APPLICATION NOTE

Los Alamos National Laboratory
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FRAM Plutonium Isotopic Analysis System

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Safeguards Assay Group N-1

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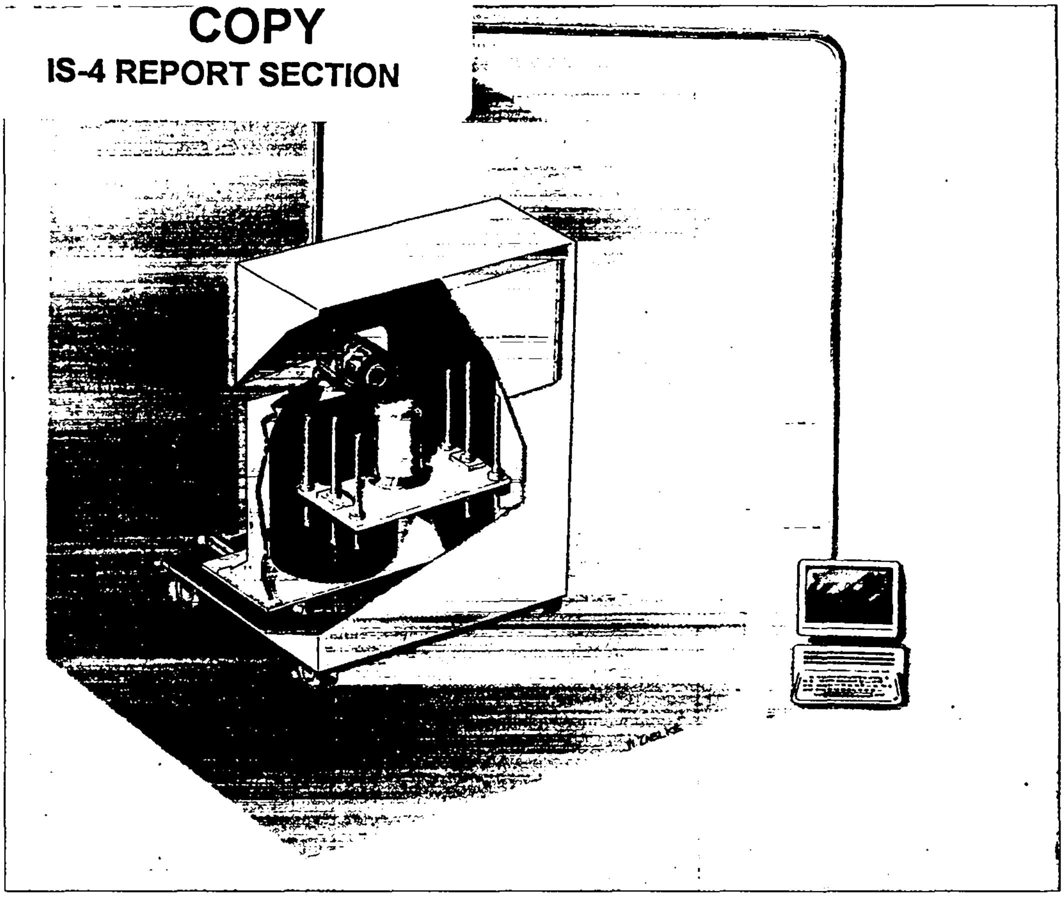

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Figure 1. This cutaway drawing shows the FRAM scanner and connection to the computer. The liquid nitrogen Dewar flask (green) cools the detector (silver) in its shield (brown). The material (gold), shown in the measurement position, is moved up and down and rotated in front of the detector.

FRAM (Fixed-Energy, Response Function Analysis with Multiple Efficiency)* is a code developed by Los Alamos to analyze pulse-height spectra generated by high-resolution gamma-ray detectors. It has been used primarily to determine the isotopic composition of plutonium in special nuclear materials. Its design and flexibility allow it to easily measure ratios and distributions of isotopes other than plutonium in arbitrary items. The measurement technique in FRAM is used world-wide for process control, quality control, and nuclear safeguards in plutonium processing facilities. Figure 1 shows the hardware. FRAM has been used at the Los Alamos Plutonium Facility for about five years. It will soon be in use in the UK. In nuclear safeguards, FRAM complements calorimetry and neutron coincidence counting by allowing measurements to be interpreted in terms of total plutonium mass. With calorimetry, FRAM usually provides the most accurate and precise combination of nondestructive assay methods for measuring the total plutonium in arbitrary bulk items.

***Fram is a word in the Scandinavian languages meaning "forward" or "onward."**

Measurement Principles

In its principal application, FRAM analyzes photopeaks in the spectrum of plutonium gamma rays detected by a high-purity germanium detector. This spectrum contains peaks from the plutonium isotopes $^{238\text{-}241}$Pu, ^{241}Am and often other isotopes such as ^{235}U or ^{237}Np.

FRAM combines this information to produce isotopic ratios independent of sample size, shape, physical and chemical composition, measurement geometry, and container characteristics. The results are obtained using only the spectral data and known fundamental nuclear constants and do not require calibration with standards—a unique feature that makes the measurement technique and software easy to use. A plutonium gamma-ray spectrum from a FRAM measurement system is displayed in Fig. 2. It covers an energy range from 10–420 keV.

The fundamental relationship between the isotopic ratio and the photopeak areas from selected gamma rays from two of the isotopes in the measured item is as follows:

$$\frac{N_1}{N_2} = \frac{C_1}{C_2} * \frac{T_1(1/2)}{T_2(1/2)} * \frac{BR_2}{BR_1} * \frac{RE_2}{RE_1}$$

where N_1/N_2 is the atom number ratio of isotopes 1 and 2 in the sample; C_1 and C_2 are the photopeak areas of the selected gamma rays from isotopes 1 and 2 respectively; the $T(1/2)$ and BR are known nuclear constants—the isotope half-life and the gamma ray branching fraction; and RE is the relative detection efficiency. Because the efficiency enters as a ratio, its value need not be known absolutely and can be conveniently found from the spectral data for each measured item.

The quotient of the measured photopeak area and the branching fraction (BR) for a series of gamma rays from a single isotope, taken as a function of energy, defines the energy dependence of the relative efficiency.

A requirement that the plutonium in the sample has one isotopic composition is usually satisified. However, certain important cases in which the ^{241}Am is not homogeneous with the plutonium can also be analyzed using the "multiple efficiency" characteristics built into the FRAM code. This allows FRAM to greatly improve the results used to interpret calorimetry on elementally (Am/Pu) heterogeneous pyrochemical residues. The multiple efficiency feature of FRAM can also be applied to other types of elementally heterogeneous materials.

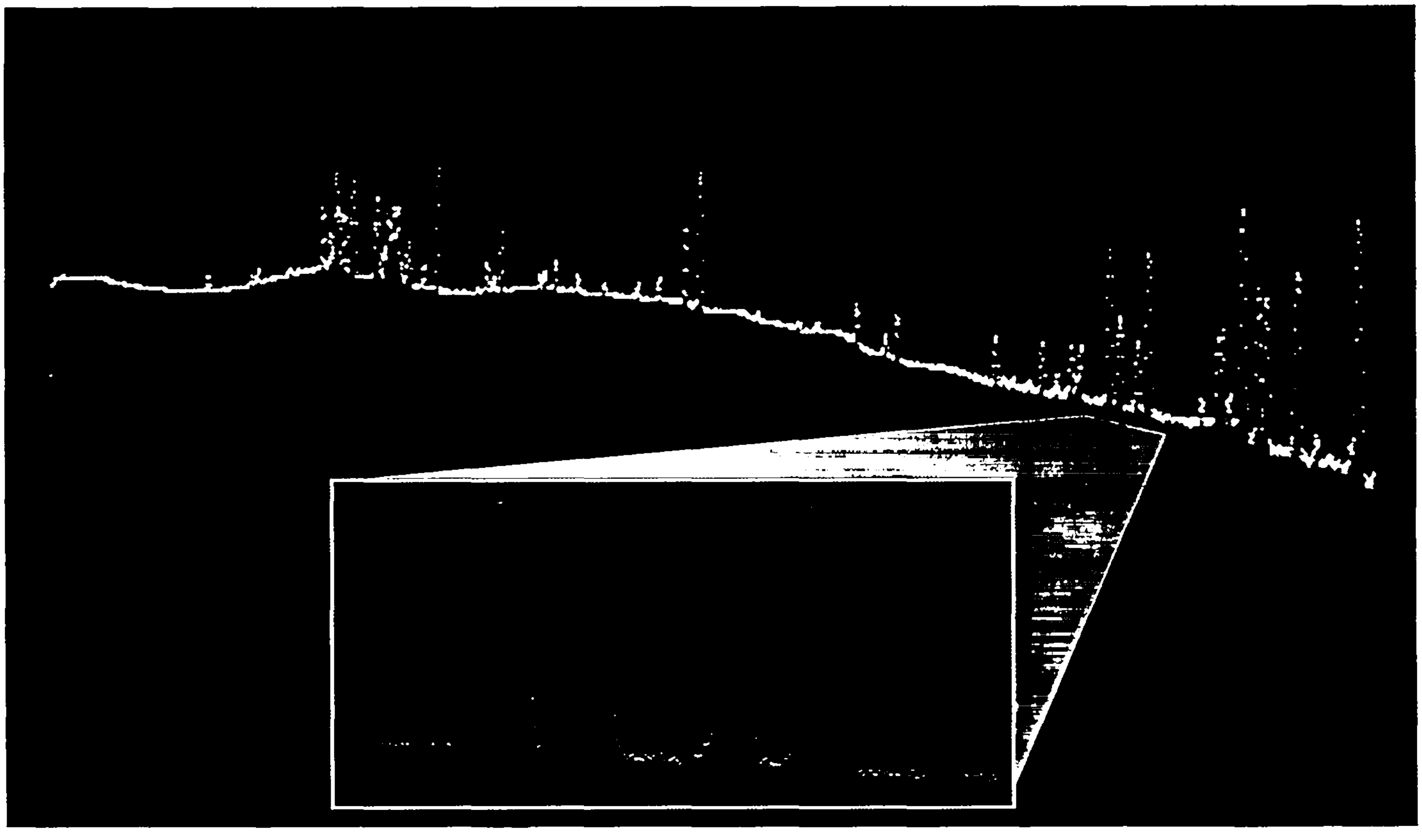

Figure 2. This figure shows a plutonium gamma ray spectrum (top) and an expanded region (bottom) with the FRAM background fit in color. The red arrows denote the portion of the spectrum shown in exploded view.

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The physical and chemical characteristics of the measured item need not be uniform or even well known. Items may contain mixtures of solids and powders with no ill effect as long as the plutonium is isotopically uniform.

Features of the FRAM Code

The FRAM code was developed for a single, high-purity germanium detector system as the simplest, most reliable, and most cost-effective approach for in-plant applications. The code was also designed to be easily adaptable to unusual measurement situations without costly, time-consuming software modifications. It is the only program of its type that can also measure the isotopic ratios in materials that contain only uranium. Table I lists some of the isotopes and concentrations in plutonium-bearing materials that have been successfully measured by FRAM.

Performance

Performance depends upon many variables; some, such as sample mass, isotopic distribution, and geometry cannot be controlled by the user. Others, such as counting time and counting rate, can be selected by the instrument operator.

The isotopic fractions themselves are usually the desired result for quality control and process control measurements. For nuclear safeguards, two additional quantities are most often the end results of the isotopic measurement process. These integral quantities, the effective specific power P_{eff} (W/g Pu) and the effective ^{240}Pu fraction $^{240}Pu_{eff}$, are derived from the measured isotopic fractions and known nuclear constants. They are used to interpret calorimetry measurements (P_{eff}) and neutron coincidence counter measurements ($^{240}Pu_{eff}$) respectively.

Table I. Material Types Measured with FRAM

- ^{240}Pu (2%–30%) of total plutonium
- ^{241}Pu-^{237}U (nonequilibrium)
- Am/Pu in heterogeneous combinations
- ^{241}Am (0.01% to 50%)
- U/Pu mixed (MOX)
- ^{243}Am-^{239}Np
- ^{244}Cm
- ^{237}Np
- ^{238}U/^{235}U (no plutonium) present
- ^{238}Pu (80%)
- ^{242}Pu (90%)

Over a wide range of measurement and sample conditions, FRAM performance falls into the ranges shown in Table II.

Table II. Typical Ranges of FRAM Measurement Characteristics

Measurement Time	20 min to 1 hr
Sample Size	100 mg to many kgs (limited by criticality)
Precision (relative standard deviation)	
P_{eff}	0.2% to 0.5%
$^{240}Pu_{eff}$	0.8% to 3.0%
Bias for isotopically homogeneous samples	
P_{eff}	<0.3%
$^{240}Pu_{eff}$	<1.0%
Individual isotopes (^{238}Pu to ^{241}Pu)	<1.0%

Measurement Hardware

While the term FRAM strictly describes the analysis software, this software must be accompanied by a detector and data acquisition hardware to provide a complete measurement system. The electronics used for data acquisition are conventional, commercially available nuclear instrumentation modules and consist of a linear amplifier, analog-to-digital converter, digital stabilizer, and a separate computer-based (or computer-controlled) multichannel pulse-height analyzer. The high-resolution, high-purity germanium detector, required for measurements, is also available from several commercial sources. These measurements *cannot* be performed with low-resolution detectors such as sodium iodide (NaI).

The measurement systems used with the FRAM software are computer-based and compatible with popular mini-computer and personal computer hardware. Some systems may include simple scanning mechanisms to rotate and translate the item in front of the shielded detector. Figure 3 shows a system that incorporates all these features. The nature of the measurement process, not requiring calibration or standards, makes sample presentation very flexible. Sample positioning need not be reproducible or fixed.

Figure 3. A FRAM system showing the scan mechanism (left), the data acquisition electronics and computer (center), and console and printer (right).

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Additional Sources of Information

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