2. PLUTONIUM ISOTOPIC ANALYSIS USING PC/FRAM
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I. INTRODUCTION

A. Purpose of This Chapter

It has been 20 years since Chapter 8, "Plutonium Isotopic Composition by Gamma-Ray Spectroscopy" in the *Passive Nondestructive Assay of Nuclear Materials* (Sampson 91) book was first drafted in 1983. This book, commonly referred to as the "PANDA Manual," is one of the principal resources for passive Nondestructive Assay (NDA) measurements throughout the worldwide nuclear community. Since 1986, when the material in Ch 8 was expanded and updated (Sampson 86), there has been an explosion of activity in the field of gamma-ray isotopic analysis. New codes and new analysis methods have been developed and applied worldwide to complement improvements in both detectors and data acquisition devices.

This chapter will describe the developments that have taken place at the Los Alamos National Laboratory concentrating on the PC/FRAM* isotopic analysis software. A review of the principles of gamma ray isotopic analysis will expand on the principles developed in PANDA to include the physics behind the current techniques used in FRAM. This chapter will cover all aspects of the FRAM software, including usage, development principles, algorithms, parameter files, performance, and measurement applications.

B. Isotopic Analysis Applications in Nondestructive Assay

1. Calorimetry.

A calorimeter (Likes 91a) determines the power produced by a sample of special nuclear material (SNM), the power arising primarily from the alpha decay of the isotopes making up the SNM. Elemental plutonium usually contains a mixture of isotopes with ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, and ^{241}Am present in most plutonium-bearing items. Each isotope produces a characteristic amount of heat proportional to its decay energy. This decay heat, when quantified per gram of isotope, is called the specific power, has the customary units of (W or mW)/gram isotope, and is denoted by P_i. The specific powers can be calculated from fundamental principles and can also be directly measured from pure isotopes. The accepted values for the specific powers of the plutonium isotopes and ^{241}Am are given in Table I-1.

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* FRAM is the name of the gamma-ray isotopic analysis software developed over the years in the Safeguards Science and Technology group, N-1 at the Los Alamos National Laboratory. FRAM is a word of Scandinavian origin meaning "forward" or "onward." In addition, it can be viewed as an acronym, <u>F</u>ixed energy, <u>R</u>esponse function <u>A</u>nalysis with <u>M</u>ultiple efficiencies, describing the general features of the code.

Table I-1.   Specific Power Values for the Isotopes of Plutonium (ANSI 1987)

Isotope	Half Life (yr)	Specific Power (mW/g isotope)	Standard Deviation (mW/g isotope)
^{238}Pu	87.74	567.57	0.26
^{239}Pu	24119	1.9288	0.0003
^{240}Pu	6564	7.0824	0.002
^{241}Pu	14.348	3.412	0.002
^{242}Pu	376300	0.1159	0.0003
^{241}Am	433.6	114.2	0.42

The sum of the specific powers of the individual heat-producing isotopes, weighted by their relative abundances, yields the effective specific power P_{eff} in units of mW/g Pu (Likes 91a).

P_{eff} is the important factor required to convert the measured Watts from the calorimeter to grams elemental plutonium. The mass of elemental plutonium (all the heat-producing isotopes) is given by

$$M = W/P_{eff} \quad , \tag{I-1}$$

where W is the measured Watts from the calorimeter and P_{eff} in units of Watts/g Pu is the effective specific power.

The measurement of P_{eff} is one of the most important applications of gamma-ray isotopic analysis. Calorimetric Assay, the combination of a calorimetric measurement of Watts and a gamma ray isotopic measurement of P_{eff}, provides the most accurate and precise method available for nondestructively determining the mass of elemental plutonium in bulk samples.   As a result, tabulating the accuracy and precision of the measurement of P_{eff} is one of the principal methods for characterizing isotopic analysis software. The characteristics and errors involved in the measurement of P_{eff} by FRAM will be discussed in detail later in this document.

2. Neutron Coincidence Counting.

The even isotopes of plutonium (^{238}Pu, ^{240}Pu, and ^{242}Pu) have large spontaneous fission yields that dominate the fission neutron output from plutonium. The spontaneous fission yields of the plutonium isotopes are shown in Table I-2 (Ensslin 91a). Spontaneous fission neutron emission in conjunction with sensitive neutron coincidence counting systems provides a widely used measurement technique (Ensslin 98, Reilly 91). All three of the even isotopes contribute to the response of a neutron coincidence counter with the contribution from ^{240}Pu dominating for most plutonium-bearing materials. For this reason it is customary to define the effective ^{240}Pu mass by

$$^{240}Pu_{eff} = 2.52 * {}^{238}Pu + {}^{240}Pu + 1.68 * {}^{242}Pu, \tag{I-2}$$

where ^{240}Pu$_{eff}$ is the mass of ^{240}Pu that would give the same coincidence response as that observed from the actual measured item.

We define the effective ^{240}Pu fraction in an analogous fashion by

$$\text{fract}^{240}Pu_{eff} = 2.52 * \text{fract}^{238}Pu + \text{fract}^{240}Pu + 1.68 * \text{fract}^{242}Pu \tag{I-3}$$

Table I-2.   Spontaneous Fission Neutron Yields From the Isotopes of Plutonium

Isotope	Spontaneous Fission Yield (n/s-g)
^{238}Pu	2.59×10^{3}
^{239}Pu	2.18×10^{-2}
^{240}Pu	1.02×10^{3}
^{241}Pu	5×10^{-2}
^{242}Pu	1.72×10^{3}

The isotopic fractions are obtained from either mass spectrometry or from nondestructive gamma-ray isotopic analysis. Combining the measured effective ^{240}Pu mass with the effective ^{240}Pu fraction yields the plutonium mass in a fashion completely analogous to calorimetry as

$$M \;=\; \text{grams } ^{240}\text{Pu}_{eff} \,/\, \text{fract}^{240}\text{Pu}_{eff} \tag{I-4}$$

where the grams ^{240}Pu$_{eff}$ comes from the neutron coincidence counting of the bulk item and fract^{240}Pu$_{eff}$ comes from the isotopic analysis. In addition to the isotopic information required in computing fract^{240}Pu$_{eff}$, coincidence counting requires knowledge of the complete isotopic distribution, including ^{241}Am, for computing (α,n) rates for multiplication corrections.

Characterizing the accuracy and precision of the measurement of fract^{240}Pu is important for characterizing the performance of isotopic analysis software.

3. Other Bulk Measurement Techniques.

The FRAM isotopic analysis software has been applied to essentially every bulk measurement problem that quantifies individual isotopes.

The Active Well Coincidence Counter (AWCC) is an instrument commonly used to assay ^{235}U (Menlove 91). Thus, for application to ^{235}U in the AWCC, we require knowledge of the ^{235}U isotopic fraction. The FRAM software was the first gamma-ray isotopic analysis code to demonstrate measurements on uranium.

The Segmented Gamma Scanner (SGS) uses transmission-corrected passive assay techniques (Parker 91) to quantify individual isotopes (usually ^{239}Pu or ^{235}U) in items of scrap and waste. FRAM is widely used to support this common measurement technique, as well as the Tomographic Gamma Scanner (TGS) especially for application to permanent waste disposal at the Waste Isolation Pilot Plant.

4. Process Control

There are numerous applications of gamma-ray isotopic analysis in providing information necessary for the control of various fabrication processes. Isotopic analysis may be required anytime material from two different batches is mixed to produce a product that must meet designated specifications.

One example might be that of blending materials with different ^{240}Pu fractions to meet a given "weapons grade" specification. Another application is that of blending plutonium from different batches to produce MOX fuel where the fissile isotope concentrations are of interest. The FRAM isotopic analysis software has the capability for verifying isotopic composition with accuracy requisite for performing these blending operations. Another capability of FRAM allows the quantification, relative to plutonium, of the concentration of fission products in the mixture or its components. This capability is also proving to be useful for rapidly and economically characterizing materials that must meet production specifications.

5. Treaty Verification.

Treaty verification is an application that did not exist when the PANDA chapter on isotopic analysis was written. Arms control and plutonium disposition negotiations between the United States (US) and the Russian Federation (RF) consider the disposition of plutonium from dismantled Russian nuclear weapons. The treaties and agreements arising from these negotiations contain requirements to verify the amount and isotopic composition of the plutonium declared as coming from dismantled weapons. A problem arises because in Russia the isotopic composition of the plutonium used in Russian weapons is classified. Nondestructive gamma-ray isotopic analysis techniques are applied behind an information barrier to verify this ratio without revealing the detailed isotopic composition.

The Russian Weapons Plutonium Conversion Line will take plutonium from dismantled weapons, mix it with fuel grade plutonium and produce PuO_2 for MOX fuel. Gamma-ray isotopic analysis provided by the FRAM software will be used to determine the proper mixing ratios and also to verify the isotopic composition of the output PuO_2 (Sampson 98).

II. BASIC PRINCIPLES OF GAMMA-RAY ISOTOPIC ANALYSIS FOR THE ARBITRARY SAMPLE

A. Gamma Ray Measurement of Isotopic Ratios

The development of the fundamental relation governing the measurement of isotopic ratios using gamma-ray spectrometry has been shown in previous publications (Sampson 91, Sampson 03). The well-known result is given in eq. II-1

$$\frac{N^i}{N^k} = \frac{C(E_j^i)}{C(E_l^k)} \times \frac{T_{1/2}^i}{T_{1/2}^k} \times \frac{BR_l^k}{BR_j^i} \times \frac{RE(E_l)}{RE(E_j)} \tag{II-1}$$

where

$C(E_j^i)$ = photopeak area of gamma ray j with energy E_j emitted from isotope i,

λ^i = decay constant of isotope i, $\lambda^i = \ln 2/T_{1/2}^i$ where $T_{1/2}^i$ is the half life of isotope i,

N^i = number of atoms of isotope i,

BR_j^i = branching ratio (gamma rays/disintegration) of gamma ray j from isotope i,

RE_j = relative efficiency for photopeak detection of gamma ray with energy E_j. This includes detector efficiency, sample self-absorption, and attenuation in packaging and materials between the sample and the detector.

The relative efficiency RE includes the effects of sample self-absorption, attenuation in materials between the sample and the detector, and detector efficiency. The half-lives, $T_{1/2}$ and the branching ratios, BR are known nuclear data. The $C(E)$ term is determined from the gamma ray spectral data, leaving only the ratio of the relative detection efficiencies to be determined. The need for only an efficiency ratio removes the problems associated with the geometric and sample reproducibility associated with absolute measurements and makes the method applicable to samples of arbitrary size, shape, and composition.

B. Ratio Measurements for the Arbitrary Sample—Without Efficiency Corrections

The earliest application of isotopic ratio measurements on plutonium made the assumption that the two gamma-ray peaks were close enough in energy that the differences in sample self-attenuation, absorption in packaging materials, and the detector efficiency could be neglected. The early applications of this technique at the Mound Laboratory recommended using gamma-ray pairs with energy spacing less than 10 keV.

Efficiency differences even with closely spaced peak pairs can be significant and were the cause of some of the biases observed with this early method.

C. The Intrinsic Self-Calibration Technique

In 1974 Jack Parker and Doug Reilly at Los Alamos proposed the first practical method for accurately measuring the isotopic composition of an arbitrary (size, shape, composition, measurement geometry) plutonium sample via analysis of its gamma-ray spectrum (Parker 74). 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.

Parker and Reilly noted that you could determine the ratio of the relative efficiency at the selected energies from the measured gamma-ray spectrum of the unknown sample. From eq. II-1, considering a series of gamma rays from a single isotope, we see that the quotient of the photopeak counts at energy E_j^i and the branching ratio BR_j^i is proportional to the efficiency at energy E_j.

$$\frac{C(E_j^i)}{BR_j^i} \propto \left[\frac{N^i \ln 2}{T_{1/2}^i} \right] \varepsilon(E_j) \tag{II-2}$$

Thus, this quotient defines the shape of the relative efficiency as a function of energy for the measurement in question. Gamma rays from several isotopes may be used to define the relative efficiency as long as all the isotopes used have the same physical distribution (an important restriction!). The curves from different isotopes with the same physical distribution have the same shape and differ only in their amplitude, the term in brackets in eq. II-2.

The development of Parker and Reilly forms the basis for most isotopic analysis applications that are in use today, including the FRAM code.

D. The Relative Efficiency Concept

The concept of the intrinsically determined self-calibration of the measurement's relative efficiency is the key feature of modern gamma ray isotopic analysis methods. Equation II-2 is used to determine the relative efficiency at the gamma-ray energies used in the peak pair ratio expression of eq. II-1.

The relative efficiency is viewed as a function of energy. Almost any variable that perturbs the absorption or relative intensity of gamma rays emitted from the sample can affect the shape or energy dependence of the relative efficiency curve. Some of these are

- The size, configuration, and efficiency of the HPGe detector.
- The mass of plutonium in the sample.
- The areal density of plutonium in the sample.
- The density and absorption properties of any matrix material.
- Material properties and thickness of the container(s).
- Absorbers between the sample and the detector.

Figure II-1 shows the overall effect seen as the sample gets larger which also usually means thicker (more mass and more areal density). The curves in Fig II-1 are normalized to unity at their maximum value. Because the mean free path increases as energy increases, an isotopic measurement will "see" farther into the item being measured and hence sample more volume and mass at high energy than at low energy. This means that relative to low energies, large samples emit more high-energy gamma rays than low energy gamma rays and the relative efficiency tends to increase with increasing energy more strongly than for small samples. This is illustrated numerically in Table II-1 for plutonium at low density approximating that of PuO_2. The "thickness" or areal density of the plutonium in a sample must be several mean free paths in magnitude to take full advantage of the intensity available at a specific measurement energy.
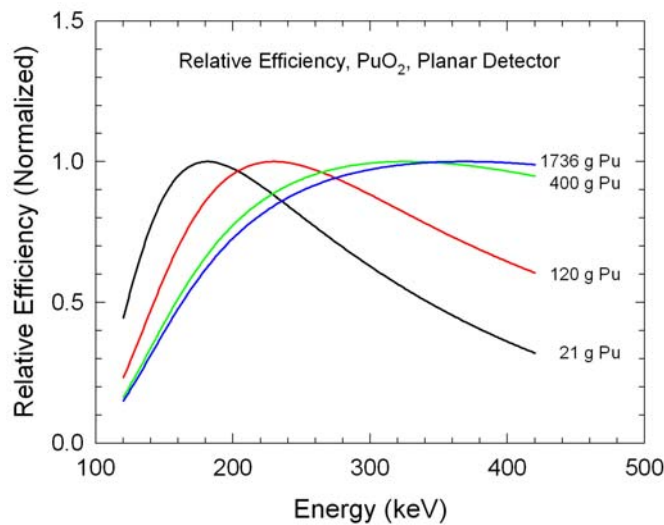
Fig. II-1. The relative efficiency varies for different size samples using the same 16 mm dia x 13 mm deep planar detector. The curves are normalized at their maximum value.

Table II-1. Mean Free Path for Various Gamma Rays in Plutonium of Density 3.0 g/cm^3

Pu-238	λ (cm)	Pu-239	λ (cm)	Pu-240	λ (cm)
152 keV	0.13	129 keV	0.083	104 keV	0.19
766 keV	3.0	414 keV	1.2	160 keV	0.15
				642 keV	2.4

Figure II-1 shows that the user can get qualitative information on the plutonium areal density in the sample just by examination of the shape of the relative efficiency curve.

E. Relative Efficiency Models

After relative efficiency values have been determined for a specific measurement using eq II-2, the user has a series of relative efficiency vs energy points that might appear to look like Fig. II-2. The user then needs to find relative efficiency values for energies not defined by a specific relative efficiency point and sometimes, even outside the range defined by the relative efficiency points. This requirement has led to the development of several models to parameterize the relative efficiency curve.
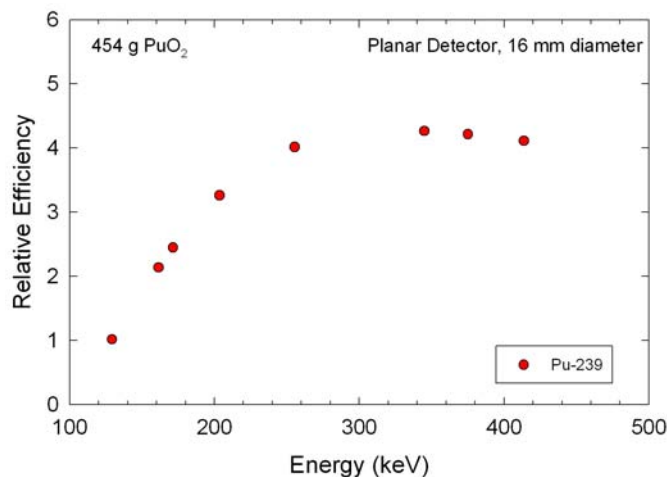

Fig. II-2. "Raw" relative efficiency points from ^{239}Pu for a specific measurement.

Several models are discussed in (Sampson 03) including simple models used in early Los Alamos codes. The models discussed below are more commonly used in modern isotopic analysis applications.

1. Empirical Model

Fleissner and Ruhter used linear least squares fitting of polynomial expressions in lnE in the early 1980s to parameterize the relative efficiency vs energy relationship. This represented the "state of-the-art" when the PANDA chapter on isotopic analysis was drafted.

Fleissner in his GRPAUT software (Fleissner 81a) used the form

$$\ln \varepsilon_i = a_0 + \sum_{j=1}^{2}\left(a_j E_i^{-j}\right) + \sum_{j=1}^{3}\left(a_{j+2}(\ln E_i)^j\right) + a_6 \delta_6 + a_7 \delta_7 \tag{II-3}$$

in the energy range from 120 keV to above 450 keV with points from ^{239}Pu, ^{241}Pu and ^{241}Am. The delta function terms, a_6 and a_7, normalize the ^{241}Pu and ^{241}Am data points to the ^{239}Pu data points. This form was expanded to include a second relative efficiency curve for cases where ^{241}Am was not isotopically homogeneous with plutonium.

Ruhter's form was similar and was used to fit from 120 keV to 210 keV if the spectrum was limited to that range or 120 keV to 380 keV if the data included the 375-keV region. His expression was simpler because it was used on a system with limited computing resources.

$$\ln \varepsilon_i = a_0 + a_1 \delta_1 + \sum_{j=1}^{2 \, or \, 3}\left(a_{j+1}(\ln E_i)^j\right) \tag{II-4}$$

The delta function term, a_1 normalizes the ^{241}Pu data points to the ^{239}Pu data points. Ruhter did not use any points from ^{241}Am to determine efficiency.

All versions of FRAM use or have available an empirical relative efficiency curve of essentially the same polynomial form (Sampson 89). FRAM's empirical relative efficiency is

$$\ln \varepsilon_i = C_1 + C_2 E_i^{-2} + \sum_{j=1}^{3}\left(C_{j+2}(\ln E_i)^j\right) + \sum_{k=1}^{m} C_{k+5}\delta_{k+5} + \sum_{l=1}^{n} C_{l+5+m}\delta_{l+5+m}E_i^{-1}, \tag{II-5}$$

where the k summation is the normalization for each isotope after the first and the l summation is the normalization for each additional relative efficiency curve. The FRAM empirical relative efficiency curve is not limited with regard to the number of isotopes (We use as many as 6), nor is it limited to the number of different relative efficiency curves applied for isotopic heterogeneity although our principal experience is still with only one additional relative efficiency curve for isotopic heterogeneity.

All applications of this empirical, polynomial-based relative efficiency curve work very well over the range of definition. This form can run into trouble if it extrapolated outside its range of definition or if the relative efficiency data is statistically poor.

2. Physical Model

A physics-based relative efficiency model has been widely used by Gunnink and co-workers at the Lawrence Livermore National Laboratory (Gunnink 90).

$$\varepsilon_j = \exp\left(-\mu_j^{Cd} * x_{Cd}\right) * \frac{1}{\mu_j^{Pu} * x_{Pu}}\left(1 - \exp\left(-\mu_j^{Pu} * x_{Pu}\right)\right) * eff_j * \left(1 + bE_j + cE_j^2\right), \qquad \text{II-6}$$

where

μ_j^{Cd} = mass absorption coefficient of cadmium for peak j,

μ_j^{Pu} = mass absorption coefficient of plutonium for peak j,

x_{Cd} = thickness (g/cm2) of cadmium absorber,

x_{Pu} = thickness (g/cm2 of plutonium in the sample,

eff_j = detector efficiency for peak j from a "generic" efficiency curve,

$b, c,$ = coefficients in a quadratic function to account for small deviations in the efficiency from the generic value as well as other slowly varying effects, such as absorption from low Z matrix materials.

This model explicitly accounts for self-absorption in the plutonium in the sample, absorption in a cadmium filter between the sample and detector, and the intrinsic detector efficiency. It has been used very successfully in the region from 59 keV to 300 keV with a planar detector. The variables are determined by iterative non-linear least squares techniques. Because the model is based on physical principles, it can give valid results outside its range of definition in cases where simpler models fail.

Vo at Los Alamos has implemented a very versatile physical model for relative efficiency that allows for multiple absorbers, multiple efficiency curves, and uses a wide-ranging correction factor for slowly varying effects.

$$\begin{aligned}
\varepsilon = &\frac{1}{\mu_{Pu} * x_{Pu}}\left(1 - \exp\left(-\mu_{Pu} * x_{Pu}\right)\right) \\
&* \exp\left(-\mu_{Cd} * x_{Cd}\right) * \exp\left(-\mu_{Fe} * x_{Fe}\right) * \exp\left(-\mu_{Pb} * x_{Pb}\right) \\
&* (I_i) * \exp\left(\frac{c_j}{E}\right) * (Det\ Eff) * (Correction\ Factor)
\end{aligned} \qquad \text{(II-7)}$$

The first term is self absorption in the plutonium; the second line is the absorption in up to three different materials (out of a choice of seven—Al, Fe, Cd, Er, Pb, H_2O, Concrete); I_i is the activity of isotope i; $\exp[c_j/E]$ accounts for isotopic heterogeneities (see next section); *Det Eff* is a generic detector efficiency parameterized in the software; and *Correction Factor* corrects for variations of the actual detector efficiency, nuclear material, and matrix from that specified in the model. The *Correction Factor* is a modified Hoerl function.

$$Correction\ Factor = \left(E^b * c^{\frac{1}{E}}\right) \qquad \text{(II-8)}$$

This physical efficiency function is available in FRAM version 4 (Kelley 02) and has been used in various applications in the energy range from below 40 keV to above 1500 keV.

3. Isotopic Heterogeneity

Equation II-1 is very general, but it contains the important assumption that all the measured isotopes in the sample are homogeneous with respect to each other. Another way of saying this is that gamma rays of the same energy from different isotopes must suffer the same attenuation as they escape from the sample. Failure of this assumption is called isotopic heterogeneity.

An example of isotopic heterogeneity occurs in pyrochemical plutonium processing applications. This process produces pure plutonium metal with Am and U removed. The waste Am and U that have been separated reside as a chloride salt along with small amounts of residual plutonium as metal fines in the residue stream. The proper quantification by calorimetric assay of the plutonium in this residue stream is complicated by the isotopic heterogeneity of the plutonium and americium present. The gamma rays from [241]Am suffer attenuation predominately in a low-Z chloride salt matrix while the plutonium gamma rays suffer attenuation characteristic of the high-Z plutonium metal fines. The relative-efficiency curve for gamma rays from [241]Am is different than that from plutonium. The isotopic ratio expression of eq. II-1 does not work in this instance.

Fleissner first proposed a second relative efficiency curve for [241]Am in pyrochemical residues—the [241]Am curve being related to the main plutonium relative efficiency by a multiplicative factor of $\exp[\beta/E]$, E being energy and β being a fitted constant . The Empirical (Eq. II-5) and Physical (Eq. II-7) relative efficiency formalisms contained in version 4 of FRAM both include a heterogeneity terms as proposed by Fleissner.

Testing of this heterogeneous model has involved comparison of isotopic measurements on heterogeneous pyrochemical residues with destructive chemical analysis of the entire item (Sampson 89). These destructive chemical analysis studies are very lengthy and extremely expensive so comparison data is limited. The most important parameter determined in the isotopic measurement is P_{eff} in mW/gPu. This is used directly to convert a calorimetry measurement of total sample power to grams elemental plutonium. With the heterogeneous relative-efficiency model of Fleissner, both Fleissner's GRPAUT code and the Los Alamos FRAM code determined P_{eff} with a bias that usually did not exceed 5%. Analysis of the same data without the heterogeneous relative-efficiency correction yielded biases from 10% to 200%.

III. PC/FRAM

A. Development

The first version of FRAM (Sampson 89) was fielded in 1988 at the Los Alamos Plutonium Facility running on Digital Equipment Corporation MicroVAX computers. The FRAM code represented a major advance in measurement flexibility as it was designed to address the shortcomings of the software described in PANDA and also included significant upgrades in the measurement and analysis hardware to the state of the art at that time.

By the early 1990s, computer hardware and software developments made the VAX/VMS-based 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 facilities that did not support the previous VAX system. This change has resulted in FRAM becoming commercially available through several vendors and now being used worldwide. Some of the major features of FRAM are described below. [Note: We use the names PC/FRAM and FRAM interchangeably throughout this document.]

B. Single Detector System

Like all previous Los Alamos isotopic analysis systems, PC/FRAM uses only a single detector to acquire its data. We made a conscious choice to keep PC/FRAM a single detector system because single detector systems are inherently

- Easier to use
- More versatile
- More reliable
- Less expensive
- Occupy less facility space

C. Choice of Detector Type

PC/FRAM is the only isotopic analysis software system that can obtain a complete isotopic analysis using either a single planar, a single coaxial HPGe detector, or a CdTe 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. PC/FRAM has been used with a single planar detector to measure uranium isotopic composition in the energy range from 120–1024 keV. Recent developments fielded in version 4 of PC/FRAM now allow the analysis of planar detector spectra in the 100-keV region and the 40-keV region (Vo 01a).

The most widely used mode of operation with a single coaxial detector is to acquire a spectrum from 0–1024 keV. Various analysis modes can then be used. If the region between 120 and 200 keV is available, PC/FRAM will work best analyzing 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. We have also found that the optimum analysis of coaxial detector data from some samples may come from the 200–800 keV region even when the region between 120 and 200 keV is available.

The optimum choice of planar or coaxial detectors is made only after considering all possible measurement applications. The planar detector is usually chosen 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 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.

With a CdTe detector, one collects data in the 125-414 keV range, just like a planar HPGe.

D. Shielded Samples

Most isotopic analysis codes (including the original Vax-based FRAM) require the presence of spectral peaks in the region below 200 keV, regardless of whether they use one or two detectors. When this region is not available, perhaps because the sample is shielded to lower radiation exposure or because it is inside a very heavy-walled container, some isotopic analysis codes may not function. PC/FRAM 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 mm of lead or about 10 mm of steel. FRAM measurements have been made through as much as 25 mm of lead and very easily through 25 mm of steel.

E. Uranium Isotopic Analysis

Up until 1990 the isotopic analysis techniques originally proposed by Parker and Reilly were applied only to plutonium. There was always the need for uranium isotopic analysis but the features of the uranium gamma-ray spectrum precluded the easy application of the "peak pair" ratio method used in early isotopic analysis applications.

The uranium gamma-ray spectrum is essentially divided into two regions. The low-energy region up to about 200 keV contains only gamma rays from ^{235}U with the major gamma rays at 143.76, 163.33, 185.72, 202.11, and 205.31 keV, all from ^{235}U. The sole gamma ray from ^{234}U above 100 keV is at 120.90 keV and ^{236}U has no measurable gamma rays. The intense ^{238}U gamma rays arise from its ^{234m}Pa daughter with energies of 742.81, 766.36, 786.27, and 1001.03 keV. The wide separation between ^{235}U and ^{238}U gamma rays stymied the application of the early arbitrary-sample isotopic analysis techniques. The weak ^{234m}Pa gamma ray at 258.26 keV plays an important role in current applications of FRAM to uranium.

The formalism of FRAM does not require closely spaced peak pairs. Thus, in the late 1980s, we applied the original VAX version of FRAM to analyze uranium. This required a coaxial detector and data analysis in the 120-1200 keV region. We demonstrated FRAM's ability to measure, *with no code modification,* the $^{238}U/^{235}U$ ratio in samples of arbitrary physical and chemical composition, geometry, and mass, containing only uranium.

In PC/FRAM, uranium analysis was expanded to include ^{234}U and in the latest version we include a correlation to predict ^{236}U and a correction for cases where ^{234m}Pa is not in equilibrium.

F. Version 4

Sampson (Sampson 03) describes the features and improvements in all of the released versions of FRAM. The features and upgrades in the most recent release, version 4, mainly concern enhancements to the physics algorithms, new measurement capabilities, and a new structure to make derivative applications easier to implement.

Version 4.2

- **Relative Efficiency**. This version incorporates a new physical model for calculating the relative efficiency curve.
- **Analysis Engine**. The analysis algorithms have been placed in their own library. This makes it easier for other users to adapt FRAM for their own applications.
- **New Menus for Uranium Analysis**. There are separate dialog boxes for measuring plutonium and uranium and for analyzing Pu and U data files.
- **Intelligence**. There is a selectable capability of automatically switching, in a limited way, from one parameter set to another depending on analysis results.
- **Uranium Analysis Enhancements**. Enhancements for uranium analysis include 1) correction for ^{234}Th nonequilibrium, 2) isotopic correlation to predict ^{236}U, and 3) corrections for coincidence summing effects.
- **CdTe**. FRAM can analyze spectra taken with a CdTe detector in the 120-414 keV energy range (Vo 02).
- **100-keV Region Analysis**. FRAM can analyze plutonium using the 100-keV region (Vo 01a).
- **40-keV Region Analysis.** FRAM can analyze freshly separated plutonium using the 40-keV region (Vo 01a).

IV. HOW FRAM WORKS

A. Obtain Data

There are two basic types of data sources for FRAM.

1) "Live" data from a multichannel analyzer (MCA) acquiring a gamma-ray spectrum from a high-resolution detector (usually HPGe). FRAM can control the data acquisition from several commercial MCA families. The ORTEC line of Multichannel Buffers (MCB) operating with the Maestro MCA emulator can all be controlled via FRAM. Canberra MCAs operating under Genie 2000 can also be controlled from FRAM. Control is limited to the basic functions of count time, start, stop, and readout to a disk file. The user must invoke the appropriate MCA emulator other functions (high voltage, amplifier gain, etc.). The analysis of "live" data acquired under FRAM control proceeds automatically after the acquisition terminates.

2) Data from a disk file. FRAM can read and analyze data from disk files recorded in several data formats. These data formats include, for version 4, the following formats:

- N-1 standard
- Canberra S100
- Ortec 'spc'
- Ortec 'chn'
- Canberra CAM
- IAEA MCRS
- IAEA MMCA
- Green Star
- ASCII

If FRAM is purchased from a licensee it will contain only the control and data formats appropriate to the vendor's own products or that are publicly available. In a version of FRAM from Los Alamos, the user will have access to everything that was current at the version release date. The user will have to purchase and install the appropriate MCA emulator to control the setup of the MCA and make full use of the commercial formats.

B. Perform Analysis

The analysis of a gamma-ray pulse-height spectrum by the PC/FRAM code proceeds in two steps, 1) the internal calibration, and 2) the analysis of the spectral data.

1. Internal Calibrations

The internal calibration uses peaks in the spectrum under analysis to provide a calibration of energy vs. channel, full width at half maximum (FWHM) vs. channel, and peak shape (tailing parameters) vs. channel. These calibrations do not depend on parameters determined from other measurements that may have been taken with different conditions of count rate, resolution, or electronic adjustment. In some cases, there are insufficient peaks to use the unknown spectrum for its own calibrations. In these cases, one can fix the peak calibration parameters to their initial values in the parameter set.

a. Energy Calibration

The first portion of the internal procedure calibrates energy vs. channel number from a list of calibration peaks in the parameter set. A piecewise linear calibration is made between successive pairs of peaks. The algorithm locates the peak at the maximum count found in a region of 10 channels on either side of the default peak position located using the default gain and zero values from the parameter set. FRAM is not constrained to any particular energy calibration. Within the constraints of spectral quality, FRAM can analyze spectra at any gain given that the energy calibration is known well enough to find the calibration peaks within a ± 10 channel window. The peak centroid is found using a least-squares fit of a quadratic function to the logarithm of the counts. Calibration outside the range of the energy calibration list is linearly extrapolated from the nearest two points.

b. Initial Background

Next, a background is calculated for all peak regions in the parameter set. The calculation uses the background functional shape for each region that is specified in the parameter set.

c. FWHM Calibration

The parameter set contains a user-editable list of peaks for use in the internal calibration of FWHM vs. energy. The FWHM of each peak in the list is calculated after a channel-by-channel subtraction of the initial background. The FWHM is calculated from a least-squares fit of a quadratic to the logarithm of the net counts over a range of channels in which the counts exceed

75% of the peak maximum on the low-energy side and 25% of the peak maximum on the high-energy side (for CdTe, because of the larger tails, the fit starts from 85% on the low-energy side). The FWHM as a function of energy that is used in calculating the response function for an arbitrary fitted peak is found from a least squares fit to the function:

$$FWHM(E) = SQRT\left[A_1 + (A_2 * E) + \left(A_3 / E\right)\right] \qquad \text{(IV-1)}$$

The first two terms are physics-based while the third term accounts for the observation that the FWHM for some detectors tends to "level out" at low energies.

d. Peak Shape/Tailing Calibration

The gamma-ray peak shape is described by a central Gaussian component with a single exponential tail on the low-energy side of the peak.

$$Y(J) = Ht * \exp\left[\alpha * (J - x_0)^2\right] + Tail(J), \qquad \text{(IV-2)}$$

where

$Y(J)$ = Net counts in channel J,
Ht = Peak height at the peak centroid x_0,
α = $2.77259/FWHM^2$ is the peak width parameter,

and the tailing parameter Tail(J) is given by

$$Tail(J) = Ht * \exp\left[(T1 + T2 * E) + (T3 + T4 * E) * (J - x_0)\right] * \left[1 - \exp\left(-0.4 * \alpha * (J - x_0)^2\right)\right]. \quad \text{(IV-3)}$$

Both the amplitude and slope of the tailing function are permitted to be a function of energy. However, in practice, we set T4 to zero reducing the number of unknowns to three. After subtracting the Gaussian portion of the peak (known because we have calibrations for energy and FWHM), we combine the data from all the FWHM peaks using the net channel contents on the low energy side of the peak from 0.5 to 1.5 FWHM from the peak center to determine the slope and amplitude constants from a least-squares fit.

This completes the internal calibration giving all the parameters necessary to calculate the shape of a gamma-ray peak at any location in the spectrum.

2. Analysis of Spectral Data

After the internal calibration is complete, the analysis proceeds on a region-by-region basis in the order that the regions are entered in the parameter set. The program makes three iterations through all the regions. A very detailed description of the analysis procedure may be found in (Kelley 02, Sampson 03).

The analysis starts by subtracting the initial background to get the net counts in a region. The background for the first iteration was calculated during the internal calibration phase. The analysis is iterative because of the interdependence of the peak areas and relative efficiencies calculated in separate steps.

a. Calculate Peak Areas Using Response Functions

For each of the regions defined in the parameter set, FRAM calculates the peak areas using response functions. The procedure allows peak areas to be fixed to peaks within or outside of the region. The results obtained after the final iteration consist of peak areas and uncertainties for all of the peaks in each region.

b. Calculate Relative Efficiencies

Relative efficiencies are calculated for all of the designated relative efficiency peaks in the parameter set. The individual relative efficiency points are fit via least squares to either the empirical relative efficiency function (Eq. II-5) or the physical relative efficiency function (Eq. II-7). The empirical function is fit by linear least squares methods while the physical relative efficiency function is determined by non-linear least squares using the Levenberg-Marquardt method.

c. Calculate Relative Activities

The relative activities are calculated for each peak by summing over all of the isotopes that contribute to the peak. Some of the peaks from the decay of the ^{237}U daughter of ^{241}Pu also contain an unresolved (exact energy match) gamma ray from ^{241}Am. The model is:

$$Area_i = \sum_j A_j (BR_{ij})(RE_i), \qquad (IV\text{-}4)$$

where,

A_j is the activity ratio for the jth isotope,
BR_{ij} is the branching ratio for the gamma ray emitted by the jth isotope contributing to
    the area of the ith photopeak
RE_i is the relative efficiency at the energy of the ith peak

The relative activities, the half lives, and atomic masses of the isotopes are then combined to yield the relative masses for each isotope.

d. Calculate Isotopic Fractions

After the third iteration is complete, the final relative masses (relative to the first isotope in the isotope list) are combined to give the absolute isotopic fractions without ^{242}Pu (^{236}U). The fractions are renormalized accounting for ^{242}Pu (^{236}U) computed by correlation or fixed by operator entry. Non-plutonium (uranium) isotopes are quantified relative to total plutonium (uranium). For samples containing no plutonium or uranium, the final results are the relative masses themselves. Auxiliary results such as the effective specific power and effective ^{240}Pu fraction are computed from the plutonium isotopic fractions and the appropriate constants in the parameter set.

e. Calculate Isotopic Correlation for ^{242}Pu and ^{236}U

Plutonium-242 and ^{236}U cannot be measured directly with gamma-ray spectroscopy techniques. It is customary to introduce an empirical isotopic correlation (Gunnink 90, Bignan 95) to predict their concentrations from the measured ratios for the other isotopes.

FRAM predicts ^{242}Pu from:

$$^{242}Pu = A \times \left[\left(^{238}Pu\right)^B \times \left(^{239}Pu\right)^C \times \left(^{240}Pu\right)^D \times \left(^{241}Pu + ^{241}Am\right)^E \right] \qquad (IV\text{-}4)$$

where the five constants, A–E, are user-editable values in the parameter file.

In a similar manner, Vo has developed a correlation to predict ^{236}U in uranium-bearing samples. It is of the form

$$^{236}U = A \times \left[\left(^{235}U\right)^B \times \left(^{238}U\right)^C \right] \qquad (IV\text{-}5)$$

The constants in eq. IV-5 have been determined from mass spectrometry values for US uranium produced by the gaseous diffusion process.

V. PARAMETER FILES, THE KEY TO FRAM'S VERSATILITY

The FRAM code has been structured to give the user as much control as desired over the analysis to increase versatility and applicability. This is accomplished by using Parameter Files (or Parameter Sets). A Parameter File contains the all the parameters required to analyze a gamma-ray spectrum. This includes information on the isotopes to be analyzed, the gamma-ray peaks to use, the nuclear data for the isotopes and gamma-rays, data acquisition conditions such as gain, zero, number of channels, spectral regions to analyze, and diagnostic test parameters.

These parameters reside in a custom-designed database within FRAM. This database can accommodate multiple parameter sets. The Change Parameter Utility, accessed from FRAM, gives the user access to all sets. The utility allows the user to add a new parameter set, delete a set, or modify the values in any set. The utility also allows the user to export the information in a parameter set to a text file on disk and subsequently to import this information back into the database allowing different systems to share parameter sets. Formally, "parameter set" refers to the information residing in the database in computer memory. "Parameter file" refers to the information in a text file residing on a disk.

FRAM is delivered with a variety of parameter files suitable for nearly all routine analyses. These parameter files usually do not need editing or changes to use FRAM for the first time. Routine FRAM analyses can be started with as few as three mouse clicks.

VI. FRAM USER INTERFACE

The FRAM user interface for version 4.2 provides a wide range of options governing data acquisition, data analysis, data display, results output, and parameter set manipulation and editing. We will briefly illustrate the extensive capability available to the user of FRAM.

The main menu of FRAM appears with four major options, File, Edit, Measure, and Options.
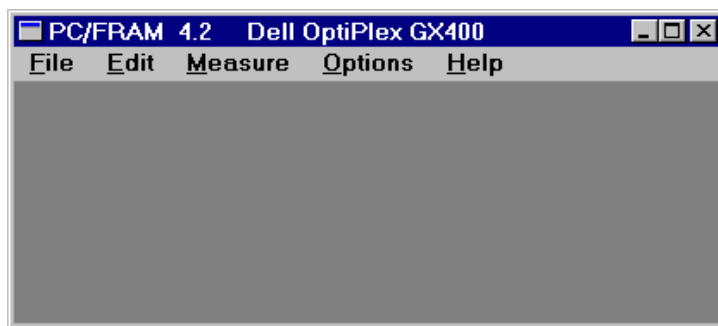
A. File

The File option allows the user to open a spectral data file where it can be viewed under Options. The file can also be saved in any of the supported data formats. Saving the file in the ASCII text format makes it easy to plot the data in most graphical applications.

B. Edit

The Edit menu has three groups of options. The first group allows password-protected access to the Change Parameter Utility. It also contains General Defaults allowing the user to set up

global parameters that govern the data handling in the system such as default paths for accessing FRAM, storing the spectral data and results files, and access to supported MCAs.

The second group of parameters under the Edit menu allows the user to set up default entries in many of the application windows. With the use of these defaults, the user is able to start a measurement with only three clicks of the mouse.

The third group is the password-protected User List. The User List controls access at three levels of password protection to all of the password-protected options.

C. Measure

The Measure menu governs the acquisition and analysis of data from both "live" MCA sources and disk files.
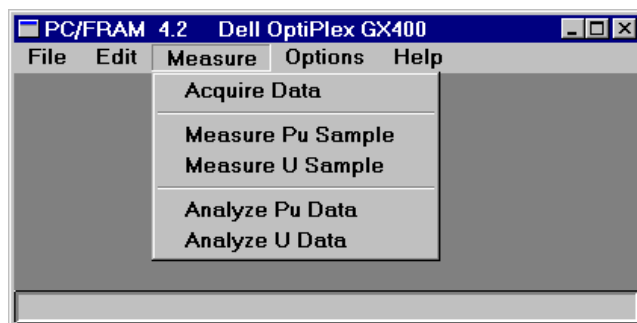
The Acquire Data option controls acquisition of data from a supported MCA and the storage of the data (without analysis) in a disk file. The Measure Pu (U) Sample options follow the data acquisition with an immediate analysis of the acquired spectrum. Analyze Pu (U) Data analyzes the spectral data from an existing disk file. The Measure Pu Sample window appears below as an example.
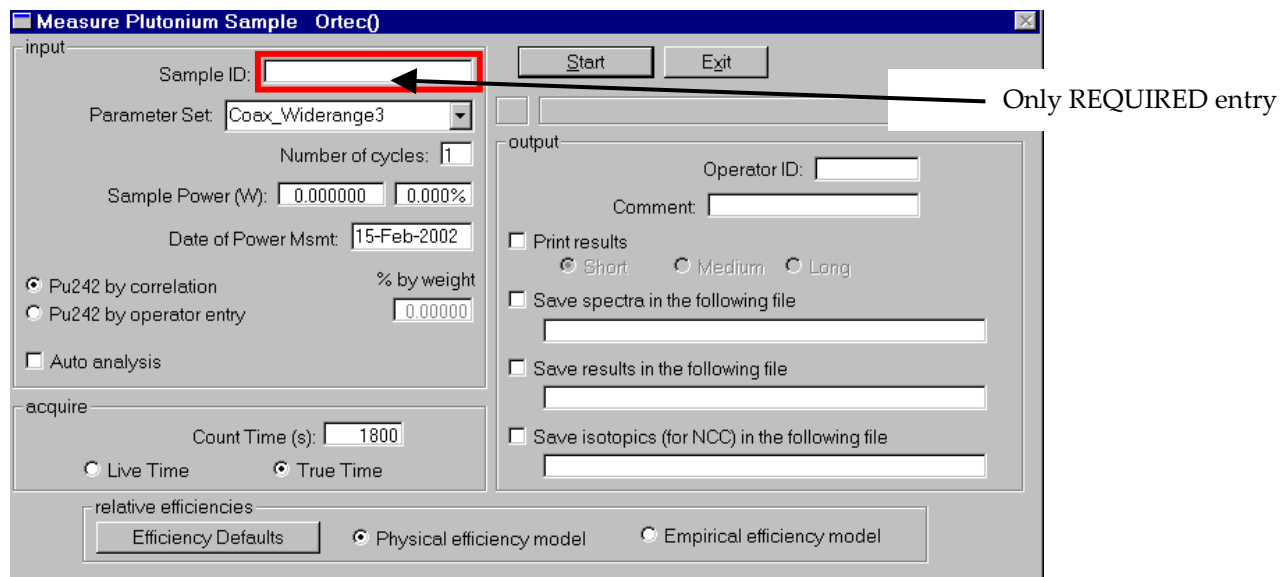
The window above has some of the entries defaulted from the Edit | Measure Pu Sample Defaults option. At this point the only entry *required* to start the measurement and complete the analysis is the Sample ID. With the use of defaults successive measurements may be completed with only identification of the sample and clicking the Start button. In many cases the user will also want to utilize some of the optional output options. To facilitate this, the Sample ID is defaulted as the filename for data storage.
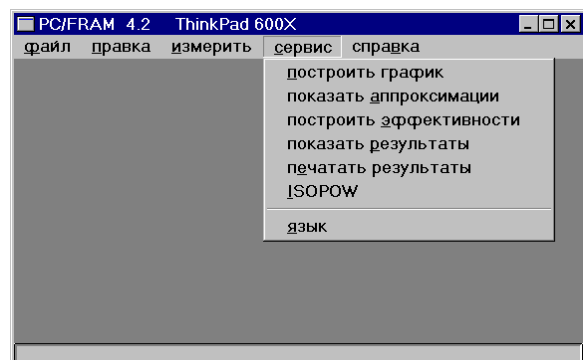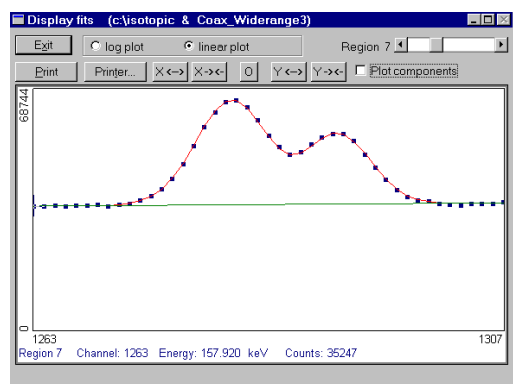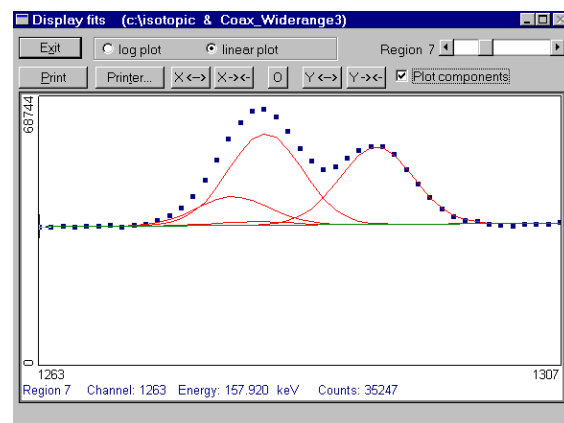
D. Options

The Options menu allows the user several ways to display and view the spectral data and the results of the analysis. The entire spectrum can be displayed and manipulated from the Plot Spectrum option with versatility similar to that of a commercial MCA emulator. The user can also display the peak fits and view the relative efficiency curve (below). These options are
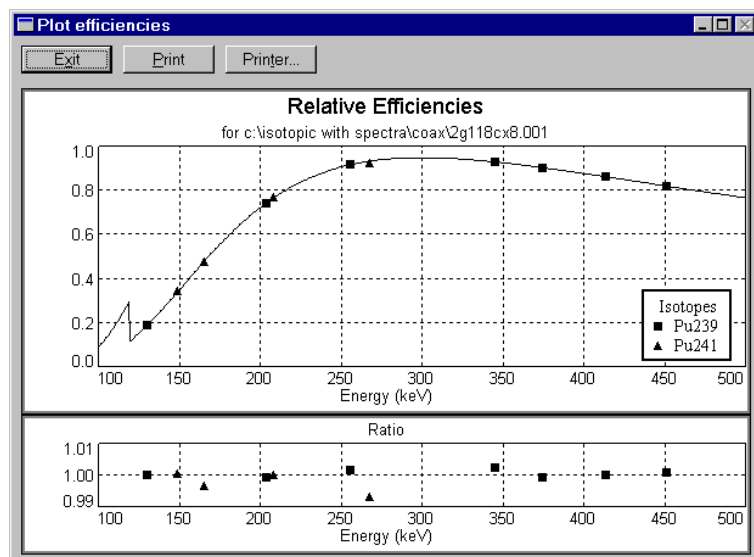
invaluable when troubleshooting a suspect analysis. The results of selecting Plot Efficiencies and Display Fits are shown below with fits being displayed showing only the fit envelope (left) or with the individual components (right).

The ISOPOW option gives access to a plutonium and americium decay correction program (Sampson 86a) that can be used on line or off line.

The option Language under the Options menu allows the user to display the operator interface, FRAM program messages, and results in a language other than English. The language strings are kept in a text file allowing any European language to be used as the second language merely by editing the second language text file. The current second language used with

FRAM is Russian. When Russian is selected the Main menu with "Options" pulled down looks like the screen to the left. The user can switch between the two languages with a single mouse click.

VII. FRAM PERFORMANCE

A. Measurement Precision or Repeatability

1. Definitions

In this section, we describe the many interrelated factors that govern the statistical precision of an isotopic measurement. In this context, precision or repeatability refer to the variability arising from counting statistics and are usually denoted by the relative standard deviation (RSD) in per cent.

$$\%RSD = 100 \times \frac{sigma}{measured\ value}. \tag{VII-1}$$

Here *sigma* is the absolute standard deviation of the *measured value*.

Sigma can be determined in at least two ways. First, the sigma from counting statistics is estimated within FRAM using standard error propagation techniques. This is difficult given mathematical analysis involved, the presence of correlated variables, and the wide range of the magnitude of the measured values. However, this method gives an estimate of sigma for every measurement and is invaluable when one has only a single measurement. The second method uses repeated measurements. From n repeated measurements of the variable x, we determine s, the standard deviation of the sample and use it as an estimate of the standard deviation of the population.

$$\left(s^2\right) = \frac{1}{n-1} \sum_{1}^{n} (x_i - \bar{x})^2 \tag{VII-2}$$

This expression is useful when comparing the estimated standard deviation with the standard deviation (eq VII-2) observed from repeated measurements. One has to perform many repeated measurements to verify propagated error estimates accurately as Table VI-1 illustrates.

Sigma estimated in this fashion is also a random variable. That is, if the series of n measurements of x is repeated, s will be different. The mean value of s will be the population sigma. The relative standard deviation of s values is given by the formula

$$RSD(s) = \frac{1}{\sqrt{2(n-1)}} \tag{VII-3}$$

Table VII-1   The RSD of Sigma (Error of the Error)

No. of Measurements	RSD of Sigma
10	0.235
15	0.189
25	0.144
50	0.101

2. Influencing Factors

In this section we will discuss some of the many, often interrelated factors, which influence the precision or repeatability of the isotopic measurement.

a. Count Rate and Throughput

The net counts in the photopeaks of the analyzed spectrum are the primary factors determining the measurement precision. The count rate directly influences the net photopeak counts. While the count rate in the detector is the parameter that is often observed, it is the actual data storage rate in the MCA that is of direct importance; this depends upon electronics settings including shaping time and the use of pulse-pileup rejection. Measurement systems are usually optimized by simultaneously measuring the throughput and resolution vs. incoming count rate and choosing the compromise settings best for the application at hand. An excellent discussion of these compromises may be found in (Parker 91a).

Throughput curves show a maximum throughput rate beyond which throughput decreases and counting precision worsens. The best compromise for throughput and resolution is usually chosen to be at a counting rate significantly below the throughput maximum. Operating at input counting rates that are 50-60 % of the count rate at the throughput maximum usually yields throughput values that 80-90 % of maximum while simultaneously preserving detector resolution.

Figure VII-1 shows the throughput and resolution measured with a 25% relative efficiency coaxial HPGe detector coupled with first generation digital signal processing electronics operated with a rise time of 4µs (equivalent to a 2µs shaping time in an analog amplifier). Here the throughput maximum occurs at an input rate of 60 kcps but we usually choose to operate at a maximum input rate of around 40 kcps where the resolution is better.

Optimizations performed in this manner affect the primary results of an isotopic analysis measurement. Figure VII-2 shows how the precision of the FRAM result for ^{240}Pu and P_{eff} varies for the same data set presented in Figure VII-1. Collection of spectral data at an input rate of 40 kcps gives essentially the same precision as operating at the 60 kcps peak of the throughput curve. The precision does not change very rapidly in a broad range about the throughput maximum, but it does worsen significantly at low count rates. In the range where the throughput curve is linear at rates below 15 kcps, the precision varies with the square root of the number of counts.
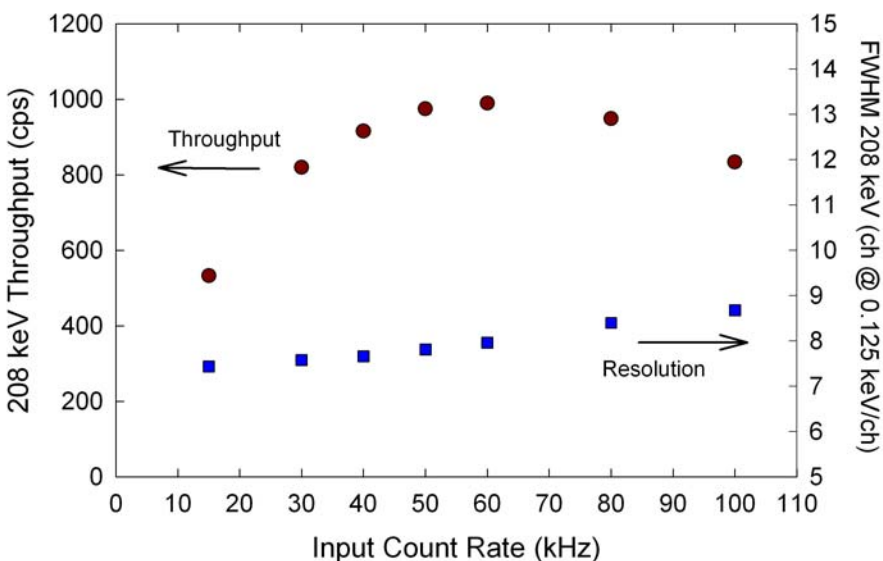

Fig. VII-1. Throughput and resolution for 208 keV peak of ^{241}Pu-^{237}U from 965 g PuO₂ with 16.85 % ^{240}Pu, and a 25%-relative-efficiency HPGe detector.
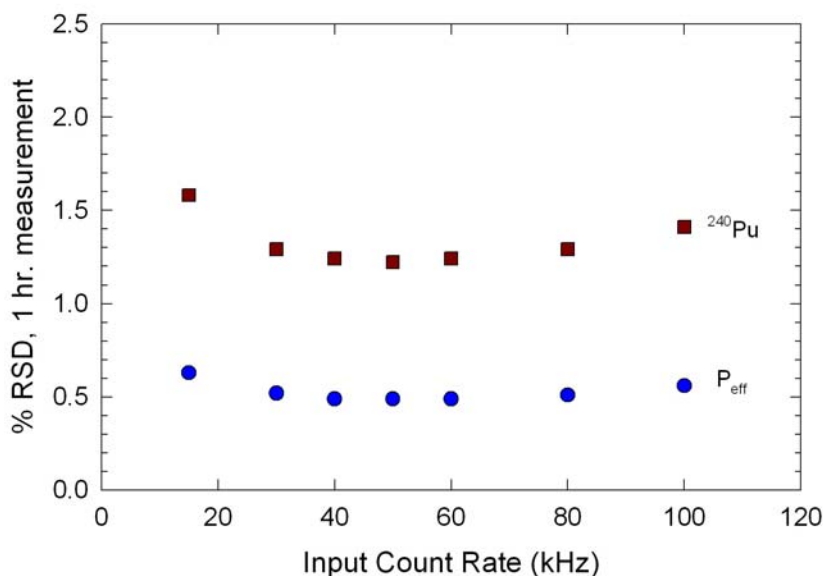
Fig. VII-2. % RSD of FRAM measurement of ^{240}Pu and P$_{eff}$ for 965 g PuO$_2$ with 16.85 % ^{240}Pu, a 25%-relative-efficiency HPGe detector, an ORTEC DSpec operated at 4-μs rise time with analysis in the 120-450 keV region. Count time was 1 hour.

b. Electronic Settings

The amplifier shaping time is the single most influential electronic setting affecting system performance. The shaping time-resolution tradeoff is well known and is discussed in detail by Parker (Parker 91a). The term "rise time" is usually used in characterizing digital spectroscopy systems with the rise time being about three times the analog shaping time. Throughput generally varies inversely with shaping time. A shorter shaping time improves throughput.

Longer shaping times usually correspond to better resolution and lower throughput, although for any specific system, the user will find that the resolution-shaping time curve goes through a broad minimum that is dependent upon the type of detector being characterized. One always operates on the low or shorter shaping time side of this minimum, giving up a little resolution in order to improve throughput. Fortunately the minimum is broad and resolution does not suffer too much.

For the small coaxial detectors (25-30 % relative efficiency) often used with FRAM, we obtain good results with analog systems using 2-μs Gaussian or triangular shaping. This corresponds to a rise time of 4 μs for digital systems. With these settings, good resolution is obtained at suggested maximum count rates of approximately 30 kcps (analog) and 40 kcps (digital).

For the planar detectors most often used with FRAM (16-25 mm dia. by 13-15 mm deep), we recommend a 1-μs triangular shaping with an analog system or a 2-μs rise time with a digital system.

Since the first Los Alamos isotopic systems in the early 1980s, improvements in amplifiers and pulse processing methods have led to improvements of about a factor of 3 in the precision for ^{240}Pu or P$_{eff}$ for measurements with the same count time.

c. Count Time

Poisson counting statistics are an appropriate model to represent the influence of counting time on the precision of isotopic analysis measurements. That is, the % RSD of a measured isotopic fraction varies inversely with the square root of the counting time, T.

$$\%RSD \approx \frac{1}{\sqrt{T}} \tag{VII-4}$$

Increasing the counting time by a factor of 2 improves the % RSD by a factor of 1.4.

d. Energy Range

The energy range used in the analysis is often the single largest factor in determining the precision or repeatability of an isotopic measurement. The foremost factor here is the intrinsic intensity of the gamma rays used in the analysis. Table VII-2 displays the intensities for the principal gamma rays from each of four energy regions that have been used historically. The intensities of the principal gamma rays from the isotopes [238]Pu and [240]Pu drop by roughly an order of magnitude with each successively higher energy region. This means that the best precision measurements, at least for the important [240]Pu and [238]Pu isotopes, come from the lowest energy regions.

The 40–60 keV energy region is used only in the special case of freshly reprocessed ([241]Am and [237]U removed) plutonium-bearing solutions, mainly in reprocessing plants. This region is not used widely for isotopic analysis because the Compton continuum from the 59.5-keV [241]Am peak swamps the plutonium peaks in the 40-keV region for aged materials and these low energy gamma rays are easily absorbed by many types of containers.

Table VII-2 Intrinsic Gamma-Ray Intensities of Major Gamma Rays in Principal Energy Regions

Region	^{238}Pu		^{239}Pu		^{240}Pu		^{241}Pu-^{237}U(*)		^{241}Am	
(keV)	(keV)	γ/s/g	(keV)	γ/s/g	(keV)	γ/s/g	(keV)	γ/s/g	(keV)	γ/s/g
40–60	43.5	2.5 e8	51.6	6.2 e5	45.2	3.8 e6			59.5	4.5
90–	99.9	4.6 e7	98.8	2.8 e4	104.2	5.9 e5	103.7	3.9 e6	98.9	2.6 e7
									103.0	2.5 e7
120–	152.7	6.1 e6	129.3	1.4 e5	160.3	3.4 e4	148.6	7.2 e6	125.3	5.2 e6
			375.0	3.6 e4			*208.0	2.0 e7	335.4	6.3 e5
			413.7	3.4 e4			*332.4	1.1 e6		
450–	766.4	1.4 e5	646.0	3.4 e2	642.5	1.0 e3			662.4	4.6 e5
									722.0	2.5 e5

The 90–105 keV region has been widely used for isotopic analysis and often offers the best precision for the measurement of [240]Pu. Strong attenuation of these low energy gamma rays does preclude their use for samples in thick-walled or shielded containers. An absorber of 10 mm of steel is usually enough to stop measurements using this region.

The 120–450 keV region is the most versatile region for plutonium isotopic analysis and is historically the region used at Los Alamos. Measurement precision for [240]Pu in the 120–450 keV region is usually poorer than in the 100-keV region. The advantage of this region is that measurements can easily be performed through as much as 12 mm of steel and even 0.3 mm of lead.

FRAM was the first code to carry out a complete Pu isotopic analysis using the 200–800 keV region. This capability allows measurements on samples in heavy-walled containers or in containers with internal shielding that prevents gamma rays below 200 keV from reaching the detector. Variants of this allow a complete plutonium isotopic analysis through shielding up to 25 mm of lead (Hypes 00).

The ability to analyze the 200-800 keV region for the complete isotopic distribution gives FRAM several more options for the analysis of large samples. The inherent advantage in precision of the 100-keV energy region for [240]Pu and [238]Pu analysis is reduced for large samples analyzed at high energy because of the increased penetrability of gamma rays in the 600-800

keV range relative to the 100-keV gamma rays. In effect, the high-energy gamma rays sample a larger volume of the sample. This increased penetrability is apparent principally for items with larger areal plutonium densities. Table VII-3 shows the parameters that affect penetrability for the three ^{240}Pu gamma rays at 104, 160, and 642 keV. This is the effect causing the relative efficiency curves for large items to increase in magnitude as one moves to higher energies (see Fig. II-1).

Table VII-3   Absorption Properties for PuO$_2$ at ρ = 3g/cm^3 for Gamma Rays from ^{240}Pu

Energy (keV)	μ/ρ (cm^2/g)	μ (cm^{-1})	Mean Free Path (cm)
104	1.62	4.9	0.21
160	2.01	6.0	0.17
642	0.131	0.39	2.5

The self-absorption at 160 keV is actually greater than at 104 keV because the plutonium K edge falls at 121.8 keV, between the two energies. The mean free path (mfp) at 642 keV is 15x greater than at 160 keV. Samples with a thickness greater than three mfp at 642 keV will have an emission rate increase at 642 keV over that at 160 keV that nearly compensates for the intrinsic intensity(γ/s/g) difference (Table VII-2) at the two energies. When this is coupled with the lower background continuum present at higher energies, one often obtains better precision for measurement of ^{240}Pu at 642 keV than at 160 keV.

Another consideration for using the 200–800 keV range for larger samples is that one is often able to improve the precision still further by filtering the detector with additional lead. The additional lead removes gamma rays below 200 keV that do not contribute to the 200–800 keV analysis. The sample counts at a constant input counting rate are shifted preferentially to higher energies and a greater fraction of the analyzed gamma rays will fall in the 600-keV region. Measurement precision improves as the analysis moves into the 200–800 keV range and improves further when the spectrum is filtered. Pu-241 does not improve in every case because its analysis is carried out at the lowest energies (200-340 keV) of the analysis range.

e. Detector Type
The influence of the detector type is directly related to the energy range used in the analysis. Two types of HPGe detectors have been most often used for isotopic analysis with FRAM.
Planar Detectors   Planar detectors of dimensions 16-mm dia by 13-mm thick and 25-mm dia by 15-mm thick are commonly used for FRAM measurements. These detectors have historically been used in the 120-420 keV energy range and, with version 4, can now be used in the 100-keV region. For most measurements on samples in thin containers analysis in the 100-keV region will provide better precision for all isotopes than the 120–420 keV region. For samples in containers with steel thicknesses in the range 5-10 mm, the optimum analysis region becomes less clear. Above a wall thickness of about 10-mm steel, the 100-keV region analysis fails leaving the 120-420 keV region as the only viable option.
Coaxial Detectors   Coaxial detectors of 25-30% relative efficiency (relative to a 7.6-cm-dia x 7.6-cm-thick NaI(Tl) detector for ^{60}Co at a distance of 25 cm) have been often used with FRAM. Coaxial detector measurement precision depends upon the energy range and shielding as discussed previously. It is not always possible to predetermine which detector, planar or coaxial, will have the better precision in the 120-420 keV region. Suffice it to say that the measurement precision in this energy range is often similar for the two detector types and the choice is often made empirically with measurements under realistic conditions. When samples are shielded,

planar detectors will not be viable and the coaxial detector choice often comes down to whether or not to use additional filters.

    <u>CdTe Detectors</u>   Although version 4 can analyze spectra from CdTe, the precision of CdTe results is not as good as with HPGe detectors. This is because of the small detector size (a few mm^3) giving volumes over a thousand times smaller than a coaxial HPGe detector (Vo 02).

f. Sample Characteristics

    Characteristics of the measured item, such as mass, density, and shape, affect measurement precision. Compare a sample with a large area presented to the detector, but with a low areal density (g/cm^2) of plutonium, to a sample of the same mass with less surface area and higher areal density. For the former case, the lower energy gamma rays will be enhanced relative to those at higher energy and the 120–450 keV region may produce the best precision. Conversely, the second sample will have an enhanced high-energy region relative to the low areal density sample. These characteristics can often be observed in the shape of the relative efficiency curve (see Fig. II-1) which tends to fall with energy more rapidly for low areal density samples while samples with a greater areal density tend to have a higher relative efficiency continuing to higher energies.

3. Prediction of Precision in the FRAM Code

    Every FRAM measurement includes a predicted value for *sigma*, the absolute error in the measured mass %, from the propagation of counting uncertainties in the photopeak areas. This is also reported as a RSD(%). These fundamental errors are propagated to produce the absolute and relative errors in other parameters. We do not include any systematic error components in *sigma* or RSD(%) so that we may check our purely statistical error prediction with repeated measurements.

    We confirm the correctness of the FRAM error propagation by analyzing many sets of repeated measurements on many different types of samples. We compare the sigma predicted by FRAM with that calculated from repeated measurements. The ratio of these two sigmas should be near unity within the uncertainty of the observed sigma (Eq. VII-3). Table VII-4 displays the average ratio, over many data sets, of the predicted sigma to the estimate of sigma observed from repeated measurements on plutonium samples. Table VII-5 displays the same information for repeated measurements on uranium.

    We predict the statistical uncertainty very well for ^{238}Pu, ^{239}Pu, and ^{240}Pu. FRAM underestimates the statistical error by about 25% (relative) for ^{241}Pu and ^{241}Am. This likely arises from the difficulty in correctly apportioning the uncertainties for the co-energetic peaks that contain contributions from both ^{241}Pu (and daughter ^{237}U) and ^{241}Am. The error in P_{eff} is overestimated in FRAM likely arising from the correlations resulting from the normalization condition that all isotopic fractions must sum to unity.

Table VII-4   Comparison of Predicted and Observed Uncertainties for Pu Analysis Using
FRAM v. 4

Detector	Region (keV)	No. Data Sets	No. Meas.	Average Ratio: Predicted/Observed					
				^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{241}Am	P_{eff}
Coaxial	120–450	46	751	0.97	0.97	0.98	0.65	0.74	1.28
Coaxial	200–800	53	828	0.73	0.94	0.94	0.59	0.82	1.18

Table VII-5   Comparison of Predicted and Observed Uncertainties for U Analysis Using
FRAM

Detector	Region (keV)	No. Data Sets	No. Meas.	Average Ratio: Predicted/Observed		
				^{234}U	^{235}U	^{238}U
Coaxial	120–1001	33	415	1.35	0.84	0.80

The error predictions for uranium are reasonably good. When displayed as a function of ^{235}U enrichment (Fig. VII-3), we do see some ^{235}U dependence. Fig. VII-3 shows the uncertainty from the finite number of measurements (Eq. VII-3) for the observed uncertainty in each data set.
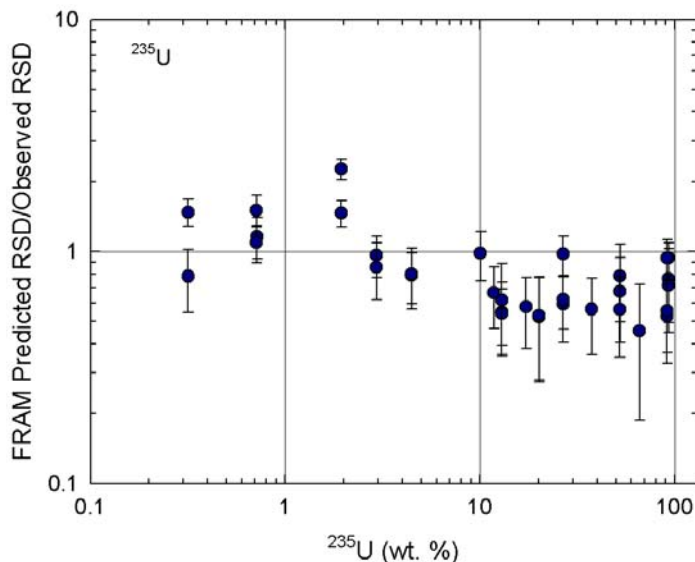

Fig. VII-3. U-235 dependence of FRAM estimate of ^{235}U statistical uncertainty.

4. Examples of FRAM's Statistical Precision

In this section, we present examples of FRAM's statistical precision taken from sets of repeated measurements. In Fig. VII-4 and -5, we display the precision for P_{eff} and ^{240}Pu, plotted vs. sample mass. These data represent not only different samples, masses, and isotopic compositions, but also different detectors, counting times, counting rates, and data acquisition electronics. Counting times for most data sets are 1 h, but some are 30 min or 2 h. The data display the wide range of

precisions that one might expect from the many different measurement conditions applicable to FRAM analysis.

One should also note that these are laboratory measurements, in most cases, where one was able to attain an optimum count rate from the sample. In many field applications, detector-to-sample distance constraints prevent this and lead to poorer measurement precision.

The coax and planar data in the 120–450 (420) keV region show similar results, at least at lower masses. We do see a definite trend (precision becomes poorer as mass decreases) in the precision vs. mass for the 200–800 keV region. This precision is worse than in the 120–450 keV region for small masses but can be, in many cases, the best choice for large mass samples.
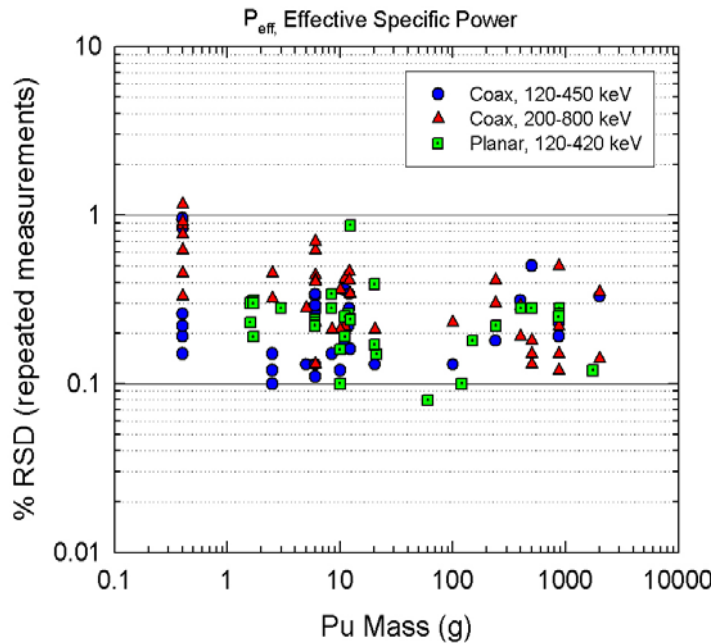
Fig. VII-4. Observed precision for P_{eff} for different detectors, different isotopic compositions, different count times, and different analysis regions as a function of plutonium mass.
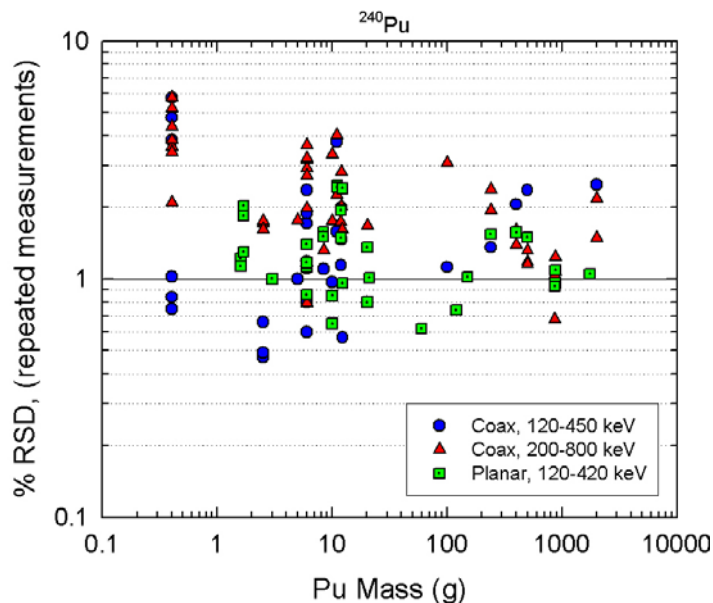

Fig VII-5. Observed precision for ^{240}Pu for different detectors, isotopic compositions, count times, and analysis regions as a function of plutonium mass.

Figure VII-6 extracts data for a single analysis method, coaxial detector in the 120–450 keV region. Each sample's data is analyzed for precision of ^{240}Pu and P_{eff}. The plotted ratio shows that the precision for P_{eff} is better than that for ^{240}Pu for every sample. Figure VII-7 displays the

precision of the measurement of the ^{235}U isotopic fraction in pure uranium samples. The precisions are those from repeated measurements and the data sets represent both 30-min and 1-h data. The detectors used are 25–30 % relative efficiency coaxial detectors. The measurement precision is pretty much constant for ^{235}U enrichments below approximately 10 %. In this case, the measurement precision is driven by the intensity of the 258-keV ^{238}U daughter peak that effectively controls the normalization of the relative efficiency curve. The intensity of this peak does not change very much for enrichments below 10 wt%.
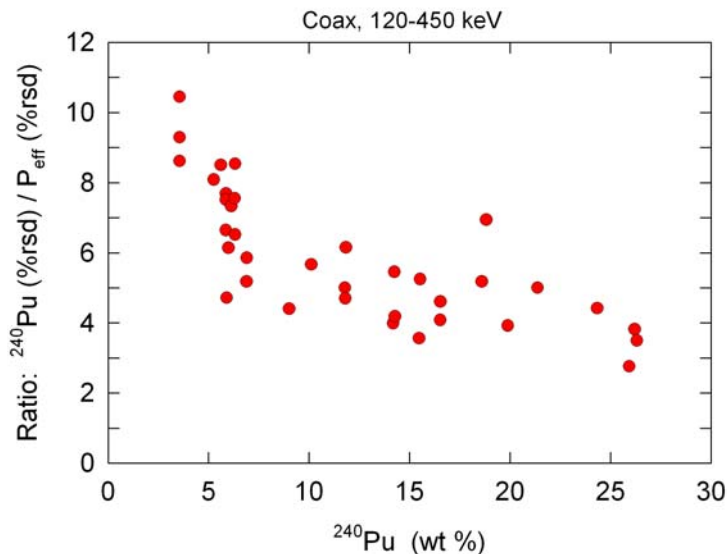

Fig. VII-6. The ratio of the observed precision for ^{240}Pu to that for P_{eff} for a coaxial detector with analysis in the 120-450 keV region is plotted vs % ^{240}Pu in the sample. Every sample shows better precision for P_{eff} than for ^{240}Pu (Ratio > 1) for the same data.

The precision of the measurement of ^{234}U is also of interest for higher enrichment samples as this isotope provides the overwhelming majority of the heat produced for calorimetry measurements. Figure VII-8 displays the ^{234}U precision for all ^{235}U enrichments for which you can measure ^{234}U. Only the enrichments above ~ 90% ^{235}U, coupled with kg-size samples, can currently be measured by calorimetry. For these samples, measurement precision can be in the 1-2 % range for ^{234}U.
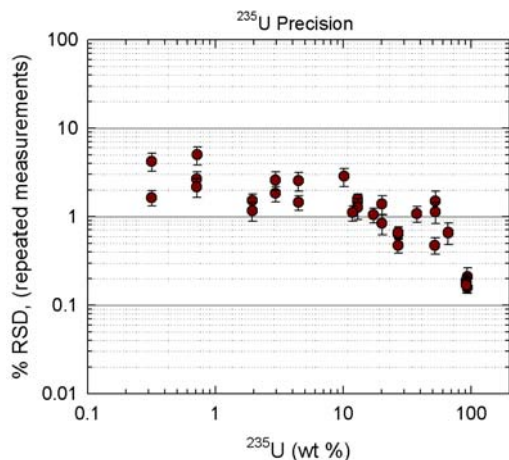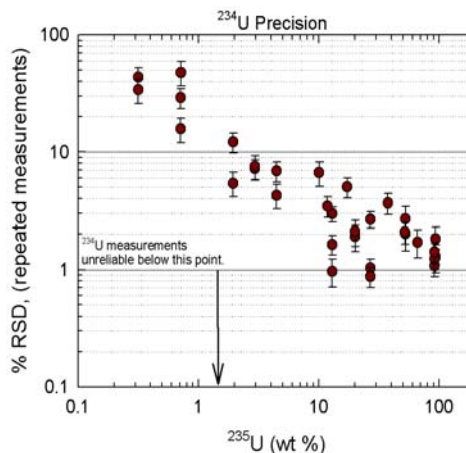

Fig. VII-7. ^{235}U measurement precision

Fig. VII-8. ^{234}U measurement precision

B. Measurement Bias

1. Introduction

Bias is defined as the difference between the measured value and the true value. For all samples, we estimate the true value by using the best available value for the isotopic composition and ^{241}Am content. The best available values almost always are derived from mass-spectrometry measurements, sometimes supplemented with alpha counting for ^{238}Pu and/or ^{241}Am. In this document, bias is expressed as a ratio of the measured value divided by the best available or "accepted value."

Los Alamos has a large number of well-characterized plutonium, uranium, and MOX-bearing items having well-known isotopic distributions and ^{241}Am content. Several items are Certified Reference Materials (CRM) traceable to the national measurement system and are national or international standards. Many items have well documented mass-spectrometry values from routine analytical characterizations. Others have mass-spectrometry values determined by several different laboratories or by extensive measurements at a single laboratory. We find that routine mass-spectrometer measurements on modern instruments are nearly always adequate for characterization of the bias of a gamma-ray isotopic composition measurement. Nevertheless, we always examine the mass-spectrometer and gamma-ray measurements closely. We especially look for problems with ^{238}Pu, ^{241}Pu, and ^{241}Am.

Pu-238 This isotope is one of the most difficult to measure by mass spectrometry. First of all, its low concentration, in the range of 0.01 % for weapons-grade plutonium, approaches the sensitivity limit for mass spectrometry. Second, it has the same mass as ^{238}U. The most careful procedures are necessary to keep the plutonium sample (typically nanograms) from being contaminated with uranium. Any such contamination will result in too large a value for the ^{238}Pu isotopic fraction. We have observed these biases from old (20–30 years) mass-spectrometer measurements of plutonium. Alpha counting can obviate these problems. Under the best circumstances analytical uncertainties for ^{238}Pu fall in the 1-3 % (relative) range and can be the limiting factor in characterizing the ^{238}Pu bias for gamma-ray isotopic composition measurements.

Pu-241 The measurement of ^{241}Pu by mass spectrometry requires a chemical separation of the mass-spectrometry sample to remove isobaric ^{241}Am. If this chemical separation is not complete, the mass-spectrometer measurement of the ^{241}Pu fraction may be biased high.

Am-241 There are no CRMs for characterizing the concentration of ^{241}Am in plutonium. This limits the ability of the analytical chemists to make traceable ^{241}Am measurements. Typical characterizations may be biased by several percent.

We discover these problems by comparing gamma-ray measurements of the sample in question with gamma-ray measurements on another sample of similar isotopic composition that is known to have a bias-free analytical characterization. We have not observed any problems with mass-spectrometry measurements on uranium.

2. Plutonium Measurement Bias

Los Alamos has a large archive of plutonium spectral data going back to 1988. These data encompass many different detectors and varying electronic configurations including NIM, portable MCA, and digital spectroscopy systems. Some of the samples have been measured on different dates, separated in some cases by eight or more years. An archival data set of over 800 coaxial HPGe detector measurements exists for plutonium. The plutonium masses vary from 0.4

to 869 g and the ^{240}Pu fractions vary from 3.56 - 26.4%. Similar, though less extensive, data sets exist for plutonium measurements with planar HPGe detectors as well as coaxial and planar detector measurements on uranium. Analysis consists of averaging results for the multiple runs for each sample and then averaging the averages for all samples to obtain an overall bias for the entire data set. We use the accepted value for ^{242}Pu so the correlation does not affect the results. We have omitted the averages for ^{238}Pu for samples known to have incorrect accepted values or have measurements so statistically poor as to be unreliable. In these cases we also omitted the value for P_{eff} that can depend strongly on ^{238}Pu. Table VII-6 shows the average bias and standard deviation in the bias for the archival coaxial measurements using the 200-800 keV region for analysis. The standard deviation in the bias is a parameter that captures the fluctuations in the measurements caused by different sample (size, isotopic composition) characteristics, container characteristics, detector and data acquisition conditions as well as errors in the accepted values. This becomes the minimum error or uncertainty that could be quoted a priori for an arbitrary measurement on an arbitrary sample and is the limiting uncertainty or minimum bias that should be assigned to an arbitrary measurement. This parameter is similar in interpretation to the between-sample variance from an analysis-of-variance calculation.

   Analysis of this type of archival data is available for the 120–450 keV analysis with coaxial detectors and also for planar detector analysis in the 120–420 keV region. The bias analysis for all three data sets is summarized in Table VII-6 for all isotopes. The ^{240}Pu$_{eff}$ bias is plotted in Figure VII-9 for coaxial detector data taken in the 120–450 keV region.

Table VII-6.   FRAM Version 4, Bias for Plutonium

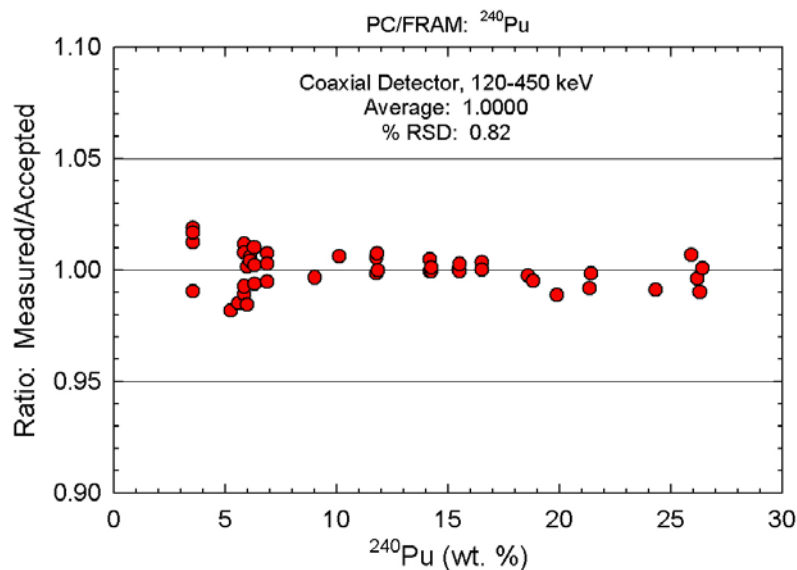|		^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{241}Am	P_{eff}
Coax 200–800	Bias	1.0049	1.0003	0.9999	1.0013	0.9982	0.9994
	% RSD	5.83	0.37	1.58	0.76	0.81	0.47
Coax 120–450	Bias	0.9958	1.0001	1.0000	0.9999	0.9955	0.9993
	% RSD	1.81	0.12	0.82	0.59	0.93	0.21
Planar 120–420 keV	Bias	1.0002	1.0002	0.9995	0.9979	0.9965	0.9995
	% RSD	1.91	0.12	0.77	0.61	1.30	0.24

Fig VII-9. Measurement bias for ^{240}Pu, 120-450 keV analysis with coaxial detectors.

3. Uranium Measurement Bias

Just as for plutonium, Los Alamos has a large number of well-characterized, uranium-bearing items used routinely as standards. The extent of these standards and the quality of their characterization is unique in the DOE complex.

For comparison with standards, we use an operator-entered, accepted value for ^{236}U since it does not have detectable gamma rays and is determined by isotopic correlation in FRAM. This is analogous to the method used to analyze plutonium standards for ^{242}Pu.

We also have an extensive archive of uranium measurements from different samples, detectors, acquisition systems, and measurement geometries. The latter is particularly important to note. It demonstrates that version 4 of FRAM does indeed correct properly for the coincidence summing effects that affected earlier uranium measurements. The values for individual items come from the average of repeated measurements of typically 30-min or 1-h duration. For the important ^{235}U isotope, the average bias for all samples, over the enrichment range from 0.3-93 % is 0.25%. The RSD(%), in the same fashion as discussed for plutonium, is just under 1%. Recall that the interpretation of this parameter is the a priori measurement uncertainty for an arbitrary sample attributed to sample and measurement specific conditions. The measurement bias data for ^{235}U are plotted in Fig. VII-10.
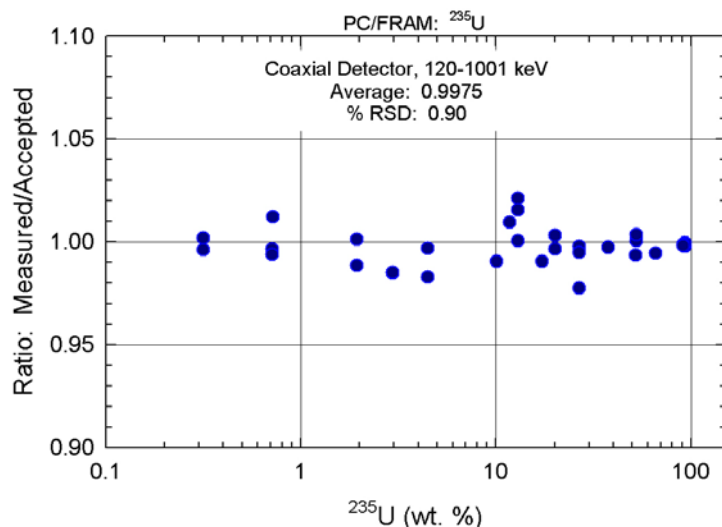

Fig. VII-10. FRAM Version 4 Measurement Bias for ^{235}U.

4. MOX Measurement Bias

Version 4 of FRAM can analyze the gamma-ray spectra from MOX samples giving results, in many cases, for the complete plutonium isotopic distribution, the ^{235}U/Pu ratio and the ^{238}U/Pu ratio.

MOX samples can be measured in several ways by FRAM depending upon the energy region analyzed and the amount of shielding surrounding the sample. U-235 is always analyzed at 185.7 keV. This gamma ray may not be present in a shielded sample and is not visible above the continuum for ^{235}U/Pu ratios below about 0.005. If the sample is lightly shielded, FRAM can obtain both the ^{235}U/Pu ratio at 185.7 keV and the ^{238}U/Pu ratio at 1001 keV. For heavily shielded samples, the ^{238}U/Pu ratio at 1001 keV may be the only uranium measurement available.

Tables VII-7 to 9 display the results of the analysis of five samples in three different ways. Results from unshielded sample measurements analyzed in the 120–450 keV range are given in Table VII-7. In the 120–450 keV analysis range, we only have access to ^{235}U at 185.7 keV. In Table VI-8, we show the results from the analysis of the unshielded samples using data in the 200–800 keV energy range. With this analysis, we are still able to analyze for ^{235}U at 185.7 keV as well as analyzing for ^{238}U at 1001 keV. In Table VII-9, we show results from the third

possible MOX data analysis using data from coaxial detectors. Here the samples are shielded with 3 mm of lead making the 185.7-keV gamma ray undetectable. We can only analyze for ^{238}U/Pu.

All of the MOX measurements tabulated in Tables VII-7 –VII-9 have a one-hour count time at rates between 30 and 40 kcps and were analyzed with FRAM version 4. We typically made 12–15 repeat measurements on each of the five samples. The Average is the average of the five individual sample averages. The % RSD is calculated from the distribution of the five individual averages of the repeated measurements. It can be thought of as the RSD of the bias. The MOX samples contained a few hundred grams of plutonium and had a ^{235}U/Pu ratio that varied from 1.4–6. U-235 enrichment ranged from 0.2–1%.

Table VII-7.   Unshielded Measurements, 120–450 keV Analysis

| | Ratio: Measured/Accepted | | | | | | | |
	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{241}Am	Spec.	^{240}Pu$_{eff}$	^{235}U/Pu
Average	0.9765	0.9966	1.0250	1.0022	0.9938	1.0005	1.0238	1.0083
% RSD	0.79	0.15	1.11	0.30	1.67	0.48	1.06	3.96

Table VII-8.   Unshielded Measurements, 200–800 keV Analysis

| | Ratio: Measured/Accepted | | | | | | | | |
	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{241}Am	Spec.	^{240}Pu$_{eff}$	^{235}U/Pu	^{238}U/Pu
Average	1.0322	0.9986	1.0101	1.0055	0.9938	1.0024	1.0100	1.0136	1.0152
% RSD	5.08	0.04	0.32	0.30	1.66	0.51	0.31	3.67	1.42

Table VII-9.   Shielded Measurements, 3-mm Lead, 200–800 keV Analysis

| | Ratio: Measured/Accepted | | | | | | | | |
	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{241}Am	Spec.	^{240}Pu$_{eff}$	^{235}U/Pu	^{238}U/Pu
Average	1.00245	1.00077	0.99391	1.00816	0.99373	0.99792	0.99415		1.04488
% RSD	4.29	0.05	0.33	0.21	1.71	0.49	0.34		0.87

C. Intercomparison Exercises

1. The PIDIE Exercise

The Plutonium Isotopic Determination Intercomparison Exercise (PIDIE) was the first exercise designed to characterize gamma-ray isotopic analysis on a worldwide basis. This exercise was organized in Europe and was notable in that the organizers provided identical sets of seven, nominal 0.4 g plutonium as PuO_2, samples to each participant. The international shipment of the sample sets proved to be extraordinarily difficult because of shipping regulations and compliance issues.

These samples came to Los Alamos in 1980 at the time when we were developing our first isotopic analysis software. The Los Alamos measurement results are so obsolete that they will not be discussed, but they were published (Morel 1991). The samples used in PIDIE are a well-documented legacy of the exercise and are still used today at Los Alamos and elsewhere to characterize gamma-ray plutonium isotopic composition measurements.

2. Uranium Enrichment Measurement Exercise, IRMM 1996

The Uranium Enrichment Measurement Exercise, sponsored by the European Safeguards Research and Development Agency (ESARDA) was held at the Institute of Reference Materials and Measurements (IRMM) in Geel, Belgium in 1996. For this exercise, participants brought their own equipment to IRMM and had one week to measure a set of well-characterized standards (Morel 00).

Although the exercise was organized primarily to test "classical" infinite sample ^{235}U enrichment measurements, Los Alamos participated with FRAM giving results for ^{234}U, ^{235}U, and ^{238}U. The organizers only reported certified values for ^{235}U. Table VII-10 displays the average of all FRAM measurements. The biases displayed are very similar to those shown in Fig. VII-10. Samples X and Y are of interest because they are freshly separated and the daughter products ^{234}Pa and ^{234m}Pa used by FRAM to characterize ^{238}U have been removed from the sample. They grow back in with the 24.1-day half-life of ^{234}Th. For samples X and Y in Table VII-10, we made a correction using the known separation time. This correction reduced the error from about 20% to about 4%. The residual error may arise from an incomplete separation. A separation time correction has been incorporated into FRAM v. 4.

Table VII-10   FRAM Results for IRMM Uranium Enrichment Exercise

Sample	Measured wt% ^{235}U mean ± sigma(mean)	Certified wt% ^{235}U	% Difference 100*(M -	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 %	

3. The Pu-2000 Exercise

The most recent intercomparison exercise sponsored by the ESARDA NDA Working Group was conducted at IRMM in 2000. This exercise was organized similar to the Uranium Enrichment Measurement Exercise with participants using their own equipment in a one-week measurement window. The purpose of this exercise, known as the Pu-2000 exercise, was to test the performance of recent isotopic analysis methods over a wide range of abundances and investigate possible sources of error. Los Alamos was one of eight participating laboratories. Twenty unknown samples characterized by IRMM were available for measurement. Four certified samples were available as references. Seventeen of the twenty samples contained plutonium or MOX with ^{240}Pu ranging from 1.6 – 26.9 wt %. The remaining three samples were isotopically pure samples of ^{239}Pu, ^{240}Pu, and ^{241}Am.

Table VII-11 displays the average and relative standard deviation (%) for the ratio (%) of the FRAM result to the IRMM certified value. Table VII-12 displays FRAM results for the isotopically pure samples. FRAM was the only code to report results for all three samples. It's versatility allowed us to modify parameter files for these very special samples.

Table VII-11   FRAM/IRMM Results for Pu-2000 Exercise, 17 MOX samples

FRAM/IRMM certified	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{241}Am
Average	0.9943	1.022	0.9940	1.0098	1.0194
RSD (%)	4.55	0.41	1.13	1.67	3.05

Table VII-12   FRAM Results for Isotopically Pure Samples (Wt. % Relative to Total Plutonium)

Sample	Type		^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{241}Am (µg/gPu)
R	Pu-239	FRAM	0.00031	99.964	0.0325	0.0029	75
		Certified	0.0000	99.979	0.0210	0.0001	3
S	Pu-240	FRAM	0.1027	0.0084	99.860	0.0001	13.6
		Certified	0.0119	0.023	99.935	0.00098	20.2
T	Am-	FRAM					100.00
		Certified					100.00

D. Factors Influencing Measurement Bias

Many factors influence the bias of a gamma-ray isotopic measurement.

1. Sample Composition Characteristics

The elemental makeup of the sample influences self-absorption thus influencing the relative efficiency curve. Version 4 accounts for this by allowing up to three different elements or compounds to define the matrix. These elements or compounds may be chosen from Al, Fe, Cd, Er, Pb, H_2O, and Concrete. These materials were chosen not only for their likely presence in the sample, but also to cover the range of atomic number. If the matrix contains elements other than the three designated in the parameter file, FRAM will use a linear combination of these three to approximate the relative efficiency curve. Differences from the true relative efficiency curve are likely to be compensated by the Hoerl function correction factor and the result is almost always a good fit to the relative efficiency points, even if the three chosen components do not match the actual sample matrix.

2. Branching Ratios

The branching ratios used in the analysis directly affect the bias in the measured isotopic ratios (Eq. II-1). Most of the branching ratios have been measured by gamma-ray spectrometry techniques. Absolute measurements of this type are very difficult and the very best results usually have an associated uncertainty of no better than 1% (relative).

3. Coincidence Summing

Coincidence summing occurs when a gamma-ray decay includes cascades of two or more gamma rays that are emitted simultaneously. These two gamma rays may be detected simultaneously and the single pulse will not represent either gamma ray. This takes events away from the full energy peak in a manner that is dependent upon the measurement geometry.

Summing effects depend upon the square of the detector solid angle (Knoll 00) and can be reduced by increasing the sample-to-detector distance. For coincidence summing to be a

problem, the coincident gamma rays must be of high enough intensity to be useful in the analysis and must be of high enough energy to escape the sample and its containment.

a. Uranium

Coincidence summing is present in low-enriched uranium measurements in the 120–1001 keV range; it particularly affects the 258-keV gamma ray from ^{234m}Pa. In general, measurements on high-enriched uranium are not affected because the 258-keV gamma ray is not used in the analysis. The ^{235}U isotopic result can be biased by as much as 15%, depending upon the sample-to-detector distance. The effects are pronounced for sample-to-detector distances below 5 cm and become small above about 15 cm.

FRAM version 4 incorporates a correction factor for coincidence summing. It is derived by examining the intensities of several gamma rays affected by coincidence summing in different proportions while knowing their correct branching ratios. This allows a correction to be made by comparing intensity ratios of the affected peaks to the ratios expected without coincidence summing.

b. Plutonium

Coincidence summing is not a problem with plutonium measurements because the filtering present for plutonium measurements removes the low energy coincident gamma rays.

4. Peak Area Determination

a. Background Shape

A parameter file is set up for a wide range of measurement conditions such as variable isotopic composition, sample mass, composition, container and shielding, detector resolution, as well as measurement geometry. Because measurement conditions affect the shape of the background continuum, the parameter file setup for the region must allow for a background continuum shape that will adapt itself to different conditions. Some "tricky" regions are illustrated and discussed below.

<u>Continuum at 160 keV</u>   The definition of the background type for the region around 160 keV is critical as this peak is the only one available for analysis of ^{240}Pu in the 120-450 keV region. We see in Fig. VII-11 that the background has an inflection point. We customarily define a quadratic background for this region with background ROIs below 160 and above 165 keV.

<u>Continuum at 148 keV</u>   The background continuum underneath the 148.6-keV peak from ^{241}Pu is usually positively sloping as is shown in Fig. VII-12. One would like to use a linear background with a smoothed step function. However, since the background is higher above 148 keV than it is below, the "step" goes the wrong way and becomes unphysical. FRAM can sense this condition and automatically recalculates the background continuum with the simpler linear form that does not incorporate a step.
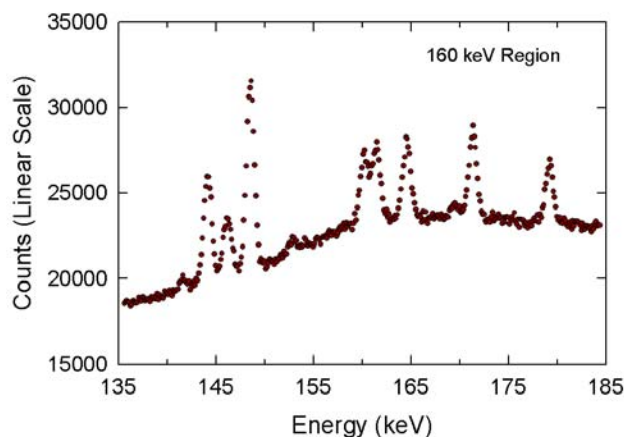
Fig. VII-11. The background in the region around 160 keV shows curvature making definition of the background continuum difficult. Poor definition can bias the ^{240}Pu peak area at 160.3 keV.
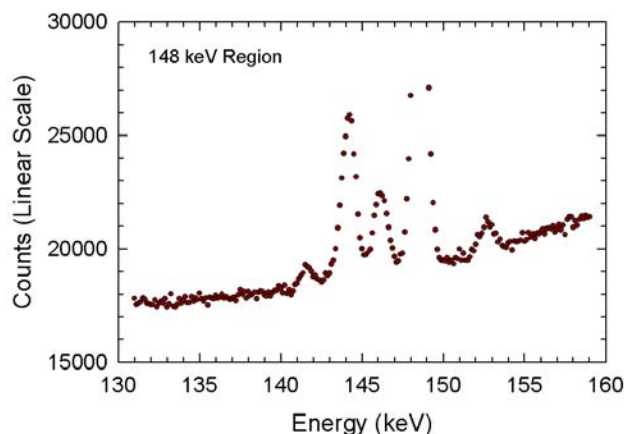

Fig. VII-12. The positive sloping background makes it difficult to calculate an accurate background continuum for the 148.6 keV from ^{241}Pu

Continuum at 208 keV   The ^{241}Pu–^{237}U peak at 208 keV can vary over a factor of 500 in intensity relative to neighboring peaks, depending upon isotopic composition. This peak usually presents the classic case of the linear step function background type. We see in Figs VII-13, -14 that the underlying continuum can change slope above and below 208 keV. While a linear step function works well in Fig. VII-13 the continuum in Fig VII-14 is better characterized by a bilinear step having a different slope above and below the 208-keV peak. FRAM automatically switches from a bilinear step background to a linear step, if it finds the bilinear step inappropriate.

b. Interferences

FRAM can be characterized as a response function code. The peak shapes are determined in the internal calibration phase of the analysis and the peaks for analysis are predetermined by the setup of the parameter file. An unexpected peak (one not listed in the parameter file) appearing in an analyzed region, may bias the analysis of the designated peaks in the region. This bias can arise by direct interference with the peak or the interference may appear in a region used to define the background continuum thus biasing the peak area via an incorrect background subtraction. Once an interference peak is recognized, we can easily modify the parameter file to and then reanalyze the data.
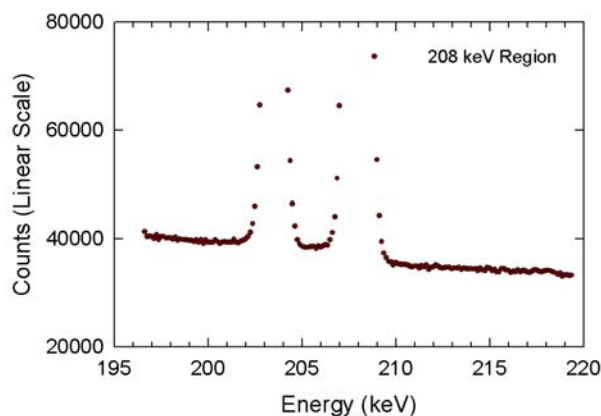
Fig. VII-13. The 208-keV region from a low burnup plutonium sample. The background continuum has the same slope above and below 208 keV.
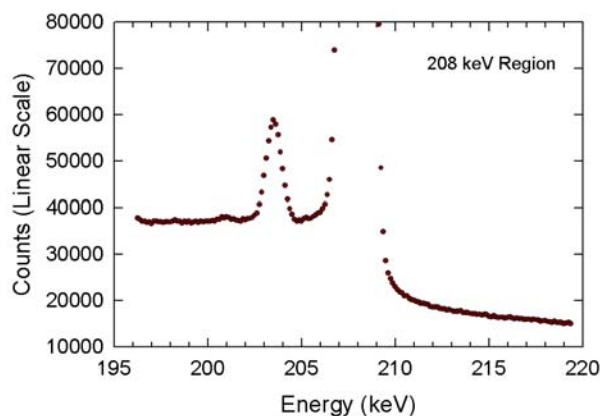

Fig. VII-14. The 208-keV region from a high burnup plutonium sample. The background continuum has different slopes above and below 208 keV.

The general-purpose plutonium parameter files delivered with version 4 of FRAM are set up to include common interferences from ^{235}U, ^{243}Am–^{239}Np, and ^{237}Np. The general-purpose parameter files are usually adequate for low concentrations but may require modification for very high concentrations. The FRAM user has another tool to discover unexpected interference peaks by displaying the analysis and background regions from the parameter file with color highlights overlaid on the spectrum. The highlighted spectrum will reveal if any unexpected interference peaks appear in a background region. Highlighting the analysis regions can show if the energy calibration matches that in the parameter file. If the highlighted peak regions do not contain the peaks, the energy calibration in the parameter file may not be appropriate for the spectrum.

E. Bias Correction

We have developed several procedures for identifying and correcting biases. In general, the procedures rely on measurement of standards, identification of the source of the bias, and correction of the bias, usually by adjustment of branching ratios.

The first step is to carefully set up the parameter file. This includes identifying the full range of measurement conditions, isotopic compositions, ^{241}Am content, interferences, and any other parameters that might affect the gamma-ray spectrum. The user then establishes the peak and background ROIs based on representative spectra. This is the most important step in making bias corrections.

1. Adjustment of Branching Ratios

If we still observe biases, even after careful setup of the parameter file, we may adjust the branching ratios to reduce or eliminate the bias. There is good justification for this approach, although it is questioned by some as inappropriate because branching ratios are imperfectly known constants of nature. Our reasoning is that we are making corrections for imperfections in the analysis in addition to correcting for uncertainties in our knowledge of the branching ratios.

One type of bias arises from peak area uncertainties caused by imperfect background subtraction. A second uncertainty comes from imperfections in the response function fitting of closely lying, overlapping peaks. A third uncertainty arises from biases in the published branching intensities. We have previously mentioned imperfections in modeling the relative

efficiency curve as a contributor to bias. Finally, the model (Gaussian plus exponential tail) used to fit the peaks is itself imperfect.

There are two principle sources of branching ratio values for plutonium, the work of Gunnink and coworkers (Gunnink 76a) and the Table of the Isotopes (Firestone 96). Any adjustment of plutonium branching ratios usually starts from these sources.

2. Observation of Peak Area Biases

We examine a FRAM analysis to look for inconsistencies in the activities calculated for gamma rays from a single isotope. We look at the consistency with consideration of the counting statistics for analyses from different samples. Any inconsistency that shows up regularly is examined in closer detail.

The bias correction for an isotope with only a single gamma ray (^{238}Pu and ^{240}Pu) can be straightforward. We examine the bias as a function of isotopic fraction. A simple branching ratio adjustment is usually indicated when the data do not show a trend with changing isotopic composition. The situation is more complex, however, if there is a trend. This could indicate unresolved or imperfect subtraction of interferences or a background continuum subtraction that does not adapt well over the full range of data.

3. Least-Squares Adjustment of Branching Ratios

A more general adjustment method utilizes high precision data from multiple measurements of samples with well-characterized isotopic compositions. A "Downhill Simplex Method" is incorporated into a special developer's version of FRAM and used to minimize the weighted chi square of the chosen isotopic ratio(s).   Multiple branching ratios are varied iteratively to find a minimum.

With this method we can obtain accuracies of a percent (relative) or less for the individual isotopic ratios. This fine-tuning usually does not make large adjustments from the published values. Indeed, if the adjustment is large (> 5 %), one should carefully examine the data to see if it is justified.

We have compared the adjusted branching ratios used in FRAM with published (Gunnink 76a, Firestone 96) values to verify the appropriateness of this approach. The FRAM values agree with the published values within the differences and uncertainties in the published values.

4. Use of Standards

All of the adjustment processes ultimately depend upon comparison of a measured value with a reference value. Thus, while gamma-ray isotopic composition measurements do not directly depend upon standards (Eq. II-1), we do rely heavily on comparison with standards for ultimate validation of the technique as well as the "fine tuning" of the analysis process. We have discussed analytical characterization issues in section VII. B. 1 and noted that they still contribute to gamma-ray isotopic analysis uncertainties for ^{238}Pu and ^{241}Am.

VIII. MAKING MEASUREMENTS FOR FRAM ANALYSIS

The FRAM analysis of a gamma ray spectrum can only be as good as the data will permit. Poor quality spectral data may limit the ability of FRAM to derive accurate results from the measurement, although the flexibility of FRAM's analysis gives one a better chance of a good analysis than with any other analysis method. This section will present information on choosing and setting up equipment to collect high quality spectral data for FRAM analysis.

A. Choice of Detector

FRAM has been used with a wide variety of HPGe detectors, both planar and coaxial, as well as CdTe detectors. The detector procurement question will usually revolve around what size and type of HPGe detector.

First consider the mix of samples to be measured and their packaging. A coaxial detector is the appropriate choice for the user that makes a wide range of measurements on samples contained both in thin-walled and heavy-walled or shielded containers. Coaxial detectors can literally "do it all" with the exception of analyzing the 100-keV region. Specifications for a small coaxial detector often used with FRAM are shown in Table VIII-1. Planar detectors can be a good choice for samples in thin-walled containers and are required if one wishes to make measurements in the 100-keV region. Table VIII-1 also gives specifications of a planar detector successfully used with FRAM.

Many other HPGe detectors have been used with FRAM. Waste applications have used coaxial detectors of nearly 100% relative efficiency. The poorer resolution of these larger detectors may introduce some biases and require ROIs to be enlarged. We have used large semi-planar detectors (70-mm dia by 30-mm thick) very successfully for both plutonium and uranium. Measurements through thick-walled containers are usually improved if the detectors have higher efficiency than the "standard" 25%.

Table VIII-1. HPGe Detector Specifications for General Purpose FRAM Usage

Coaxial Detector	
Geometry	Coaxial, P-type germanium [1]
Efficiency	At least 25% relative efficiency at 1.33 MeV in the usual definition
1000 cps FWHM (122 keV)	≤ 750 eV at ≤ 6μs shaping time [2]
1000 cps FWHM (1.33 MeV)	≤ 1.75 keV at ≤ 6μs shaping time
30 kcps SWHM (122 keV)	≤ 880 eV at ≤ 2μs shaping time
30 kcps FWHM (1.33 MeV)	≤ 2.00 keV at ≤ 2μs shaping time
Peak Shape	[FW1/50M]/FWHM ≤ 2.50 at rates ≤ 30 kcps and time constants from 2 – 6 μs
Preamplifier/Energy Rate	Resistive feedback preamplifier with Energy Rate ≥ 50,000 MeV/s
Planar Detector	
Geometry	Planar
Crystal Size	≥ 25 mm diameter, ≥ 13 mm thick
1000 cps FWHM (122 keV)	≤ 520 eV at ≤ 6μs shaping time
50 kcps FWHM (122 keV)	≤ 580 eV at ≤ 1μs shaping time
Peak Shape	[FW1/50M]/FWHM ≤ 2.50 at rates ≤ 50 kcps and time constants from 1 – 6 μs
Preamplifier/Energy Rate	Resistive feedback preamplifier with Energy Rate ≥ 10,000 MeV/s [3]

[1] We specify P-type germanium because the resolution is usually better than that of N-type for the same detector size. N-type detectors have also been used successfully with FRAM.

[2] Shaping times are specified for analog amplifiers.

[3] The preamplifier energy rate for a planar detector is a trade off with low-rate resolution. This specification assures that the preamplifier will allow a maximum counting rate above 50 kcps for a typical plutonium spectrum. A more stringent low rate resolution specification may be achieved at the expense of the maximum preamplifier count rate.

B. Choice of Energy Range

 Planar Detector With a planar detector one has the choice of 1) analyzing data above 120 keV (typically taken in 4096 channels at a gain of ~ 0.1 keV/chan to include 120–420 keV) or 2) analyzing the 100-keV region (typically taken in 4096 channels at a gain of ~0.075 keV/chan to include 0–300 keV). The 120–420 keV analysis provides more penetrability and will work through 12–15 mm of steel. The 100-keV region analysis provides better precision for plutonium in thin-walled containers at shorter counting times, compared to the higher energy analysis.

 Coaxial Detector A coaxial detector is the most versatile choice if one is limited to a single detector. It can analyze data from all the samples that a planar detector can plus it can analyze data from samples contained in very substantially-shielded containers, up to and including about 25 mm of lead (Hypes 00). The amount of shielding governs the energy range used in the analysis. "Bare" samples are analyzed starting at about 120 keV. Samples shielded with about 6 mm lead or less are analyzed starting at about 200 keV. Analysis of samples with about 12 mm lead starts at about 300 keV as all gamma rays below that energy are removed by the shielding.

 We typically acquire coaxial detector data in 8192 channels at a gain of 0.125 keV/chan spanning the energy range 0 – 1024 keV. This range is very convenient as it allows analysis of both uranium and plutonium spectra with the same electronic settings.

C. Collection of Pulse Height Spectra

1. Electronics

 Data acquisition electronics used to collect spectral data for isotopic analysis span the range from simple analog to modern digital systems. Both types of systems have been implemented in mains-powered and battery-powered versions. The multichannel pulse height analyzers (MCA) that record the spectra have advanced over the years from large, dedicated, standalone desktop units to small, portable units consisting of a computer, an MCA board, and MCA emulator software. All MCA functions are integrated into small, portable, battery-powered systems suitable for field use.   The portable MCAs also operate from mains power making them useful in both the laboratory and the field. The small size and low power requirements of portable, battery-powered analog systems usually extract a toll in system performance when compared to NIM module systems (Sampson 01).

 Digital data acquisitions systems have now reached full commercial application. These systems digitize the signal directly out of the preamplifier. They offer an expanded selection of time constants and improved performance for both resolution and throughput over corresponding analog systems (Sampson 01). Digital systems, even in their portable, battery-powered form, demonstrate performance superior to the best analog systems and are recommended for any new procurement. Vo has performed extensive testing of the performance of many commercially available data acquisition systems (Vo 02).

2. Count Rate Considerations

 The importance of count rate in the collection of high quality spectra cannot be overestimated. Too high a count rate may lead to degraded detector resolution, tailing from pulse pileup, and sum peaks, all of which can lead to measurement bias. Conversely, very low count rates yield spectra with too few counts and poor statistical precision. Increasing the count time to compensate for low count rate often leads to unacceptably low throughput. The count rate–resolution–throughput tradeoff is one that needs to be evaluated for every measurement system and situation. Many of the factors contributing to this question have been discussed in section VII.A.2.a. and also described by Parker (Parker 91a).

One of the first tests is the simultaneous measurement of throughput and resolution under realistic measurement conditions. The user selects a prominent peak and measures its net peak area and FWHM for a fixed count time (true or real) while varying the input count rate for a constant value of the time constant. The required peak areas and FWHM can be obtained from the MCA. The peak can be from a standard test source, ^{57}Co at 122 keV, ^{137}Cs at 661.6 keV, or from plutonium at 129.3 keV or 208.0 keV. The throughput and resolution can be plotted as shown in Fig. VII-1. The user then repeats the measurements for other time constants.

Short time constants will usually increase throughput, move the throughput maximum to a higher input rate, and make the resolution poorer. For analog systems, the resolution may worsen very rapidly at high count rates. You should operate at an input rate about 50-70% of the peak throughput rate which captures over 80% of the maximum throughput.

Sometimes the sample-to-detector distance is constrained. A large sample may produce an unacceptably high count rate with resolution that is too poor for successful analysis. These cases benefit greatly from the use of a digital spectrometer. FRAM has been successfully used with a digital spectroscopy system operating with input count rates up to 100 kcps and has made bias-free analyses for all of the conditions displayed in Fig. VII-1.

The detector and analysis region chosen can influence whether one chooses to optimize resolution or throughput. We give some general suggestions below.

100 keV region, planar detector: Optimization for measurements in the 100-keV region should concentrate on obtaining the best resolution at the expense of throughput. The larger intensities in this region make up for lowered throughput from increasing the time constant to 2 μs or even 3 μs from 1 μs.

120-420 keV region, planar detector: Optimization in this case usually concentrates on maximizing throughput by use of a 1-μs time constant and perhaps increasing the area of the detector. Resolution is secondary because we routinely use coaxial detectors with great success in this same region. Coaxial detector resolution at 122 keV is typically 1.5 times larger than that of a planar detector.

120-450 keV region, coaxial detector: We optimize for throughput in this region because of the lower intrinsic intensity of the gamma rays.   Digital spectrometers can simultaneously improve resolution and throughput. Analog time constants can be as low as 2 μs for top quality 25-30 % relative efficiency coaxial detectors. Higher efficiency detectors usually require a longer time constant.

200-800 keV region, coaxial detector: This region is difficult to optimize. The intrinsic plutonium gamma-ray intensities are low which calls for larger, generally poorer resolution detectors. The peak fitting in the 637-646 keV peak region, containing five peaks including the important ^{240}Pu peak at 642.5 keV, demands the best possible resolution. Digital spectrometers improve the measurements in this region allowing FRAM to work at input rates as high as 100 kcps.

120-1024 keV region, coaxial detector, U: The uranium spectrum is not as complex as plutonium and does not demand as good resolution. Uranium systems can usually be optimized for throughput.

3. Pulse Pileup

a. Coincidence Summing

Coincidence summing occurs when gamma rays are emitted and detected in coincidence. It is possible for such gamma rays with energies E_1 and E_2 to be detected simultaneously in a time much shorter than the resolving time of the pulse processing electronics. The result is a new

peak at the energy of the sum of the two gamma rays. More important, for isotopic analysis, is the diminution of the individual peak areas at E_1 and E_2 .

Coincidence summing is isotope dependent and does not cancel out as random summing does. The effect is proportional to the solid angle of the detector as seen from the sample and can be reduced by increasing the sample-to-detector distance.

b. Random Summing

Random pulse pileup occurs when two pulses from independent decays are detected with a time separation shorter than the pulse-processing time of the analysis system. The result is a single distorted pulse with a pulse height unrelated to either input pulse. Such pileup can be minimized by proper pole-zero adjustment on the analog amplifier, or proper pulse-shape optimization on a digital spectroscopy system. Pileup rejection circuitry in modern amplifiers can reject the storage of pileup pulses if they can be resolved by the pileup rejecter, which is typically about 0.5 μs.

Pulse pileup can affect an isotopic measurement. The principal effect of random pulse pileup is to add misshapen peaks and structure to the spectrum. If these additional peaks fall inside a peak or background region defined in the parameter file, the analysis of that region will likely be biased. An example of random pulse pileup is shown in Fig. VIII-1. This example shows the summing of the 59.54-keV ^{241}Am peak with several peaks in the 100-keV region. The arrows show the location of the sum peaks. The sum peak that falls directly under the 160.3-keV ^{240}Pu peak is especially important. It will cause a bias in the 160.3-keV peak area and it may not be noticed if the other sum peaks are weak.

Random summing is assumed to result in equal losses throughout the entire spectrum. Thus, isotopic ratio methods should be unaffected. This assumption may not hold precisely because peak widths increase with energy, but it appears that the losses occurring from random summing do not materially affect isotopic ratio measurements. Two setup conditions also affect pulse pileup. Pileup can be reduced by operating at a shorter time constant or by operating at a lower count rate.
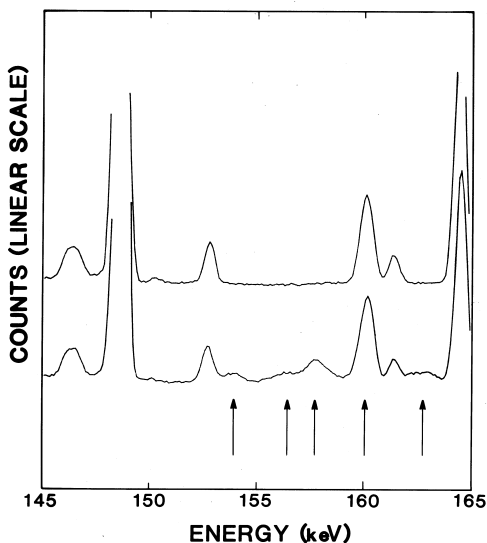

Fig. VIII-1. The effect of random pulse pileup on the 160-keV region can be dramatic. The top trace is a spectrum without pileup peaks. The bottom trace shows the effects of pileup of the 59.54 - keV ^{241}Am peak with other peaks in the 100-keV region. The arrows mark the location of the pileup peaks.

4. Filtering

Spectroscopists place absorbing materials or "filters" in front of the detector to preferentially absorb low energy radiation that is not useful for the measurement. Filters are typically chosen to have their K-absorption edge near but below the energy of the radiation to be absorbed. This maximizes the filter absorption and minimizes its effect on high-energy gamma rays. The

removal of unwanted low energy gamma rays eliminates a source of pileup summing, both random and coincidence summing.

The 59.54-keV gamma ray of ^{241}Am is almost always filtered. Its activity, 3-5 orders of magnitude greater than any other Pu gamma ray, dominates the spectrum of any Pu in a thin-walled container. If not filtered, the dead time from this gamma ray will prevent any useful information from being recorded. The unfiltered count rate from this gamma ray can often paralyze the detector and electronics.

Thin sheets of cadmium (Cd, Z=48, K shell binding energy = 26.71 keV) are most widely used to filter plutonium spectra. Tin (Sn, Z=50, K shell binding energy = 29.20 keV) is used in situations where health concerns preclude the use of cadmium. Lead (Pb, Z=82, K shell binding energy = 88.00 keV) is also used because of its wide availability.

A widely used "rule of thumb" is that the filter should be thick enough to reduce the intensity of the 59.5-keV gamma ray to the same magnitude as the intensity of the 100-keV complex of gamma and X rays. When this is met, one does not observe pileup in the 160-keV region (top trace in Fig. VIII-1).

The FRAM systems used at the Los Alamos Plutonium Facility measure the isotopic composition of plutonium over a very wide range of ^{241}Am content (< 100 ppm to $> 500,000$ ppm) where ppm is parts per million with respect to total plutonium. This dynamic range of nearly 10,000 makes it difficult to optimize the filter for all measurements. At Los Alamos, we use a single filter for all measurements so that the operator does not have to make filter decisions. This filter consists of about 2.0 mm of Cd backed by 0.25–0.51 mm of Cu. The copper (or like Z material) absorbs cadmium x rays. This graded filter keeps the spectrum below 80 keV as free as possible of photopeaks. It minimizes pileup summing from strong low-energy peaks. We adhere to the philosophy that any low-energy peak (below 100 keV) that is not used in the analysis is a "bad" peak. It cannot help the measurement and may hurt by contributing to sum peaks.

The "one size fits all" filter does degrade the intensity of useful photopeaks below 200 keV. The measurement precision of the ^{241}Am peak at 125.3 keV and the ^{240}Pu peak at 160.3 keV can be improved if one uses less filtering when ^{241}Am is less than 10,000 ppm. The measurement precision using the 100-keV region will be improved with filters that are less than 2.0 mm of Cd because the sum peaks do not fall into an energy region that is used in the analysis. Table VIII-2 displays the transmission of common filter components at several important gamma-ray energies.

Table VIII-2. Transmission of Common Filter Components

Energy	Cadmium			Lead	
	0.41 mm	0.81 mm	2.0 mm	0.41 mm	0.81 mm
59.5	0.127	0.016	0.000032	0.118	0.014
104.2	0.638	0.407	0.105	0.109	0.012
125.3	0.756	0.571	0.247	0.251	0.063
160.3	0.856	0.733	0.461	0.474	0.224

5. Shielding

The shielding used around the HPGe detector for analysis of plutonium isotopic composition is usually tailored to the energy range of the analysis to reduce direct photopeak interferences from other plutonium nearby. A system measuring in the 600—800 keV range requires more shielding than one used in the 100-keV region. If size and weight constraints permit, the

shielding around the side of the detector should provide an attenuation of at least 100 for the highest energy critical to the analysis.

The shielding for photopeak interferences is of primary importance, as these will directly bias the measurement. Shielding for continuum gamma rays is of secondary importance. Background gamma rays falling in the continuum will worsen precision but not the bias.

Lead (Pb) and machinable tungsten are commonly used for shields. Lead is a health hazard and is often canned in steel to alleviate that concern. Machinable tungsten is used where maximum shielding is required with minimum size; it does not require canning. Table VIII-3 shows the thickness of Pb and W shields required to provide a factor of 100 attenuation at common analysis energies.

Table VIII-3. Thickness in cm for Factor of 100 Attenuation

Energy (keV)	Pb ($\rho = 11.35$)	W ($\rho = 15$)
104	0.084	0.080
185	0.362	0.351
208	0.476	0.461
414	2.00	1.82
662	3.94	3.33
766	4.61	3.84
1001	5.97	4.83

The shield weight becomes an overriding consideration for portable systems. Because FRAM uses energies up to 1001 keV, a hand-held HPGe detector measuring isotopic composition of stacked UF_6 cylinders suffers from inadequate shielding even using the thickest practical shield (~ 12.7 mm Pb).

FRAM has been used at the Los Alamos Plutonium Facility in a mobile, mains-powered system that is easily transportable (Fig. VIII-5). This system uses about 19 mm of machinable tungsten shielding which Table VIII-3 shows provides a factor of 100 attenuation at 414 keV. Fixed FRAM systems are usually configured with at least a 5-cm-thick Pb shield around the lateral surface whether with a coaxial or planar HPGe detector. This provides a factor of 160 attenuation at 766 keV, the highest energy analyzed in a shielded-sample plutonium measurement.

Fig. VIII-5. A mobile PC/FRAM system configured on a commercial thyroid scanner cart for use at the Los Alamos Plutonium Facility. The detector shielding is 12.7 mm of machinable tungsten.

IX. DIFFICULT MEASUREMENT SITUATIONS

A. Using FRAM With Rate-Loss Correction Sources

We have successfully analyzed data taken with a rate-loss correction source attached to the detector (this correction source is used in Segmented and Tomographic Gamma Scanners performing transmission-corrected assays). The rate-loss correction source may have gamma rays that interfere directly with plutonium gamma rays used by FRAM. In addition, the gamma or x rays from the rate-loss source may randomly sum with plutonium gamma rays to produce unwanted interferences. The versatility of FRAM has allowed analysis of data from both situations (Sampson 03).

B. Simultaneous FRAM/AWCC Measurements

An AWCC user wished to perform passive coincidence measurements with the AWCC while simultaneously making an isotopic measurement with an HPGe detector. It was possible to make direct measurements using a 25% relative efficiency coaxial detector inside an *unmodified* AWCC. The sample contained 847g PuO_2 with a nominal 16% ^{240}Pu. The detector face was positioned 3.8 cm from the outer surface of the AWCC giving a gross count rate of 40 kcps. The total sample-to-detector distance was 30 cm. Each spectrum (10 of 30 min) was analyzed in the 120–450 keV and 200–800 keV regions. Table IX-1 below gives the results of the two analyses of the 10 measurements.

The measurement results are clearly more precise using the 200–800 keV analysis. Both sets of results show that spectra suitable for FRAM analysis can be obtained by collecting data through the body of the neutron counter.

Table IX-1   Results through AWCC Wall (operator ^{242}Pu) 847g PuO_2 with 16% ^{240}Pu

Analysis	Pu-238	Pu-239	Pu-240	Pu-241	Am-241
120–450 keV Analysis					
Avg: Measured/Accepted	0.9745	0.9999	1.0010	0.9842	1.0127
% RSD distribution	13.0	1.3	6.4	1.4	2.5
200–800 keV Analysis					
Avg: Measured/Accepted	1.0163	1.0049	0.9747	1.0163	0.9868
% RSD distribution	3.5	0.26	1.4	0.90	0.91

C. Measurements Through Thick Shielding

FRAM's ability to measure through very thick shielding is a well known characteristic. While we cannot document all possible measurement scenarios, we will give some examples of the extreme measurement conditions encountered for shielded samples.

1. Steel Shielding

10–12 mm of steel is the limit for reliable measurements using the 100-keV region.

a. Plutonium

Very early, it was shown that FRAM could easily analyze plutonium isotopic data from the 120–450 keV region taken through 12.5 mm of steel. Somewhat later, it was demonstrated that FRAM could analyze a plutonium spectrum taken through 25 mm of steel using the standard parameter file for the 200–800 keV region. It is clear that FRAM can measure through steel much thicker than 25 mm, but the experiments have not been performed to define a higher limit.

b. Uranium

A common problem is the measurement of low-enriched uranium in UF_6 cylinders. Cylinders containing UF_6 with enrichments in the 1 – 5 % range typically have a wall thickness of 13 mm. The wall material may be steel or a nickel alloy possessing about the same absorption properties as steel. Depleted or natural UF_6 is often found in cylinders with 16-mm-thick walls.

FRAM has no problem analyzing coaxial detector data from uranium shielded with 16 mm of steel (Sampson 01). Numerous measurements were made with a 28%-relative-efficiency coaxial detector on enrichment standards in a geometry simulating a UF_6 cylinder. Figure IX-1 displays this geometry.
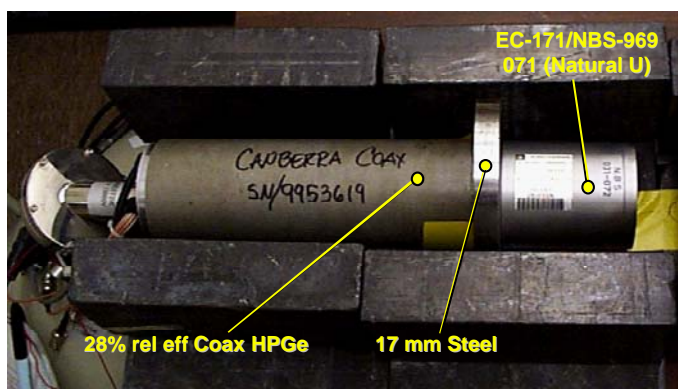

Fig. IX-1. Measurement geometry with detector, steel absorber, and sample simulating the measurement of a UF_6 cylinder.

Figure IX-2 displays, as a function of enrichment and count time, the relative standard deviation of a single measurement obtained from 15 repeat measurements. This was repeated using a semi-planar detector (70-mm diam by 30-mm thick) that yielded the same results as the coaxial detector.
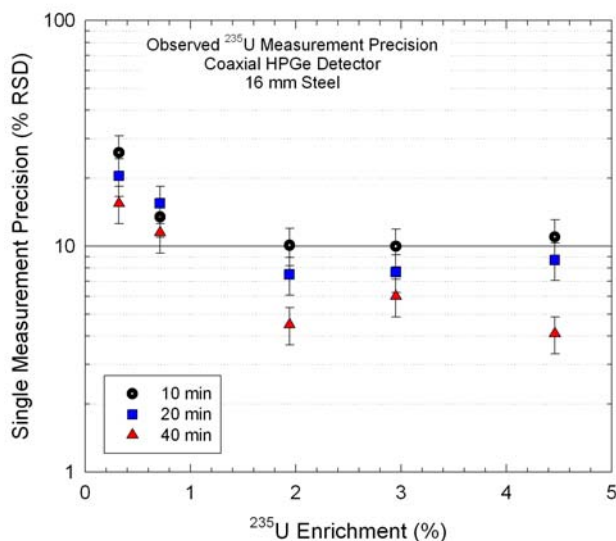

Fig. IX-2. Single measurement precision of ^{235}U calculated from 15 replicates. Five standards were measured in 8192 channels with a 28%-relative-efficiency coaxial HPGe. Measurements made in the geometry of Fig. IX-1.

FRAM has analyzed spectra acquired from type 30B UF_6 cylinders. This cylinder has 13-mm-thick steel walls. The measurements, made with a 26%-relative-efficiency coaxial detector in 8192 channels, had a precision of 6-8 % (1 RSD) for 20–30 min covering enrichments from natural to 4.7%. This is consistent with Fig. IX-2. One should consider the following when choosing between FRAM or the classical enrichment method for UF_6 cylinder assay.

The advantages of using FRAM are:

- Because FRAM does not require calibration, it is faster for a limited number of measurements than the classical enrichment method (infinite-thickness).
- The classical method requires a measurement of the cylinder wall thickness that adds time and uncertainty to the measurement and requires additional equipment.
- FRAM has no geometric reproducibility requirement. The classical enrichment method requires the geometry of the unknown be the same as for the calibration.
- FRAM can measure other materials in the facility such as scrap, waste, and oxide powder. The classical method is not able to do the other measurements without a new calibration and the assurance of a geometry that is "infinitely thick".

There are situations in which it is advantageous to use classical enrichment methods:

- The classical method has a shorter measurement time after calibration is complete.
- FRAM requires decay equilibrium (~100 days from separation) in the ^{238}U decay. The classical enrichment method does not because it measures ^{235}U directly.
- Shielding requirements are more stringent because FRAM measures up to 1 MeV. The classical enrichment method measures at 185 keV.

The discussion of gamma-ray isotopic measurements on UF$_6$ cylinders would not be complete without noting the capability of planar HPGe detectors. Such detectors have long been considered too small and inefficient for FRAM which measures up to 1 MeV. The IAEA asked if FRAM could analyze data from 25-mm-dia. x 16-mm-thick planar HPGe measurements of UF$_6$ cylinders; data that could not be analyzed with their standard technique using the 100-keV region. The data were acquired in 4096 channels at 0.25 keV/chan to match the IAEA's standard MCAs. The measurement geometry was as pictured in Fig. VIII-1 but with a planar detector in place of the coaxial detector. These measurements were very difficult for FRAM to analyze. The peaks were weak because of the small detector. Even more important was the low number of chan/keV resulting in very narrow peaks at low energy. The FWHM for the 185-keV ^{235}U peak was less than 3 channels. Measurements were made on enrichments of 0.31, 0.71, 1.94, 4.46, and 10.09 wt. % using steel thicknesses of 13 and 16 mm and count times of 15, 30, and 60 min. Each combination was measured 24 times for a total of 720 measurements.

The results (Sampson 01) from the most difficult measurement (0.31 wt. % ^{235}U for 15 min) are shown in Table IX-2. The % RSD is for a single measurement determined from the 24 replicates. The measurements in Table IX-2 demonstrate that FRAM can perform successfully in as little as 15 min, albeit with quite large uncertainties. Coaxial detector measurements are still preferred as the precision is a factor of two better than that of the planar detector (Fig. IX-1).

Table IX-2.   Twenty-Four 15-min Measurements, Planar HPGe (25 mm x 16 mm)

Sample	Accepted wt% ^{235}U	16-mm Steel Absorber			13-mm Steel Absorber		
		Measured wt% ^{235}U	Meas./Accept.	% RSD	Measured wt% ^{235}U	Meas./Accept.	% RSD
EC-171-	0.3166	0.3131	0.9889	37.4	0.3556	1.1232	27.9
A1-408-1	0.7135	0.7231	1.0135	16.9	0.7100	0.9951	14.1
EC-171-	1.9420	1.9495	1.0039	18.6	1.9968	1.0282	15.7
EC-171-	4.4623	4.5950	1.0297	20.9	4.3954	0.9850	11.9
A1-324-1	10.086	10.093	1.0007	14.9	10.453	1.0364	8.0
		Average	1.0073		Average	1.0336	

2. Lead Shielding

The ability to analyze data acquired through lead shielding was one of the first "difficult applications" demonstrated for FRAM. Numerous examples of FRAM performance on lightly lead-shielded plutonium samples have already been mentioned. Hypes (Hypes 00) demonstrated that FRAM can obtain a complete Pu isotopic analysis through as much as 25 mm of lead. Figure IX-3 displays spectra for lead thickness of 0, 12, and 25 mm.

This requires some parameter file modification because low energy gamma rays "disappear" as the lead thickness increases. The 203- and 208-keV peaks of ^{239}Pu and ^{241}Pu-^{237}U are present and can be analyzed for lead thicknesses up to 6 mm. At 12 mm, we see gamma-ray peaks only above about 300 keV. This still allows analysis for ^{241}Pu using the 330- and 370-keV regions. With 25 mm of lead, the 330-keV region becomes very weak and one must measure ^{241}Pu using the 370-keV region. If the 370-keV region is not available, one cannot measure the ^{241}Pu isotopic fraction as 370.94 keV is the highest energy
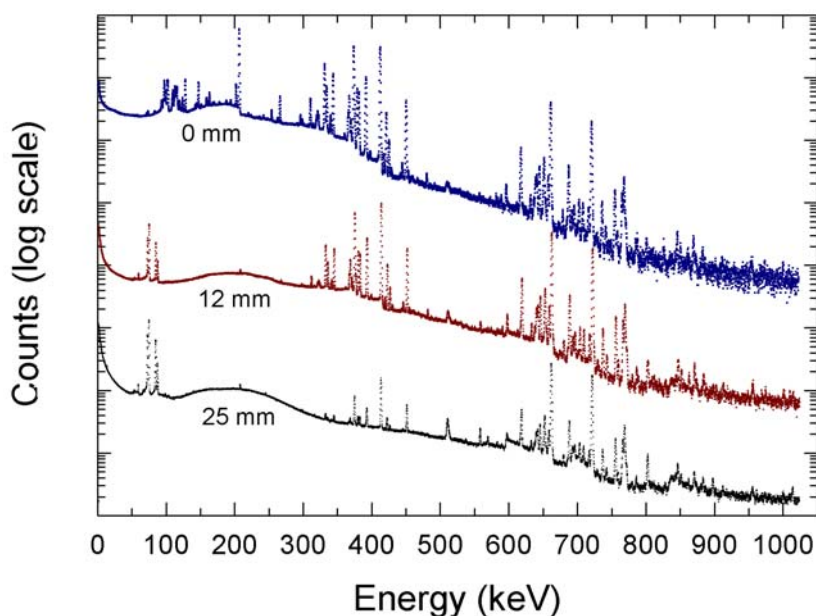

Fig. IX-3. HPGe gamma-ray spectra from a plutonium sample shielded with 0, 12, and 25 mm of lead. The peaks around 75 keV are Pb x rays from the lead shielding in front of the detector.

gamma ray emitted by ^{241}Pu or its ^{237}U daughter. It is also of interest to note the differential attenuation of the 375- and 414-keV gamma rays from ^{239}Pu. They are the most intense peaks in the 375-425 keV region in the plot below. Visually, the peaks are of about equal height with no shielding. The differential attenuation is apparent for 12 mm of lead and is approximately a factor of two for 25 mm of lead.

Note that 148.6 keV is the highest measurable gamma ray directly from ^{241}Pu. All the higher energy gamma rays come from the ^{237}U daughter. This means that ^{241}Pu-^{237}U decay equilibrium (> 45 days from chemical separation) must be present for measurement of ^{241}Pu for any samples shielded with enough lead to remove the 148-keV gamma ray (~ 1 mm of lead).

3. 9975 Shipping Container

The 9975 shipping container is difficult to measure. This container is used by DOE facilities to ship and store plutonium-bearing materials slated for disposition or long-term storage. The drum holds the nominal 13-cm-diameter DOE 3013 plutonium storage container. An interior view of a 9975 is shown in Figure IX-4.

Fig. IX-4. A view of the interior of a 9975 shipping drum showing four different layers of materials surrounding the central cavity. These layers consist of approximately 1.9 cm steel, 1.3 cm lead, and 13 cm of Cellutex.

A coaxial detector gamma-ray spectrum from approximately 4 kg of weapons plutonium in a 9975 is presented in Figure IX-5 below. It is not much different from the spectrum seen through 25 mm of lead shown in Figure IX-3. Data from samples in a 9975 are analyzed with a parameter set that uses gamma rays above 300 keV. The sawtooth structure at energies above 600 keV arises from neutron inelastic scattering from germanium in the detector.
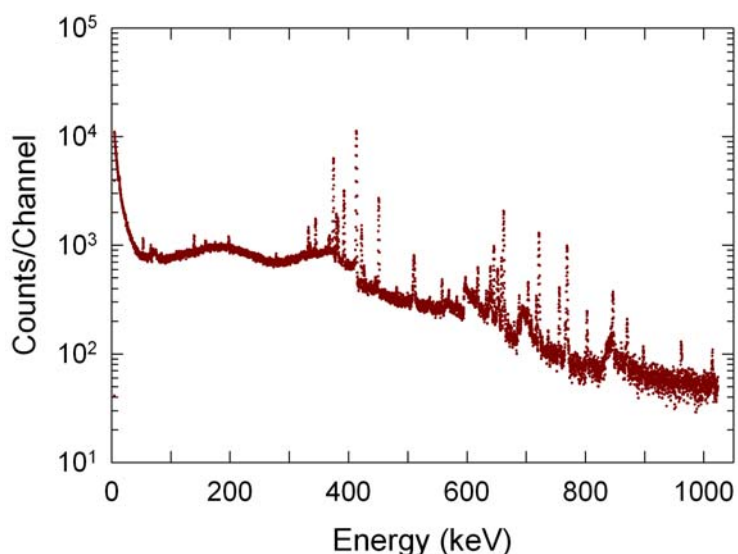

Fig. IX-5. A spectrum taken with a coaxial detector from 4 kg of weapons plutonium inside a 9975 shipping container.

D. Measurements of Am-Be Neutron Sources

FRAM has analyzed AmBe neutron sources for other heat-producing isotopes than [241]Am. These measurements supported calorimetry of the sources prior to disposal. It analyzed [239]Pu, [243]Am, [237]Np, and [239]Np relative to the main heat-producing isotope, [241]Am. Measuring neutron sources is a problem, in the long term, because of potential neutron damage to the HPGe crystal. These measurements did not result in any noticeable neutron damage and successfully demonstrated the ability of FRAM to analyze spectra containing neither plutonium nor uranium.

E. Measuring Samples with [237]Np

Neptunium-237 is present in all plutonium as a decay product of [241]Am and the [237]U daughter of [241]Pu. The level varies as a function of age, burnup (initial [241]Pu content), and chemical processing history. The Los Alamos archival plutonium samples used to characterize FRAM performance have [237]Np/Pu ratios in the range from 10^{-5} to 10^{-3}. The standard parameter files for the 120–450 keV and 200–800 keV regions delivered with FRAM quantify the [237]Np/Pu ratio on every measurement.

We characterize the concentration of ^{237}Np in plutonium by analysis of the gamma rays from its ^{233}Pa daughter ($T_{1/2}$ = 27.0 d). The gamma-ray activity from ^{233}Pa is in secular equilibrium with ^{237}Np after about 150 days. The decay of ^{233}Pa produces intense gamma rays at 271.5, 300.1, 311.9, 340.5, 375.4, 398.5, and 415.8 keV that can be used for the quantification of the ^{237}Np/Pu ratio and are interferences for the plutonium isotopic composition measurement.

For "normal" ^{237}Np/Pu ratios from 10^{-5} to 10^{-3}, we quantify ^{237}Np/Pu using the 312.2-keV peak. We also account for the 375.4- and 415.8-keV peaks because of their interference with prominent ^{239}Pu gamma rays. Plutonium-bearing samples containing ^{237}Np at levels above a ^{237}Np/Pu ratio of about 10^{-2} usually require additional analysis. At this level, the 312-keV line is the most intense gamma ray in the spectrum. The ^{233}Pa gamma rays contribute significantly to the dose from the sample. At Los Alamos, samples containing significant quantities of ^{237}Np are usually packaged in lead-shielded containers to reduce the dose to the sample handlers. The shielding eliminates all gamma rays below 200 keV necessitating analysis in the 200–800 keV region.
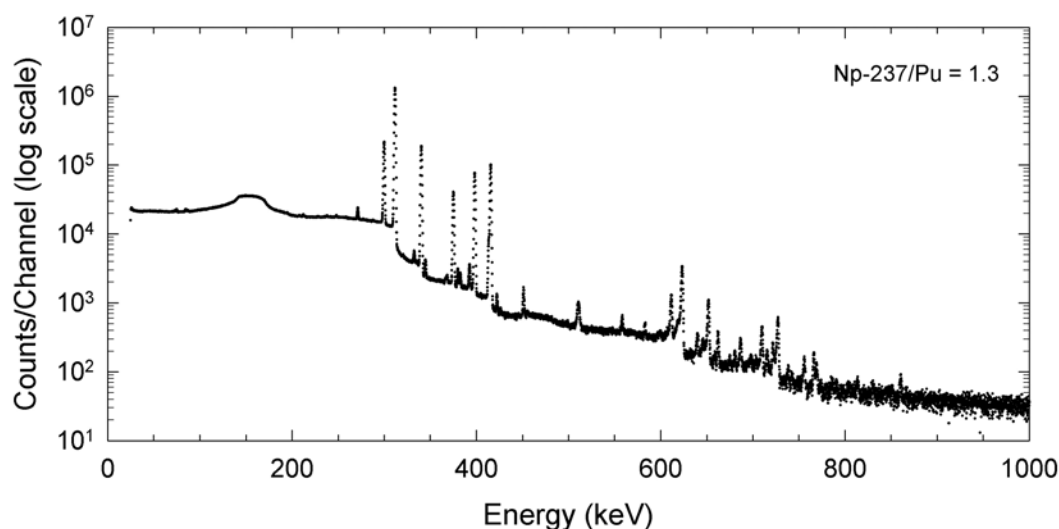

Fig. IX-6. A plutonium spectrum, measured with a coaxial detector, from a shielded sample with a ^{237}Np/Pu ratio of 1.3. This concentration is at the upper limit for neptunium analysis. The six major peaks in the 300–400 keV region are all ^{237}Np related; they overpower the plutonium peaks in the region. The three most intense peaks in the 600–650 keV region are sum peaks from ^{233}Pa gamma rays. The sum peaks also overpower the plutonium gamma rays in this region.

With a suitably modified parameter file this works up to ^{237}Np/Pu ratios of ~ 1 for samples shielded with 3 mm of lead. At a ^{237}Np/Pu ratio of about 1, the ^{233}Pa gamma rays dominate the spectrum making the plutonium analysis very difficult (see Figure IX-6). Sum peaks from the intense ^{233}Pa gamma rays dominate the region around 640 keV. FRAM can analyze spectra from samples shielded with 3 mm of lead with ^{237}Np/Pu ratios up to about 2. With extra lead filtering, we would expect to analyze spectra with ^{237}Np/Pu ratios up to about 10.

F. Measuring Samples with Very High ^{241}Am

The concentration that qualifies as "very high ^{241}Am" is ill defined, but falls in the range of ^{241}Am/Pu above 0.1. This is a convenient dividing line because most samples containing ^{241}Am solely from the decay of ^{241}Pu are below this limit. Americium-241 concentrations above 10% relative to plutonium usually arise only in residues and wastes containing ^{241}Am concentrated from purification processes. While the ^{239}Pu isotopic fraction may change by less than a factor of two over the entire range of samples encountered, the ^{241}Am/Pu ratio may change over a range of 10^6 considering all of the material streams in a plutonium processing facility. The ^{241}Am/Pu

ratio may approach 1:1 in process residues. There are several problem areas for analyzing gamma-ray spectra from samples with very high americium.

1. Americium gamma-rays dominate the spectrum. The peaks remaining from plutonium that haven't been swamped by americium ride on a large continuum from americium gamma rays. The resulting signal/background ratio is poor and the plutonium gamma-ray peaks have poor statistical precision.

2. Americium peaks normally too weak to be visible will suddenly appear in the spectrum. These "new" peaks interfere with the ROIs established for analyses of samples with "normal" americium concentrations.

The second problem can be handled by using a spectrum with very high [241]Am to guide the ROI selection. The first problem is more or less intractable. Items containing [241]Am/Pu above 0.1 are often in lead-lined containers making it necessary to analyze the 200–800 keV region, extracting [240]Pu at 642 keV. Figure IX-7 shows that the analysis for [240]Pu in this region can be difficult to impossible for samples with very high [241]Am because the [241]Am peak at 641.5 keV swamps the [240]Pu peak at 642.6 keV.

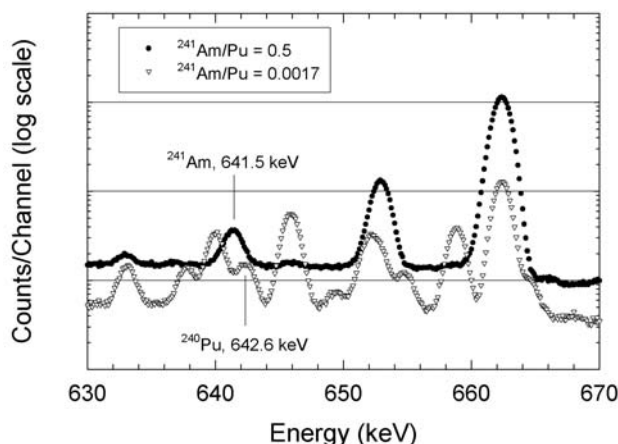

Fig. IX-7. The [240]Pu peak in the 600-keV region is hidden for samples with very high [241]Am. The FRAM analysis (200–800 keV) of the spectrum ([241]Am/Pu = 0.5) produces a result for [240]Pu with a RSD of 30%. The analysis can still be used to interpret a calorimetry measurement because over 95% of the power arises from [241]Am. The RSD(%) for P_{eff} is < 1% for the same analysis.

G. Measuring Heat-Source Grade [238]Pu

FRAM has analyzed gamma-ray spectra from numerous samples of heat-source grade [238]Pu. The first waste shipments to WIPP from Los Alamos contained such items and all were successfully analyzed by FRAM. Such plutonium typically contains about 80% [238]Pu, and 2-4% [240]Pu with the remainder being mostly [239]Pu. Plutonium-241 and [241]Am are also present and can be analyzed. The [236]Pu that is originally present in parts per million (ppm) produces strong gamma-ray peaks from its thorium daughter products. We use these peaks to help define the relative efficiency curve.

We analyze the energy range from 140 keV to 860 keV from a coaxial detector operating in 8192 channels at the standard gain of 0.125 keV/chan. The analysis includes the well-known [238]Pu peaks at 152.7 and 766.4 keV and weaker peaks at 201.0, 742.8, and 786.3 keV. The analysis for [239]Pu and [241]Pu is straightforward using the major high-energy peaks. The usually strong 129.3-keV [239]Pu peak is not visible and the 203.5-keV peak is also not useful. Plutonium-240 is the most difficult isotope to analyze, as its peaks at 160.3 and 642.5 keV are undetectable. This results in very large RSD(%) values for [240]Pu.

X. FRAM APPLICATION WITH CADMIUM TELLURIDE (CDTE) DETECTORS

The application of an unmodified version 4 of FRAM to data taken with a 10-mm x 10-mm x 1.5-mm, Peltier-cooled, CdTe detector is another demonstration of the extreme versatility of

FRAM (Vo 02). This is the first successful application of a general-purpose isotopic analysis code to a non-cryogenic semiconductor detector. Several characteristics of CdTe spectra provide an extreme challenge for FRAM. The energy calibration is nonlinear in contrast to the highly linear calibration from HPGe. This is easily handled by the piecewise, linear calibration in FRAM. The very large tails on the photopeaks (Vo 02) are the greatest limitation to using the unmodified version 4 of FRAM.

Spectra from NaI, CdZnTe, and HPGe are overlaid with a CdTe spectrum for two different isotopic compositions in Figures X-1, -2. Note the region around 203–208 keV. The gamma-ray peaks at 203.5 keV and 208.0 keV are completely resolved for both samples. The peaks are partially resolved by CdTe for the 94% ^{239}Pu sample but the 203.5-keV peak is completely hidden by the tail of the 208-keV peak from the 64% ^{239}Pu sample. This difficult analysis situation contributes to the larger errors seen in the CdTe analysis when compared to the HPGe analysis. Nevertheless, FRAM successfully analyzes the complete isotopic distribution for samples with ^{240}Pu ranging from 3% to 26%.
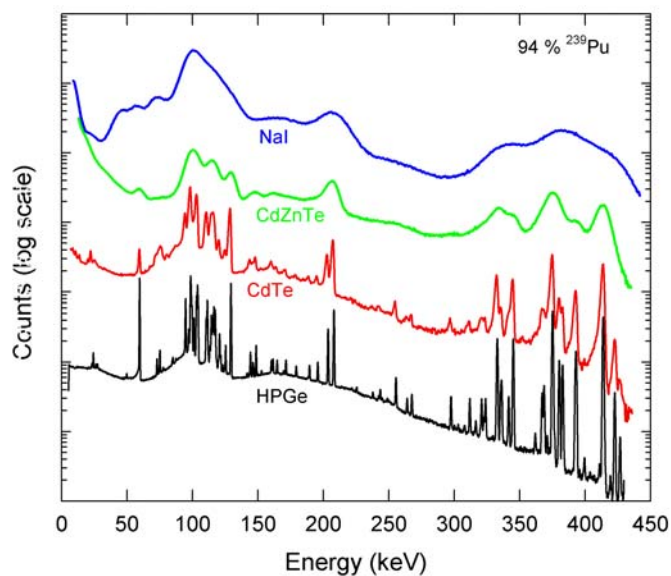

Fig. X-1. Comparison of the gamma-ray spectra from a sample containing 94% ^{239}Pu and 6% ^{240}Pu for four different detectors. Version 4 of FRAM can obtain the complete isotopic distribution from HPGe and CdTe.
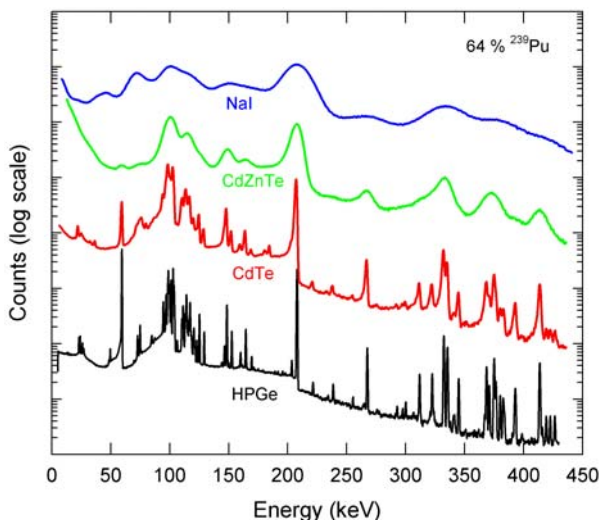

Fig. X-2. Comparison of gamma-ray spectra from a sample containing 64% ^{239}Pu and 26% ^{240}Pu for four different detectors. Version 4 of FRAM can obtain the complete isotopic distribution from HPGe and CdTe.

Table X-1 displays the average bias for measurements using CdTe on standards with ^{240}Pu in the range 3.56 - 26.39 %. The larger bias for CdTe measurements compared to HPGe arises from two sources. First, the counting statistics are poorer because of the small size of the crystal;

over a factor of 500 times smaller in volume than a 25%-relative-efficiency coaxial HPGe detector. The second source is the imperfect fit of the HPGe peak-shape model to the severely tailed CdTe peaks. There is room for significant improvement in the CdTe results by including peak-shape models that are more applicable to CdTe and applying this new analysis to the 100-keV region. Note that FRAM was successfully applied to CdTe spectra, not CdZnTe. While CdZnTe is in more common use, its resolution is too poor to allow analysis of its spectra by an unmodified version of FRAM (see Figures X-1, -2).

Table X-1.   Percent Bias in FRAM Analysis of CdTe Spectra from Reference Standards

Sample	^{240}Pu mass %	Percent Bias = 100 * (Measured – Accepted)/Accepted					
		^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{241}Am	P_{eff}
STDISO	3.56	-30.23	-0.33	8.98	-1.00	-3.06	0.19
PIDIE6_	5.99	-76.71	-0.06	0.97	3.00	-9.33	-2.77
STDISO	6.13	6.32	-0.67	10.24	-1.04	9.25	2.18
CBNM9	6.31	8.69	-0.45	6.67	0.68	-24.23	-1.25
STDISO	6.90	-0.53	1.41	-18.93	-2.74	-0.83	-2.65
STDISO	11.85	-6.31	0.81	-5.79	-3.13	3.25	-0.91
PIDIE6_	14.20	-17.7	-0.37	2.31	-1.32	2.26	0.03
CBNM8	14.27	-10.67	-0.14	1.06	-5.40	-4.95	-1.86
STDISO	15.52	-6.56	0.33	-1.61	-1.08	2.12	-1.07
CBNM7	18.81	-6.99	-3.75	16.38	-6.98	-6.21	-3.65
PIDIE6_	21.41	-15.82	1.99	-6.80	-4.43	-6.59	-5.68
CBNM6	26.29	-4.08	-0.37	1.50	-3.19	-3.66	-2.92
PIDIE6_	26.39	-4.44	1.16	-2.39	-1.64	-0.32	-2.01
	Avg Bias	-12.69	-0.03	0.97	-2.17	-3.25	-1.72
	Std Dev	21.73	1.39	8.86	2.59	8.02	1.96

XI. FRAM APPLICATIONS IN AUTOMATED SYSTEMS

A. ROBOCAL

FRAM is now applied in robotic systems designed to operate continuously and unattended, 24 hours/day. This places an additional burden on FRAM because may not know anything about the type of material being measured. ROBOCAL is robotic system for remote calorimetric and gamma-ray isotopic analysis of special nuclear materials operating at the Los Alamos Plutonium Facility (Hurd 89). ROBOCAL, operating since 1990, has recently been upgraded with new robotics, new computers and software, and a FRAM isotopic analysis system called the Intelligent Isotopic Unit (IIU).

1. Intelligent Isotopic Unit Autoanalysis

The IIU is designed to assay normal samples and pyrochemical residues in shielded or unshielded containers, all without operator intervention. Normally, when a range of sample types is presented to FRAM in a manual mode, the operator must choose an appropriate parameter set for each sample. The sample knowledge may be imperfect and the analysis may have to be repeated if the initial choice was bad.

In FRAM for the IIU, we have incorporated additional logic and diagnostic tests, based on the spectrum under analysis, to choose the best parameter set. The automated logic or intelligence in the modified FRAM for the IIU, automatically chooses from six different parameter sets that accommodate all combinations of unshielded, shielded, homogeneous, or heterogeneous Am/Pu, and very high [241]Am. The IIU acquires data using a 25-30%-efficient coaxial HPGe. The spectrum is first analyzed in the 120–450 keV region assuming it is unshielded. The data is reanalyzed, if necessary, with other parameter sets based on the results of three diagnostic tests.

- Test for Shielded Sample
  This test forms the ratio of the relative efficiency of a high-energy peak to one at low energy. The ratio is tested against an empirical limit. If it is less than the limit, the sample is not shielded, if it is greater than the limit, the sample is shielded. The test is based on the fact that absorbers attenuate low-energy peaks (decreased relative efficiency) more than high-energy ones.
- Test for Am/Pu Heterogeneity
  This test forms the ratio of Am/Pu from a low-energy peak to the ratio from a high-energy peak. If Am and Pu are homogeneous, this ratio should be unity. If Am and Pu are heterogeneous, the ratio will not be unity because of the different attenuation suffered by Am and Pu gamma rays at the same energy. The test also accounts for the uncertainties in the Am/Pu ratios. If Am/Pu heterogeneity is detected, the spectrum is reanalyzed with an Am relative-efficiency curve.
- Test for High Americium
  This test compares the measured Am/Pu fraction against a user-defined limit. A different parameter file is used for high americium because of the additional peaks that are not visible at lower concentrations. A typical user-defined limit for high Am is Am/Pu greater than 0.1.

2. Intelligent Isotopic Unit Hardware

The IIU hardware provides sample rotation and vertical translation. In addition, the detector platform varies the sample-to-detector distance to optimize count rate. The rotation and translation even out the response from heterogeneous samples. Figure XI-1 shows ROBOCAL with a sample about to be placed on the sample table.

Fig. XI-1. The ROBOCAL robot is about to place a container on the sample table of the IIU at the Los Alamos Plutonium Facility

B. ARIES NDA System

ARIES (Advanced Retirement and Integrated Extraction System) is a series of processes designed to extract plutonium metal from retired weapons components, convert it to oxide, and

package it for long-term storage, disposition, or feed for MOX fuel fabrication. The ARIES NDA system consists of three instruments and a robot, under computer control, that nondestructively quantify the plutonium mass in the product oxide containers produced by the ARIES processes. The instruments are a heat-flow calorimeter, an active-passive neutron multiplicity counter, and a FRAM isotopic analysis system. The FRAM system in ARIES uses a 25-mm dia. x 16-mm thick planar HPGe detector and collects and analyzes data in the 120–420 keV region.

   The isotopic analysis system is similar in concept and execution to ROBOCAL. One major departure is that the ARIES NDA system was designed for glove-box operations. While this is no longer applicable, the original requirement is responsible for the size and configuration of the NDA system pictured in Fig. XI-2. A close-up of the FRAM detector is seen in Fig. XI-3.

   Los Alamos is continuing to refine this system and plans to test other measurement regimes using different detectors and energy regions. The data in section VI indicate that other detector-energy region combinations may offer improved precision over the results in Table XI-1.

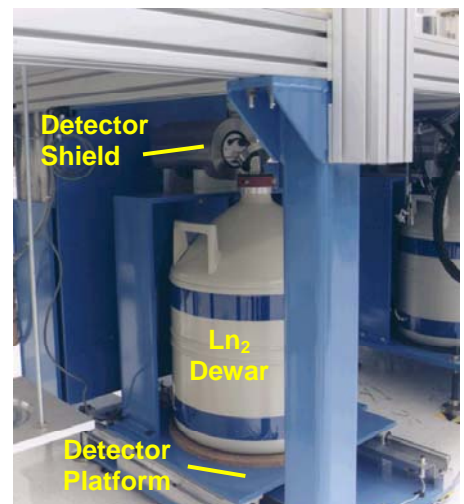*Fig. XI-2. The ARIES NDA system installed at the Los Alamos Plutonium Facility.*

Fig. XI-3. A closeup of the FRAM isotopic analysis station.

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