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GAMMA RAY EMISSION DURING THE DECAY OF ²³⁵U
FOR THE PURPOSES OF NUCLEAR FORENSICS**

The value of absolute emission probabilities from the decay of ²³⁵U in the energy range of 200–450 keV was refined by using semiconductor gamma spectrometry with a reference material containing highly enriched uranium and mathematical modelling of the measurement geometry. This refinement is essential for the accurate gamma-spectrometric determination of characteristics (signatures) of highly enriched uranium that are crucial for nuclear forensics, such as model age and ²²⁸Th impurity. This clarification is also useful from the point of view of fundamental nuclear physics, as it introduces significant changes in the data concerning the ²³¹Th level scheme from the alpha decay of ²³⁵U.

Keywords: ²³⁵U, nuclear forensics, highly enriched uranium, model age, gamma spectrometry, absolute emission probability.

1. Introduction

The precise determination of the nuclear-physical characteristics (signatures) of uranium-bearing materials is critical for nuclear forensics and nuclear safeguards. One of the most common methods for sufficiently accurate and rapid determination of many of these signatures is high-resolution gamma spectrometry. The method is characterised by its simplicity and efficiency in determining uranium enrichment and ²³⁴U content, a process facilitated by the utilisation of the well-established MGAU or FRAM [1] software. Furthermore, the FRAM software facilitates the determination of ²²⁸Th, a daughter radionuclide of ²³²U. In the context of gamma spectrometric studies conducted within a laboratory environment, it is feasible to ascertain the model age of uranium with a reasonable degree of expediency. This is achieved without the necessity of performing radiochemical procedures. The model age of uranium, which corresponds to the cutoff date of the final separation of uranium from protactinium and thorium, can be determined for both the radionuclide pairs ²³⁵U–²²³Ra and ²³⁴U–²²⁶Ra. The model ages of the material for these pairs may not coincide with each other or with the actual date of the separation procedure if there was incomplete separation of protactinium or thorium from uranium, or in the case of a mixture of varied materials. However, from the standpoint of nuclear forensics, these time characteristics are significant signatures that are indicative of the material in question. It is evident that, given the

knowledge of the model age of the material in question and the relevant data on enrichment, ²³⁴U content, and ²²⁸Th admixture, the material can be identified and its origin established.

The calculation of model age is based on determining the activity ratios of radionuclides in the ²³⁵U and ²³⁴U decay chains. One of the main stages of the calculation is the construction of an internal relative curve of gamma-quantum detection efficiency. The most convenient analytical gamma peaks for determining the model age for the pairs ²³⁵U–²²³Ra and ²³⁴U–²²⁶Ra are the gamma peaks 269.5 keV ²²³Ra and 351.9 keV ²¹⁴Pb, respectively. The ²²⁸Th admixture can be determined by measuring the 238.6 keV ²¹²Pb gamma peak. The utility of these gamma peaks lies in two key aspects. Firstly, they exhibit high absolute emission probabilities within the relevant decay chains, which makes them particularly amenable to analysis. Secondly, the multiplets containing these gamma peaks can be reliably decomposed using contemporary gamma spectrum processing software, facilitating efficient data analysis. Furthermore, these gamma peaks do not cascade in their decay schemes, a property that renders it possible to perform measurements in “close” measurement geometries using large-volume detectors. To cover the energy range required for analysis, it is necessary to construct an internal relative curve of gamma-quantum detection efficiency in the energy range of 230–350 keV. For this purpose, it is convenient to use intense and conditionally non-cascading (provided that the 100 keV gamma peaks of the region

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are shielded with absorbers) gamma peaks of 221.4 keV, 246.8 keV, the doublet 275 keV, 291.7 keV, and 345.9 keV from the decay of ^{235}U . To construct the internal curve of relative efficiency for these gamma peaks with any degree of precision, it is first necessary to know their absolute emission probabilities with the same degree of precision. However, in modern reference tables, these values are given with large uncertainties ranging from 5 to 25 %.

The authors conducted experimental studies to refine these values. During the refinement process, it was determined that, in certain instances, the values obtained by the authors exhibited substantial discrepancies from the tabulated values for both these gamma peaks and several additional peaks from the decay of ^{235}U . These discrepancies at times exceeded one hundred percent.

The primary outcome of this study is the establishment of the refined values of absolute emission probabilities for a series of gamma peaks emanating from the decay of ^{235}U . The paper describes the research conducted and provides a comparative table of current tabulated values of absolute emission probabilities from the decay of ^{235}U in the energy range of 200–450 keV and those obtained by the authors.

2. Research methodology

The research methodology comprised the measurement of the gamma spectrum of certified high-enriched uranium material, which was characterised by well-known properties (uranium isotopic composition, mass, geometric and physicochemical characteristics). The measurement was conducted using a detector that had also been thoroughly characterised. Concurrently, mathematical modelling of the experiment was conducted using the Monte Carlo method in the MCNP [2] and Geant4 [3] environments, and a model curve of the absolute efficiency of gamma-quantum detection was constructed. An experimental internal curve of relative efficiency was also constructed using the mathematical apparatus of the FRAM program, normalized to the experimentally determined absolute detection efficiency for an energy of 185.7 keV for the given measurement geometry.

Following the processing of the measured gamma spectrum, the count rates at the total absorption peaks were divided by the activity of ^{235}U and by the calculated absolute detection efficiencies for these energies. This process resulted in the corresponding absolute emission probabilities.

3. Detector, reference material, and measurement geometry

A high-purity germanium detector manufactured by CANBERRA was used to measure the gamma

spectrum. Specifically, the BE6530 model was employed, which possesses a crystal diameter of 91 mm and a thickness of 31 mm. In addition to its relative detection efficiency of 65 %, this model demonstrates real energy resolution over a substantial energy range (FWHM is equal to 0.76 keV for 185.7 keV energy and 1.49 keV for 1.001 MeV energy).

The reference material employed in this study was a standard with 93 % enrichment for ^{235}U from the CRM-146 uranium standard set, manufactured at the New Brunswick Laboratory. The standard was selected based on 230 g of U_3O_8 powder, since it possesses well-known uranium isotope masses (with an uncertainty of less than 0.1 %), as well as the geometric dimensions, elemental composition, and densities of the radioactive material and container. These are necessary for the mathematical modelling of the measurement geometry in the MCNP and Geant4 environments. Additionally, in the investigated energy range (200–450 keV), the Compton background from high-energy gamma rays of $^{234\text{m}}\text{Pa}$ is low. This significantly reduces the statistical uncertainty in peak area determination. This, in turn, serves to reduce the statistical uncertainty of peak areas during the processing of the gamma spectrum. The activity of ^{235}U in the reference material is 14.5 MBq, whereas the activity of ^{238}U is 100 times lower.

The reference material was measured in a configuration referred to as “close”, with 4 cm from the detector crystal. This configuration was selected to ensure the maximisation of the counting rate in gamma peaks within the specified energy range. Concurrently, to reduce the counting load of the spectrometric system caused by X-rays and gamma quanta with energies in the range under 200 keV, as well as to minimise the effects of random and cascade summation, a system of filters made of lead, cadmium, and copper was placed between the detector and the reference material.

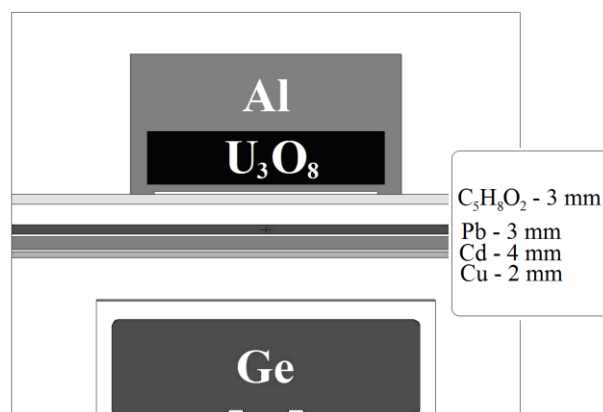


Fig. 1. Measurement geometry.

As illustrated in Fig. 1, the measurement geometry is delineated as follows. Furthermore, the

detector was shielded on the sides by a ring system of cadmium and copper filters. The purpose of this shielding was to reduce the registration of X-rays and gamma rays scattered and induced in the lead shielding of the detector.

4. Gamma spectrum measurement and processing

The gamma spectrum of the reference material was measured over a period of 10 days. The spectro-

metric system demonstrated a data upload rate of 1600 counts/s, while the dead time was recorded at 2.4 %. The energy range up to 1024 keV was studied at a conversion factor of 0.125 keV/channel. A region of the measured gamma spectrum, spanning up to 500 keV, is presented in Fig. 2. The figure uses arrows to indicate gamma peaks, which are proposed for use in constructing a curve of internal relative efficiency to determine the model age of highly enriched uranium samples.

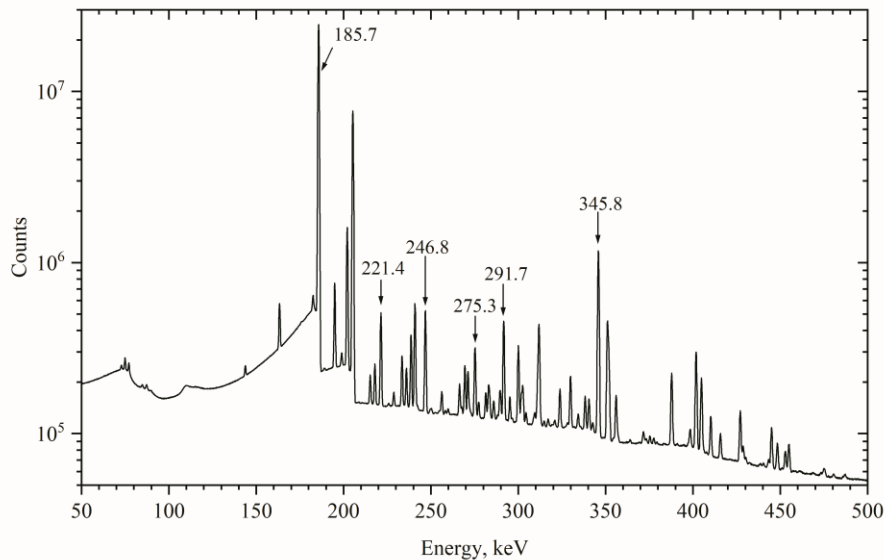


Fig. 2. Gamma spectrum of the reference material with 93 % enrichment.

5. Determination of absolute detection efficiencies and spectrum processing

Based on a mathematical model of measurement geometry in MCNP5 and Geant4 environments, the absolute efficiencies of gamma-quantum detection were calculated for transition energies in the ^{235}U level scheme in the range of 200–450 keV. The mathematical model was verified using measured values of absolute detection efficiency for well-known gamma peaks: 163.4, 185.7, 205.3 keV ^{235}U [4], and 1001 keV $^{234\text{m}}\text{Pa}$. The discrepancy between the calculated and measured values did not exceed 1 %. Furthermore, a relative efficiency curve was

constructed using the mathematical apparatus of the FRAM v5.2 program. The construction of this curve necessitated the creation of a parameter file, which incorporated the intense gamma peaks 269.4 keV, 323.8 keV, and 445.0 keV ^{223}Ra and 401.8 keV ^{219}Rn from the ^{223}Ra decay chain. This parameter file encompassed the entire range of 200–450 keV and incorporated well-known absolute emission probability values. Table 1 presents data on the absolute gamma-ray intensities used for the validation of the mathematical model (V) and for constructing the intrinsic relative efficiency curve (Eff_{rel}) using the FRAM software.

Table 1. Absolute gamma-ray intensities

Radionuclide	E_γ , keV	I_γ , %	Destination	Reference
^{235}U	163.33	5.08 4	V	[4]
	185.72	57.2 5	V, Eff_{rel}	[4]
	205.3	5.01 5	V, Eff_{rel}	[4]
$^{234\text{m}}\text{Pa}$	1001.03	0.837 10	V, Eff_{rel}	[4]
^{223}Ra	269.46	13.31 7	Eff_{rel}	[5]
	323.87	3.65 3	Eff_{rel}	[5]
	445.03	1.22 1	Eff_{rel}	[5]
^{219}Rn	401.81	6.4 2	Eff_{rel}	[4]
^{212}Pb	238.63	43.3 3	Eff_{rel}	[4]
^{212}Bi	727.33	6.58 5	Eff_{rel}	[4]

In the parameter file created for the purpose of constructing an internal curve of relative detection efficiency, the gamma peak 1001 keV ^{234m}Pa was artificially “assigned” to ^{235}U with a change in the absolute emission probability based on the known ratio of ^{235}U to ^{238}U activity in the measured reference material. The corresponding FRAM program window with the internal relative efficiency curve is shown in Fig. 3. The experimental values of relative detection efficiencies and values after fitting with an empirical theoretical curve, calculated by the FRAM program, differed by no more than 1%. As illustrated in Fig. 4, the absolute efficiencies of ^{235}U gamma-quantum detection have been calculated in the Geant4, MCNP5, and FRAM environments. In the

case of FRAM, the relative detection efficiencies were normalized to the absolute efficiency of the 185.7 keV gamma peak. The final values of the absolute efficiencies of ^{235}U gamma-quantum detection in the energy range of 200–450 keV were obtained after averaging the results, which differed from each other by no more than 1%. Using the experimental intensities of the reference gamma-ray peaks and the values of the absolute detection efficiency, we obtained the following absolute gamma-ray emission probabilities for these peaks (in percent): 57.2(6) for the 185.7 keV gamma peak, 5.03(6) for the 205.3 keV gamma peak, and 0.840(11) for the 1001 keV gamma peak.

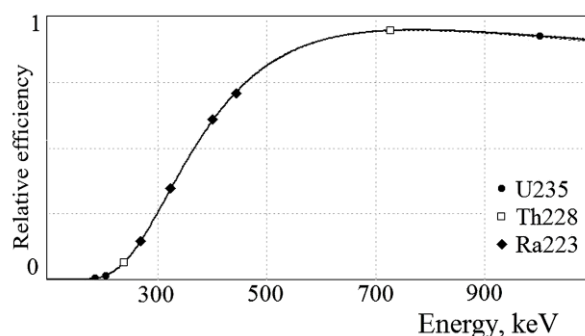


Fig. 3. Internal relative efficiency curve constructed using the FRAM v.5.2 program.

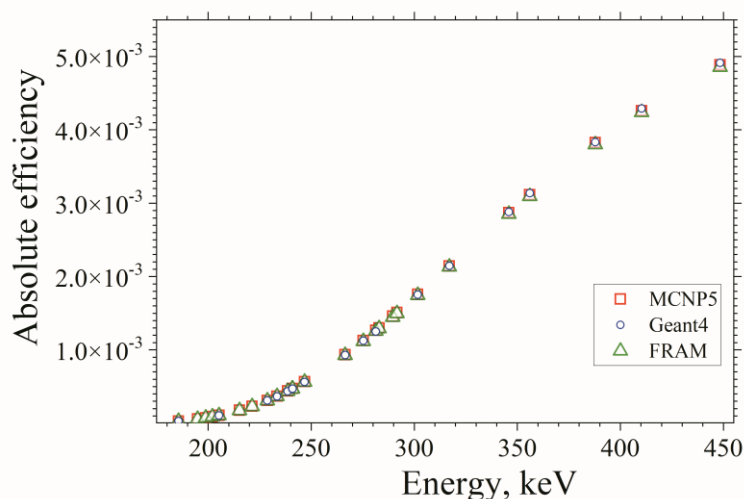


Fig. 4. Absolute efficiencies of ^{235}U gamma-quantum detection calculated in Geant4 and MCNP5 environments, as well as using the FRAM v.5.2 program. (See color Figure on the journal website.)

The processing of the gamma spectrum and the determination of absolute detection efficiencies were carried out using three independent approaches: gamma-spectrum analysis using the WinSpectrum v3.24 software [6] with subsequent calculation of absolute efficiencies in the Geant4 environment; gamma-spectrum analysis using the Interactive Peak Fit v2.1 software from the Genie 2000 package by CANBERRA, with efficiency calculations performed in the MCNP5 environment; and gamma-spectrum processing along with the determination of

the intrinsic relative efficiency curve using the FRAM v5.2 software.

During the analysis of the gamma spectrum, potential interferences from gamma lines of other radionuclides were taken into account. The effects of cascade and random summation were eliminated at the stage of measurement geometry design, except for the contribution of cascade and random summation of the 185.7 and 202.1 keV gamma peaks of ^{235}U to the intensity of the 387.8 keV gamma line. Under the given measurement conditions, their com-

binned contribution was less than 0.3 % and was therefore taken into consideration.

If, during gamma-spectrum processing, the Win-Spectrum and Interactive Peak Fit software reported peak area uncertainties of less than 0.5 %, a value of 0.5 % was adopted in subsequent calculations of total uncertainties.

Energy calibration based on a linear function does not provide sufficient accuracy for the spectrometer; therefore, a second-order polynomial calibration was employed. The most precise gamma peaks of uranium isotopes (^{235}U , ^{238}U) and their progeny were used for the calibration procedure.

Table 2. Verification of the spectrometer's energy calibration

E_γ , keV			
$^{230}\text{Th}(n, \gamma)^{231}\text{Th}$ [7]	^{223}Ra [8]	^{211}Pb [9]	Our work
185.712 2			185.715 5
198.928 3			198.921 9
202.111 3			202.109 4
205.309 2			205.310 5
228.785 6			228.790 10
233.469 3			233.466 8
240.875 3			240.868 6
281.441 9			281.443 8
448.339 18			448.334 13
	338.282 10		338.284 9
	445.033 12		445.017 13
		404.853 10	404.856 11
		427.088 10	427.091 12

For verification purposes, reference data obtained using a crystal-diffraction spectrometer for the $^{230}\text{Th}(n, \gamma)^{231}\text{Th}$ reaction were employed, along with several gamma lines from the decay of ^{223}Ra and ^{211}Pb . The comparison results are presented in Table 2.

6. Results

To calculate the values of absolute emission probabilities, the counting intensity in the gamma

peaks of ^{235}U in the measured spectrum was divided by the absolute efficiencies of ^{235}U gamma-quantum detection that had been obtained, and by the activity of ^{235}U in the measured reference material. The uncertainties of the obtained absolute emission probabilities arise mainly from statistical uncertainties in peak area determination ($\leq 1\%$) and uncertainties in the calculated absolute efficiencies ($\approx 1\%$). The calculation results are presented in Table 3.

Table 3. Refined absolute emission probabilities from the decay of ^{235}U in the energy range 200–450 keV compared with data from NDS

E_γ , keV Our work	I_γ , % Our work	I_γ , % NDS	I_γ , % Ref. [13]
194.937 6	0.632 7	0.637 13	0.625 6
198.921 9	0.0464 6	0.042 3	0.0451 13
202.109 4	1.100 12	1.07 3	1.14 6
215.258 5	0.0296 4	0.0295 20	0.0300 17
221.387 7	0.1155 13	0.115 6	0.115 5
228.790 10	0.00690 9	0.0069 10	0.0069 4
233.466 8	0.0280 3	0.0332 23	0.0283 17
240.868 6	0.0658 8	0.068 4	0.065 7
246.838 4	0.0522 6	0.051 3	0.0511 16
266.421 7	0.00556 6	0.0066 10	0.00547 15
275.3*	0.01448 16	0.032 8	0.0145 4
275.11 2	0.0062 4	0.023 8	0.00665 26
275.42 2	0.0084 4	0.0086 23	0.00786 26
281.443 8	0.00315 4	0.0063	0.00305 13
282.964 7	0.00376 5	0.0045 13	0.00385 28
291.697 6	0.0177 2	0.037 5	0.0179 6
301.733 9	0.00231 3	0.0053	0.00227 7
317.09 3	0.000457 9	0.0011	0.000450 22

E_γ , keV Our work	I_γ , % Our work	I_γ , % NDS	I_γ , % Ref. [13]
345.868 6	0.0314 4	0.034 5	0.0316 8
356.010 7	0.00211 3	0.0024 8	0.00214 7
387.823 7	0.00325 4	0.0277 24	0.00335 15
410.300 10	0.001102 12	0.0023 6	0.00112 6
448.334 13	0.000489 6	0.0011	0.000495 18

Note. * Summary absolute emission probability of peaks 275.129 keV and 275.428 keV.

The same table contains data from Nuclear Data Sheets (NDS) [10] (based mainly on [11, 12]), which has been normalized to an absolute emission probability of 57.2 % for the 185.7 keV gamma

peak, and work [13]. In a multitude of instances, discrepancies between the absolute emission probabilities obtained and those tabulated in NDS have been observed to exceed 100 %.

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ВИЗНАЧЕННЯ АБСОЛЮТНИХ ІМОВІРНОСТЕЙ ВИПРОМІНЮВАННЯ ГАММА-ПРОМЕНІВ НИЗЬКОЇ ІНТЕНСИВНОСТІ ПІД ЧАС РОЗПАДУ ^{235}U ДЛЯ ЦІЛЕЙ ЯДЕРНОЇ КРИМІНАЛІСТИКИ

Значення квантових виходів у розпаді ^{235}U в діапазоні енергій 200–450 кеВ було уточнено за допомогою напівпровідникової гамма-спектрометрії з використанням еталонного матеріалу, що містить високозбагачений уран, та математичного моделювання з врахуванням геометрії вимірювання. Таке уточнення є важливим для коректного гамма-спектрометричного визначення характеристик (сигнатур) високозбагаченого урану, які мають вирішальне значення для ядерної криміналістики, таких як вік моделі та домішки ^{228}Th . Це уточнення також є корисним з точки зору фундаментальної ядерної фізики, оскільки воно вносить істотні зміни в дані щодо схеми рівнів ^{231}Th при альфа-розпаді ^{235}U .

Ключові слова: ^{235}U , ядерна криміналістика, високозбагачений уран, модельний вік, гамма-спектрометрія, квантовий вихід.

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