ORNL/LTR-2015/419

Making Quality HPGe Gamma Ray Spectrum Measurements for Uranium: The Role of FRAM for Analysis, Quality Control and Enrichment Measurements, and Opportunities for Improved Quantification Documentation

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October 2015

OAK RIDGE NATIONAL LABORATORY
MANAGED BY UT-BATTELLE FOR THE US DEPARTMENT OF ENERGY

ORNL/LTR-2015/419

Nuclear Security & Isotope Technology

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Date Published: May 2015

Revision 4, 21-October-2015

Prepared by
OAK RIDGE NATIONAL LABORATORY
Oak Ridge, TN 37831-6283
managed by
UT-BATTELLE, LLC
for the
US DEPARTMENT OF ENERGY
under contract DE-AC05-00OR22725

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ABSTRACT

Enrichment measurements are used in uranium safeguards verification and accountancy as both an attribute signature and in the interpretation of other nondestructive assay information, such as active interrogation for mass. Accurate and reliable assessments require quality information. The practices used to collect the raw data are therefore the crucial first step in managing the overall quality and uncertainty of the enrichment determination.

This report discusses several topics associated with gamma-ray spectrum measurements of uranium using HPGe detectors. First a discussion of good data acquisition practices including physical setup, detector types and configurations, detector shielding and filtering, spectrum stabilization, and data acquisition protocols.

Next is a discussion of analysis options to obtain isotopic composition and/or enrichment centered on the FRAM gamma ray isotopic analysis software. The report discusses the results of the development of a FRAM parameter file to perform classical Enrichment Meter Principle (EMP) analysis. Possible analysis and data acquisition issues (random summing, x-ray fluorescence peaks, peak interferences, shielding) that can affect FRAM analyses are also discussed. There is also a brief discussion of FRAM measurements in the field.

Under the topic of uncertainty propagation and quantification in FRAM, uncertainty propagation verification methods and gaps in available uncertainty quantification data for uranium is covered. Suggestions are made for areas where ORNL can contribute to closing the identified uncertainty quantification gaps, and also to contributing nuclear data quality spectra to the research community.

1. DATA ACQUISITION PRACTICE

This section discusses topics that must be mastered in order to acquire quality data that can be readily analyzed by FRAM and other gamma spectrum analysis software without the need for making ad hoc pre-analysis corrections to the data. The overriding theme is that one must have high quality data in order to have any hope of obtaining a quality analysis. Many analyses are ruined from the on-set by poor data collection practices. The review by Gehrke and Davidson is an excellent report on good data acquisition practices (Gehrke 05).

1.1 DETECTOR CHOICE

The detector selected for a particular set of measurements should be chosen and specified with the end goal of the measurements and also the capacity and specifications of the data acquisition electronics in mind. The energy range of the analysis must be compatible with the resolution and efficiency of the detector in the chosen energy range. Detailed descriptions of the available detector types from the major manufacturers and the manufacturers' names for them may be found at http://www.ortec-online.com/ (ORTEC) and http://www.canberra.com/ (Canberra). Several types of High Purity Germanium (HPGe) have commonly been used for various types of uranium isotopic abundance or enrichment measurements.

Planar Detectors: These detectors, often called Low Energy Photon Spectrometers (LEPS), generally have the best resolution of the commonly used detector types. Detectors with a diameter of ~25 mm and

thickness of ~15 mm are typically used for gamma ray isotopic measurements in the low energy range—100 keV region (both U and Pu) and up to ~500 keV (mid-range measurements of Pu). While this type of detector has historically been thought to be limited to measurements < 500 keV, they have been shown (Sampson 01) to be useful for full range uranium isotopic measurements, up to 1010 keV, through as much as 16 mm of steel, which covers UF_6 cylinder measurements. This type of detector could be used for Enrichment Meter Principle (EMP) measurements but somewhat larger diameter detectors are usually preferred for this technique.

Coaxial Detectors: This type of detector is used for uranium isotopic measurements in the 120-1010 keV range. While the first measurements of this type used relatively small coaxial detectors of about 25% relative efficiency (in the usual definition, relative to the 1333 keV full energy peak efficiency of a 3"x3" NaI(Tl) scintillator at 25 cm) all sizes of coaxial detectors have been used successfully for uranium isotopic measurements in this energy range including detectors of 110% relative efficiency (Harker 11). Coaxial detectors may be used for EMP measurements or full range (120-1010 keV) full uranium isotopic analysis measurements but generally do not have good enough resolution to measure in the 100 keV region which involves the deconvolution of numerous overlapping features.

Semi Planar Detectors: These detectors maintain their efficiency to low energies while maintaining good low energy resolution and also have reasonable high energy efficiency. They present a reasonable compromise single detector allowing use for 100 keV x-ray region measurements as well as full energy range (120-1010 keV) uranium isotopic measurements - although they are not optimum for either domain. They may be used for EMP measurements.

One of the most important specifications when choosing a detector is the energy resolution and peak shape. Consult the detector manufacturer's literature for extensive discussion of this topic. For best application and best measurement practice, always choose a detector with the best resolution that the budget will allow, even at the expense of efficiency because "An ounce of resolution is worth a pound of computer code."

1.2 DETECTOR/CRYOSTAT/DEWAR CONFIGURATION

The basic choice here is usually between a side-looking detector and an up-looking detector. Other choices or variants include portable, small dewar configurations that can be used in all attitudes. Many configurations of the detector end-cap are available. The user should purchase a configuration that allows the use of a backshield behind the end-cap improve the detector shielding to ambient sources of radiation.

For the highest quality EMP measurements use an up-looking detector. This allows the user to more easily establish geometric reproducibility. Several diagrams of successfully implemented EMP geometries are given in Fig 1 below. The low-enriched uranium isotopic standard set (NBL CRM 969) and the high-enriched uranium isotopic standard set (NBL CRM 146) are ideally suited for measurements in an up-looking geometry because the aluminum bottom window of the containers is very accurately specified and certified. These isotopic standards can conveniently be used in a stable up-looking geometry when performing measurements designed to test and/or characterize an enrichment measurement or analysis method or technique.

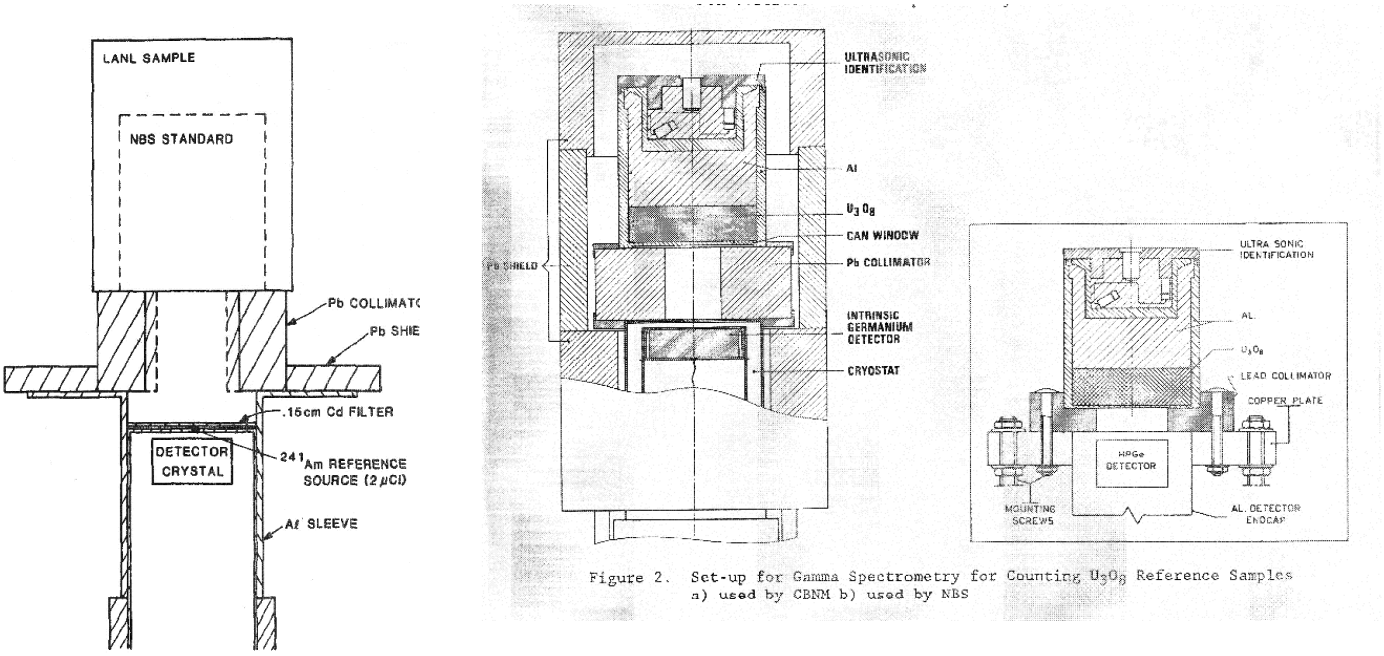

Fig. 1. Examples of EMP measurement geometries featuring up-looking HPGe detectors and reproducible geometry. Left: (Parker 88), Right (Carpenter 86).

1.3 DATA ACQUISITION ELECTRONICS

Modern digital spectroscopy systems incorporating pileup rejection, digital gain and zero stabilization, and 16,384 channel capacity (16K) ADC are recommended for isotopic analysis or EMP data collection. While 16K channel capacity is now widely available and version 5 of FRAM can handle up to 32K channels of data, most analyses seldom need to use more than 8K channels. The older version 4 of FRAM has a maximum channel capacity of 8K channels.

1.3.1 Data Acquisition Electronics: Gain Settings

Most gamma ray spectrum acquisitions from uranium fall into one of three energy ranges.

"100 keV Region," 60-300 keV: This region of the uranium spectrum is analyzed for isotopic information and uses x-ray and gamma-ray peak information in the 100 keV-region to supplement the ^{235}U information typically obtained at 185.7 keV. Analysis of the x-ray region puts a premium on good detector resolution essentially requiring the use of planar HPGe detectors or very good resolution semi-planar detectors. A gain setting of 0.075 keV/ch is very commonly used with 4K channels of data. Extending the data acquisition to higher energies by using 8K (600 keV) or 16K (1200 keV) channels usually does not produce additional quality data because the high resolution detectors required for 100 keV region analysis usually lack sufficient efficiency at the higher energies for good statistical quality. In any case, using 8K channels at 0.075 keV/ch (range up to 600 keV) does not provide any additional useful isotopic data over the 4K data acquisition. An exception to this arises for waste assay measurements where quantitative uranium and, in addition, other isotopic masses (activation and fission products) are determined.

120–1010 keV Region: This region of the spectrum is commonly used to obtain a complete uranium isotopic analysis. More efficient coaxial detectors are best for this region and a gain setting of 0.125 keV/ch in 8K channels is a standard measurement condition for FRAM. Conversion gains as high as 0.2 to 0.25 keV/ch have been used in this range although when the gain is as high as 0.25 keV/ch the peaks

below 200 keV become more difficult to analyze with FRAM because they are too narrow (contain too few channels of data) to get a good internal peak shape calibration. FRAM has been shown to work for uranium and plutonium isotopic analysis (using the original ORTEC Detective) with a fixed gain of 0.366 keV/ch (Sampson 06) although working at this gain is definitely not recommended.

"Enrichment Meter" Region: Because this region, centered around 185.7 keV is fairly small, a range of gain settings from 0.075 to 0.125 keV/ch may be successful with a planar, semi-planar, or small coaxial detector.

1.3.2 Data Acquisition Electronics: Digital Stabilizer Settings

Using Gain and Zero stabilization will preserve spectral quality against spectral shifts caused by ambient temperature fluctuations and will accurately preserve the spectrum energy calibration making it easier to find and observe known peaks or accurately identify the energy of unknown or unexpected peaks.

For spectra concentrating on the 100 keV-region for isotopic analysis or for spectra collected for EMP measurements it is usually sufficient to set the Gain stabilization on the 185.7 keV ^{235}U peak. The Zero stabilizer does not need to be used assuming the 185.7 keV peak is in the top half of the spectrum. However, if a ^{241}Am rate-loss-correction source is used with EMP measurements one should use its 59.54 keV peak for Zero stabilization.

For broader range data collections that extend to or above 1 MeV, set the Gain stabilization on the 1001.03 keV ^{234m}Pa (^{238}U daughter) and set the Zero stabilization on the 185.7 keV ^{235}U peak.

1.3.3 Data Acquisition Electronics: Amplifier Time Constant Settings

Older amplifiers with analog (Gaussian or Triangular shaping) were specified differently than the current generation of digital spectrometers (e.g. rise time or rise time and flat-top in case of trapezoidal filtering). The value of the rise time on a digital spectrometer is approximately 2x the equivalent Gaussian shaping time on an analog amplifier. The setting of an amplifier time constant is an important trade-off between resolution, throughput, and spectrum distortion from pulse pileup. The user is advised to consult nuclear instrumentation vendor's web sites for a detailed discussion of the trade-offs. Another recommended resource is (Parker 91).

Every regularly-used detector should have the resolution vs. count rate|time constant thoroughly characterized as well as the throughput vs. count rate|time constant. The visualization of the best operating point becomes easier after viewing these curves.

A suggested amplifier time constant that will handle nearly all measurements is a rise time (digital spectrometer) setting of 4 μs. Going to a higher setting may gain a little better energy resolution but at the loss of throughput and, more importantly, an increase in random summing pileup. This setting may be increased for very low count rates (< 1000 cps). The highest resolution, even in the 100 keV region, is not as critical for uranium measurements as it is for some other measurements with more overlapping peaks. For the most part, uranium produces a comparatively simple spectrum.

1.4 SHIELDING

The amount of shielding needed depends upon the purpose of the measurement and is usually governed by the energy range of the data to be analyzed.

1.4.1 Lead Shielding

For lead, the most commonly used shielding material; Table 1 shows the transmission for various energies as a function of the lead thickness.

Table 1. Transmission of Lead as a Function of Thickness at Indicated Energies (keV).

Lead Thickness		Transmission at Indicated Energy (keV), ( ) = Exponent						
Inches	**mm**	**75**	**100**	**185.7**	**300**	**766.4**	**1001**	**2614**
0.001	0.0254	0.921	0.852	0.966	0.988	0.997	0.998	0.999
0.005	0.127	0.664	0.449	0.842	0.943	0.987	0.990	0.994
0.0197	0.50	0.199	0.0429	0.508	0.0.796	0.949	0.960	0.976
0.03125	0.794	0.0772	0.0067	0.341	0.695	0.920	0.938	0.962
0.03937	1.00	0.0397	0.00184	0.258	0.633	0.900	0.923	0.952
0.0625	1.587	0.00596	4.55(-5)	0.117	0.484	0.846	0.880	0.925
0.0787	2.0	0.00157	3.38(-6)	0.0667	0.400	0.810	0.851	0.907
0.125	3.175	3.55(-5)	2.07(-9)	0.0136	0.234	0.715	0.774	0.856
0.25	6.35	1.26(-9)	4.28(-18)	1.84(-4)	0.0547	0.512	0.600	0.733
0.5	12.7			3.4(-8)	0.0030	0.262	0.360	0.537
0.75	19.0				1.64(-4)	0.134	0.216	0.393
1.0	25.4					0.0686	0.129	0.288
1.5	38.1					0.0180	0.0465	0.155
2.0	50.8					0.00471	0.0167	0.0829
4.0	101.6					2.22(-5)	2.8(-4)	6.88(-3)

Measurements in the 100 keV energy range, extending up to approximately 300 keV should have at least 0.5 inches (12.7 mm) of lead shielding to assure that no full energy gamma rays with energy less than 300 keV can penetrate the shield. This advice also applies to EMP measurements which concentrate on the 185.7-keV ^{235}U gamma ray.

The shielding requirements for uranium isotopic measurements in the 120–1010 keV region are more formidable. Over 1.5% of 1001 keV gamma rays will be transmitted through 2 inches of lead. This means that any measurement in this energy range must have at least 2 inches of lead shielding unless it can be assured that that there are no ^{238}U materials in the vicinity of the measurement.

1.4.2 Tungsten Shielding

The use of a machinable tungsten alloy for shielding can reduce the shielding mass by up to 1/3 compared to lead for the same absorption properties. Unlike lead tungsten alloy not considered a hazardous (toxic) material. Facility safety requirements may require lead shields to be encapsulated to prevent contact with the skin. Machinable tungsten alloy shields are more compact, easier to handle, and less hazardous than lead shields.

1.4.3 Shielding for Dual Range Measurements

The definition of a "dual range" measurement is a measurement made over a wide energy range that has sufficient energy resolution and efficiency such that it could be analyzed in two energy ranges. One example is the use of a "100 keV region" detectors (Planar or Semi-Planar) set up to take 16,384 ch of data at 0.075 keV/ch (0 - 1228 keV) with the idea of analyzing and comparing the analyses in both the energy region 60–250 keV and 120–1010 keV (FRAM standard parameter file analysis ranges).

For this type of measurement the shielding must be adequate for the highest energy range to be analyzed. Unfortunately one often sees this type of measurement made with a "100 keV region" detector that is only shielded for "100 keV region" measurements. Because the data spans the 120–1010 keV uranium isotopic analysis region it is assumed (mistakenly) that the isotopic analysis can be done in this region. It cannot if there is any uranium background that will not be properly shielded with only "100 keV region" shielding. Such a dual range measurement must have shielding adequate for 1001 keV.

An example of this can arise when EMP measurements are made through a collimator that is not thick enough to completely attenuate all the gamma rays of interest and these measurements are then analyzed with a FRAM analysis in the 120–1010 keV range. For example suppose the EMP measurements were made through a 15-mm diameter hole in a 20-mm thick lead collimator. All gamma-ray energies could pass unattenuated through the 15 mm diameter hole in the 20 mm thick collimator. However, at 1 MeV, the most intense ^{238}U line, the transmission of 20 mm of Pb is 0.2. So part of the 1 MeV gamma rays reaching the detector are unattenuated while others reach the detector having passed through 20 mm of Pb. The fraction passing through the Pb varies with energy being about 11% at 742 keV and essentially zero at 258 keV and all lower energies. The attenuation model in FRAM cannot handle this type of varying attenuation. This situation would not affect an EMP analysis at 185.7 keV but has a pronounced effect for a FRAM analysis in the 120–1010 keV range.

1.4.4 Graded-Z Shielding

A graded-Z shield is a shield that has layers of Cd (or Sn) on the inside next to the detector. The Cd is usually covered with another layer of even lower atomic number (typically Cu). The purpose of these additional materials is to absorb the characteristic x-rays produced in the bulk shielding material. The Cd (Sn) thickness is chosen to completely absorb the 72–85 keV K-shell fluorescent x-rays produced in the lead shield. The Cu absorber, in turn, is scaled to absorb the fluorescent K x-rays produced in the Cd (Sn) in the 23–28 keV range.

Table 2 shows the transmission properties of Cd and Cu at energies encountered in graded shielding applications.

A graded-Z shield of Cd of thickness 3/32 inch (2.38 mm) is sufficient to absorb essentially all fluorescent K x-rays from a lead shield. Adding an additional Cu layer 0.020 inch (0.5 mm) will absorb any fluoresced Cd x-rays. Removing the K x-rays removes spurious features from the spectrum which can cause interference. This is discussed next.

Table 2. Transmission Properties of Cadmium (Cd) and Copper (Cu).

Cd, Cu Thickness		Transmission at Indicated Energy (keV), ( ) = Exponent						
		Cd			Cu			
Inches	mm	75	100	185.7	25	75	100	185
0.001	0.0254	0.931	0.967	0.992	0.661	0.980	0.990	0.996
0.005	0.127	0.698	0.846	0.962	0.126	0.903	0.949	0.981
0.0197	0.50	0.243	0.517	0.859	2.88(-4)	0.670	0.814	0.927
0.03125	0.794	0.106	0.351	0.785	2.39(-6)	0.529	0.722	0.887
0.03937	1.00	0.0590	0.268	0.738				
0.0625	1.587	0.0112	0.123	0.617				
0.0787	2.0	0.00348	0.0716	0.544				
0.09375	2.381	0.00118	0.0433	0.484				
0.125	3.175	1.25(-4)	0.0152	0.380				

1.4.5 Graded-Z Shielding for Uranium Isotopic and EMP Measurements

While a graded shield should be used in all gamma ray measurements, the effects of having an inadequate graded shield are different in the different analysis energy regions.

FRAM Uranium Isotopic Measurements in 120–1010 keV region: The lack of a graded-Z shield can seriously affect FRAM measurements in this energy region. Figures 2 and 3 below shows the effect of the random summing of fluoresced Pb K x-rays from a bare Pb shield without any graded Z additions. The Pb K x-rays sum with the strong 185.72 keV ^{235}U gamma ray to produce peaks in the region around 258 keV, a region important for defining the relative efficiency curve for 120–1010 keV FRAM uranium analysis.

$$\text{PbK}\alpha_1 \;\; 74.97 \text{ keV} \;+\; 185.72 \text{ keV } ^{235}\text{U} \;=\; 260.69 \text{ keV}$$

$$\text{PbK}\alpha_2 \;\; 72.81 \text{ keV} \;+\; 185.72 \text{ keV } ^{235}\text{U} \;=\; 258.53 \text{ keV}$$

^{234m}Pa at 258.26 keV is used for uranium relative efficiency and is important because it "connects" the high-energy and low-energy portions of the relative efficiency curve. See Section 2.1.1.

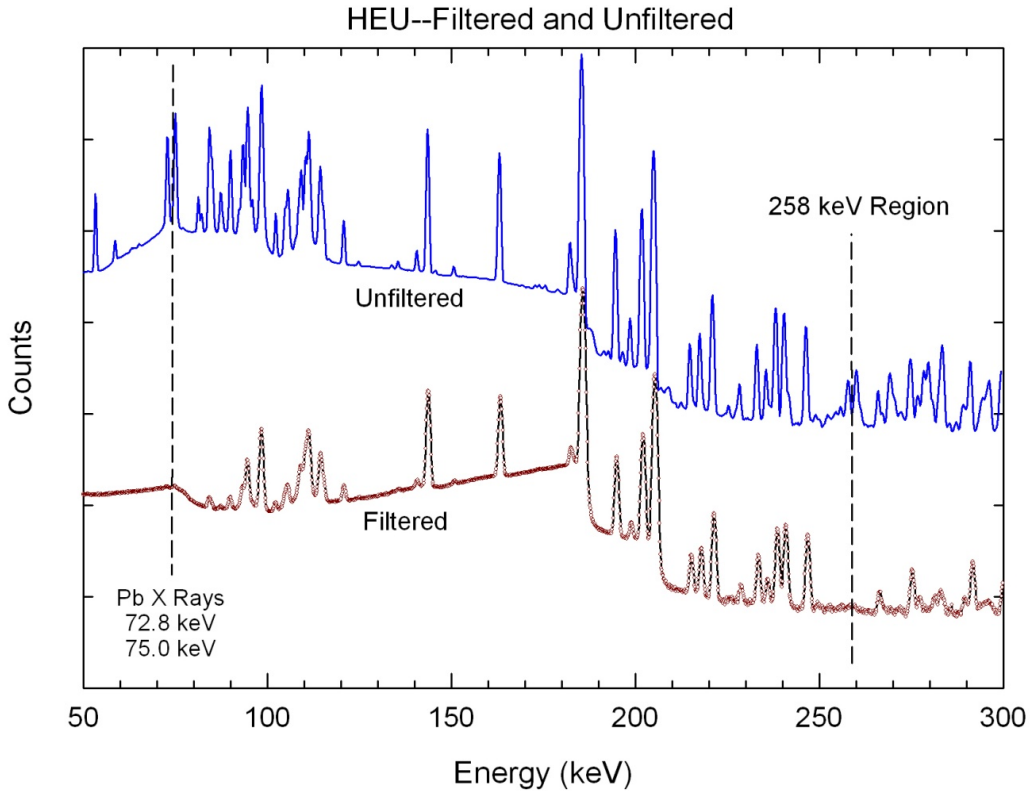

Fig. 2. Spectra from two HEU samples showing how the presence of Pb x-rays from an ungraded Pb shield (top spectrum labeled Unfiltered) can sum with the 185.7 keV ^{235}U peak to produce an interference in the 258-keV region.

Another pair of peaks, although not Pb x-ray related, that can sum to produce interference in the 258–260 keV region are:

$$UK\alpha_1 \ 98.43 \text{ keV} + 163.36 \text{ keV } ^{235}U = 261.79 \text{ keV}$$

$$UK\alpha_2 \ 94.65 \text{ keV} + 163.36 \text{ keV } ^{235}U = 258.01 \text{ keV}$$

FRAM corrects for this interference in the 258 keV region but the correction will be imperfect because sum peaks are usually misshapen (do not conform to the full energy peak shape model) and cannot be fitted well.

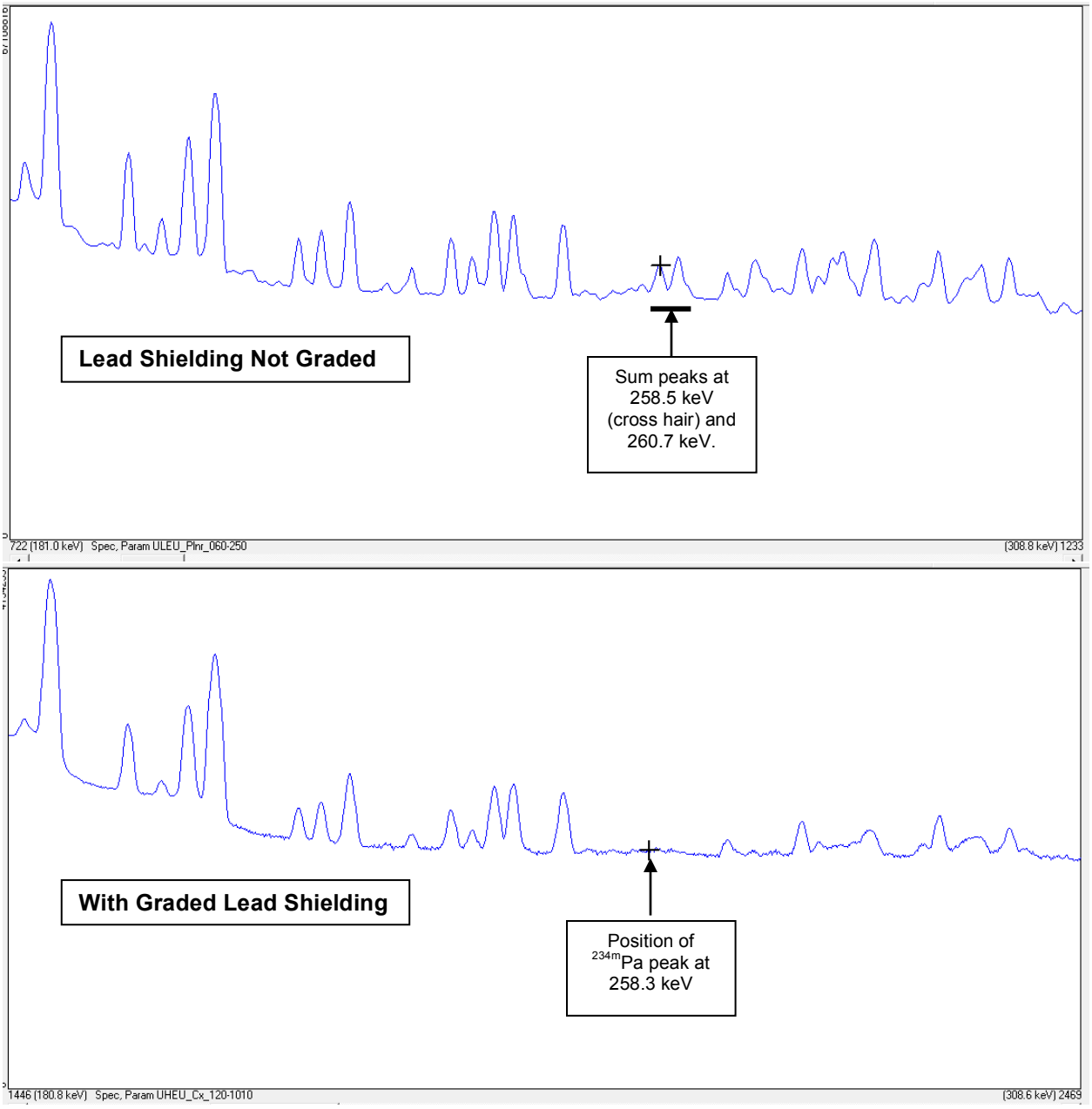

Fig. 3. Expanded view of 258 keV region with bare lead shielding (top) and graded-Z lead shielding (bottom).

FRAM Uranium Isotopic Measurements in 60–250 keV region: The presence of lead x-rays affects this energy region differently than the higher energy region just discussed. In this analysis FRAM uses the 84.22 keV ^{231}Th (^{235}U equilibrium daughter) to anchor the low end of the relative efficiency curve. A problem arises if significant lead x-rays are present in the spectrum. Lead Kβ x-rays appear in two main groups centered around ~ 84.8- and ~87.5 keV. The complex at ~84.8 keV interferes directly with 84.22 keV ^{231}Th peak. Figure 4 shows three different cases for the energy region around 84 keV. The Pb x-rays in Fig 4a are too strong for FRAM to make an accurate correction.

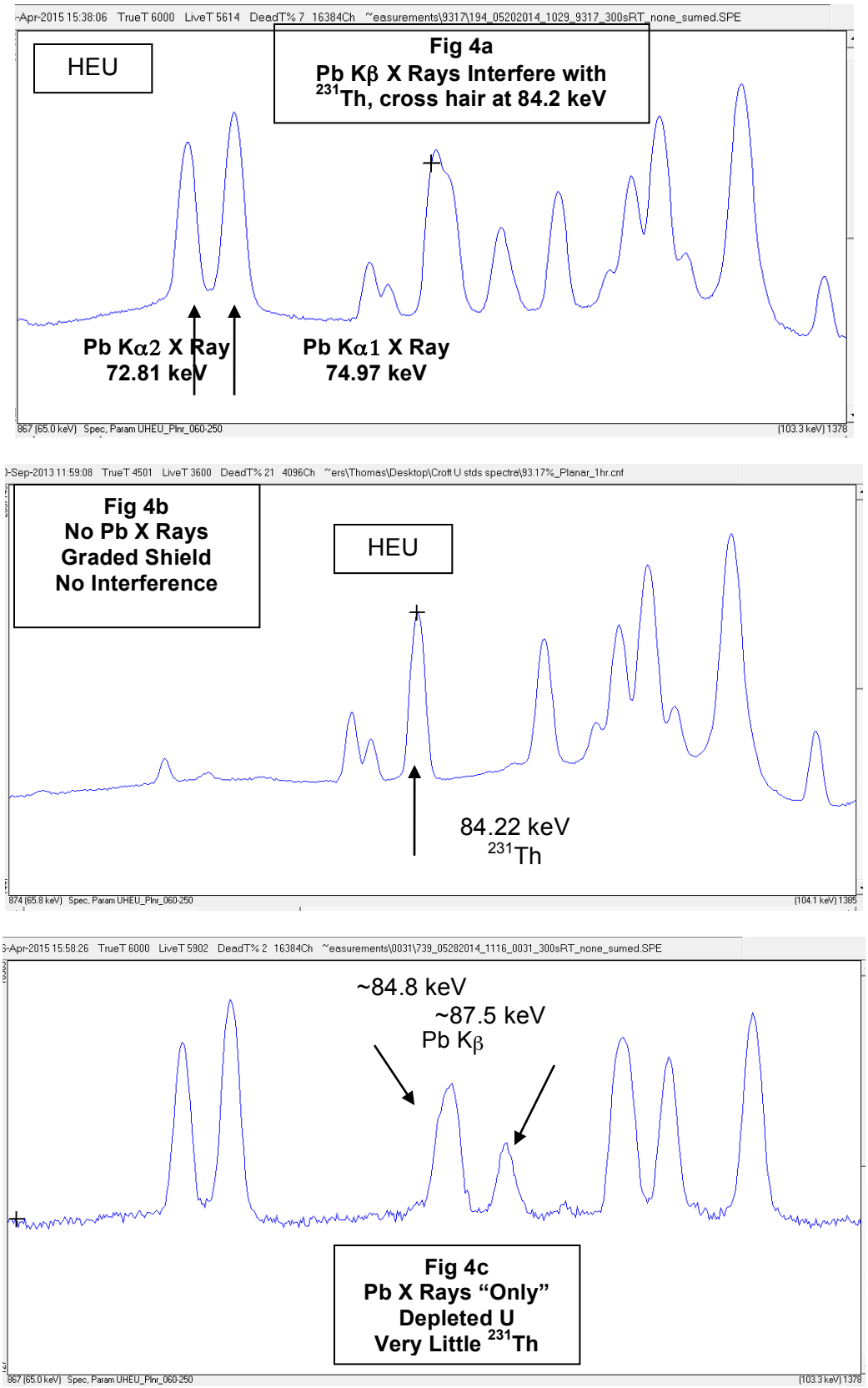

Fig. 4. Effect of lead x-rays.

1.4.6 Filtering

In addition to graded-Z shielding, a quality data acquisition setup must always consider the need for filters over the entrance face of the detector. Filters are designed to remove lower energy gamma rays, not needed in the analysis, from the spectrum. They can help manage the overall counting rate and improve useful throughput. Proper filtering can often prevent unwanted low energy gamma rays from producing sum peaks that could compromise the analysis in higher energy regions of the spectrum.

While this report concentrates on uranium measurement issues, the classic example for filtering arises in the measurement of plutonium. Essentially all plutonium spectra have a strong (often overwhelmingly strong) 59.54 keV gamma ray from ^{241}Am produced in the decay of ^{241}Pu. This Am gamma ray is often $> 10^3$ times as strong as the next strongest peak in the spectrum. The 59.54 keV gamma ray sums with numerous peaks in the 100 keV region to produce interferences in the 160-keV region, an important analysis region for medium-energy (120–450 keV) FRAM analyses. Such an overpowering peak increases the deadtime and makes it impossible to acquire sufficient statistics in the desirable portions of the spectrum.

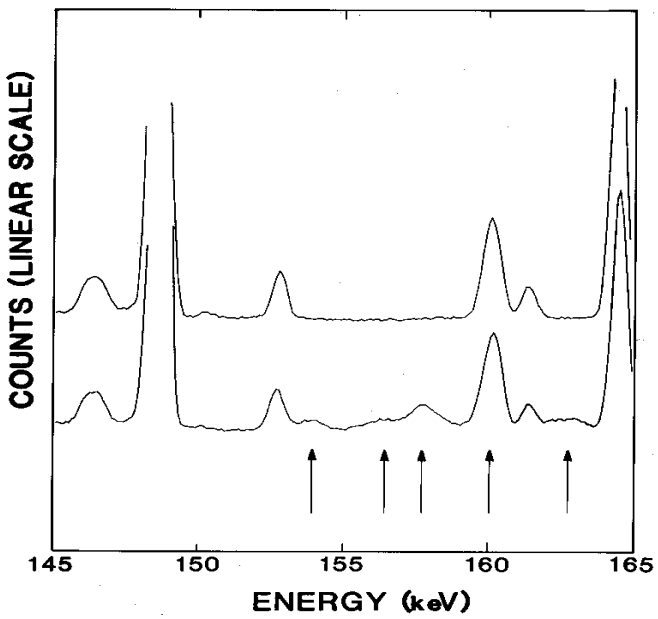

Fig. 5. Pileup and summing in the 160-keV region of a plutonium spectrum caused by a strong 59.54 keV ^{241}Am peak that has not been filtered out.

For Pu a filter of 2 mm of Cd (or Sn, which is non-toxic) is usually sufficient for most any ^{241}Am concentrations. For Pu data acquisitions that require analysis in the 100 keV region and may also require the presence of the 59.54 keV peak, the Cd filter thickness should be adjusted so that the 59.54 keV peak is approximately the same height as the peaks in the 100 keV region.

Just like graded-Z shields, filters (Cd or Sn) should also be backed on the detector side by a Cu filter of 0.5 mm thickness (see Table 2). Figure 6 shows the low energy portion of a spectrum of essentially pure ^{241}Am. This spectrum is filtered with Sn that almost completely suppresses the 59.54 keV ^{241}Am gamma ray. However, the filter does not have a low-Z component because the fluoresced Sn K x-rays are strong in the 25 keV region. There is no need for the Sn K x-rays to be in the spectrum. They can only cause trouble by summing with other peaks to produce unwanted interferences. Get rid of them by using a graded-Z filter.

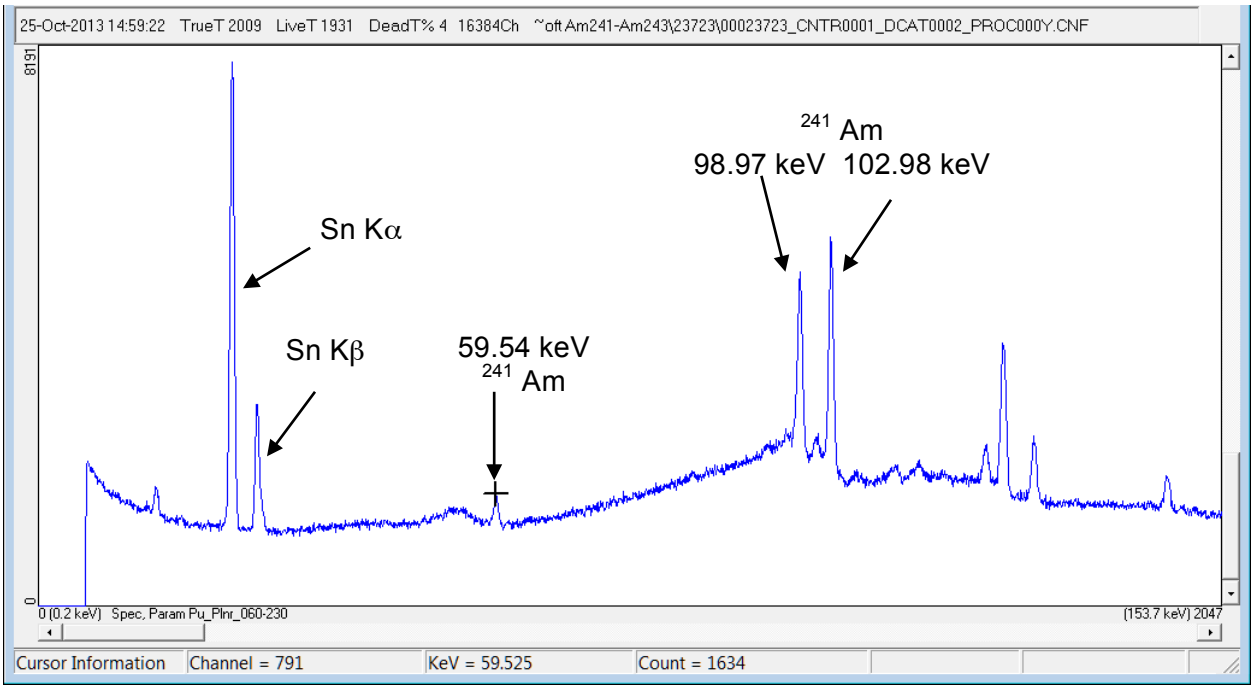

Fig. 6. The low energy portion of a spectrum from pure ^{241}Am. The spectrum is filtered with Sn effectively suppressing the 59.54 keV ^{241}Am gamma ray. The filter does not have a Cu graded-Z component hence we see fluorescent Sn x-rays.

Filtering for uranium spectra usually has fewer requirements than for plutonium. In general to analyze the 100 keV region, one should minimize the filter thickness. A semi-planar or planar detector will require a graded-Z shield as these detectors are very sensitive to low energy gamma rays. With a p-type coaxial detector one might 'get away' without the Cu layer because the thick Ge dead layer on the crystal will serve the same purpose.

1.4.7 Implementing Shielding and Filtering

The easiest way to implement graded-Z shielding is to start by making a graded-Z wrap to completely cover the cylindrical surface of the detector end cap. A successful wrap can consist of 3 layers of 1/32 in. thick (total thickness = 0.094 in. (2.4 mm)) Cd wrapped tightly around the detector end cap. Use a 0.020 in. (0.5 mm) thick Cu wrap next to the detector end-cap inside the Cd. This will be suitable for any type of detector. Fabricate a custom configuration to match the end-cap length and diameter for each available detector. Figure 7 shows such a Cd wrap implemented on a right angle dipstick detector in a 30 liter Dewar.

Assorted thicknesses of circular Cd end-cap filters should be machined to match the outer diameter of the filter wrap. Select a suitable assortment of thickness to make a filter sandwich of any thickness in 0.016 in. thick increments up to 3/32 in. Machine at least one Cu end-cap filter 0.020 in. thick. The Cu-Cd sandwich can be fastened to the cylindrical wrap with tape.

A backshield should be placed behind the detector to provide shadow shielding against background gamma rays incident from behind. This may have to be set up for each individual measurement. For certain types of cold-finger/end-cap configurations it is possible to install a split ring backshield around the cold-finger behind the end-cap. A tungsten split disk covered with Cd and Cu facing the detector is one way to implement a backshield whose thickness may be tailored to the available space.

Fig. 7 A Cd-wrapped end cap for a 28 % relative efficiency right angle dipstick coaxial detector.

1.5 CONFIGURATIONS FOR HIGH ACCURACY EMP MEASUREMENTS

The work of (Parker 88) is probably the single, most comprehensive and complete description of high accuracy EMP measurements available. A portion of Fig. 1 showing Parker's experimental setup is expanded below in Fig. 8.

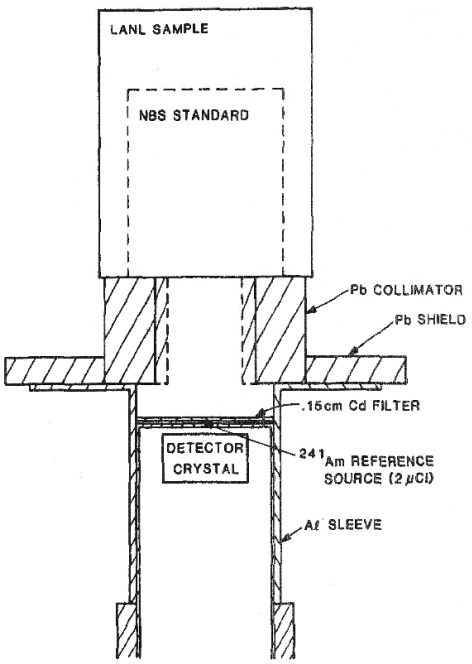

Fig. 8. Original caption in LA-11277-MS reads: "A mechanical schematic of the actual counting arrangement used for gamma-ray measurements on both the LANL oxides and the NBS-969 certified reference standards".

Details of Parker's measurements follow below:

- He used "medium-sized" coaxial HPGe detectors. The detector size is not given but given the date of the measurements (1988) the detector is undoubtedly quite small by today's standards, likely a 15–20 % relative efficiency coaxial crystal.

- The detector resolution was ~ 750 eV FWHM at 122 keV.

- He used high quality NIM format electronics with an amplifier analog time constant of 3 μs and pileup rejection.

- His system was operated at a gain of 0.20 keV/ch with data stored in 1024 channels.

- Digital gain and Zero stabilization was used with Zero stabilization on the 59.5 keV ^{241}Am reference peak and Gain stabilization on the 185.7 keV ^{235}U assay peak.

- The approximately 2 μCi ^{241}Am reference source was taped directly to the end cap to provide a constant reference rate (constant save for the slow decay of its ~433 y half life).

- The 1.5 mm thick Cd filter placed on the measurement item side of the reference source above the detector was designed to assure complete absorption of the 58.6 keV gamma from ^{235}U to prevent any interference with the Am reference peak. (The NuDat2 database gives a 58.5700 (24) keV gamma from ^{231}Th which would be in equilibrium with ^{235}U.)

- The lead collimator, 5.08 cm (2.00 in.) deep, was mounted on the detector end-cap flange to minimize any movement of the collimator and detector with respect to the measurement item. Different sized collimator inserts were used with the different-sized items. The collimator was 3.8 cm (1.5 in.) in diameter for use with the NBS 969 series standards and 5.08 cm diameter for measurements on the larger LANL standards.

- Data extracted from the measurements were the net full energy peak areas of the ^{241}Am reference source peak and the 185.7 keV ^{235}U peak. Data analysis was performed on the ratio of the 185.7 peak area/59.5 keV peak area counted for a true time long enough to give an estimated 0.1% rsd in the ratio.

Consult (Parker 88) for a detailed explanation of all facets of the measurement and data analysis.

2. ANALYSIS OPTIONS WITH FRAM FOR COMPLETE ISOTOPIC ANALYSIS

The FRAM gamma ray isotopic analysis program can measure the complete isotopic distribution of uranium-bearing samples in two energy regions. Either the 120 keV to 1010 keV region can be used or, with an appropriate detector, FRAM can also measure the complete uranium isotopic distribution using gamma rays and x-rays in the 60-250 keV region.

2.1 FRAM URANIUM ISOTOPIC COMPOSITION MEASUREMENTS IN THE 120- TO 1010-KEV REGION

The energy region from 120–1010 keV is used (usually with a moderate to large-sized coaxial detector) to measure the complete isotopic distribution of uranium including direct measurement of ^{234}U, ^{235}U, and ^{238}U. ^{236}U, having no detectable gamma rays, is estimated using an isotopic correlation based on the production of the material. FRAM can measure all enrichments from < 0.2 % to > 97 % ^{235}U in this energy range. Fig. 9 below shows the general characteristics of the coaxial detector gamma ray spectrum for three uranium enrichments.

One of the main features of FRAM is its user-editable set of parameters that governs all phases of a FRAM analysis. A complete group of parameters is called a parameter file or parameter set. A parameter set is developed to apply to a large range of isotopic analysis characteristics and is easily modified (requires moderate gamma-ray spectroscopy experience) to accommodate varied measurement and analysis conditions. Two standard parameter sets, delivered with the FRAM version 5 software, are available for the analysis of uranium gamma-ray spectra in the 120 to 1010 keV range. The low-enriched uranium (LEU) parameter set, ULEU_Cx_120-1010, is designed to analyze enrichments up to approximately 40% ^{235}U. The parameter set for high-enriched uranium (HEU), UHEU_Cx_120-1010, is designed for ^{235}U enrichments above 10%. Either parameter set may be used in the region where they overlap.

2.1.1 FRAM Measurements on Low-Enriched Uranium in the 120- 1010-keV Energy Range

The coaxial detector LEU parameter set, ULEU_Cx_120-1010, uses ^{235}U gamma rays at 143.7, 163.4, 185.7, and 205.3 keV and ^{234m}Pa (^{238}U daughter) gamma-rays at 258.3, 742.8, 766.4, 880.5, 883.2, 946.0, and 1001.0 keV to define the relative efficiency curve. A relative efficiency curve from a uranium standard with 4.46% ^{235}U is shown in Fig. 10.

With the exception of the 258.3 keV relative efficiency point, all the ^{238}U relative efficiency points are above 700 keV and all ^{235}U relative efficiency points are below 210 keV with a large gap in between. The normalization of the two sets of data points depends almost solely on the 258.3 keV ^{234m}Pa (^{238}U daughter) relative efficiency point meaning that the ^{235}U/^{238}U ratio and its uncertainty is determined mainly by this single peak for LEU.

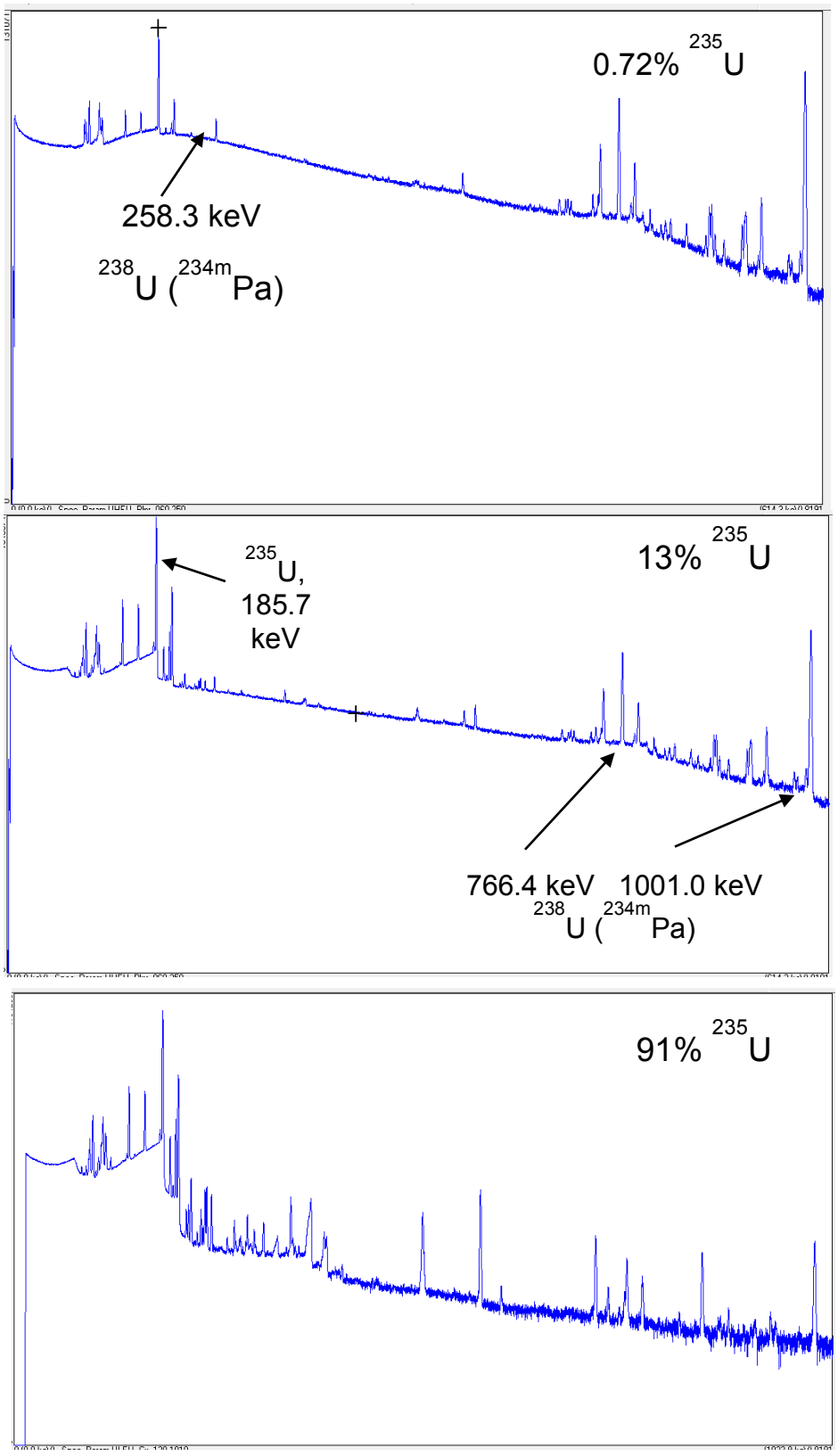

Fig. 9. Coaxial detector spectra of three uranium enrichments in the 0–1024 keV range. The vertical scale is logarithmic.

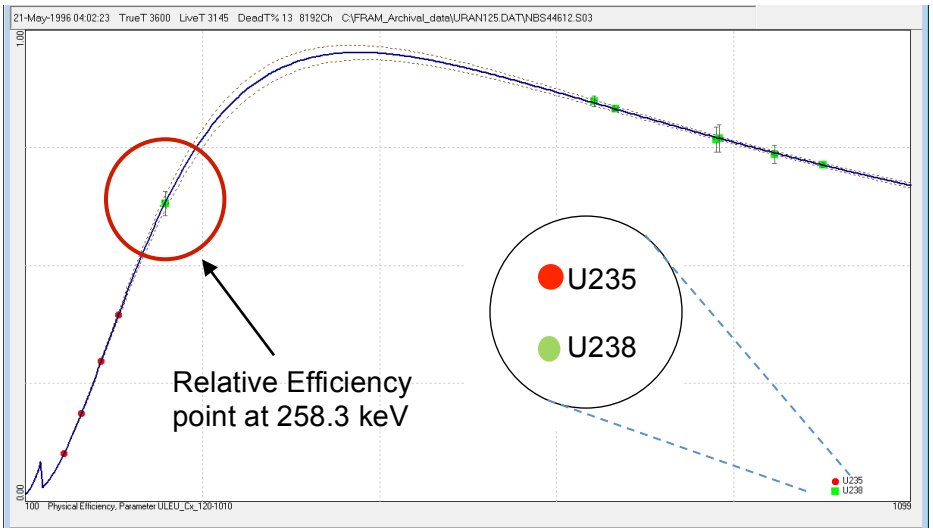

Fig. 10. FRAM relative efficiency curve for an item containing ~ 200 g uranium in the form of U_3O_8 powder with 4.46% ^{235}U. The important relative efficiency point at 258.3 keV is indicated.

2.1.2 Coincidence Summing for Uranium Isotopic Measurements in the 120-1010-keV Range

The decay scheme of ^{234}Pa shows that many of the gamma rays useful for ^{238}U isotopic ratio measurements are in true coincidence with each other. This means that there is an increased probability of two coincident gamma rays entering the detector at the same time producing a pulse that does not correspond to either energy but to the sum of their energies or Compton-scattered energy. This true coincidence summing effect depends upon the subtended solid angle of the detector as seen from the sample, thus leading to a strong sample-to-detector distance dependence. This distance dependence is shown in Fig. 11.

Vo has extensively studied this problem as it relates to FRAM (Vo 98, Vo 99, Vo 99a). The results of these studies, while in the context of an earlier version of FRAM, are basically valid for the current version 5 of FRAM. A correction for this effect has been implemented in versions 4 and 5 of FRAM.

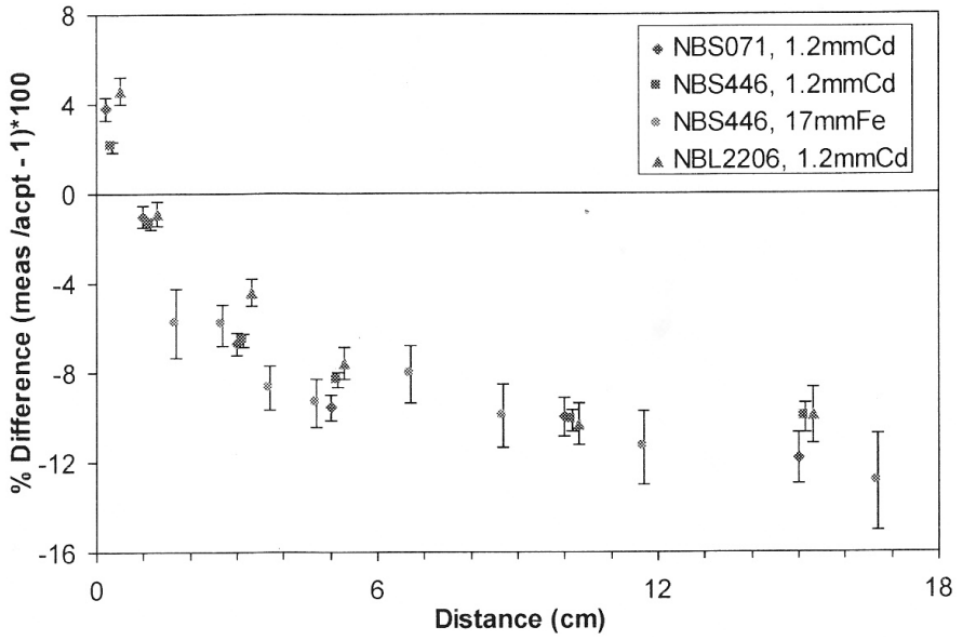

Fig. 11. Without a coincidence summing correction the results can vary by as much as 14 % over a 0–15 cm distance (Vo 99).

The coincidence summing correction removes the distance dependence of Fig. 11 at the cost of increasing the statistical uncertainty as shown in Fig. 12.

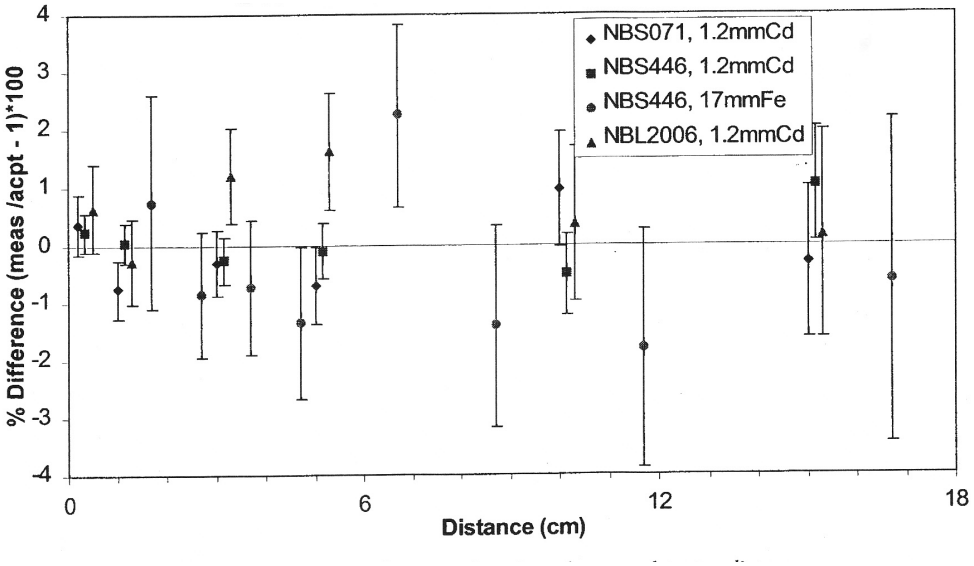

Fig. 12. With coincidence summing correction the distance dependence is removed (Vo 99).

2.1.3 FRAM Measurements on High-Enriched Uranium in the 120 – 1010 keV Energy Range

Most of the HEU seen in the US contains small amounts of ^{232}U, arising from recycling reactor returns through the enrichment cycle. The ^{232}U decays to ^{228}Th whose decay chain gives rise to familiar gamma rays also found in natural background. The ^{228}Th chain gamma rays are present in the spectrum of HEU in varying amounts depending upon the specific production history. In general, the amount of ^{228}Th decreases with decreasing enrichment. While some ^{228}Th and daughters may be found even in power reactor fuel enrichments (< 5 % ^{235}U), their measurement at low enrichments is complicated by their low abundance and the presence of the same gamma ray peaks in natural background.

For these reasons FRAM uses a different standard parameter file for 120 – 1010 keV range coaxial detector measurements. This parameter file is UHEU_Cx_120-1010 and it is recommended for use over the enrichment range > 10%.

The same ^{228}Th daughter peaks at 238.6, 583.2, 727.3, 763.3, and 860.6 keV are included in both the UHEU_Cx_120-1010 and ULEU_Cx_120-1010 parameter files. However, in the UHEU_Cx_120-1010 parameter file the ^{228}Th daughter peaks at 238.6, 583.2, 727.3, and 860.6 keV are used also to help define the relative efficiency curve.

2.1.4 The Role of 238.6 keV ^{228}Th Daughter and 258.3 keV ^{234m}Pa Peaks in the FRAM Relative Efficiency Curve for Uranium

For HEU the intensity of the 258.3 keV ^{238}U daughter peak is very weak essentially disappearing for enrichments > 90%. For these enrichments this peak is no longer available to effectively perform the "normalization" between the high energy relative efficiency points from ^{238}U and the low energy relative efficiency points from ^{235}U. Instead, for HEU the ^{228}Th daughter peak at 238.6 keV is used along with the higher energy ^{228}Th daughter peaks at 583.2, 727.3, and 860.6 keV. The intensities of the ^{228}Th daughter peak at 238.6 keV and the 258.3 keV ^{238}U daughter peak are shown for three different enrichments in Fig. 13.

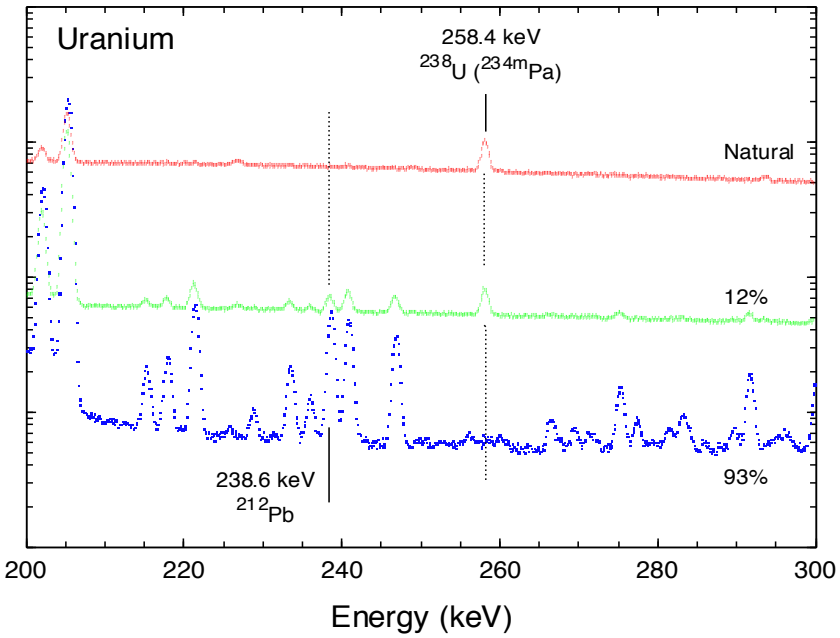

Fig. 13. The contributions from the ^{212}Pb-Th daughter and the ^{234m}Pa-^{238}U daughter change with enrichment. Th daughters disappear for low enrichments (<~10%) and the ^{234m}Pa peak disappears for high enrichments (>~75%).

Figs. 14 and 15 show the relative efficiency curves for two enrichments at the ends of the applicable range of the UHEU_Cx_120-1010 parameter file.

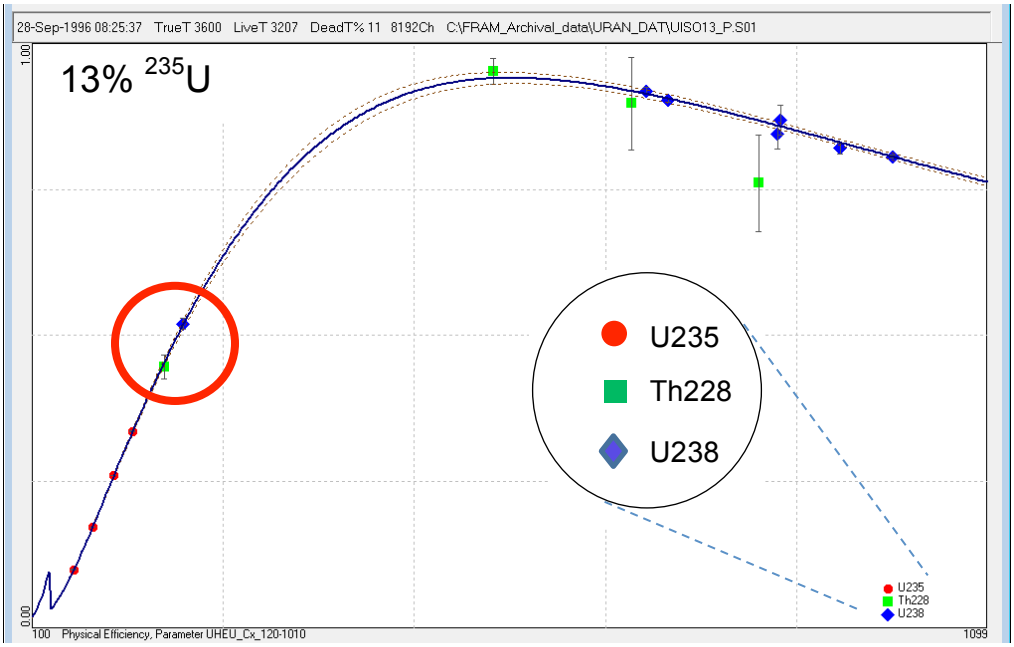

Fig. 14. For this item of 13 % ^{235}U, both the 238 keV ^{228}Th daughter and the 258 keV ^{238}U daughter peaks provide good relative efficiency results.

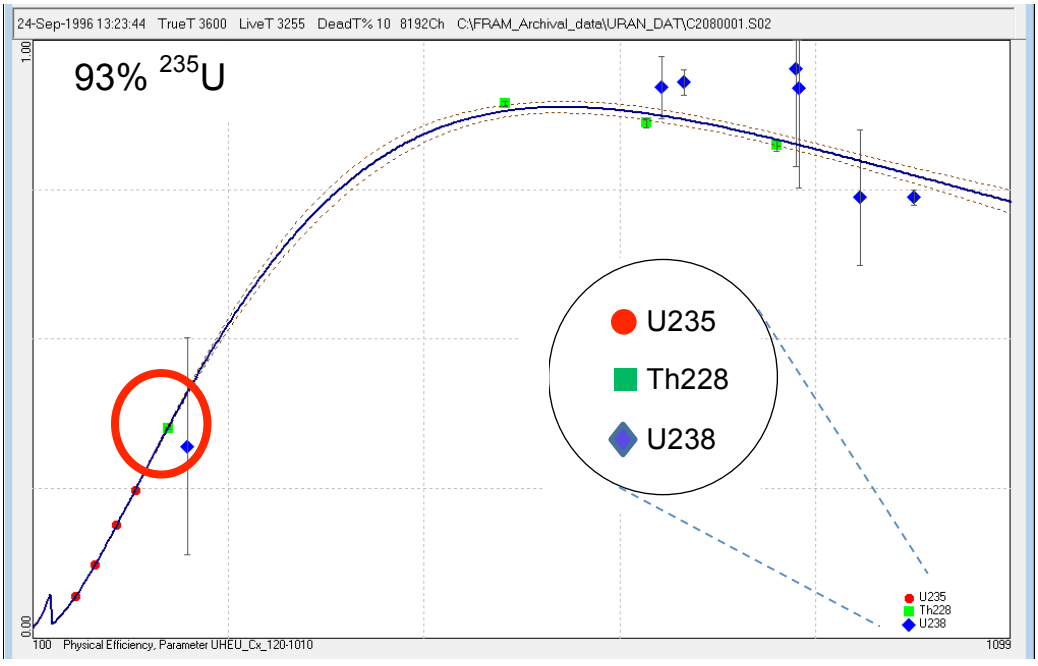

Fig. 15. For HEU the Th daughter peaks help determine the relative efficiency at higher energies as well as providing a "connection" at 238 keV. The 258 keV ^{238}U daughter peak does not contribute to relative efficiency for HEU (too weak).

2.1.5 Acquisition Conditions for FRAM Isotopic Composition Measurements in the 120- to 1010-keV Region

FRAM measurements in this energy region are most often carried out with coaxial HPGe detectors with relative efficiencies of at least 25% (an exception is discussed in sec 1.1). Larger detectors also work well because the resolution demands for uranium analysis in this energy region are not great.

Historically, for these measurements and FRAM analyses, spectra are acquired in 8192 channels at a conversion gain of 0.125 keV/ch, covering the energy range 0–1024 keV. The standard parameter files, UHEU_Cx_120-1010 and ULEU_Cx_120-1010, built into version 5 of FRAM are set up for a gain of 0.125 keV/ch although this gain may be easily changed on an analysis-by-analysis basis to meet spectrum requirements.

Uranium analyses in this energy region have been successfully carried out at gains from 0.075 to 0.25 keV/ch. When the gain is very high (0.075 keV/ch) and coupled with a poorer resolution detector, the peaks at high energies become very broad. FRAM has a maximum limit of 32 channels for the FWHM of an analyzed peak. FRAM will not analyze any peak with a FWHM >32 ch. and the analysis will be completed with just the peaks that can be analyzed. It is not uncommon to see spectra that cannot be completely analyzed because the high-energy peaks have a FWHM of >32 ch.

It is recommended that the spectrum gain be set up so that the analysis peaks of interest have a FWHM that falls in the range from 8–12 ch.

Uranium analyses in this energy region must be carried out with shielding and filtering as discussed in Section 1.4. At least 2 inches of Pb shielding is needed if the measurements are to be carried out in a location with any significant ambient background either from uranium or natural sources. Graded-Z shielding and filters should be used to suppress fluorescent x-rays from any Pb shielding and low-energy gamma rays (< 80 keV). There is no need to have any visible peaks in the spectrum below 80 keV or so. However, avoid using excessive filtering because it is necessary to retain measurable intensity for the 120 keV peak from ^{234}U.

2.2 FRAM URANIUM ISOTOPIC COMPOSITION MEASUREMENTS IN THE 60- TO 250-KEV REGION

The energy region from 60 to 250 keV is used (usually with a high resolution planar or semi-planar detector) to measure the complete isotopic distribution of uranium including direct measurement of ^{234}U, ^{235}U, and ^{238}U. ^{236}U, having no detectable gamma rays, is estimated using an isotopic correlation. FRAM can measure all enrichments from < 0.2 % to > 97 % ^{235}U in this energy range. Fig. 16 below shows the general characteristics of the planar detector gamma ray spectrum for three uranium enrichments.

FRAM covers the entire enrichment range with two standard, built-in parameter sets, ULEU_Plnr_060-250 for enrichments <40% and UHEU_Plnr_060-250 for enrichments >10%. Either may be used in the range where they overlap.

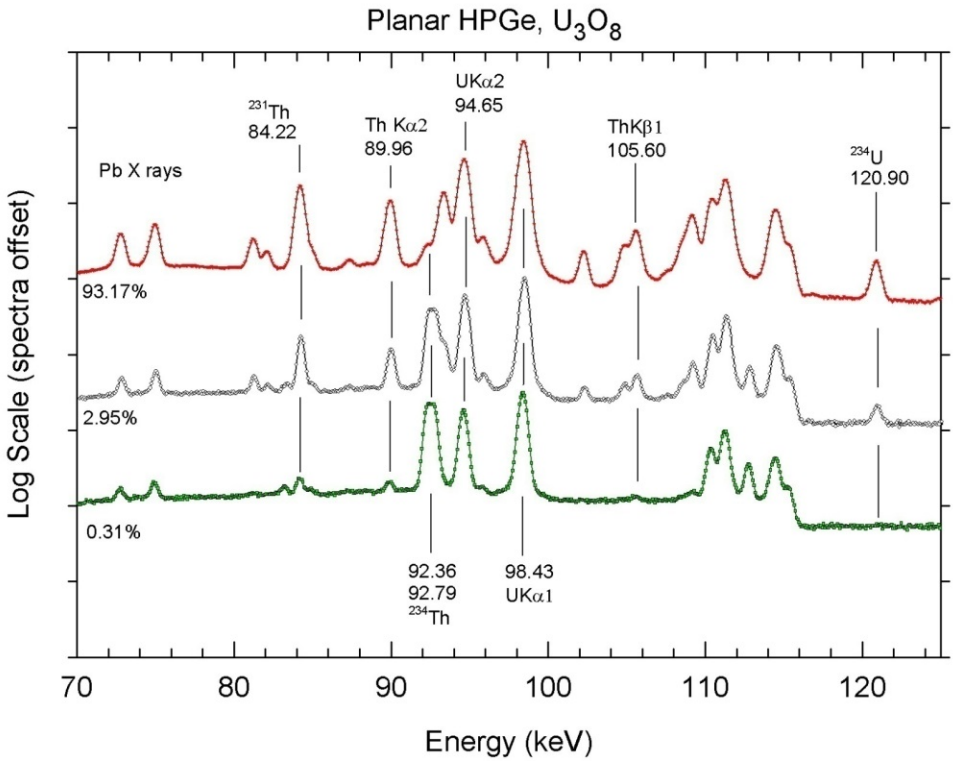

Fig. 16. The 100 keV region of the gamma ray spectrum changes greatly as a function of ^{235}U enrichment.

2.2.1 FRAM Measurements on Low-Enriched Uranium in the 60-250-keV Energy Range

The LEU parameter set for the 60–250 keV energy range, ULEU_Plnr_060-250, uses the ^{235}U-related peaks at 84.2 keV (^{231}Th), 89.96 keV (ThKα_2), 94.65 keV (UKα_2), 98.43 keV (UKα_1), 105.60 keV (ThKβ_1), and peaks directly from the decay of ^{235}U at 143.8, 163.4, 185.7, and 205.3 keV to define the relative efficiency curve. The branching intensities and energies of some of the peak data have been empirically adjusted for best results judged against known U-standards from the initial starting nuclear data values in the NuDat 2 database from the National Nuclear Data Center at Brookhaven. Fig. 17 shows these relative efficiency peaks highlighted on a spectrum (vertical log scale) from a LEU sample.

The activity of ^{234}U is determined from the single peak at 120.90 keV. The activity of ^{238}U is determined from the 92.36 keV gamma ray of ^{234}Th (^{238}U daughter). The activity of ^{235}U is determined from the ^{235}U-related peaks at 84.2 keV (^{231}Th), 89.96 keV (ThKα_2), 105.60 keV (ThKβ_1), and peaks directly from the decay of ^{235}U at 143.8, 163.4, 185.7, and 205.3 keV.

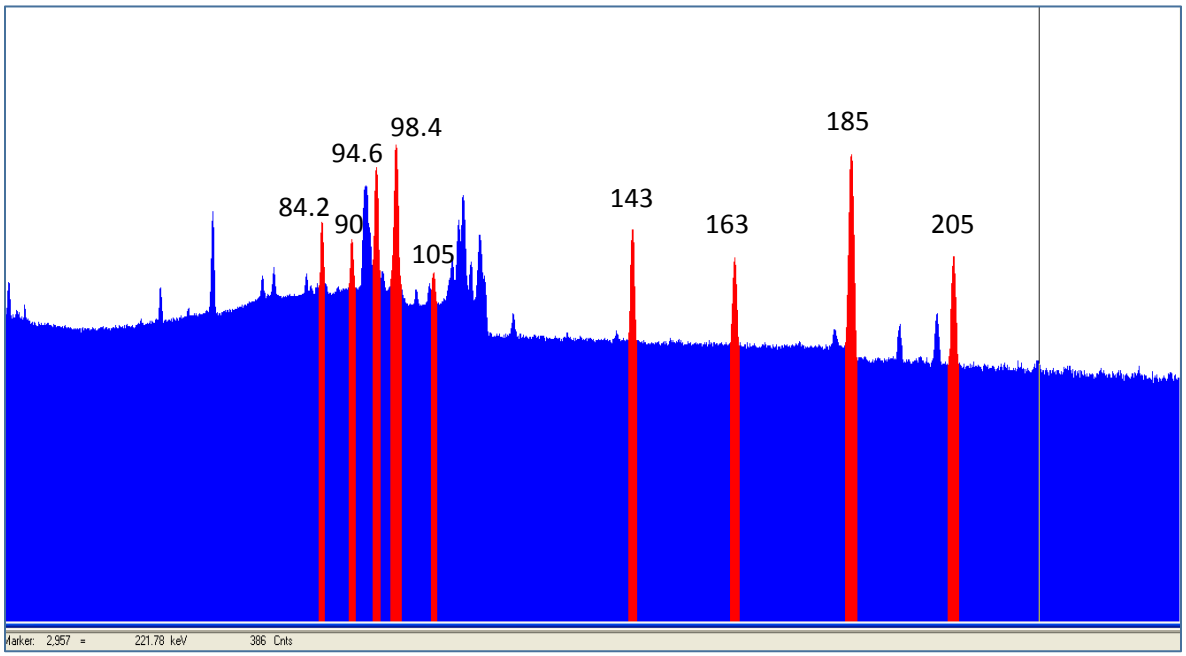

Fig. 17. FRAM spectrum of LEU showing the relative efficiency peaks. Peak energies in keV are noted.

A relative efficiency curve from a uranium standard with 4.46 % ^{235}U is shown in Fig. 18.

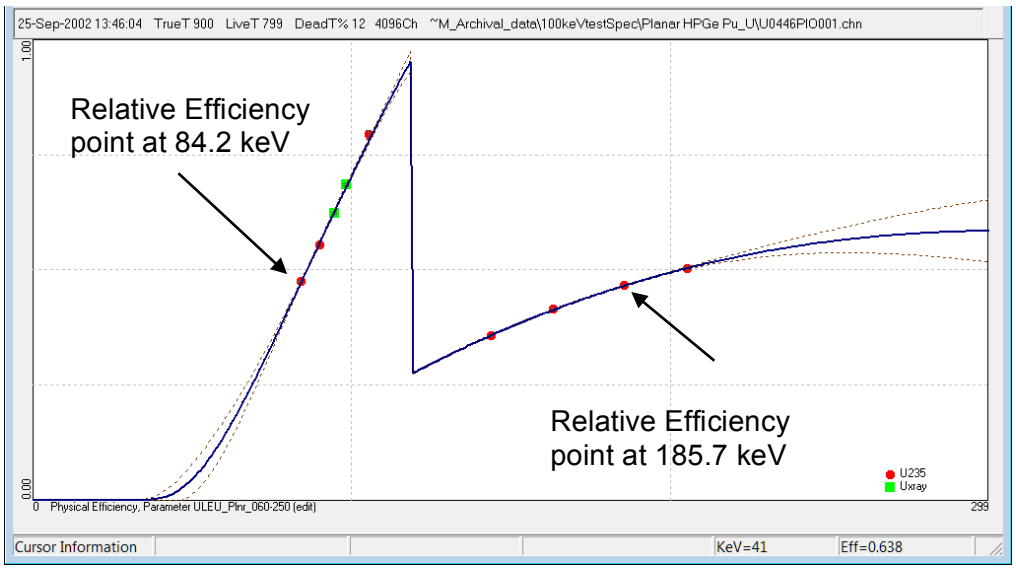

Fig. 18. FRAM relative efficiency curve for an item containing ~200 g uranium in the form of U_3O_8 powder with 4.46 % ^{235}U.

The spectrum that produced the above relative efficiency curve is shown in Fig. 19. The vertical scale is logarithmic and the horizontal scale spans the energy range 0–307 keV.

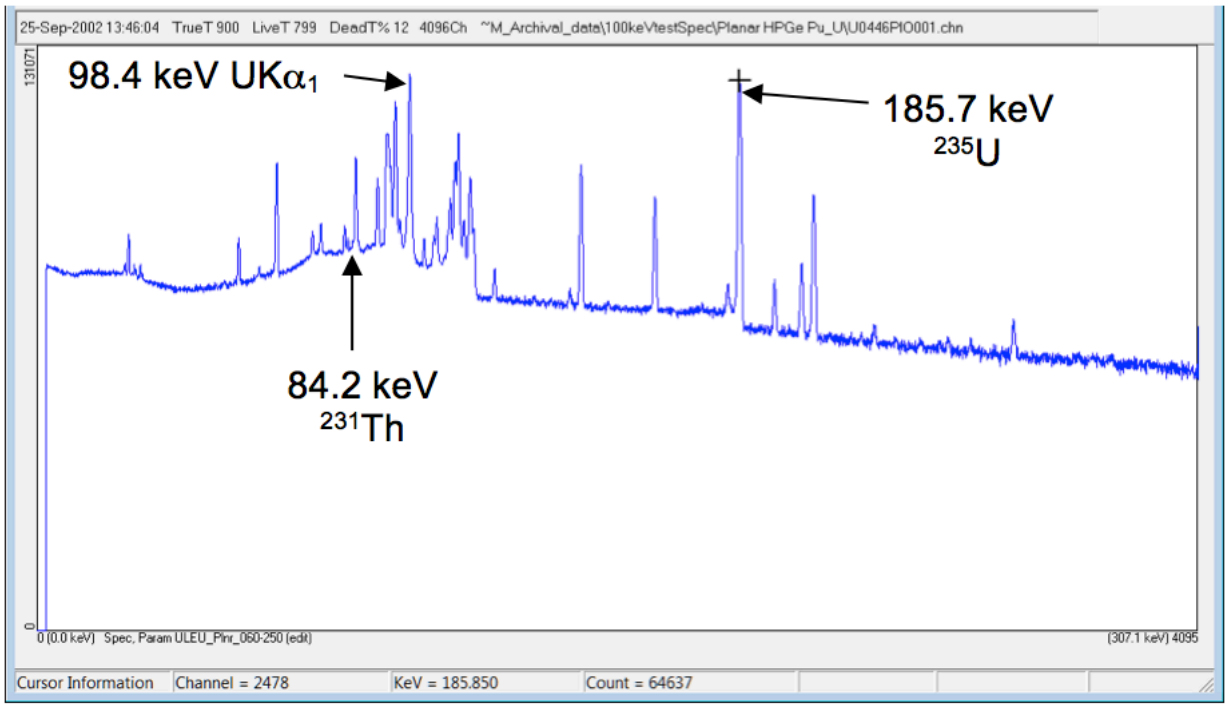

Fig. 19. Planar detector spectrum from an item containing ~200 g uranium in the form of U_3O_8 powder with 4.46 % ^{235}U. Vertical scale is logarithmic.

Note that the spectrum in Fig. 19 has little or no filtering, contrary to what has been recommended previously in this document. Observe there are weak Pb x-rays in the spectrum. FRAM can correct for weak Pb Kβ x-ray interference at 84.22 keV, as in this spectrum, but cannot make an accurate correction when the interference is large as it is in Fig. 4a. Fig. 20 shows the FRAM display of the fitting in the 84 keV region of the spectrum in Fig. 19.

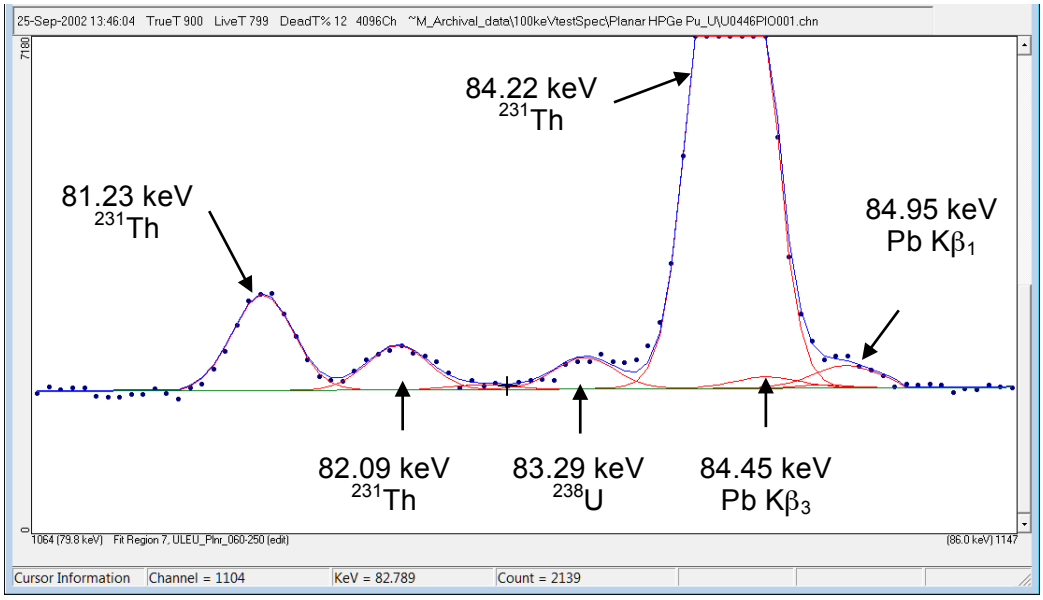

Fig. 20. FRAM display of the fitting of Pb Kβ x-ray interferences in the 84 keV region of the spectrum in Fig. 19. Vertical scale is linear.

LEU-product items, in general, are not difficult to analyze in the 100 keV region given good quality data because the peaks are not heavily overlapping.

2.2.2 FRAM Measurements on High-Enriched Uranium in the 60-250-keV Energy Range

The HEU parameter set for the 60–250 keV energy range, UHEU_Plnr_060-250, uses the ^{235}U-related peaks at 84.2 keV (^{231}Th), 89.96 keV (ThKα_2), 94.65 keV (UKα_2), 98.43 keV (UKα_1), 105.60 keV (ThKβ_1), and peaks directly from the decay of ^{235}U at 143.8 , 163.4 , 185.7 , and 205.3 keV to define the relative efficiency curve. The branching intensities and energies of some of the peak data used in FRAM have been adjusted for best results from the initial starting values in the NuDat 2.6 database from the National Nuclear Data Center at Brookhaven.

The activity of ^{234}U is determined from the single peak at 120.90 keV. The activity of ^{238}U is determined from the 92.36 keV gamma ray of ^{234}Th (^{238}U daughter). The activity of ^{235}U is determined from the ^{235}U-related peaks at 84.2 keV (^{231}Th), 89.96 keV (ThKα_2), 105.60 keV (ThKβ_1), and peaks directly from the decay of ^{235}U at 143.8, 163.4, 185.7, and 205.3 keV. In addition ^{228}Th is quantified from the activity at 238.6 keV from its ^{212}Pb daughter.

A planar-detector gamma-ray spectrum from a Certified Reference Material with ^{235}U enrichment of 52.49 % is shown in Fig. 21.

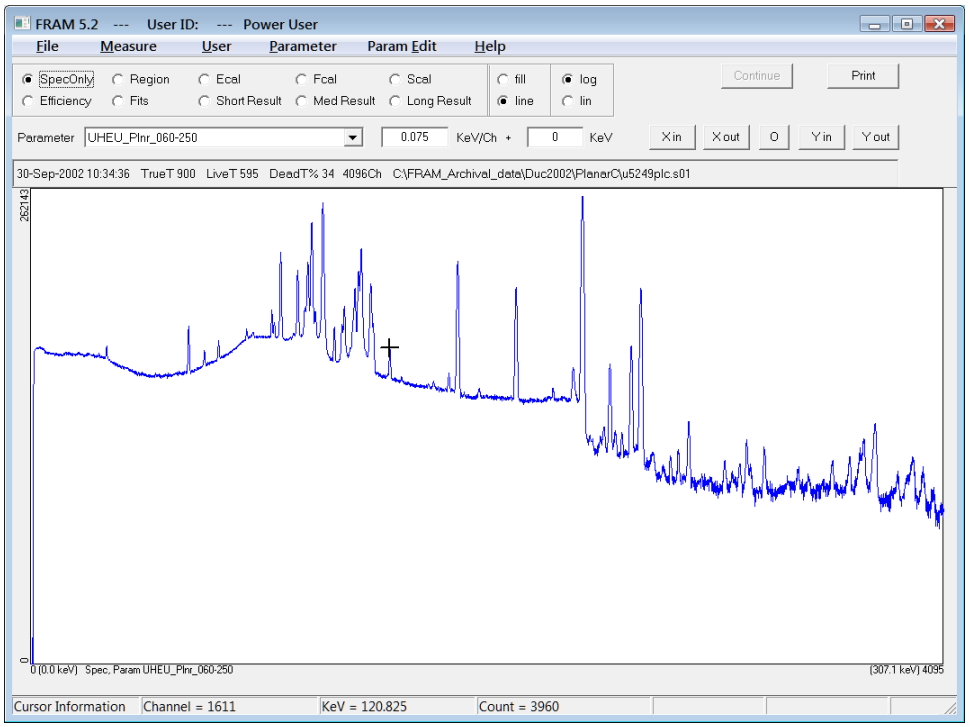

Fig. 21. Planar detector spectrum from an item containing ~ 200 g uranium in the form of U_3O_8 powder with 52.49% ^{235}U. Vertical scale is logarithmic. The cross hair marks the 120.9 keV peak of ^{234}U.

Uranium isotopic analysis using the 100 keV region becomes difficult at the high end (> 90% ^{235}U) of the HEU range because of the difficulty of extracting the 92.36 keV peak of the so-called “Thorium Doublet” (92.36 and 92.79 keV from ^{234}Th), (Sampson 73) from the stronger interferences from ThKα_1 at 93.35 keV and Pa Kα_2 at 92.29 keV. The 92.36 keV peak of ^{234}Th determines the ^{238}U isotopic fraction. A detector that exhibits low-side tailing will exacerbate this problem. Good peak shape is important for analysis in this energy range. Fig. 22 shows the thorium doublet for a mid-range enrichment.

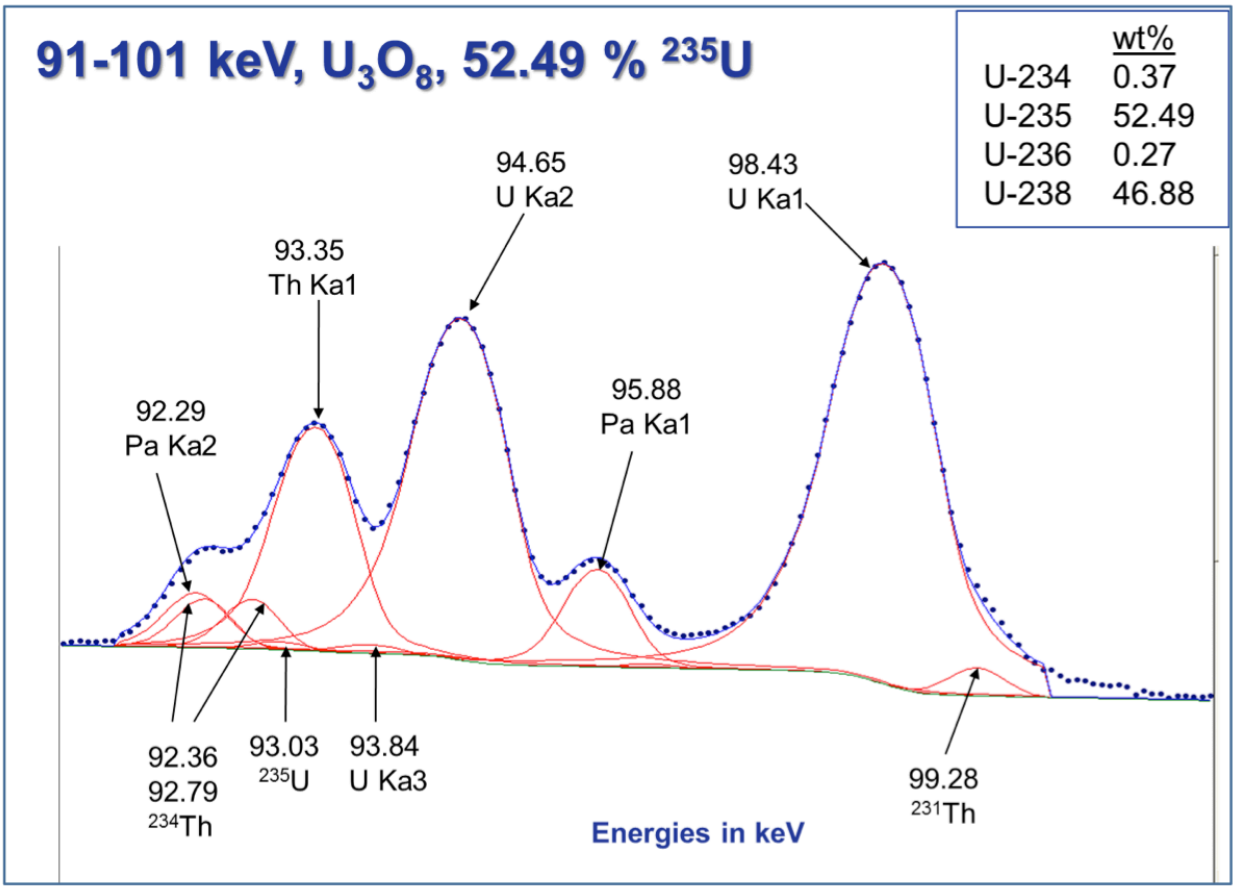

Fig. 22. Planar detector spectrum from an item containing ~ 200 g uranium in the form of U_3O_8 powder with 52.49 % ^{235}U. Vertical scale is logarithmic. The strong interferences with the thorium doublet are apparent at this enrichment.

For comparison Figs. 23 and 24 show the thorium doublet at the extremes of the enrichment range.

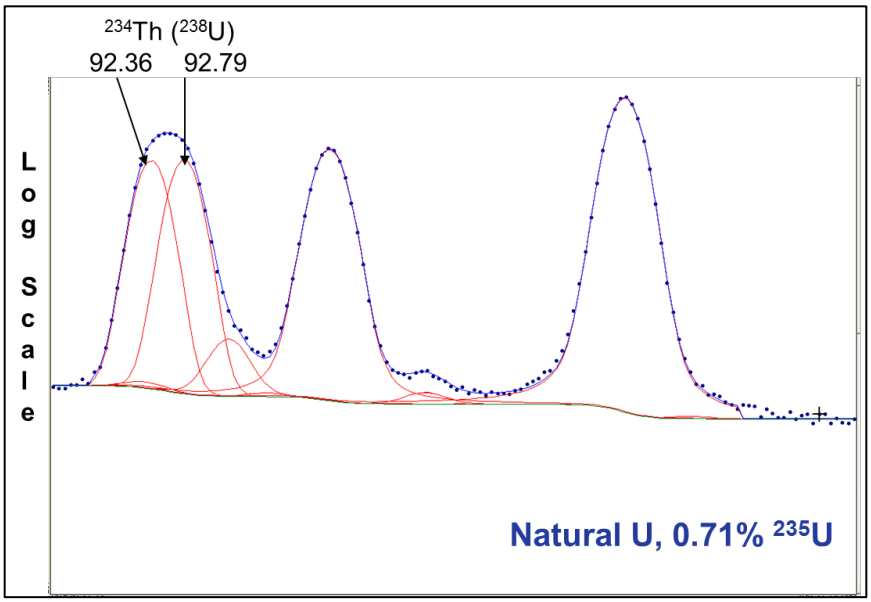

Fig. 23. Planar detector spectrum from an item containing ~ 200 g uranium in the form of U_3O_8 powder with 0.71% ^{235}U. Vertical scale is logarithmic. The thorium doublet is easily analyzed at this enrichment.

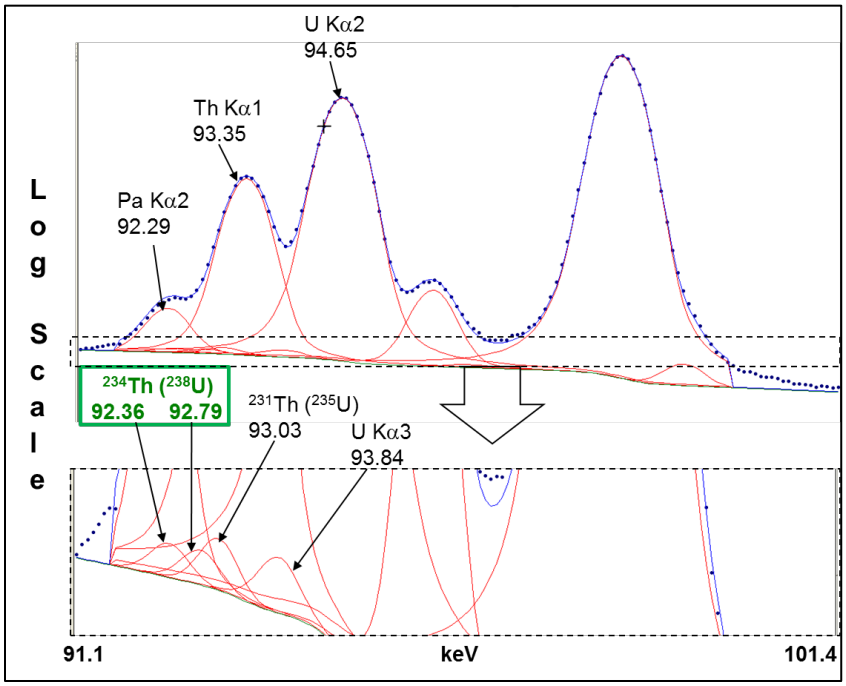

Fig. 24. Planar detector spectrum from an item containing ~ 200 g uranium in the form of U_3O_8 powder with 93.17 % ^{235}U. Vertical scale is logarithmic. The thorium doublet is completely swamped at this enrichment.

The Pb x-ray interferences that were discussed for LEU analysis are usually not as significant for HEU because the 84.22-keV ^{231}Th peak is much stronger than for LEU.

3. FRAM ANALYSIS WITH SPECIAL PARAMETER FILE FOR ENRICHMENT METER PRINCIPLE (EMP) MEASUREMENTS

In this section we will also discuss the possibility of using FRAM as the basis for classical EMP measurements of ^{235}U enrichment using the direct 185.72 keV ^{235}U gamma ray.

The principles of the measurement of ^{235}U enrichment using the EMP were outlined as early as 1971 (Reilly 71). The most complete description of the salient procedures to ensure quality data and of the underlying principles was published by Matussek in 1985 (Matussek 85) as a user's manual for the Certified Reference Materials EC-NRM-171/NBS-SRM-969. Although no longer available, these standards were available from New Brunswick Laboratory as CRM 969. A practical and probably the most comprehensive description of accurate and precise wide-range EMP measurements was published by Parker and Brooks (Parker 88). The reader is urged to consult these documents before undertaking EMP measurements and these details will not be duplicated or discussed here. However, it is noted that EMP measurement techniques must be mastered before high-accuracy measurements are undertaken. This section describes the preliminary steps taken to permit the FRAM (Sampson 03) gamma-ray isotopic analysis software to be used as an "engine" to extract the necessary information from a properly-configured uranium sample measurement to determine the ^{235}U enrichment. The details of the FRAM software are beyond the scope of this document and it is assumed that the reader has some familiarity with the software and its use.

3.1 WHY USE FRAM AS BASIS FOR EMP MEASUREMENTS?

This is a logical question given that FRAM is a proven and widely used tool that can already perform complete isotopic measurements on an arbitrary uranium item (as noted in Section 2) while the constraints on EMP measurements are numerous.

One reason for using FRAM that has been expressed is that the documentation, diagnostic and reporting structure, and relative transparency of the inner workings of the FRAM code make it easier to understand the data extraction process – especially when a user may already be familiar with FRAM and less so with some other spectrum analysis tools.

As will be discussed below there are also complications that can arise because FRAM's inner workings require information from the spectrum that is not needed for simpler EMP analyses.

3.2 EXTRACTION OF PEAK AREAS WITH FRAM

The extraction of peak areas (185.7 keV from ^{235}U and perhaps 59.5 keV from ^{241}Am rate-loss correction source) and their uncertainties would be the output from a FRAM "engine" analysis. FRAM determines peak areas using a response function fitting process. To use a response function to obtain an accurate peak area the algorithm must know the position of the peak (precise energy calibration), the width (FWHM) of the peak, and the shape (tailing) of the peak. FRAM obtains this from the spectral data itself on every measurement by analyzing strong single peaks spread throughout the spectrum. The shape is energy-dependent as is the width of the peak. It is desirable to have these internal calibration peaks at both high and low energies.

EMP measurements are usually fairly short and given the constrained, collimated geometry, may not have strong peaks over the entire uranium spectrum. The upside, here, is that ^{235}U has strong single peaks at 143, 163, 186, and 205 keV that can be analyzed in most spectra to establish the response function

constants. An ^{241}Am reference source peak added at 59.5 keV will further improve the response function internal calibration. These peaks over this limited energy range should make it possible to use a FRAM response function analysis.

FRAM also has the ability to fix the response function constants at user-editable default values. This would be of value when performing measurements with poor counting statistics using a detector whose default constants had been previously determined from high quality spectra.

3.3 A FRAM PARAMETER FILE FOR EMP MEASUREMENTS

A standard built-in uranium parameter file in version 5 of FRAM was modified to produce a parameter file for uranium EMP measurements. This new parameter file, u_enrichment_emp, contains eight ^{235}U peaks from 140 to 205 keV. The four principal ^{235}U peaks at 143.76, 163.36, 185.715, and 205.311 keV are used for the internal calibration of the response function fitting parameters.

Two groups of data supplied by ORNL were analyzed with this new parameter file. Both sets of data were from the standard uranium enrichment CRMs, CRM 146 and CRM 969 with the first set of data omitting the 1.95 % and 2.95 % enrichment items.

The first set of data was taken at 0.075 keV/ch in 4K channels, spanning the energy range 0–307 keV. The detector resolution at 185.7 keV appeared to be approximately 670 eV and the peak shape visually appeared to be excellent (almost pure Gaussian). The measurements were made in a horizontal-looking geometry, a configuration that somewhat increases the difficulty of obtaining reproducible results. The acquisition time constants used were not stated while the counting rates (also not stated) produced dead times ranging from 2% to 21%. Each item was measured for a fixed live time of 3600 s.

Analysis of the raw data from the first data set, presented in Table 3, looks at the consistency of the ratio of [185.7 keV peak area/^{235}U atom%/Live Time] as a measure of how well the measurement results follow the EMP assumptions.

Table 3. First EMP Dataset.

File Name	True Time (s)	Live Time (s)	Dead Time (%)	U-235 Enrichment (atom %)	% RSD	Area 185 keV	% RSD	A/Enrich/LT	% RSD
Depleted_Planar_1hr.cnf	3664	3600	2%	0.3206	0.030	39441	0.65	34.1729	0.65
Natural_Planar_1hr.cnf	3666	3600	2%	0.7209	0.035	85447	0.36	32.9245	0.36
4.46%_Planar_1hr.cnf	3698	3600	3%	4.5168	0.036	533514	0.19	32.8105	0.19
20.11%_Planar_1hr.cnf	3835	3600	7%	20.311	0.050	2405052	0.13	32.8920	0.14
52.49%_Planar_1hr.cnf	4108	3600	13%	52.800	0.040	6272748	0.15	33.0006	0.16
93.17%_Planar_1hr.cnf	4501	3600	21%	93.2330	0.0028	11098571	0.15	33.0670	0.15

Omitting the depleted uranium result gives a maximum difference of less than 0.8% between the highest and lowest value. In Fig 25 the results with enrichment expressed in mass % and atom % are shown. The 1-sigma error bars (not shown) for the atom % data are the same as those for the mass % results.

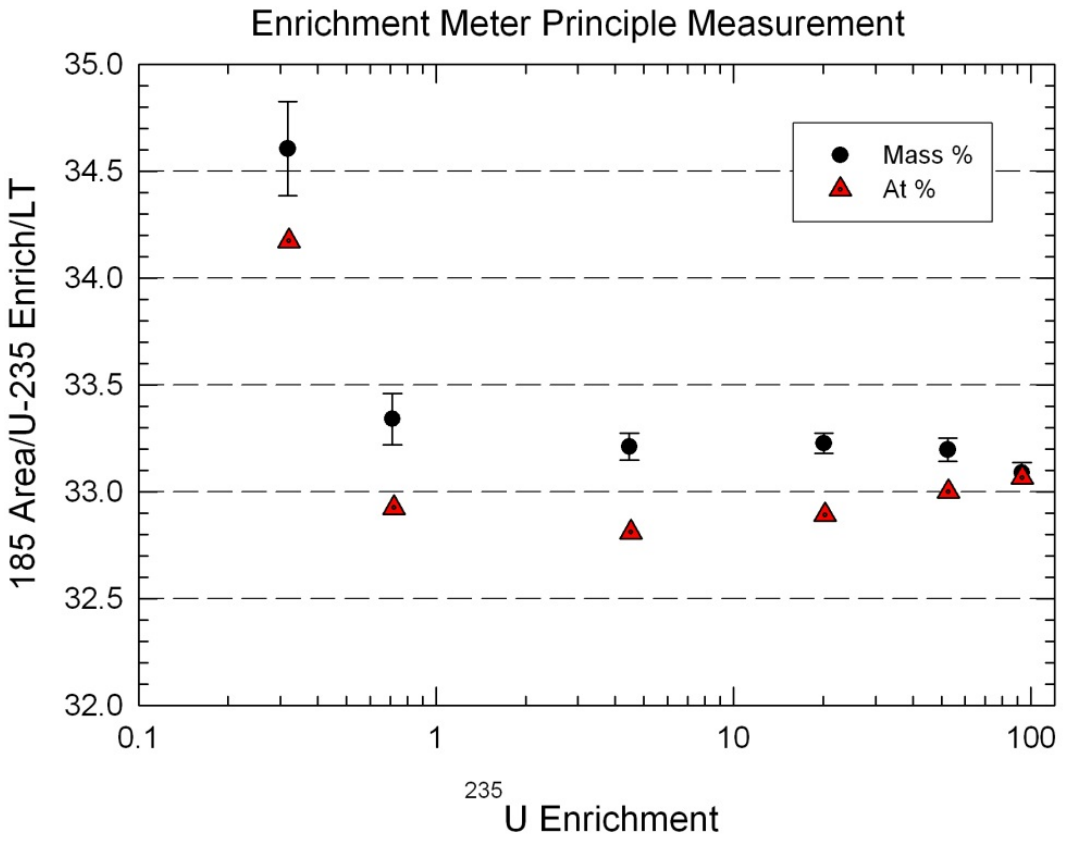

Fig. 25. Enrichment Meter Principle Data from first dataset.

The second set of data consisted of one spectrum for each item that was the summation of twenty 300 sec true time measurements. The geometry is clearly not the same as in the first dataset because the peak count rates are much lower for the same measured items. The data in tabular form are shown in Table 4 while plots of the data (with enrichment expressed both as atom % and mass %) are shown in Fig 26. As in the previous plot the atom % 1-sigma error bars (not shown) are the same size as those for the mass % points.

The atom % data for enrichments above 1% are very consistent.

Table 4. Second EMP Dataset.

File Name	True Time (s)	Live Time (s)	Dead Time (%)	U-235 Enrichment (atom %)	% RSD	Area 185 keV	% RSD	A/Enrich/LT	% RSD
739_none_sur	6000	5902	2%	0.3206	0.03	22463	0.89	11.871	0.89
650_none_sur	6000	5902	2%	0.7209	0.035	49914	0.52	11.731	0.52
563_none_sur	6000	5898	2%	1.9664	0.036	133679	0.29	11.526	0.29
476_none_sur	6000	5894	2%	2.9857	0.036	201326	0.23	11.44	0.23
389_none_sur	6000	5885	2%	4.5168	0.036	306184	0.19	11.519	0.19
139_none_sur	6000	5815	2%	20.311	0.05	1359769	0.15	11.513	0.16
281_none_sur	5745	6000	5%	52.8	0.04	3489720	0.12	11.504	0.13
194_none_sur	5614	6000	7%	93.233	0.0028	6023506	0.17	11.508	0.17

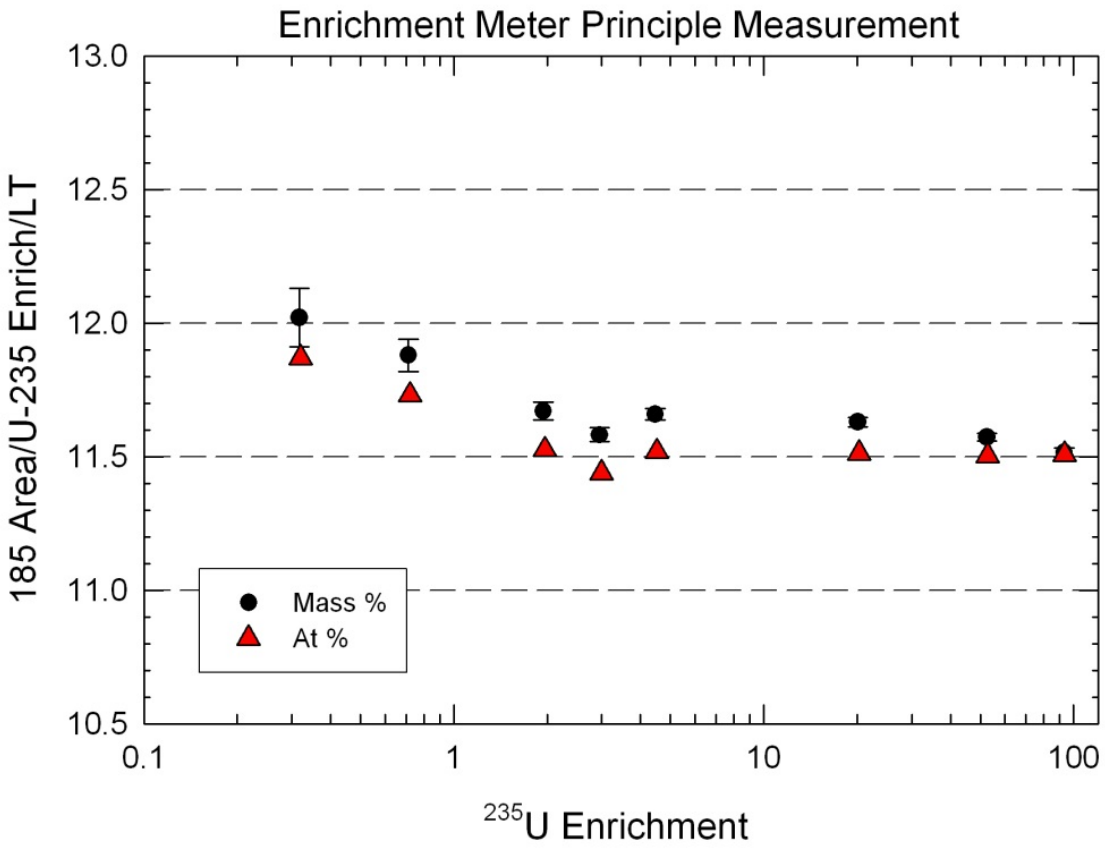

Fig. 26. Enrichment Meter Principle Data from second dataset.

3.3.1 FRAM Peak Fit at 185.7 keV

In Fig. 27 we show how well FRAM fit the 185.7 keV EMP peak for the 20.11 % enriched ^{235}U standard in the first dataset.

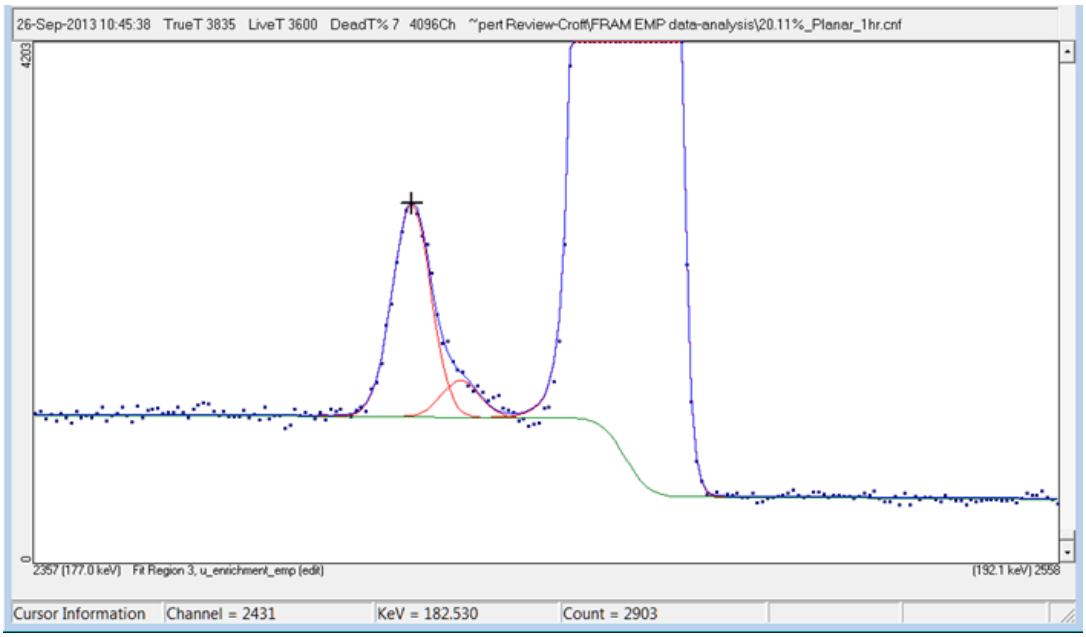

Fig. 27. 185 keV peak fitting for the 20.11 % enriched ^{235}U standard from the first dataset.

This spectrum exhibits very good peak resolution and very little low-energy peak tailing. The 182.5-keV ^{235}U peak and the weaker 183.3-keV ^{231}Th peaks are resolved from the much larger 185.7 EMP peak. A good quality detector such as this enhances the ability of FRAM to fit the peaks well.

3.3.2 Interferences with the 185.7-keV ^{235}U Peak

The parameter file u_enrichment_emp used in these initial analyses did not include any direct interference peaks at 185.7 keV. Mattusek (Matussek 85) discusses potential interferences in great detail. Parker (Parker 88) also discusses interferences in the context of his high-precision EMP measurements and concludes that the interferences are negligible for enrichments > 5%. His conclusion is reproduced below.

> A final correction, which should be considered here, is that for the interference to the 185.7 keV gamma rays of ^{235}U by low-intensity gamma rays of ~186 keV energy from ^{238}U daughter products. For the methods used to extract spectral peak areas in the work reported here, the effective (emission) rate of the interfering gamma rays is 0.32/s-g ^{238}U, while the emission rate for ^{235}U is 4.60×10^{4}/s-g ^{235}U. The factor by which the apparent measured enrichments should then be multiplied to compensate for this interference is:
>
> $$CF = 1 - [\text{at\% U238}/\text{at\% U235}] \times [0.32/4.6\ 10^{4}]$$
>
> For enrichments of 0.7, 2, 3, and 5% the CF values are very nearly 0.9990, 0.9997, 0.9998, and 0.9999, respectively. For enrichments above 5% the influence of this interference is clearly negligible.

Note that Parker's correction is applied after-the-fact. With his ROI summation peak area extraction this was the only way he could do it. One might be tempted to suggest that with FRAM (or any peak fitting code) one could fit the interference peaks directly. However, reliable fitting of directly overlapping free peaks with a 1000:1 or greater peak area ratio cannot be done.

With FRAM one could also consider fixing the ^{238}U daughter interference that Parker considers (186.15 keV from ^{234}Pa) to the 258.26-keV ^{234m}Pa daughter peak that is fairly easy to measure for low enrichments where the correction would be made. But this would require an extrapolation of the relative efficiency curve beyond its range of definition. This could be explored in the future but is beyond the scope of this current effort. Keep in mind that this correction is <0.1% relative for enrichments above natural U and vanishes for practical consideration for enrichments >5 %.

3.4 POTENTIAL DIFFICULTIES WITH FRAM EMP ANALYSIS

The above data are very preliminary and the lack of agreement at the very lowest enrichments shows that more work is needed. One measurement regime that has not been investigated is the measurement with short data acquisition times (10 min or less). Short measurement times are very important for practical use in the field. The potential issue with FRAM would be lack of statistics in the 143, 163, and 205 keV peaks that are used (along with 185 keV) to establish the response function fitting constants. This is likely to be a problem for low enrichments. Fixing the values of the response function constants is an established way to circumvent this issue. This might reduce the accuracy of the peak area extraction and compromise the attainable accuracy.

3.5 PREPARATIONS FOR ADDITIONAL MEASUREMENTS

The following suggestions draw upon material previously discussed in this report and thus will not be greatly elaborated.

3.5.1 Establish an Up looking, Reproducible Measurement Geometry

A solid, reproducible measurement geometry (as pictured in Fig. 1 and Fig. 8) is the first requirement before any additional EMP measurements are undertaken.

3.5.2 Provide Adequate, Graded-Z Shielding and Filtering

The geometry established in 3.5.1 must be provided with sufficient shielding (Table 1) to accommodate all measurement scenarios. This shielding must be completely covered on the detector side with a graded-Z filter to prevent any fluorescent x-rays from the shield and the filter from reaching the detector. Sometimes it is easier to put a graded-Z filter around the detector (Fig. 7). These requirements have been discussed in Section 1.4.

Filter the spectrum entering the detector with a graded-Z filter placed in front of the end cap (Table 2). Filter thickness is chosen based on the purpose of the measurements. Every detector should be provided with a range of filters applicable to all measurement scenarios.

3.5.3 Plan for Repeated Measurements

By performing repeated measurements, each measurement with the target count time, one will obtain data that can be analyzed to give the observed statistical repeatability in addition to providing accuracy results for the parameters of interest.

Set up the file names for the stored spectra to be compatible with FRAM's automated analysis of multiple spectra capability (see Section 3.4.1 in the FRAM Version 5 User Manual).

When analyzing EMP data with the u_enrichment_emp, or successor, parameter file, be sure to check the Save Results box in the FRAM Measure | Analyze window

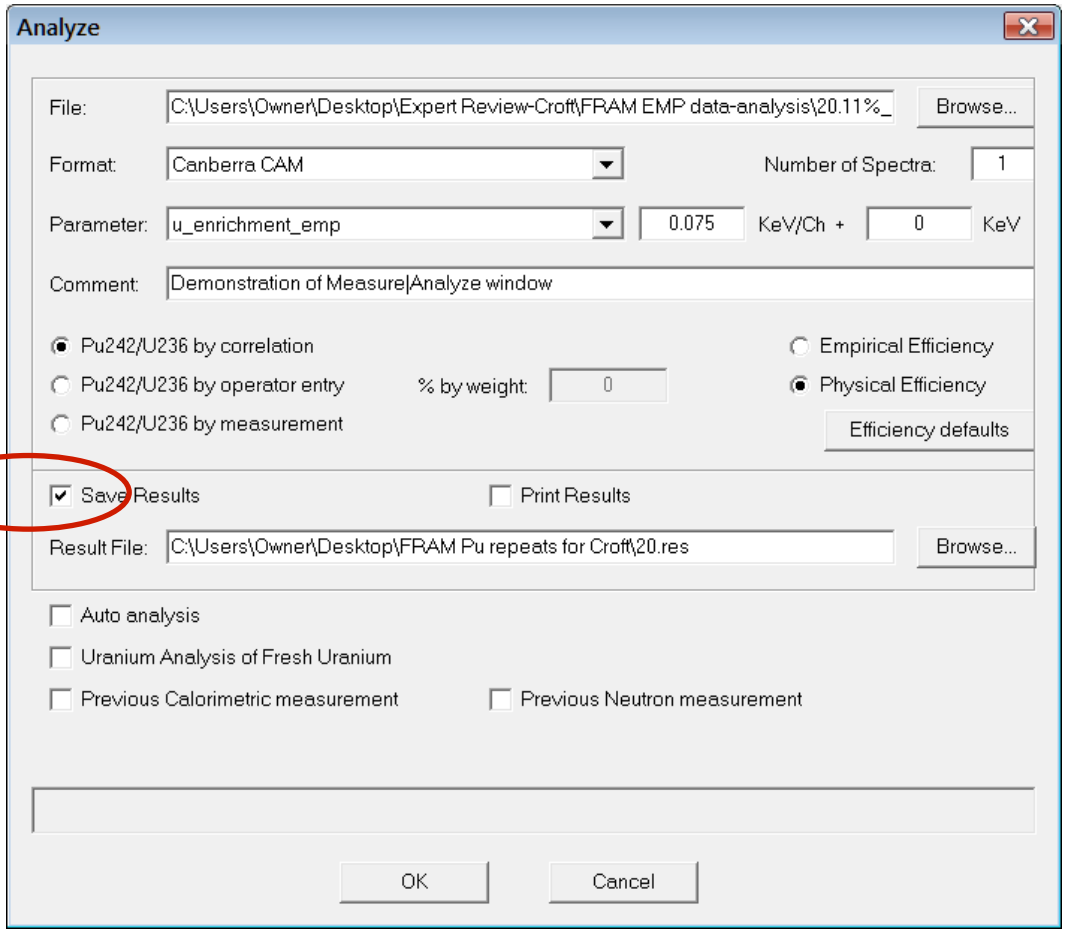

Fig. 28. The FRAM Measure|Analyze window showing how to save a FRAM results file.

FRAM results for peak areas and uncertainties are buried in the middle of a FRAM results file which is a text file containing all information about the analysis. It is unwieldy to extract these values manually when analyzing multiple repeated runs. It is straightforward for a programmer to write some rather simple software to extract these results to an Excel spreadsheet from a series of repeated analyses. This is recommended for any extensive data analysis campaigns of FRAM analysis of repeated measurements.

3.6 WHAT IS REQUIRED TO MAKE A STAND-ALONE, FRAM-ENGINE BASED, EMP CODE?

3.6.1 FRAM Command Line Mode

The FRAM Command Line Operation is described in the FRAM Version 5 User Manual in Appendix E, starting at p. 147.

The Command-Line Operation mode is meant for users who want to incorporate FRAM into their own applications. This mode allows the user to make a very simple interface to the FRAM analysis engine. When FRAM is run with a string argument it will switch to the command-line operation mode. This mode of operation will execute the commands supplied in the argument and when the analysis is completed will automatically terminate. It will not open a window.

Any stand-alone FRAM EMP code would likely start here.

3.6.2 Additional Software to be Developed

Some of the additional software that would be needed includes

- Establish Setup and Input routines
- Extract peak areas and uncertainties from FRAM results
- Establish Calibration routines
- Establish Enrichment calculation routines
- Establish routines to Output results

4. FRAM URANIUM MEASUREMENTS IN THE FIELD

4.1 FRAM COMPLETE ISOTOPIC ANALYSIS COMPARISON WITH EMP MEASUREMENTS

Field measurements with FRAM were carried out at a uranium fuel fabrication facility in Europe (Sampson 99). These measurements were undertaken to test FRAM as a method to supplement the calibration-intensive, classical ^{235}U EMP method and also to prove its capability on thick-walled UF_6 cylinders where other arbitrary sample/geometry methods using the 100 keV region fail.
FRAM used a nominal 25 % relative efficiency coaxial detector to make complete U isotopic measurements in the 120–1010 keV region. EURATOM inspectors used a very small, coaxial HPGe detector to make classical ^{235}U EMP method measurements alongside the FRAM measurements. The dual sets of measurements allowed the following useful conclusions to be drawn from the 1999 exercise.

1. 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 ($\lesssim$6) were required.
2. After calibration, the classical enrichment method can complete verification measurements on the samples appropriate to the calibration more quickly (100 sec count time) than FRAM (10–20 min count time). This method is therefore more appropriate when a large number of the same type of items have to be measured.

3. 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.
4. 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 making it easier to set up the measurement geometry.
5. FRAM was able to demonstrate measurements on additional container types (UO_2 powder, scrap container) that could not be measured by the classical enrichment method using the calibration for 30B cylinders.
6. FRAM had no difficulty with measurements on the thick-walled (13 mm) UF_6 cylinders. Other arbitrary sample measurement techniques that use low-energy gamma rays cannot measure these UF_6 cylinders.
7. FRAM measurement precision (typically 3 %–8 % rsd), 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 for the need.
8. The current implementation of FRAM for uranium measurements demonstrates some sensitivity to the sample-detector distance. Pending a new algorithm a simple change of one parameter in the parameter file could remove this bias for UF_6 cylinder measurements. *Note--2015: The coincidence summing correction alluded to here is now an integral part of FRAM, see Section 2.1.2 of this document.*
9. The analysis method used by FRAM currently requires secular equilibrium between ^{238}U and its ^{234m}Pa daughters. This process may be further complicated by processing steps that only partially remove daughter products and by daughter product deposits on cylinder walls.

4.2 SHIELDING FOR FIELD MEASUREMENTS WITH PORTABLE EQUIPMENT

The physical requirements for moving shielded portable detectors to perform FRAM isotopic analysis measurements in a field of UF_6 cylinders (4.1 above) are formidable. Practical Pb shield thickness may be only 0.75–1.0 in. This thickness range of Pb will pass 10-20 % of incident 1001 keV gamma rays, increasing the likelihood of a biased measurement. Be very aware of this potential problem and make judicious use of additional portable shields, shadow shields, and the like. The lack of geometric positioning requirements also makes it possible to reduce background by detector positioning.
EMP techniques that rely on the 185.7 keV gamma ray have much less severe shielding requirements. Complete isotopic distribution measurement techniques using the 100 keV region (including FRAM) also have less severe shielding requirements (see Section 1.4.1) but generally cannot measure UF_6 cylinders (wall thickness 13–16 mm steel or equivalent) so the advantage is moot.

5. UNCERTAINTY QUANTIFICATION IN FRAM

5.1 COUNTING STATISTICS UNCERTAINTIES

A short FRAM output for uranium is shown in Fig. 29 with the measurement uncertainties marked.
It is important to understand what uncertainty component(s) are captured in the listed uncertainty. The uncertainty labeled "sigma" is a 1-σ absolute uncertainty propagated from counting statistics only! The "%RSD" line is the 1-σ relative standard deviation expressed as a %, calculated as %RSD = [sigma/mass% x 100].

That the uncertainties shown above include only counting statistics was a deliberate choice made during the development of FRAM. This gives the user an opportunity to verify the correctness of the counting statistics uncertainty prediction in FRAM by carrying out repeated measurements (without sample replacement) and comparing the standard deviation observed from the repeated measurements with the "sigma" estimated by FRAM.

```
*******************************************************************************
 PC FRAM (5.2)     Isotopic Analysis     04-May-2015 09:09:59
 (Fixed energy Response function Analysis with Multiple efficiencies)
 Operator ID:

 spectrum source:  C:\FRAM_Archival_data\URAN125.DAT\NBS44612.S02
 spectrum date:  21-May-1996 03:02:22
 live time:  3145 s
 true time:  3600 s
 num channels:  8192

 parameter set:  ULEU_Cx_120-1010 (2013.07.25 11:16)
      U235 < 40% or high Thorium BG, 0.125 keV/ch, Coax Detector
     Physical Efficiency,  Gain 0.125 keV/ch, Offset 0 keV
 comment:
*******************************************************************************
*******************************************************************************
 at   185.715 keV, high E tail %(meas-fit)/area =   0.455 +-   0.012 [? <  0.400]

                              (By Corr)

              U234       U235       U236       U238
 mass%       0.0368     4.4749     0.0266    95.4618
 sigma       0.0029     0.0668     0.0004     0.0670
 %RSD        7.83%      1.49%      1.46%      0.07%

 %TotPwr    85.37       3.48       0.60      10.53

 Specific Power (W/gU):     (     0.0772 +/-     0.0052)e-006   (  6.68%)

 Relative mass (Th228 / U):     8.921e-014   (  4.03%)
*******************************************************************************
```

Fig. 29 A FRAM Short output for a uranium item. The measurement uncertainties are boxed.

Note that the sigma determined from the observed standard deviation from repeated measurements is also a random variable (statistic) with its own sampling uncertainty. The relative standard deviation (rsd) of the observed sigma (s) is given by rsd(s) = [1/sqrt(2(n-1))] where n is the number of repeat measurements. This is obviously a slowly-varying function of n with the rsd for 50 repeats being 0.101 while the rsd for 15 repeats is 0.189. This uncertainty should be kept in mind when comparing the sigma estimated in FRAM with the sigma observed from repeated measurements.

The estimate of sigma for an individual FRAM measurement is challenging effort in error propagation. There are approximations made and correlations are neglected in the code in order to make the calculation tractable. Nevertheless, we will see later that the FRAM prediction of counting statistics-related uncertainty is fairly good.

5.2 SYSTEMATIC UNCERTAINTIES IN FRAM

Some of the general areas that can give rise to systematic uncertainties are:

- Sample composition characteristics
- Knowledge of photon emission probabilities
- Peak area determination process
- Unrecognized peak interferences
- Coincidence summing for uranium (see sec. 2.1.2)

While an extensive discussion of sources of systematic uncertainties in FRAM is beyond the scope of the present report, the reader is referred to (Sampson 03) and (Burr 05) for more information and guidance on this important topic.

In addition to the random uncertainty from counting statistics that is reported for every FRAM measurement, FRAM also has a provision for incorporating an additional systematic uncertainty component and reporting a combined uncertainty for each individual measurement.

The systematic uncertainty components are specified in the Application Constants section of FRAM Version 5 User Manual (Vo 11). These application constants are listed in Table 5.

Table 5. Application Constants Governing Uranium Systematic Uncertainties.

Command	Type	Default	Description
syst_error_u234	Real	0	Systematic uncertainty of U234. A positive number represents the percent uncertainty. A negative number would invoke the built-in bias. The absolute value of the negative number is the multiplication factor of the built-in bias.
syst_error_u235	Real	0	Systematic uncertainty of U235. See description for U234 above.
syst_error_u236	Real	0	Systematic uncertainty of U236. See description for U234 above.
syst_error_u238	Real	0	Systematic uncertainty of U238. See description for U234 above.

The built-in parameter sets in version 5 of FRAM come with the systematic uncertainty application constants set to a value of "-1". This invokes a built-in systematic uncertainty (or bias as it is termed in the application constant description). The documentation of this built-in bias may be found in (Vo 11a). If a non-zero systematic uncertainty has been set up, the results showing the combined uncertainty may be viewed in FRAM's Med Result output appended to the normal Short Result. This is shown below in Fig. 30.

5.3 QUANTIFICATION OF BIAS AND STATISTICAL UNCERTAINTIES IN FRAM

The characterization of FRAM's bias is obtained by analyzing repeated measurements and looking at the average over all measurements of the mass% result in comparison with the known or certified values. Certified Reference Materials are the first choice for these measurements and you have access to two sets spanning the enrichment range. Other materials that are not CRMs may also be used as long as they have a traceable mass spec. It might be useful for US applications to look at additional highly-enriched items that have ^{235}U percentages characteristic of national defense materials.

```
*****************************************************************************
 PC FRAM (5.2)     Isotopic Analysis     05-May-2015 09:48:08
 (Fixed energy Response function Analysis with Multiple efficiencies)
 Operator ID:

 spectrum source:  C:\FRAM_Archival_data\URAN125.DAT\NBS44612.S02
 spectrum date:  21-May-1996 03:02:22
 live time:  3145 s
 true time:  3600 s
 num channels:  8192

 parameter set:  ULEU_Cx_120-1010 (2013.07.25 11:16)
      U235 < 40% or high Thorium BG, 0.125 keV/ch, Coax Detector
     Physical Efficiency,  Gain 0.125 keV/ch, Offset 0 keV
 comment:
*****************************************************************************
*****************************************************************************
 at   185.715 keV, high E tail %(meas-fit)/area =   0.455 +-   0.012 [? <  0.400]

                              (By Corr)

              U234      U235      U236      U238
 mass%       0.0368    4.4749    0.0266   95.4618
 sigma       0.0029    0.0668    0.0004    0.0670
 %RSD        7.83%     1.49%     1.46%     0.07%

 %TotPwr    85.37      3.48      0.60     10.53

 Specific Power (W/gU):    (     0.0772 +/-     0.0052)e-006  (  6.68%)

 Relative mass (Th228 / U):    8.921e-014  (  4.03%)
*****************************************************************************
*****************************************************************************
 Isotopic Results with Systematic errors included:
                              (By Corr)

              U234      U235      U236      U238
 mass%       0.0368    4.4749    0.0266   95.4618
 sigma       0.0031    0.0681    0.0027    0.0684
 %RSD        8.39%     1.52%    10.11%     0.07%

 %TotPwr    85.37      3.48      0.60     10.53

 Specific Power (W/gU):    (     0.0772 +/-     0.0055)e-006  (  7.17%)

 Relative mass (Th228 / U):    8.921e-014  (  4.03%)
*****************************************************************************
```

Fig. 30. The Short output of Fig. 29 with systematic uncertainties appended from a Med Result.

These same data sets can be analyzed to obtain the observed standard deviation of the repeated measurements. This value will be dependent upon the counting conditions and you should characterize it for all appropriate measurement conditions of interest. The other vital comparison that is carried out with the same data is to compare the observed standard deviation from repeated measurements with the statistical uncertainty (sigma, % rsd) (sec. 5.1) predicted by FRAM. This informs on how well FRAM predicts the statistical measurement uncertainty which is vital when you have only a single measurement result to rely on.

The comparison of observed precision from repeats to the predicted FRAM precision can be performed with any item. It does not have to be a CRM and it does not even have to have any mass spec values. All that is being looked at is the repeatability and knowledge of the actual mass% values for the item is not required.

Analysis of this type of data is enhanced in version 5 of FRAM by the fact that the principal isotopic results of every analysis are automatically written to an Excel spreadsheet. For uranium measurements this spreadsheet is `Uranium.csv` which is found in the `C:\FRAMdata` folder. The stored variables are listed in Table 6.

Table 6. Results Stored in Uranium.csv Excel File for Every Measurement.

File name	U236
Acq. Date	U238
Anal. Date	SpecPow
Parameter	U234sigma
Eff. Model	U235sigma
Gain	U236sigma
Offset	U238sigma
U234	SpecPow sigma
U235	

The appropriate results from `Uranium.csv` may be copied to another spreadsheet where the appropriate averages and standard deviations can be calculated. An example of this is shown in Table 7.

The FRAM statistical precision prediction is compared against the observed precision from repeats by comparing the Average, Uxxx sigma cell value (solid outline) with the Std Dev, Uxxx cell value (dashed outline).

This comparison for uranium has been carried out for FRAM version 4 using 33 sets of repeated measurements, a total of 415 measurements, on items spanning the enrichment range from 0.3 % to 93 % (Sampson 03, 03a). This comparison is reproduced in Table 8.

Table 7. EXCEL Spreadsheet Calculating FRAM Performance Parameters—Basic Data is Taken From Uranium.csv.

NBS44612.S01-S14								
Run	U234	U234 sigma	U235	U235 sigma	U236	U236 sigma	U238	U238 sigma
1	0.0378	0.0028	4.3298	0.0665	0.0258	0.0004	95.6066	0.0667
2	0.0368	0.0029	4.4749	0.0668	0.0266	0.0004	95.4618	0.0670
3	0.0378	0.0029	4.4282	0.0685	0.0263	0.0004	95.5077	0.0687
4	0.0305	0.0027	4.1772	0.0638	0.0249	0.0004	95.7673	0.0640
5	0.0398	0.0028	4.4175	0.0657	0.0263	0.0004	95.5164	0.0659
6	0.037	0.0028	4.4177	0.0683	0.0263	0.0004	95.5190	0.0685
7	0.0329	0.0027	4.3064	0.0677	0.0256	0.0004	95.6350	0.0678
8	0.0359	0.0028	4.4175	0.0635	0.0263	0.0004	95.5203	0.0637
9	0.0367	0.0029	4.3267	0.0710	0.0258	0.0004	95.6108	0.0712
10	0.0388	0.0028	4.4300	0.0654	0.0263	0.0004	95.5049	0.0656
11	0.0321	0.0027	4.4428	0.0618	0.0264	0.0004	95.4988	0.062
12	0.0349	0.0028	4.3471	0.0699	0.0259	0.0004	95.5922	0.0701
13	0.0382	0.0028	4.4644	0.0634	0.0265	0.0004	95.4709	0.0636
14	0.0394	0.0028	4.5309	0.0591	0.0269	0.0003	95.4028	0.0593
Average	0.0363	0.0028	4.3937	0.0658	0.0261	0.00039	95.544	0.0660
Std Dev	0.00280		0.0887		0.000499		0.09103	
% RSD	7.71		2.02		1.91		0.095	
Accepted	0.0359		4.4623		0.0068		95.495	
Average/Accepted	1.0119		0.9846		3.8435		1.0005	

Table 8. Comparison of Predicted and Observed Statistical Uncertainties for Uranium Analysis using FRAM version 4.

Detector	Region (keV)	No. Data Sets	No. Meas.	Average Ratio: Predicted/Observed		
				^{234}U	^{235}U	^{238}U
Coaxial	120–1001	33	415	1.35	0.84	0.80

We determine the average FRAM bias for a particular parameter file by analyzing the results from repeated measurements (as in Table 7) for a large number of items and then averaging the Average/Accepted ratio over all the items. This "average of the averages" is a measure of the FRAM bias for the parameter file in question as seen in Table 9.

Table 9. Uranium Bias for Version 4 of FRAM.

Filename	Accepted Wt % ^{235}U	Measured/Accepted ^{234}U	^{235}U	^{238}U
NBL93_03	93.1703	0.99495	0.99953	1.00875
NBL93_00	93.1703	1.00583	0.99777	1.03636
C2080001	93.1700		0.99901	
U91C2G20	91.3356	0.98896	0.99890	1.01493
U91C2T30	91.3356	0.97910	0.99823	1.02431
UISO91_N	91.3356	1.01151	0.99819	1.02087
U66C2G20	66.0405	1.00983	0.99437	1.01106
NBL53_10	52.4880	0.99612	1.00049	0.99948
NBL53_03	52.4880	1.01046	1.00351	0.99598
U52C2G20	52.1174	1.02696	0.99345	1.00704
U38C2G15	37.5518	1.01110	0.99733	1.00157
UISO_27	26.7519	1.02056	0.99782	1.00074
UISO27PS	26.7519	1.01997	0.99494	1.00180
UISO27	26.7519	0.99837	0.97735	1.00833
NBL20_01	20.1070	1.00905	0.99662	1.00084
NBL20_05	20.1070	0.99096	1.00318	0.99921
U17C2T10	17.2386	1.02646	0.99036	1.00197
UISO_13	12.9543	1.04940	1.02127	0.99678
UISO13_P	12.9543	1.04751	1.01551	0.99764
UISO13	12.9543	1.02076	1.00058	0.99989
UISO1212	11.7974	1.00117	1.00961	0.99871
A1324PS	10.0863	1.00801	0.99039	1.00107
NBS44612	4.4623	0.98043	0.99669	1.00016
NBS446PS	4.4623	1.00557	0.98285	1.00080
NBS29512	2.9491	0.96287	0.98473	1.00047
NBS295PS	2.9491	0.96205	0.98514	1.00046
NBS19412	1.9421	1.03156	1.00125	0.99997
NBS194PS	1.9421	0.97662	0.98849	1.00023
A11127PS	0.7167	1.29268	1.01235	0.99990
NBS07112	0.7119	1.07897	0.99649	1.00002
NBS071PS	0.7119	1.02303	0.99389	1.00004
NBS03112	0.3166	1.43220	0.99606	1.00000
NBS031PS	0.3166	1.03553	1.00190	0.99999
Average		1.0054 **	0.9975	1.0040
Std. Dev.		0.0221	0.0089	0.0088
% RSD		2.20	0.90	0.88

Notes:

** U-234 Average and Std. Dev. exclude enrichments with $^{235}U < 1.9$ %. This enrichment value is at the sensitivity limit of FRAM for ^{234}U.

Some additional information regarding Table 9:

- In version 4 of FRAM a single coaxial detector parameter file was used for all items and all enrichments.
- The individual entry averages in Table 9 do not necessarily arise from the same number of assays. In general the number of repeated measurements per sample ranged from 10 to 15. From Table 8 we see that the average number of repeats over all items was 12.6 (415 measurements over 33 items). The "Average of the Averages" in Table 9 is un-weighted and does not account for the different number of measurements per entry.
- The Std. Dev. (% RSD) (boxed in Table 9) is thought of as an item-dependent bias and would be the number that would be cited for version 4 when asked the question, "What is the typical bias of a FRAM uranium measurement?"

The type of data represented in Table 9 has been used for plutonium measurements to carry out a more sophisticated ANOVA statistical analysis, also from version 4 of FRAM (Burr 05). This analysis was carried out for three different plutonium parameter files representing different detector types and different analysis energy ranges. The details of this analysis are beyond the scope of this report but it should be noted that the item-dependent bias that Burr identified for plutonium was around 1 %, the same as that identified above for uranium. On this basis, for field quality measurements, there is perhaps little to be gained for counting longer than is needed to achieve about 1 % rsd precision.

5.4 GAPS IN PERFORMANCE VERIFICATION FOR VERSION 5 OF FRAM

5.4.1 Uranium Coaxial Detector Performance Verification, 120–1010 keV Range

The tabulation of bias and uncertainty verification in the previous section was rigorous for uranium analysis with a coaxial detector in the 120–1010 keV range for version 4 of FRAM. While one expects that these values will generally apply to version 5, there is little documented performance data for uranium for version 5 of the software. For version 5 the coaxial detector analysis in the 120–1010 keV range is split into two standard, built-in parameter files. These parameter files are new for version 5; they incorporate changes and modifications over the single coaxial detector parameter file used in v4. The version 5 coaxial detector LEU parameter set, ULEU_Cx_120-1010 is designed to measure enrichments up to 40%. For high enrichments (see sec 2.1.3) the parameter file UHEU_Cx_120-1010, whose recommended enrichment range is enrichments > 10%, is used.

The performance of these two built-in FRAM version 5 parameter files should be verified against the same coaxial detector spectra that are tabulated in Table 9. This verification would produce both bias results and produce comparisons of the observed RSD from the repeated measurements to the RSD predicted by FRAM.

All the spectral data presented in Table 9 still exist in the author's (Sampson) personal files. These data are available for reanalysis using version 5 of FRAM. This reanalysis is, however, beyond the scope of this report.

5.4.2 FRAM v5 Uranium Performance Verification in the 60–250 keV Range

While version 5 performance data for uranium in the 60–250 keV range exists in many places, there has been no published summary like that in Table 9 for version 4 coaxial detector analysis. Also, since FRAM analysis in the 100 keV region is newer, more stringent regulation governing the handling and use

of SNM have essentially made it impossible, e.g. at LANL, to get ready access to the wide range of samples represented in Table 9.

FRAM version 5 performance for uranium in the 60–250 keV range needs to be tabulated in the format of Table 9 using existing data. There is a significant amount of uranium 100 keV region data in the author's personal files. The eight reference materials form the majority of the data but there are other items that are not represented in Table 9. Unfortunately the author does not currently have any 100 keV region spectra from the intermediate enrichment standards (Parker 88) that are represented in Table 9. Analysis of the existing data with version 5 would be straightforward but is beyond the scope of this report.

5.4.3 ORNL Role in Closing Gaps in FRAM v5 Uranium Performance Verification

ORNL can provide high-quality spectra using existing BEGe detectors that could go a long way toward closing the gaps in the documentation of FRAM 100 keV region performance. The data would be repeat measurements on the eight U_3O_8 uranium reference materials, DU to HEU, that ORNL possesses. The importance of perfecting and optimizing data acquisition procedures has been emphasized in this report. Uranium data acquisition should address the following good practices:

- Use a high quality planar or semi-planar (BEGe for example) HPGe detector.
- Use graded-Z shielding and filtering to suppress Pb x-rays from the shielding (sec. 1.4).
- Use wide open "collimation" and vary count rates by sample-to-detector distance.
- Optimize the filter thickness to preserve 100 keV region intensity as much as possible while suppressing any peaks below 80 keV.
- Use a rise time of 4 μs for acquisition with a digital spectrometer. This is a good compromise to minimize random summing while still maintaining good resolution. Verify that this is a good compromise from the count rate / throughput / resolution / time constant parameter study (sec. 1.3.3) performed on the particular detector.
- Concentrate data acquisition on the 100 keV region. There is no need to use more than 8K ch at 0.075 keV/ch. It is OK to acquire data in 16K channels, but recognize that analysis above the 100 keV region may not be optimum because of different filter optimization.
  - Design the measurement times to cover the range of practical data acquisition scenarios. Production facility measurement times seldom exceed 1 h. Measurement times for 100 keV region analysis are often ~ 30 min and could be shorter in an inspection scenario. At a minimum carry out repeated measurements for several times in this range in order to characterize the precision for practical count times.
  - Make sure that the individual measurements are preserved. Incremental summed spectra are not useful in this regard.
  - Try to collect at least 15-20 repeats on each item. More repeats are even better.

Try to obtain additional items with enrichments that fill the gaps in the existing standards. It is entirely good enough if the additional items have just a single traceable mass spectrometry measurement to characterize their isotopic composition. Items without documented isotopic knowledge may also be used to characterize precision from repeated measurements but, of course, will not provide any bias information.

6. REFERENCES

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APPENDIX A. Acquisition of Nuclear Data Quality Spectra

APPENDIX A. ACQUISITION OF NUCLEAR DATA QUALITY SPECTRA

Management of data acquisition conditions, detector selection, use of shielding, and graded filtering have all been discussed at length earlier in this report. The specifications for high-quality measurements, such as those that might be used for the extraction of fundamental nuclear parameters, are highlighted below, based on these earlier discussions. The acquisition of nuclear data quality spectra requires great attention to detail in order that the measurements can be fully described, appropriately analyzed and reliable results reported. The report by Jack Parker (Parker 88) is probably the best single reference available detailing the care needed for high-precision, high-accuracy measurements.

As mentioned previously, FRAM makes use of adjusted parameters - that is adjusted from the values in nationally recognized evaluated nuclear data files - in order to obtain the best agreement with measurements on Certified Reference Materials (of uranium, for the purposes of this report). The fact that these adjustments are made does not necessarily mean that the evaluated nuclear data files are in error (although that may be the case in some instances). It likely means, for the most part, that we are correcting for imperfections in the FRAM spectral analysis in addition to correcting for uncertainties in our knowledge of the gamma-ray branching intensities. One type of bias in a FRAM (or any other isotopic analysis software) analysis arises from net peak area uncertainties caused by imperfect background (continuum) subtraction. A variety of empirical methods are used to represent the background based on experience and implemented using application specific assumptions. There is no exact way to describe the shape of the background beneath the extended (in energy) peak groupings. A second peak area uncertainty comes from imperfections in the response function fitting of closely-lying, overlapping peaks. A third uncertainty contribution arises from biases and random errors in the branching intensities in the nuclear data files. Other analysis uncertainties can arise from imperfections in the modeling of the relative efficiency fitting function (the physical model description is built on a simple layered picture) as well as the imperfect modeling of the gamma-ray peak shapes themselves.

Acquisition of nuclear data quality spectra and the analysis of these spectra by FRAM contribute to the better understanding of these uncertainty contributions and their interaction with the fundamental nuclear parameters used by FRAM. The varied, extensive, and detailed outputs available from FRAM enable careful examination of the consistency of branching intensities from different gamma rays which is a strong indicator that the relative values of the nuclear data are good. An important point about using CRM's is that the mixture of nuclides serve as internal references so that, for example, ^{235}U lines can compared to ^{238}U lines directly on a per gram basis. And this is ultimately the sort of relationships that are need for isotopic analysis – although the nuclear data are not entered into the code this way. Turning this argument around, it is important to realize that basic nuclear data is seldom measured or evaluated with specific applications in mind. Isotopic analysis codes applied to safeguards, non-proliferation, arms control etc. use nuclear data in a specific way and the opportunity exists to revisit nuclear data generation to improve practical performance. The guidance below is provided with this goal in mind.

Samples: The measurements will use the NBS CRM 969 and the NBL CRM 146 isotopic standard sets. These materials are very-well characterized physically, chemically, and isotopically. The containers are precisely manufactured and measured to make container attenuation corrections straightforward and accurate.

Detector: The type of detector to be used is best chosen to match the desired energy range of the fundamental parameters to be measured. Semi-planar detectors have been widely used at ORNL for uranium measurements and would be the detector of choice for measurements in the energy range below ~300 keV. However, if measurements of fundamental parameters in the 700- to 1100 keV range are desired, they would be best made using a fairly large volume coaxial detector (perhaps in the 50 % relative efficiency range or higher). The increased efficiency for higher energy gamma rays is definitely needed for fundamental parameter measurements.

Measurement Geometry, Filtering: No collimation, with the measured item on the detector axis, is desirable to collect the maximum signal (and keep the description of the efficiency curve simple). The flat bottom of the containers should face the detector. The use of low-energy filters over the detector face is an open question. However, for measurements concentrating on the energy range above 120 keV light filtering is recommended to remove the possibility of any gamma rays < 80 keV causing random summing. Any filters should be of the minimum thickness required and should be accurately fabricated and measured to facilitate transmission corrections. The chemical composition of the filters should be well known, again, to facilitate the accurate calculation of any necessary transmission corrections.

Measurement Distance: It is a good idea to choose a measurement distance that will minimize coincidence summing effects. From Fig. 11 it would appear that ~ 15 cm is a good choice for the closest measurements. Of course, a much larger diameter detector might require a larger distance since this is a solid angle effect.

Measurement Distance, Fixtures: Any precision measurement campaign where position reproducibility is required must use well-designed fixtures to position the measured items in an exactly reproducible position relative to the detector crystal. This can be more challenging for side-looking geometries.

Shaping Time: Use a rise time (digital spectroscopy system) of 4 μs. Using this fairly short rise time is a compromise, but is specified to reduce random summing, over ultimate resolution.

Count Rate: The parameters of count rate, count time, measurement distance, and dead time are all interdependent and a balance must be achieved. The requirement to use a minimum measurement distance of about 15 cm is probably the controlling factor. Keeping the maximum count rate under 10 kcps is probably a good idea. Counting time is dependent upon the peak area precision desired. Stability and control of the ambient background become more important when the run times become long.

Other Amplifier/ADC Settings: Use pileup rejection and digital gain and zero stabilization. A conversion gain of 0.075 keV/ch is a good (and conventional) choice for data concentrating on energies < 300 keV. However, for data at higher energies (> 700 keV) it is much better to use a coarser conversion gain. A value of 0.1 to 0.125 keV/ch makes the data much easier to handle, plot, fit, and visualize. Don't try to extract too much information from a single spectrum with a large number of channels. Make the size (number of channels) commensurate with the desired energy range of the data and conversion gain so that the peaks are not too broad or too narrow. See Section 2.1.5 for information on peak widths desirable for FRAM analysis.

Shielding: Much has been written in this report regarding the importance of adequate, graded-Z shielding, appropriate for the energy range of the data to be analyzed. All of these suggestions/requirements are also applicable to obtaining nuclear data quality spectra. The filtering and graded-Z shielding should completely suppress x-rays from the shielding material in order to avoid undesirable artifacts.

GAMMA-RAY SPECTROSCOPY

RESOURCE INFORMATION

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MANUFACTURER'S WEB SITES

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