

Optimization and Validation of the Developed Uranium Isotopic Analysis Code

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1. Introduction

γ -ray spectroscopy is a representative non-destructive assay for nuclear material, and less time-consuming and less expensive than the destructive analysis method. The destructive technique is more precise than NDA technique, however, there is some correction algorithm which can improve the performance of γ -spectroscopy. For this reason, an analysis code for uranium isotopic analysis is developed by Applied Nuclear Physics Group in Seoul National University [1]. Overlapped γ - and x-ray peaks in the 89-101 keV X_{α} -region are fitted with Gaussian and Lorentzian distribution peak functions, tail and background functions. In this study, optimizations for the full-energy peak efficiency calibration and fitting parameters of peak tail and background are performed, and validated with 24 hour acquisition of CRM uranium samples.

2. Parameter optimization

In the previous study, the full-energy peak efficiency of interested peaks in the 89-101 keV X_{α} -region was determined by 4th order polynomial function calibrated by using 59-391 keV γ -ray peaks in the standard source spectrum or 84-205 keV γ -ray peaks in the uranium spectrum [1] as shown in figure 1. The efficiency in 89-101 keV X_{α} -region is affected by the efficiency data points in high- and low-energy region, however, the full-energy peak efficiency varies slightly in the region. Hence, the full-energy peak efficiency is determined with a 2nd order polynomial function calibrated by using efficiency data points near the 89-101 keV X_{α} -region.

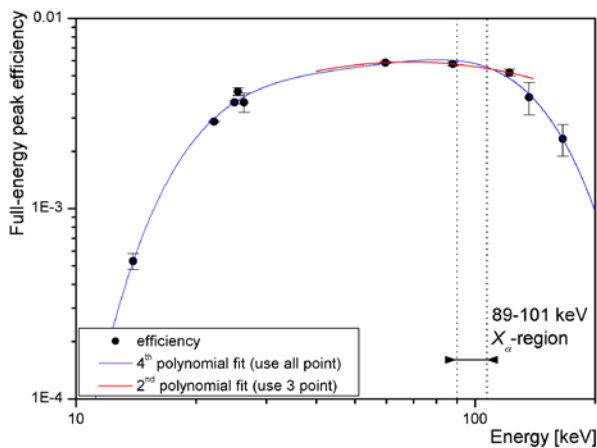


Figure 1. The full-energy peak efficiency calibration curves of the planar HPGe detector.

Uranium samples have a lot of types of geometry, density and chemical forms, then, it is difficult to prepare the same type of the analysis sample for the full-energy peak efficiency calibration. Hence, coefficients of the polynomial calibration function can be searched for optimum values simultaneously with peak fitting parameters in the spectrum fitting procedure. The function for the energy-efficiency calibration is

$$\log \epsilon = P_1 \times (\log E - P_2)^2 + P_3 \quad (1)$$

where ϵ is a detection efficiency, P_n are the coefficients, E is a γ -ray energy. Boundary conditions of P_n is set to the generic efficiency calibration curve which decreases and has a concave down shape in the 89-101 keV energy region [2-4].

All fitting parameters of peak tail and background were searched for optimum values, and the enrichment of CRM uranium samples (²³⁵U enrichment 2-80%) detected for 1 hour was analyzed in 8% precision in the previous study [x]. In the analysis of 97% enriched uranium sample, however, the area of γ - and x-ray peaks of ²³⁸U becomes zero and ²³⁵U enrichment is calculated to 100% because the curvature of the fitted background function is too high to eliminate the γ - and x-ray peaks of ²³⁸U. In the analysis of LEU samples, the curvature of the background function and the amplitude of peak tails is almost zero as shown in figure 2. These optimum values are applied to the spectrum fitting procedure in this study.

Table 1. The isotopic characteristics of CRM uranium samples.

Code	²³⁴ U (%)	²³⁵ U (%)	²³⁶ U (%)	²³⁸ U (%)
CRM-U010	0.00532	0.9911	0.00675	98.997
CRM-U030	0.0187	3.009	0.0202	96.953
CRM-U050	0.0275	4.949	0.0476	94.975
CRM-U100	0.0666	10.075	0.0376	89.821
CRM-U200	0.1229	19.811	0.2103	79.856
CRM-U500	0.5126	49.383	0.0754	50.029
CRM-U850	0.6399	84.988	0.3713	14.001
CRM-U970	1.6582	97.663	0.1497	0.5296

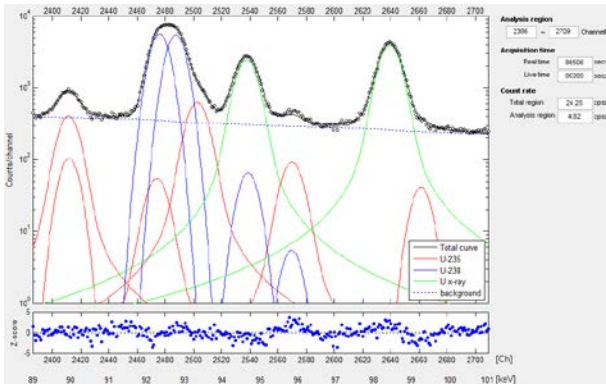


Fig. 2. 89-101 keV region of uranium spectrum for a 1% enriched uranium CRM sample.

4. Validation

Certified reference material (CRM) uranium samples with ^{235}U enrichment of 1-97 mass% are used in this study. Samples contain 1 g of highly purified U_3O_8 powder encased in a 1.5-dram glass bottle. Front wall thickness of the container is 1mm and the source is inserted into 1 mm-thick PE carrier. The characteristics of CRM uranium samples determined in New Brunswick Laboratory by using a thermal ionization mass spectrometer equipped with an ion-multiplier detection system are shown in Table 1.

An ORTEC planar HPGe detector GLP-36360 is used for measurements. The detector has a Be window of 0.25 mm thick and shielded by a low background lead shield. CRM uranium samples are mounted in front of HPGe detector, and source-to-detector distance is 5 cm. All measurements were made with the sample for 24 hours counting.

Fig. 3 shows the measurement results analyzed by using the developed code and a commercial analysis code MGAU. In MGAU analysis, results of 1-85% uranium samples are biased only about 0.8%, however, 97% uranium sample is analyzed to 94.421% with poor statistics of ^{238}U peaks. In the developed code analysis, the results are biased about 8% at 50% and 85% enriched samples, and the 97% enriched uranium sample is analyzed to 100% ^{235}U enrichment before the optimization of tail and background fitting parameters. The bias of analysis results are improved with the fixed tail and background parameters, however, low enrichment samples are still underestimated about 2%.

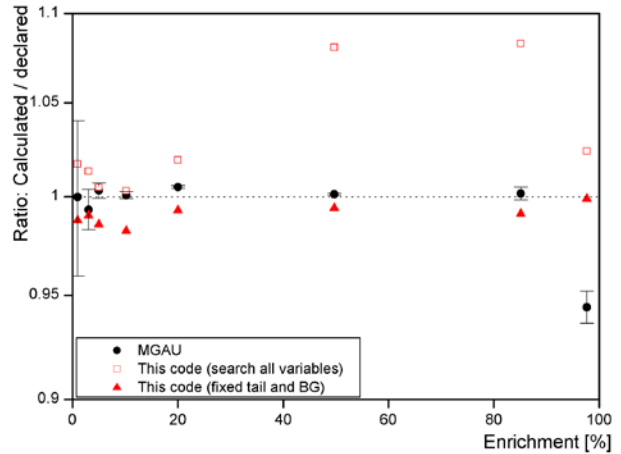


Figure 3. Measurement bias ratio: measured/declared values.

Acknowledgements

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5. Conclusion and further works

The optimization of peak tail and background parameters are performed with the validation by using CRM uranium samples. The analysis performance is improved in HEU samples, but more optimization of fitting parameters is required in LEU sample analysis.

In the future, the optimization research about the fitting parameters with various type of uranium samples will be performed. ^{234}U isotopic analysis algorithms and correction algorithms (coincidence effect, self-attenuation effect) will be developed.

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