DETERMINATION OF URANIUM ISOTOPIC COMPOSITION VIA THE PEAK RATIO METHOD USING HIGH-RESOLUTION GAMMA-RAY SPECTROSCOPY

I.O. Afanasieva[1,2], S.N. Afanasiev[1], D.Y. Lobantsov[2]
[1]NSC "Kharkiv Institute of Physics and Technology", Kharkiv, Ukraine;
[2]V.N. Karazin Kharkiv National University, Kharkiv, Ukraine
E-mail: afaninna@i.ua

This work proposes a procedure for calculating the enrichment and isotopic composition of uranium samples using the peak ratio method in the 120...260 keV gamma-ray energy range. The calculation utilized the measured intensities of the ^{234}U and ^{238}U gamma lines at energies of 120.9 and 258.3 keV, respectively. The ^{235}U intensities for these specific energies were determined by extrapolating the fitting results of five of its lines measured within the 142...242 keV range. To automate the determination of uranium isotopic composition and enrichment, an open-source graphical application was developed.

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Gamma-ray spectrometry is advantageous primarily for three reasons: 1) it requires a relatively minimal amount of equipment to perform measurements; 2) it involves virtually no complex sample preparation; and 3) it allows for 'bulk' rather than surface analysis. Furthermore, the gamma-spectroscopic method is a passive, non-destructive technique compared to destructive radioanalytical methods, such as alpha-spectrometry or mass spectrometry. It is particularly suitable for cases where the studied objects are enclosed in containers and cannot be opened or sampled.

Currently, the isotopic composition of uranium is determined from gamma-ray spectrometry measurements using commercial software products, such as MGAU (Multi-Group Analysis for Uranium) and FRAM (Fixed-energy Response-function Analysis with Multiple efficiency), which were developed at U.S. National Laboratories (LLNL, LANL) [1, 2]. As a result of processing the gamma-spectrometric data, the software calculates the content of ^{234}U, ^{235}U and ^{238}U isotopes. A significant disadvantage of commercial software packages is their high cost and the inability to modify them independently. Therefore, the development of alternative, open-source methods for processing gamma-ray spectrometry measurements remains a highly relevant task.

This work presents the results of gamma-ray spectra analysis for uranium samples, obtained using a custom-developed graphical application. The software incorporates the necessary procedures and mathematical algorithms for determining the isotopic composition of uranium samples via the peak ratio method in the 120…260 keV energy range. The gamma-ray spectra of the uranium samples were acquired at the NSC KIPT using a BEGe 3830 Broad Energy High-Purity Germanium detector with an active area of 3800 mm² and a thickness of 30 mm. The detector is equipped with a 0.6 mm thick carbon composite entrance window. The energy resolution (FWHM) of the detector is 720 eV at 122 keV.

Uranium enrichment, or the relative ^{235}U content (%), is defined as [3, 4]:

$$\eta_{235U} = \frac{m_{235U}}{m_{234U} + m_{235U} + m_{238U}} \cdot 100\%, \qquad (1)$$

where m_{234U}, m_{235U} i m_{238U} – are the masses of the ^{234}U, ^{235}U and ^{238}U isotopes, respectively.

The activity of a uranium isotope is directly related to its mass as $A = \lambda N$, where λ – is the decay constant of the isotope, related to the half-life as $\lambda = \ln 2/T_{1/2}$, and N is the number of nuclei, calculated as $N = mN_A/\mu$ (where m is the mass of the isotope in the sample, N_A is Avogadro's number, and μ is the atomic mass of the isotope). Taking all this into account, equation (1) can be rewritten as:

$$\eta_{235U} = \frac{1}{0.00035\frac{A_{234U}}{A_{235U}} + 1 + 6.43\frac{A_{238U}}{A_{235U}}} \cdot 100\%. \qquad (2)$$

Thus, uranium enrichment can be calculated after determining the activity ratios $A(^{234}\text{U})/A(^{235}\text{U})$ and $A(^{238}\text{U})/A(^{235}\text{U})$.

The activity of an isotope in a sample is defined as $A = S/(I_\gamma \cdot \varepsilon^{abs})$, where S is the net area of the selected photopeak, I_γ is the gamma-ray emission probability (the number of gamma rays emitted per 100 decays), and ε^{abs} is the absolute detection efficiency. The primary challenge in measuring absolute isotope activities is determining the value of ε^{abs}, which depends on numerous factors (gamma-ray energy, detector and sample characteristics, geometry, shielding, etc.). When determining the activity ratios of isotopes, this problem can be avoided as ε^{abs} cancels out. This technique is known as 'intrinsic calibration' [5], where all information necessary for the analysis is obtained directly from the measured gamma-ray spectrum. However, it should be noted that ε^{abs} cancels out effectively only if the energies of the selected peaks are sufficiently close to each other, or if a relative efficiency function is employed to account for the energy dependence of the detector response.

Table 1 presents the characteristics of the uranium isotope gamma lines (1st column) used for calculating the isotopic composition via the peak ratio method; the 2nd column shows the gamma-ray energy (E), and the 3rd column lists the gamma-ray emission intensity (I_γ, expressed as the number of gamma-rays emitted per 100 decays). In the 120...260 keV range, only one line is observed for each of the ^{234}U and ^{238}U isotopes.

The uranium enrichment and isotopic composition were determined using a custom-developed Python application based on the Tkinter graphical library. The

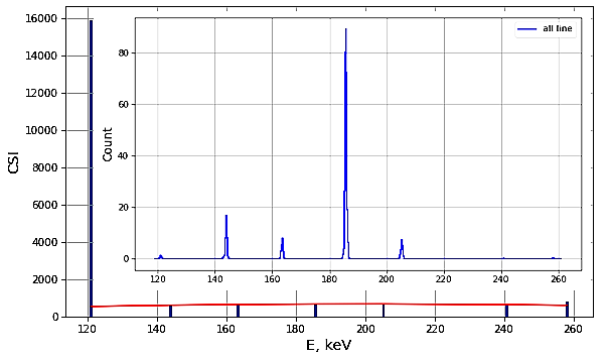
net peak area (S) for each selected line was calculated by Gaussian fitting, accounting for the background contribution.

Table 1

The characteristics of the uranium isotope gamma lines

	E, keV	I_γ
^{234}U	120.9	$3.42\cdot10^{-4}$
^{235}U	143.8	0.1096
	163.4	0.0508
	185.7	0.572
	205.3	0.0501
	240.9	$7.4\cdot10^{-4}$
^{238}U	258.3	$7.54\cdot10^{-4}$

Figure (inset) shows the gamma-ray spectrum in the 110...270 keV energy range, covering all analytical lines of interest. The main plot illustrates the energy dependence of the parameter CSI = S/I_γ which correlates with the activity A.



Fragment of the gamma-ray spectrum utilized for enrichment and isotopic composition analysis

The ^{235}U data points were fitted with a second-order polynomial (solid red line in Fugure). The CSI values for ^{235}U at 120.9 and 258.3 keV were determined by extrapolating the fit results. The activity ratios $A(_{234}\text{U})/A(^{235}\text{U})$ at 120.9 keV and $A(^{238}\text{U})/A(^{235}\text{U})$ at 258.3 keV were then calculated at these respective energy points.

The relative contributions of ^{234}U and ^{238}U isotopes (η_{234U} and η_{238U}, respectively) were determined by accounting for their specific activities A^{spec}:

$\eta_{234U} = A(^{234}\text{U})/A(^{235}\text{U})\ \eta_{235U}\ (A^{spec}{}_{235U}/\ A^{spec}{}_{234U})$,

$\eta_{238U} = A(^{238}\text{U})/A(^{235}\text{U})\ \eta_{235U}\ (A^{spec}{}_{235U}\ /\ A^{spec}{}_{238U})$,

where $A^{spec}{}_{234U} = 231.2\cdot10^6$ Bq/g, $A^{spec}{}_{235U} = 79.99\cdot10^3$ Bq/g and $A^{spec}{}_{238U} = 12.44\cdot10^3$ Bq/g [6].

Table 2 provides a comparison of the enrichment values (η_{235U} and the relative ^{234}U isotope contribution (η_{234U}) calculated by commercial software packages and the custom-developed application. Low-enriched uranium (LEU) samples with an enrichment range of 2.0 to 10.0% were selected as test specimens. The gamma-ray spectra were acquired over a live time of 734400 s.

As follows from the table, the ^{235}U enrichment data obtained by the three programs are in good qualitative agreement, despite some numerical inconsistencies. Regarding the ^{234}U content, the values calculated by FRAM differ slightly from those obtained by MGAU and the custom application. Furthermore, it should be noted that the values generated by our software exhibit higher uncertainty, which presents an objective for further refinement and optimization.

In conclusion, this study proposes a methodology for calculating the enrichment and isotopic composition of uranium samples using the peak ratio method within the 120...260 keV gamma-ray energy range. The obtained results were compared with those from specialized commercial software packages, demonstrating that the calculated data are in good qualitative agreement.

Table 2

Relative contributions of ^{235}U (η_{235U} and ^{234}U (η_{234U}) isotopes obtained using different software packages

Збагачення, %	η	MGAU	FRAM	Our_app
2	^{235}U	1.98±0.01	2.03±0.01	2.01±0.04
	^{234}U	0.0174±0.0002	0.0216±0.0002	0.0178±0.0005
4	^{235}U	3.97±0.02	3.90±0.02	4.08±0.06
	^{234}U	0.0372±0.0003	0.0439±0.0004	0.0379±0.0006
6	^{235}U	5.98±0.03	5.96±0.03	6.06±0.07
	^{234}U	0.0572±0.0005	0.0679±0.0005	0.0579±0.0008
10	^{235}U	9.99±0.05	9.74±0.04	10.04±0.08
	^{234}U	0.0967±0.0009	0.1124±0.0008	0.0959±0.002

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ВИЗНАЧЕННЯ ІЗОТОПНОГО СКЛАДУ УРАНУ МЕТОДОМ ПІКОВИХ СПІВВІДНОШЕНЬ ІЗ ВИКОРИСТАННЯМ ГАММА-СПЕКТРОСКОПІЇ ВИСОКОЇ РОЗДІЛЬНОЇ ЗДАТНОСТІ

І.О. Афанасьєва, С.М. Афанасьєв, Д.Ю. Лобанцов

Запропоновано методику розрахунку збагачення та ізотопного складу уранових зразків із використанням методу пікових співвідношень в області енергій гамма-квантів 120…260 кеВ. Для розрахунку використовувались виміряні інтенсивності гамма-ліній ^{234}U і ^{238}U при енергіях 120,9 і 258,3 кеВ відповідно. Інтенсивності ^{235}U для цих енергій вираховувалися шляхом екстраполяції результатів фітування інтенсивностей п’яти його ліній, виміряних в області енергій 142…242 кеВ. Для автоматизації процесу визначення ізотопного складу та збагачення урану створено графічний додаток з відкритим кодом.