*Plutonium Isotopic Abundance
Measurements on CBNM NRM 271
Analyzed with the FRAM and MGA Codes*

Los Alamos

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PLUTONIUM ISOTOPIC ABUNDANCE MEASUREMENTS ON CBNM NRM 271 ANALYZED WITH THE FRAM AND MGA CODES

by

Robert J. Friar and Thomas E. Sampson

ABSTRACT

We report results of gamma-ray spectrometry measurements of the isotopic distributions of plutonium in the reference-material set CBNM NRM 271 as analyzed by the FRAM and MGA plutonium isotopic codes. We acquired high-quality spectral data under measurement conditions approximating field-use conditions recommended by the code developers. Bias and precision results from these measurements are presented for both codes. Both codes performed very well for these measurements. These standards have proven to be very useful for testing the Los Alamos FRAM code in the high-burnup region where well-characterized materials have been unavailable at Los Alamos.

I. INTRODUCTION

On May 17, 1991, the Safeguards Assay Group (N-1) at Los Alamos National Laboratory received set O/8 of the Central Bureau for Nuclear Measurements Certified Nuclear Reference Material 271. The Central Bureau of Nuclear Measurements in Geel, Belgium is one of the Joint Research Centres of the Commission of the European Communities and is well known for its mass spectrometry program. The isotopic distributions of the set of four samples, CBNM Pu93, CBNM Pu84, CBNM Pu70, CBNM Pu61, have been very accurately determined making them ideal for checking the accuracy of gamma-ray spectrometry codes for plutonium isotopic composition analysis. We used the samples to characterize both the accuracy and precision of FRAM,¹ a Los Alamos plutonium isotopics code, and MGA,² a Lawrence Livermore plutonium isotopics code. In addition, the high ²⁴⁰Pu abundance of Pu70 and Pu61 allowed us to characterize FRAM performance in a high-burnup range previously untested at Los Alamos with high-quality standards.

II. THE CBNM STANDARDS

The CBNM samples consist of a plutonium oxide pellet sealed in a stainless steel can. The pellet contains approximately 6.6 g of sintered PuO_2 and has nominal dimensions of 14.7 mm in diam by 3.7 mm thick. The bottom of the can has a 0.780-mm-thick window over the pellet diameter to minimize attenuation for measurements viewing the sample from the bottom. This window is protected by a removable plastic cover that prevents damage during handling. Figure 1 shows a diagram of the can with external dimensions in millimeters.

Fig. 1. CBNM nuclear reference material 271.

CBNM determined the isotopic distribution of the plutonium by thermal ionization mass spectrometry after first bringing the sample into solution to ensure isotopic homogeneity. The ^{241}Am concentration was measured by a combination of calculated ^{241}Pu decay from the time of chemical separation and the spectrometry of alpha particles and low-energy gamma rays on liquid samples. The isotopic homogeneity of the finished samples was verified by high-resolution gamma-ray spectrometry. The isotopic distributions for the four samples, updated to June 15, 1991, are shown in Table I with absolute uncertainties at the 95% confidence level. The absolute uncertainties are those quoted on the original certificate for June 20, 1986, except for ^{241}Pu and ^{241}Am that are assigned the same *relative* uncertainty as on the certificate.

III. MEASUREMENTS

The primary purpose of the measurements was to provide data from additional well-characterized materials that would allow us to adjust and fine tune the FRAM analysis parameters in routine use at the Los Alamos Plutonium Facility. A secondary purpose was to characterize the performance of both FRAM and MGA on these samples. We purposely did not make these measurements an identical side-by-side comparison. The acquisition conditions, count rates, count times,

TABLE I. CBNM NRM 271 Isotopic Composition on June 15, 1991

		Mass Percent Relative to Total Plutonium (June 15, 1991)					
		^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	^{241}Am
CBNM Pu93		0.0113	93.4596	6.3139	0.1758	0.0395	0.1516
95% confidence		0.0002	0.0040	0.0039	0.0003	0.0003	0.0030
CBNM Pu84		0.0678	84.5304	14.2339	0.8095	0.3585	0.4357
95% confidence		0.0006	0.0084	0.0085	0.0014	0.0010	0.0044
CBNM Pu70		0.8232	74.2157	18.5111	4.3471	2.1029	2.3546
95% confidence		0.0018	0.0098	0.0087	0.0027	0.0023	0.0235
CBNM Pu61		1.1682	63.4675	25.7787	5.3295	4.2562	2.9009
95% confidence		0.0025	0.0283	0.0241	0.0069	0.0064	0.0289

and filters were independently selected for each code to approximate field-use conditions suggested by the developer. These conditions were not the same for the two codes because of the very different spectral features emphasized in the respective analysis algorithms of the codes. The field-use conditions approximated those in United States facilities for accountability measurements involving the combination of calorimetry and gamma-ray plutonium isotopic measurements. These conditions were not intended to represent inspection measurements such as those that might be performed by European Atomic Energy Commission or the International Atomic Energy Agency.

Many of the same electronic components were used in acquiring data for both codes. These components are listed in Table II.

We used a single high-quality planar detector for all acquisitions except those described in Sec. VI. Along with its near perfect peak shape, it exhibited a maximum count-rate capacity (at Los Alamos's elevation of 2190 m) of about 70 kHz for ^{57}Co and resolution characteristics as shown below in Table III.

TABLE II. Principal System Components

Detector	Canberra LEGe, 16-mm diam by 13 mm deep aluminum end cap. Model GL0213S
Resolution	See Table III
MCA	Canberra Series 90
Digital Stabilizer	Canberra 8232
ADC	Canberra 8077
NIM Bin	Built into Series 90
Bias Supply	Ortec 459
Amplifier	Ortec 672
Count Rate Monitor	Canberra 2071
Oscilloscope	Tektronix T922R (FRAM), Tektronix 2225 (MGA)
Computer	MicroVax II (FRAM), IBM Industrial 7561 (MGA)
FRAM version	2.1
MGA version	7.4

TABLE III. Resolution Characteristics of Planar Detector

Time Constant (μ s)	Count Rate (khz)	FWHM at 122 keV (eV)
6	1	512.3
6	1	511.9
3	1	513.3
2	50	526.4
1	50	558.8

The measurement conditions for each of the two acquisitions for each analysis code are shown in Table IV. The letters after the name of the code signify the order in which the data were acquired. These letters are also used for identification in tables and plots presented later in this report. All four sets of acquisitions consisted of a 20 run auto-cycle for each sample with the single exception that FRAM B for CBNM Pu93 had 25 runs. The decay-corrected certified value of ^{242}Pu was entered by the operator for all measurements. FRAM data were collected over an energy range of 10 to 420 keV at a gain of 0.1 keV/channel while MGA data were acquired from 0 to 307 keV at 0.075 keV/channel—both standard operating conditions for the codes.

TABLE IV. Measurement Conditions

	FRAM B	FRAME	MGA C	MGA D
Shaping time constant (μ s)	1	1	3	2
Count Rate (kHz) CBNM 93	10	11	10	14
Spectrum Integral	6.83 E06	2.96 E06	2.68 E07	3.18 E07
Count Rate (kHz) CBNM 84	10	15	10	20
Spectrum Integral	6.91 E06	4.37 E06	2.72 E07	3.81 E07
Count Rate (kHz) CBNM 70	25	30	10	20
Spectrum Integral	1.42 E07	7.39 E06	2.79 E07	3.88 E07
Count Rate (kHz) CBNM 61	25	30	10	20
Spectrum Integral	1.34 E07	7.82 E06	2.76 E07	3.90 E07
Count Time [True Time (s)]	3600	1800	1670 1500 s (live)	1755 1500 s (live)
Filters	1.9 mm Cd 0.38 mm Cu	1.9 mm Cd 0.38 mm Cu	1.14 mm Cd 0.38 mm Cu	1.14 mm Cd 0.38 mm Cu

The filters for the FRAM acquisitions were chosen to reduce the 59.5-keV ^{241}Am peak and the 100-keV x-ray region sufficiently to eliminate sum peaks in the 150- to 170-keV region. Because filters are not optimized for each sample in routine field use, we chose the same filter conditions for all samples to approximate worst case field conditions.

Filter conditions for MGA were different from those of FRAM to emphasize the 100-keV region, where MGA obtains most of its results. Also, because MGA requires the 59.5-keV peak in its analysis, we used filtering only to reduce deadtime from this peak and remove lower energy x rays and gamma rays.

We chose the shaping time of 1 μ s for both sets of FRAM data to duplicate field conditions. For MGA, we initially acquired data at 10 kHz with a shaping time of 3 μ s, but after discussions with the developer of MGA, we made a second set of acquisitions at 20 kHz and 2 μ s to further enhance MGA's precision.

Typical field measurement conditions for FRAM use 1800-s to 3600-s (true time) acquisitions so we ran all the CBNM samples for both times. We also wished to acquire MGA data sets for

1800 s of true time. Unfortunately, the MGA data acquisition system only allowed us to preset the live time. We set a live time of 1500 s that gave about 1700 s of true time, closely approximating the 1800-s true time often used in MGA field measurements.

The samples were small enough (~6.6 g plutonium) that we could not always attain the target count rate for each sample on each measurement series. Target count rate is loosely defined as the rate you would use if there were no rate-restricting limitations. This was especially true for samples 93 and 84. The target count rate was 25 kHz for FRAM B, 40 kHz for FRAM E, 10 kHz for MGA C, and 20 kHz for MGA D. The 25 kHz for FRAM B represents a “typical” fairly high count rate; the 40 kHz for FRAM E is near the upper end of the recommended count-rate range for FRAM.

The measurement conditions for the two systems were purposely not identical because we wanted to approximate field conditions for each code. The MGA time constant and count rate were chosen to emphasize resolution, while the FRAM set-up emphasizes throughput. Nevertheless, the throughput was driven by the lighter filtering used for MGA that allowed the 100-keV region to dominate the count rate; total throughput (spectrum integral in Table IV) was greater for all MGA acquisitions than for any of the FRAM runs. The higher energy region (>120 keV) emphasized in the FRAM analysis is inherently less intense than the lower energy region used in MGA. The FRAM throughput was also lower for some acquisitions because the small sample size would not allow the target count rate to be achieved.

IV. ANALYSIS

We analyzed the FRAM data, FRAM B and FRAM E, with two sets of analysis parameters. Analysis parameters are described in Ref. 1 and govern all facets of the analysis physics of the code. The first analysis was done with a general purpose parameter file developed for a wide range of isotopic compositions. This file, denoted as WIDERANGE6, has been in routine use at the Los Alamos Plutonium Facility for several years. The results from analyzing the FRAM B and FRAM E data with the WIDERANGE6 parameters are shown in Table V. Each entry in Table V is the ratio of the average measured value, averaged over the 20 repeat runs as documented in Table IV, to the CBNM certified value, decay corrected to the measurement date. P_{eff} is the effective specific power for calorimetry interpretation while $^{240}\text{Pu}_{\text{eff}}$ is the effective ^{240}Pu fraction for neutron coincidence counting applications. The Average Ratio in the table is the average of the eight average ratios for the four samples-two acquisitions displayed above in the table.

These results appear to be independent of isotopic composition, a very desirable conclusion. The small biases that do appear are approximately the same for each sample and approximately the same for the two independent acquisitions. We do not display the statistical uncertainties of the individual ratios; they may be inferred from the precision plots presented later in this report. This

TABLE V. FRAM B and FRAM E WIDERANGE6 Parameters

	Ratio: (Average Measured*/Certified)						
	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{241}Am	P_{eff}	$^{240}\text{Pu}_{\text{eff}}$
CBNM Pu 61B	1.0171	0.9963	1.0080	1.0014	0.9854	1.0057	1.0072
CBNM Pu 70B	1.0184	0.9996	1.0001	1.0022	0.9865	1.0049	1.0017
CBNM Pu 84B	1.0066	0.9982	1.0111	0.9968	0.9922	1.0019	1.0107
CBNM Pu 93B	1.0212	0.9997	1.0043	0.9963	0.9817	0.9998	1.0044
CBNM Pu 61E	1.0220	0.9986	1.0013	1.0051	0.9873	1.0080	1.0028
CBNM Pu 70E	1.0210	0.9987	1.0033	1.0039	0.9842	1.0057	1.0043
CBNM Pu 84E	0.9931	0.9990	1.0064	0.9971	0.9927	0.9995	1.0060
CBNM Pu 93E	1.0513	1.0004	0.9947	0.9962	0.9862	0.9996	0.9950
Average Ratio	1.0188	0.9988	1.0037	0.9999	0.9870	1.0032	1.0040
Standard Deviation	0.0164	0.0012	0.0051	0.0037	0.0037	0.0033	0.0046
*Average of 20 repeated runs (25 runs for series 93B).							

precision data may also be used to assess the significance of the observed biases. We do observe small and probably statistically significant biases of 1.9% for ^{238}Pu , 0.4% for ^{240}Pu (may not be significant), and 1.3% for ^{241}Am . The biases in P_{eff} and $^{240}\text{Pu}_{\text{eff}}$, of course, are not independent and reflect the biases in the individual isotopic fractions.

Because the principal goal of this work was to provide better analysis parameters for the FRAM code, we attempted to adjust some of the critical FRAM parameters to remove the average biases noted above. This small adjustment of branching ratios is a common technique in this general area of gamma-ray analysis. We realize that the branching ratios are constants of nature; however, the adjustments compensate for uncertainties in their measurement and other small biases that arise in the very complex analysis in these types of codes. The adjustment for ^{238}Pu was straightforward; a simple 1.9% change in the ^{238}Pu 152.7-keV branching ratio successfully removed this average bias. Note that we use the term “average bias,” averaged over all four samples for two independent acquisitions. There is evidence in Table V and additional evidence from the MGA results presented later that the ^{238}Pu measurements on CBNM Pu84 are inconsistent with the other three samples. Nevertheless, the Pu84 data were used with the same weight as the other three samples. For ^{240}Pu we attempted a least squares adjustment of all the branching ratios in the 160-keV region. This adjustment was not successful—it did not improve the results—because there

were data from only four independent samples. However, a 0.37% adjustment of the ^{240}Pu 160.3-keV branching ratio successfully removed this average bias. The adjustment for ^{241}Am worked out similarly to that for ^{240}Pu , that is, a least squares adjustment to the 125-keV region did not work but a 1.3% adjustment of the 125.3-keV ^{241}Am branching ratio did remove the average bias.

The results of this bias adjustment are shown in Table VI where the parameter file WRTEST identifies the adjusted parameters. All average biases are now seen to be less than 0.1%. Unless otherwise noted, FRAM results using the adjusted parameters will be used in the remainder of this report.

In Table VII below we display the results of the two series of measurements with MGA. These show only slightly more bias than the adjusted FRAM results in Table VII. We did not adjust MGA parameters because this software is not supported by Los Alamos.

TABLE VI. FRAM B and FRAME Analyzed With WRTEST Adjusted Parameters

	Ratio: (Average Measured*/Certified)						
	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{241}Am	P_{eff}	$^{240}\text{Pu}_{\text{eff}}$
CBNM Pu 61B	0.9996	0.9974	1.0059	1.0025	0.9984	1.0000	1.0043
CBNM Pu 70B	1.0005	1.0004	0.9977	1.0030	0.9993	0.9999	0.9983
CBNM Pu 84B	0.9894	0.9987	1.0080	0.9971	1.0053	1.0011	1.0075
CBNM Pu 93B	1.0021	1.0000	1.0008	0.9961	0.9950	0.9998	1.0008
CBNM Pu 61E	1.0044	0.9997	0.9993	1.0063	1.0004	1.0023	0.9998
CBNM Pu 70E	1.0030	0.9995	1.0009	1.0047	0.9969	1.0007	1.0009
CBNM Pu 84E	0.9752	0.9995	1.0033	0.9975	1.0057	0.9988	1.0029
CBNM Pu 93E	1.0318	1.0006	0.9912	0.9960	0.9995	0.9996	0.9914
Average Ratio	1.0008	0.9995	1.0009	1.0004	1.0001	1.0003	1.0007
Standard Deviation	0.0159	0.0010	0.0052	0.0042	0.0038	0.0011	0.0047
*Average of 20 repeated runs (25 runs for series 93B).							

TABLE VII. MGA C and MGA D Results

	Ratio: (Average Measured*/Certified)						
	²³⁸ Pu	²³⁹ Pu	²⁴⁰ Pu	²⁴¹ Pu	²⁴¹ Am	P _{eff}	²⁴⁰ Pu _{eff}
CBNM Pu61C	0.9988	0.9948	1.0134	0.9979	0.9988	1.0004	1.0097
CBNM Pu70C	1.0005	0.9957	1.0174	0.9994	0.9929	1.0000	1.0135
CBNM Pu84C	0.9907	0.9991	1.0054	0.9976	0.9927	0.9991	1.0050
CBNM Pu93C	1.0033	0.9996	1.0065	0.9927	0.9840	0.9998	1.0064
CBNM Pu61D	0.9965	0.9958	1.0118	0.9958	0.9935	0.9977	1.0080
CBNM Pu70D	0.9995	0.9958	1.0177	0.9972	0.9893	0.9986	1.0137
CBNM Pu84D	0.9882	0.9992	1.0049	0.9976	0.9926	0.9987	1.0045
CBNM Pu93D	0.9845	0.9995	1.0072	0.9908	0.9840	0.9994	1.0070
Average Ratio	0.9953	0.9974	1.0105	0.9961	0.9910	0.9992	1.0085
Standard Deviation	0.0067	0.0021	0.0053	0.0029	0.0050	0.0009	0.0035
*Average of 20 repeated runs							

V. RESULTS

A. Bias

Bias may be the most important parameter to characterize for a plutonium isotopic code. Comparisons between the two codes are not entirely meaningful because of the bias adjustments that have been performed on the FRAM results. Still, the consistency of results over all the samples may yield information regarding the sample-to-sample consistency of the certified values.

Measurement results on the CBNM samples with both FRAM and MGA have been shown in Tables VI and VII. We present this same data graphically in Figs. 2-8 below. The error bars on the points in Figs. 2-8 represent one standard deviation of the average ($s_{\bar{x}}$) nondestructive assay (NDA) measurement as determined from the distribution of the 20 (25) measurements for each data point. We have not included the uncertainty in the certified values of the CBNM standards (Table I) in the plotted uncertainty.

The results in Fig. 2 for ²³⁸Pu may indicate a small inconsistency in the certified values, on the order of 1% (about two sigma of the Pu84 certified value), between Pu84 and Pu61 and between Pu84 and Pu70. We also note that both code measurements (albeit with larger than desired uncertainties, see Fig. 2) agree with the certified values for the low ²³⁸Pu content of Pu93. This

Fig. 2. ^{238}Pu : ratio of FRAM- and MGA-measured values to CBNM certified values. Error bars are $(s_{\bar{x}})$ for the measured value.

Fig. 3. ^{239}Pu : ratio of FRAM- and MGA-measured values to CBNM certified values. Error bars are $(s_{\bar{x}})$ for the measured value.

Fig. 4. ^{240}Pu : ratio of FRAM- and MGA-measured values to CBNM certified values. Error bars are $(s_{\bar{x}})$ for the measured value.

Fig. 5. ^{241}Pu : ratio of FRAM- and MGA-measured values to CBNM certified values. Error bars are $(s_{\bar{x}})$ for the measured value.

Fig. 6. ^{241}Am : ratio of FRAM- and MGA-measured values to CBNM certified values. Error bars are $(s_{\bar{x}})$ for the measured value.

Fig. 7. P_{eff} : ratio of FRAM- and MGA-measured values to CBNM certified values. Error bars are $(s_{\bar{x}})$ for the measured value.

Fig. 8. $^{240}\text{Pu}_{\text{eff}}$ ratio of FRAM- and MGA-measured values to CBNM certified values. Error bars are $(s_{\bar{x}})$ for the measured value.

observation will become important later when some additional FRAM measurements are discussed in Sec. VI. FRAM and MGA use entirely different gamma-ray peaks and analysis methods to arrive at their results. Agreement between the two codes adds confidence to observations made from the NDA results.

Differences between FRAM and MGA for ^{239}Pu in Fig. 3 merely reflect the differences that are observed for ^{240}Pu illustrated in Fig. 4. Here the MGA values are biased, relative to the CBNM certified values, by about 1%.

For ^{241}Pu , in Fig. 5, all measured averages agree with the certified values to better than 1%. The FRAM values seem to indicate a monotonic increase with ^{241}Pu content of nearly 1% over the range of values represented by these samples. The MGA results exhibit a similar pattern with only a slightly smaller range of increase.

The ^{241}Am results in Fig. 6 indicate similar trends for both codes; this observation was also made for previous measurements on the PIDIE³ samples carried out at Los Alamos and analyzed with the two codes. It is difficult to assess the significance of an approximately 1% difference between the MGA measurements and the CBNM certified values for ^{241}Am , again, because it is at about the two sigma level of the certified value.

The P_{eff} results in Fig. 7 are excellent, indicating biases of less than 0.25% for all measurements. The $^{240}\text{Pu}_{\text{eff}}$ results in Fig. 8 merely reflect the pattern shown in Fig. 4 for ^{240}Pu .

Plutonium-242 has been removed as a variable by using the certified value as an operator-entered parameter in all analyses.

B . Precision

Measurement precision* is shown in Table VIII for all reported data. We report two numbers for each measurement series on each sample. The first number reported is called “Observed” and is the percent relative standard deviation (RSD) of a single measurement calculated from the distribution of the 20 (25) measurements for each run. The second reported number is called “Estimated in Software.” This is the percent RSD estimated from propagated counting statistics in the analysis software and is part of the output for every measurement. Refer to Table IV for the measurement conditions producing these precisions.

The observed measurement precisions are also presented for each isotope, P_{eff} , and $^{240}\text{Pu}_{\text{eff}}$ in Figs. 9-15. It is expected that the MGA code will show better precision than the FRAM code. This arises because the primary data analysis in the MGA code arises from a more intense region of the spectrum than that analyzed by FRAM. This has been borne out by the spectrum integrals in Table IV and the ensuing discussion of that table.

Table VIII and Figs. 9-15 are quite informative. We observe generally good agreement between the observed and software-estimated precisions in Table VIII. The agreement gives one confidence that the software-estimated precisions are reasonable representations of the true measurement precision. Exhaustive study of this data would undoubtedly yield some inconsistencies—FRAM software appears to underestimate the ^{241}Pu RSD and overestimate the P_{eff} RSD, for example—but the general agreement is pleasing.

There were surprises for the authors when examining some of the FRAM precisions vs those of MGA under the ground rules of these measurements as discussed previously. These surprises were that for the higher burnup samples (Pu70 and Pu61) the FRAM precisions for ^{238}Pu , ^{241}Pu , and ^{241}Am were very similar to those of MGA. This meant that FRAM’s P_{eff} precision for these samples was as good as or better than that of MGA. The MGA precision for ^{240}Pu and $^{240}\text{Pu}_{\text{eff}}$ was better than FRAM’s for all measurements on all samples: more in line with our own expectations.

*The term “precision” as used here and throughout the report may be more correctly described as “repeatability” because all measurement series were completed in less than one day without changing the sample-detector geometry. We do find that “reproducibility” (single measurement repeated with sample replacement on a daily basis for a period of a month) is essentially the same as repeatability for the FRAM system.

TABLE VIII. Observed and Software-Estimated Measurement Precision

	Percent RSD of a Single Measurement						
	²³⁸ Pu	²³⁹ Pu	²⁴⁰ Pu	²⁴¹ Pu	²⁴¹ Am	P _{eff}	²⁴⁰ Pu _{eff}
CBNM Pu93							
FRAM B							
Observed	3.82	0.11	1.58	0.37	1.05	0.22	1.56
Estimated in software	5.94	0.12	1.72	0.19	1.03	0.36	1.70
FRAM E							
Observed	8.89	0.16	2.39	0.54	1.70	0.42	2.37
Estimated in software	9.06	0.18	2.70	0.30	1.59	0.56	2.66
MGA C							
Observed	4.25	0.04	0.64	0.28	0.59	0.16	0.63
Estimated in software	3.27	0.04	0.54	0.41	0.48	0.13	0.54
MGA D							
Observed	3.16	0.03	0.42	0.28	0.28	0.10	0.41
Estimated in software	2.77	0.03	0.47	0.34	0.40	0.11	0.46
CBNM Pu84							
FRAM B							
Observed	1.21	0.16	0.94	0.33	0.75	0.26	0.90
Estimated in software	1.53	0.17	1.04	0.20	0.57	0.36	0.99
FRAM E							
Observed	1.51	0.25	1.51	0.45	0.79	0.30	1.44
Estimated in software	1.87	0.21	1.25	0.24	0.69	0.43	1.19
MGA C							
Observed	1.01	0.07	0.39	0.31	0.44	0.17	0.37
Estimated in software	1.08	0.08	0.45	0.35	0.41	0.19	0.43
MGA D							
Observed	0.70	0.05	0.30	0.18	0.20	0.13	0.29
Estimated in software	0.80	0.06	0.34	0.26	0.31	0.14	0.33
CBNM Pu70							
FRAM B							
Observed	0.56	0.34	1.46	0.48	0.49	0.23	1.10
Estimated in software	0.47	0.34	1.50	0.35	0.42	0.31	1.16
FRAM E							
Observed	0.88	0.41	1.77	0.74	0.51	0.35	1.32
Estimated in software	0.63	0.46	2.01	0.47	0.56	0.42	1.56
MGA C							
Observed	0.54	0.22	0.79	0.62	0.77	0.43	0.62
Estimated in software	0.76	0.27	0.93	0.67	0.69	0.55	0.74
MGA D							
Observed	0.56	0.19	0.68	0.42	0.40	0.38	0.56
Estimated in software	0.56	0.20	0.69	0.49	0.51	0.40	0.56
CBNM Pu61							
FRAM B							
Observed	0.48	0.51	1.32	0.53	0.60	0.22	0.95
Estimated in software	0.50	0.42	1.19	0.42	0.48	0.33	0.87
FRAM E							
Observed	0.66	0.53	1.40	0.60	0.68	0.31	0.99
Estimated in software	0.66	0.55	1.58	0.56	0.63	0.43	1.15
MGA C							
Observed	0.73	0.45	0.95	0.76	0.70	0.59	0.73
Estimated in software	0.84	0.47	0.93	0.78	0.79	0.65	0.71
MGA D							
Observed	0.38	0.22	0.51	0.37	0.43	0.31	0.37
Estimated in software	0.60	0.34	0.67	0.56	0.57	0.47	0.53

Fig. 9. ^{238}Pu precision from repeated measurements.

Fig. 10. ^{239}Pu precision from repeated measurements.

Fig. 11. ^{240}Pu precision from repeated measurements.

Fig. 12. ^{241}Pu precision from repeated measurements.

Fig. 13. ^{241}Am precision from repeated measurements.

Fig. 14. P_{eff} precision from repeated measurements.

Fig. 15. $^{240}\text{Pu}_{\text{eff}}$ precision from repeated measurements.

Another very important observation was that the precision for P_{eff} was better than that for $^{240}\text{Pu}_{\text{eff}}$ for all measurements on all samples by both codes. This means that gamma-ray isotopic measurements used to interpret calorimetry should give more precise measurement results for total plutonium than gamma-ray isotopic measurements used to interpret coincidence counting. This observation does not consider the biases that may be present in any of the methods; it is based only on the precision of the measurements. Typical measurement biases for gamma-ray isotopics are seen in Tables V - VII and are similar in magnitude to the measurement precisions reported in Table VIII. This general result on measurement precision for P_{eff} and $^{240}\text{Pu}_{\text{eff}}$ is expected to hold for all sample types and all reasonable isotopic distributions. Section VI confirms this result for FRAM measurements on larger samples.

Precisions are always best (smallest RSD) for small samples assuming the throughput can be optimized. A larger sample may improve the FRAM precision because counting rates would be nearer the target values. Yet, a larger sample tends to exhibit poorer precision for both codes (relative to the CBNM samples) because the pulse height spectrum will have a more intense low-energy continuum, hence a poorer signal-to-background ratio, caused by scattering within the sample of the higher energy gamma rays from plutonium. This means that it is difficult, if not impossible, to extrapolate these results to other samples. Some examples of FRAM measurements on larger samples are given in the next section.

VI. FRAM MEASUREMENTS ON LOS ALAMOS WORKING STANDARDS

We also measured three Los Alamos working standards currently in use at the Los Alamos TA-55 Plutonium Facility and analyzed the data with the FRAM code. These standards were measured at the TA-55 facility with the same hardware that is used for routine FRAM measurements at that facility, thus representing actual field-use conditions. We acquired these data to determine if the branching ratio adjustments made to FRAM from the CBNM sample measurements (the WRTEST parameter file) improved actual field measurement results. There were no MGA measurements made on these samples.

In Table IX we list the sample material, sample mass, and isotopic distribution for these three samples as of the measurement date. The CALEX standard is the United States DOE Calorimetry Exchange Sample used by all DOE facilities doing plutonium calorimetry. Its isotopic distribution was characterized by several laboratories in 1979. Some remeasurements were carried out a few years ago in an attempt to answer concerns about the original characterization of ^{238}Pu and ^{241}Am . The results of the remeasurements were not conclusive and did not support any changes to the original values. STD40 and LAO225BS have been extensively characterized and recharacterized at Los Alamos by two different mass spectrometry groups.

As with the CBNM samples, we acquired data for 20 repeat measurements for each sample. We analyzed the data set for each sample with two FRAM parameter files—WIDERANGE6 that is in routine use at TA-55 and WRTEST, the parameter file incorporating adjustments from the CBNM sample measurements as discussed in Sec. IV above. Table X displays, similar to Table IV for the CBNM measurements, the counting conditions for the TA-55 working standards. For STD40 and LAO225BS, we were able to attain the target count rate of 40 kHz that we were unable to attain with the CBNM samples. The count rate and count time used for the CALEX measurements were the same as those used every day as a measurement control sample at TA-55. We did not characterize the detector resolution with ^{57}Co as we did for the detector used with the CBNM samples, but the FRAM analyses showed that it was about 580 eV at 129 keV and 712 eV

TABLE IX. Characteristics of Los Alamos TA-55 Working Standards

Standard	Material	Weight Percent with Respect to Total Plutonium					
		^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	^{241}Am
CALEX	400 g, PuO_2	0.0093	93.89	5.86	0.207	0.029	0.1691
STD40	1000 g, PuO_2	0.0635	87.25	11.78	0.710	0.201	0.5508
LAO225BS	1000 g, PuO_2	0.0590	82.12	16.51	0.952	0.354	0.6425

TABLE X. Measurement Conditions for TA-55 Working Standards

	CALEX	STD40	LAO225BS
Shaping time constant (μ s)	1	1	1
Count Rate (kHz)	24	40	40
Spectrum Integral	6.64 E07	4.85 E07	4.83 E07
Count Time (True Time) (s)	3600	1800	1800
Filters	1.9 mm Cd 0.38 mm Cu	1.9 mm Cd 0.38 mm Cu	1.9 mm Cd 0.38 mm Cu

at 208 keV under the measurement conditions of Table X. In comparison, the detector used for the CBNM sample measurements in Table IV gave a resolution from the FRAM analyses of 570 eV at 129 keV and 700 eV at 208 keV.

Table XI(a) displays the ratio to the accepted values of the average of the 20 measurements for the FRAM data analyzed with the WIDERANGE6 parameters in routine use at TA-55. With the exception of the ^{238}Pu value for CALEX and, perhaps ^{241}Am , the results are very similar to those displayed in Table V from using the same parameters on the CBNM data. The WIDERANGE6 analysis of these measurements is also consistent with the measurements on the same samples reported in Ref. 1.

Table XI(b) displays the results of using the adjusted parameters from the CBNM standards in the analysis of the data from the TA-55 working standards. The WRTEST parameters improved the results for ^{238}Pu (CALEX was still bad) but gave poorer agreement for ^{241}Am leaving the small bias in P_{eff} unchanged. The new parameters improved the bias in $^{240}\text{Pu}_{\text{eff}}$ by about 0.3%.

TABLE XI(a). FRAM WIDERANGE6 Parameters with TA-55 Working Standards

Sample ID	Ratio: (Average Measured/Accepted)						
	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{241}Am	P_{eff}	$^{240}\text{Pu}_{\text{eff}}$
CALEX	1.0745	0.9994	1.0101	1.0010	0.9904	1.0021	1.0105
STD40	1.0118	0.9997	1.0019	0.9984	1.0029	1.0020	1.0019
LAO225BS	1.0172	0.9990	1.0049	0.9968	1.0043	1.0034	1.0048
Average Ratio	1.0345	0.9994	1.0056	0.9987	0.9992	1.0025	1.0057

TABLE XI(b). FRAM WRTEST Parameters with TA-55 Working Standards

Sample ID	Ratio: (Average Measured/Accepted)						$^{240}\text{Pu}_{\text{eff}}$
	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{241}Am	P_{eff}	
CALEX	1.0546	0.9996	1.0065	1.0008	1.0029	1.0022	1.0066
STD40	0.9933	1.0002	0.9987	0.9986	1.0155	1.0019	0.9991
LAO225BS	0.9989	0.9996	1.0019	0.9972	1.0170	1.0036	1.0018
Average Ratio	1.0156	0.9998	1.0024	0.9988	1.0118	1.0025	1.0025

These data indicate a possible bias between the CBNM ^{241}Am measurements and the Los Alamos measurements on STD40 and LAO225BS at the level of about 1%. Because the CBNM standards have a 1 RSD uncertainty of about 0.5% for ^{241}Am , a 1% bias is difficult to demonstrate for these measurements. These data and data in Ref. 1 continue to support the explanation of the ^{238}Pu bias for CALEX as a characterization error in spite of several analytical results to the contrary. We also note that MGA measurements on CALEX as reported to the Calorimetry Exchange support the accepted value, not the FRAM-measured value.

At the level of 0.01 wt%, ^{238}Pu presents a difficult measurement problem for both destructive and nondestructive analysis. Apparent biases at the level of a few percent are difficult to resolve and are not a significant source of error at this concentration.

The final data that we will show in Table XII is the FRAM measurement precision for the three TA-55 working standards. As in Table VIII we display the observed and software-estimated precision at the one standard deviation level. Refer to Table X for the measurement conditions.

A comparison of these precisions with the FRAM precisions in Table VIII for the CBNM samples will reinforce some of the comments made at the end of Sec. V. CALEX and CBNM Pu93 have similar isotopic distributions. Both FRAM B and CALEX were counted for one hour, the small CBNM FRAM B measurement at 10 kHz and the 400-g CALEX at 24 kHz. Yet the larger, higher-count-rate CALEX sample had slightly poorer precision for ^{238}Pu and ^{240}Pu than the smaller, lower-count-rate CBNM Pu93 sample. Compare also CBNM Pu84 FRAM E with STD40. Both are counted for 1800 s, STD40 at 40 kHz and FRAM E at 15 kHz. Again the ^{238}Pu and ^{240}Pu precisions are poorer for the higher-count-rate sample although ^{241}Am precision is better at the higher count rate yielding similar precisions for P_{eff} in both cases.

TABLE XII. Observed and Software-Estimated Measurement Precision

	Percent RSD of a Single Measurement						
	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{241}Am	P_{eff}	$^{240}\text{Pu}_{\text{eff}}$
CALEX							
Observed	6.80	0.13	2.08	0.30	0.98	0.30	2.05
Estimated in software	6.55	0.11	1.74	0.16	0.81	0.34	1.72
STD40							
Observed	2.01	0.23	1.72	0.46	0.57	0.33	1.71
Estimated in software	1.86	0.19	1.45	0.22	0.51	0.42	1.40
LAO225BS							
Observed	1.76	0.20	0.98	0.36	0.60	0.23	0.94
Estimated in software	1.96	0.20	1.00	0.22	0.45	0.37	0.96

VII. SUMMARY

We have completed a series of gamma-ray plutonium isotopic composition measurements on the CBNM NRM 271 standards produced by the European community and have analyzed the measurements with two isotopic analysis codes, the Los Alamos FRAM code and the Lawrence Livermore MGA code. The purpose of the measurements was to see if these standards would enable us to reduce biases in the routine use of the FRAM code at Los Alamos. Separate data acquisitions were made for each code emphasizing acquisition conditions thought to optimize each code's performance under realistic field conditions.

We analyzed measurements taken to test the FRAM code with two different parameter files, the first being that in routine use at Los Alamos, and the second being a parameter file whose parameters were adjusted to remove all significant biases found with the first file analysis. Measurements taken under MGA conditions were not adjusted in any fashion because the MGA code is not supported by Los Alamos.

Several small inconsistencies in the sample characterization or the NDA measurements were tentatively identified but not positively confirmed because of the uncertainties in the certified values for ^{238}Pu and ^{241}Am .

We also used FRAM parameters, adjusted from the CBNM measurements, to analyze a separate set of in-plant measurement data on Los Alamos working standards. The results were somewhat inconclusive as we saw improved agreement between measured and accepted values for ^{238}Pu and ^{240}Pu and poorer agreement for ^{241}Am . The question of a FRAM measurement bias or a characterization error in the ^{238}Pu abundance in the Calorimetry Exchange sample was not resolved.

These standards will be very useful to Los Alamos in providing certified reference values for higher burnup materials with ^{240}Pu isotopic fractions >16%—a range not addressed with materials currently available at Los Alamos.

VIII. ACKNOWLEDGMENTS

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