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FRAM Version 7.1's Bias

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ABSTRACT

The Fixed-Energy Response-Function Analysis with Multiple Efficiency (FRAM) code was developed at Los Alamos National Laboratory to measure the gamma-ray spectrometry of the isotopic composition of plutonium, uranium, and other actinides. For FRAM versions 4 and earlier, the reported uncertainties of the results come from the propagation of the statistics in the peak areas only. No systematic error components are included in the reported uncertainties. For FRAM versions 5 and 6, we examined the FRAM analytical results of both the archival plutonium data and the data specifically acquired for the isotopic uncertainty analysis project and found the relationship between the bias and other parameters. We worked out the equations representing the biases of the measured isotopes from each measurement using internal spectral parameters, such as peak resolution and shape, region of analysis, and burnup (for plutonium) or enrichment (for uranium). The resulting biases were included in the reported uncertainties of FRAM v.5 and v.6.

For the FRAM version 7.1, we are doing the same study that we did for FRAM v.5 and v.6 The resulting biases are included in its reported total uncertainties.

A. INTRODUCTION

The Fixed-Energy Response-Function Analysis with Multiple Efficiency (FRAM) software was developed and continues to be refined by Los Alamos National Laboratory (LANL). The code was developed for gamma-ray spectrometry measurements of the isotopic composition of plutonium, uranium, and other actinides [1–3]. In 2005, we studied FRAM's bias as a function of peak resolution and shape, intending to apply the results to the upgraded version of FRAM [4]. The software in that study was FRAM version 4. In 2011, we did another bias study on FRAM version 5 [5] and applied the bias function to the FRAM v.5 code. In 2019, we did a bias study on FRAM version 6 [6] and applied the bias function to the FRAM v.6 code. In that study, we used different bias formulae than those in v.5. For the bias study of FRAM v.7, we use the same bias formulae as those of FRAM v.6.

In the 2005 study, experiments were set up to obtain spectra of various peak resolutions and shapes to study the relationships of the biases with the spectra's internal parameters (peak resolution and shape, region of analysis, plutonium burnup, or uranium enrichment). The same data were used for the FRAM v.5 study in 2011 and FRAM v.6 study in 2019. For the study reported here, we used the same spectra as in the previous studies.

B. PLUTONIUM (HPGe)

FRAM can analyze a spectrum using any energy region, including the very narrow or very wide energy region, if the region contains the peaks of every isotope. The disadvantage of a very small region analysis is that the statistics may not be very good, since the analysis uses only a small number of peaks in a small region. The disadvantage of a very large region analysis is that, due to the very wide energy range, the efficiency curve may not be completely represented by the efficiency models used by FRAM. This will lead to a bad efficiency curve, which will then lead to bad results.

FRAM normally analyzes a plutonium spectrum using one of the three energy regions: low energy (60–230 keV), medium energy (120–420 keV), and high energy (180–1010 keV). Figure 1 shows a typical plutonium spectrum with these three overlapping analytical regions, which are depicted with three thick horizontal bars.

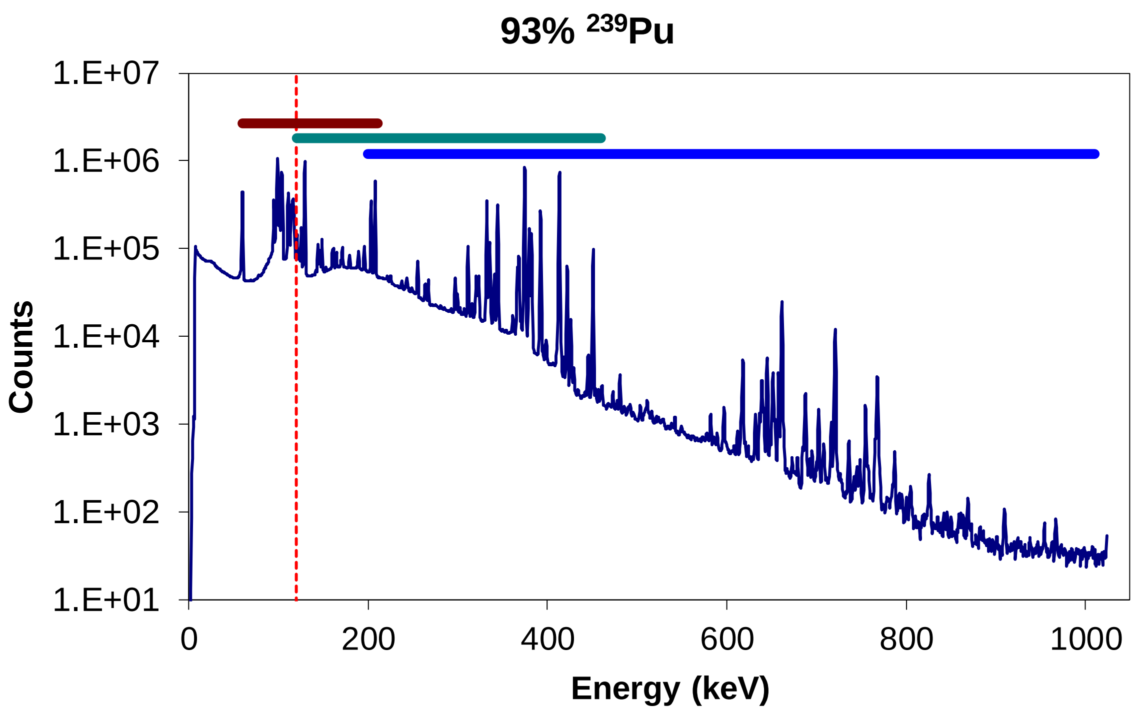

Figure 1. A low-burnup plutonium spectrum. The vertical dashed line denotes the plutonium K-edge. The three overlapping analytical regions that FRAM normally uses for the analysis are shown as three thick horizontal bars above the spectrum.

1. Data acquisition

The spectra were acquired with the electronics adjusted so that each set would have distinctive peak resolutions and shapes. Two detector systems were used for the experiments, one planar germanium detector system and one coaxial germanium detector system. The planar detector system consisted of a 16-mm-diameter × 13-mm-long planar detector from Canberra and the DSPEC Plus multichannel analyzer (MCA) from Ortec. The coaxial detector system consisted of a 58-mm-diameter × 53-mm-long coaxial detector (32% relative efficiency) and the DSPEC Plus MCA, also from Ortec.

The samples for these measurements included four of the seven samples from the plutonium isotopic determination inter-comparison exercise (PIDIE) set: PIDIE-1, PIDIE-3, PIDIE-5, and PIDIE-7. These samples were small, only 0.4 g each. In the planar detector system, the input rates for the four samples were 3, 5, 8, and 10 kHz, respectively, from low to high burnup. For the coaxial detector system, the input rates were 16 kHz for the PIDIE-1 sample and 20 kHz for the other three samples.

The data for the planar detector were acquired in 8-K channels at 0.075 keV/ch up to >600 keV so that the data could be analyzed in two different energy ranges: 60–230 keV (low energy) and 120–500 keV (medium energy). For the coaxial detector, the spectra were acquired in 8K channels at 0.125 keV/ch, covering the entire 0–1,024-keV energy range. These spectra can be analyzed using two separate parameter sets employing the 120–500-keV (medium energy) and 180–1,010-keV (high energy) regions.

In both detector systems, we varied the rise time of the DSPEC Plus to obtain spectra with various resolutions. The rise times used were 0.2, 0.4, 0.6, 1.0, 1.4, 2.0, 2.8, 4.0, and 8.0 µs. The flattop was 1.0 µs, and the cusp value was 0.8. The acquisition time for each spectrum was 15 minutes live time, and 16 spectra were obtained for each dataset. The full-width at half-maximum (FWHM) at 208 keV for the planar detector varied from about 0.64 keV at 8-µs rise time to 0.82 keV at 0.2-µs rise time; for the coaxial detector, the variation was from 1.04 keV to 2.28 keV for the same span of rise times. The peak tails were all small for these measurements. In general, the FWHM is larger for higher-energy peaks and smaller for lower-energy peaks.

To obtain spectra with various shapes, we used a rise time of 4.0 µs and manually adjusted the pole zero (PZ) to produce peaks with low-energy tails of various sizes. For each sample, six sets of data, with 16 spectra (15 minutes true time) in each set, were obtained, with the peak tail percentages varying from approximately zero to about 17%. For a spectrum, the tail of a peak can be either larger or smaller at different energy. On average, the tail is about the same at all different energies.

2. Analysis

a. Correlation

The bias correlation for an isotope is

$$\text{Bias} = a \cdot \boldsymbol{F}^{b} \cdot (\boldsymbol{W} \cdot (1 + c \cdot \boldsymbol{T}))^{d}, \qquad \text{(Eq. 1)}$$

where bias = |Measured/Accepted – 1|; a, b, c, and d are variables, with a, c, and d being nonnegative; ***F*** is the isotopic percent of the isotope; ***W*** is the FWHM (keV) of the selected peak; and ***T*** is the tail percent of that selected peak. For ^{241}Am, the isotopic percent is defined as 100 times the ratio of ^{241}Am to plutonium. The selected peaks are chosen to be the 129-keV peak of ^{239}Pu for the low-energy region analysis, the 208-keV peak of ^{241}Pu and ^{241}Am for the medium-energy region analysis, and the 662-keV peak of ^{241}Am for the high-energy-region analysis.

b. Bias fitting

To obtain the bias for any measurement, we would need to fit the data points to a model, which is described by Equation 1. Normally, when a curve is fitted through some data points, half of the points, on average, will be above the curve and half will be below. Here, we are trying to obtain a curve that would represent the standard deviation or the bias of the data. For a Gaussian distribution, about 32% of the points will be outside one standard deviation, or sigma, and 68% will be inside. So, in fitting Equation 1, we gave the points above the curve a weight of 0.68, and the points below the curve a weight of 0.32. Then the bias curve is obtained such that roughly 32% of the points are above it and 68% are below it. In addition to the 32/68 weight distribution, each data point is given a weight equal to the inverse of the statistical error of that data point.

For the low-energy-region analysis, we use the parameter set HPGe_Pu_060-230 to analyze the data of both planar and coaxial detectors. For the medium-energy-region analysis, we use the parameter set HPGe_Pu_120-420 to also analyze the data of both planar and coaxial detectors. For the high-energy-region analysis, the parameter set HPGe_Pu_180-1010 was used to analyze the coaxial data. The physical efficiency model was used for these analyses.

We grouped the average results (of 16 runs at each setup) based on the energy region used in the analysis, regardless of the detector type: low-energy, medium-energy, and high-energy. We fitted the data from each group to Equation 1 to obtain the values for a, b, c, and d.

c. Results

Table 1 shows the results of the fits.

We see that if the widths of the peaks are not zero, some bias will always be associated with the finite resolutions. (The tail can be made zero or very near zero with a good detector system and careful measurements.)

The resolution for a typical planar detector is about 0.55 keV FWHM at 129 keV and 0.7 keV FWHM at 208 keV. For a typical coaxial detector, the FWHM is about 1 keV at 208 keV and 1.45 keV at 662 keV. The tails are about 1% for peaks at all energies. Figure 2 shows the plots of the biases calculated using such FWHM and tail parameters.

Table 1. Results of the fits of the equation Bias = |Measured/Accepted – 1| = $a \cdot \boldsymbol{F}^b \cdot (\boldsymbol{W} \cdot (1 + c \cdot \boldsymbol{T}))^d$, where $\boldsymbol{F}$ is the isotopic percent of the isotope, and $\boldsymbol{W}$ and $\boldsymbol{T}$ are the FWHM and tail of the reference peak. The reference peak is the 129-keV peak for the low-energy region analysis, 208-kev peak for the medium-energy region analysis, and 662-keV peak for the high-energy-region analysis.

Isotope	Low-Energy-Region Analysis			
	a	b	c	d
^{238}Pu	7.46E-02	0.112	0.0000	2.006
^{239}Pu	2.67E+00	-1.291	0.0015	6.360
^{240}Pu	1.90E-02	0.343	0.0029	4.884
^{241}Pu	3.68E-02	0.559	0.0280	2.905
^{241}Am	4.80E-02	0.065	0.0000	2.006
Isotope	**Medium-Energy-Region Analysis**			
	a	b	c	d
^{238}Pu	1.17E-02	-0.247	0.0000	4.541
^{239}Pu	2.31E+02	-2.573	0.0017	4.779
^{240}Pu	8.83E-04	1.160	0.0050	2.514
^{241}Pu	1.04E-02	0.244	0.0123	3.142
^{241}Am	9.29E-03	0.461	0.0000	1.551
Isotope	**High-Energy-Region Analysis**			
	a	b	c	d
^{238}Pu	1.99E-02	0.073	0.1003	0.276
^{239}Pu	4.33E+00	-1.920	0.3043	0.789
^{240}Pu	1.17E-03	0.944	0.7483	0.122
^{241}Pu	1.08E-02	0.409	0.0501	0.101
^{241}Am	5.35E-03	0.614	1.9824	0.074

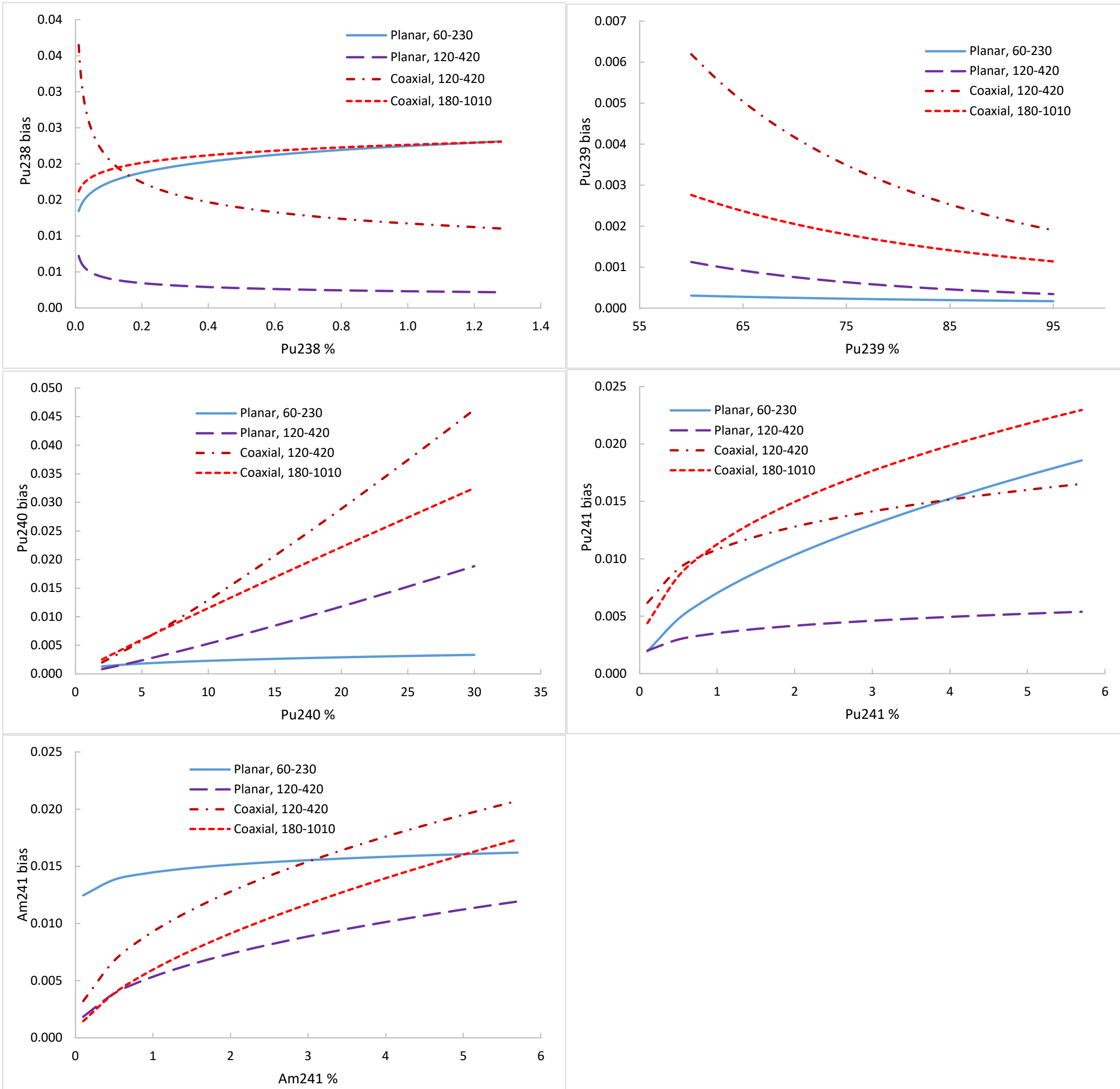

Figure 2. Calculated biases for typical planar and coaxial detector systems.

C. URANIUM (HPGe)

Figure 3 shows an example of a uranium spectrum with two regions, one below the K-edge and one above the K-edge, separated by the dashed line. The two thick horizontal bars above the spectrum represent the two overlapping analytical regions (low and high) that FRAM normally uses for the analysis.

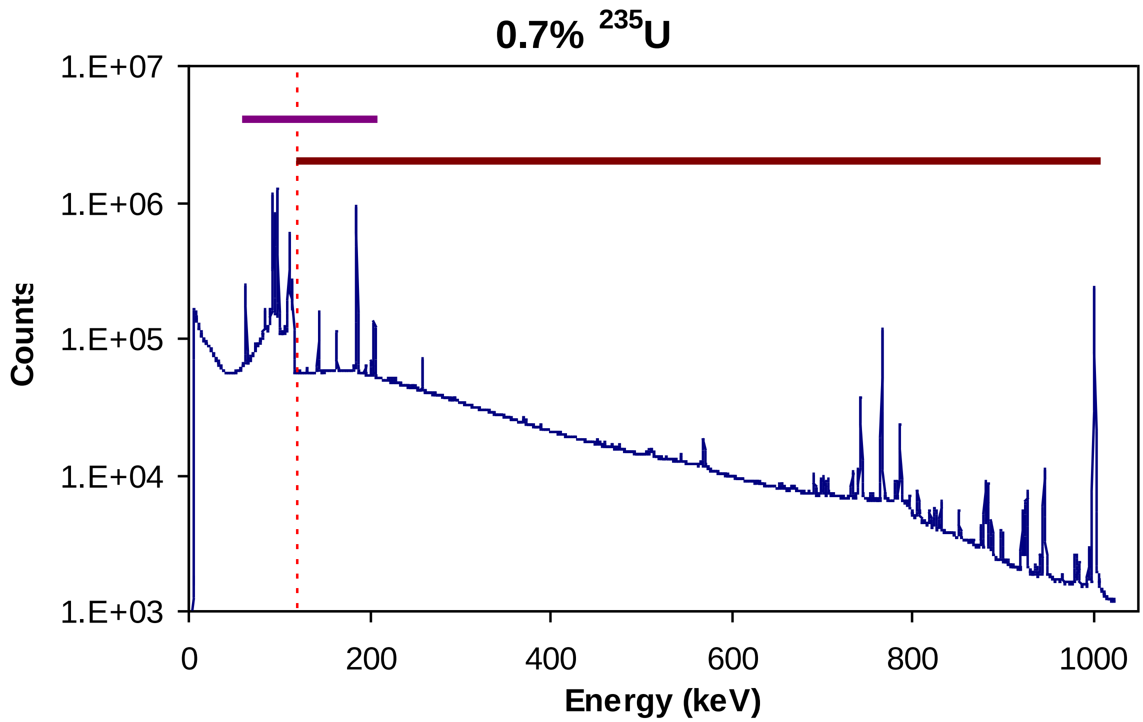

Figure 3. A natural uranium spectrum. The vertical dashed line denotes the uranium K-edge. The two overlapping analytical regions that FRAM normally uses for the analysis are shown as two thick horizontal bars above the spectrum.

1. Data acquisition

The data acquisition system was set up the same way as it was for the plutonium bias determination (Section B). For these measurements, five low-enriched uranium (LEU) samples of the NBS-SRM* 969 set and three highly enriched uranium (HEU) samples of the NBL-CRM† 146 set, ranging from 0.3% to 93.2% ^{235}U enrichment, were used. These samples weighed about 200 g each. For the planar detector system, the input rates for the five LEU samples were small, ranging from 1.8 kHz for the 0.3% ^{235}U sample to 3.5 kHz for the 4.5% ^{235}U sample. For the three HEU samples, input rates were at 10 kHz. For the coaxial detector system, the input rates were about 20 kHz for all eight samples.

The data for the planar detector were acquired in 4-K channels at 0.075 keV/ch and analyzed using the peaks in the 60-keV to 210-keV energy range. For the coaxial detector, the spectra were acquired in 8-K channels at 0.125 keV/ch and analyzed using the parameter set employing the 120-keV to 1,010-keV region.

Just as we did for plutonium (Section B), we varied the rise time of the DSPEC Plus to obtain spectra with various resolutions for both detector systems. The rise times used were 0.2, 0.4, 0.6,

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* NBS-SRM - National Bureau of Standards - Standard Reference Materials.
† NBL-CRM - New Brunswick Laboratory - Certified Reference Materials.

1.0, 1.4, 2.0, 2.8, 4.0, and 8.0 µs. The flattop was 1.0 µs, and the cusp value was 0.8. The acquisition time for each spectrum was 15 minutes of live time. The FWHM at 186 keV for the planar detector varied from about 0.61 keV at 8-µs rise time to 0.77 keV at 0.2-µs rise time, and for the coaxial detector, it was from 1.00 keV to 2.32 keV for the same span of rise times. The peak tails were all small for these measurements.

To obtain spectra with various shapes, we used a rise time of 4.0 µs and manually adjusted the PZ to produce peaks with low-energy tails of various sizes. For each sample, six sets of data, with 16 spectra (15 minutes true time) for each set, were obtained, with the 186-keV peak-tail percents varying from approximately zero to about 15%.

2. Analysis

For the low-energy-region analysis, we analyzed the planar and coaxial data using the GePlnr_ULEU_060-250 and GePlnr_UHEU_060-250 parameter sets. For the high-energy-region analysis, we analyzed the coaxial data using the GeCoax_ULEU_120-1010 and GeCoax_UHEU_120-1010 parameter sets. The efficiency model for these analyses was the physical model.

a. Correlation

The correlation equation relating the resolutions and tails of the peaks to the bias of the uranium isotopes is the same as that of plutonium. It is Equation 1,

$$\text{Bias} = a \cdot \boldsymbol{F}^{b} \cdot (\boldsymbol{W} \cdot (1 + c \cdot \boldsymbol{T}))^{d},$$

where bias = |Measured/Accepted – 1|; a, b, c, and d are variables, with a, c, and d being nonnegative; ***F*** is the isotopic percent of the isotope; ***W*** is the FWHM (keV) of the selected peak; and ***T*** is the tail percent of that selected peak. The selected peak is chosen to be the 186-keV peak of ^{235}U for both low- and high-energy region analyses.

The bias fittings of Equation 1 for ^{234}U, ^{235}U, and ^{238}U are done the same way as for plutonium, as described in Section B.2.

b. Results

Table 2 shows the results of the fits.

Like the plutonium analysis, we see that if the widths of the peaks are not zero, some bias will always be associated with the finite resolutions.

The typical FWHM at 186 keV is about 0.65 keV for the planar detector and 0.95 for the coaxial detector. The tails are about 1% for peaks at all energies. Figure 4 shows the plots of the biases calculated using such FWHM and tail parameters.

D. NON-HPGE

We obtained a limited number of spectra with two $LaBr_3$ detectors, two 500-mm^3 CZT detectors, and a large (16-cm^3), pixelized CZT detector (made by H3D). The spectra were of many certified plutonium and uranium items we have in our group at LANL. For the $LaBr_3$ detectors, the average FWHM at 186 keV was 10.5 keV and at 662 keV was 19.3 keV. The peak tail percentage was about zero. One CZT detector was used to measure uranium with an average 4.5 keV FWHM and 22.6% peak tail at 186 keV. Another CZT detector was used to measure plutonium. At 208 keV, the average FWHM was 4.6 keV and the average low energy tail was 16.0%. For the pixelized CZT detector, the average FWHM was 2.6 keV and total (low energy plus high energy) tail was 13.5% at 186 keV and 3.6 keV FWHM and 34% total tail at 662 keV.

Table 2. Results of the fits of the equation Bias = |Measured/Accepted – 1| = a · $\boldsymbol{F}^b$ · $(\boldsymbol{W} \cdot (1 + c \cdot \boldsymbol{T}))^d$, where $\boldsymbol{F}$ is the isotopic percent of the isotope, and $\boldsymbol{W}$ and $\boldsymbol{T}$ are the FWHM and tail of the reference peak. The reference peak is the 186-keV peak of ^{235}U for all the region analysis.

Isotope	Low-Energy-Region Analysis			
	a	b	c	d
^{234}U	3.07E-02	-0.0206	0.0130	2.9748
^{235}U	1.50E-02	0.3174	0.0058	4.3300
^{238}U	1.12E+01	-2.4148	0.1841	0.8870
Isotope	**High-Energy-Region Analysis**			
	a	b	c	d
^{234}U	9.30E-03	-0.1140	0.0000	2.7597
^{235}U	1.08E-02	0.0550	0.0274	0.8143
^{238}U	3.49E-01	-1.4788	0.0988	0.0557

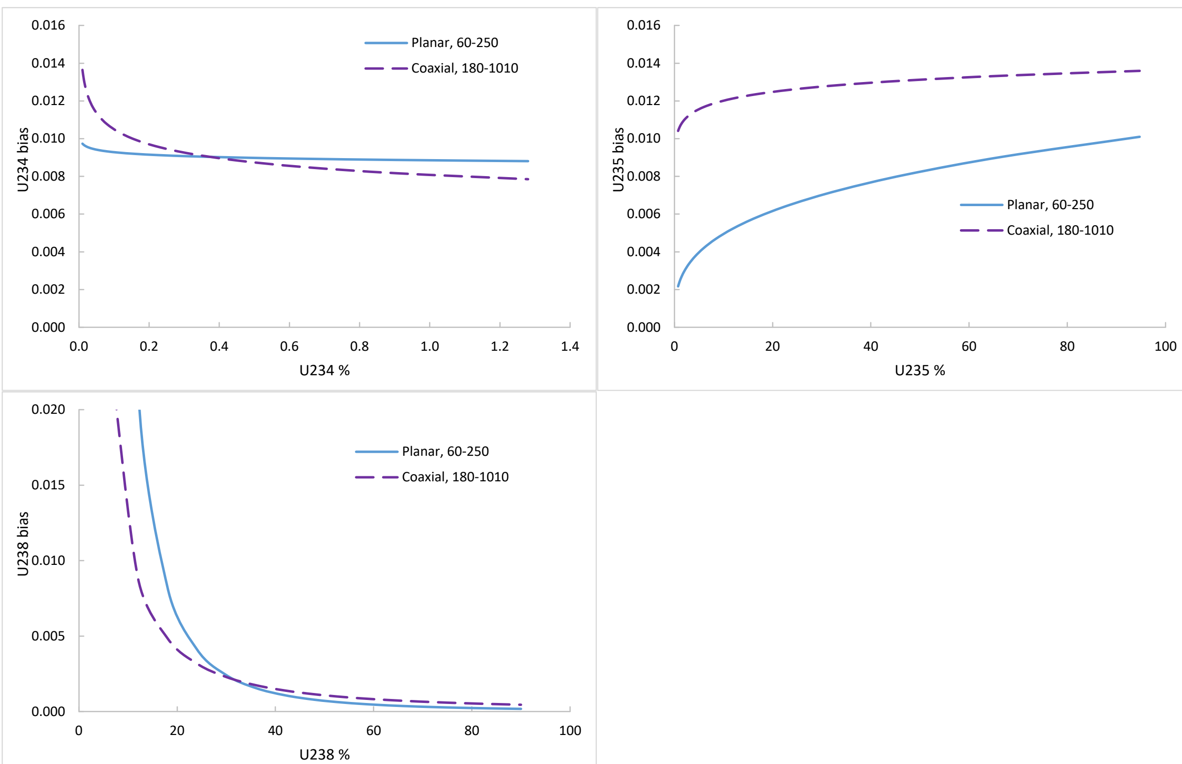

Figure 4. Calculated biases for good planar and coaxial detector systems.

We analyzed the spectra using the corresponding parameter sets LaBr_Pu_200-750, LaBr_U_120-1010, CZT500_Pu_120-500, CZT500_U_120-1010, CZT-H3D_PuRG_060-500, CZT-H3D_PuWG_060-500, CZT-H3D_PuRG_180-800, CZT-H3D_PuWG_180-800, CZT-H3D_ULEU_120-1010, and CZT-H3D_UHEU_120-1010. The measured biases were then compared with the biases calculated from Equation 1. Table 3 shows the measured-bias to calculated-bias ratios. These ratios are entered in the user-designated systematic component for the isotopes in FRAM (under the command syst_error_xxyyy in the Application Constants section of the FRAM parameter set, where xxyyy is the isotope name).

Table 3. Measured-bias to calculated-bias ratios.

Parameter set	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{241}Am
LaBr_Pu_200-750	1.8	7.5	5.4	10.4	16.6
CZT500_Pu_120-500	0.084	0.014	0.21	0.14	5.2
CZT-H3D_PuR(W)G_060-500	0.24	0.073	0.40	0.85	18
CZT-H3D_PuR(W)G_180-800	21	1.7	5.4	21	14

Parameter set	^{234}U	^{235}U	^{238}U
LaBr_U_120-1010	0.020	5.3	21
CZT500_U_120-1010	0.12	1.4	15
CZT-H3D_UL(H)EU_120-1010	1.2	2.0	8.0

E. CONCLUSION

We have studied the bias of FRAM analysis by employing various parameter sets using gamma rays and x-rays in various energy regions of data taken with the HPGe, LaBr3, single crystal CZT, and pixelized CZT detectors. We determined the biases as functions of the resolutions and the tails of the peaks. This method considers the specific measurement conditions for every measurement and estimates the bias based on those measurement conditions.

FRAM v.7.1 includes the systematic uncertainties in addition to the random uncertainties in its results. These systematic uncertainties are based on the biases shown in this paper. The results, with the systematic uncertainties, are shown in the medium and long display modes of FRAM.

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