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The measurements of uranium enrichment by using X rays and gamma rays below 100 keV

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Abstract. In this work the non-destructive gamma - spectroscopic method for determination of uranium enrichment is presented. The method is applicable to material of any physical form and geometrical shape, and does not require the use of reference materials or the use of efficiency calibrated geometry. The X Rays and gamma rays below 100 keVfrom ^{235}U were used to determine their relative detection efficiency internally. The activity ratios of ^{234}U/^{235}U and U^{238}/U^{235}in studied sample were calculated based on the measurents of the count rate of 53.2keV (0.123%) gamma ray of ^{234}U and 92.6 keV (5.16%) of ^{234}Th, a daughter of ^{238}U. As a test of this method, a highly enriched uraniumsample was measured. The obtained result was in agreement with the estimated value.

Keywords: Uranium enrichment, gamma-spectrometry, intrinsic efficiency calibration, MGA method.

1. Introduction

Nowadays, uranium is most commonly used fuel for nuclear power plantsfor production of electricity. The determination of the uranium enrichment is very important in various fields such as nuclear power generation, nuclear safeguards, radiation protection and especially fight against illicit international traffic of radioactive materials and nuclear terrorism. Gamma-ray spectrometry has found increased application in resent years for determining uranium enrichment [1,2].

The object of uranium enrichment measurement is to determine the activity ratios of uranium isotopes. Normally, the activity ratio of ^{238}U/^{235}U in the studied samples were calculated based on the count rate of 185.72 keV gamma line of ^{235}U; 111 keV- X rays (^{238}U) and 1001 keV gamma line of ^{234m}Pa, a first daughter of ^{238}U. Before the analysis, the efficiency calibration of the system was carried out by using some standard sources [1,3]. The method doses not require the use of standard samples or

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the knowledge of the absolute detector efficiency, but uranium enrichment were measured using gamma ray above 100 keV [4,5,6].

In this work,the uranium enrichment of investigated sample is derived from the activity ratios $^{234}U/^{235}U$, $U^{238}/^{235}U$ using gamma rayswith energy below 100 keV. The activity of ^{234}U is determined from 53.200 keV (0.00123) photopeak area of ^{234}U. The activity of ^{235}U is determined from 58.57 keV(0.0048), 81.228 keV (0.0085), 82.087 keV (0.073), 84.214 keV (0.0671) peaks of ^{231}Th, daughter of ^{235}U and 89.956 keV (0.041), 93.356 keV (0.055), 95.86 keV (0.0088) X-ray peaks of ^{235}U- IC. The activity of ^{238}U can be determined based on the 92.365 keV (0.026) and 92.79 keV (0.0256) peaks of ^{234}Th, daughter of^{238}U.

2. Method for determination of uranium enrichment

Uranium enrichment is determined based on the measuring the activity ratios A_{U234}/A_{U235} and A_{U238}/ A_{U235}. To determine the isotopic activity ratio, the multigroup gamma-rays method (MGA method) was used [4]. The method is to measure basically the intensity of two or more peaks from gamma-rays with similar energy but from different isotopes. Then the activity ratio of two different (1 and 2) isotopes can be expressed as follows:

$$\frac{A_1}{A_2} = \frac{n_1.Br_2.\Omega_2.\varepsilon_2.\tau_2}{n_2.Br_1.\Omega_1.\varepsilon_1.\tau_1} \quad (1)$$

where A_1,A_2 are the activities of two isotopes 1 and 2 respectively; n_1,n_2 are the net count rates of the photopeak corresponding to gamma rays γ1 and γ2 with a specific energy E_1 and E_2 from isotopic 1 and 2 respectively; Br_1 and Br_2 are branching ratios for γ_1, γ_2 rays; Ω_1, Ω_2 are the fractional solid angle of detector. Here, they are the same of both γ_1 and γ_2 and can be ignored; ε_1, ε_2 are the efficiency for the energies E_1 and E_2 of γ_1 ray, γ_2-ray from two isotopes respectively; τ_1 and τ_2 are gamma transmission to detector of γ_1 and γ_2 respectively. If the two γ1-ray,γ2 ray are close to the same energy, it gets $\tau 1.\varepsilon 1 \cong \tau 2.\varepsilon 2$. Now Eq. (1) becomes:

$$\frac{A_1}{A_2} = \frac{n_1.Br_2}{n_2.Br_1} = \frac{n_1 / Br_1}{n_2 / Br_2} = \frac{n_1 / Br_1}{f(E)} \quad (2)$$

where $f(E) = \frac{n(E_2^i)}{Br(E_2^i)}$, with E_2^i is energy of γ_i from isotope 2, is called intrinsic efficiency function, which depend on energy of gamma rays [5,6,7].

The uranium enrichment of ^{235}U, q_{235} (%) is calculated by using the following equation:

$$q_{235} = \frac{m_{235}}{m_{235} + m_{234} + m_{238}} = \frac{1}{1 + m_{234}/m_{235} + m_{238}/m_{235}} .100\% \quad (3)$$

where m_{234},m_{235} and m_{238} are the masses of ^{234}U,^{235}U and ^{238}U respectively in investigated sample.

The enrichment of uranium isotopes can be expressed as a function of activity of ^{234}U, ^{235}U and ^{238}U as follows:

$$q_{235} = \frac{1}{1 + 3.479.10^{-4}.\frac{A_{U234}}{A_{U235}} + 6.43.\frac{A_{U238}}{A_{U235}}}.100\% \quad (4)$$

To determine ^{235}U isotope enrichment, we have to measure the activity ratios ofA_{u234}/A_{235} and A_{u238}/ A_{235} .

3. Experimentaland results

3.1. Uranium sample measurements

The enriched uranium sample was in the form of oxide (U_3O_8), which is sent to Institute of Isotopes of the Hungarian Academy of Sciences by the International Technical Working Group on Combating Illicit Trafficking of Nuclear Materials (ITWG). All measurements were carried out at Institute of Isotopes of the Hungarian Academy of Sciences.The data were analyzed at Nuclear Department of Physics, University of Sciences, VNU. The U_3O_8 powder was placed within a thin, closed polyethylene cylinder of 2.9cm inner diameter. The sample was measured at 10cm distance from the detector. The gamma spectra were taken by using a planar HPGe detector model GLP-10180/07 (ORTEC) with active diameter of 10mm and thickness of 7mm. The gamma spectra were being recorded until the statistical error of the 53,2 keVand 92.6 keV gamma peaks dropped below 1.5%. The gamma spectra were measured and analyzed by using the Gamma-Vision and Genie2000 program. Typical gamma spectrum of high enriched uranium is shown in Fig.1. From Fig.1 we can see that: Counts of the 53.2 keV and 58.57 keV gamma ray peaks of ^{234}U are stongest (table 1) and isolated. The 81.228 keV (0.0085), 82.087 keV (0.073), 84.214 keV (0.0671) gamma peaks of ^{231}Th and 89.956 keV (0.041), 93.356 keV (0.055), 95.86 keV (0.0088) X ray peaks of ^{235}U-IC have high counts, but many of them are overlapped.

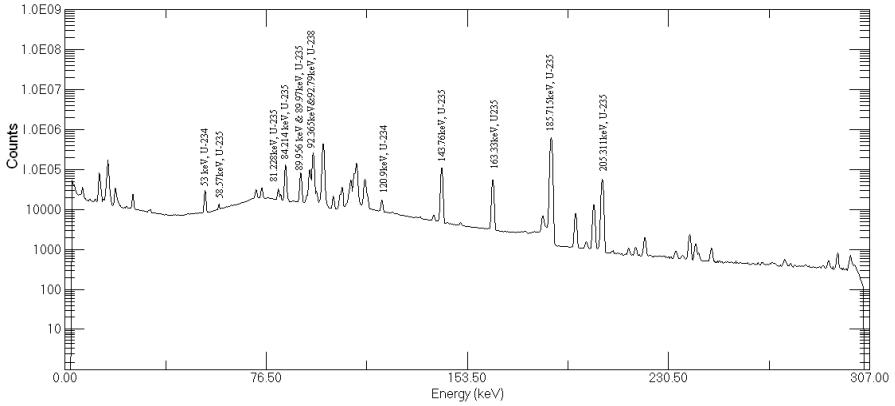

Fig.1. Gamma ray spectrum of 5 g enriched uranium sample with the measuring time 43.56 h

3.2. Analyze multipletphotopeak into components

To analyze multiplet photopeak into components, the least squares fitting method was used. In The deconvolution treatment of x- ray and γ- ray peaks in the spectra are carried out by fitting with a Gaussian function for γ- ray and pseudo Voigt function for x-rays [7,8,9,10]. The Voigt Function$V_p(E)$, which is expressed by a weighted sum of Gaussian, G(E) and, Lorentzian, L(E) functions, both added to linear background, B(E):

$$V_p(E) = kG(E) + (1-k)L(E) + B(E) \tag{5}$$

where G(E) and L(E) functions with the same full-width at half – maximum, $\Gamma = FWHM$ and the factor, k = 0.57 in a pseudo Voigt function are defined as function of the energy, E as:

$$G(E) = N_{\max} \exp\left[-\ln 2\left(\frac{E-E_j}{\Gamma/2}\right)^2\right] \tag{6}$$

$$L(E) = \frac{N_{\max}}{1+\left[(E-E_j)/(\Gamma/2)\right]^2} \tag{7}$$

where E_j is peak energy and N_{max} is peak counts, respectively.

A highly- enriched uranium γ- ray spectrum in the energy region of 80 keV -100 keV taken by a planar HPGe detector and fitted with Gaussian function for γ- ray and pseudo Voigt function for X-rays using Origin 7.5 (Fig 2). The net peak areas are determined (Table1). The branching ratios for γ- rays in Table 1 were taken from reference [9,10].

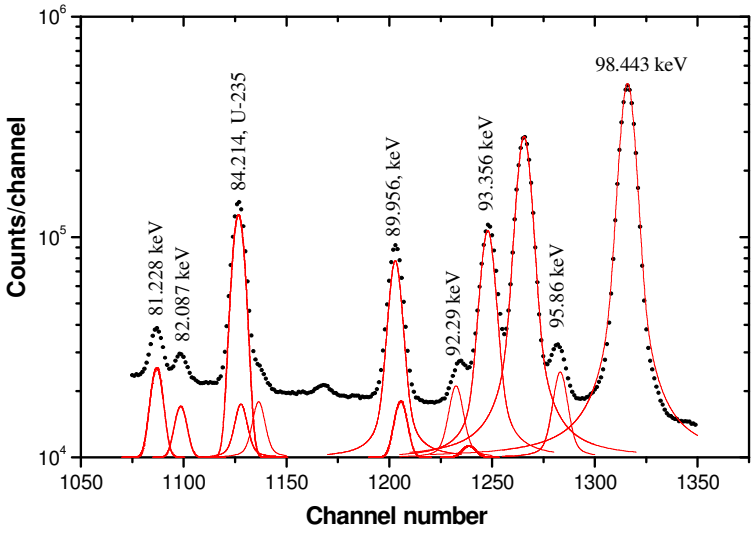

Fig. 2. A highly- enriched uranium γ- ray spectrum taken by a planar HPGe detector and fitted with Gaussian function for γ- ray and pseudo Voigt function for X-rays.

Table.1. Characteristic of rays-gamma and measured results in below 100keV region

Energy(keV)/	Branching ratio –Br	Photopeak area (count)	Net count rates of the photopeak n(cps)	n/Br (cps)	Parent
53.2	0.00123	144860	0.924÷0.002	751.1 ±11.3	^{234}U
58.57	0.0050	23073	0.147÷0.001	29.42±1.7	^{235}U
81.228	0.0085	104857	0.669÷0.002	78.67±2.78	^{235}U
82.087	0.0038	47478	0.303÷0.001	78.45±3.1	^{235}U
84.214	0.0671	834449	5.321÷0.006	79.30±1.2	^{235}U
89.944	0.0094	551636	3.518÷0.005	85.80±5.9	^{235}U
92.6	0.0516	7893	0.024÷0.004	0.975 ±0.048	^{238}U
93.356	0.055	847300	5.403÷0.006	98.49±9.0	^{235}U
95.86	0.0088	148320	0.946÷0.002	107.48±5.3	^{235}U

3.3. Determination of the uranium enrichment of material

The ^{234}U activity is determined directly from 53.2 keV gamma ray. By using relative efficiency calibration and from the equation (2), the A_{U234}/A_{U235}ratio was derived as follows:

$$A_{U234}/A_{U235} = \frac{n(53.2)/Br(53.2)}{f(53.2)}. \quad (8)$$

Determination of the ^{238}U activity is based on92.6 keV gamma peak (92.365 keVpeak (0.0260) and 92.79 keV (0.0256) of ^{234}Th. According to Eq. (2), the A_{U238}/A_{U235} was determined using relative efficiency calibration, as follows:

$$A_{U238}/A_{U235} = \frac{n(92.6)/Br(92.6)}{f(92.6)} \quad (9)$$

In this case, the function f(E) is obtained by fitting experimental data of relative efficiencies at the 58.57 keV,81.228 keV, 84.214 keV peaks of^{231}Th and 89.956 keV, 93.356 keV, 95.86 keVX- peaks of ^{235}U- IC (Fig. 3).

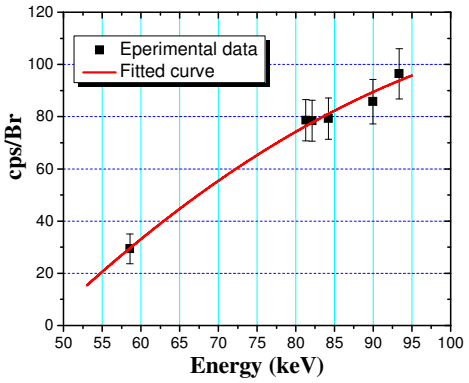

Fig. 3.The relative efficiency curve using the gamma peaks of U^{235}, $f(E) = 0.01774E^2 + 4.54148E - 175.50361$, with the value of $R^2 = 0.989$

The activity ratios $A_{U234}/A_{U235}$and A_{U238}/A_{U235}were then calculated using the Eqs. (8) and (9) respectively.The obtained values *are* as follows:

$A_{U234}/A_{U235} = 30.2 \pm 2.1$. (Bq/Bq)

$AU238/A_{U235} = 0.0102 \pm 0.0012$ (Bq/Bq).

Uranium enrichment of the investigated sample, q_{235}, was determined based on the activity ratio A_{U234}/A_{U235} and A_{U238}/A_{U235} and using Eq. (4). Experimental result for the investigated uranium sample is: **q_{235} = (92.9 ± 5.1) %**. According to IAEA uranium enrichment of the investigated sample is 90%.

4. Conclusion

From the results in this work, we can see that the gamma-spectrometric technique can be used to determine precisely the uranium enrichment of highly enriched uranium samples.The uranium enrichment of investigated sample was calculated from the activity ratio $^{234}U/^{235}U$and $^{238}U/^{235}U$. The activity ^{234}U, ^{235}U, U^{238} can be determinedby using X - rays and gamma rays below 100 keV. The X – rays and γ- rays spectra were analyzed by fitting a Gaussian function for γ- ray and pseudo Voigt function for X-rays respectively.The main sources of the errors are statistical error, net peak area determination, gamma ray self absorption, fitting procedure and nuclear data used. The result obtained is in good agreement with estimated value from IAEA.

Acknowledgments

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