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Analysis of FRAM Measurements to Establish Methodology Uncertainty

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OAK RIDGE National Laboratory

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Nuclear Nonproliferation Division

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1. INTRODUCTION

Qualification measurements were performed at Oak Ridge National Laboratory (ORNL) on a variety of measurement standards. These measurements will be used to qualify the Fixed-Energy, Response Function Analysis with Multiple Efficiency (FRAM) code with a high purity germanium (HPGe) detector for use at ORNL. Measured items include uranium isotopic standards, U_3O_8 line sources, and a uranium tetrafluoride source. The ^{235}U enrichment was estimated using the FRAM isotopic analysis software for each source and compared with the declared isotopic values.

1.1 DESCRIPTION OF FRAM

FRAM is a code developed by Los Alamos National Laboratory to analyze pulse-height spectra generated by high-resolution gamma-ray detectors. FRAM calculates the isotopic ratios of U and Pu using a peak ratio method and relative efficiency curve, thus a calibration is not required for isotopic fractions of ^{234}U ^{235}U, and ^{238}U. The calculations are theoretically independent of sample size, shape, physical and chemical composition, and measurement geometry. There is a dependence on the material's separation date, as peaks from ^{234m}Pa (the daughter of ^{238}U) are used in the calculation; but the software includes a correction if the separation date is known. Determination of ^{236}U isotopic fraction does require a calibration as it is an isotopic correlation to the ^{235}U and ^{238}U isotopic fractions [1].

Gamma-ray isotopic analysis is often performed in conjunction with other nondestructive assay techniques, such as calorimetry, neutron coincidence counting, and other bulk measurement systems (for example, active well coincidence counter, segmented gamma scanner, and solution assay instrument), which require foreknowledge of a material's isotopic composition. It is used extensively at Los Alamos National Laboratory for process control and nuclear material control and accountability.

The primary components of uncertainty are counting statistics and fitting of the relative efficiency curve. Validation and extensive determination of the uncertainty of the technique was performed by its creators and published in two application guides by Los Alamos National Laboratory [1, 1]. The uncertainty of ^{235}U enrichment was <5% with a 0.25% bias.

Additional published data on highly enriched uranium (HEU) items indicate impurities in the materials may result in poor peak fitting, thereby increasing the uncertainty. Thick absorbing materials between the detector and the material, such as containers or process equipment, can also increase uncertainty [3]. Methods for reducing the uncertainty are presented in Myers et al. [4] and will be consulted when needed.

1.2 INTENDED USE

For ORNL, it is envisioned that FRAM will be used with high-purity germanium detectors to determine isotopics and combined with other techniques to determine the ^{235}U and total U mass. FRAM may also be used in combination with a weight measurement if the characteristics of the material are well known.

Note that these measurements were performed in a geometry that is not considered optimal for FRAM.

2. MEASUREMENT CONTROL

2.1 METHODOLOGY

Measurement control limits with a point source are established for each individual detector and shall be conducted before use. Results will be written in a registered logbook and entered in a spreadsheet for monitoring via control chart. The statistical control rules established in Department of Energy (DOE) Standard 1194-2019 [5], regarding warning and alarm limit violations, will be applied. If a control measurement exceeds the "Warning" limit, two repeat measurements will be performed. If the second measurement is outside the "Warning" limit, the detector owner will be contacted, and the detector will be removed from service until the issue has been identified and resolved. Similarly, if an "Alarm" limit is violated, the detector will immediately be removed from service until the issue has been identified and resolved.

2.2 MEASUREMENT GEOMETRY

A ^{137}Cs point source was used for measurement control. Two peaks were used to monitor the performance of the detector, the 32.2 keV peak and the 661.6 keV peak. The control measurement was taken at the start of each day on which qualification measurements were taken. The source was placed 10 cm away from the face of the detector and measured for 120 seconds. The source was placed in a metal holder designed for a source of its size and centered with the detector face.

When measurements were taken on the ISOCS (In Situ Object Counting System) cart and shield assembly, the source was placed 30 cm from the detector. To account for the reduced count rate, the count time was extended to 360 seconds. Figure 1 shows the setup for the control measurements taken at 30 cm. This geometry is not ideal and will be used only when necessary.

Figure 1. Measurement control setup.

2.3 DETERMINATION OF CONTROL LIMITS

Control limits for the HPGe detector have been set according to measurable characteristics that demonstrate the functionality of the detector and electronics. Three characteristics were chosen: the peak

full width at half-maximum (FWHM), the peak centroid channel, and the peak centroid in energy. The FWHM is monitored to determine the effective operation of the germanium detector. Monitoring of the peak centroid in terms of energy monitors the energy calibration and the linearity of the multi-channel analyzer. Finally, monitoring the peak centroid channel provides information regarding the stability of the electronics.

The FWHM is treated as a random variable that will follow a normal distribution. Therefore, the mean and standard deviation of the peak FWHM were determined from the nine control measurements collected during the qualification measurement campaign, although 30 control measurements are best practice. The warning limit was set as the mean plus two standard deviations and the alarm limit as the mean plus three std. Only upper limits were established, as smaller FWHM values are not indicative of a malfunction. The control limits for the FWHM of the ^{137}Cs peaks are shown in Table 1.

Table 1. Control limits for the FWHM of ^{137}Cs peaks

Parameter	FWHM (eV)	
	32.2 keV	661.6 keV
Mean	0.809	1.263
Standard deviation	0.064	0.041
Warning limit	0.936	1.346
Alarm limit	1.000	1.387

In general, germanium detectors and their multi-channel analyzers are fairly stable. Therefore, the position of a gamma ray in the multi-channel analyzer does not fluctuate in a random fashion and control limits can be set as fixed upper and lower bounds instead of using the 2x and 3x standard deviation control limits. The control limits are selected based on analysis settings to ensure gamma rays of interest can be identified by the identification routines; the peak centroid must be within 1 keV of the nominal peak energy. Therefore, the control limits have been established so that the warning limits are the nominal peak energy ±0.5 keV and the alarm limits are ±1 keV. The control limits for the peak centroid in channels have been established based on simply the conversion of these limits into channels using the energy calibration. The control limits on the peak centroid, in both energies and channels, are presented in Table 2 and Table 3.

Table 2. Control limits for the peak centroid in energy (keV)

Nominal peak energy	Lower control limits		Upper control limits	
	Alarm	Warning	Warning	Alarm
32.2 keV	31.19	31.69	32.69	33.19
661.6 keV	660.66	661.16	662.16	662.66

Table 3. Control limits for the peak centroid in channels

Nominal peak energy	Expected channel	Lower control limits		Upper control limits	
		Alarm	Warning	Warning	Alarm
32.2 keV	87.89	85.15	86.52	89.26	90.63
661.6 keV	1811.04	1808.30	1809.67	1812.40	1813.77

3. QUALIFICATION MEASUREMENTS

Qualification measurements were taken using a liquid nitrogen–cooled Broad Energy Germanium Detector (BEGe), serial number 13211, along with an Inspector 2000 Digital Signal Processing Portable Spectroscopy Workstation, both of which are produced by Canberra Industries (now Mirion Technologies). Data acquisition and analysis for qualification measurements followed an internal operating procedure, summarized in Section 3.1.

Several measurement standards of varying forms and enrichments were used for the qualification measurements. Measured items included uranium enrichment standards, filter papers coated with U_3O_8, U_3O_8 line sources, and a uranium tetrafluoride source.

3.1 OVERVIEW OF MEASUREMENT PROCEDURE

Measurements were taken using the BEGe detector, the Inspector 2000, and the Genie 2000 Gamma Acquisition and Analysis software. The hardware settings used for these measurements are shown in Table 4. The energy calibration was performed using three sources: ^{241}Am, ^{137}Cs, and ^{60}Co.

Table 4. Inspector 2000 settings used for qualification measurements

Parameter	Value
High voltage	+3000 V
Conversion gain	8192
Course gain	×10
Fine gain	1.3361
Input polarity	Negative
Pole zero	3103

The detector was placed on a flat surface facing the center of the source being measured. A control measurement was performed each day for 120 seconds with a ^{137}Cs point source before any qualification measurements were taken. Most measured items were placed either 10 or 30 cm away, with a few exceptions depending on the number of counts, dead time, and size of the item. The source-to-detector distance and the number of measurements for each item are defined in the following sections. The average dead time was under 25% for each item. Each qualification measurement was acquired for a live time of 600 seconds. Several background measurements were taken overnight for 12 hours each.

Before continuing to the analysis, the background can be subtracted from the measurement spectra at this point using the Genie 2000 Gamma Acquisition and Analysis software; but this step is not always necessary. The background measurements taken showed no significant peaks, so this step was omitted.

The FRAM isotopic analysis software version 5.2 was used to determine the measured enrichment of each source. The analyses were performed using the coaxial parameter sets covering a range of 120 to 1010 keV. The HEU and low-enriched uranium (LEU) parameter sets were selected, depending on whether the declared enrichment was greater than 20%. These parameter sets are called *UHEU_Cx_120-1010* and *ULEU_Cx_120-1010* in the FRAM software. The energy calibration slope and offset values were set to 0.3653 keV/Ch and 0.0885 keV, respectively. These values can be found in the Genie 2000 Gamma Acquisition and Analysis software by using the *Calibrate/Energy Show* feature. In FRAM, the *Ecal* radio button can be selected to verify the peaks are properly aligned. Figure 2 shows an example of the spectrum in FRAM with the *Ecal* peaks identified and the proper parameters set. The spectrum was analyzed and the report saved for each measured source.

For most sources, the default attenuator, cadmium, was used for the FRAM analysis as it is part of the default parameter sets. The analysis will iterate over the areal density and thickness of the attenuator material to determine the appropriate corrections for attenuation, if any are needed. For measurements collected with the line sources inside a copper pipe, the iron attenuator more closely matched the physical attenuation properties of the metal. The mass attenuation coefficient for cadmium is significantly different for gamma ray energies < 400 keV, so using the iron attenuator should produce more accurate results. The attenuator material is changed by selecting the "Efficiency defaults" button on the *Analyze* window.

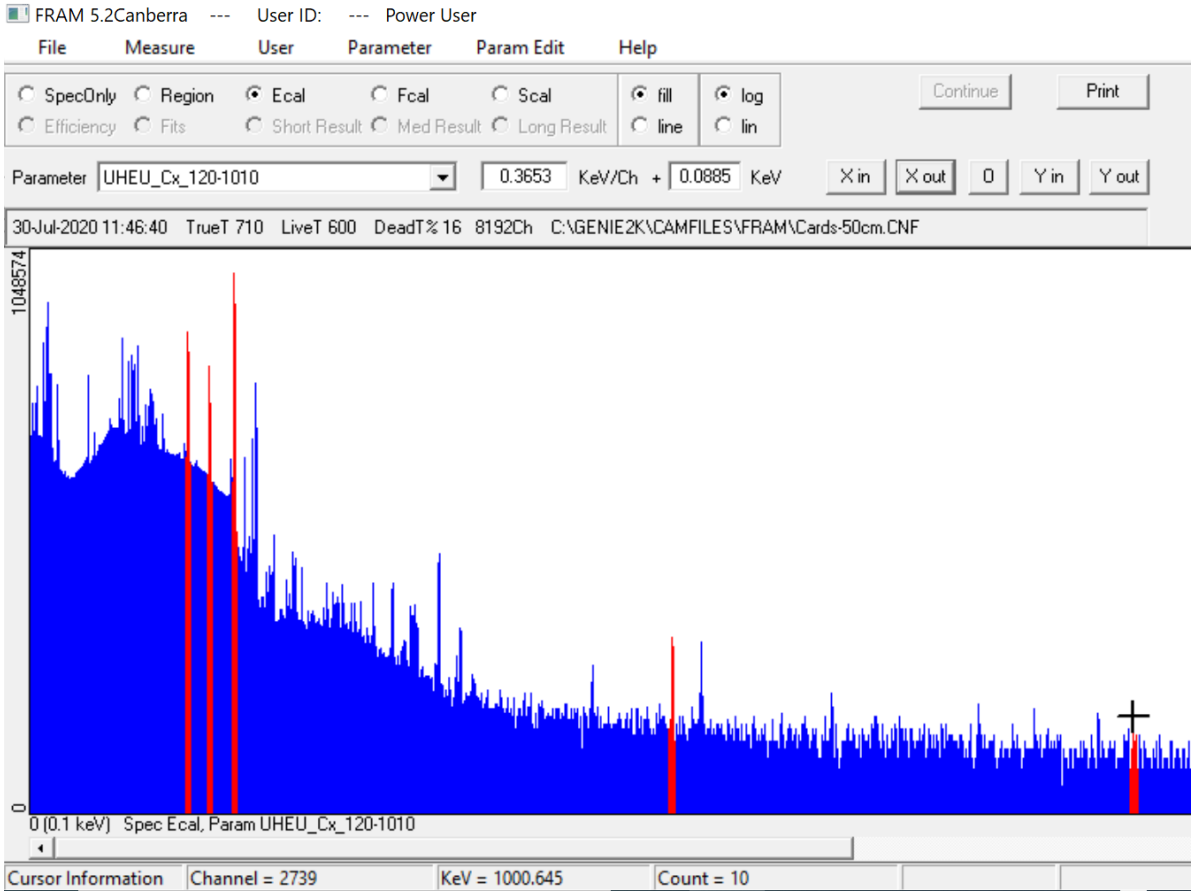

Figure 2. FRAM isotopic analysis software with an example spectrum open. The *Ecal* feature is selected to highlight the relevant peaks, and the proper parameters are selected.

3.2 MEASUREMENTS OF URANIUM ISOTOPIC REFERENCE STANDARDS

3.2.1 Description of Standards

The New Brunswick Laboratory and National Bureau of Standards uranium standards used have enrichments ranging from depleted uranium to HEU. These standards contain U_3O_8 in sealed aluminum cans. These Certified Reference Materials and Standard Reference Materials are intended for use for calibration and evaluation of gamma-ray counting for nondestructive assay of uranium isotopic analysis in uranium bulk materials. The associated certificates issued by New Brunswick Laboratory [6, 7] include certified values for the isotopic fractions of all the uranium isotopes, as well as their masses. Each standard has a 2 mm–thick wall of aluminum in front of the U_3O_8 source material. The material present in the cans represents infinite thickness for 186 keV gamma rays. The declared isotopics of these standards are shown in Table 5.

Table 5. Isotopic fractions of the uranium isotopic reference standards [6, 7]

Name	^{235}U fraction (wt %)	^{234}U fraction (wt %)	^{236}U fraction (wt %)	^{238}U fraction (wt %)
Standard A	0.3166 ± 0.0002	0.0020 ± 0.0002	0.0146 ± 0.0003	99.6668 ± 0.0004
Standard B	0.7119 ± 0.0005	0.0052 ± 0.0002	<0.00002	99.2828 ± 0.0004
Standard C	1.9420 ± 0.0014	0.0171 ± 0.0002	0.0003 ± 0.0001	98.0406 ± 0.0018
Standard D	2.9492 ± 0.0021	0.0279 ± 0.0004	0.0033 ± 0.0002	97.0196 ± 0.0029
Standard E	4.4623 ± 0.0032	0.0359 ± 0.0003	0.0068 ± 0.0002	95.4950 ± 0.0032
Standard F	20.107 ± 0.020	0.14861 ± 0.00037	0.1973 ± 0.0013	79.547 ± 0.020
Standard G	52.488 ± 0.042	0.3718 ± 0.0010	0.26451 ± 0.00060	46.876 ± 0.043
Standard H	93.1703 ± 0.0052	0.9800 ± 0.0029	0.2937 ± 0.0022	5.5559 ± 0.0053

3.2.2 Measurement Geometries

Standards C and F were chosen to be measured 30 times each, and the remaining 6 standards were measured once each. Each measurement had a live time of 600 seconds. Standards with an enrichment below 20% were placed 10 cm away from the face of the detector. Standards with an enrichment above 20% were placed 30 cm away. This helped to keep the average dead time below 25%. The measurement setup for Standards F-H is shown in Figure 3. For the FRAM analysis, the LEU parameter set was used for Standards A through E, and the HEU parameter set was used for Standards F through H.

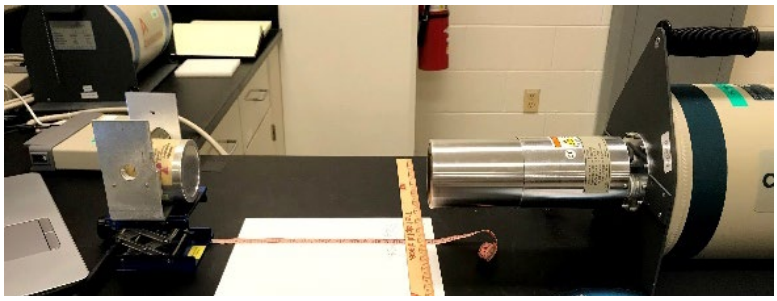

Figure 3. Side view of measurement setup for New Brunswick Laboratory Isotopic Standards F, G, and H. The source-to-detector distance is 30 cm.

3.3 UF_4 WORKING STANDARD

3.3.1 Description of Standard

The UF_4 working standard is a plastic vial containing UF_4 mixed with a slow-setting epoxy. The plastic vial is wrapped in approximately 2 mm of plastic, as sealed sources require two layers of encapsulation. The vial is approximately 7 cm tall and 2 cm in diameter. The isotopics of this standard are shown in Table 6. Note that this standard is not infinitely thick to 186 keV gamma rays.

Table 6. Uranium enrichment values for UF_4 working standard

Isotopes	Isotopic fraction (wt %)
^{234}U	1.015
^{235}U	93.165
^{236}U	0.436
^{238}U	5.385

3.3.2 Measurement Geometry

The UF_4 vial was placed on a stand 10 cm directly in front of the detector, as shown in Figure 4. It was measured 30 times for a live time of 600 seconds each. For the analysis of this standard, the HEU parameter set was used with the default Cd attenuator.

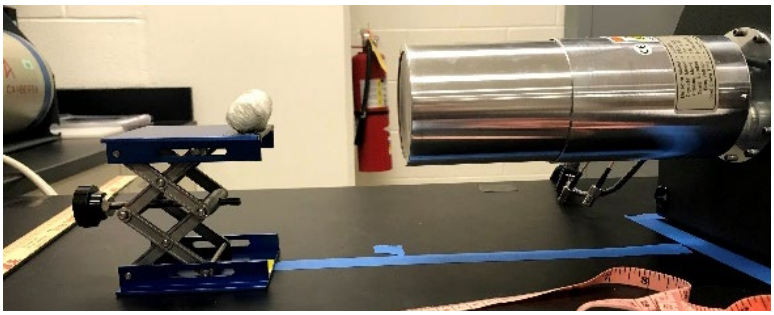

Figure 4. Measurement setup for the UF_4 working standard. The source to detector distance is 10 cm.

3.4 65% ENRICHED LINE SOURCES

3.4.1 Description of Standard

Several U_3O_8 line sources are used in different measurement setups. These long, thin "sticks" consist of U_3O_8 sealed in a plastic tube. Each line source is 65.75% enriched and is 30 cm in length. Figure 5 shows an image of similar line sources. The 65.75% enriched line sources look nearly identical. These line sources are not infinitely thick to 186 keV gamma rays. The uranium isotopics are shown in Table 7.

Figure 5. Photograph of line sources.

Table 7. Uranium enrichment values for the line sources

Isotope	Enrichment (wt %)
^{234}U	1.190
^{235}U	65.747
^{236}U	19.697
^{238}U	13.366

3.4.2 Measurement Geometry

Three 65.75% enriched U_3O_8 line sources were placed in a line inside a copper pipe. The pipe was centered horizontally in front of the detector. The pipe was 89 cm long and was placed 50 cm away from the detector. A standard ISOCS shield/collimator assembly was used for this setup. For this item, 30 measurements were taken for a live time of 600 seconds each. Figure 6 shows the setup using the copper pipe. The HEU parameter set and the Fe attenuator were used for the analysis of this measurement setup.

Figure 6. Measurement setup for copper pipe with uranium oxide.

3.5 URANIUM DISC STANDARD

3.5.1 Description of Standard

A uranium enrichment standard in the shape of a disc was measured. This uranium disc standard is typically used as a holdup standard. The disc consists of uranium metal alloy sealed with epoxy in a 2.381 cm radius stainless-steel cavity behind a stainless-steel window of 0.159 cm thickness. There is a stainless-steel back welded to the lip of the cavity. A diagram of this standard is shown in Figure 7 and Table 8 shows the uranium isotopics. Note that this item is not infinitely thick to 186 keV gamma rays.

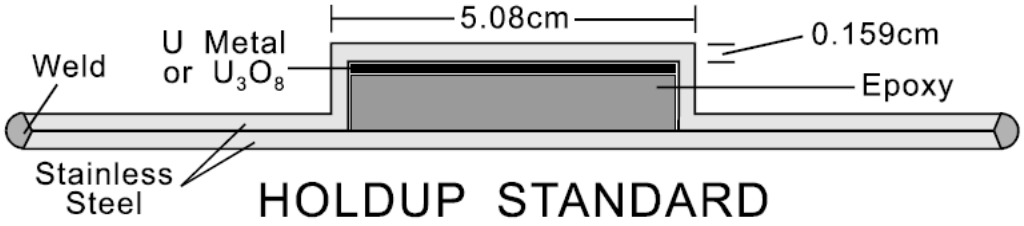

Figure 7. Side view diagram of the uranium disc standard.

Table 8. Uranium enrichment values for the uranium disc standard

Isotope	Isotopic fraction (wt %)
^{234}U	1.016
^{235}U	93.162
^{236}U	0.400
^{238}U	5.421

3.5.2 Measurement Geometry

The uranium disc was placed on a stand and centered 15 cm directly in front of the detector. One measurement was taken for this item with a live time of 600 seconds. Figure 8 shows the measurement setup for the uranium disc. For the FRAM analysis, the HEU parameter set was used with the default Cd attenuator.

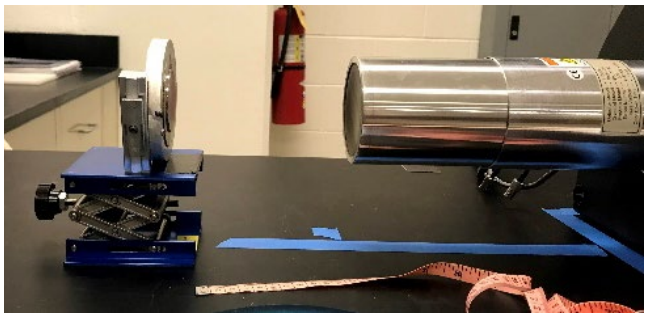

Figure 8. Measurement setup for the uranium disc standard. The source-to-detector distance is 15 cm.

3.6 PERFORMANCE

The replicate measurement results for Standard C, Standard F, the UF_4 vial, and the 65% enriched line sources in the copper pipe are shown in Appendix A. These items were measured 30 times each, so an average measured enrichment is compared with the declared enrichment for these items.

The first analyses were performed using the coaxial parameter sets identified in Section 3.1. Table 10 presents the mean analysis values for each of the primary uranium isotopes using the indicated parameter set. There was a slight bias for each, which was especially noticeable for the line sources with the default physical efficiency parameters. The attenuation material was then changed for the line source measurements to Fe from Cd, as the attenuation cross section of Fe is closer to Cu than Cd is at 186 keV. As the isotopic fractions are certified only for the New Brunswick Laboratory isotopic standards, the replicate measurements of Standard C and Standard F were used to estimate the bias and standard

deviation (equivalent to the root mean square error) presented in Table 9. Performance for ^{236}U isotopic fraction is not included due to the very small content of ^{236}U present in the standards. As a result of the Cu pipe measurement results, we do not recommend the use of FRAM when thick containers are present, or when the containers or attenuators are not in the default list of attenuating materials.

One item of note in the analysis results is that ^{236}U is determined by correlation, as it does not have prominent gamma-ray signatures. The line sources in the copper pipe included a large amount of ^{236}U, as the source material was derived from recycled uranium. The correlation algorithm is based on isotopic correlations found in material enriched by gaseous diffusion. The measurements reflect that the correlation coefficients are inappropriate for the material measured. The analysis results for the ^{236}U isotopic fractions are included in the reported results, but FRAM is not recommended for ^{236}U isotopic fraction determination.

Results for the remaining items, which were measured once, are shown in Table 11 and Table 12. The measured enrichment is compared with the declared enrichment along with the reported relative uncertainty and relative error. The uncertainty reported by FRAM includes only the random error contributors.

Table 9. Standard deviation and bias based on the pooled variance of Standard C and Standard F replicate measurements using the coaxial parameter sets

	^{234}U	^{235}U	^{238}U
Isotopic fraction range (wt %)	0.017–0.149	1.942–20.107	79.547–98.041
Bias	1.24%	1.04%	–0.30%
Estimated measurement standard deviation	9.15%	6.98%	1.44%

Table 10. Results from FRAM analysis using coaxial parameter sets of the replicate measurements

Measurement ID		Standard C	Standard F	UF_4 Vial	Copper pipe
FRAM parameter set		ULEU_Cx_120-1010	UHEU_CX_120-1010	UHEU_CX_120-1010	UHEU_CX_120-1010 with Fe attenuator
^{234}U	Declared isotope fraction	0.0171	0.1486	1.015	1.190
	Avg measured isotope fraction	0.0175	0.1492	1.025	1.316
	Relative standard deviation	9.35%	8.77%	3.77%	14.3%
	Avg measurement error	2.28%	0.37%	0.94%	10.6%
^{235}U	Declared isotope fraction	1.942	20.107	93.165	65.747
	Avg measured isotope fraction	1.929	20.671	93.306	78.095
	Relative standard deviation	6.12%	7.65%	1.32%	13.6%
	Avg measurement error	–0.68%	2.81%	0.15%	18.8%
^{236}U	Declared isotope fraction	0.0003	0.197	0.436	19.697
	Avg measured isotope fraction	0.0122	0.108	0.354	0.326
	Relative standard deviation	5.69%	6.91%	0.65%	8.24%
	Avg measurement error	3972%	–45.3%	–18.7%	–98.3%
^{238}U	Declared isotope fraction	98.041	79.547	5.385	13.366
	Avg measured isotope fraction	98.041	79.072	5.315	20.262
	Relative standard deviation	0.12%	2.02%	23.3%	53.5%
	Avg measurement error	0.001%	–0.60%	–1.30%	51.6%

Table 11. Results from FRAM analysis of LEU items measured once using coaxial parameter set ULEU_Cx_120-1010

	Measurement ID	Standard A	Standard B	Standard D	Standard E
^{234}U	Declared isotope fraction	N/A	N/A	0.0279	0.0359
	Measured isotope fraction	Below	Below	0.0236	0.0345
	Reported uncertainty	Sensitivity	Sensitivity	7.69%	7.43%
	Measurement error	Limit	Limit	–15.4%	–3.90%
^{235}U	Declared isotope fraction	0.3166	0.7119	2.9492	4.4623
	Measured isotope fraction	0.2974	0.8070	2.5079	4.3288
	Reported uncertainty	4.46%	4.28%	3.20%	2.84%
	Measurement error	–6.06%	13.4%	–15.0%	–2.99%
^{236}U	Declared isotope fraction	0.0146	0.00002	0.0033	0.0068
	Measured isotope fraction	0.0022	0.00550	0.0156	0.0258
	Reported uncertainty	4.22%	4.17%	3.13%	2.80%
	Measurement error	–84.9%	27400%	373%	279%
^{238}U	Declared isotope fraction	99.6668	99.2828	97.0196	95.495
	Measured isotope fraction	99.6991	99.1821	97.4529	95.611
	Reported uncertainty	0.01%	0.04%	0.08%	0.13%
	Measurement error	0.03%	–0.10%	0.45%	0.12%

Table 12. Results from FRAM analysis of HEU items measured once using the coaxial parameter set UHEU_Cx_ 120-1010

	Measurement ID	Standard G	Standard H	U Disc
^{234}U	Declared isotope fraction	0.3718	0.9800	93.1620
	Measured isotope fraction	0.3732	0.9730	91.3080
	Reported uncertainty	12.8%	14.8%	2.43%
	Measurement error	0.38%	–0.71%	–1.99%
^{235}U	Declared isotope fraction	52.49	93.17	1.016
	Measured isotope fraction	51.89	92.95	1.215
	Reported uncertainty	6.50%	1.66%	23.6%
	Measurement error	–1.15%	–0.24%	19.6%
^{236}U	Declared isotope fraction	0.2645	0.2937	0.4000
	Measured isotope fraction	0.2433	0.3559	0.3559
	Reported uncertainty	8.50%	6.58%	11.3%
	Measurement error	–8.02%	21.2%	–11.0%
^{238}U	Declared isotope fraction	46.876	5.5559	5.421
	Measured isotope fraction	47.495	5.7205	7.121
	Reported uncertainty	7.15%	27.1%	31.0%
	Measurement error	1.32%	2.96%	31.4%

The detector used for the measurements is a semi-planar p-type HPGe, which has higher efficiency at lower energies than coaxial HPGe detectors. Therefore, the FRAM analyses were repeated using the planar parameter sets available in FRAM. These parameter sets, ULEU_Plnr_060-250 and UHEU_Plnr_060-250, evaluate the x-ray region and low-energy gamma rays. Table 13 through Table 16 present the measurement results and uncertainty analysis using the planar parameter set.

Table 13. Standard deviation and bias based on the pooled variance of Standard C and Standard F replicate measurements using the planar parameter sets

	^{234}U	^{235}U	^{238}U
Isotopic fraction range (wt %)	0.017–0.149	1.942–20.107	79.547–98.041
Bias	−1.04%	0.63%	−0.28%
Estimated measurement standard deviation	0.30%	6.34%	4.46%

Table 14. Mean results from FRAM analysis using planar parameter sets of the replicate measurements.

Measurement ID		Standard C	Standard F	UF_4 Vial	Copper pipe
FRAM parameter set		ULEU_Plnr_060-250	UHEU_Plnr_060-250	UHEU_Plnr_060-250	UHEU_Plnr_060-250 with Fe attenuator
^{234}U	Declared isotope fraction	0.0171	0.14861	1.015	1.19
	Avg measured isotope fraction	0.0168	0.15	1.01	1.51
	Relative standard deviation	7%	1%	2%	8%
	Avg measurement error	–1.56%	–1.01%	–0.13%	26.95%
^{235}U	Declared isotope fraction	1.942	20.11	93.17	65.75
	Avg measured isotope fraction	1.914	20.66	94.47	84.52
	Relative standard deviation	1%	1%	1%	5%
	Avg measurement error	–1.45%	2.73%	1.40%	28.54%
^{236}U	Declared isotope fraction	0.0003	0.20	0.44	19.70
	Avg measured isotope fraction	0.0121	0.11	0.35	0.35
	Relative standard deviation	1%	1%	1%	3%
	Avg measurement error	3943%	–45%	–19%	–98%
^{238}U	Declared isotope fraction	98.0406	79.547	5.385	13.366
	Avg measured isotope fraction	98.0571	79.09	4.16	13.63
	Relative standard deviation	0.029%	0.3%	26.3%	34.5%
	Avg measurement error	0.017%	–0.58%	–22.70%	1.95%

Table 15. Results from FRAM analysis of LEU items measured once using the planar parameter set ULEU_Plnr_060-250

	Measurement ID	Standard A	Standard B	Standard D	Standard E
^{234}U	Declared isotope fraction	N/A	N/A	0.2790	0.0359
	Measured isotope fraction	Below	Below	0.0282	0.0328
	Measurement uncertainty	Sensitivity	Sensitivity	6.03%	5.49%
	Measurement error	Limit	Limit	–89.89%	–8.64%
^{235}U	Declared isotope fraction	0.3166	0.7119	2.949	4.462
	Measured isotope fraction	0.3095	0.6949	2.947	4.407
	Measurement uncertainty	5.49%	3.66%	1.79%	1.37%
	Measurement error	–2.24%	–2.39%	–0.09%	–1.25%
^{236}U	Declared isotope fraction	0.0146	0.0055	0.0033	0.0068
	Measured isotope fraction	0.0023	0.0048	0.0181	0.0262
	Measurement uncertainty	4.35%	4.17%	1.66%	1.53%
	Measurement error	–84.25%	–13%	448%	285%
^{238}U	Declared isotope fraction	99.6668	99.2828	97.020	95.611
	Measured isotope fraction	99.6871	99.2954	97.007	95.534
	Measurement uncertainty	0.02%	0.03%	0.06%	0.06%
	Measurement error	0.02%	0.01%	–0.01%	–0.08%

Table 16. Results from FRAM analysis of HEU items measured once using the planar parameter set UHEU_Plnr_060-250

	Measurement ID	Standard G	Standard H	U Disc
^{234}U	Declared isotope fraction	0.3718	0.9800	1.0160
	Measured isotope fraction	0.3792	0.9282	1.0557
	Reported uncertainty	5.70%	7.34%	7.53%
	Measurement error	1.99%	–5.29%	3.91%
^{235}U	Declared isotope fraction	52.490	93.170	93.162
	Measured isotope fraction	55.138	94.134	94.489
	Reported uncertainty	4.47%	7.06%	7.29%
	Measurement error	5.04%	1.03%	1.42%
^{236}U	Declared isotope fraction	0.2645	0.2937	0.4000
	Measured isotope fraction	0.2560	0.3542	0.3525
	Reported uncertainty	4.53%	13.50%	15.15%
	Measurement error	–3.21%	20.60%	–11.88%
^{238}U	Declared isotope fraction	46.876	5.556	5.421
	Measured isotope fraction	44.227	4.584	4.103
	Reported uncertainty	5.64%	146.9%	170.5%
	Measurement error	–5.65%	–17.50%	–24.32%

When one compares the bias and relative standard deviation of the replicate measurements using the isotopic standards, Table 9 and Table 13, the performance is similar without a discernible improvement from switching between the coaxial and planar parameter sets. However, consider the measurement error on the ^{235}U isotopic fraction from the analysis of all measurements. Table 17 presents the minimum and maximum measurement errors analyzed with both the coaxial and planar parameter sets. Significant reduction is evident in the analysis using the planar parameter sets for all but the UF_4 vial, which already had little measurement error.

Table 17. Minimum and maximum measurement errors of ^{235}U enrichment

Measurement scenarios	Coaxial parameter sets		Planar parameter sets	
	Minimum error	Maximum error	Minimum error	Maximum error
Standard C	–16.7%	10.5%	–4.37%	1.25%
Standard F	–11.4%	22.2%	–0.11%	4.67%
UF_4 Vial	–2.2%	2.4%	–2.08%	2.75%
Copper pipe (with Fe attenuator)	–16.2%	46.6%	3.74%	33.9%
All other standards	–15.0%	19.6%	–2.39%	5.04%

Based on the presented data, it is recommended that FRAM be qualified for use in determining the ^{235}U enrichment of uranium materials with an accuracy of 1% and a precision of 6% using the planar parameter sets with a BEGe detector. Thick-walled containers or containers that contain a material that is not an option for attenuator material in the FRAM analysis, are not captured under this proposed accuracy and precision. Further study should be conducted taking into consideration the work performed previously by Sampson, Hypes and Vo [3].

4. SUMMARY

Measurements were performed at ORNL to qualify the FRAM isotopic analysis software with an HPGe detector for use at ORNL. A variety of measurement standards were used for these qualification measurements, including uranium isotopic standards, a U_3O_8 line source, and a uranium tetrafluoride source. The ^{235}U enrichment was estimated using FRAM for each source and compared with the declared isotopic values. The methodology uncertainty for FRAM in determining the ^{235}U enrichment of uranium materials was determined to be an accuracy of 1% and a precision of 6% using the planar parameter sets with a BEGe detector in non-optimal geometries. Use of FRAM for the determination of the ^{236}U isotopic fraction is not recommended.

5. REFERENCES

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APPENDIX A. RESULTS FROM REPLICATE MEASUREMENTS

APPENDIX A. RESULTS FROM REPLICATE MEASUREMENTS

A.1 Results from Analysis Using Coaxial Parameter Set over Energies 120 keV to 1010 keV

Table 18. Complete enrichment analysis results for the UF_4 vial with declared enrichment of 93.17%

Measurement number	Measured enrichment (%)	Measurement relative uncertainty	Measurement relative error
1	91.33	2.85%	-1.97%
2	94.45	1.78%	1.38%
3	94.62	1.61%	1.56%
4	95.33	1.64%	2.32%
5	93.20	1.50%	0.04%
6	92.60	1.67%	–0.61%
7	94.06	1.80%	0.96%
8	91.54	2.36%	–1.75%
9	93.08	2.42%	–0.09%
10	95.01	1.54%	1.98%
11	93.16	1.70%	0.00%
12	92.82	3.52%	–0.37%
13	94.67	1.55%	1.61%
14	92.49	2.16%	–0.73%
15	93.54	1.75%	0.40%
16	92.11	1.75%	–1.14%
17	94.09	1.59%	0.99%
18	93.24	1.46%	0.08%
19	93.64	1.59%	0.52%
20	95.42	1.40%	2.42%
21	92.97	1.67%	–0.21%
22	91.10	2.75%	–2.22%
23	93.17	1.30%	0.01%
24	93.40	1.60%	0.26%
25	91.28	1.87%	–2.02%
26	92.75	1.47%	–0.45%
27	92.52	1.80%	–0.69%
28	94.48	1.35%	1.41%
29	92.02	2.29%	–1.23%
30	95.09	1.63%	2.06%

Table 19. Complete enrichment analysis results for Standard C with declared enrichment of 1.9420%

Measurement number	Measured enrichment (%)	Measurement relative uncertainty	Measurement relative error
1	1.95	2.98%	0.57%
2	2.00	3.17%	3.20%
3	1.88	3.19%	–3.30%
4	1.94	3.08%	–0.29%
5	1.95	3.37%	0.16%
6	2.04	3.10%	5.22%
7	1.93	3.25%	–0.84%
8	1.90	3.76%	–2.40%
9	1.82	3.07%	–6.06%
10	1.95	3.29%	0.56%
11	1.62	3.17%	–16.69%
12	1.95	2.98%	0.39%
13	1.90	3.35%	–1.94%
14	1.95	3.18%	0.25%
15	1.74	3.75%	–10.58%
16	2.06	3.26%	6.09%
17	2.05	3.48%	5.31%
18	1.69	3.67%	–12.91%
19	1.98	2.99%	2.07%
20	2.02	3.80%	3.89%
21	1.90	3.04%	–2.27%
22	1.91	3.14%	–1.48%
23	1.96	3.43%	0.70%
24	2.06	3.15%	5.91%
25	2.10	3.08%	8.27%
26	1.84	3.25%	–5.47%
27	1.92	3.37%	–1.33%
28	1.76	3.67%	–9.54%
29	2.15	3.79%	10.53%
30	1.97	3.73%	1.66%

Table 20. Complete enrichment analysis results for Standard F with declared enrichment of 20.11%

Measurement number	Measured enrichment (%)	Measurement relative uncertainty	Measurement relative error
1	19.72	6.05%	–1.95%
2	24.58	7.72%	22.20%
3	20.52	6.62%	2.06%
4	18.91	6.47%	–5.94%
5	19.57	6.54%	–2.70%
6	22.80	6.03%	13.38%
7	21.42	5.44%	6.50%
8	18.70	6.50%	–7.02%
9	19.55	8.98%	–2.78%
10	19.46	6.51%	–3.22%
11	21.07	9.07%	4.78%
12	18.00	6.81%	–10.51%
13	20.50	7.95%	1.92%
14	19.65	8.42%	–2.31%
15	21.80	7.25%	8.38%
16	19.53	7.26%	–2.88%
17	21.26	5.91%	5.73%
18	22.39	7.92%	11.35%
19	21.04	12.60%	4.64%
20	21.17	7.72%	5.28%
21	17.82	7.11%	–11.39%
22	18.24	5.60%	–9.27%
23	21.70	9.03%	7.92%
24	21.60	7.47%	7.40%
25	22.86	5.18%	13.69%
26	20.74	4.99%	3.12%
27	22.16	11.34%	10.21%
28	22.16	5.67%	10.18%
29	20.15	5.21%	0.20%
30	21.07	7.42%	4.80%

Table 21. Complete enrichment analysis results for line sources in copper pipe with declared enrichment of 65.75%

Measurement number	Measured enrichment (%)	Measurement relative uncertainty	Measurement relative error
1	80.87	20.71%	23.00%
2	76.67	28.08%	16.61%
3	91.21	40.67%	38.73%
4	92.73	24.44%	41.03%
5	65.30	49.02%	–0.68%
6	74.15	30.59%	12.79%
7	95.68	29.80%	45.52%
8	82.40	22.75%	25.33%
9	55.12	22.36%	–16.16%
10	81.95	22.69%	24.64%
11	96.37	23.24%	46.58%
12	86.27	28.05%	31.22%
13	79.20	24.68%	20.46%
14	79.41	27.38%	20.78%
15	56.84	28.08%	–13.54%
16	76.46	27.32%	16.30%
17	82.26	28.20%	25.12%
18	83.18	19.97%	26.51%
19	68.77	31.79%	4.60%
20	73.68	28.47%	12.06%
21	83.33	36.91%	26.75%
22	80.85	25.72%	22.97%
23	69.75	29.71%	6.09%
24	79.06	26.20%	20.25%
25	65.91	24.06%	0.24%
26	82.95	21.90%	26.16%
27	67.05	20.45%	1.98%
28	62.19	26.86%	–5.42%
29	90.53	26.97%	37.70%
30	82.73	24.82%	25.84%

Note: Efficiency default modified by changing attenuator material to Fe.

A.2 Results from Analysis Using Planar Parameter Set over Energies 60 keV to 250 keV

Table 22. Complete enrichment analysis results for the UF_4 vial with declared enrichment of 93.17%

Measurement number	Measured enrichment (%)	Measurement relative uncertainty	Measurement relative error
1	93.20	6.3%	0.03%
2	92.70	6.2%	–0.51%
3	95.73	5.1%	2.75%
4	93.92	6.2%	0.80%
5	95.23	6.2%	2.21%
6	94.99	6.6%	1.95%
7	94.27	6.8%	1.18%
8	95.19	6.2%	2.17%
9	95.13	6.4%	2.10%
10	95.05	6.6%	2.02%
11	94.98	6.5%	1.94%
12	95.02	6.7%	1.99%
13	94.35	6.4%	1.27%
14	93.92	6.6%	0.81%
15	92.94	6.2%	–0.24%
16	95.21	6.2%	2.19%
17	95.02	6.7%	1.99%
18	95.67	5.4%	2.69%
19	95.73	5.3%	2.74%
20	91.23	14.2%	–2.08%
21	92.93	6.5%	–0.26%
22	93.36	6.1%	0.20%
23	95.28	6.1%	2.27%
24	94.15	6.4%	1.06%
25	95.21	6.2%	2.19%
26	95.16	6.4%	2.14%
27	95.09	6.5%	2.06%
28	95.10	6.2%	2.07%
29	93.34	5.9%	0.18%
30	95.06	5.9%	2.03%

Table 23. Complete enrichment analysis results for Standard C with declared enrichment of 1.9420%

Measurement number	Measured enrichment (%)	Measurement relative uncertainty	Measurement relative error
1	1.898	1.4%	–2.25%
2	1.954	2.0%	0.62%
3	1.887	2.0%	–2.84%
4	1.966	2.0%	1.25%
5	1.932	1.9%	–0.52%
6	1.930	1.8%	–0.62%
7	1.929	2.2%	–0.67%
8	1.913	2.4%	–1.48%
9	1.903	1.4%	–2.01%
10	1.892	2.2%	–2.57%
11	1.904	1.4%	–1.98%
12	1.961	1.8%	0.96%
13	1.905	1.4%	–1.89%
14	1.960	2.1%	0.95%
15	1.910	2.6%	–1.65%
16	1.888	1.9%	–2.77%
17	1.876	2.3%	–3.41%
18	1.900	2.1%	–2.15%
19	1.896	2.5%	–2.39%
20	1.857	1.9%	–4.37%
21	1.893	1.8%	–2.50%
22	1.933	1.8%	–0.45%
23	1.862	1.6%	–4.12%
24	1.911	1.2%	–1.62%
25	1.906	1.8%	–1.83%
26	1.908	1.3%	–1.77%
27	1.941	2.4%	–0.04%
28	1.930	1.3%	–0.61%
29	1.919	2.5%	–1.18%
30	1.953	2.1%	0.56%

Table 24. Complete enrichment analysis results for Standard F with declared enrichment of 20.11%

Measurement number	Measured enrichment (%)	Measurement relative uncertainty	Measurement relative error
1	20.59	2.6%	2.38%
2	20.57	2.6%	2.27%
3	21.03	2.8%	4.60%
4	20.73	2.6%	3.08%
5	20.67	2.5%	2.80%
6	20.82	2.6%	3.51%
7	20.85	2.5%	3.69%
8	20.50	2.6%	1.95%
9	20.32	2.8%	1.05%
10	20.72	2.5%	3.05%
11	21.03	2.5%	4.57%
12	20.09	2.8%	–0.09%
13	20.62	2.7%	2.54%
14	20.86	2.8%	3.73%
15	21.05	2.8%	4.67%
16	20.46	2.6%	1.74%
17	20.19	2.5%	0.41%
18	20.96	2.6%	4.23%
19	20.68	2.6%	2.82%
20	20.86	2.5%	3.72%
21	20.43	2.5%	1.57%
22	20.29	2.7%	0.91%
23	20.87	2.6%	3.80%
24	20.93	2.5%	4.07%
25	20.90	2.6%	3.92%
26	20.73	2.5%	3.07%
27	20.66	2.7%	2.74%
28	20.09	2.7%	–0.11%
29	20.66	2.8%	2.74%
30	20.63	2.7%	2.58%

Table 25. Complete enrichment analysis results for line sources in copper pipe with declared enrichment of 65.75%

Measurement number	Measured enrichment (%)	Measurement relative uncertainty	Measurement relative error
1	81.31	19.6%	23.66%
2	81.91	19.6%	24.57%
3	84.21	27.2%	28.08%
4	86.84	20.6%	32.08%
5	86.93	21.3%	32.21%
6	79.49	18.4%	20.90%
7	87.64	19.4%	33.30%
8	81.31	30.6%	23.66%
9	75.22	16.8%	14.40%
10	84.74	19.2%	28.88%
11	81.45	18.0%	23.88%
12	87.99	19.7%	33.83%
13	84.85	19.2%	29.04%
14	79.73	18.5%	21.26%
15	87.80	19.7%	33.54%
16	87.60	20.4%	33.23%
17	87.46	20.3%	33.02%
18	88.00	19.6%	33.85%
19	87.91	19.5%	33.70%
20	85.60	19.5%	30.19%
21	68.21	15.1%	3.74%
22	87.80	19.8%	33.54%
23	87.90	19.8%	33.69%
24	87.68	20.1%	33.35%
25	87.36	21.0%	32.86%
26	87.25	20.9%	32.70%
27	80.14	18.4%	21.88%
28	87.69	20.0%	33.37%
29	85.92	20.2%	30.68%
30	87.57	20.2%	33.18%

Note: Efficiency default modified by changing attenuator material to Fe.