Yields of Gamma Radiation of ^{235}U.

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1. Introduction

Precise knowledge of gamma emission yields of uranium isotopes is vital for solving of practical tasks of determination of the isotopic composition of uranium. The most accurate and common used gamma spectrometric method for measuring of the enrichment degree is the method proposed by R. Gunnink [1]. It is based on measuring of the ratio of the count rates of 92.38 and 92.80 keV gamma lines of ^{234}Th (assuming equilibrium with ^{238}U) and the characteristic line $K_{\alpha1}$ (Th) 93.35 keV from the decay of ^{235}U. The ratio of the yields of these lines under ENSDF [2] on 2004 are

$$\frac{I_{92.38}}{I_{93.35}} = 0.484(45) \qquad \frac{I_{92.80}}{I_{93.35}} = 0.477(45).$$

These ratios were used to calculate the enrichment of uranium and their reliability is confirmed by comparison of the results of gamma spectrometric measurements with mass spectrometry data [3]. The results of recent measurements of the yields of ^{234}Th lines [4], which differ from the previous ones by ~ 30%, cast doubt on the value of the yield of the $K_{\alpha1}$ characteristic line (^{231}Th) 93.35 keV from the decay of ^{235}U.

Moreover, the yields of ^{235}U lines in equilibrium with ^{231}Th with energies above 205 keV are still lacking accuracy. In our opinion it's due to the problem of accounting of peaks summation of intense lines of ^{235}U + ^{231}Th with energies below 205 keV.

The aims of this study were to clarify the yield of the $K_{\alpha1}$ characteristic line (^{231}Th) 93.35 keV from the decay of ^{235}U; to measure the ratio of the yields of lines ^{234}Th (the child of ^{238}U) 92.38 and 92.80 keV ^{231}Th X-ray line from the decay of ^{235}U 93.35 keV; to measure the yield of the lines of ^{235}U in equilibrium with ^{231}Th with energies above 205 keV.

2. Materials and Methods

2.1. Samples

^{235}U yields measurements have been performed with standard samples of the National Bureau of Standards, USA: Standard Reference Material SRM 969 at a concentration of ^{235}U (0.32 – 4.5%) and Certified Reference Material CRM 146 at a concentration of ^{235}U (20 – 93%), certified by the isotopic composition of uranium with high accuracy. A detailed description of the samples can be found in [5].

Table 1. SRM 969 samples. Here and after m.f. is mass fraction.

Sample name	SRM031	SRM071	SRM194	SRM295	SRM446
^{235}U m.f., %	0,3166	0,7119	1,942	2,9492	4,4623
^{234}U m.f., %	0,002	0,0053	0,0174	0,0284	0,0365
^{238}U m.f., %	99,6668	99,2828	98,0406	97,0196	95,495
^{236}U m.f., %	0,0146	0,00002	0,0003	0,0033	0,0068
mass, g	200,1	200,1	200,1	200,1	200,1

Table 2. CRM 146 samples. Here and after m.f. is mass fraction.

Sample name	CRM20	CRM52	CRM93
^{235}U m.f., %	20,107	52,488	93,1703
^{234}U m.f., %	0,15	0,37	0,98
^{238}U m.f., %	79,55	46,88	5,56
^{236}U m.f., %	0,2	0,26	0,29
mass, g	230,01	230,00	230,05

2.2. Hardware

The measurements have been performed in the Institute of Nuclear Research, Kiev (INR) and Kharkiv Institute of Physics and Technology (KIPT) in the "U-Pu InSpector" - spectrometers of Canberra.

The gamma-ray spectrometer in INR consisted of a planar HPGe detector of GL1015R type with area of 1000 mm^2 and a thickness of 15 mm with a FWHM resolution of 560 eV for the energy of 122 keV and analyzer Model 1200UPU. Energy range was 10 – 305 keV.

The gamma-ray spectrometer in KIPT consisted of a planar HPGe detector of GL0515R type with area of 500 mm^2 and a thickness of 15 mm with a FWHM resolution of 580 eV for the energy of 122 keV and analyzer Model 1200UPU. Energy range was 10 – 1200keV.

Measurements in INR and KIPT have been performed on the fully identical samples.

2.3. Software

The spectra processing has been performed using the software package SpectraLineUltimate of LSRM Ltd for processing of complex gamma spectra with a large number of multiplets [6]. The features of the software package are listed below:

- An adequate description of the gamma lines is provided by the algorithm which uses the real shape of the apparatus line [7].
- Consideration of the Lorentz broadening for the description of the X-ray lines. Data on the natural width of the lines are taken from [8].
- The yields ratio between the lines of one nuclide is taken into account at the approximation of informative intervals of the spectrum.
- **All** the informative intervals of the spectrum are processed simultaneously. At the boundaries of informative intervals the background steps under them are smoothed under the condition of continuity of the spectrum and its first derivative.
- The contribution of the peaks of the random summation is taken into account.

2.4. Gamma radiation Efficiency Registration Calibration

The precise values of the samples mass and the mass fractions of the content of uranium isotopes provide the calculation of activity reference values for each sample.

The registration efficiency of gamma radiation has been calculated separately for each type of CRM and SRM samples due to the differences in their geometrical dimensions and densities. The “live" time correction has been used for all spectra in the same geometry to account differences in the source-detector distances and spectrometers different loading for different samples. The measurement time has been corrected for the counting rate for ^{238}U reference lines of 63,290 keV for SRM sample and 185.7 keV ^{235}U for CRM sample.

$$t_{corr}^{i} = t^{i} \cdot \frac{\frac{S^0}{A^0}}{\frac{S^i}{A^i}} \qquad (1)$$

where S is the count rate at the peak of the reference line, and A is the activity of the corresponding nuclide.

The registration efficiency of gamma radiation in the energy range below the K-edge of absorption has been calculated for the single lines of ^{231}Th, ^{234}U, ^{234}Th using the reference values of the activity of standard samples, calculated on the base of the isotope uranium concentrations and sample weights. Moreover, the relative values of the registration efficiency for the lines of the uranium fluorescence have been used. The lines used to calculate the efficiency of the gamma-radiation and the resulting relative uncertainty of the registration efficiency (k = 1) are listed in the Table 3.

Table 3. The lines used for calculation of the registration efficiency below the K-edge

Nuclide	Energy, keV	I (Intensity), gammas per 100 decays	ΔI, gammas per 100 decays	Δε, %
^{231}Th	84.214	6.7	0.07	1.7
	102.270	0.441	0.011	2.5
^{234}U	53.2	0.123	0.002	3
^{234}Th	63.290	3.75	0.08	2.5

For the energies above the K-edge the efficiency has been calculated by the lines of ^{235}U, ^{238}U using the relative efficiency of ^{228}Th lines (see Table 4).

Table 4. The lines used for calculation of the registration efficiency higher the K-edge

Nuclide	Energy, keV	I (Intensity), gammas per 100 decays	ΔI, gammas per 100 decays	Δε, %
^{235}U	143.767	11.00	0.08	0.8
	163.356	4.97	0.04	0.9
	185.715	57.0	0.3	1.0
	205.31	5.01	0.05	1.1
^{234}U	120.900	0.0342	0.0005	6
^{238}U	766.321	0.323	0.004	2
	1001.026	0.847	0.008	1.5
^{228}Th	238.62	42.2	0.556	2.5
	583.191	29.6	0.49	3

The yields of the lines have been taken from the ENSDF-data file and LNHB [9].

The calculated values of efficiency for the values below the K-edge of absorption (115.6 keV) have been corrected for the jump in the mass absorption coefficient of the sample material U_3O_8, which was

$$\frac{\varepsilon_{E<115.6}}{\varepsilon_{E>115.6}} = 3.916 \qquad (2)$$

This allowed us to approximate the transformed registration efficiency by one logarithmic

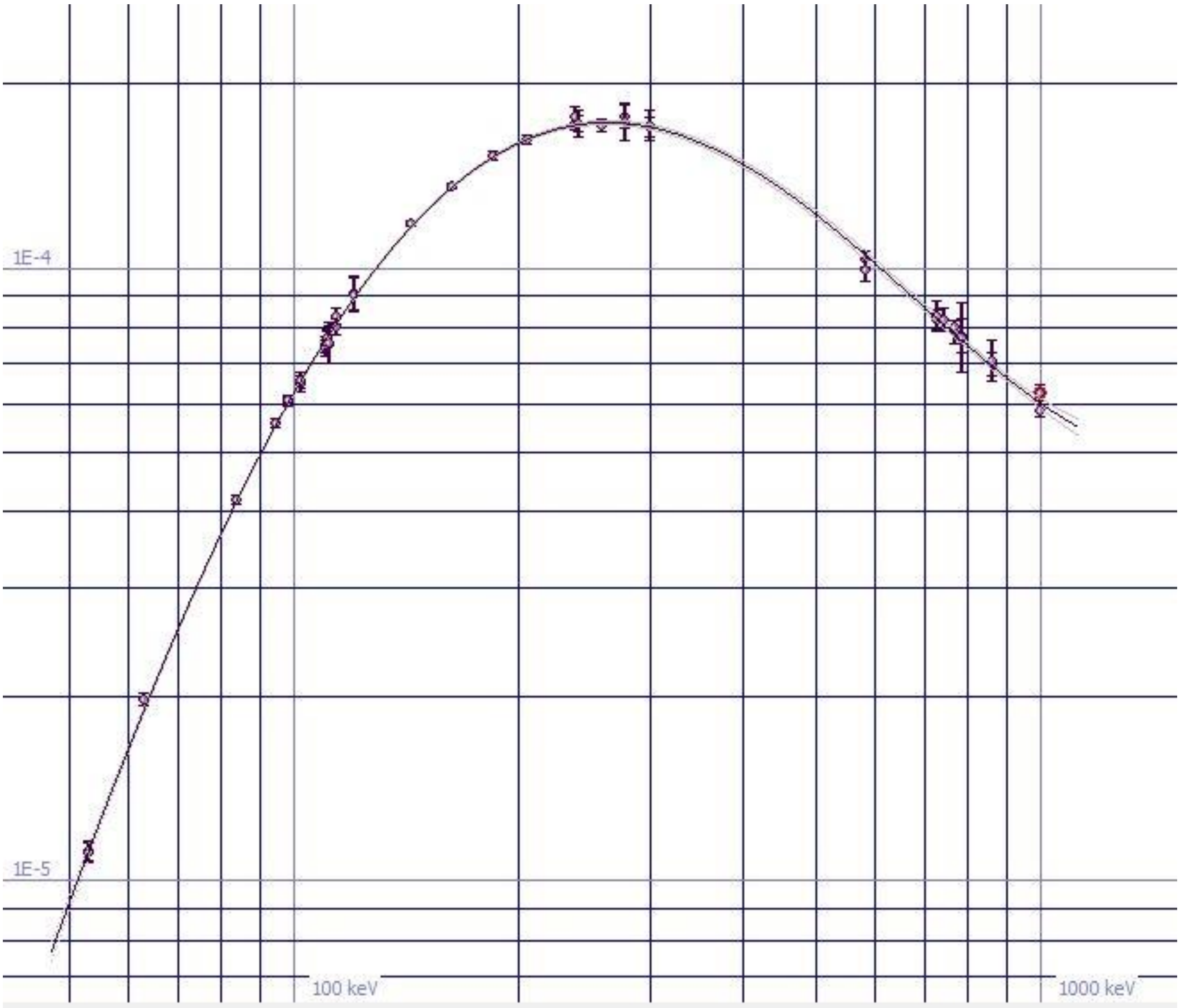

Fig. 1. Approximating curve of the registration efficiency of gamma rays in the energy range of 50 – 1000 keV, taking into account the K-edge of radiation absorption in the sample.

polynomial of the 4 degree (Fig. 1):

$$\lg(\varepsilon(E)) = \sum_{i=0}^{4} a_i \cdot (\lg(E))^i \quad (3)$$

The uncertainty of the efficiency curve in the range of 90 – 110 keV did not exceed 1.5%.
The inversed transformation for the jump has been performed below K-edge (Fig. 2) at the calculation of the real efficiency registration of the gamma radiation.

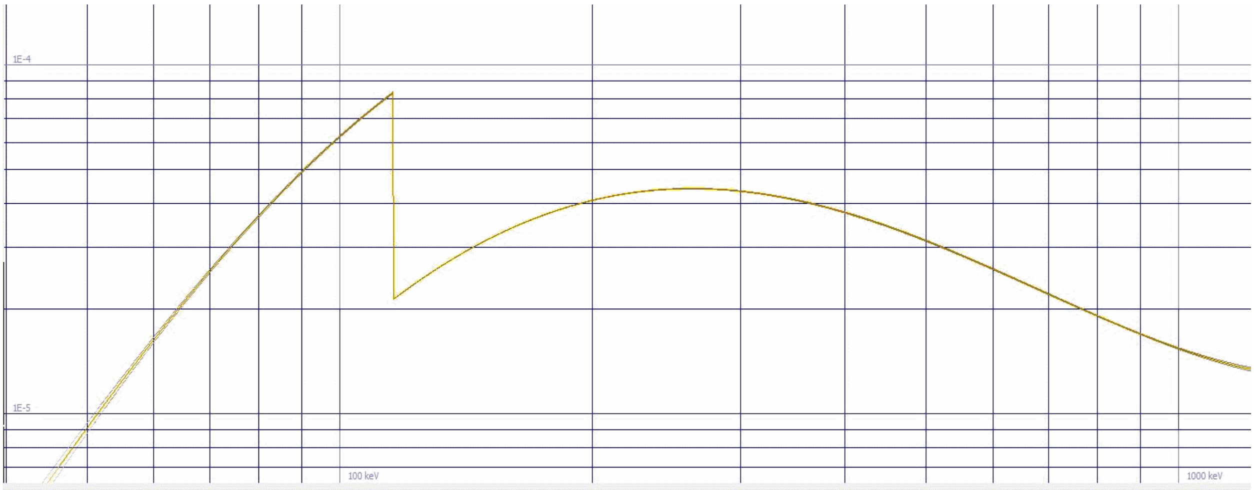

Fig. 2. Approximating curve of the registration efficiency of gamma rays in the energy range of 50 - 1000 keV, without K-edge of radiation absorption in the sample.

2.5. Calculation of ^{235}U Yields of Gamma Radiation

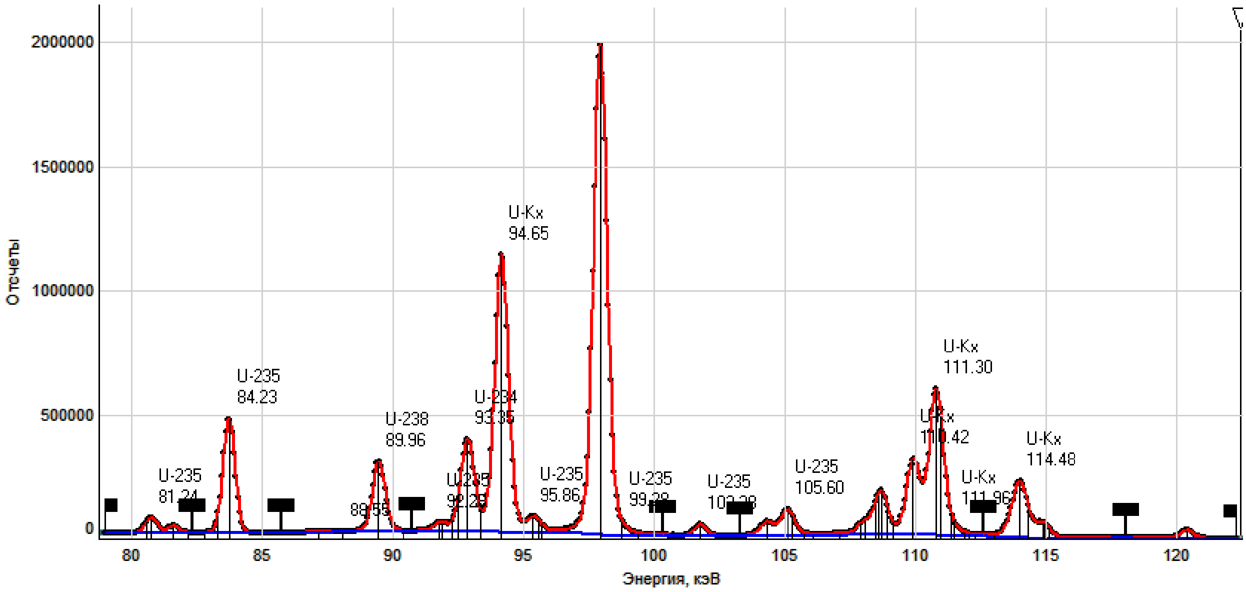

Fig. 3. Markup of spectrum of CRM93 sample.

The yield I_i of the i-th line with the energy E_i has been calculated in accordance with the following equation

$$I_i = \frac{S_i}{t_{corr}\varepsilon(E_i)A} \cdot 100, \qquad (4)$$

where

Si is the area of this line,

A is ^{235}U activity of the measured sample,

ε (E_i) is the registration efficiency.

The spectra of the samples with the highest enrichment CRM93 and CRM52 have been used for the yields determination. All significant gamma and X-ray lines, which appeared to be registered in the spectrum of the sample, have been used for the approximation of the spectrum informative interval. The example of the spectrum markup is shown in Fig.3. As it has been noted above the ratio of yields for the lines of the same nuclide has been considered at the approximation. The yields ratios of the measured lines have been excluded. The lines areas were the results of the approximation. They have been used for the yields calculation in accordance with the equation (4).

3. The Results

3.1. Measurement of the Yield of the Line of 93.35 keV

The results of yield measurement of 93.35 keV which have been got using both spectrometers are shown in the Table 5.

Table 5. the results of measurement of $I_{93.35}$

Spectrometer	Sample	$I_{93.35}$
GL1015R	CRM52	4.83(6)
GL1015R	CRM93	4.83(6)
GL0515R	CRM52	4.88(6)
GL0515R	CRM93	4.85(6)
average value		**4.85(6)**

3.2. Measurement of the Yields Ratio

The yields ratio of ^{234}Th of 92.38 and 92.80 keV and Th characteristic line 93.35 keV from the decay of ^{235}U ($\frac{I_{92.38}}{I_{93.35}}$ and $\frac{I_{92.80}}{I_{93.35}}$), which have been used to measure the degree of enrichment, can be measured with greater accuracy than the yields of the lines themselves.

The counting rates $R_i = \frac{S_i}{A_i}$ for each line normalized for the activity have been calculated on the base of measurements results of low-enriched SRM samples using the GL1015R spectrometer. The average values for four SRM samples are listed in the table.

Table 6. The normalized counting rates of ^{234}Th lines of 92.367 and 92.792 keV

$\frac{S_{92.38}}{A_{u238}}$, 10^{-5}	$\frac{S_{92.80}}{A_{u238}}$, 10^{-5}	$\frac{S_{93.35}}{A_{u235}}$, 10^{-5}
1.5864(10)	1.650(10)	3.3446(20)

Using the ratios of registration efficiency calculated on the basis of the efficiency curve

$$\frac{\varepsilon(92.38)}{\varepsilon(93.35)} = 0.9789(30), \qquad \frac{\varepsilon(92.80)}{\varepsilon(93.35)} = 0.9873(18),$$

we have resulted in the yields ratio of the lined of ^{238}U(^{234}Th) and ^{235}U:

$$\frac{I_{92.38}}{I_{93.35}} = 0.4845(15) \qquad \frac{I_{92.80}}{I_{93.35}} = 0.4997(30)$$

Table 7. The yields of the lines 92.38 keV, 92.80 keV ^{238}U(^{234}Th) and 93.35 keV ^{235}U and their ratios

	ENSDF Up to 2004 and NuDat [2, 10]	ENSDF and NuDat [2, 10]	LNHB 2011/53	This study
$I_{92.38}$	2.81(26)	2.13(20)	2.18(19)	2.350(30)
$I_{92.80}$	2.77(26)	2.10(20)	2.15(19)	2.423(30)
$I_{93.35}$	5.81(11)	5.54(14)	5.76(14)	4.850(33)

$\frac{I_{92.38}}{I_{93.35}}$	0.484(45)	0.384(40)	0.370(40)	0.4845(15)
$\frac{I_{92.80}}{I_{93.35}}$	0.477(45)	0.379(40)	0.373(40)	0.4997(30)

3.3. Measurement of the Yield of Lines of ^{235}U in Equilibrium With ^{231}Th with Energies above 205 keV

The calculated approximation curve of the registration efficiency of gamma radiation provides a measurement of the yields of the lines of ^{235}U in equilibrium with ^{231}Th in the energy range from 50 to 450 keV. The measurement of ^{235}U lines with low-yields with energies above 220 keV is complicated by the peaks of random summation of ^{235}U lines with high yields. The specified procedure in SpectraLineUltimate software allows to calculate the peak energy and area of the random summation and thus to take into account their contribution to the area of the measured lines. Control of correct accounting of summation effects is performed for 371 keV summary peak (185.7 + 185.7 keV). The results of measuring of these lines yields are shown in the Table 8.

The yields of the characteristic radiation have been calculated using the yield values for $K\alpha_1$ line 93.35keV Th $I_{93.95}$ = 4.85 and relative fluorescence yields of Th [9].

Table 8. Yields of gamma-radiation of ^{235}U and ^{231}Th

E,keV	(Intensity), gammas per 100 decays		E,keV	(Intensity), gammas per 100 decays	
	[9] LNHB 2011/53	This study	[9] LNHB 2011/53		This study
58.57	0.477(6)	0.471(7)	202.11	1.08(2)	1.09(1)
74.94	0.051(6)	0.036(9)	205.31	5.02(3)	5.01(5)
89.95	1.01(3)	1.02(3)	215.28	0.029(3)	0.0295(5)
89.957	3.56(9)	3.04(6)	221.38	0.118(5)	0.116(10)
92.288	0.37(4)	0.41(4)	228.78	0.0074(4)	0.070(2)
93.351	5.76(14)	4.85(6)	233.5	0.038(4)	0.0287(4)
95.869	0.59(7)	0.69(7)	240.87	0.074(4)	0.067(1)
96.09	0.091(11)	0.093(9)	246.84	0.055(3)	0.054(1)
99.278	0.137(6)	0.149(12)	266.47	0.0078(6)	0.060(3)
104.819		0.606(18)	275.35	0.051(6)	0.0153(4)
105.604	2.06	1.16(3)	275.49	0.032	
106.239		0.0411(19)	275.4 (sum)	0.083	0.0153(4)
108.582		0.443(13)	281.42	0.0063	0.039(5)
108.955	0.685	0.0119(5)	282.94	0.0063	0.043(5)
109.442		0.097(6)	289.56	0.0074	0.004(2)
109.18	1.66(13)	1.45(3)	291.65	0.040(6)	0.019(2)
115.45	0.03(1)	0.03(1)	301.7	0.0053	0.0022(3)
194.94	0.63(1)	0.63(4)	345.9	0.04	0.034(5)
198.9	0.036(2)	0.044(2)	387.84	0.04(6)	0.037(3)

4. Results and Discussion

As the result of this study we have obtained a new value of the yield of the $K\alpha_1$ (^{231}Th) characteristic lines 93.35 keV from the decay of ^{235}U – 4.85 (6) which differs from the previous value of 5.76 (14) for almost 20%. The reasons for this discrepancy are not clear for us. But it should be noted that the previous value has been obtained by calculation.

The yields ratios of ^{234}Th lines 92.38 and 92.80 keV and Th characteristic line of ^{235}U decay 93.35 keV ($\frac{I_{92.38}}{I_{93.35}}$ and $\frac{I_{92.80}}{I_{93.35}}$), used for the enrichment degree measurement, have been measured with high accuracy (~ 0.5%). The ratios have been estimated with greater accuracy than for the case of using the yields ratio of the tabular data.

The yields of other lines of ^{235}U (in equilibrium with ^{231}Th) above 205keV have been obtained. Tabular data for these lines have a lot of variations. In our opinion the reasons of these variations are neglection or incorrect account of the contribution of the random summation peaks to the spectrum.

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