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*Using the FRAM Plutonium Isotopic
System for ^{242}Pu Isotopic Verification*

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USING THE FRAM PLUTONIUM ISOTOPIC SYSTEM FOR ^{242}Pu ISOTOPIC VERIFICATION

by

Thomas E. Sampson

ABSTRACT

This report describes how to use the FRAM plutonium isotopic system to verify the isotopic composition of plutonium with very high percentages of ^{242}Pu .

I. INTRODUCTION

Plutonium highly enriched with ^{242}Pu is a valuable material. It is important, to preserve the research value of this material, that this material not be contaminated with other grades of plutonium.

A ^{242}Pu Verification Station¹ using gamma-ray spectroscopy techniques has been used for the past 10 years to assure the isotopic integrity of these high ^{242}Pu materials. This station measures the unique isotopic ratios of $^{238}\text{Pu}/^{241}\text{Pu}$ and $^{239}\text{Pu}/^{241}\text{Pu}$ that are characteristic of these materials. The simple algorithms used in the analysis of the gamma-ray spectroscopy data are adequate for material with low concentrations of ^{241}Am (below a few thousand ppm), but do not give quality results for samples with higher americium concentrations. Most of the samples that are difficult to measure are reprocessing residues and materials that have been in storage for several years. The measurement difficulties arise from closely spaced americium interferences that the simple region-of-interest (ROI) peak summation techniques cannot adequately treat. The FRAM plutonium isotopic code²

employs more sophisticated peak analysis algorithms that can eliminate these problems. This report briefly describes the use of the FRAM code for the analysis of these materials.

II. THE FRAM PLUTONIUM ISOTOPIC CODE

This versatile code is designed to measure the ratio of isotopic fractions present in plutonium-bearing materials. For plutonium with "normal" isotopic distributions, approximately 3-20% ^{240}Pu , values are calculated for the ratios of ^{238}Pu , ^{240}Pu , and ^{241}Pu to ^{239}Pu . For high ^{242}Pu materials, the gamma rays from ^{241}Pu and its ^{237}U daughter are the most prominent peaks in the spectrum; it is therefore most desirable to measure the isotopic ratios of ^{238}Pu , ^{239}Pu , and ^{240}Pu to ^{241}Pu .

Response function fitting allows for proper analysis of peak complexes containing ^{241}Am , the interference that presents difficulties in the old code. The FRAM code can analyze data from samples with an americium content exceeding several percent (upper limit unknown). The old code with its ROI summation technique could not do this primarily because the americium peak at 376.6 keV interfered with the weak, but critically important, 375-keV ^{239}Pu peak.

FRAM also allows direct input of the ^{242}Pu percentage, if known (^{242}Pu cannot be directly measured with gamma-ray techniques because its gamma rays are too weak). With this option, FRAM calculates the complete isotopic distribution and thus one can observe the magnitude and distribution of all the isotopic fractions. The comparison of the isotopic fractions with known stream averages or absolute limits set from experimental requirements will tell the user if the material is acceptable for experimental purposes. The comparison also will show if the batch has been contaminated with any other grades of plutonium. The direct comparison of isotopic fractions is more desirable than just comparing isotopic ratios, as done in the old code. But there is a limitation; if ^{242}Pu is entered directly, its value must be accurate. An incorrect value will produce large errors in the minor isotopes, the content of the minor isotope ^{239}Pu being extremely important in many cases. An error of only 1% (absolute) in the ^{242}Pu percentage will yield fractional errors in the ^{239}Pu content ranging from 6% to 22%, depending on the ^{242}Pu percentage. This is shown in Fig. 1. However, it is very important to note that the incorrect entry of the ^{242}Pu percentage does not cause a corresponding error in the measured values of the isotopic ratios, that is, $^{239}\text{Pu}/^{241}\text{Pu}$, $^{238}\text{Pu}/^{241}\text{Pu}$, etc. These ratios are independent of the operator-entered ^{242}Pu value and will be as accurate as the basic measurement technique will allow.

III. PARAMETER FILE FEATURES

The parameter file for ^{242}Pu analysis contains the major peaks from ^{241}Pu and ^{241}Pu - ^{237}U at 148, 164, 208, 267, 332, 335, 368, and 370 keV. It uses the 152-keV peak for ^{238}Pu and the 160.3-keV peak for ^{240}Pu . Most of the major ^{239}Pu peaks are also in the parameter file, but only the 375-keV peak is an important contributor because of the low ^{239}Pu content of this material. For example, the usually prominent 129-keV ^{239}Pu peak is barely visible in most spectra of these materials. Americium-241 peaks are also included if they interfere with other peaks.

The relative efficiency is defined with ^{241}Pu and ^{241}Pu - ^{237}U peaks, so the analysis is only valid for equilibrium ^{241}Pu - ^{237}U materials. Peak shapes are calibrated with the major peaks at 148, 152, 164, 208, 267, and 332 keV. With more normal plutonium materials, the 332 complex would contain peaks at 332.4 keV from ^{241}Pu - ^{237}U and 332.8 keV from ^{239}Pu . However, for these materials with low ^{239}Pu , in the worst case, the 332.8-keV ^{239}Pu peak area is $<0.5\%$ of the ^{241}Pu - ^{237}U peak at 332.4 keV, so the 332-keV "complex" is essentially a clean single peak.

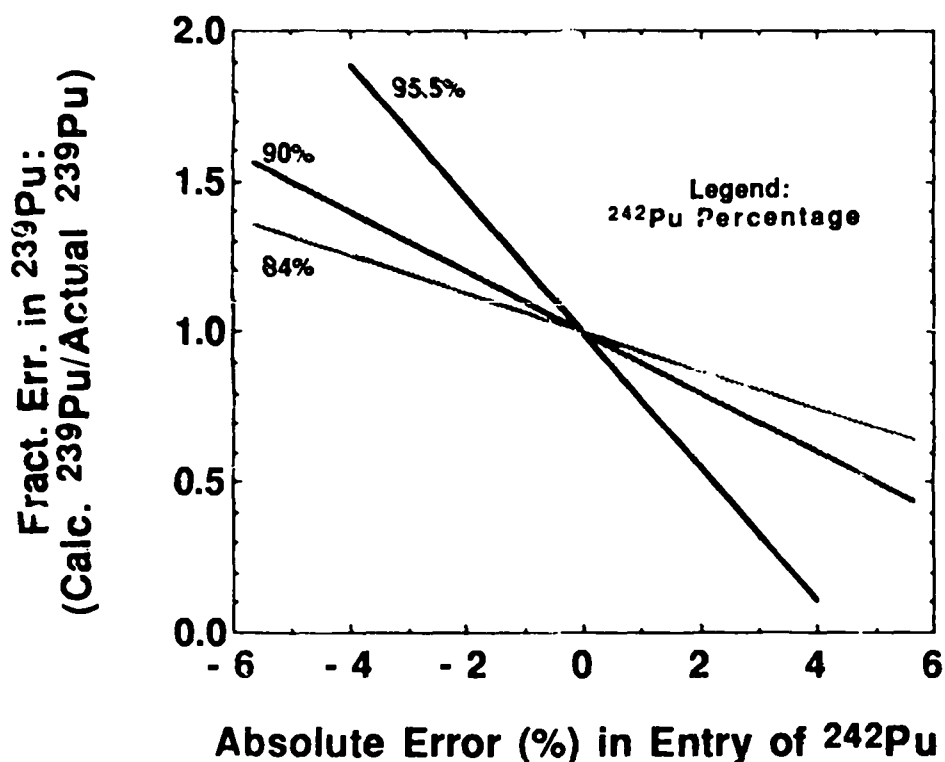


Fig. 1. Error in ^{239}Pu fraction resulting from an error in the entry of ^{242}Pu fraction.

Energy calibration, as well as relative efficiency fitting, is done from 148 keV to 370 keV. It is felt that the extrapolation of these functions to 375 keV can be done with little error.

IV. MEASUREMENT SETUP

Measurement setup is similar to that for normal FRAM plutonium isotopic measurements. However, there are a few additional steps that must be taken before starting a measurement on a ^{242}Pu sample. These steps are listed below. The description assumes that the user is familiar with the routine operation of the FRAM code.

- (a) Use the parameter file named PU242.
- (b) Change digital stabilizer settings to the following: gain peak = 3224, zero peak = 1386. Be sure to change stabilizer settings back to normal after completing the ^{242}Pu measurements.
- (c) Set printout to level 4 (Supervisor menu, Change parameters, Change defaults).

Do not change any other analysis parameters or diagnostic parameters. You will probably also wish to change back to level 5 after completing the measurement.

- (d) Manually enter the ^{242}Pu percentage. Do not use the correlation. You may interpret the results using the final isotopic percentages or the intermediate isotopic ratios, depending upon your knowledge of the accuracy of the entered ^{242}Pu percentage.

V. MEASUREMENT RESULTS

A limited number of proof-of-principle measurements have been done on samples with ^{242}Pu fractions of 84% and 95%. In Table I, we compare the measured with the accepted values for the isotopic composition of two samples. In both cases in Table I, the ^{242}Pu percentage was known and was entered by the operator.

VI. MEASUREMENT INTERPRETATION

The first step in interpreting the measurement results is to look at the summary isotopics. If you are confident of the accuracy of the ^{242}Pu entry, then check the absolute magnitude of the minor isotopes for acceptable values. If you are unsure about the ^{242}Pu entry, recognize that the absolute

TABLE I. Measurement Results

| | ²³⁸ Pu (wt %) | ²³⁹ Pu (wt %) | ²⁴⁰ Pu (wt %) | ²⁴¹ Pu (wt %) | ²⁴² Pu (wt %) | ²⁴¹ Pu (μg/g Pu) | P _{eff} (mW/g Pu) |
|-----------|-----------------------------|-----------------------------|-----------------------------|-----------------------------|-----------------------------|--------------------------------|-------------------------------|
| Measured: | 0.4523(7) | 0.4369(70) | 2.403(41) | 0.9137(10) | 95.794 ^a | 5 404(14) | 3.5052(51) |
| Accepted: | 0.4500 | 0.4364 | 2.401 | 0.9185 | 95.794 | 5 525 | 3.5057 |
| Measured: | 1.054(3) | 1.332(25) | 10.36(16) | 3.252(6) | 83.999 ^a | 19 672(73) | 9.197(20) |
| Accepted: | 1.035 | 1.389 | 10.34 | 3.214 | 83.999 | 19 856 | 9.106 |

^aOperator entry

fractions of the minor isotopes may have large errors, but the isotopic ratios should be accurate. Check the isotopic mass ratios on the fifth page of the level 4 medium output. Look particularly at the ²³⁹Pu/²⁴¹Pu and ²³⁸Pu/²⁴¹Pu isotopic ratios. A typical output is shown in Table II. These are the same ratios that are measured and checked with the old program. These values should fall into the same ranges as used for testing the results of the old ²⁴²Pu Verification Station.

TABLE II. Isotopic Ratio Output for Relative Activity Least-Squares Fit

| RESULTS OF RELATIVE ACTIVITY CALCULATIONS | | | |
|---|----------------|-------------|---------------|
| Results of relative activity least-squares fit: | | | |
| ISOTOPIC RATIOS | ACTIVITY RATIO | MASS RATIO | PERCENT ERROR |
| PU-241/PU-241: | 9.99960E-01 | 9.99960E-01 | 0.03% |
| PU-239/PU-241: | 2.44135E-04 | 4.06978E-01 | 1.94% |
| AM-241/PU-241: | 2.00289E-02 | 6.05278E-01 | 0.25% |
| PU-238/PU-241: | 5.36753E-02 | 3.24137E-01 | 0.16% |
| PU-240/PU-241: | 7.00926E-03 | 3.19329E+00 | 1.69% |
| Reduced chi-square = | | 9.269E+00 | |

If you believe that your operator-entered ^{242}Pu value is accurate, perhaps obtained from the paperwork on the sample container, consider how old that value may be. The relatively high ^{241}Pu content of these materials and the short 14.35-yr half-life of ^{241}Pu couple with the normalization condition governing plutonium isotopic composition decay corrections to produce a significant change in the ^{242}Pu isotopic fraction with time. Two examples illustrate this. Nominal 84% ^{242}Pu material that has decayed for 5.75 years can typically have its ^{242}Pu fraction changed from 84% to 84.7%. This alone will produce a 4% (relative) error in the ^{239}Pu fraction if the old 84% value is entered rather than the current 84.7% value. A 95.8% ^{242}Pu sample that has decayed for 5.75 years will have a ^{242}Pu fraction of 96.0%. This will also produce about a 4% (relative) error in the ^{239}Pu fraction. In the first case, the total plutonium mass in the sample will have decreased by over 0.8%, probably requiring a decay correction update in the accountability system.

REFERENCES

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