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Enrichment determination of low – enriched uranium material by gamma spectroscopic method

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Abstract. In this work the non - destructive gamma spectroscopic method for determination of uranium enrichment is presented. In order to determine the uranium enrichment the activity ratios of $^{234}U/^{235}U$ and $^{238}U/^{235}U$were measured. The activity ratios$^{234}U/^{235}U$ and $^{238}U/^{235}U$ were determined by using intrinsic efficiency calibration. As a test of this method, low - enriched uranium standard was measured, the obtained result was in good agreement with the estimated value.

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

I. INTRODUCTION

The enriched uranium can be classified into two main types: highly- enriched uranium (more than 20% of ^{235}U) and low–enriched uranium (less than 20% of ^{235}U). In recent years, gamma ray spectrometry has found increased application for determining uranium isotopic abundance [1,2,3,4,5].

The traditional uranium enrichment measurement method is based on the measurement of the 185.75 keV peak and is commonly referred to as the "enrichment meter" method. The enrichment in material samples was obtained by measuring the activity ratio of $^{235}U/^{238}U$. The activity of ^{235}U was determined from 185.75 keV peak of ^{235}U and the activity of ^{238}U was determined from 1001.03 keV of ^{234m}Pa from decay of ^{238}U [2].

The our purpose is to use a non-destructive, gamma-spectrometric method using intrinsic efficiency calibration for determining the uranium enrichment of samples with of arbitrary size, shape and composition [6,7,8]. In this work, low-enriched uranium samples were measured, intrinsic efficiency calibration was obtained using the peaks of ^{231}Th and U^{235}. The half- life of ^{231}Th is 25.52 h and therefore it is practically always in equilibrium with its parent, ^{235}U. The activity of ^{238}U is determined based on the 63.29 keV (0.0447)peak of ^{234}Th. The activity of ^{234}U is determined from 120.9 keV (0.000342) peak of ^{234}U.

II. METHODOLOGY

A. The uranium enrichment

The objective of Uranium enrichment measurement methods is to determine the $^{235}U/U$ isotopic ratios. The uranium (atom) enrichment of ^{235}U, q_{235} (%) is defined as follows:

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

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where N_{234}, N_{235} and N_{238} are the numbers of ^{234}U, ^{235}U and ^{238}U nuclei 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. In consideration of the basis relation between the activity A and number of radioactive nuclei N in sample, we have:

$$A = \frac{\ln(2).N}{T_{1/2}} \qquad (2)$$

where A is activity; $T_{1/2}$ is half-life of isotope, $T_{1/2,234} = 2.46.10^5$ years, $T_{1/2,235} = 7.04.10^8$ years and $T_{1/2,238} = 4.47.10^9$ years, which are half-life of isotopes ^{234}U, ^{235}U and ^{238}U respectively [9].

From the formulas (1) and (2), we can derive the uranium enrichment as follows [8]:

$$q_{235} = \frac{1}{1 + 3.49x10^{-4}\frac{A_{234}}{A_{235}} + 6.435\frac{A_{234}}{A_{235}}}100\% \qquad (3)$$

where A_{234}, A_{235} and A_{238} are the activity of ^{234}U,^{235}U and ^{238}U respectively.

Thus uranium enrichment will be determined based on the measuring the activity ratios A_{234}/A_{235} and A_{238}/A_{235} .

B. Determination of the isotopic activity ratio

We can write an expression for the net count of photopeak of specific gamma ray with energy E_j emitted from a given isotope in a sample of arbitrary configuration, as [2] as:

$$n(E_j) = A.Br_j.\varepsilon(E_j) \qquad (4)$$

where: A is activity of given isotope; $n(E_j)$ is net count rates of the photopeak of specific gamma ray with energy E_j emitted from isotope.

Br_j is branching ratio for γ with energy E_j, $\varepsilon(E_j)$ is total detection efficency for a gamma ray with energy E_j. This includes detector efficiency, fractional solid angle of detector and sample, self absorption of gamma rays in sample, and attenuation in packaging and materials between sample and the detector.

To determine the isotopic activity ratio, the multigroup gamma rays method (MGA) was used [3,4,5,9]. This method is based on measuring (basically) the intensity of two or more gamma peaks from different isotopes that have (from gamma-ray of) similar energy. (but from different isotopes). Then the activity ratio of two different (1 and 2) isotopes can be expressed as follows:

$$\frac{A_1}{A_2} = \frac{n(E_i^1)Br_2(E_k)\varepsilon(E_k)}{n(E_k^2)Br_1(E_i)\varepsilon(E_i)} \qquad (5)$$

where: A_1, A_2 are the activities of two isotopes 1 and 2 respectively;

$n(E_i^1)$ and $Br_1(E_i)$ are net count rates of the photopeak of specific gamma ray with energy E_i and branching ratio for γwith energy E_i emitted from isotope 1.

$n(E_k^2)$ and $Br_2(E_k)$ are count rates of the photopeak of specific gamma ray with energy E_k and branching ratio for γ with energy E_k emitted from isotope 2.

$\varepsilon(E_i)$, $\varepsilon(E_k)$ are total efficency for photopeak detection of a gamma ray with energy E_i and E_k respectively. If the two γ rays with E_i and E_k are close to the same energy, it gets $\varepsilon(E_i) = \varepsilon(E_k)$. Now formula (5) becomes:

$$\frac{A_1}{A_2} = \frac{n(E_i^1)/Br_1(E_i)}{f(E)} \qquad (6)$$

Where $f(E) = \frac{n(E_k^2)}{Br(E_k)}$, with E_k^2 is energy of γ_k ray emitted from isotope 2, is called intrinsic efficiency calibration function, which depend on energy of gamma ray.

From formula (4), considering a series of gamma rays from a single isotope, we see that the quotient of the photopeak counts at energy E_k and branching ratio Br, (n/Br), is proportional to the efficiency at energy E_k. The

function f(E) is obtained by fitting experimental data (n/Br) of efficiencies at peaks of only isotope 2 [6,7,8].

III. EXPERIMENTAL RESULTS

A. Uranium sample measurements

The low - enriched uranium standard 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 2.9 cm inner diameter.

An amount of 5 g of U_3O_8 powder was placed within a thin, closed polyethylene cylinder of 2.9 cm inner diameter. The sample was measured at 20 cm distance from the detector. The gamma spectrum was taken by gamma spectrometry using a planar HPGe detector model GLP-10180/07 (ORTEC) with active diameter of 10 mm and thickness of 7 mm. The gamma spectra were measured and analyzed by using the Gamma Vision program. The spectra were being recorded until the statistical error of counts of the 120.9 keV of ^{234}U and 63.29 keV of ^{238}U dropped below 1%. A typical gamma spectrum of low – enriched uranium sample is shown in Fig.1. (From that gamma spectra) we can see that the peaks of : counts of 58.57 keVof ^{231}Th, 63.29 keV of ^{234}Th, 120.90 keV of ^{234}U and 143.764 keV, 163.358 keV, 185.57 keV and 205.309 keV of ^{235}U are high count rate stronggest and separated isolated.

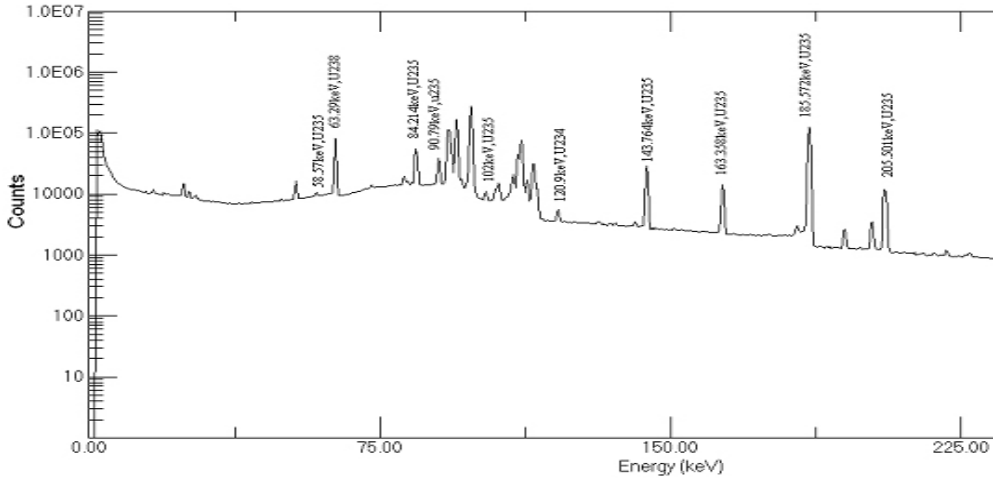

Fig.1. *Gamma ray spectrum of low - enriched uranium sample with counting time 25.14 h.*

Meanwhile, (But) the peaks of 81.228 keV(0.0085) and 82.087 keV (0.0037) of ^{231}Th(^{235}U); 83.3 keV (0.00073) of ^{234}Th(^{238}U) , 84.214 keV (0.0671) of ^{231}Th(^{235}U) and 84.930 keV (0.107) Pb- Kβ_1 X-ray (fluorescence); 92.290 keV (0.0047) Pa Kα2 ^{235}U – IC decay, 92.356 keV (0.0260) , 92.790 keV (0.0256) of ^{234}Th (/^{238}U) and X-rays of 93.356 keV(0.11) (0.00\550) of Th Kα1 of ^{235}U (-IC), 94.660 keV (0.0916) U Kα2 (fluorescence) and 98.443 keV (0.1480) U Kα1 (fluorescence) have high counts, but many of them are overlapped.

Gamma ray spectrum in the energy region of 80 keV – 94 keV taken by a planar

HPGe detector and fitted [8] with Gaussian function for γ – ray and pseudo Voigt for x – rays to determine photopeak areas (Fig.2). The of gamma photopeak areas are determined.

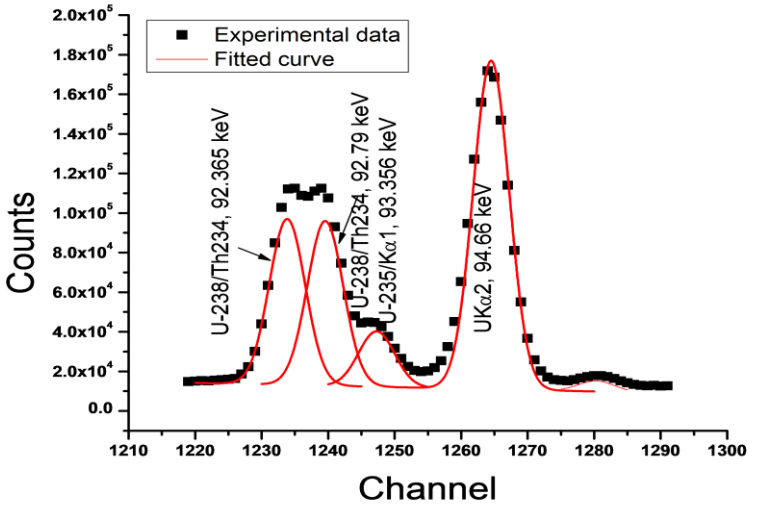

Fig.2. The 92 keV region of investigated sample.

B. Determination of the activity ratios A_{234}/A_{235} and A_{238}/A_{235}

The ^{234}U activity was determined directly from its 120.9 keV peak. According to formula (6), the ratio A_{234}/A_{235} was determined by using relative efficiency calibration, as:

$$\frac{A_{234}}{A_{235}} = \frac{n(120.9keV)/Br(120.9keV)}{f(120.9keV)}$$

In this case, the function f(E) is obtained by fitting a second order polynomial to relative efficiencies at 143.764 keV, 163.358 keV, 185.57 keV and 205.309 keV peaks of ^{235}U (Fig 3).

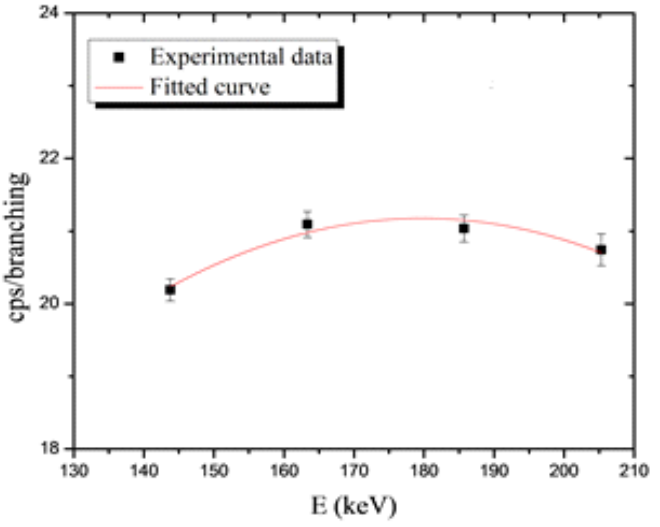

Fig 3. The relative efficiency curve is constructed based on gamma peaks of ^{235}U

f(E)= - 0.00073E^2+ 0.26162E - 2.34247, with the value R^2= 0.953.

We obtained value of activity ratio:

$$\frac{A_{234}}{A_{235}} = 27.7 \pm 1.2(Bq/Bq)$$

The ^{238}U activity was determined from 63.29 keV peak of ^{234}Th. According to the formula (6), the ratio A_{234}/A_{235} was determined by using relative efficiency calibration, as:

$$\frac{A_{238}}{A_{235}} = \frac{n(63.29keV)/Br(63.29keV)}{f(63.29keV)}$$

In this case, the function f(E) is obtained by fitting a second order polynomial to relative efficiencies at 81.228 keV, 82.087 keV, 84.214 keV, 89.96 keV and 93.56 keV of ^{235}U and 53.2 keV of ^{234}U (Fig 4).

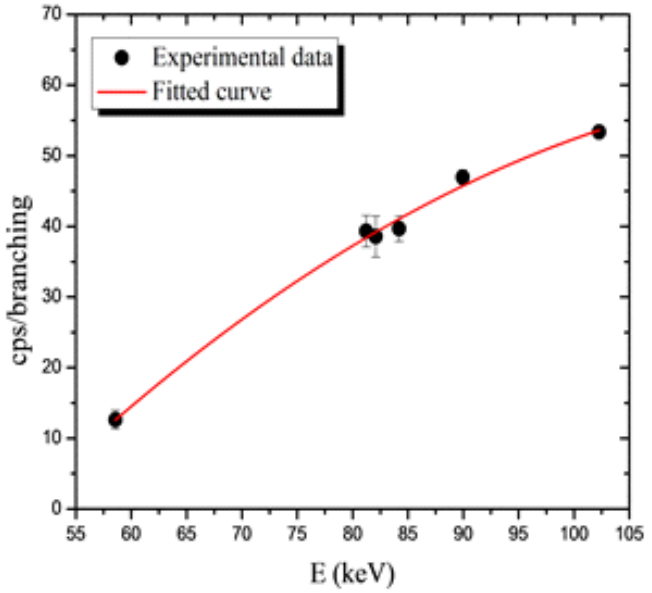

Fig 4. The relative efficiency curve is constructed based on gamma peaks of ^{235}U

f(E)= - *0.0095E*^{2}+ *2.46722E* – 99.326; with the value R^2= 0.9951.

We obtained value of activity ratio:

$$\frac{A_{238}}{A_{235}} = 5.2 \pm 0.2\left(Bq/Bq\right)$$

Uranium enrichment of investigated sample was determined based on the obtained results of activity ratios A_{234}/A_{235} and A_{238}/A_{235} and using formular (3), the obtained result of uranium enrichment:

$$q_{235} = (2.89 \pm 0.12)\%$$

The uranium enrichment of the representative sample was analyzed to be 2.89%. This result is in agreement with estimated value (2.96%) from IAEA. The main sources of the uncertainties for the present results were estimated due to statisticcal errors: $(0.5 \div 1.5\%)$; the detection efficiency: $(1.0 \div 1.5\%)$; the gamma branching ratio.

IV. CONCLUSION

In this work, the gamma-spectrometric technique was applied to determine precisely the uranium enrichment of low enriched material. The uranium enrichment of investigated sample was calculated from the activity ratios ^{234}U/^{235}U and ^{238}U/^{235}U. The intrinsic efficiency calibration was used in determining the activity ratios of ^{234}U/^{235}U and

$^{238}U/^{235}U$. The gamma spectra were analyzed by fitting a Gaussian function for γ- ray and pseudo Voigt function for X-rays respectively. This method doses not require the use of standard samples nor the knowledge of the detector absolute efficiency. The method can be used for samples of arbitrary size, shape and composition.

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