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Title: **PEAK FITTING APPLIED TO LOW-RESOLUTION
ENRICHMENT MEASUREMENTS**

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PEAK FITTING APPLIED TO LOW-RESOLUTION ENRICHMENT MEASUREMENTS

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Abstract

Materials accounting at bulk processing facilities that handle low enriched uranium consists primarily of weight and uranium enrichment measurements. Most low enriched uranium processing facilities draw separate materials balances for each enrichment handled at the facility. The enrichment measurement determines the isotopic abundance of the ^{235}U , thereby determining the proper strata for the item, while the weight measurement generates the primary accounting value for the item.

Enrichment measurements using the passive gamma radiation from uranium were developed for use in U.S. facilities a few decades ago.¹ In the U.S., the use of low-resolution detectors was favored because they cost less, are lighter and more robust, and don't require the use of liquid nitrogen. When these techniques were exported to Europe, however, difficulties were encountered. Two of the possible root causes were discovered to be inaccurate knowledge of the container wall thickness and higher levels of minor isotopes of uranium introduced by the use of reactor returns in the enrichment plants. The minor isotopes cause an increase in the Compton continuum under the 185.7 keV assay peak and the observance of interfering 238.6 keV gamma rays. The solution selected to address these problems was to rely on the slower, more costly, high-resolution gamma ray detectors when the low-resolution method failed.

Recently, these gamma ray based enrichment measurement techniques have been applied to Russian origin material.² The presence of interfering gamma radiation from minor isotopes was confirmed. However, with the advent of fast portable computers, it is now possible to apply more sophisticated analysis techniques to the low-resolution data in the field. Explicit corrections for Compton background, gamma rays from ^{236}U daughters, and the attenuation caused by thick containers can be part of the least squares fitting routine. Preliminary results from field measurements in Kazakhstan will be discussed.

INTRODUCTION

Materials accounting at bulk processing facilities that handle low enriched uranium consists primarily of weight and uranium enrichment measurements. Most low enriched uranium processing facilities draw separate materials balances for each enrichment handled at the facility. The enrichment measurement determines the isotopic abundance of the ^{235}U , thereby determining the proper strata for the item, while the weight measurement generates the primary accounting value for the item.

Recently, these gamma ray based enrichment measurement techniques have been applied to Russian origin material.² The presence of interfering gamma radiation from minor isotopes was confirmed. However, with the advent of fast portable computers, it is now possible to apply more sophisticated analysis techniques to the low-resolution data in the field. Explicit corrections for Compton background, gamma rays from ^{236}U daughters, and the attenuation caused by thick containers can be

part of the least squares fitting routine. An evaluation of a new response function fitting technique is presented and compared to the more traditional enrichment meter method which uses two Regions-of-Interest (ROIs) to determine ^{235}U enrichment.

ENRICHMENT METER PRINCIPLE BASICS

The enrichment of a uranium bearing item is the fraction of ^{235}U to the total uranium present in the item. This enrichment value can be expressed as a weight fraction or an atom fraction of ^{235}U to the total uranium. The enrichment meter principle is applicable to items containing depleted uranium (i.e. $<0.7\%$ ^{235}U fraction) up to highly enriched uranium (HEU) with greater than 90% ^{235}U enrichment. The 185.7-keV gamma ray is the most frequently used signature to measure ^{235}U enrichment.

In general, three sample conditions must be met for the enrichment meter principle to be applicable. First the sample must have a uniform distribution of material that is isotopically uniform and a uniform matrix distribution. Secondly, the daughter activities must be in secular equilibrium with the parent uranium activities. Finally, the material must be infinitely thick with respect to the 185.7-keV gamma peak throughout the entire field of view of the detector as defined by the sample attenuation, collimator, and detector.

When the above sample conditions are met the total 185.7-keV count rate is proportional to the enrichment of the sample. The uranium enrichment as a function of total 185.7-keV count rate is given as:

$$E_w = \left[\frac{\mu_U}{\epsilon SA} \right] R \left[\frac{[1 + (\mu_m \rho_m / \mu_U \rho_U \cdot \exp(\mu_c \rho_c t_c))]}{1 - \exp(-\mu \rho D)} \right] \quad (1)$$

Where:

- μ_x = mass attenuation coefficient at 185.7-keV; subscripts m, c, and U are for matrix, container and uranium, respectively.
- ρ_x = density of material
- t_c = single wall thickness of the sample container
- R = total 185.7-keV count rate
- ϵ = detection efficiency at 185.7-keV
- S = specific activity of the 185.7-keV gamma ray
- D = sample thickness.

The exponential in the denominator tends to zero as D reaches the infinite-thickness criterion. The first bracket in the above equation contains constants dependent only on the instrument properties and intrinsic properties of uranium. R is measured experimentally and the terms in the numerator of the second bracket can be corrected for as appropriate depending on container wall thickness and matrix material type.

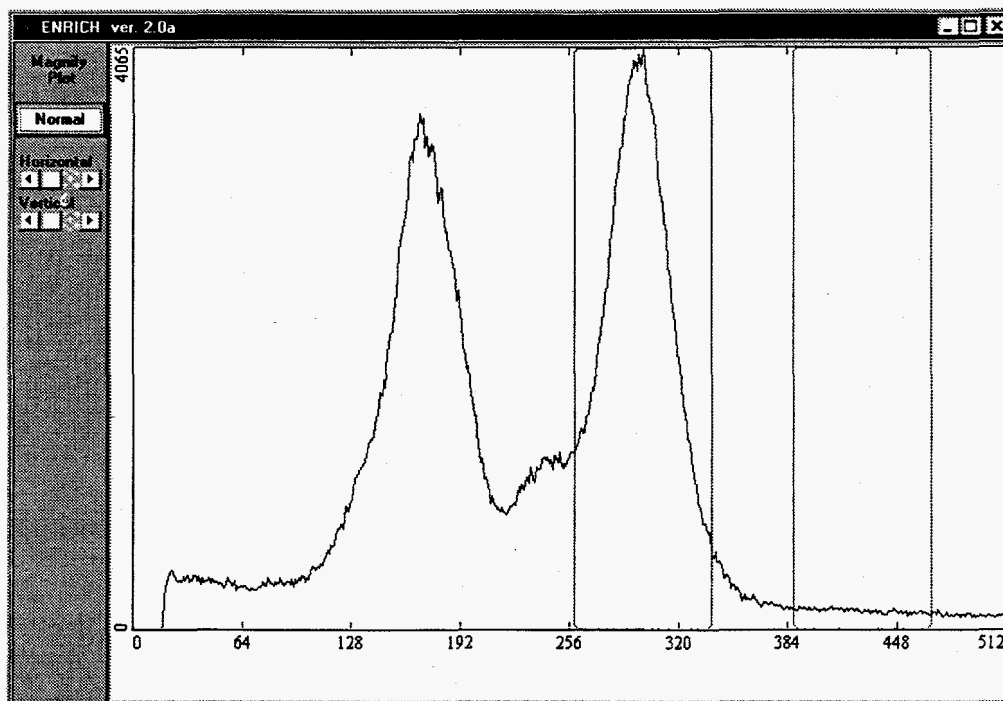


Fig. 1. Gamma spectra of 20.264 at.% ^{235}U as displayed in the enrichment meter software Enrich. The two ROIs used for the determination of uranium enrichment are defined by the rectangular boxes. The peak ROI is centered around the 185.7 keV peak which corresponds to a channel number of 300. (note to self spectra sprinkle\lan\nbl_0005.chn)

ENRICHMENT METER USING NAI DETECTORS

The enrichment meter principle using low resolution NaI scintillator detectors has been in use in the U.S. for more than 25 years.¹ Using equation 1 as the basis for the technique two regions of interest (ROI) are defined. A typical uranium spectra taken with a 1 in. dia. x $\frac{1}{2}$ in. thick NaI detector is displayed in Fig. 1, with two ROIs defined by the rectangular boxes. One ROI surrounds the 185.7-keV peak which in practice includes the 163.4-keV and 205.3-keV peaks as well as other less intense peaks in the region. Typical NaI detector resolution of better than 8% FWHM at 661-keV allow the identification of the 143.8-keV peak as a shoulder on the peak region of interest. A second ROI is set in the higher energy region outside the peak ROI. This ROI is used to subtract background counts from the peak ROI. Each ROI is typically 50 keV wide. This measurement technique is calibrated using two or more reference samples of know enrichment. Using references that are representative of unknowns with respect to container thickness, material type and enrichment, it is possible to make measurements with a relative accuracy of 0.25% (1σ).³ The method becomes less accurate when non representative references are available or the material type, packaging and gamma ray background is varying from sample to sample. Also, a large gamma ray background can reduce the accuracy of the results.

Although the mathematical simplicity of the technique allows for easily made hand calculations, many software programs have been developed for uranium enrichment meter analysis. More sophisticated algorithms allow for corrections due to different matrix types and container wall thickness based on operator input parameters and equation 1.

IMPROVEMENTS TO ENRICHMENT METER METHOD

An improvement to the traditional enrichment-meter method of determining ^{235}U enrichments applicable to material containing reactor returns, is made by fitting computed response profiles to the observed data of NaI spectra in the 130 to 290 keV region. This new method has been incorporated into a computer analysis code called NaIGEM (NaI Gamma Enrichment Measurements).⁴ Some analysis improvements that are included in NaIGEM are automatic correction for changes in gain and detector resolution between and during data acquisition. Additionally, the calibration of the system requires only a single reference sample spectrum. The calibration can be permanent and be applied to any detector/collimator assembly of identical design. Since peak fitting is applied to the spectrum, large Compton continuum and interference peaks can be accounted for.

The energy region of the NaI spectrum used in NaIGEM is from about 130 keV to 290 keV. Typically, spectra are taken with a 512 channel analyzer, using a gain of 0.6 to 0.7 keV/channel. Good enrichment meter method gamma ray counting practices should be used, which was reviewed in the previous section.

Attenuation due to the sample matrix and container wall thickness are corrected for based on operator input into the code and the appropriate terms in equation 1. A calibration constant (ϵ in equation 1) is determined based on the count rate of a single reference sample.

The method used to analyze the data is to compute a response profile for each of the components contributing to the region of the spectrum that is analyzed. The major contributions to the 130-300 keV region are ^{235}U photoelectric peaks, ^{238}U Compton continuum, low angle Compton scattering and in the case of LEU partially enriched from reactor returns Compton continuum due to minor uranium isotopes. A response function is also included for thorium. The responses are iteratively fit to the observed data by the method of least-squares.

RESULTS (2 ROI VS. NAIGEM)

Three broad groups of spectra types were taken in order to make comparisons of the ability of Enrich and NaIGEM to measure uranium enrichments under varying conditions. Two sets of spectra were taken at LANL using well characterized reference standards and the third data set measured Russian origin material stored at the Ulba facility in Kazakhstan. Some of the Ulba material contains a large Compton continuum contribution due to reactor returns increasing the concentration of the minor uranium isotopes. This material is not well characterized, that is the reference values are not traceable to the international measurement system and container wall thickness are not well know. Therefore, data was collected at LANL that simulated the large Compton background in the Ulba material, in an effort to study this measurement issue.

All uranium enrichment spectra were taken using a PC controlled M³CA and 1 in. diameter and in. thick NaI detectors fitted with lead collimators. In most cases the collimator was 1 in. thick with a 1 in. diameter hole, unless otherwise noted. The 2 ROI enrichment meter software Enrich was used to collect the spectra. Analysis was done using both Enrich and NaIGEM in order to make comparisons between the two techniques.

Table I. Measured enrichments using the two ROI code Enrich and the response function fitting software NaIGEM for spectra collected at the Ulba facility in Kazakhstan. Also listed are % bias ((measured - accepted/accepted)*100) and the one sigma uncertainty determined by the software for each measurement. The first 12 spectra were taken October 23rd and 24th, 1997 and the rest of the spectra were taken December 11th, 1997.

File name	Thickness steel (mm)	Count time (seconds)	Reference enrichment	Enrich	sigma	% Bias	NaIGEM	sigma	% Bias
gfu1	13.2	500	2.4	1.79	0.69	-25.4	2.53	0.15	5.3
gfu2	7.2	500	4.4	4.12	0.53	-6.4	4.32	0.10	-1.8
pel	8.9	500	2.4	-0.07	0.38	-102.8	0.35	0.16	-85.6
pel2	8.9	500	2.4	0.41	0.35	-83.0	0.36	0.076	-85.2
pel3	10.8	500	3.6	1.64	0.52	-54.4	2.04	0.119	-43.5
pel4	3.9	500	2.0	1.18	0.5	-41.0	2.04	0.071	2.0
plav1	3	500	2.51	1.39	0.99	-44.6	2.20	0.075	-12.2
plav2	3.1	500	2.09	0.51	0.68	-75.6	1.91	0.086	-8.6
pow1	3.8	500	3.3	2.48	0.5	-24.8	3.31	0.078	0.3
pow2	4.9	500	2.4	1.51	0.49	-37.1	2.37	0.075	-1.2
ulba2_4	3 glass	100	2.4	2.15	0.13	-10.4	2.23	0.078	-7.0
ulba4_4	3 galss	100	4.4	3.97	0.13	-9.8	4.13	0.090	-6.1
tuk1	11.1	100	2.4	0.62	0.48	-74.2	2.44	0.28	1.6
tuk2	11.1	100	2.4	0.03	0.7	-98.8	3.42	0.38	42.5
tuk3	8.8	100	2.4	0.05	0.31	-97.9	0.90	0.18	-62.5
tuk4	8	100	2.4	0.80	0.41	-66.7	1.63	0.23	-32.1
tuk5	8.8	100	4.4	2.22	0.22	-49.5	3.18	0.17	-27.7
tuk6	10.8	100	4.4	2.31	0.28	-47.5	3.28	0.25	-25.5
tuk7	8.9	100	4.4	2.24	0.23	-49.1	3.38	0.19	-23.2
tuk8	8.8	100	4.4	2.00	0.24	-54.5	3.10	0.19	-29.5
tuk9	10.8	100	3.6	1.63	0.33	-54.7	2.69	0.24	-25.3
tuk10	10.7	100	3.6	1.29	0.32	-64.2	2.76	0.24	-23.3
tuk11	11	100	3.6	2.25	0.3	-37.5	3.71	0.26	3.1
tuk12	11	100	3.6	1.81	0.35	-49.7	3.10	0.29	-13.9
tuk13	2.9	100	0.7	-0.29	0.32	-142.0	0.81	0.14	15.7
tuk14	2.9	100	0.7	-0.38	0.32	-154.3	0.55	0.14	-21.4
tuk15	2.4	100	3.59	2.80	0.13	-22.1	4.16	0.11	15.9
tuk16	1.3	100	3.59	2.74	0.1	-23.7	3.68	0.09	2.5
tuk17	2	100	3.59	2.89	0.12	-19.5	4.32	0.11	20.3
tuk18	1.3	100	3.59	2.45	0.1	-31.8	3.49	0.09	-2.8
pov19	5.1	100	2.4	2.10	0.32	-12.3	3.52	0.18	46.7

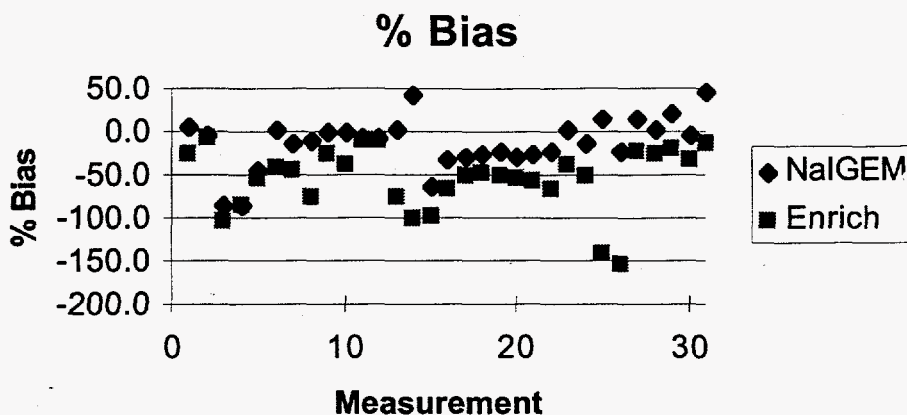


Fig. 2. Enrichment measurement % bias. The diamonds are biases determined using NaIGEM and the squares are the biases associated with using Enrich.

Numerous LEU material types were measured at the Ulba facility including UF_6 , UO_2 , nitrate, and U_3O_8 in solid form. The measured enrichments as determined by Enrich and NaIGEM are presented in Table I. The average absolute % bias are 53.7% and 22.4% for Enrich and NaIGEM, respectively. The bias for each of the measurement techniques is displayed in Fig. 2. Nearly all of the enrichments determined by NaIGEM were within $\pm 50\%$ of the true value with an overall negative bias. All enrichments determined using Enrich were low and many were greater than 50% low.

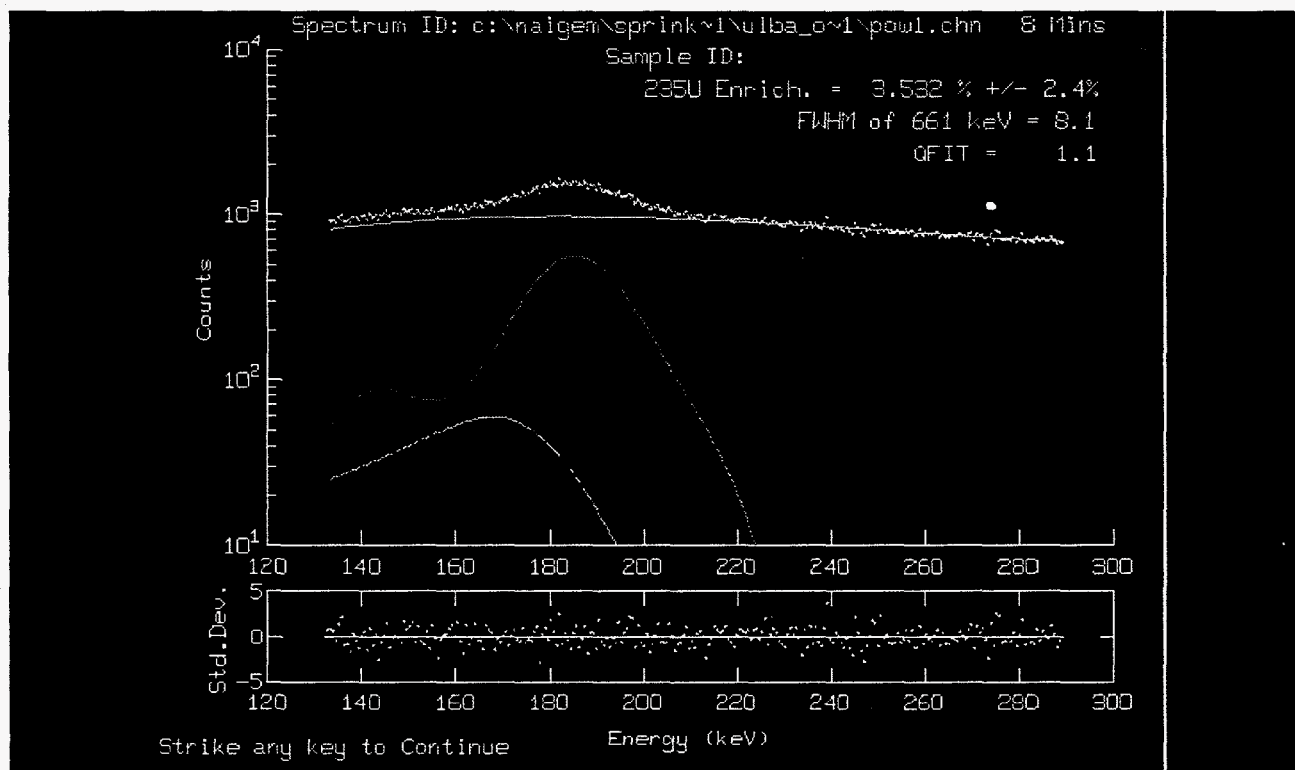


Fig. 3. NaIGEM fitted output spectra of file pow1. The individual response functions for ^{235}U , ^{238}U , and low-angle Compton scattering are displayed as well as the total fit to the data are displayed in the top plot. The lower plot is the residuals from the fit.

The bias in these results are very large due to the large Compton background that is present, uncertainty in the determination of container thickness and the low enrichment of the material. Figure 3 contains a display of the fitted output from NaIGEM for file name pow1. This is also the spectra that was used as in NaIGEM as the calibration spectra for the October data. It can be seen in Fig. 3 that the counting statistics are dominated by the Compton continuum.

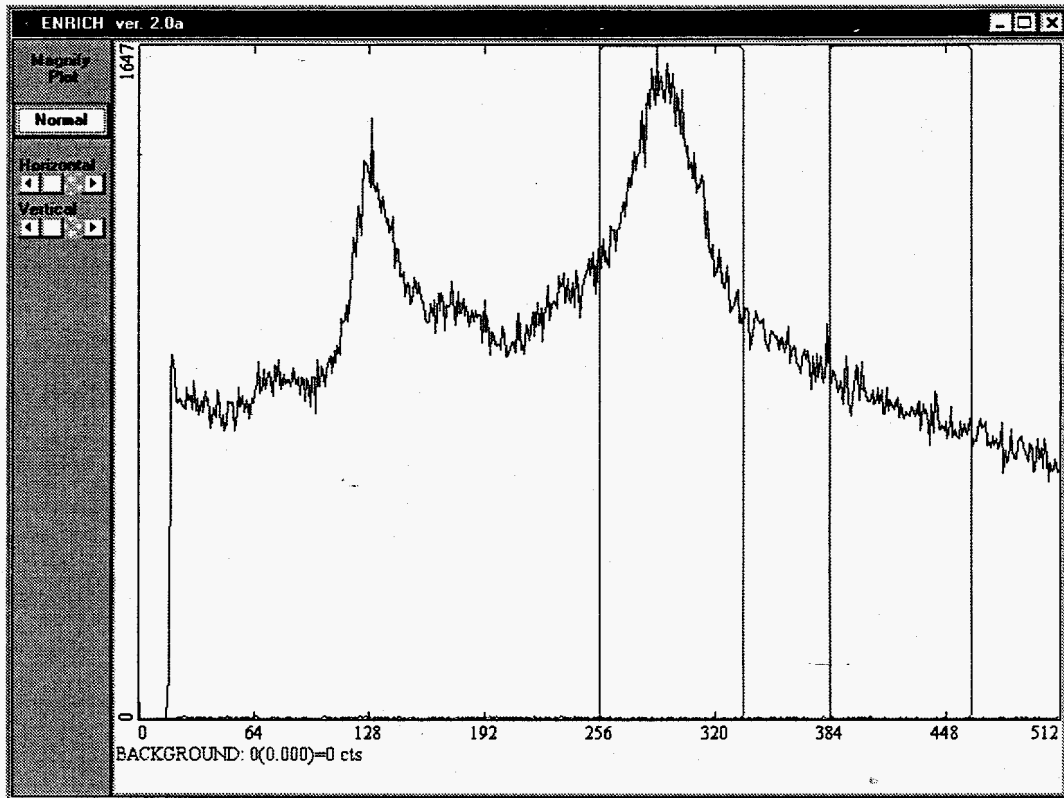


Fig. 4. Output from Enrich of spectra pow1. As in Figure 1 the rectangular boxes define the peak and background ROIs.

The output from Enrich of spectra pow1 is displayed in Fig. 4. The rectangular boxes define the 185.6 keV peak ROI and background ROI. A comparison of the background ROI in Fig. 4 to Fig. 1 shows the difficulty in dealing with a large and/or varying Compton background when enrichment measurements are made using the 2 ROI method. Using response function fitting NaIGEM has the ability measure and correct for widely varying Compton contributions this is well illustrated by the fits to the spectra in Fig. 3.

Table II. Measured enrichments using the two ROI code Enrich and the response function fitting software NaIGEM. Also listed are % bias (measured-accepted/accepted]*100) and the one sigma uncertainty determined by the software for each measurement. The listed averages for % bias are average absolute % bias.

Reference standard	reference enrichment	Enrich U-235 at. %	sigma	% Bias	NaIGEM U-235 at. %	sigma	% Bias
5 min. data							
nbl0005	20.264	20.18	0.14	-0.4	20.215	0.178	-0.2
nbs295	2.9857	2.997	0.038	0.4	3.007	0.026	0.7
nbs446	4.5168	4.499	0.045	-0.4	4.562	0.042	1.0
nbs194	1.9664	1.938	0.039	-1.4	1.986	0.034	1.0
nbs071	0.7209	0.716	0.039	-0.7	0.724	0.033	0.4
nbl0006	52.56	52.332	0.31	-0.4	52.217	0.406	-0.7
nbs031*	0.3206	0.378	0.037	17.9	0.372	0.033	16.0
50 min. data							
nbl0005	20.264	20.06	0.13	-1.0	20.154	0.202	-0.5
nbl0006	52.56	52.05	0.3	-1.0	52.315	0.709	-0.5
nbs446	4.5168	4.513	0.045	-0.1	4.539	0.039	0.5
nbs295	2.9857	2.98	0.038	-0.2	2.989	0.03	0.1
nbs194	1.9664	1.969	0.035	0.1	1.993	0.02	1.4
nbs071	0.7209	0.708	0.032	-1.8	0.74	0.012	2.6
nbl031*	0.3206	0.333	0.0311	3.9	0.331	0.01	3.2
average			0.0940	0.66		0.13	0.80
*Data for this standard is excluded from the average							

Several different well characterized uranium reference standards were measured and analyzed using both Enrich and NaIGEM to gauge the relative performance on the most basic enrichment measurements, clean samples with thin container walls (2 mm aluminum). The reference material enrichments ranged from depleted uranium (0.3 atom % (at.%) ²³⁵U) to HEU (>50 at. % ²³⁵U). The software determined enrichments are tabulated in Table II. Both codes performed well when care was taken in determining the calibration constants used within the software. The Enrich calibration was software determined using three 50 min. data files nbl0005, nbs446, and nbs295 with enrichments of 20.264, 4.5168, and 2.9857 at. % ²³⁵U, respectively. In order to minimize the single point effect in NaIGEM an average calibration constant was used based on several spectra of different enrichment. Best results were obtained by inputting by hand an average calibration constant. Calibration constants were determined for the 5 min. and 50 min. files nbl0005, nbs446, and nbs295 and the six values were averaged together and input into the program to give an average calibration constant. If a calibration constant was used based on any one spectra the overall bias for the complete data set would be increased

The average absolute % bias using Enrich was 0.66% and 0.80% using NaIGEM. The main difference between the codes was that Enrich was overall biased low by 0.23% and NaIGEM gave an overall bias that was high by 0.70%. In general, the 2 ROI method performs better if the uranium oxide has very little minor isotopes of uranium and is measured in a container that is the same material type and thickness as the container used during calibration. Note that both codes gave poor accuracy

results based on the 5 min. spectra of NBS031 which had an enrichment of 0.321 at.%. This is not surprising due to the low concentration of ^{235}U in the sample providing poor counting statistics as input into the analysis codes. It should be noted that NaIGEM had less bias for both NBS031 spectra. The NBS031 spectra results were not included in the above mentioned average absolute % bias determination. Difficulty arises in applying the 2 ROI method when there are large differences in container thickness, or if the minor isotope fraction is larger.

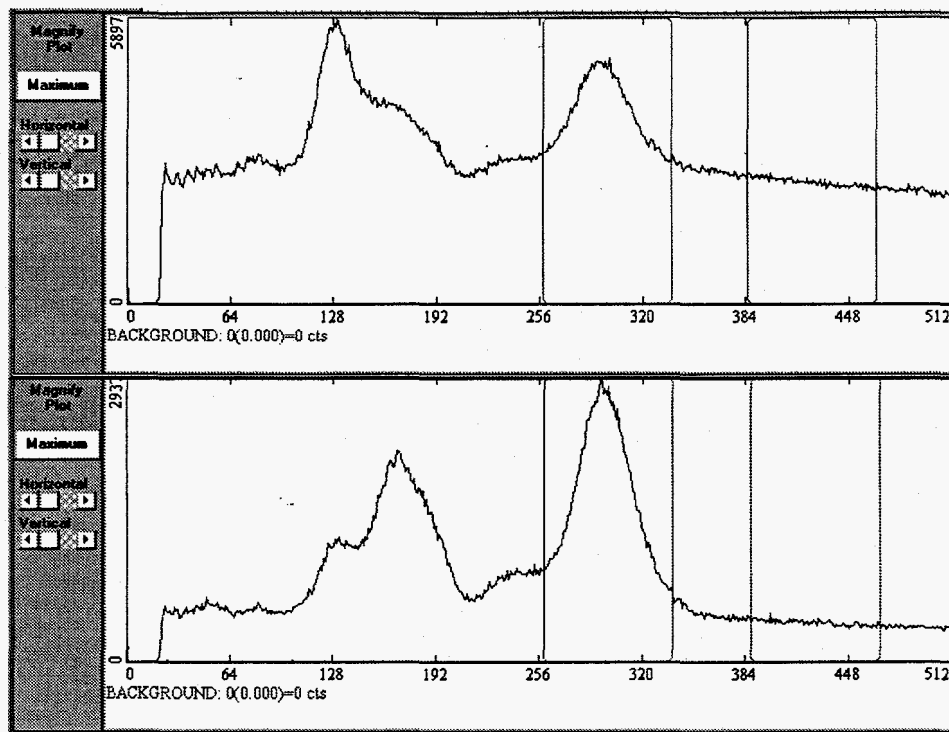


Fig. 5. The top panel displays a spectra taken of reference standard A1-324-2 (10.04 at.% ^{235}U) with 427.5 grams of ^{232}Th placed next to the lead shielding around the circumference of the detector. The bottom panel is a spectra of the same reference standard but with only 227 grams of ^{232}Th .

In order to study these effects under controlled conditions, measurements were made at Los Alamos. Spectra were collected while the NaI crystal was exposed to ^{232}Th background penetrating a 1 in. thick lead shield surrounding the detector. This was done to simulate the large Compton background present in some of the Ulba spectra. The gram quantity of thorium is directly proportional to the magnitude of Compton continuum in the spectra. The relative difference in Compton continuum contribution for the maximum and minimum amount of thorium is presented in Fig. 5. Presented in the top panel of Fig. 5 is a spectrum taken of reference standard A1-324-2 (10.04 at.% ^{235}U) with 427.5 grams of ^{232}Th placed outside the lead shield, the only difference in the bottom panel is the amount of ^{232}Th is only 227 grams. Clear differences between these spectra exists. The intensity of the x-ray region relative to the 186.5 keV peak region is roughly the reciprocal of each other for each of the gram quantities. Also the intensity of the high energy Compton continuum relative to the 186.5 keV peak region is much larger for the 427.5 grams of thorium spectra.

Table III. Measured enrichment, one sigma uncertainties and % bias ($[(\text{measured}-\text{accepted})/\text{accepted}]*100$) as determined by Enrich and NaIGEM. The ^{232}Th metal and powder was placed adjacent to the 1 in. thick lead shielding surrounding the NaI crystal as a simulation of the large Compton continuum found in the Ulba spectra. The count time for each of the spectra was 5 min.

Reference standard	Reference enrichment	Enrich U-235 A.T.%	sigma	% Bias	NaIGEM U-235 A.T.%	sigma	% Bias
lead collimator 2.54 cm dia. x 2.54 high							
Standards only							
UIISO-17	17.42	17.281	0.078	-0.8	17.562	0.152	0.8
A1-423-2	10.2	10.197	0.06	0.0	10.365	0.094	1.6
A1-1126-1	3.063	3.055	0.046	-0.3	3.097	0.47	1.1
A1-1125-1	1.96	2.002	0.045	2.1	1.919	0.04	-2.1
227 g Th powder							
UIISO-17	17.42	17.138	0.098	-1.6	17.638	0.159	1.3
A1-423-2	10.2	9.972	0.082	-2.2	10.369	0.098	1.7
A1-1126-1	3.063	2.877	0.073	-6.1	3.124	0.054	2.0
A1-1125-1	1.96	1.701	0.073	-13.2	2.14	0.05	9.2
227 g Th powder + 15 g Th metal							
UIISO-17	17.42	15.799	0.227	-9.3	17.368	0.161	-0.3
A1-423-2	10.2	8.322	0.22	-18.4	10.208	0.114	0.1
A1-1126-1	3.063	1.476	0.212	-51.8	3.065	0.078	0.1
A1-1125-1	1.96	0.394	0.216	-79.9	1.743	0.078	-11.1
227 g Th powder + 15 g Th metal (2)							
UIISO-17	17.42	16.652	0.124	-4.4	17.379	0.169	-0.2
A1-423-2	10.2	9.653	0.111	-5.4	10.3	0.102	1.0
A1-1126-1	3.063	2.51	0.103	-18.1	3.137	0.061	2.4
A1-1125-1	1.96	1.426	0.103	-27.2	1.92	0.053	-2.0
227 g Th powder + 31.5 g Th metal							
UIISO-17	17.42	16.368	0.147	-6.0	17.488	0.157	0.4
A1-423-2	10.2	9.247	0.136	-9.3	10.059	0.107	-1.4
A1-1126-1	3.063	2.279	0.128	-25.6	3.197	0.068	4.4
A1-1125-1	1.96	1.164	0.129	-40.6	1.8	0.056	-8.2
227 g Th powder + 77 g Th metal							
UIISO-17	17.42	15.982	0.196	-8.3	17.435	0.171	0.1
A1-423-2	10.2	8.984	0.185	-11.9	10.248	0.11	0.5
A1-1126-1	3.063	1.806	0.181	-41.0	3.142	0.078	2.6
A1-1125-1	1.96	0.575	0.182	-70.7	1.997	0.069	1.9
227 g Th powder + 77 g Th metal (2)							
UIISO-17	17.42	15.679	0.218	-10.0	17.28	0.163	-0.8
A1-423-2	10.2	8.5	0.21	-16.7	10.034	0.112	-1.6

Table III.
(cont.)

A1-1126-1	3.063	1.46	0.205	-52.3	3.155	0.077	3.0
A1-1125-1	1.96	0.546	0.205	-72.1	1.833	0.069	-6.5
227 g Th powder + 108.5 g Th metal							
UIISO-17	17.42	15.469	0.246	-11.2	17.224	0.179	-1.1
A1-423-2	10.2	8.428	0.24	-17.4	10.297	0.116	1.0
A1-1126-1	3.063	1.24	0.237	-59.5	2.999	0.079	-2.1
A1-1125-1	1.96	0.174	0.237	-91.1	1.756	0.077	-10.4
227 g Th powder + 154 g Th metal							
UIISO-17	17.42	14.598	0.327	-16.2	17.279	0.185	-0.8
A1-423-2	10.2	7.65	0.322	-25.0	10.222	0.125	0.2
A1-1126-1	3.063	0.346	0.32	-88.7	2.778	0.089	-9.3
A1-1125-1	1.96	-0.247	0.317	-112.6	1.811	0.091	-7.6

Average absolute % bias

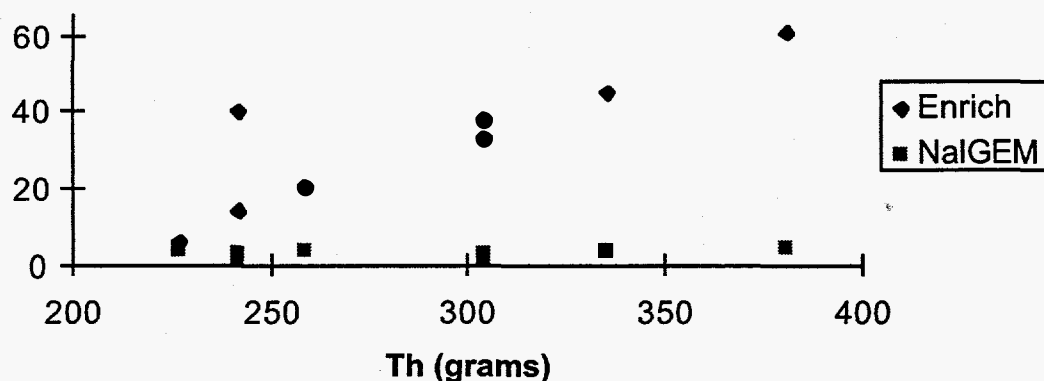


Fig. 6. Average absolute % bias as a function of grams ^{232}Th . The thorium was used to simulate the large Compton continuum present in the Ulba data.

The results obtained using Enrich and NaIGEM on the simulated large Compton background spectra are presented in Table III. As expected, due to ^{235}U counting statistics, the absolute bias increases with decreasing enrichment. Better results for the lowest enrichments could be obtained by increasing count time.

A concise comparison of each methods capabilities is presented in Fig. 6. The plot displays the average absolute enrichment % bias as a function of grams thorium derived from the data in Table III. The absolute % bias results obtained for each of the four different enrichments listed in Table III were averaged together at each thorium gram quantity to give the average absolute enrichment % bias as a

function of grams thorium. The distribution calculated from NaIGEM results and presented in Fig. 6 is not dependent on the amount of Compton continuum present. Therefore, enrichment precision results determined using NaIGEM will not be influenced by varying background. The results determined using Enrich show a strong bias dependence as the Compton background is changed. The bias becomes more and more negative as the amount of Compton continuum increases. The reason for this is apparent from the background ROIs displayed in Fig. 5 for the two quantities thorium interference. In the case of larger thorium interference a severe over estimate of the background under the 186.5 keV peak is made leading to enrichment values that are too low.

ADDITIONAL NAIGEM PERFORMANCE TESTS

Performance tests were done using NaIGEM to determine the applicability of one calibration constant to several different detectors utilizing the same measurement and crystal geometry and replicate measurements were made to test the accuracy of the software reported uncertainty in the enrichment determination.

Table IV. NaIGEM determined calibration constants for several different detector models. All detectors are manufacture specified as 1 in. dia. by $_$ in. thick NaI crystals with 1 in. dia. by 1 in. thick lead collimator. Alphanumeric model numbers starting with G or H are from Bicon Corporation⁵ with the last two purchased from EFC Company.⁶

Detector	Calibration Constant (xE-4) 300 sec	% Error (1RSD)	Calibration Constant (xE-4) 3000 sec	% Error (1RSD)
GP644	2.425	0.9	2.436	1.1
GP645	1.935	0.8	1.932	1.2
GP647	1.982	0.9	1.985	1.3
GP648	1.888	0.8	1.882	1.1
GG494R	2.237	0.8	2.247	0.9
HO-384	1.997	0.9	2.001	1.2
HX-517	1.861	0.8	1.85	1
EFC11	1.95	0.9	1.953	1.1
EFC12	2.168	0.8	2.15	1
average	2.049	0.84	2.048	1.10
stdev	0.188		0.192	

Nine different detectors from two manufactures covering five model types were used to measure a 20.264 at. % ²³⁵U enrichment standard (NBL-0005). Sample counting times of 5 min. and 50 min. were made using each detector. The calibration constant determined for each spectra by NaIGEM are presented in Table IV along with the relative uncertainty associated with the constant. The average calibration constants for the two counting times were 2.05E-04±9% (1RSD) for both. The uncertainty between detectors is an order of magnitude larger that the uncertainty on the calibration constant for a given detector. All of these detectors have the same declared NaI crystal size, lead collimator size and used the same electronics to collect the spectra therefore it is recommended that a calibration for each detector be performed. The variation in calibration constant is apparent even within manufacturer and model type. Two factors are likely contributors to the variation in detector efficiency differences. Variations in the physical size of the detectors from the specified 1 in. dia. x $_$ in. thick and alignment of the detector with the collimator could also increase the variability of detector response. The detector collimator geometry is extremely important for these detectors because the diameter of the detector matches that of the collimator.

Table V. Uranium enrichment measurements calculated using NaIGEM including one sigma uncertainty predicted by software. NBL standard 146-0005 with an accepted enrichment of 20.264 at.% was used for all 25 runs. The average enrichment value for the 25 runs is 20.328% \pm 0.100 (1 σ). The average software predicted uncertainty is 0.174% \pm 0.0075 (1 σ).

Run #	% U-235	Software predicted Uncertainty	Run #	% U-235	Software predicted Uncertainty
1	20.285	0.167	14	20.552	0.172
2	20.216	0.188	15	20.357	0.176
3	20.174	0.178	16	20.332	0.167
4	20.426	0.168	17	20.367	0.17
5	20.35	0.178	18	20.368	0.187
6	20.264	0.167	19	20.281	0.175
7	20.333	0.176	20	20.298	0.167
8	20.369	0.168	21	20.099	0.166
9	20.444	0.172	22	20.161	0.18
10	20.444	0.172	23	20.382	0.17
11	20.356	0.171	24	20.367	0.183
12	20.313	0.193	25	20.416	0.181
13	20.242	0.167			

A series of 25 5 min. spectra were taken using the same detector, electronics and sample to determine the accuracy of the software reported measurement uncertainty. The book value ^{235}U enrichment of the reference sample used was 20.264 at.%. The average ^{235}U enrichment determined from the 25 spectra was 20.32 at.%, giving a relative bias of -0.3%. A complete listing of the individual spectra results is presented in Table V. Detector EFC12 was used to collect all 25 spectra. Run #1 was used to determine the calibration of NaIGEM for the complete set of spectra. The calibration factor was determined to be $2.155\text{E-}4 \pm 0.8\%$ (1RSD) with a gain of 0.70 (keV/channel) and a zero of -26.81 (keV). The standard deviation of the data was 0.5% (1RSD), while the average software predicted uncertainty was nearly a factor of two higher at 0.9% (1RSD). The software predicted uncertainty will be adjusted accordingly by the developer, once these precision tests have been confirmed by more complete testing at different enrichment values and count times.

FUTURE PLANS

In order to develop the NaIGEM code more completely test should be made with different detector collimator combinations. These tests would better determine the intrinsic uncertainty of the analysis technique and allow for improvement in the response function fitting, further improving the code. It would be useful to develop the ability to fit absorber thickness based on user input container material and a first guess of thickness. This would reduce much of the uncertainty encountered during field measurements where container thickness are not well know and have to be measured ultrasonically. Finally, the application of this response function fitting technique to CdZnTe detectors should be investigated.

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