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FRAM's Isotopic Uncertainty Analysis

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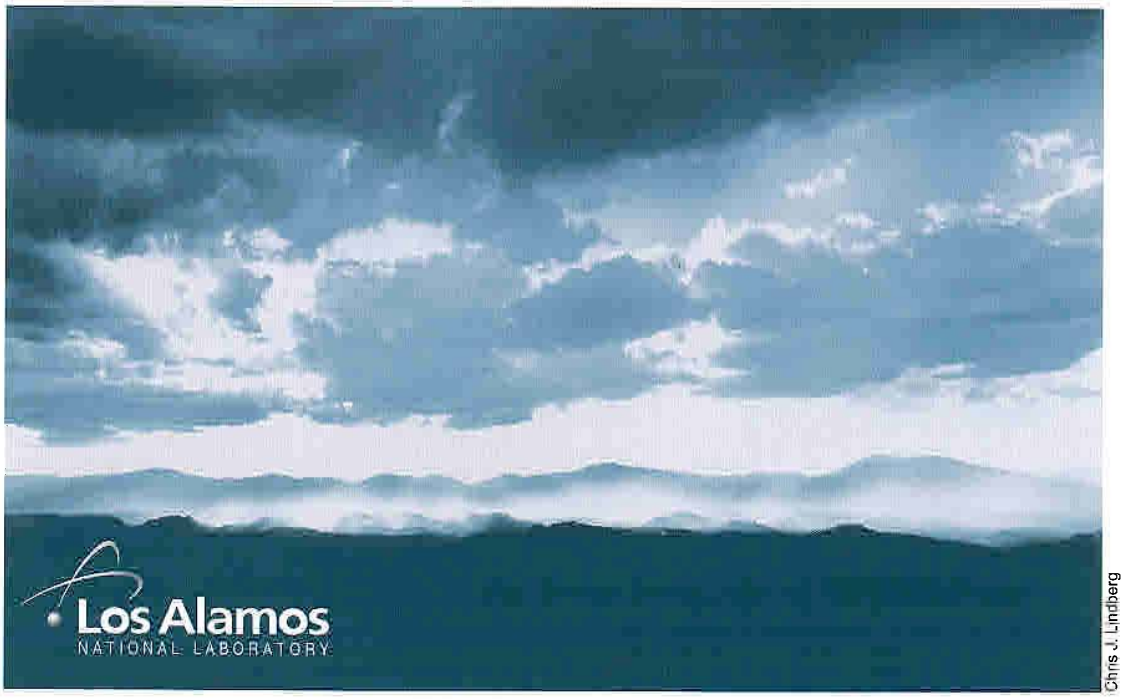

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ABSTRACT

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. We have studied and identified two different kinds of errors from FRAM analysis: random and systematic. The random errors come mainly from statistics and are easily determined. The systematic errors can come from a variety of sources and can be very difficult to determine.

We carefully examined the FRAM analytical results of the archival plutonium data and of the data specifically acquired for this isotopic uncertainty analysis project, and found the relationship between the systematic errors and other parameters. We determined that the FRAM's systematic errors could be expressed as functions of the peak resolution and shape, region of analysis, and burnup (for plutonium) or enrichment (for uranium). All other parameters such as weight, matrix material, shape, size, container, electronics, detector, input rate, etc., contribute little to the systematic error or they contribute to the peak resolution and shape and then their contributions can be determined from the peak resolution and shape.

A. INTRODUCTION

The Fixed-Energy Response-Function Analysis with Multiple Efficiency (FRAM) software has been developed and is continuing to be refined at the Los Alamos National Laboratory for the gamma-ray spectrometry measurement of the isotopic composition of plutonium, uranium, and other actinides [1–2].

FRAM can obtain a complete plutonium or uranium isotopic analysis using either a single planar or coaxial germanium detector or a CdTe detector. The actual detector type is not really important; what matters most are the resolution and the shapes of the peaks in the spectrum. The current version of FRAM (v4) can analyze a plutonium or uranium spectrum with a maximum peak resolution of 1.8 keV FWHM (full width at half maximum) at 122 keV for analysis of the 100-keV region, 2.2 keV at 122 keV for analysis of the low-energy region above 120 keV, and 3 keV at 1.3 MeV for analysis of the energy region above 500 keV. As for the peak shape, FRAM requires that the fraction of the peak area that is present in the low-energy tail of the peak must be less than 0.20 for all the peaks in the spectrum. Note that FRAM can analyze a peak with a large tail on the low-energy side of the peak. However, it cannot determine the tail on the high-energy side of a peak. Therefore, in spectra with peaks that have large high-energy tails, FRAM will not be able to accurately determine the isotopic compositions of the plutonium or uranium.

Although FRAM can analyze spectra with very broad peaks and with large-tail peaks, such as those near the limits just mentioned, common sense would tell us that the results from those

spectra may not be very good or they may have large errors. In our previous study, we found this is very true [3]. In general, a spectrum with good resolution and peak shape would give results with a smaller bias and better precision than a spectrum with bad resolution and peak shape.

For this study, we set up experiments to obtain the relationships of the biases with the internal parameters (peak resolution and shape, region of analysis, plutonium burnup or uranium enrichment) of the spectra. This paper shows only the results of the work done using the physical-efficiency curve. The systematic errors that occur with the empirical-efficiency model of FRAM will be presented in another paper.

B. PLUTONIUM

Figure 1 shows an example of a plutonium spectrum with four regions separated by three vertical dashed lines. Each of these four regions has one, and only one, measurable gamma ray from ^{240}Pu decay. The four overlapping analytical regions that FRAM normally uses for the analysis are shown as four thick, horizontal bars above the spectrum. The top bar corresponds to the analytical region for freshly separated plutonium. The other three are for aged plutonium.

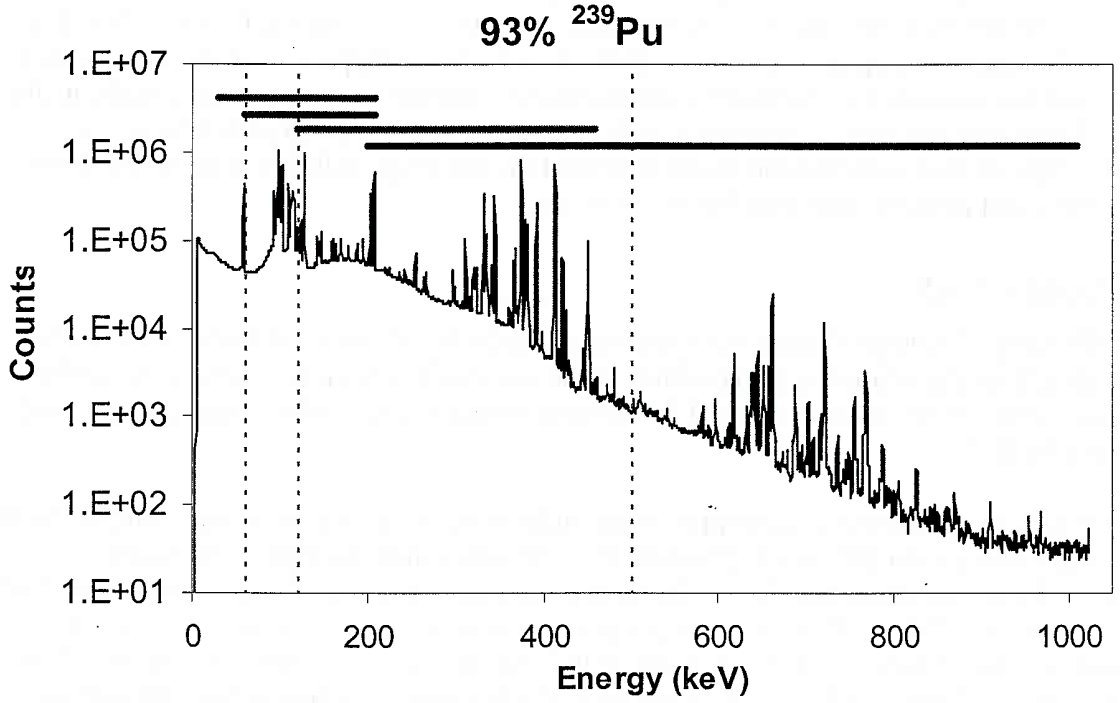

Figure 1. A low-burnup plutonium spectrum. The three vertical, dashed lines separate the four regions. The four overlapping analytical regions that FRAM normally uses for the analysis are shown as four thick horizontal bars above the spectrum.

1. Data acquisition

We set up the data acquisition system to obtain spectra with various resolutions and tails. We used two detector systems for the experiments, one planar germanium detector system and one

coaxial germanium detector system. The planar detector system consists 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 consists of a 58-mm-diameter × 53-mm-long coaxial detector (32% relative efficiency) and the DSPEC Plus MCA, both from Ortec.

The samples for these measurements included four of the seven samples from the PIDIE (plutonium isotopic detonation intercomparison exercise) set: PIDIE-1, PIDIE-3, PIDIE-5, and PIDIE-7. These samples are 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 8K channels at 0.075 keV/ch (keV per channel) up to more than 600 keV so that the data could be analyzed in two different energy ranges: 60 to 210 keV and 120 to 500 keV. In the coaxial detector, the spectra were acquired in 8K channels at 0.125 keV/ch, covering the entire 0- to 1,024-keV energy range. These spectra can be analyzed using two separate parameter sets employing the 120- to 500-keV and 200- to 1010-keV 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 of live time. The reason live time was chosen over true time was so the statistics would be the same for all the spectra and they could be directly compared. Sixteen spectra were obtained for each data set.

The variations of the FWHM for different samples with the same rise time are small, so they can be averaged together. Table 1 shows the average resolutions of the 129-keV peak of the spectra for each rise time setting for both detectors. In general, the resolution becomes better as the rise time increases from 0.2 μs to 8.0 μs.

Table 1. Average resolution of the 129-keV peak

Rise time (μs)	Planar FWHM (eV)	Coaxial FWHM (eV)
0.2	710	2151
0.4	656	1883
0.6	676	1720
1.0	607	1478
1.4	574	1343
2.0	555	1218
2.8	543	1125
4.0	533	1057
8.0	522	965

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 129-keV peak tail percentages varying from approximately zero to about 16%.

2. Analysis

We analyzed the data using the appropriate parameter sets for the data: parameter sets employing the 60- to 210-keV and 120- to 500-keV regions for the planar detector and the 120- to 500-keV and 200- to 1,010-keV regions for the coaxial detector. For the 60- to 210-keV region analysis, we analyzed the data using both the current FRAM v4 version and an upgraded FRAM v4 [4], which significantly improves the bias in the analysis of plutonium and uranium in the X-ray regions. Note that each parameter set uses only one gamma ray to determine ^{240}Pu: 104.2 keV for

the 60- to 210-keV region, 160.3 keV for the 120- to 500-keV region, and 642.5 keV for the 200- to 1,010-keV region. Both the planar and coaxial detectors use the same 120- to 500-kev region, and the results of the analyses of this region are combined from both detectors.

In typical plutonium, more than 90% of the plutonium comes from two isotopes: ^{239}Pu and ^{240}Pu. The activity of ^{239}Pu can be accurately determined due to the high intensities of the ^{239}Pu gamma rays. The activity of ^{240}Pu is much less accurately determined because the peaks of ^{240}Pu are much weaker. The normalization of the isotopic fraction process (i.e., the sum of all the plutonium isotopes must equal one) would pass on the large uncertainty of the ^{240}Pu activity to other isotopes, regardless of how accurately the activities of the other isotopes are determined. This would, in effect, make the weak peaks of ^{240}Pu the most important peaks in the plutonium spectrum. In other words, the accuracy of the ^{240}Pu activity determines the overall accuracy of all other isotopes.

The fact that the uncertainty of the ^{240}Pu isotope is passed on to the other isotopes significantly simplifies the determination of the systematic error in the FRAM analysis. That is, instead of finding the systematic error for every single isotope, we can just obtain the systematic error for ^{240}Pu, and the normalization process will transfer this systematic error to other isotopes.

At first, we tried to determine the correlation between the ^{240}Pu biases and the FWHMs and tails of the ^{240}Pu peaks. However, we found that it was difficult to find a good correlation connecting the ^{240}Pu biases to both the FWHMs and tails of the peaks. We then worked out two separate correlations: one for the bias and the FWHM of the ^{240}Pu peak and the other for the bias and the tail of the ^{240}Pu peak. They can be expressed as

$$^{240}\text{Pu bias} = 1 - \exp(a\boldsymbol{W}^2),$$

and

$$^{240}\text{Pu bias} = 1 - \exp(b\boldsymbol{T}),$$

where bias = |Measured/Accepted – 1|, a and b are some constants, $\boldsymbol{W}$ is the FWHM of the ^{240}Pu peak, $\boldsymbol{T}$ is the tail percent of the ^{240}Pu peak, and the ^{240}Pu peak is the 104.2-keV peak for the 60- to 210-keV region analysis, the 160.3-kev peak for the 120- to 500-keV region analysis, and the 642.5-keV peak for the 200- to 1,010-keV region analysis.

Figure 2 shows the ^{240}Pu biases as the functions of the FWHMs and of the tails of the ^{240}Pu peaks. The biases denoted as "100keVFixX" are for the analysis in the 60- to 210-keV region by the upgraded FRAM. The others are for the analysis using the current FRAM.

We see that the ^{240}Pu bias gets worse as the FWHM or the tail of the ^{240}Pu peak increases. Because the ^{240}Pu peak is different for different analyses, we cannot compare the analyses of different energy regions directly or look at the graphs in Figure 2 and readily say that one analysis using a certain energy region has better bias than the other. However, we can compare the 60- to 210-keV region analysis that was done with the current FRAM to the one done with the upgraded FRAM. In this comparison, we see that the upgraded FRAM gives somewhat better bias as a function of resolution and much smaller (about a factor of 4.5 smaller) bias as a function of the size of the tail of the 104.2 keV peak.

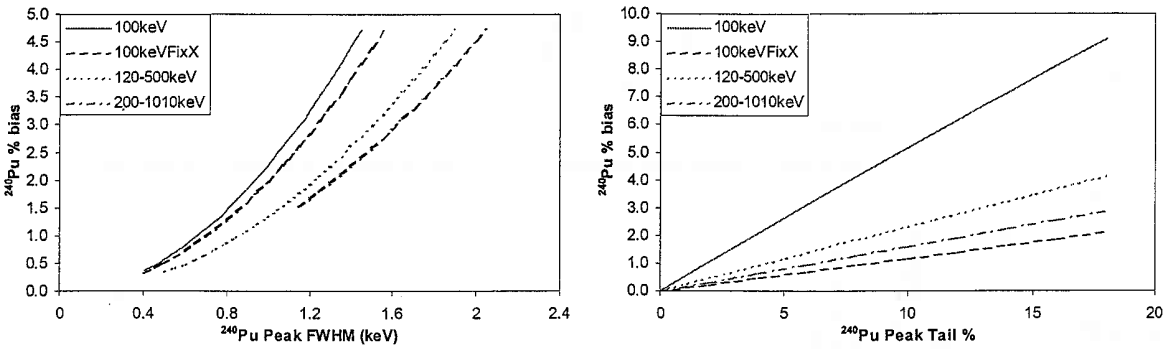

Figure 2. Pu-240 biases as the functions of the FWHMs and of the tails of the ^{240}Pu peaks. The ^{240}Pu peak is the 104.2-keV peak for the 100-keV region analysis, 160.3-kev peak for the 120- to 500-keV region analysis, and 642.5-keV peak for the 200- to 1,010-keV region analysis.

It is interesting to note that as long as the widths of the peaks are not zero, there will always be some bias associated with the finite resolutions. From the resolution graph in Figure 2, we see that the best germanium detector would have a ^{240}Pu bias of about 0.4% with a planar detector in both the 60- to 210-keV region (~0.45 keV FWHM at 104 keV) and 120- to 500-keV analysis region (~0.55 keV FWHM at 160 keV), about 0.9% for a coaxial detector with a resolution of about 0.8 keV at 122 keV in the 120- to 500-keV analysis region, and about 1.5% for a coaxial detector with a resolution of about 1.2 keV at 642 keV in the 200- to 1,010-keV analysis region.

C. URANIUM

FRAM can also obtain a complete isotopic analysis for uranium. If a planar detector is used, FRAM normally analyzes data from 80 to 210 keV. In low-enriched uranium (LEU), where the 63.3-keV peak from the ^{238}U decay-chain may be visible, FRAM may also use that peak. If a coaxial detector is used, the analytical energy range is normally from 120 to 1,010 keV.

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 that FRAM normally uses for the analysis.

1. Data acquisition

The data acquisition system is set up the same way it is for plutonium bias determination (Section B). For these measurements, five LEU samples of the NBS-SRM* 969 set and three high-enriched uranium (HEU) samples of the NBL-CRM† 146 set, ranging from 0.3 to 93.2% ^{235}U enrichment, were used. These samples weight 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.

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

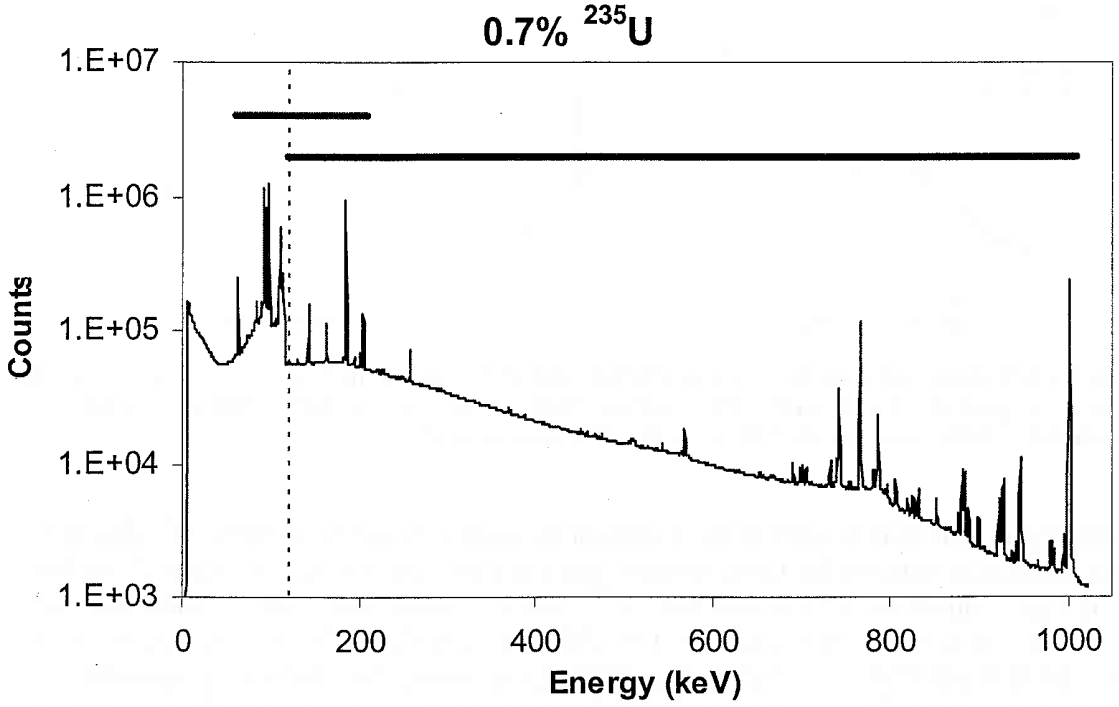

Figure 3. A natural-uranium spectrum. The vertical dash line separates the two regions: one below the K-edge and one above the K-edge. The two overlapping analytical regions that FRAM normally uses for the analysis are shown as two thick horizontal bars above the spectrum.

The data for the planar detector were acquired in 4K channels at 0.075 keV/ch and analyzed using the peaks in the 60- to 210-keV energy range. For the coaxial detector, the spectra were acquired in 8K channels at 0.125 keV/ch and analyzed using the parameter set employing the 120- 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, 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. Table 2 shows the average resolutions of the 186-keV peak ^{235}U decay of the spectra for each rise-time setting for both detectors.

In order 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 minute true time) for each set, were obtained, with the 186-keV peak-tail percents varying from approximately zero to about 16%.

Table 2. Average resolution of the 185.7-keV peak

Rise time (μs)	Planar FWHM (eV)	Coaxial FWHM (eV)
0.2	766	2325
0.4	731	1976
0.6	707	1783
1.0	670	1538
1.4	652	1394
2.0	637	1271
2.8	629	1188
4.0	618	1098
8.0	610	999

2. Analysis

We analyzed the data using the appropriate parameter sets for the data: parameter sets using the 60- to 210-keV region for the planar detector and the 120- to 1010-keV regions for the coaxial detector. In the 60- to 210-keV region analysis, we analyzed the data using the current FRAM v4 version and the upgraded FRAM v4, like we did with the plutonium analysis in Section B. For the coaxial analysis, we used two different analyses: one that used the gamma rays of ^{235}U, ^{238}U, and ^{228}Th for the relative efficiency curve, and the other used only the peaks of ^{235}U and ^{238}U decay for the relative efficiency curve. There is normally some ^{228}Th in uranium if the uranium is enriched from recycled uranium. Therefore, we may be able to use the gamma rays from ^{228}Th decay to determine the relative efficiency curve. However, in LEU or HEU enriched from natural uranium where the gamma rays from ^{228}Th decay chain may be weak or absent, only the peaks of ^{235}U and ^{238}U decay can be used to determine the relative efficiency curve.

Figure 4 shows the ^{235}U biases of the 60- to 210-keV analysis of the planar data as functions of the FWHM and of the tail of the 92.7-keV peak. The biases denoted as "100keVFixX" are for the analysis in the 60- to 210-keV region by the upgraded FRAM, and the others are for the analysis using the current FRAM.

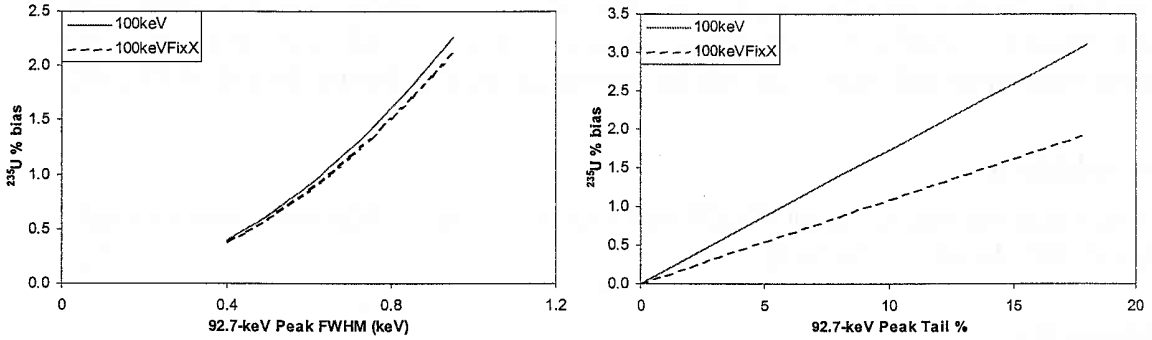

Figure 4. The biases of the ^{235}U enrichment as the functions of the FWHMs and of the tails of the 92.7-keV peak.

Figure 5 shows the ^{235}U biases of the 120- to 1,010-keV analysis of the coaxial data as a function of the FWHM and of the tail of the 185.7-keV peak.

We see that the ^{235}U bias of the analysis using ^{228}Th is better than that of the analysis without using the ^{228}Th. This is expected since extra peaks of ^{228}Th would improve the accuracy of the relative efficiency curve and thus improve the biases.

D. CONCLUSION

We have studied the systematic errors of FRAM analysis by employing various parameter sets using gamma and X-rays in various energy regions of data taken with the planar and coaxial detectors. We determined the biases as functions of the resolutions and the tails of the peaks. The modified FRAM code includes a new command, **system_error_factor,** in the **Application Constants** section of the FRAM parameter set. The default value for the command is **0.0**, which will give no systematic error and will automatically turn off the calculations of systematic error.

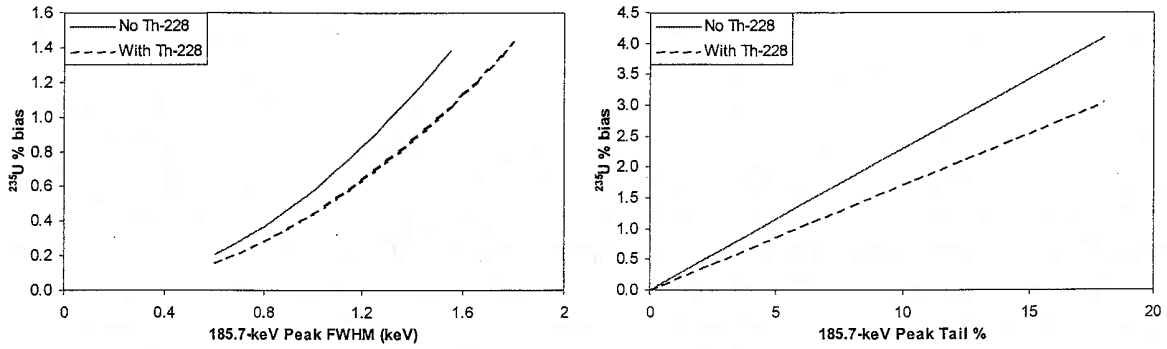

Figure 5. The biases of the ^{235}U enrichment as the functions of the FWHMs and of the tails of the 185.7-keV peak.

When it is set at value **1.0,** the systematic error is set to be equal to the biases shown in Figures 2, 4, and 5. The FRAM user may change it to a different value if the user finds the new value better corresponds to the performance of a particular system. The determined systematic errors are added to the statistical errors and stored in the result file of FRAM. The total errors are currently embedded into the secondary layer of the FRAM result file and can be viewed or printed out using the "Medium result" of FRAM output. The upgraded FRAM v4 code that includes this systematic uncertainty and other fixes will be released, hopefully, before the end of FY 2005.

Acknowledgement

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