Modification of the Developed Code for Isotopic Analysis of Uranium

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1. Introduction

To measure and determine the activities of radioactive samples by using γ -ray measurement, certified radioactive sources of known geometry and energies of radiations are required to determine the energy-efficiency calibration curve. In case of nuclear material, however, there are various types of chemical composition with different phase and containments, hence, it is hard to find the reference radioactive source for every case.

The isotopic analysis code for uranium – being developed by the Applied Nuclear Physics Group in Seoul National University – applied certified standard radioactive source to calibrate energy-fwhm and energy-efficiency before [1]. In this study, the isotopic analysis results calibrated by using peak data of the uranium spectrum itself is discussed.

2. Material and equipment

Certified reference material (CRM) uranium samples with 235 U enrichment of 2~80 mass% as shown in Fig. 1 (a) is used in the study. Samples contain 1 g of highly purified U₃O₈ powder encased in a 1.5-dram glass bottle with internal dimensions 20 mmØ. Front wall thickness of the container is 1 mm 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.

The radiation source which was used for calibration is a nuclide mixed gamma standard. Radioactive isotopes – ²⁴¹Am, ⁵⁷Co, ¹³⁹Ce, ²⁰³Hg, ¹¹³Sn – are dissolved in 20 ml HCl acid.

Table 1. The isotopic characteristics of CRM uranium samples.

Code	²³⁴ U (%)	²³⁵ U (%)	²³⁶ U (%)	²³⁸ U (%)
CRM-U020	0.01703	2.0011	0.01169	97.9702
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-U800	0.6519	80.088	0.2450	19.015

An ORTEC planar HPGe detector GLP-36360 (S=1000 mm², d=13 mm, FWHM=585 eV at 122 keV) is used for measurements. The detector has a Be window of 0.25 mm thick and shielded by a low background lead shield (Pb 100 mm, Cu 0.9 mm, tin 0.5 mm) as shown in Fig. 1 (b). 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 an hour counting.

3. Modification and results

 γ -ray peaks of ²³⁵U (144, 163, 186, 202, 205 keV) and ²³¹Th (84 keV) in the γ -ray spectrum of CRM uranium sample are used for energy-efficiency calibrations. These peaks are not overlapped with other γ - and x-ray peaks, and strong enough to detect for all measured CRM uranium samples (2~80% enrichment). The function for the energy-efficiency calibration is

$$\log \epsilon = A_0 + A_1 \log E + A_2 \log^2 E + A_3 \log^3 E \quad (1)$$

where ϵ is detection efficiency, A_n are the coefficients, E is γ -ray energy.

The peak resolution of HPGe spectrum is varying with γ -ray energy [2]. The function for the energy-fwhm calibration is

$$fwhm = \sqrt{C_1 + C_2 E + C_3 E^2}$$
(2)

where *E* is γ -ray energy, C_n are the coefficients [2,3]. The C_n are determined by least square fit for each spectrum.

Fig. 2 shows the measurement results obtained by using intrinsic calibrations. Measurement biases are improved after applying the intrinsic calibrations. The level of accuracy is still similar with MGAU code.



Figure 1. CRM uranium sample and carrier (a) and HPGe detector (b).

4. Conclusion and further works

The intrinsic calibration method is applied to the developing code for uranium isotopic analysis, and the measurement bias is improved. Difference in volume and geometry of sources effects the calibration curves.

In the future, an uncertainty calculation will be handled, and correction algorithms (coincidence effect, attenuation effect) will be developed.



Figure 2. Measurement bias ratio: measured/declared values.

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