649	0.99923	1.05954*	1.00436*
J00132501	1.01024	0.99976	1.00384	0.99697	0.99186	1.00019
STD117	0.90523*	1.00030	0.99608	0.98445	1.00344	0.99647*
STD8	1.00909	1.00007	0.99895	0.99650	0.99061	0.99949
STD6	0.99428	0.99967	1.00495	0.99608	0.99265	1.00008
STD3	1.00962	0.99935	1.00730	1.01220	1.00197	1.00174
STD118	0.97280	1.00006	1.00014	0.98647	1.00612	0.99926
PEO382C	0.94194*	0.99850	1.01270	1.02590	0.99307	0.99831*
STD40	1.00004	0.99906	1.00637	1.00801	1.00750	1.00234
STD119	1.01116	0.99912	1.00692	0.99304	0.99625	1.00137
NBS946	0.98228	1.00382	0.97614	0.98635	1.01228	0.99969
STD116	0.98894	1.00430	0.97877	0.99674	1.00019	0.99427
STD120	0.99301	1.00433	0.97792	0.99944	0.99099	0.99245
LAO256C10	0.98140	0.99752	1.01175	1.01258	1.02016	1.00345
LAO225BS	1.00249	0.99754	1.01175	1.00760	1.00792	1.00419
STD121	1.00489	1.00016	0.99922	0.99942	1.00542	1.00102
NBS947	0.99058	1.00389	0.98523	0.98961	1.0009	0.99661
Average	0.99824	1.00031	0.99948	0.99999	1.00538	0.99981
Std. Dev.	0.01554	0.00198	0.01093	0.00972	0.02225	0.00312
%RSD	1.55697	0.19772	1.09368	0.97208	2.21265	0.31161

*Values not used in average.

relative efficiency for gamma ray emission is different for plutonium gamma rays and americium gamma rays.

Fleissner⁷ has developed a method for analyzing these materials 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. The limited comparison with chemistry showed an average bias of only 1.5%. It may seem large compared to results on other materials shown above in this report, but it must be put in proper context. If conventional nondestructive assay (NDA) plutonium isotopic 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 expect identical results because the two codes use different peak-fitting methods, analysis regions, and branching ratios.

Because only a single comparison with chemistry was available, an additional comparison program was established at Los Alamos, 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 analytical chemistry techniques by the Los Alamos analytical chemistry group, CLS-1. Some of these same samples were also measured by Fleissner at Rocky Flats prior to the crushing and blending. These comparison results for measurements on the heterogeneous samples are presented in Table VII. Table VIIa shows the accepted values from mass spectrometry (IDMS for americium) on the crushed and blended samples. In Table VIIb the measurement results for the specific power, P_{eff} , are compared to the accepted values from chemical

TABLE VII(a)
ACCEPTED VALUES OF COMPOSITION OF HETEROGENEOUS SALT RESIDUES

Sample ID	Accepted Values (wt%)					²⁴¹ Am (ug/g Pu)	P_{eff} (mW/g Pu)
	238	239	240	241	242		
MSE-1	0.00673	93.9735	5.8530	0.1467	0.0200	53 627	8.39452
MSE-2	0.01093	93.8035	5.9496	0.2032	0.0328	48 108	7.79358
MSE-3	0.01033	93.7982	5.8655	0.2636	0.0624	7 153	3.10914
MSE-4	0.00928	93.8893	5.8720	0.2044	0.0250	45 473	7.47955
MSE-5	0.00947	93.8374	5.8517	0.2700	0.0314	3 373	2.67261
ARF876642	0.00741	94.4436	5.4180	0.1160	0.0150	45 768	7.47810
XBLP121	0.01034	93.9815	5.7255	0.2456	0.0370	36 135	6.41189
XBLP278	0.00761	94.6056	5.2540	0.1088	0.0240	35 463	6.29360

TABLE VII(b)
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	

assay. For these samples with much 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 two XBLP samples came from Los Alamos and may be typical of Los Alamos process residues, so it is encouraging that the analysis by FRAM produced good results. The molten salt extraction (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. Note also that the FRAM measurements on the Rocky Flats MSE samples are at least as good as Fleissner's measurements on the identical samples. The average agreement on P_{eff} is plotted vs specific power in Fig. 2. 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% $^{235}\text{U}/\text{U}$, and the other with 12% ^{240}Pu and high concentrations of 93% enriched uranium. The accepted values and measurement conditions are in Tables VIII and IX.

The data were analyzed with two analysis parameter files; one was for high-enriched uranium samples defined by $^{235}\text{U}/\text{Pu} > 0.2$. This file used the strong ^{235}U peaks at 143, 163, 185, and

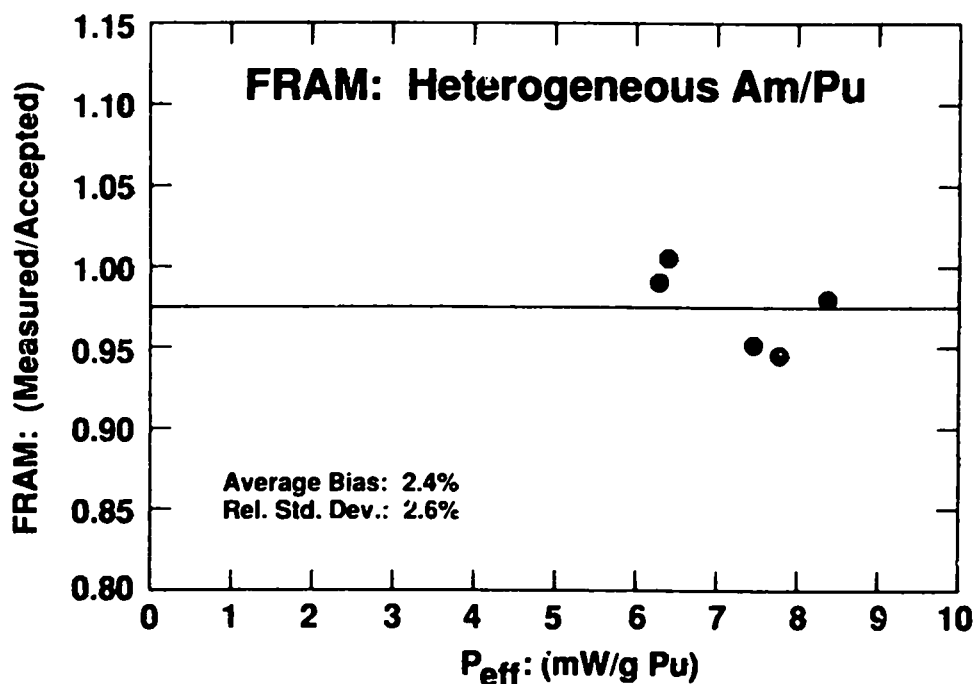


Fig. 2. Agreement with accepted values for FRAM measurements on heterogeneous salt residues.

TABLE VIII
ACCEPTED VALUES FOR PLUTONIUM ISOTOPIC
COMPOSITION OF MOX SAMPLES

Sample ID	238	239	240	241	242	$\mu\text{g/g Pu}$ ^{241}Am	mW/g Pu P_{eff}
HUA5301	0.0499	87.339	11.639	0.787	0.184	5 097	3.4015
HUA5065	0.0664	87.076	11.797	0.883	0.176	12 691	4.3717
HUA5062	0.0661	87.076	11.797	0.885	0.176	12 508	4 3493
HUA8971	0.0590	87.057	11.792	0.883	0.209	6 005	3.5653
HUA5069	0.0604	87.243	11.662	0.850	0.185	10 703	4.1028
SRPSTDPUEU2-6	0.01439	93.782	5.862	0.2761	0.0659	234	2.3419

TABLE IX
CHARACTERISTICS OF MOX SAMPLES/MEASUREMENTS

Sample ID	g Pu	g U	% ^{235}U	$^{235}\text{U}/\text{Pu}$	Ct. Time (hours)	No. of Meas.	Single Meas. Precision (%RSD) from Repeated Runs		
							^{240}Pu	P_{eff}	$^{235}\text{U}/\text{Pu}$
HUA5301	373.1	820.6	0.225	0.00496	1	6	1.2	0.20	3.8
HUA5065	306.1	441.4	0.769	0.01112	1	6	0.69	0.16	4.3
HUA5062	243.5	397.9	0.727	0.01191	1	6	1.4	0.12	2.9
HUA8971	236.3	625.9	1.022	0.02714	1	6	1.3	0.36	1.1
HUA5069	114.3	679.1	1.073	0.06397	1	6	2.5	0.23	1.2
SRPSTDPUEU6	1308.1	422.4	93.08	0.3007	1	7	1.0	0.19	0.48
SRPSTDPUEU5	348.7	1351.9	93.11	3.610	1	10	1.5	0.27	0.30
SRPSTDPUEU4	174.5	1522.8	93.11	8.125	2	9	2.0	0.30	0.29
SRPSTDPUEU3	87.14	1605.8	93.11	17.16	4	6	1.6	0.36	0.21
SRPSTDPUEU2	43.64	1646.7	93.11	35.13	3.3	5	4.3	0.51	0.20

205 keV in the relative efficiency curve. For $^{235}\text{U}/\text{Pu} < 0.2$ we did not include the ^{235}U peaks in the efficiency curve. The $^{235}\text{U}/\text{Pu}$ ratio varied from 0.005 to 35, a factor of 7500.

The two samples with the largest $^{235}\text{U}/\text{Pu}$ ratios, SRPSTDPUEU2,3 were extremely difficult to measure because of low count rates for plutonium gamma rays and intense background continua from the strong ^{235}U gamma rays. Even with long count times, measurement precision was still poor. These large $^{235}\text{U}/\text{Pu}$ ratios probably represent a practical limit to the method. Table X shows the measurement results for all the samples. The results for P_{eff} are also graphed in Fig. 3. We see a small bias of $\sim 1\%$ for ^{240}Pu on the SRPSTD samples. This probably arises from the shape of the background continuum underneath the 160- to 165-keV region and the necessity for fitting it with a single function over this broad range. There is also a bias on americium on the SRPSTD samples. These samples only have a little over 200 ppm americium, which falls at the reliability limit of the FRAM analysis although we believe that FRAM is accurate to ~ 300 ppm in the absence of perturbing effects described below. The analysis for americium in the important 125-keV region is further complicated by an interference from the ^{231}Th daughter of ^{235}U . More "tuning" needs to be done on the branching ratios in this region to reduce the bias. The bias of 1.6% for $^{235}\text{U}/\text{Pu}$ from the SRPSTD samples could not be removed by simply adjusting the branching ratio, probably because of the interaction with the use of ^{235}U peaks in the relative efficiency curve. Correction of this bias for the HUA samples is straightforward. Even with all these small bias problems, P_{eff} was still accurate, on the average, to 0.1% over this enormous range of uranium concentrations as shown in Fig. 3.

TABLE X
MEASUREMENT RESULTS--MOX SAMPLES

Ratio: Measured/Accepted

Sample ID	238	239	240	241	Am	P_{eff}	$^{235}\text{U}/\text{Pu}$
HUA5301	1.0105	0.9991	1.00062	0.9982	0.9542	0.9941	0.9924
HUA5065	0.9999	0.9984	0.9986	0.9983	1.0045	1.0006	0.9881
HUA5062	1.0077	0.9985	0.9978	0.9938	1.0184	1.0057	0.9804
HUA8971	1.0049	0.9997	1.0005	0.9936	1.0086	1.0021	0.9701
HUA5069	1.0114	0.9996	0.9945	0.9914	0.9996	0.9996	0.9863
SRPSTDPUEU6	0.9922	0.9992	1.0130	0.9970	0.9258	1.0006	0.9922
SRPSTDPUEU5	1.0039	0.9992	1.0125	1.0013	0.9213	1.0008	0.9917
SRPSTDPUEU4	1.0205	0.9991	1.0146	1.0051	0.9786	1.0023	0.9831
SRPSTDPUEU3	0.9474	1.0004	0.9932	1.0064	1.0752	0.9982	0.9854
SRPSTDPUEU2	1.0086	0.9992	1.0122	1.0040	1.400	1.0064	0.9853

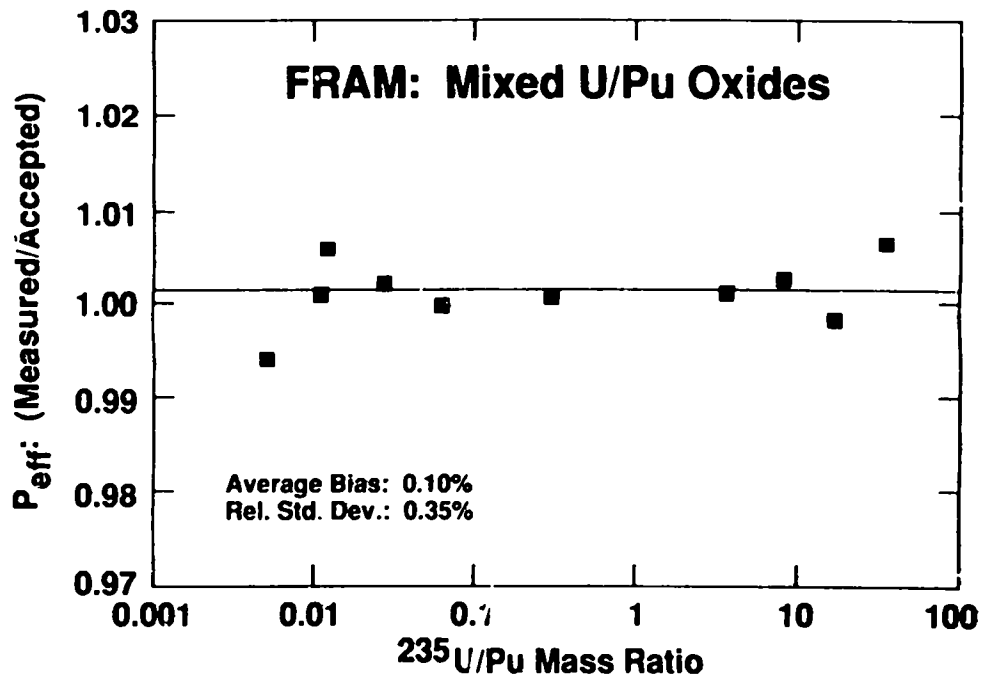


Fig. 3. Accuracy for P_{eff} from MOX samples..

E. Special Materials

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

1. Plutonium-238. 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 $^{239}, ^{240}\text{Pu}$. Limited measurements have been done but the analysis proceeded routinely. The biases that remain can be improved with more data.

2. Plutonium-242. 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. Neptunium-239. 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 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 >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. We have also analyzed materials in which the ^{243}Am - ^{239}Np is heterogeneous with respect to the plutonium in the sample.

VII. CONCLUSION

The FRAM code has demonstrated its versatility for measuring 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 is improved significantly in accuracy, precision, and throughput over previous LANL codes.

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APPENDIX A

ANALYSIS PHYSICS OF FRAM SOFTWARE

INTRODUCTION

FRAM is a fixed-energy code in contrast to some general-purpose computer codes that use a variable peak location--found by a least-squares fit of the centroid of the peak. Because the energies of the plutonium, americium, and uranium gamma rays are well known, we can know the location of the peak accurately in advance. This information reduces the number of variables in the problem, because it is not necessary to fit the peak location. Predetermining other variables governing the peak shape allows the fitting to become a linear fitting problem instead of the nonlinear, iterative problem that arises in general-purpose peak fitting codes. The general approach taken in FRAM is response function analysis.

PEAK SHAPE AND RESPONSE FUNCTIONS

The response function, which is fit to the peak by linear least-squares techniques, is a simple Gaussian with a short-term tail of the form:

$$R(X) = HT*[EXP(-A*(X-X0)**2)] + T(X)$$

with

$$T(X) = HT*[EXP(T1 + T2*ENERGY + T3*(X-X0))* \\ [1 - EXP(-T4*A*(X-X0)**2)]$$

where $A = 2.77259/FWHM**2$.

BACKGROUND DETERMINATION

The background is determined by linear least-squares fitting of selected channels in the spectrum to the function:

$$BKG(CH. NO.) = C1 + C2*(CH. NO.) + C3*(SUM OF CTS FROM \\ CH. NO. TO END OF REGION) + C4*(CH. NO.)**2$$

The number of these terms is specified in the parameter file. The third term gives the step background. Specifying a -2 for the number of terms in the parameter file gives the first and third terms only, that is, a step function on a constant, zero slope continuum.

The background is computed and stored in an array for later use. After the first iteration, when peak areas have been determined, the background is recomputed by least-squares fitting of

the same function to the (spectrum - tail) data. This is useful when the background regions are very close to a peak such that the Gaussian has gone to zero in the background channels, but some short-term tail counts are still in the background channels.

The first step in FRAM is to determine the tailing constants. This is done by first fitting a Gaussian to the selected tailing peaks from a point where the count is greater than $0.75 \cdot HT$ on the left side to $0.25 \cdot HT$ on the right side. This fit is done on the natural log of the (spectrum - background), and gives a value for the centroid, FWHM, and height (HT) of the Gaussian. This is a linear fit, because the natural log of a Gaussian is a quadratic in X . The Gaussian is then computed, and subtracted from the (spectrum - background), so that the residual is the tail. The log of the residual divided by the roll-off function $[1 - \text{EXP}(-0.4 \cdot A \cdot X - X_0)^2)]$, is least squares fit from the (centroid channel - $1.8 \cdot \text{FWHM}$) through the (centroid channel - $0.3 \cdot \text{FWHM}$). Channels with negative residuals are not included in the fit. Using data from several peaks gives the three tailing constants T1, T2, and T3. The fourth tailing constant, T4, is fixed at 0.4, a value that appears to be in general use.

The next step is the determination of the function for FWHM as a function of channel number X_0 in the form:

$$\text{FWHM}(X_0) = \sqrt{F1 + F2 \cdot X_0 + F3/X_0}$$

This proceeds by subtracting the tail function, $T(X)$ above, from the (spectrum - background) data for a peak. The result should be pure Gaussian, and its logarithm is fit by linear least squares to a Gaussian in channel number. The fitting range is from $0.5 \cdot HT$ on the left of the peak to $0.25 \cdot HT$ on the right of the peak. This range was selected because the peak is better defined in the center.

This fit gives the FWHM for a single peak. The FWHMs for all of the FWHM calibration peaks are fit by linear least-squares to the function of channel number given above. The three constants, F1, F2, and F3 are determined.

PEAK CENTROIDS FOR ENERGY CALIBRATION

Peak centroids for all peaks in the energy calibration list are needed to carry out the segment-by-segment linear energy calibration. The peak centroids are found by fitting a quadratic in channel number to the logarithm of (spectrum - background - tail). The fitting range is from $0.25 \cdot HT$ on the left of the peak to $0.25 \cdot HT$ on the right of the peak. The energy calibration is then assumed to be linear between each pair of energy calibration peaks in the list.

DIAGNOSTIC CHECKS

Four types of diagnostic checks are done after the tail, FWHM, and energy calibration have been determined. These checks compare certain parameters against predetermined limits and provide warning messages if the limits are exceeded. The centroid check does a full nonlinear fit of a (Gaussian + tail) to (spectrum - background). If the background has not been determined for that peak (peak is not in a previously defined region) a linear background is interpolated from the minimum three-channel count sums in regions of width $8 \times \text{FWHM}$ above and below the peak. This nonlinear fit applied to an energy calibration peak gives the same channel centroid as the linear procedure outlined above to within about 0.001 channels.

The FWHM check does a linear interpolation of the (spectrum - background) on each side of the peak at half maximum. Because the tail is not subtracted, the FWHM thus determined by this interpolation is slightly greater than the FWHM determined for the FWHM calibration.

The tail area fraction determines the ratio of the tail area to total (tail + Gaussian) area. This is done by evaluating the function, which is the analytical integration of the tail function.

The diagnostic check for interfering peaks is done by getting a segment of spectrum of width $10 \times \text{FWHM}$ about the suspected interference, calculating a linear background from the minimum three-channel sum over a $5 \times \text{FWHM}$ region on each side of the peak, and summing the peak area as the spectral data minus this linear background from $-1 \times \text{FWHM}$ to $+1 \times \text{FWHM}$ on either side of the peak location. This is done for two peaks, and the ratio of peak areas is printed and compared to the test limit.

A fifth diagnostic test is done after the isotopics analysis has been completed. The general form of this test compares the ratio of the mass ratios of two selected peaks against a limit. This type of test can be customized to give various diagnostics such as nonequilibrium, heterogeneous Am/Pu, or high americium data.

FITTING RESPONSE FUNCTIONS TO SPECTRAL DATA

The (spectrum - background) is fit to response functions, which are composed of one or more peaks evaluated at the corresponding channel number. There are provisions for up to ten response functions, but testing has been done with at most five response functions. A response function for a single peak is the Gaussian plus tail function, $R(X)$ given above, centered at the channel number specified by the known gamma ray energy and the linear segment-by-segment energy calibration. It is set up so that the area of the Gaussian portion of the response function is 1.0, and so that the coefficient giving the amount of the response function is itself the area of the Gaussian. The response function for multiple peaks includes a major peak with Gaussian area = 1.0 as above, but with other peaks added in and ratioed to the major peak by a factor composed of ratios of branching ratios, efficiency ratios, and (if a different isotope) activity ratios. The coefficient giving the amount of the

response function in the fit to (spectrum - background) is the area of the major peak. The areas of the minor peaks are determined from the major peak using the ratio factors.

The relative efficiency depends on the peak areas, which in turn weakly depend on the relative efficiency if peaks are ratioed to other peaks. For this reason an iterative process is performed to find peak areas, relative efficiency, and then peak areas again.

RELATIVE EFFICIENCY DETERMINATION

The relative efficiency vs energy is fit by linear least squares to:

$$\text{LOG(EFF)} = C1 + C2/E^{**2} + C3*\text{LOG}(E) + C4*[\text{LOG}(E)]^{**2} + C5*[\text{LOG}(E)]^{**3} + \text{DELTA FCN NORMALIZATION FOR EACH ISOTOPE AFTER THE FIRST} + \text{DELTA FCN NORMALIZATION FOR EACH ADDITIONAL RELATIVE EFF. FCN.}$$

RELATIVE ACTIVITY DETERMINATION

The relative activity of each isotope can be determined by linear least squares, if the relative efficiency is known, by the function:

$$\text{COUNTS} = \text{RATIO}(1)*\text{BR}(1)*\text{EFF1} + \text{RATIO}(2)*\text{BR}(2)*\text{EFF2} + \dots$$

where COUNTS is the sum of counts from all contributing peaks and the terms on the right-hand side are the products for each of the peaks in that sum. This is useful for co-energetic peaks, but also for any peaks, which overlap so that the individual peak areas are not well known while the sum of the peak areas is reliably known. There is, however, no requirement that the peaks be close together, or even in the same region.

The fitting of COUNTS to the terms above determines the activity ratios, RATIO(I), ratios of the activity of each isotope to the main isotope, isotope no. 1.

A final calculation uses the mass ratios of the isotopes to compute the isotopic percentages of the plutonium isotopes and americium. This is the only part of FRAM, besides inputting ²⁴²Pu data, that is not completely general. The code also calculates the mass ratios of any other isotope, which is not plutonium, and lists them relative to plutonium. Also specific to plutonium samples, the code calculates and lists the specific power and the amount of ²⁴⁰Pu effective.

APPENDIX B

SOFTWARE OPTIONS

Program options have been designed so that all routine operations can be performed without leaving the program. Only a few seldom-used housekeeping functions have to be accessed from an offline parameter change program, not discussed here. The options and features briefly discussed below will give the reader a good idea of the versatility of the FRAM software.

OPERATOR MENU

A--Assay

Performs a complete assay sequence including operator dialog, data acquisition, data analysis (typically takes ~ 25 s on MicroVax II), and output of results. Operator dialog can be customized to minimize the number of questions.

MC--Measurement Control

This unique feature of FRAM allows essentially "one button" initiation of a measurement control run on a standard sample. A file contains all necessary information on acceptable measurement control samples including their isotopic compositions, their dates, sample descriptions, and count times. Also included is a historical standard deviation for each measured isotope and P_{eff} . The operator merely selects the proper sample from a menu list and starts the assay. At the completion of the count time, the data are analyzed and compared to the stored, decay-corrected, accepted values. The comparison is made in units of the historical standard deviation. Deviations outside control limits are flagged and the results are produced in a form convenient for direct control charting.

Q--Quit Assay

Allows the operator to stop an assay in progress and return to the operator menu without analyzing the data.

R--Resume Assay

Allows the operator to restart data acquisition on a measurement that had been suspended with the "S" command.

S--Suspend Assay

Stops data acquisition until resumed manually with the "R" command. Useful for filling LN dewars during data acquisition.

SU--Supervisor Functions

Allows users with proper authority to access the supervisor menu. See description of supervisor functions below.

T--Terminate Assay

Allows the operator to stop an assay in progress and return to the operator menu with analysis of data collected to that point.

X--Exit the Program

Returns the user to the VMS operating system. Must have proper authority to carry out this operation.

SUPERVISOR MENU

AD--Assay From Disk

Analyzes spectral data files stored on the system disk. Data are read directly into the computer, bypassing the MCA. This mode can be used for off-line analysis.

AU--Autocycle

Enables an operator to repeat an assay sequence a selected number of times without requiring intervention between measurements.

AUD--Autocycle From Disk

Analyzes multiple data files from the system disk that have been stored with sequential file-name extensions with the AU option. No operator intervention is required between analyses.

CH--Change Parameters

Allows the user to access nearly every parameter that governs the physics analysis and operator interface. Discussed more fully in a separate section below.

ISO--Run ISOPOW

Runs the ISOPOW code which calculates decay corrections for plutonium and americium isotopic results. Can be used concurrently with other system operations.

RD--Read Spectrum into MCA

Reads a spectrum from a data file on the system disk into the MCA memory.

WD--Write Spectrum to Disk

Writes spectral data from MCA memory to a data file on the system disk.

X--Return to Previous Menu

Returns the user to the Supervisor menu.

CHANGE PARAMETERS

Change Defaults. Eight different flags or parameters can be set with this option, without depending on the analysis parameter file in use.

Ask operator for sample power?

Ask operator for 242Pu information?

Database name for analysis parameters (default database)

Instrument identification

Number of iterations in analysis

Level of detail printed in analysis results

Should spectral counts be automatically saved on disk?

Is operator terminal a VDT (Video Display Terminal)?

Change Analysis Parameters. This part of the CH option controls the physics analysis. These parameters are keyed to a specific analysis parameter file or database. Selection of most of these options invokes another menu to allow editing of the associated parameters.

Database name

Peaks for relative activity and isotopics calculation

Initial values for constants

Display a spectrum in the Series 90 MCA

Peaks and isotopes for efficiency calibration

Peaks for FWHM calibration

Isotope names and data

List parameter values

Peaks for energy calibration

Peak data

Region data

Peaks for shape calibration

Switch to another parameter database

Exit

Change Diagnostic Parameters. These options set parameters that control measurement diagnostics made on each spectrum to assure proper spectral quality.

Database name

Stabilization peaks

FWHM peaks

Tail peaks

Interference peaks

Mass ratios

APPENDIX C

THE NAME "FRAM"

It has been a longstanding custom in the Safeguards Assay Group for the project software engineer to name the analysis code that he/she is developing. In this case the software developer also developed the physics of the code. The author (GWN) being of Scandinavian descent chose the name FRAM for this code as explained below.

FRAM is the acronym for **F**ixed-energy **R**esponse-function **A**nalysis with **M**ultiple efficiency, and as such describes the details of this spectrum analysis code. At the same time, FRAM, in the Scandinavian languages, means "forward" or "onward."

FRAM "Onward!" was the battle cry of the Vikings in the struggles for unification of the Nordic lands. FRAM was also the name chosen by Fridtjof Nansen for the versatile sailing ship used on his arctic expedition (1893-96). FRAM permitted Nansen to come closer to the North Pole than anyone before him. The versatility of the polar ship FRAM was demonstrated when Otto Sverdrup used her for another arctic expedition (1898-1902), and when, after only minor modifications, Roald Amundsen sailed FRAM to and through the antarctic seas on his successful expedition to the South Pole (1910-1911).

It is hoped that spectrum analysis with FRAM may bring us closer to the goal of extracting all the information from gamma-ray spectra with accuracy and precision, and that the versatility of FRAM will be applied to as yet unforeseen analysis problems.