GEM Algorithm Advancements to Improve Reprocessed Uranium Enrichment Measurements

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Abstract

A lanthanum bromide ($LaBr_3(Ce)$) detector coupled to a digital multichannel analyser and gamma enrichment measurements (GEM) spectrum analysis software currently is the IAEA "workhorse" medium-resolution gamma spectrometer employed for uranium enrichment verification. As the associated GEM spectrum fitting algorithm provides a high-quality fitting with low systematic error, it makes only a minor contribution to the total measurement uncertainty across various enrichments and material types, including materials with moderate content of reprocessed uranium (RepU).

Recent tests on blends containing elevated amounts of RepU (^{232}U content higher than 0.5 ppb) showed a degradation of the spectrum fitting quality that led to a noticeable systematic error and, hence, biased results overestimating the ^{235}U enrichment. The issue stemmed from spectral interferences caused by gamma emissions of ^{232}U decay daughters in the spectrum analysis range from 130 to 290 keV. Apart from a set of discrete-energy gamma rays, out of which only the most intense line of ^{212}Pb at 238.6 keV was considered in the standard GEM algorithm, a substantial interference was found to be also due to a cumulative backscatter peak of intense gamma rays of ^{208}Tl and ^{212}Bi with energies from 0.51 to 2.61 MeV.

As a result of this work, the GEM algorithm was upgraded to include a complete set of discrete low-energy lines and a cumulative backscatter peak from the higher energy lines of ^{232}U decay daughters. MCNP simulations were used to determine the backscatter peak shape for the IAEA standard $LaBr_3(Ce)$ detector and collimator setup. The simulated backscatter continuum was approximated analytically and its amplitude was linked to the area of the 238.6 keV peak, thereby forming a combined ^{232}U spectrum fitting component. The modified GEM algorithm was field tested using a set of RepU material items containing substantial amounts of ^{232}U. The testing results showed a significant improvement of the spectrum fitting quality and unbiased uranium enrichment results.

1. Introduction

A large fraction of the total world inventory of separated RepU represents materials with very low residual ^{235}U enrichment, which are accumulated in long-term storage facilities, typically in solid oxide forms, either UO_3 or U_3O_8 [1]. In addition to the three naturally occurring isotopes, ^{234}U, ^{235}U and ^{238}U, RepU contains synthetic isotopes ^{232}U, ^{233}U, and ^{236}U. The RepU extracted from high-burnup nuclear fuel is considered to be low-quality material because of high content of ^{232}U and ^{236}U isotopes.

Significant absorption of neutrons by ^{236}U necessitates re-enrichment of RepU to higher levels of ^{235}U compared to normal LEU fuels. In the case of ^{232}U, the limitations are caused by its high specific activity ($T_{1/2}$ = 70.6 years) and the intense gamma and beta emissions of its decay daughters ^{212}Pb, ^{212}Bi and ^{208}Tl [2]. Therefore, even the small quantities of ^{232}U normally found to be between 0.5 and 5 ppb in RepU [3] pose a significant radiological hazard and affect its non-destructive assay by causing excessive count rates, spectral interferences and deterioration in the signal-to-noise ratio [4].

A collimated $\varnothing 2'' \times 0.5''$ $LaBr_3(Ce)$ detector coupled to the MCA527 miniature digital multichannel analyser from GBS Elektronik and a palmtop computer is the IAEA's "workhorse" MMCL system,

a medium-resolution gamma spectrometer that has been heavily utilized for uranium enrichment verifications. The high intrinsic efficiency and fast decay time of the $LaBr_3$(Ce) detector make it well-suited for measurements of both unirradiated uranium and RepU. In the latter case, it became the detector of choice due its capability to tolerate extreme count rates, while being able to acquire good counting statistics from a weak residual ^{235}U signal.

A typical $LaBr_3$(Ce) spectrum of an "infinitely thick" sample of RepU is shown in Figure 1. ^{235}U enrichment is inferred using the enrichment meter principle, which relies on the absolute intensity of counts in the 185.7 keV peak of ^{235}U. The peak counts are accurately determined by spectrum fitting implemented in the IAEA's gamma enrichment measurements software (GEM) [5].

The high accuracy of the spectrum fitting is achieved by considering several physical components, including the ^{235}U and ^{238}U(^{234m}Pa) full-energy peaks, the forward scattering continuum appearing on the low-energy side of the full-energy peaks, and the continuum due to the bremsstrahlung of beta particles and the Compton scattering of higher energy gamma rays inside the detector sensitive volume.

Because of the limited energy resolution of the $LaBr_3$(Ce) detector (FWHM ≈ 5% @ 185.7 keV) and the need to accurately delineate background under overlapping peaks, the fitting is performed in a rather wide energy interval from 130 keV to 270 keV. In the case of RepU, in order to fully accommodate the intense peak of ^{212}Pb at 238.6 keV[1], the upper boundary of the interval is extended even further to 290 keV.

Datasets obtained during recent field campaigns revealed a noticeable degradation of the spectrum fitting quality and a positive systematic bias of the measured enrichments for RepU materials with elevated amounts (≥0.5 ppb) of ^{232}U. As indicated by the data (see Figure 2), the bias increases linearly with the intensity of the 238.6 keV peak (i.e., proportional to the amount of ^{232}U), reaching 25–30% at the upper end of the ^{232}U content (≈2.6 ppb) in the measured items.

This paper presents the results of the root cause investigation of the bias. We describe modifications made to the GEM analysis algorithm together with supporting Monte Carlo computations and present the results of the validation of the upgraded code against the field data.

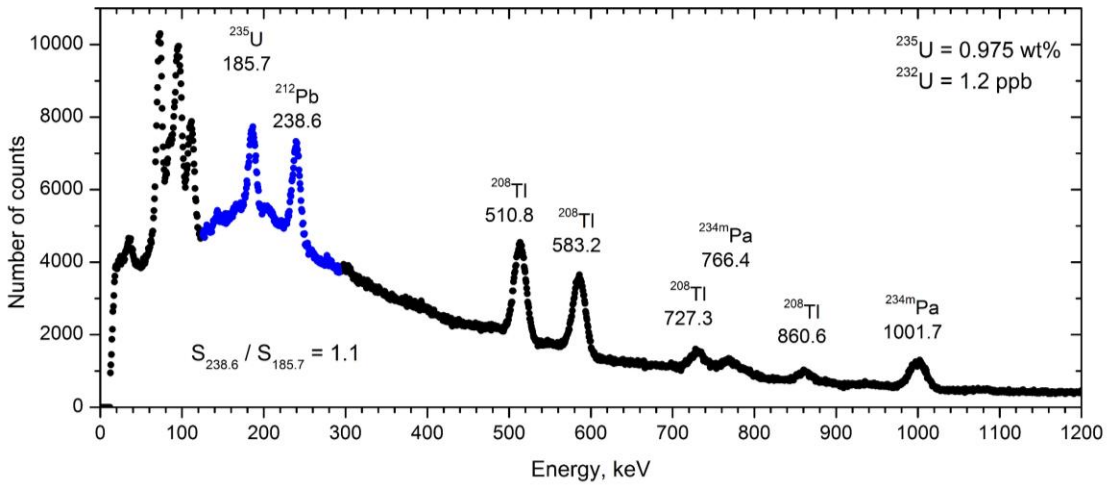

Figure 1. Gamma spectrum taken by the IAEA $LaBr_3$(Ce) detector from an "infinitely thick" RepU material with 1.2 ppb ^{232}U content and 0.975 wt% ^{235}U enrichment. The spectral region processed by the GEM software is highlighted in blue.

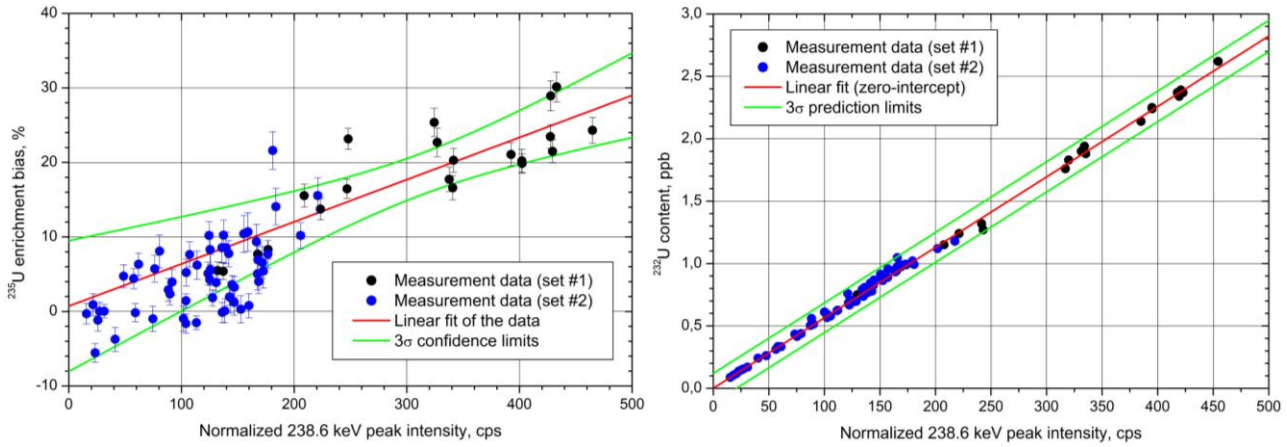

Figure 2. The ^{235}U enrichment bias and estimated ^{232}U content in the measured RepU items as function of 238.6 keV peak intensity. The raw peak intensities were renormalized to the same container wall thickness. The ^{232}U content was estimated by assuming radioactive equilibrium in the ^{232}U decay chain.

2. Root cause investigation

As clearly indicated by the data in Figure 2, the bias is related to the presence of ^{232}U in the measured items. Evaluation of the discrete-energy gamma rays of all isotopes in the ^{232}U decay chain did not reveal gamma emissions that could noticeably contribute to the region of interest around the 185.7 keV peak. Thus, the possibility of direct interference by discrete-energy gamma rays was excluded.

Another source contributing to the bias might be the specific shape of the continuum background in the spectrum fitting interval, which could be caused by the backscattering of abundant higher energy gamma rays emitted by the radionuclides in the ^{232}U decay chain and might not be adequately described by the current GEM model.

The gamma rays backscattered in the materials surrounding the detector are known to produce a wide continuum peaking at 170–250 keV (also known as a backscatter peak) [6]. The continuum shape is largely determined by the peculiarities of the measurement geometry (including source, detector, shielding etc.), while the position of its maximum is defined mainly by the energy/ies of the primary photons emitted by a source.

Because of the distinctive shape of the backscatter peak, it is reasonable to expect that its inclusion into the GEM fitting model will effectively reduce the intensity of the 185.7 keV peak (as the counts representing the backscatter peak will be effectively subtracted from the 185.7 keV peak counts). Since the amplitude of the backscatter peak is linearly proportional to the ^{232}U content, the decrease in the intensity of the 185.7 keV peak also should be proportional to the ^{232}U content. Hence, it can be expected that the positive bias in the measured ^{235}U enrichment observed in the presence of significant amounts of ^{232}U will be eliminated or at least significantly reduced.

3. Monte Carlo simulations

To support development of an improved GEM fitting algorithm, a series of Monte Carlo computations was performed on the cumulative backscatter peak continuum caused by the radiation emitted by radionuclides in the ^{232}U decay chain.

The MCNP model of the detector assembly included the $LaBr_3(Ce)$ crystal, crystal packaging, photomultiplier tube (PMT), radiation shielding and collimator. The PMT model considered the glass bulb, electrical and magnetic insulation and dynode-air mixture volume. The detector assembly model was optimized and benchmarked against measurements with a ^{137}Cs gamma point source as a reference (both unshielded and shielded). The results of the validation of the model are shown in Figure 3.

Simulations for the ^{232}U decay daughter emissions considered UO_3 powder in a steel bottle typical for RepU storage (30 cm outer diameter, 5 mm wall thickness). The source term considered gamma rays in the 510–2614 keV energy range and emission probabilities above 1%, thus encompassing the 510.8 keV, 583.2 keV, 860.6 keV and 2614.0 keV gamma rays of ^{208}Tl and the 727.3 keV, 785.4 keV and 1620.5 keV gamma rays of ^{212}Bi. The backscatter peak continuum obtained as a result of the simulations was Gaussian broadened to account for the energy resolution of the $LaBr_3(Ce)$ detector in the energy range under consideration. One example of the simulated backscatter continuum is shown in Figure 4.

In addition to the above computations, other simulation runs were performed to calculate the intensity of counts in the 238.6 keV peak of ^{212}Pb, which was used to scale the backscatter continuum in the GEM code during calculations of the ^{232}U profile.

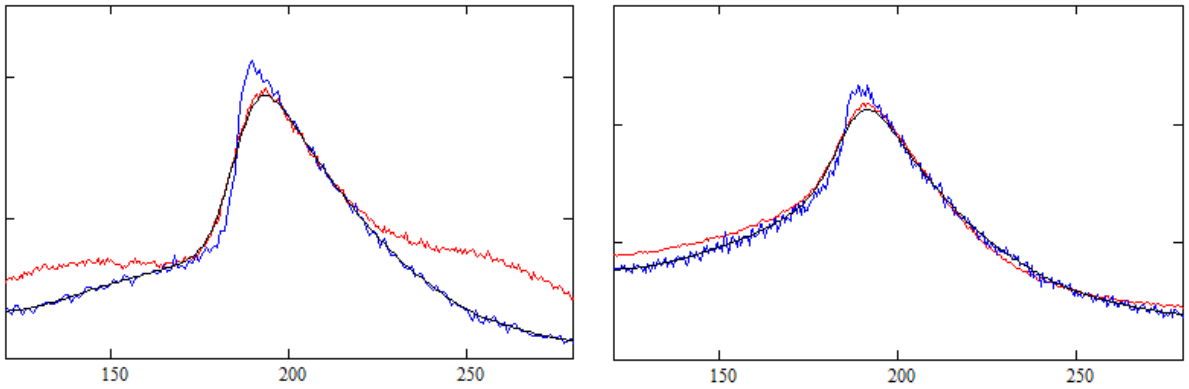

Figure 3. Backscatter peak regions of the gamma spectra acquired from an unshielded (left) and 19-mm-lead shielded (right) ^{137}Cs point source. Red curves – experimental data points; blue curves – unbroadened simulation data points; black curves – Gaussian broadened simulation data points.

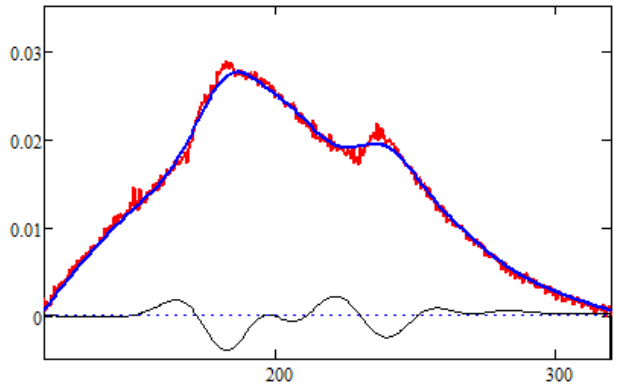

Figure 4. Backscatter peak continuum from the high-energy gamma rays emitted by radionuclides in the ^{232}U decay chain. Red curve – unbroadened simulated spectrum; blue curve – broadened simulated spectrum, black curve – the analytical approximation vs. broadened simulated spectrum residuals (×10 magnified).

Analytical functions were applied to parameterize the backscatter peak continuum obtained for the ^{232}U decay chain in the energy range of interest (120–320 keV), as follows:

$$BS(E_j) = f_{BS} \cdot S_{239} \cdot F(E_j) \cdot \exp(\Delta\mu \cdot \Delta x) \quad \text{Eq. 1}$$

where $BS(E_j)$ is the backscatter peak count density function (counts/keV), E_j is the energy of the j-th spectrum channel, f_{BS} is the backscatter peak scaling factor, S_{239} is the area of the 238.6 keV peak of ^{212}Pb and $F(E_j)$ is the backscatter peak shape function.

The exponential term in Eq. 1 accounts for the deviation $\Delta x = x - x_0$ of the container wall thickness x(cm) from the value x_0 = 0.5 cm assumed in simulations. The term $\Delta\mu$ is the difference of the linear attenuation coefficients at 238.6 keV and 583.2 keV for iron. This simplified model makes use of the fact that the 583.2 keV line is the main contributor to the backscatter hump under the 185.7 keV peak. The backscatter peak shape function in Eq. 1 has the form:

$$F(x) = b_0.(x - b_1) \cdot \left(1 - \frac{1}{1 + \exp(-(x - b_3) \cdot b_4)}\right) + b_2 + \sum_{k=0}^{3} a_k \cdot exp\left(-\left(\frac{x - c_k}{s_k}\right)^2\right) \quad \text{Eq. 2}$$

where a_k, b_k, c_k and s_k are coefficients determined by the least-squares fitting of the simulated backscatter peak. The coefficient values specific to the IAEA's MMCL system are shown in Table 1.

Table 1. Coefficients of the backscatter peak shape function defined by Eq. 2.

Index	a_k	b_k	c_k	s_k
0	0.0088001	0.51484	0.18312	0.015355
1	0.0064368	0.10501	0.23992	0.017025
2	0.0016037	-0.0047142	0.26500	0.016160
3	0.0051368	0.19264	0.20500	0.015972
4	n/a	23.651	n/a	n/a

4. GEM code upgrading

The part of the GEM algorithm responsible for the calculation of the fitted ^{232}U profile was modified as follows.

The list of discrete-energy gamma rays used to generate the ^{232}U full energy peak distribution was revised. Originally consisting of only a single gamma ray from ^{212}Pb at 238.6 keV, the list was extended to the total number of 20 gammas emitted by ^{228}Th, ^{224}Ra, ^{212}Pb, ^{212}Bi and ^{208}Tl. The extended list is shown in Table 2. The gamma ray energies and emission probabilities were taken from the latest revision of the isotope tables available from the Decay Data Evaluation Project's web site [7].

The backscatter peak model described in the previous section was implemented by adding the backscatter continuum calculated using Eq.1 and Eq.2 to the full energy peak distribution of the discrete-energy gamma rays, thereby forming a combined ^{232}U profile fitted to the spectrum. As part of fine-tuning the GEM algorithm, the scaling coefficient f_{BS} in Eq. 1 was adjusted by minimizing the overall bias for the set #1 test spectra (see Section 5). This optimization yielded a coefficient value of f_{BS} = 0.8.

Table 2. Discrete-energy gamma rays considered in the calculation of the fitted ^{232}U profile.

Isotope	E, keV	Yield[1]	Isotope	E, keV	Yield*	Isotope	E, keV	Yield[1]
^{212}Pb	115.183	0.624	^{228}Th	182.29	5.1E-06	^{224}Ra	240.986	4.12
^{228}Th	131.612	0.127	^{228}Th	205.99	0.0188	^{208}Tl	252.71	0.28
^{228}Th	142.71	1.3E-06	^{208}Tl	211.52	0.065	^{208}Tl	277.37	2.37
^{212}Bi	164.80	0.0055	^{228}Th	215.985	0.246	^{212}Bi	288.18	0.32
^{228}Th	166.41	0.1004	^{228}Th	228.42	1.8E-05	^{224}Ra	292.7	6.3E-03
^{212}Pb	176.64	0.052	^{208}Tl	233.36	0.111	^{212}Pb	300.089	3.18
^{212}Bi	180.2	0.0031	^{212}Pb	238.632	43.5			

* The number of photons emitted per 100 disintegrations of ^{228}Th[2].

5. Experimental tests

The upgraded GEM code was validated using two sets of field measurements (see Figure 2) comprising 21 (set #1) and 60 (set #2) spectra. The measurements were performed using the IAEA standard MMCL gamma spectrometry system that includes a ∅2″ × 0.5″ $LaBr_3$(Ce) detector, lead shielding and a ∅44 mm × 20 mm front collimator.

The items to be measured were packed in steel drums of three different sizes, 140 cm × 25 cm, 77 cm × 40 cm and 104 cm × 49 cm (height × diameter), which were filled with UO_3 powder from spent fuel reprocessing. The ^{232}U content varied from 0.74–2.6 ppb (set #1) and 0.09–1.2 ppb (set #2). The residual ^{235}U enrichment ranged from 0.69–0.98 wt% (set #1) and 0.71–2.50 wt% (set #2). Most (91.5%) of the items were enriched ≤1.5 wt%.

The measurements were performed following IAEA standard procedures that included equipment and measurement geometry setup, background assessment, determination of container wall thickness using an ultrasonic thickness gauge and spectrum acquisition under “infinite thickness” conditions. The measured container wall thicknesses typically ranged from 4 to 5 mm. The spectrum measurement live times were LT = 1000 s (set #1) and LT = 200 s (set #2).

The results of the tests are presented in Figure 5. The data shows relative deviations of the measured enrichments from their reference values for two cases: with and without the backscatter peak included in the ^{232}U profile. It can be seen that inclusion of the backscatter peak leads to a dramatic improvement of the measurement performance with regards to both removal of the systematic bias and reduction of the random variation of data points around their reference values. The numerical data are presented in Table 3.

To investigate the reliability and consistency of spectrum analysis using the modified GEM code, two additional subsets of test spectra were generated from the set #1 spectra. Each subset consisted of 170 spectra, statistically downgraded to shorter live times: LT = 300 s and LT = 100 s. Figure 6 shows the results of the spectrum analysis, which demonstrate the stability of the fitting algorithm and the impressive improvement in measurement performance.

A comparison of spectrum fitting with and without the backscatter peak option is presented in Figure 7. The three cases illustrate the peculiarities of spectrum analysis at low (0.14 ppb), medium (1.1 ppb) and high (1.8 ppb) ^{232}U content. It is noteworthy that enabling the backscatter peak option only slightly improves the quality of spectrum fitting for materials with medium and large ^{232}U content, which correlates with the minor decrease in the normalized residual sum of squares (NQFit) shown

in the graphs. At the same time, the derived enrichment for these materials exhibits a significant change and much better agreement with the reference values.

Table 3. Sample average (bias) and sample relative standard deviation (RSD) of the relative differences between measured and reference ^{235}U enrichment values.

Measurement data	Set #1		Set #2	
Backscatter peak	Off	On	Off	On
Bias, %	-18.2	-0.01	-4.5	0.9
RSD, %	7.2	1.7	4.9	3.0

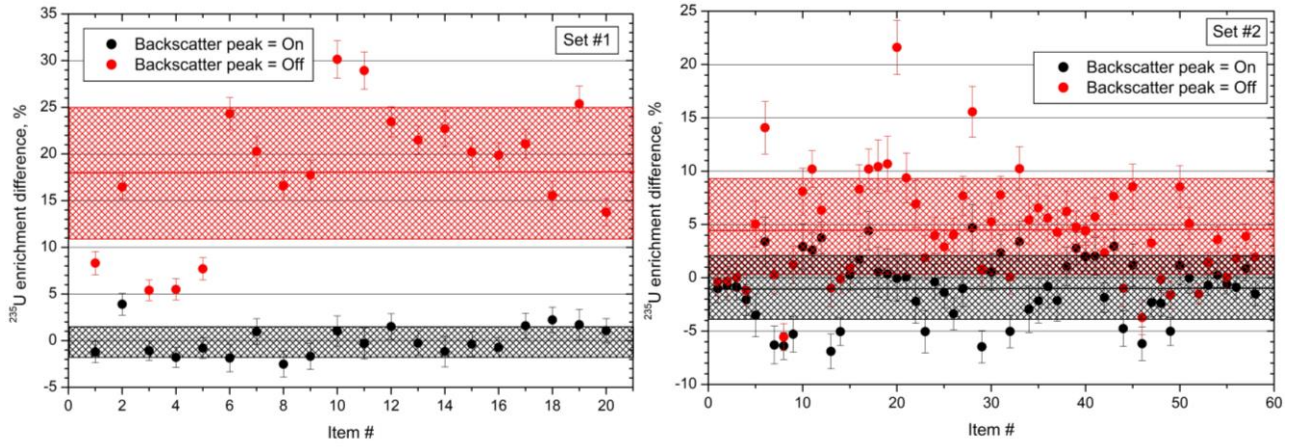

Figure 5. Relative deviation between measured and reference ^{235}U enrichment levels for set #1 (left) and set #2 (right) measurement data. Red and black horizontal lines indicate sample average deviation (bias). Shaded areas show the 68% confidence interval (relative standard deviation) around the average values.

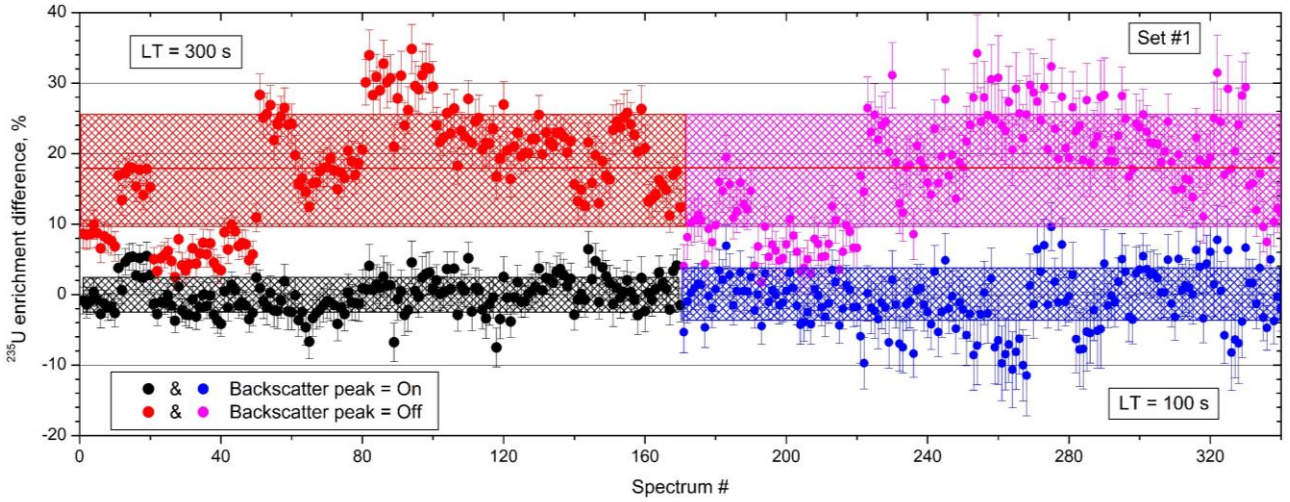

Figure 6. Results of processing two subsets of statistically downgraded spectra generated from measurement set #1. Data points show relative deviation between measured and reference enrichment levels. Red and black horizontal lines indicate sample average deviation (bias). Shaded areas show the 68% confidence interval (relative standard deviation) around the average values.

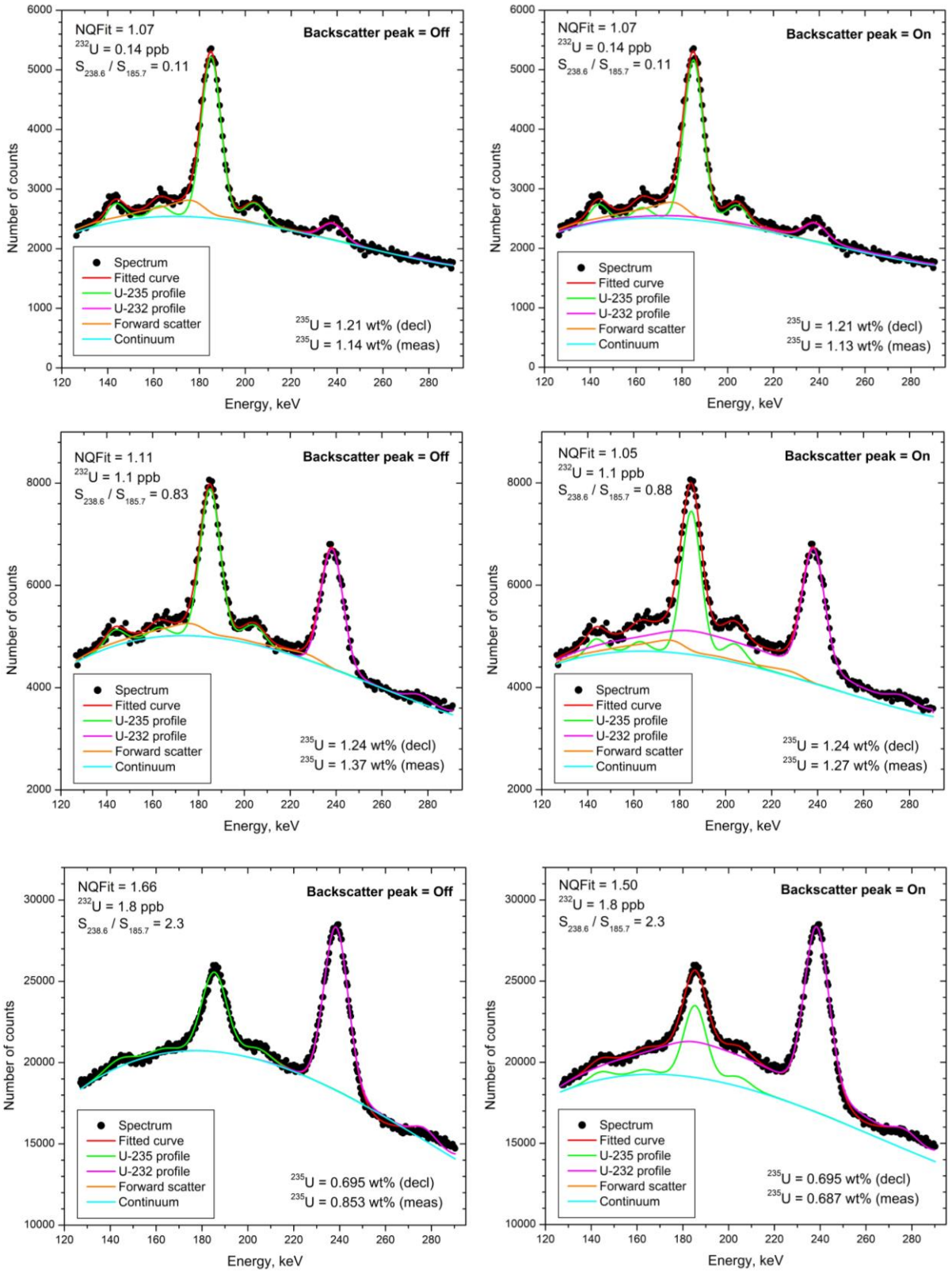

Figure 7. Spectrum fitting graphs for RepU materials with different ^{232}U content: 0.14 ppb (top), 1.1 ppb (middle), 1.8 ppb (bottom). The graphs in the left and right columns show same spectra fitted without and with the backscatter peak option, respectively.

6. Conclusions

The gamma enrichment measurements (GEM) code was upgraded to improve the performance of ^{235}U enrichment determination for RepU materials. It was shown that inclusion of the backscatter peak continuum of high-energy gamma rays emitted in the ^{232}U decay chain removes a substantial bias and decreases the random scatter of the measurement results. The upgraded GEM version 2.3.2 software delivers unbiased enrichment results with a combined RSD = 1.7% (LT = 1000 s) and RSD = 3% (LT = 200 s) in the tested range from 0.09 to 2.6 ppb of ^{232}U content. The improvement in the measurement uncertainty is especially noticeable for RepU materials containing ≥0.5 ppb of ^{232}U.

On a general note, the "infinite thickness" measurements of ^{235}U enrichment in RepU considered in this paper represent a rather unique application where the use of a higher energy resolution gamma spectrometer would be potentially beneficial, as it could allow an accurate delineation of continuum background under the critical spectral region near the 185.7 keV peak. The results presented herein demonstrate that the medium-resolution $LaBr_3(Ce)$ detectors in combination with the physics-based models for interpreting spectral data can still deliver unbiased results for RepU. Moreover, due to their high efficiency and extremely low dead time (characteristics not readily provided by the high-resolution portable gamma spectrometry systems), the $LaBr_3(Ce)$ detectors provide excellent measurement precision within short measurement times, making them more suitable for field use.

7. References

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[1] As a rule of thumb: for 1% enriched RepU containing 1 ppb of ^{232}U in radioactive equilibrium with its decay daughters, the 238.6 keV (^{212}Pb) and 185.7 keV (^{235}U) peak intensities should be nearly the same (see Figure 1). Because of ^{228}Th (the decay progeny of ^{232}U with the longest half-life, 1.9 years), the radioactive equilibrium is reached about 10 years after the chemical separation of uranium from spent nuclear fuel.

[2] Due to the unknown state of the radioactive equilibrium in the ^{232}U decay chain and since all measurable gamma emissions are produced by short-lived isotopes following the decay of ^{228}Th, it is more accurate to refer to ^{228}Th rather than ^{232}U.