*Gamma-Ray Isotopic Analysis
Development at Los Alamos*

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GAMMA-RAY ISOTOPIC ANALYSIS DEVELOPMENT AT LOS ALAMOS

by

Thomas E. Sampson

ABSTRACT

This report describes the development history and characteristics of software developed in the Safeguards Science and Technology group at Los Alamos for gamma-ray isotopic analysis. This software analyzes the gamma-ray spectrum from measurements performed on actinide samples (principally plutonium and uranium) of arbitrary size, geometry, and physical and chemical composition. The results are obtained without calibration using only fundamental tabulated nuclear constants. Characteristics of the current software versions are discussed in some detail and many examples of implemented measurement systems are shown.

FIRST PROPOSAL

In 1974, Jack Parker and Doug Reilly at Los Alamos proposed the first practical method for measuring the isotopic composition of an arbitrary (size, shape, composition, and measurement geometry) plutonium sample via analysis of its gamma-ray spectrum.¹ The key to their method was the incorporation of an internal or intrinsic self-determination of the relative efficiency curve from the gamma-ray spectrum of each unknown sample. This method has formed the basis of all development work at Los Alamos and is used worldwide in all software for the isotopic analysis of arbitrary samples.

MEASUREMENT FUNDAMENTALS

The fundamental measurement is one of isotopic ratios given by

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

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

The Cs are determined from the measured photopeak areas in the gamma-ray spectrum of the sample under study. $T_{1/2}$ and BR are fundamental known nuclear constants. Only the relative efficiency ratio remains an unknown. The relative efficiency is determined from each measured spectrum by noting the energy variation of the quotient of C/BR for a series of gamma rays from a single isotope. This quotient takes into account the variations caused by detector efficiency, sample self-absorption, sample geometry, sample composition and matrix (i.e., metal, oxide, solution, waste), and filters or absorbers placed between the sample and detector. Figure 1 shows an example of relative efficiency curves.

Fig. 1. A relative efficiency curve for the same sample measured with two types of high-purity germanium (HPGe) detectors. The curves are arbitrarily normalized at 414 keV. The different shapes are largely the result of the different intrinsic efficiency of the two detectors.

Figure 2 displays general characteristics of the relative efficiency curve as affected by sample mass for the same detector. The trends are illustrative, only, as the exact shape of the relative efficiency curve depends upon the specific details of the sample-detector measurement configuration.

Fig. 2. Relative efficiency curves for different sized samples measured with the same detector. The curves are arbitrarily normalized to a value of 1.0 at their maximum. The shape differences are largely the result of the different self-absorption characteristics of the samples.

Reference 2 develops the isotopic ratio expression in Eq. 1 and also shows how the isotopic ratios may be combined to produce a complete isotopic distribution. This technique has been used to measure all of the isotopes of plutonium (except ^{242}Pu) including ^{241}Am , the isotopes of uranium (except ^{236}U), and many other actinides contained as impurities in plutonium. The only assumption built into this method is that of isotopic homogeneity—that is, all isotopes have the same physical distribution in the sample.

MUDPI/LAPIS

The first computerized implementation of the principles developed by Parker and Reilly was accomplished in 1980.^{3,4,5} This software used on-line control of data acquisition and analysis with PDP/11 computers and the RT-11 operating system. The analysis featured simple region-of-interest (ROI) summation to obtain peak areas.⁶ Small, planar (16-mm diam x 10–13 mm deep) HPGe detectors with very high resolution were used for data collection using 4096 channels spanning the energy range from 10–420 keV. Detector resolution was typically ~500 eV at 122 keV. Specific peak ratios to be calculated in the 120–420-keV region were fixed in the code.

A system with multiple detectors (measure multiple samples simultaneously) and a single multichannel analyzer and computer was implemented at the Los Alamos Plutonium

Facility in 1981. The Multiple Detector Plutonium Isotopic (MUDPI) system was used for all nondestructive isotopic measurements at Los Alamos until the late 1980s. The MUDPI system is pictured in Fig. 3.

Fig. 3. The MUDPI analysis system implemented at the Los Alamos Plutonium Facility in the early 1980s.

A similar system with a single detector was fielded at the Savannah River site in 1981, actually preceding the MUDPI system. This system, called Los Alamos Plutonium Isotopic System (LAPIS), is pictured in Fig. 4. It was fielded in an air-conditioned instrument rack and the sample, placed on the platform above the up-looking detector, was surrounded with a shielding clamshell to reduce radiation dose to the operator.

References 7 and 8 describe both of these systems, including the data acquisition equipment, isotopic ratio algorithms, software user interface, and data from the initial operation of the systems. These systems used conventional analog NIM electronics with the amplifier time constant set at 3 μ s and a maximum recommended counting rate of 15 kHz. The measurement precision, or repeatability, for the effective specific power under these counting conditions was $\sim 0.5\%$ for a 2-h count on a 500-g plutonium sample. The precision for ^{240}Pu for the same conditions fell into the range from 2%–4% [1 relative standard deviation (RSD)].

The MUDPI system performed very well at Los Alamos throughout most of the 1980s. However, by the mid-1980s it became apparent that improvements were needed to address two measurement problems. The first problem was the presence of interference isotopes in some samples. Gamma rays from these interfering isotopes (^{235}U , ^{239}Np , ^{237}Np) were not accounted for in the simple ROI analysis, leading to analysis errors when

Fig. 4. The LAPIS plutonium isotopic analysis system implemented at the Savannah River Plant.

these gamma rays overlapped a peak or background ROI. A second problem arose from the analysis of residue samples from pyrochemical processes. These processes produce residues consisting of americium in low-Z chloride matrix with plutonium metal fines suspended in the matrix. For these samples, gamma rays from americium suffer different attenuation than plutonium gamma rays of the same energy. This violated the isotopic homogeneity assumption of the analysis method and especially led to errors in the $^{241}\text{Am}/\text{Pu}$ measurement.

FRAM DEVELOPMENT

The development of the fixed energy response function analysis with multiple efficiencies (FRAM)* as a replacement for MUDPI began in the mid-1980s with the code first fielded in 1988 at the Los Alamos Plutonium Facility. The FRAM⁹ code represented a major advance in measurement flexibility as it was designed to address the shortcomings of the MUDPI/LAPIS software and update the measurement and analysis hardware

* "FRAM" is a word in the Scandinavian languages meaning "forward" or "onward." FRAM was the name chosen by Fridtjof Nansen for the versatile sailing ship used on various Arctic and Antarctic expeditions from 1893 to 1911. The code was named by Dr. George W. Nelson, its original developer.

to the state-of-the-art at that time. Some of the features and characteristics of the FRAM code were as follows.

- MicroVAX computer and VMS operating system with software written in FORTRAN 77.
- Response function analysis for peak area determination. Allows fitting of multiple, overlapping peaks.
- Heterogeneous Am/Pu analysis capability using separate relative efficiency curves for heterogeneous isotopes.
- Capability to select any or all spectral peaks to contribute to analysis via least squares resolution of isotopic ratios.
- User-editable analysis parameters govern all facets of the analysis allowing the user to cope with arbitrary interference peaks, variable energy calibrations, different detector types, and the widest possible variety of matrix conditions.

The last bulleted feature of the FRAM code can be considered its defining characteristic, allowing FRAM to successfully analyze a wider range of materials than any other single code available.

FRAM was installed at the Los Alamos Plutonium Facility in 1988 and was put into routine operation in 1989. Figure 5 shows the FRAM system that was in use until 1997. This system used two planar HPGe detectors, allowing two samples to be measured simultaneously. The samples were placed on a scanning table in front of the detector and were rotated and translated vertically during the measurement. Scanning was performed primarily to improve measurements on the isotopically heterogeneous pyrochemical residues. The scanning mechanism was completely shielded to reduce operator exposure from the sample. The detector table and detector shield (hidden behind the shielded scanning mechanisms shown in Fig. 5) could be manually positioned to vary the sample-detector distance and hence the detector counting rate. Improvements in detectors and electronics permitted measurements at throughput rates significantly higher than those used with MUDPI/LAPIS. Triangular shaping with a 1- μ s time constant allowed a maximum recommended counting rate of 40 kHz.

In addition to implementing the capability improvements mentioned above, FRAM also allowed more rapid data collection with significantly improved precision in a shorter counting time. Tables I and II below display the summary results for Los Alamos from the Department of Energy (DOE) Calorimetry Exchange Program for the years 1988 to 1998, spanning the change over from MUDPI to FRAM.^{10, 11} The DOE Calorimetry Exchange Program tabulates the results from the facility's measurements of a standard PuO₂ sample containing 400 g of plutonium with about 6% ²⁴⁰Pu content. Each facility collects data in a manner suitable for their own operations. The Los Alamos data in Tables I and II were collected in the same manner as routine unknown samples were measured at the Los Alamos Plutonium Facility. Table I tabulates the precision or repeatability of a single measurement derived from the distribution of measurements over a period of a year. Table II tabulates the bias or difference between the measured and accepted value. The data are

tabulated for the difficult-to-measure ^{240}Pu isotope and the effective specific power (W/gPu) derived from the entire isotopic distribution.

Sample inside
shielded
measurement
station

Fig. 5. Testing a FRAM system before installation at the Los Alamos Plutonium Facility.

Table I. Los Alamos Calorimetry Exchange Results 454 g PuO_2 Calorimetry Exchange Standard, 5.86% ^{240}Pu Measurement Repeatability					
Year	# of Meas.	% RSD Single Measurement		Code*	Ct. Time (h)
		^{240}Pu	Peff		
1988	40	3.49	0.36	MP	2
1989	67	4.26	0.56	MP	2
1990	46	2.77	0.41	F	1
1991	42	2.07	0.28	F	1
1992	40	1.64	0.24	F	1
1993	44	1.73	0.20	F	1
1994	36	1.29	0.22	F	1
1995	51	1.58	0.25	F	1
1996	44	1.33	0.27	F	1
1997	91	1.60	0.25	F	1
1998	92	1.69	0.25	F	1

* MP = MUDPI, F = FRAM

Table II. Los Alamos Calorimetry Exchange Results 454 g PuO ₂ Calorimetry Exchange Standard, 5.86% ²⁴⁰ Pu Measurement Bias: [(Measured - Accepted)/Accepted] * 100					
Year	# of Meas.	Measurement Bias (%)		Code	Ct. Time (h)
		²⁴⁰ Pu	Peff		
1988	40	0.30	0.16	MP	2
1989	67	0.78	0.22	MP	2
1990	46	0.54	0.20	F	1
1991	42	-0.26	-0.05	F	1
1992	40	0.14	0.00	F	1
1993	44	0.19	0.19	F	1
1994	36	-0.14	0.11	F	1
1995	51	-0.48	0.03	F	1
1996	44	0.42	0.04	F	1
1997	91	1.27	0.09	F	1
1998	92	0.38	0.01	F	1

Table I shows not only a decrease in the counting time from 2 h to 1 h but also a simultaneous improvement in precision during the gradual switch over from MUDPI to FRAM. The precision continued to improve as the operators became familiar with FRAM. The improvement in precision arose from

- Ability to use more gamma-ray peaks in the analysis. A typical FRAM analysis uses over 60 peaks while MUDPI used about 20.
- Improved detectors and data acquisition electronics allowed data collection at higher count rates and shorter time constants, yielding greater throughput.

Table II shows, in contrast to Table I, little change in the measurement bias moving from MUDPI to FRAM. The biases for both codes are near to the limits of the technique.

PC/FRAM DEVELOPMENT^{12,13}

By the early 1990s, computer hardware and software developments made the VAX/VMS-based FRAM system obsolete. The program was recoded in C to operate on a PC under Windows 3.1. This advance was necessary to open up the applications for the FRAM code (now called PC/FRAM) at other facilities that did not support the previous VAX system. This change has resulted in FRAM becoming commercially available and used worldwide.

PC/FRAM has preserved all of the principal features of the VAX FRAM code while adding significant new capabilities.

Single Detector System. Like all previous Los Alamos isotopic analysis systems, PC/FRAM uses only a single detector to acquire its data. We have made a conscious choice to keep FRAM a single detector system because single detector systems are inherently more versatile, easier to use, more reliable, and less expensive; they also occupy less facility space.

Planar or Coaxial Detector. PC/FRAM is the only isotopic analysis system that can obtain a complete isotopic analysis using either a single planar or a single coaxial detector. When using the traditional single planar detector, PC/FRAM has most often been used to collect and analyze data in the 120–420-keV range, although it is not limited to this range. The most widely used mode of operation with a single coaxial detector is to acquire a single spectrum in the range from 0–1024 keV. Various analysis modes can then be used with this wide data range. If the widely used region between 120 and 200 keV is available, PC/FRAM will work best analyzing in an energy range from 120–450 keV. When analysis below 200 keV is precluded (sample shielding or thick-walled sample container), PC/FRAM can still obtain a complete isotopic analysis using only gamma rays above 200 keV from a single coaxial detector spectrum. A complete analysis (all measurable isotopes) using only gamma rays above 300 keV is also possible.

The optimum choice of planar or coaxial detectors is made only after all possible measurement applications are considered. The planar detector is usually the detector of choice if all measured items are unshielded or contained in “thin” containers. If shielded containers, thick-walled containers, or a mixture of thin and thick/shielded containers are encountered, then a single coaxial detector system is optimum. PC/FRAM is the only available isotopic analysis method using a coaxial detector in the energy range from 120–300 keV.

Shielded Samples. Most other isotopic analysis codes (including the original FRAM) require the presence of spectral peaks in the region below 200 keV, regardless of whether they acquire data from one or two detectors. When this region is not available to the spectroscopist, perhaps because the sample is shielded to lower radiation exposure or because the sample is inside a very heavy-walled container, other isotopic analysis codes may not function. The PC/FRAM code was the first code to demonstrate the ability to make measurements through thick-walled containers or on shielded samples. Any software that obtains its results from gamma rays and x-rays in the region around 100 keV is easily defeated by as little as a few tenths of a millimeter of lead or ~10 mm of steel. FRAM measurements have been made through as much as 13 mm (0.5 in.) of lead and very easily through 25 mm of steel. These are examples only and do not exhaust the full range of capabilities in this area.

Two containers that present no problems for FRAM measurements are shown in Figs. 6 and 7. Figure 8 shows how a coaxial detector spectrum in the energy range from 0–1024 keV is affected by various thicknesses of lead.

Fig. 6. Container for dismantled weapons components.

Fig. 7. Lead-lined, short-term storage container.

User-Editable Parameter Database. A user-editable parameter database allows the FRAM code to have the greatest possible flexibility in acquisition and analysis. The user has complete control over the setup of acquisition parameters, analysis parameters, diagnostic parameters, data storage formats, and default and global settings. All parameters may be changed from within a password-protected (three levels of protection) Change Parameter option that has the look and feel of a standard spreadsheet. This allows the user to tailor the PC/FRAM analysis to almost any imaginable set of measurement conditions. It is important to note that the user can make these changes; a dedicated computer programmer is not required, although the user should be knowledgeable in gamma-ray spectroscopy measurements on plutonium. A description of the many parameters accessible in this fashion can be found in Ref. 14.

Fig. 8. Gamma-ray spectrum from plutonium shielded by lead. The no lead and 1.6 mm lead samples are identical, 400 g of PuO_2 with 5.9 % ^{240}Pu . The 13-mm lead sample contains 300 g of plutonium as PuO_2 with 9.7 % ^{240}Pu .

Parameter Set. A complete group of parameters in the parameter database is called a Parameter Set. Parameter Sets exist for all common acquisition and analysis modes so the user is freed from the initial responsibility of setting up the system. The structure of the parameter database and its presentation to the user makes “on-the-fly” changes to the analysis a simple task. This flexible and versatile parameter database structure allows the user to analyze the widest possible variety of items without any reprogramming or software changes. Parameter Sets exist for all of the common measurement situations although no single set of parameters can tackle the complete range of possible materials. Table III displays this flexibility.

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

- 2%–95% ^{240}Pu
- 0.01%–50% ^{241}Am
- Interferences from ^{243}Am – ^{239}Np , ^{237}Np , ^{244}Cm , ^{137}Cs and other fission products or radioisotopes
- Nonequilibrium ^{241}Pu – ^{237}U
- MOX: $^{235}\text{U}/\text{Pu}$ from 0.005 - 35
- ^{234}U , ^{235}U , ^{238}U in uranium (only), no Pu, 0.2–>98% ^{235}U
- 80% ^{238}Pu (heat source grade material)
- Lead-shielded samples (up to 13 mm demonstrated for Pu)
- Steel-shielded samples (up to 25 mm demonstrated for Pu and up to 16 mm demonstrated for U)
- Heterogeneous Am/Pu
- 80%–95% ^{242}Pu (minor isotopic ratios only)
- ^{235}U : ^{241}Am : Pu = 24:1:1 as an example of waste materials

PC/FRAM PERFORMANCE

The performance of the FRAM and PC/FRAM software has been extensively documented for both bias and precision. We display Table IV to illustrate measurement bias for three types of measurement configurations. Here we define bias as the ratio of the measured value to the accepted value. The accepted values for plutonium are derived from mass spectrometry measurements (alpha counting in some cases for ^{238}Pu). The bias here is derived from repeated measurements from a number of items with well-known accepted values. The items range in size from less than 0.5 g of plutonium to about 2 kg of plutonium with the ^{240}Pu content ranging from 3% to 26%. The generally accepted goal for these measurements is to be able to measure each isotope with an accuracy of better than 1%. The averages shown here demonstrate this accuracy goal.

Table IV. PC/FRAM Measurement Bias for Three Measurement Configurations			
Ratio: Measured/Accepted Values			
Isotope	Planar Detector 120-420 keV	Coaxial Detector 120-460 keV	"Shielded" Coax Detector 200-800 keV
^{238}Pu	0.9929	1.0013	1.0075
^{239}Pu	0.9999	1.0001	0.9993
^{240}Pu	1.0005	1.0005	0.9999
^{241}Pu	0.9969	1.0005	1.0017
^{241}Am	1.0028	1.0013	1.0064
P_{eff}	1.0001	1.0005	1.0031
# of Samples	28	29	36
# of Measurements	420	367	448

Measurement biases for individual items are often not as good as the biases averaged over a group of items of different types, sizes, and isotopic distributions. Table V displays the standard deviation of the distribution of the individual item measurement biases about the mean. Each individual item's measurement bias was determined with high statistical precision from the measurements in Table IV. The standard deviation of this distribution can be thought of as an individual item bias and may be a more realistic way to think of measurement bias for an individual measurement.

Table V. Distribution of Individual Bias Values (% RSD)			
Isotope	Planar Detector 120-420 keV	Coaxial Detector 120-460 keV	"Shielded" Coax Detector 200-800 keV
^{238}Pu	4.42	1.93	3.83
^{239}Pu	0.12	0.18	0.35
^{240}Pu	0.59	0.89	1.90
^{241}Pu	0.62	0.39	2.32
^{241}Am	1.94	2.33	1.28
P_{eff}	0.30	0.37	0.62
# of Samples	28	28	36

Plots of the data from Tables IV and V are given below in Figs. 9 and 10. The plots are made for the two most widely used parameters derived from gamma-ray based isotopic measurements, the effective ^{240}Pu fraction, $^{240}\text{Pu}_{\text{eff}}$, and the effective specific power P_{eff} . The effective ^{240}Pu fraction is used to convert neutron coincidence counting measurements of the effective ^{240}Pu grams in the sample to grams of total plutonium. Equation 2 defines this parameter,⁶

$$^{240}\text{Pu}_{\text{eff}} = 2.52 \text{ } ^{238}\text{Pu} + ^{240}\text{Pu} + 1.68 \text{ } ^{242}\text{Pu} . \quad (2)$$

The effective specific power (W/g plutonium) is defined in Eq. 3. It is used to convert the Watts measured in a calorimeter to grams of total plutonium.⁶

$$P_{\text{eff}} = \sum P_i R_i , \quad (3)$$

where

- P_i = the specific power (W/g isotope) of the i th isotope
- R_i = mass fraction of the i th isotope relative to plutonium,

and the sum is taken over all heat-producing isotopes, usually the plutonium isotopes plus ^{241}Am .

Fig. 9. Bias for PC/FRAM measurement of the effective ^{240}Pu fraction for three measurement methods.

Fig. 10. Bias for PC/FRAM measurement of the effective specific power for three measurement methods.

The small differences between the precisions shown in the legend in Figs. 9 and 10 and in Table V arise from the exclusion in Table V of the coaxial detector point for ^{240}Pu with a ratio of 1.05, thought to be an outlier and also from minor differences in the data sets used between the plot and the table.

The figures show that the planar detector is still the detector of choice, if measurement conditions permit its use although the coaxial detector (120–450 keV) is nearly as good. The high points for the effective specific power between 20% and 25% ^{240}Pu for the “shielded” coaxial configuration arise from very small samples (0.4 g of plutonium). These samples showed biases of ~1.5% for ^{241}Am and ^{238}Pu , which contribute heavily to P_{eff} for these high burn-up samples

The discussion above treats measurement bias and its sample dependence. Also of concern is the measurement precision, or repeatability for a single measurement. As before, this is of most concern for the parameters P_{eff} and $^{240}\text{Pu}_{\text{eff}}$. Figures 11–13 display FRAM’s measurement precision for the three commonly used measurement/analysis configurations. Measurement precision depends upon many quantities including counting time, sample size, sample isotopic composition, measurement and sample geometry, filtering, and detector size and resolution. While not all of these parameters are controlled, the data represent a reasonable mix of conditions. The reader should also refer to the Calorimetry Exchange data in Table I for similar data.

Fig. 11. Measurement precision or repeatability (1 RSD) for a 1-h FRAM measurement using a 16-mm diameter by 13-mm thick planar HPGe detector. The precision is that observed from repeated measurements.

Fig. 12. Measurement precision or repeatability (1 RSD) for a 1-h FRAM measurement using a 25% relative efficiency coaxial HPGe detector with analysis from 120 to 450 keV. The precision is that observed from repeated measurements.

Fig. 13. Measurement precision or repeatability (1 RSD) for a 1-h FRAM measurement using a 25% relative efficiency coaxial HPGe detector with analysis from 200 to 800 keV. The precision is that observed from repeated measurements.

URANIUM MEASUREMENT CAPABILITY

FRAM was the first isotopic analysis software to demonstrate the ability to measure the isotopic composition of arbitrary items containing only uranium.¹⁵ These first measurements reported in 1990, made with the original FRAM (microVAX) software and measuring the $^{235}\text{U}/^{238}\text{U}$ ratio, demonstrated the first practical method for in-plant implementation. In addition, the uranium capability used the same FRAM software, unmodified from its normal use for plutonium measurements.

The uranium measurement capability was improved with the development of PC/FRAM and extended to the measurement of ^{234}U . The ^{236}U isotope has no gamma-ray signature (analogous to ^{242}Pu) and cannot be quantified by gamma-ray spectroscopy. Future FRAM versions will estimate ^{236}U with an isotopic correlation. FRAM uranium measurements use gamma rays in the energy range from 120–1001 keV and are typically made with a modest-sized coaxial HPGe detector (25%–30% relative efficiency) with 8K channels of data. The peak-fitting requirements for uranium are much less stringent than those for plutonium as most of the peaks are single peaks, well separated from neighboring peaks. However, fitting a relative efficiency curve to the data is more difficult because there are two widely separated groups of peaks, 140–200 keV from ^{235}U and 700–1001 keV from the $^{234\text{m}}\text{Pa}$ daughter of ^{238}U with nothing in between. A gamma-ray spectrum from items with three different enrichments is shown below in Fig. 14. Figure 15 shows the relative efficiency curve from a natural uranium sample, as displayed by the PC/FRAM software.

Fig. 14. HPGe detector spectra of three different uranium enrichments.

As the ^{235}U enrichment increases, daughter products from the decay of small amounts of ^{232}U (Th decay chain) formed in recycled uranium by reactor irradiation also increase, allowing the opportunity to use these gamma rays in the relative efficiency curve. Peaks at 238, 583, and 860 keV may be used to supplement the relative efficiency curve for enrichments above ~10% and are usually very useful for enrichments >50%. A relative efficiency curve for a 93% enriched ^{235}U sample is shown in Fig. 16. Here the triangles are the points from the ^{232}U daughters (labeled ^{228}Th). The two points at ~260 and ~950 keV that fall off the curve are peaks from ^{238}U that are very weak for highly enriched uranium.

Figure 17 displays the accuracy of FRAM for uranium. Accuracy for ^{235}U is generally in the range of 1%–3%. Measurements have been demonstrated for ^{235}U enrichments ranging from 0.2% to >97.5%. Uranium-234 can also be measured at levels above ~0.005%.

Fig. 15. Relative efficiency curve for a sample of natural uranium (0.71% ^{235}U). Figure is from the Cathode Ray Tube (CRT) display in PC/FRAM. Note the absence of relative efficiency points between 250 and 750 keV.

Fig. 16. Relative efficiency curve for a sample of 93% ^{235}U . Figure is from the CRT display in PC/FRAM.

Fig. 17. Measurement bias for ^{234}U and ^{235}U .

Coincidence summing can affect the results of uranium isotopic analysis measurements using FRAM, giving rise to a sample-detector distance measurement bias for distances close to the detector. Vo has discussed this problem in Ref. 16.

Just as for plutonium, FRAM can measure the isotopic composition of uranium through containers with thick walls or modest lead shields. The range of application is less than that for plutonium because one likes to use ^{235}U gamma rays at 143 and 163 keV in addition to the prominent gamma ray at 185.7 keV. Nevertheless, FRAM can make uranium measurements on many samples that defeat measurement methods using the 100-keV region. FRAM has made field measurements on low-enriched (<5% ^{235}U) UF_6 cylinders with steel walls 13 mm thick.¹⁷ Similar laboratory measurements have been carried out also on low-enriched uranium through 16 mm of steel. Measurements on enrichments >20% have been demonstrated through 1.6 mm of lead with lower enrichments possible but not investigated.

The FRAM code was used in the uranium enrichment measurement exercise held at the Institute of Reference Materials and Measurements (IRMM) at Geel, Belgium, in March 1997.¹⁸ Table VI shows the average results of all FRAM measurements made during the exercise.

Table VI. FRAM Results for IRMM Uranium Enrichment Exercise				
Sample	Measured wt.% ²³⁵ U mean ± sigma(mean)	Certified wt.% ²³⁵ U	% Difference 100*(M - C)/C	Comment
1614	1.521 ± 0.011 (0.71%)	1.4972	1.59 %	
1541	1.984 ± 0.018 (0.91%)	1.9952	-0.56 %	
1542	2.826 ± 0.023 (0.80%)	2.8774	-1.79 %	
1613	3.097 ± 0.061 (2.0%)	3.1094	-0.40 %	
125VA	93.35 ± 0.048 (0.051%)	93.1556	0.21 %	
X	3.316 ± 0.049 (1.48%)	3.4317	-3.37 %	freshly separated
Y	2.547 ± 0.011 (0.43%)	2.6846	-5.13 %	freshly separated
Average difference excluding freshly separated			-0.19 %	

FRAM cannot measure freshly separated uranium materials (materials for which the ^{234m}Pa daughter of ²³⁸U is not in secular equilibrium). This decay is driven by the 24.1 d half-life of the intervening ²³⁴Th daughter. For samples X and Y in Table VI, we made a correction using the known separation time. The correction reduced the error from ~20% to the ~-4% errors shown. The residual error may arise from an incomplete separation.

PC/FRAM MEASUREMENT SYSTEMS

Several types of detector/measurement hardware combinations have been implemented in the field. The FRAM system at the Los Alamos Plutonium Facility has been shown in Fig. 5. One of the two planar detectors in this system has been replaced with a coaxial detector for added versatility and the computer system has been upgraded to laptop PCs. In addition, a mobile PC/FRAM system with a coaxial detector is also in use at Los Alamos. This system is shown in Fig. 18. This mobile system can be taken to the measurement, yet provides a configurable, shielded detector for use in difficult environments. Another small system that has seen wide use is shown in Fig. 19. The system shown in Fig. 19 was used in the uranium enrichment measurement exercise at IRMM in Geel, Belgium. This system was also used at Rocky Flats to verify, for the IAEA, the isotopic content of plutonium oxide samples in lead-lined containers.

Portable, battery/mains-powered systems are increasingly used with FRAM. The field measurements on UF₆ cylinders mentioned previously¹⁷ used a system similar to that shown in Fig. 20. In the same measurement exercise, the equipment also measured isotopic composition of uranium waste containers, uranium feed hoppers, mixed oxide fuel pellets, plutonium samples through 25 mm of steel, and plutonium scrap and waste containers.

Fig. 18. A mobile PC/FRAM system used at the Los Alamos Plutonium Facility. The detector arm can be raised, lowered, swiveled, and tilted. Electronics are conventional mains-powered NIM modules. The detector is shielded with 0.75 in. of tungsten.

Fig. 19. This system is used when it is inconvenient to bring the mobile system of Fig. 18 to the measurement location.

Fig. 20. Portable, battery/mains-powered systems are used where access is restricted.

Many systems like that shown in Fig. 21 are installed in the United States and Russia. More recently the NIM bin and modules are often replaced with a single integral unit containing all the functions. Digital signal processors, with their better performance, are beginning to replace the conventional NIM modules.

Fig. 21. Systems like this are in use in the United States and at many installations in Russia.

Three systems, shown in Fig. 22, are in use at the Plutonium Fuel Processing Facility in Japan for measurement of wastes in lead-lined drums. These systems incorporate a very simple manual detector positioning system.

Fig. 22. Waste measurement systems used at the Plutonium Fuel Processing Facility in Japan.

The FRAM software is also a part of integrated nondestructive assay systems using robotic sample handling, host computer control, and integration with other nondestructive assay instruments such as calorimetry and neutron coincidence counting.^{19,20,21} Figure 23 below shows the Advanced Retirement and Integrated Extraction System (ARIES) nondestructive assay system for assaying materials from a weapons component dismantlement process. This system contains a planar HPGe detector for measurement of DOE Std 3013-96 containers of plutonium metal and oxide from dismantled weapons.

Fig. 23. The FRAM system in the ARIES system incorporates automated sample-detector positioning for counting rate optimization.

Planar HPGe detector for FRAM with the ARIES system.

SUMMARY

We have described the development of gamma-ray isotopic analysis software at Los Alamos beginning with the first exposition of the measurement principle in 1974 to the current time of September 1998. Examples of many of the systems that have been put in use have been presented. We have documented the performance of these systems and presented historic measurement control data from an operating facility. Lastly, we have described many of the features of the currently used FRAM software and its worldwide application.

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