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APPLICATION OF A COAXIAL HPGe DETECTOR AND FRAM CODE FOR DETERMINING THE ENRICHMENT OF SHIELDED URANIUM SAMPLES

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Abstract. *The determination of uranium enrichment is a critical aspect of nuclear material control, essential for assessing potential safety threats and detecting unauthorized material transfers. Gamma spectrometry using a High-Purity Germanium detector offers a non-destructive method for isotope ratio analysis. However, standard equipment often lacks precision, particularly when samples are shielded. This study aims to evaluate the capability of a portable coaxial High-Purity Germanium detector in conjunction with the FRAM software for determining uranium enrichment under both shielded and unshielded conditions. To achieve this goal, a series of gamma spectrometric measurements was performed on fuel rod samples with low uranium content. The results demonstrated that the method provides reliable and rapid enrichment estimation with an error margin of up to 10%, which is particularly significant for nuclear forensics, where timely and accurate isotope analysis is crucial.*

Keywords: nuclear forensics, uranium enrichment, shielded source, gamma spectrometry, coaxial High-Purity Germanium detector, FRAM software.

1. Introduction

Determining the isotope ratio of uranium (enrichment) is one of the key characteristics of nuclear materials, requiring prompt and accurate assessment. Such evaluations are essential for decision-making regarding whether the material poses a threat to national security and for identifying instances of unauthorized material transfers. In cases where the threat is directly associated with nuclear materials, such as enriched uranium compounds, the National Nuclear Center of the Republic of Kazakhstan (NNC RK) can provide substantial technical support through preliminary analysis of material characteristics using gamma spectrometry. It is well known that equipment used during initial inspections often lacks sufficient precision to determine the isotope ratio (e.g., the degree of enrichment or uranium type). A solution to this issue could be a non-standard gamma spectrometry method requiring the use of a high-sensitivity detector based on High-Purity Germanium (HPGe), which provides high resolution [1–3].

The NNC RK laboratory staff has extensive experience using a coaxial HPGe detector in various studies. This detector has been employed for neutron activation analysis and for measuring gamma-emitting radionuclides, such as fission and activation products, in the coolant of the IVG.1M reactor. These studies have effectively determined the composition and concentrations of radionuclides in various samples [4–6].

Gamma spectrometry is a non-destructive analytical method that can be used to measure the enrichment of nuclear materials and obtain rapid and satisfactory results on the isotopic composition of uranium or

plutonium in unknown samples. This is particularly important in nuclear forensics, where prompt provision of data on the isotopic composition of nuclear materials is required. When nuclear material is placed in a protective container (e.g., active samples or spent nuclear fuel), gamma lines in the low-energy range (90–100 keV) may not be detected by the detector. Namely, this range of photon energies is used to determine uranium enrichment by the multigroup analysis of uranium (MGAU) method. Under such conditions, calculation algorithms using the MGAU code [7] become ineffective for enrichment assessment. To solve this problem, it is advisable to use a coaxial HPGe detector in combination with the FRAM code for spectral analysis. A feature of the coaxial HPGe detector compared to the planar detector is a higher efficiency of photon registration in the high energy range of 1.2 MeV. In our work, a coaxial HPGe detector was used in tandem with the FRAM program to determine uranium enrichment. The FRAM code, unlike the MGAU program, uses gamma lines with energies in the range from 121 to 1001 keV to determine uranium enrichment, which can penetrate steel shielding up to 10 mm thick [8,9].

This study aimed to investigate the feasibility of applying gamma spectrometry to determine the enrichment of uranium samples, both unshielded and shielded with steel of varying thickness, using a portable coaxial HPGe detector and the FRAM software.

2. Methodology and equipment

To determine the enrichment of ^{235}U , eight segments of fuel rods from water-cooled technological channels with low-enriched uranium (WCTC-LEU) of the IVG.1M reactor were provided. These WCTC-LEU fuel rods, measuring 49 to 60 mm in length, are hereinafter referred to as "samples." The samples are spiral rods with a two-blade profile, consisting of a metallurgically bonded cladding and a fuel core. The fuel core comprises a zirconium alloy E110 matrix with evenly distributed uranium filaments enriched to 19.75% in ^{235}U . The appearance of the WCTC-LEU sample is shown in Figure 1.

The declared uranium enrichment in ^{235}U is 19.75%, while the ^{235}U content in the sample is 2.18% by mass, equivalent to 0.23 g. Such low uranium content complicates the enrichment determination process due to reduced gamma radiation activity, which decreases the intensity of the detectable lines, making accurate isotopic analysis challenging. To minimize errors and improve measurement accuracy, various measurement geometries were employed to assess the effects of shielding and the distance between the sample and the detector. The parameters of these measurements are summarized in Table 1.

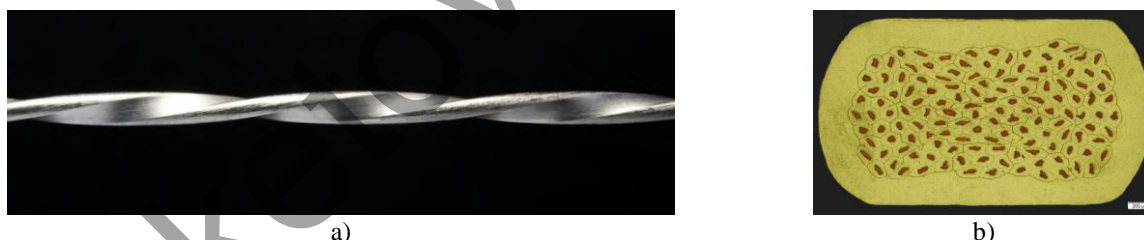


Fig.1. The appearance of the WCTC-LEU fuel rod sample (a) and cross-sectional view of the WCTC-LEU fuel rod (b).

The research methodology involved measuring the gamma spectra of the sample in four different geometries. In three geometries, the distance from the sample to the detector cap was fixed at 16 mm. The difference between these geometries was as follows: the first measurement was conducted without shielding; the second used a steel shield with a thickness of 2.5 mm; and the third used a steel shield with a thickness of 8 mm. In the fourth geometry, the sample was placed directly on the detector cap. The measurement duration was adjusted so that the peak area at energy $E = 1001$ keV was approximately $4.3 \cdot 10^3$ counts for all measurements (statistical uncertainty was estimated at 1.6–1.8%). The shield for the second geometry consisted of a steel plate measuring $90 \times 30 \times 2.5$ mm, and for the third geometry, a steel plate measuring $80 \times 40 \times 8$ mm. Additionally, background measurements were conducted in the laboratory, and no uranium isotope peaks were detected in the background spectrum.

Table 1. Measurement Parameters

Measurement Geometry Number	Distance Detector-Sample, mm	Shield Thickness, mm	Exposure Time, s	Shield Dimensions, Length×Width, Thickness, mm
1	16	-	5000	-
2	16	2,5	5500	90×30×2,5
3	16	8	7500	80×40×8,0
4	0	-	2200	-

Spectrometric studies were conducted using the GC1518 coaxial germanium semiconductor detector, which has a gamma radiation detection efficiency of 15% and an energy resolution of 1.8 keV at 1332 keV. The measurements were carried out using the DSA-LX pulse analyzer with an 8182-channel scale. The amplification of the channel was set to 0.125 keV/channel. The energy calibration of the gamma spectrometer was performed using the Eu-152 calibration source. The measurement process and the geometry of the sample placement, both without shielding and with shielding, are shown in Figures 2a and 2b, respectively.

**Fig.2.** Geometry of sample measurement without shielding (a) and with a 2.5 mm thick shield (b).

The spectra of the samples were processed using the FRAM code. Unlike the MGAU code, the FRAM code offers several different algorithms for result processing, which can be configured through an editable parameter file. The FRAM package includes a recommended set of input parameters, which vary depending on the enrichment of the sample. These parameters include specific settings for analyzing low-enriched uranium (enrichment less than 40% for ^{235}U) and highly enriched uranium (enrichment greater than 10% for ^{235}U) [8-10].

Table 2. Standard Parameter Sets Embedded in the FRAM Code.

Parameter name	Description
ULEU_Plnr_060-250	U Only, Enrichment $\leq 40\%$, 0.075 keV/ch, Planar detector
UHEU_Plnr_060-250	U Only, Enrichment $\geq 10\%$, 0.075 keV/ch, Planar detector
ULEU_Cx_120-1001	$^{235}\text{U} < 40\%$ or high thorium background, 0.125 keV/ch, Coaxial detector
UHEU_Cx_120-1001	$^{235}\text{U} > 10\%$, 0.125 keV/ch, Coaxial detector

For the analysis of the spectra, the parameter ULEU_Cx_120-1001 was used, which is oriented for low-enriched uranium and utilizes energy peaks in the range from 121 keV to 1001 keV for a coaxial detector type.

For each measurement geometry, 18 spectra were recorded. The results of processing those spectra were subjected to statistical analysis. For each data set obtained, the mean enrichment value (Enr) was calculated using formula (1), and the standard deviation (SD) was calculated using formula (2):

$$\overline{Enr} = \frac{1}{n} \sum_{i=1}^n Enr_i, \quad (1)$$

$$SD = \left(\frac{1}{n-1} \sum_{i=1}^n (Enr_i - \overline{Enr})^2 \right)^{1/2}, \quad (2)$$

where n is the number of measurements; Enr is the enrichment value obtained from the i -th spectrum.

3. Results of the study

As a result of gamma spectrometric measurements of the sample, 72 spectra were obtained (18 for each of the four measurement geometries) and processed using the FRAM code. The average enrichment values of ^{235}U in the sample (Enr) and the standard deviation (SD) for the unshielded sample at a distance of 16 mm from the detector were 20.1% and 1.8%, respectively. For the shielded sample with a 2.5 mm thick screen, these values were 20.3% and 1.2%. For the shielded sample with an 8 mm thick screen, the values were 19.65% and 1.21%. For the unshielded sample placed directly on the detector cap, the enrichment values were 20.4% and 0.8%. Table 3 presents the statistical distributions of the results for determining the enrichment of ^{235}U for both unshielded and shielded samples.

Table 3. Results of Statistical Analysis.

Interval No.	Unshielded sample		Shielded sample, d=2.5 mm	
	Enrichment % by mass	Number of Measurements	Enrichment % by mass	Number of Measurements
1	17.10 ± 0.63	1	18.44 ± 0.45	2
2	18.36 ± 0.63	5	19.34 ± 0.45	5
3	19.62 ± 0.63	5	20.24 ± 0.45	5
4	20.88 ± 0.63	1	21.14 ± 0.45	2
5	22.15 ± 0.63	6	22.04 ± 0.45	4
	Shielded sample, d=8 mm		Unshielded sample on cap	
1	18.15 ± 0.45	3	18.94 ± 0.30	2
2	19.05 ± 0.45	5	19.55 ± 0.30	5
3	19.95 ± 0.45	5	20.16 ± 0.30	2
4	20.85 ± 0.45	3	20.77 ± 0.30	5
5	21.75 ± 0.45	2	21.38 ± 0.30	4

When constructing histograms of statistical distributions, consisting of five intervals, the interval width was calculated using the formula (3):

$$w = \frac{Enr_{max} - Enr_{min}}{5} \quad (3)$$

The values for the first and subsequent intervals were calculated using formulas (4) and (5):

$$Enr_1 = Enr_{min} + \frac{w}{2} \quad (4)$$

$$Enr_i = Enr_{i-1} + w \quad (5)$$

The statistical distributions of the ²³⁵U enrichment results for the FRAM code are also presented in histograms shown in Figure 3. The point with the error interval on the abscissa axis corresponds to the average enrichment value (Enr) and the standard deviation (SD) obtained from formulas (1) and (2).

The spectra of the sample were analyzed to study the ability of shielding materials to attenuate gamma radiation intensity at various energies. The exposure time for the measurements was selected such that the area of the 1001 keV peak was approximately 4.4 · 10³ impulses. Table 4 shows the results of measuring the peak areas of gamma radiation at different energies (121, 143, 163, 185.7, 205, and 1001 keV) in four different geometries and exposure times. Analysis of the data in the table highlights the property of the absorbing screens to attenuate the low-energy part of the gamma spectrum more significantly, as well as the ability of the coaxial detector to efficiently register high-energy gamma quanta from uranium-238 at 1001 keV, which penetrate well through shielding materials. This makes the coaxial detector especially effective for analyzing samples with a high-energy component in the gamma spectrum and those shielded by steel screens.

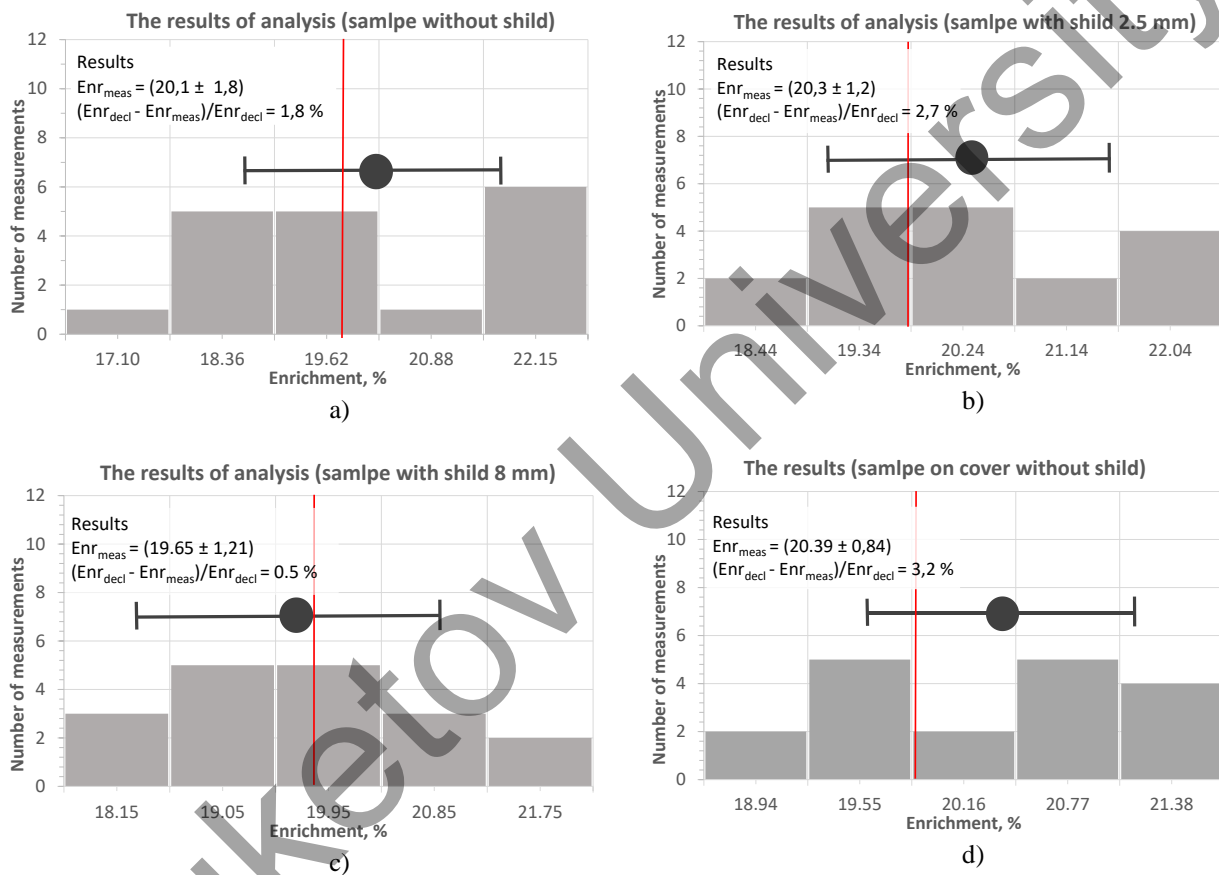


Fig.3. Statistical distribution of the results of the determination of ²³⁵U enrichment in the unshielded sample (a), the shielded sample with 2.5 mm steel (b) and 8 mm steel (c), and the sample placed on the detector lid (d).

Table 4. Results of Peak Area Measurements for Various Energies, Geometries, and Exposure Times.

Measurement Conditions	Energy, keV					
	121	143	163	185.7	205	1001
Without shield (t=2200 s)	2.74(4)E+04	3.72(6)E+05	1.87(14)E+05	2.10(2)E+06	1.78(4)E+05	4.44(9)E+03
Without shield (t=5000 s)	3.19(6)E+04	4.17(8)E+05	2.04(8)E+05	2.26(3)E+06	1.89(6)E+05	4.39(7)E+03
Shield 2.5 mm (t=5500 s)	2.10(6)E+04	3.10(7)E+05	1.61(7)E+05	1.86(2)E+06	1.59(5)E+05	4.40(7)E+03
Shield 8.0 mm (t=7500 s)	8.30(26)E+03	1.59(7)E+05	9.34(7)E+04	1.19(2)E+06	1.08(4)E+05	4.27(5)E+03

4. Discussion of results

For the FRAM code, the algorithm used for processing results was focused on low-enriched uranium, utilizing energy peaks in the range from 121 keV to 1001 keV for the coaxial detector type. It is important to note that when measuring a shielded nuclear material source, longer exposure times are required to achieve the necessary statistical accuracy of the full absorption peak area.

The study obtained statistical distributions of ^{235}U enrichment results for both unshielded and shielded samples. The distributions were divided into five intervals based on enrichment values. During the result processing, varying numbers of measurements fell into each interval, which were subsequently presented in histograms. These intervals allow for evaluating the distribution of enrichment results and how closely they align with the declared enrichment value of 19.75%.

Upon analyzing the results, it is observed that the histogram with the maximum shielding thickness and exposure time exhibits a normal distribution. The enrichment value in this case most closely matches the declared value of 19.75%. For other histograms, the distribution is uneven, likely due to an insufficient number of measurements and short exposure times. Overall, it can be stated that the results of enrichment determination using the coaxial detector are sufficiently reliable.

The count rate in the peak with energy E_i depends on the shield thickness. As shown in Table 4, gamma quanta with a "low" energy of 121 keV are attenuated to a greater extent, while gamma quanta with a "high" energy of 1001 keV are attenuated to a lesser extent. For clarity, the measurement results of the count rate as a function of steel shield thickness for various gamma-ray energies are presented in Fig. 4.

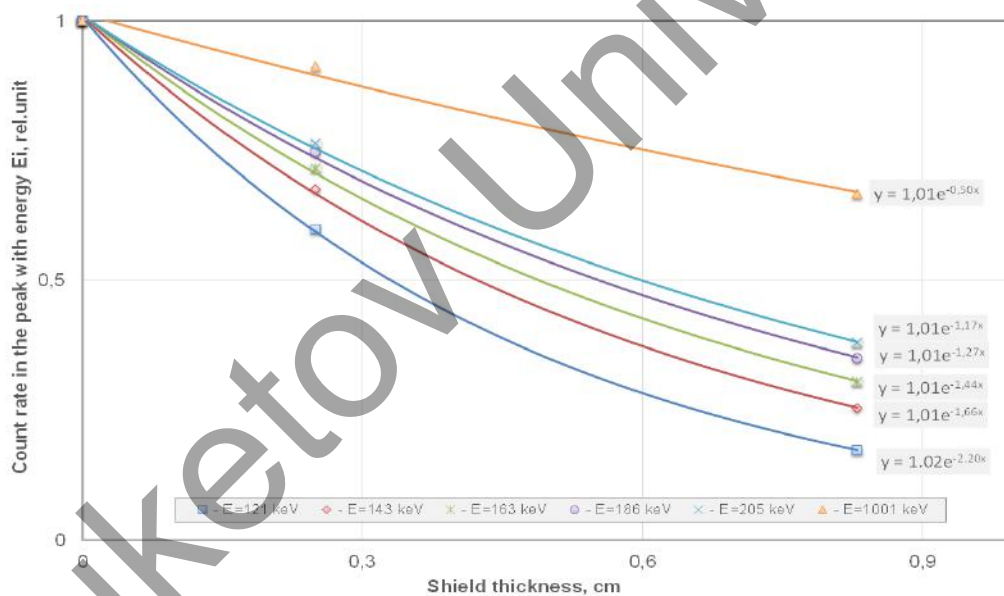


Fig.4. Results of count rate measurements as a function of steel screen thickness.

As observed, the count rate in the $E=1001$ keV peak with a 0.8 cm thick shield decreased by 1.5 times compared to the unshielded sample, while the count rate in the $E=121$ keV peak decreased by 5.8 times. This confirms the advantages of using the FRAM method compared to the MGAU method for samples shielded with thick materials. It is well-known that the MGAU method analyzes uranium enrichment using the low-energy region of the gamma spectrum in the range of 90–100 keV.

The measurements, conducted in four different geometries, indicated that the enrichment values calculated using the FRAM code had a maximum relative deviation of 3.2%. This confirms the method reliability even with changing measurement conditions such as shielding and detector distance. The use of the coaxial HPGe detector and the FRAM software code proved to be effective for analyzing uranium samples under various conditions, including shielding. The average ^{235}U enrichment value obtained from all measurements lies within the statistical error and corresponds to the declared value of 19.75%.

5. Conclusion

Gamma spectrometry measurements were performed to determine the uranium enrichment in both unshielded and shielded samples with low uranium content. Despite the low uranium content (1.2% of the total mass of 10.9 g), sufficiently reliable data on the isotopic composition of ^{235}U were obtained.

The measurements carried out in four different geometries, showed that the enrichment values calculated using the FRAM code had a maximum relative deviation of 3.2%. This confirms the method reliability even when the measurement conditions, such as shielding and detector distance, change.

The use of the coaxial HPGe detector and the FRAM software code proved effective for analyzing uranium samples in various conditions, including shielding. The average ^{235}U enrichment value, derived from all measurements, lies within the statistical error and corresponds to the declared value of 19.75%.

In perspective, gamma spectrometry using a coaxial HPGe detector with FRAM software may be an alternative method in nuclear forensics for determining uranium enrichment, where samples can be shielded (e.g. in containers with ~10 mm thick steel shielding). It allows for obtaining relatively fast and satisfactory results on the isotopic composition of uranium, which makes it an effective tool for express analysis.

Conflict of interest statement.

The authors declare that they have no conflict of interest in relation to this research, whether financial, personal, authorship or otherwise, that could affect the research and its results presented in this paper.

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Medetbekov B.: Conceptualization, Methodology, Writing Review & Editing; **Popov Yu.:** Writing- Original draft preparation, Supervision; **Sabitova R.:** Data Curation, Investigation; **Prozorova I.:** Writing- Original draft preparation and Editing; **Syssaletin A.:** Project administration. The final manuscript was read and approved by all authors.

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