

Test and Evaluation of the FRAM Isotopic Analysis Code for EURATOM Applications

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Abstract

We have made gamma-ray isotopic composition measurements on uranium and plutonium samples and analyzed the data with the FRAM isotopic analysis software. The measured samples were at two different facilities of direct interest for EURATOM safeguards inspections.

The first set of measurements were made on UF₆ cylinders. These measurements were undertaken to test FRAM as a method to supplement the calibration-intensive, classical ²³⁵U Enrichment Meter method and also to prove its capability on thick-walled UF₆ cylinders where other arbitrary sample/geometry methods using the 100 keV region fail.

The second set of measurements were made on plutonium-bearing samples to demonstrate capability through heavy-walled containers, on mixed oxide (MOX), and to make comparisons with the EURATOM version of MGA (Multi-Group Analysis).

MEASUREMENT EQUIPMENT

Nearly identical equipment was used for the FRAM measurements on both uranium and plutonium.

Detector: A nominal 25% relative efficiency coaxial detector was used for all measurements. Different detectors with similar performance were used at the two facilities for logistical reasons. The detectors were

	operated with a 2- μ s shaping time.
Data Acquisition and MCA:	An ORTEC DART portable multichannel analyzer was used for all data collection. Data from all samples [uranium, plutonium, and mixed oxide (MOX)] were acquired in 8192 channels at a gain of 0.125 keV/channel, spanning the energy range from 0–1024 keV.
Computer:	IBM Think Pad model 760ED running Windows 3.1. Current version of FRAM (version 3.2/3.3) operates under Windows NT, 98, or 95.
Software	PC/FRAM, Version 2.2. (This version is now superseded by version 3.2/3.3.)

UF₆ CYLINDER MEASUREMENTS

Our measurements concentrated on the 30B cylinders, but one measurement was made on each of two other sample types. All uranium was enriched to <5% ²³⁵U.

The uranium measurements are summarized in Table I. Most measurements were made for 20–30 min with longer measurements overnight. Laboratory measurements at Los Alamos show successful measurements through 16-mm steel (The 30B cylinders measured here have a 13-mm wall thickness.) in as few as 10 min. The statistical uncertainty for these 10-min laboratory measurements is 10%–20% (1 RSD). The longer measurements demonstrated here showed better precision.

The first measurement listed in Table I was made with a detector brought by EURATOM, not the detector described above. The combination of the smaller detector (8.5% vs 26.2% relative efficiency) and a 15-min count time gave results that were poorer than those in the rest of the table. The summary ratio and standard deviation do not include this value.

Measurements on 30B cylinders for 20–30 min show a repeatability of about 6%–8% (1 RSD). Most measurements appear to be biased low, in agreement with laboratory measurements at Los Alamos. We have not made any bias corrections in Table I. We believe that the source of the bias is coincidence summing of gamma rays from ²³⁸U daughters. This is a geometrical effect and makes the measurements sensitive to sample-to-detector distance. In the worst case, this appears to be a 6%–10% effect. We are actively working on methods to reduce this effect and a report¹ on these efforts is presented at this meeting. A recalculation of some of these results using the techniques described in Ref. 1 is presented later. In the meantime, this bias could easily be removed for a given fixed-measurement geometry as typified by UF₆ cylinder measurements.

We note that the repeated measurements shown in Table II are biased low by about 6% (Measured/Tag = 0.9384). This is in nearly exact agreement with laboratory measurements at Los Alamos using the EC171 standard set and a 16-mm diameter steel disk. For the Los Alamos measurements, a bias (Measured/Tag = 0.9422) was observed. Since this bias is distance dependent, it is appropriate to observe that the cylinder measurements and the laboratory measurements were both done with the detector in contact with the

cylinder/steel disk. The only difference is the approximately 3-mm difference in the steel thickness between the 30B cylinder and the 16-mm disk used at Los Alamos.

Table II displays the individual values of 15 measurements of 30 min each. We note that the one sigma error from propagated counting statistics displayed in the code of 5.6% (1 RSD) is in reasonable agreement with that observed (7.0%) from the spread of the 15 measurements.

A classical enrichment system composed of a small, coaxial HpGe detector and the Rossendorf Mini MCA with a palmtop computer was also set up and calibrated during this measurement exercise. This system was used along side of the FRAM isotopic system during the exercise. The dual sets of measurements allowed useful conclusions to be drawn from the exercise. These conclusions are summarized following Tables I and II.

Table I

Summary of Uranium Measurements

Measured Item	Count time (s)	Meas. ²³⁵ U (%)	1 sigma absol./(%RSD)	²³⁵ U Tag Value	Ratio Meas./Tag	Comment
30B UF ₆ Cyl	900	1.91	0.31 (16%)	1.50	1.273	EURATOM 8.5% coax, too small for best measurement
30B UF ₆ Cyl	3600	1.480	0.095 (6.4%)	1.50	0.987	Same detector, long count time
30B UF ₆ Cyl	1200	1.687	0.128 (7.6%)	1.50	1.125	Los Alamos 26% efficiency coax for this and all subsequent measurements
30B UF ₆ Cyl	1200	3.216	0.26 (8.0%)	3.20	1.005	Shield 9 cm from domed end of cylinder
30B UF ₆ Cyl	3600	4.684	0.19 (4.1%)	4.95	0.9463	Repeat
	3600	4.661	0.19 (4.1%)	4.95	0.9416	
30B UF ₆ Cyl	1200	4.467	0.34 (7.7%)	4.95	0.902	End of cylinder
UO ₂ Powder	1800	3.462	0.097 (2.8%)	3.20	1.082	Thinner wall, higher count rate
30B UF ₆ Cyl	2400	2.436	0.11 (4.6%)	2.50	0.9852	
		2.672	0.14 (5.3%)	2.50	1.069	
30B UF ₆ Cyl	1800	0.6995	0.042 (6%)	0.71	0.9852	Side of cylinder
30B UF ₆ Cyl	1800	0.598	0.036 (6%)	0.71	0.842	End of cylinder
Scrap Container	1200	2.443	0.053 (2.2%)	2.65	0.9219	
30B UF ₆ Cyl	15 x 1800	4.448 avg of 15	0.31 (7%) single meas error	4.74	0.9384	15 measurements overnight

0.979 Avg of Ratio: Meas./Tag
(exclude 1st meas. in table)

0.78 Std. Dev. Of Meas./Tag values
(exclude 1st meas. in table)

Table II
Repeated Measurements

Measurement	Measured % ^{235}U	Predicted in Code % RSD
1	4.680	5.8
2	4.475	5.5
3	4.665	5.9
4	4.263	5.4
5	4.295	5.6
6	4.282	5.5
7	4.780	5.9
8	4.120	5.2
9	4.540	5.6
10	4.997	6.2
11	4.549	5.8
12	3.830	5.0
13	4.089	5.1
14	4.385	5.6
15	4.778	6.1
Averages	4.449	5.61
Std. Dev. of single	0.31	
% RSD of single	7.0	

- Because FRAM requires no calibration, it can start actual verification measurements more quickly than the classical enrichment method. It would be most useful when only a few verification measurements ($<\sim 6$) were required.
- After calibration, the classical enrichment method can complete verification measurements on the samples appropriate to the calibration more quickly (100-s count time) than FRAM (10–20 min count time). This method is therefore more appropriate when a large number of the same type of samples have to be measured.
- Moving the detector systems (HPGe detectors) between cylinders is basically the same for the two techniques. Both techniques can benefit from special purpose fixtures to position a shielded detector to the item being measured. Moving and setting up from cylinder to cylinder consumes a significant amount of time. This reduces the throughput difference between the two methods.
- The classical enrichment method requires precise reproduction of the calibration geometry and relies on an independent estimation of the wall thickness of the container which is subsequently used as a correction factor in the evaluation procedure. FRAM has no geometry reproducibility requirements (see discussion later) making it easier to set up the measurement geometry.

- FRAM was able to demonstrate measurements on additional container types (UO₂ powder, scrap container) that could not be measured by the classical enrichment method using the calibration for 30B cylinders.
- FRAM had no difficulty with measurements on the thick-walled (13-mm) UF₆ cylinders. Other arbitrary sample measurement techniques that use low-energy gamma rays cannot measure these UF₆ cylinders.
- FRAM measurement precision (typically 3%–8%), even with its longer count time, is poorer than that from the classical enrichment method. It remains for the user to decide if the FRAM precision is adequate.
- The current implementation of FRAM for uranium measurements demonstrates some sensitivity to the sample-detector distance. We believe we understand the source of this problem and are working to correct it in the next version of the software, expected to be complete by late 1999. Preliminary results from the new algorithms required for these corrections are presented below. In the meantime, a simple change of one parameter in the parameter file could remove this bias for UF₆ cylinder measurements.
- The analysis method used by FRAM currently requires an equilibrium condition between ²³⁸U and its ²³⁴Pa daughters. This process may be further complicated by processing steps that only partially remove daughter products and by daughter product deposits on cylinder walls.

COINCIDENCE SUMMING CORRECTIONS

Reference 1 discusses the coincidence summing corrections that are being studied for inclusion in future versions of FRAM. A preliminary version of these corrections has been applied to the data of Table II and is presented below in Table III. This preliminary analysis is seen to correct the analysis very well for this data set at the expense of somewhat poorer measurement precision. Study of these correction methods will continue for inclusion in the next version of FRAM.

Table III
Reanalysis with Coincidence Summing Corrections

Coincidence Summing Corrections		
Repeated Measurements on 30B UF ₆ Cylinder		
15 measurements at 1800 s each		
Tag Value = 4.74%		
Measurement	Measured (Table II) % ²³⁵ U	Corrected for Summing % ²³⁵ U
1	4.680	5.296
2	4.475	4.729
3	4.665	5.218
4	4.263	4.389
5	4.295	4.538
6	4.282	4.507
7	4.780	5.128
8	4.120	4.422
9	4.540	4.914
10	4.997	5.228
11	4.549	4.517
12	3.830	4.114
13	4.089	4.523
14	4.385	4.804
15	4.778	5.146
Averages	4.4485	4.765
Std. Dev. of single	0.310	0.371
% RSD of single	7.0	7.8

PLUTONIUM-BEARING SAMPLE MEASUREMENTS

For the plutonium measurements, we compare the FRAM measurements with EURATOM's Pu Meter results. The Pu Meter is version 7.4 of MGA with a custom EURATOM user interface and these results are referred to as "MGA" in the following tables and discussion.

The FRAM measurements used the operator-declared values for ²⁴²Pu where available.

The measurement results are displayed in Table IV.

Table IV
Summary of Plutonium Measurements
Declared values updated to measurement dates

Sample No.	Material Type	Mass g Pu	Measurement	Count time (s)	²³⁸ Pu (%)	²³⁹ Pu (%)	²⁴⁰ Pu (%)	²⁴¹ Pu (%)	²⁴² Pu (%)	²⁴¹ Am (%)	P _{eff} mW/g Pu	Comment	
1.	Pu + SS	468	Declared	Declared	0.0368	85.103	14.201	0.438	0.222	1.790	4.916		
			MGA	1800	0.0374	85.397	13.867	0.455	0.243	1.626	4.714		
			FRAM	1200	0.0302	85.08	14.188	0.482	0.222	1.646	4.714		
			(FRAM %RSD)		(11.4%)	(0.59%)	(3.6%)	(0.63%)	declared	(1.0%)	(NA)		
2.	PuO ₂	1114	Declared	Declared	0.0338	83.182	16.066	0.464	0.254	1.683	4.873		
			MGA	1800	0.0398	85.41	13.807	0.487	0.254	1.64	4.741		
			FRAM	3600	0.0366	84.891	14.326	0.492	0.254	1.648	4.759		
			(FRAM %RSD)		(5.6%)	(0.35%)	(2.1%)	(0.37%)	declared	(0.63%)	(0.58%)		
3.	??	131.5	Declared	Declared	0.0439	84.915	14.343	0.490	0.208	1.685	4.845	Declaration of 07-Apr-1991 updated	
			Declared	Declared	0.0417	84.790	14.439	0.496	0.233	1.678	4.829	Declaration of 01-Jan-1968 updated	
			MGA	1800?	0.0415	85.233	14.007	0.490	0.2295	1.684	4.812		
			FRAM	1800	0.0406	85.139	14.092	0.495	0.233	1.682	4.809		
(FRAM %RSD)		(3.6%)	(0.29%)	(1.6%)	(0.32%)	declared	(0.49%)	(0.43%)					
4.	MOX 29% ²³⁵ U	54 g Pu 129 g ²³⁵ U 440 g U	Declared	Declared	0.0209	91.934	7.766	0.226	0.053	1.000	3.591	<u>²³⁵U/Pu</u> 2.39	29% enriched uranium in MOX
			MGA	1800	0.0251	91.858	7.81	0.254	0.053	0.955	3.566	2.13	
			FRAM	1800	0.0189	92.770	6.927	0.227	0.053	0.987	3.522	2.27	
			(FRAM %RSD)		(13%)	(0.4%)	(5.7%)	(0.5%)	declared	(1.0%)	(0.98%)		
5.	MOX	569.6	Declared	Declared	0.0925	78.658	19.677	1.008	0.564	2.936	6.824	<u>²³⁵U/Pu</u>	
			MGA	600	0.107	78.493	19.731	1.104	0.565	2.363	6.258	0.0241	
			FRAM	1800	0.0984	79.304	18.958	1.078	0.562	2.445	6.261		Analyzed without uranium
			FRAM	1800	0.0956	79.427	18.869	1.046	0.562	2.486	6.287	0.0279	Analyzed including uranium
(FRAM %RSD)		(4.1%)	(0.6%)	(2.7%)	(1.05%)	declared	(1.0%)	(0.8%)					
6.	MOX + SiC residues	336	Declared	Declared	0.207	77.862	20.113	1.23	0.587	2.935	7.108	<u>²³⁵U/Pu</u>	
			MGA	1200	0.3306	77.002	20.546	1.53	0.591	3.68	9.072	0.0203	
			FRAM	1800	0.226	77.826	20.088	1.288	0.571	3.226	7.935		Analyzed without uranium
			FRAM	1800	0.222	77.82	20.121	1.266	0.571	3.256	7.946	0.0187	Analyzed including uranium
(FRAM %RSD)		(1.9%)	(0.6%)	(2.4%)	(0.6%)	declared	(0.8%)	(0.67%)		FRAM results thought better for P _{eff}			
7.	MOX 48.5% enrich U 1864 g U	531	Declared	Declared	1.029	65.972	24.34	4.753	3.906	4.946	14.653	<u>²³⁵U/Pu</u> 1.702	48.5% Enriched U in MOX
			MGA	1200	1.045	65.915	24.342	4.791	3.906	4.791	14.699	1.675	
			FRAM	1383	1.023	65.052	25.23	4.789	3.903	4.900	14.614	1.732	
			(FRAM %RSD)		(2.5%)	(2.1%)	(6.2%)	(2.1%)	declared	(2.4%)	(1.6%)		
8.	Pu + SS	468	Declared	Declared	0.0368	85.103	14.201	0.438	0.222	1.790	4.916	MGA can not measure shielded sample	
			MGA	1200	0.036	84.99	14.29	0.47	0.22	1.65	4.758	MGA measurement <u>without</u> steel	
			FRAM	1800	0.032	84.847	14.389	0.4775	0.254	1.638	4.727	Shield sample with 24 mm steel and	
			(FRAM %RSD)		(7.2%)	(0.7%)	(4.4%)	(0.9%)	declared	(0.8%)	(1.1%)	1.5 mm Cd	
9.	Scrap	50	Declared	Declared	NA	NA	NA	NA	NA	NA	NA		
			MGA	1200	0.243	71.61	24.66	2.44	0.978	1.45	6.250		
			FRAM	3600	0.242	71.496	24.763	2.499	1	1.461	6.260		
			(FRAM %RSD)		(1.25%)	(0.44%)	(1.35%)	(0.46%)	declared	(0.83%)	(0.53%)		

10.	Scrap	226	Declared	Declared	NA	NA	NA	NA	NA	NA	NA	
			MGA	1800	0.232	71.75	24.67	2.39	0.95	1.44	6.175	
			FRAM	1800	0.241	71.298	24.959	2.502	1.00	1.448	6.252	Possible Am/Pu heterogeneities
			(FRAM %RSD)		(1.8%)	(0.65%)	(1.95%)	(0.66%)	declared	(1.3%)	(0.77%)	

The first two samples in Table IV were said to be from the same batch of plutonium. Their declared values are significantly different from each other. The FRAM measurements and MGA measurements generally agree with each other and tend to support the declared value for Sample 1 as the better of the two declared values.

For the most part, the FRAM measurements confirm the MGA results. The precision (RSD) of the FRAM measurements is given for each measurement. For the important ^{240}Pu isotope, the FRAM precision is poorer than that of MGA (not shown) as is well known, arising from the intrinsic intensity of the gamma rays analyzed.

For MOX Sample 4, the FRAM value for ^{240}Pu differed from the MGA value and the declared value. With only one exception, the P_{eff} (mW/gPu) values for FRAM and MGA agreed to 1% or better with most agreeing to better than 0.5%. The outlier was Sample 6), where FRAM differed from MGA by 14%. The FRAM results were thought to be best, based on the judgment of the EURATOM personnel present.

Sample 1 was measured a second time (third entry from the bottom of Table IV, listed as Sample 8) with a "shield" of 24 mm of steel interposed between the sample and the detector to mock up a thick-walled container. The Pu Meter MGA software will not work under these conditions. The FRAM measurement using the more penetrating gamma rays above 200 keV was in very good agreement with the previous unshielded measurements of this sample.

Four of the samples we measured (Samples 4, 5, 6, and 7) were MOX with two of these having ^{235}U enriched to 29% and 49%. The results for the measurement of $^{235}\text{U}/\text{Pu}$ are in the Comment column in Table IV. The FRAM measurements appear to be in general agreement with the MGA results and with the declared values (where available). It should be noted that gamma-ray isotopic measurements of the $^{235}\text{U}/\text{Pu}$ ratio in MOX can be biased if the particle sizes of the uranium and plutonium are different.

Table V displays the results of six repeated measurements (not shown in Table IV) on Sample 1. These measurements illustrate the repeatability of FRAM for short measurement times and demonstrate the degree of agreement between the uncertainties estimated in the code and those actually observed from the repeated measurements. The reader may also compare the averages with the other two measurements of Sample 1 and Sample 8 for this item shown in Table IV.

Table V
Six 300 s Measurements of Sample 1

Measurement	Measured ^{238}Pu	Est. in Code % RSD	Measured ^{239}Pu	Est. in Code % RSD	Measured ^{240}Pu	Est. in Code % RSD	Measured ^{241}Pu	Est. in Code % RSD	Measured ^{241}Am	Est. in Code % RSD	Measured P_{eff}	Est. in Code % RSD
1	0.0252	21	84.989	0.93	14.29	5.6	0.474	1.0	1.686	1.7	4.736	1.57
2	0.0382	15	84.195	0.96	15.06	5.4	0.481	1.0	1.688	1.7	4.852	1.55
3	0.0431	13	85.387	0.94	13.87	5.9	0.478	1.0	1.774	1.7	4.916	1.54
4	0.0357	16	85.925	0.95	13.33	6.2	0.488	1.0	1.703	1.7	4.766	1.60
5	0.0326	14	85.621	0.77	13.64	4.9	0.483	0.83	1.665	1.4	4.720	1.30
6	0.0381	12	85.332	0.78	13.93	4.8	0.483	0.83	1.625	1.4	4.721	1.30
Average	0.0355	15.2	85.2415	0.89	14.0200	5.47	0.4812	0.94	1.6902	1.60	4.7852	1.48
Std. Dev. of single	0.0061		0.60		0.60		0.0048		0.049		0.08	
% RSD of single	17.20		0.70		4.29		1.00		2.91		1.69	

Figure 1 displays the ratio of FRAM/MGA results for ^{240}Pu and P_{eff} for the ten samples listed in Table IV. The error bars are those from the FRAM measurements. Adding the error bars from MGA (not available but generally smaller than those from FRAM) would only increase the confidence that the two codes are in agreement with each other. The two outliers have been mentioned earlier. The low value of the ^{240}Pu ratio for Sample 4 may arise from FRAM. We note that the value is about 2 sigma from agreement. The low value for Sample 6 is thought to arise from the MGA measurement.

An interesting aspect of these measurements is the higher precision of the ratios for P_{eff} relative to those for ^{240}Pu . This is a general result that arises from the structure of the equations governing P_{eff} and appears to hold for both FRAM and MGA and most isotopic distributions although it may be more pronounced for lower burnup plutonium. This characteristic argues strongly for the use of calorimetry in instances where the increased measurement time can be tolerated or in instances where a referee measurement is needed.

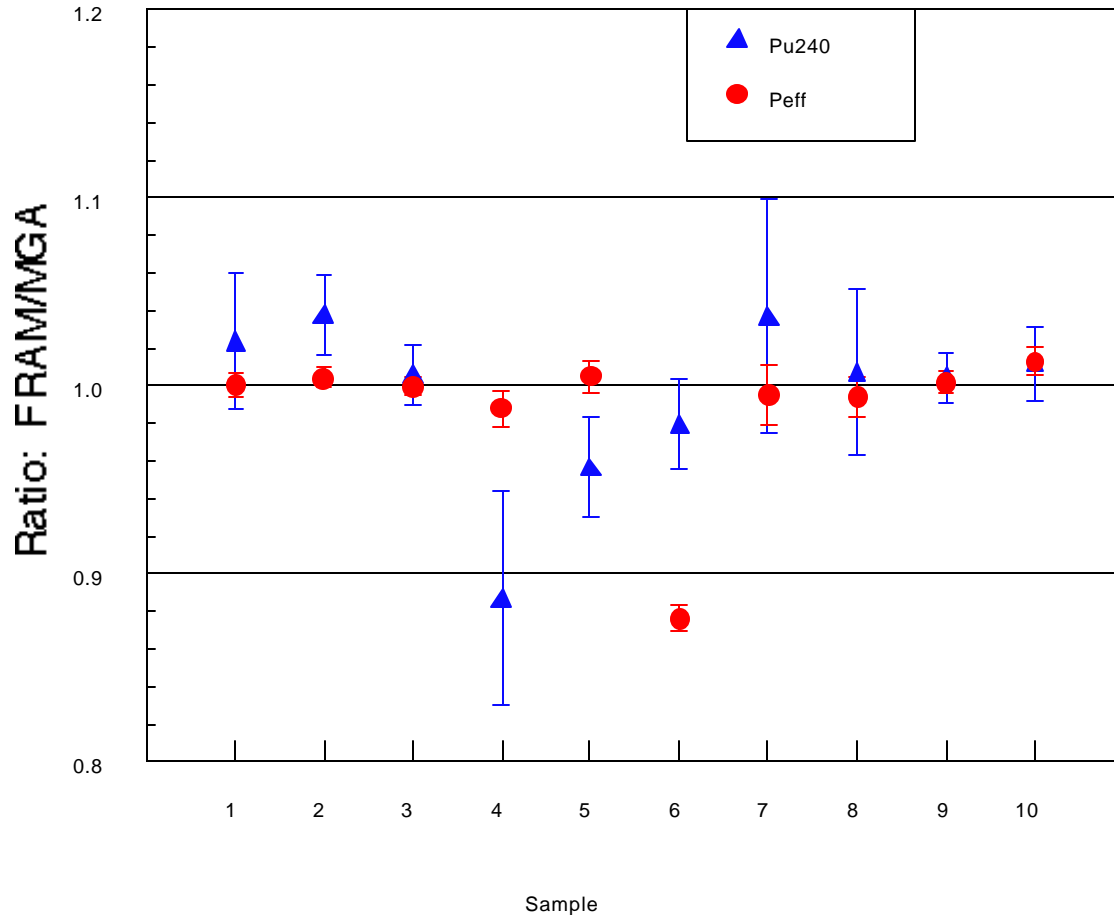


Fig. 1. Ratio of FRAM to MGA measurement results for ^{240}Pu and P_{eff} .

The averages over all samples (two outliers excluded) are 1.0069 for ^{240}Pu and 0.9997 for P_{eff} . The standard deviation of the ^{240}Pu ratios is 0.026 while that of the P_{eff} ratios is 0.007.

SUMMARY

We have demonstrated the use of a data-acquisition system using a single coaxial HPGe detector, a single set of electronics settings, and the FRAM isotopic analysis software to measure the isotopic composition of a wide variety of uranium- and plutonium-bearing items. This single system has a wider range of applicability than any other system available today and, by itself, is capable of all the isotopic composition measurements currently made by EURATOM inspectors using several different systems.

We demonstrated measurements on low-enriched uranium in thick-walled (13-mm) 30B UF_6 cylinders as well as in other, thinner containers. Although not demonstrated in these measurements, the same system has been used to measure ^{235}U enrichments as high as 98%. The measurements also pointed out the known, but correctable, distance-dependent measurement bias for ^{235}U in low-enriched uranium. The same system was

used, also, with no change in operating conditions, to measure plutonium with ^{240}Pu content from 7%–24% and MOX with $^{235}\text{U}/\text{Pu}$ from 0.02–2.3. We also demonstrated the ability to measure plutonium isotopic composition through 24 mm of steel and have, on other occasions, demonstrated the same capability through as much as 13 mm of lead. The measurements also demonstrated that the versatility of FRAM also comes at a price of poorer counting precision in some instances, compared to the several individual methods currently in use.

The test and evaluation of this system was undertaken at the request of EURATOM to ascertain if FRAM can effectively be used in their inspection regimes and supported under the DOE/EURATOM Agreement by the International Safeguards Division, NN44.

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REFERENCE

1. Duc T. Vo and Thomas E. Sampson, "Methods for Uranium Isotopic Analysis with the FRAM Isotopic Analysis Code," to be presented at the 40th Annual INMM to be held in Phoenix, Arizona, July 25-29, 1999, Los Alamos National Laboratory document LA-UR-99-3541.