VNU Journal of Mathematics – Physics, Vol. 29, No 2 (2013) 33-39

Experimental determination of enrichment of uranium material by gamma-spectroscopic technique

Bui Van Loat[1,*], Le Tuan Anh[1], Nguyen Cong Tam[2]
Pham Duc Khue[3], Bui Minh Hue[3]

[1]*Department of Nuclear Physics, Faculty of Physics, VNU University of Science*
334 Nguyen Trai, Hanoi, Vietnam

[2]*Institute of Isotopes of Hungarian Academy of Sciences, Budapest, Hungary*

[3]*Institute of Physics, Vietnam Acadermy of Science, 18 Hoang Quoc Viet, Hanoi, Vietnami*

Received 03 November 2012
Revised 24 December 2012; Accepted 15 March 2013

Abstract: During the last years, as the international illicit traffic of radioactive/ fissionable materials have increased, it became important to be able to apply fast reliable methods for the uranium enrichment determination. In order to determine the uranium enrichment the activity ratios of $^{234}U/^{235}U$ and $^{238}U/^{235}U$ was measured. Uranium isotopic abundance can be determined by alpha spectrometry and mass- spectrometry methods, which are destructive methods. In this work the non-destructive gamma – spectroscopic method for 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 nor the use of an efficiency calibrated geometry. The activity $^{234}U/^{235}U$ was determined by using intrinsic efficiency calibration. The 63.29 keV photopeak of ^{234}Th and 58.57 keV of ^{231}Th were used for determination of activity $^{238}U/\ ^{235}U$. As a test of this method, a highly enriched uranium standard was measured, the obtained result was in agreement with the estimated value.

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

1. Introduction

Uranium is probably the most important radioactive element present in the nature. The isotopic abundance of natural uranium is 99.2742% ^{238}U, 0.7204% ^{235}U and 0.0054% ^{234}U [1]. Most of applications of uranium are based on the energy generated by the fission of the ^{235}U nuclide. Nowadays uranium is often used in nuclear power plants to produce electricity. The enriched uranium can be classified into two main types: highly- enriched uranium (more than 20% of ^{235}U) and low–

* Corresponding author. Tel.: 84- 912865869
E-mail: loatbv@vnu.edu.vn

enriched uranium (less than 20% of ^{235}U) [1]. 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 [1,2].

Normally, enrichment of uranium material is determined by alpha spectrometry and mass - spectrometry methods, which are destructive methods. In the last few years, method for the absolute determination of uranium enrichment by high-resolution gamma spectrometry, which is non-destructive method [1-6]. 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 [1-4]. Before the analysis, the efficiency calibration of the system was carried out by using some standard sources.

The purpose of this work is to develop a non-destructive, gamma-spectrometric method using intrinsic efficiency calibration for determining the uranium enrichment of highly-enriched samples with small volume. The uranium enrichment of investigated sample is derived from the activity ratios ^{234}U/^{235}U, ^{238}U/^{235}U. The activity of ^{234}U is determined from 120,9 keV (0.040%) photopeak area of ^{234}U. The activity of ^{235}U is determined from 58.57 keV (0.462%) peak of ^{231}Th and 143.8, 163.3, 185.7, 205.3 keV peaks of ^{235}U. ^{231}Th has a short half- life (25.52 h) and therefore it is practically always in equilibrium with its parent, ^{235}U. The activity of ^{238}U can be determined based on the 63.29 keV peak of ^{234}Th. ^{234}Th with half-life 24.1 d and its daughter, ^{234m}Pa with half-life 6.7 h, therefore secular equilibrium was established within reasonable time.

2. Methodology

2.1. The uranium enrichment

The uranium enrichment or content of ^{235}U, q_{235} (%) is define as:

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

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. Starting from the basis relation between the activity A and mass of radioactive isotope in sample, we have:

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

where A is activity; m is mass in sample; μ is the atomic mass of isotope; $T_{1/2}$ is half-life of isotope and N_A is the Avogadro number.

From formula (2), the masses of ^{234}U, ^{235}U and ^{238}U were calculated as:

$$m_{234} = \frac{234.T_{1/2,U234}.A_{U234}}{\ln 2.N_A};\ m_{235} = \frac{235.T_{1/2,U235}.A_{U235}}{\ln 2.N_A}\ and\ m_{234} = \frac{238.T_{1/2,U238}.A_{U238}}{\ln 2.N_A} \quad (3)$$

where Au_{234}, A_{U235} and A_{U238} are the activity values of ^{234}U,^{235}U and ^{238}U respectively; $T_{1/2,U234}$ = $2.46.10^5$ years, $T_{1/2,U235}$ = $7.04.10^8$ years and $T_{1/2,U238}$ = $4.47.10^9$ years [9,10].

From the formulas (1) and (3), we can derive the uranium enrichment as follows:

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

Uranium enrichment is determined based on the measuring the activity ratios A_{u234}/A_{235} and A_{u238}/A_{235} .

2.2. Determination of the isotopic activity ratio

To determine the isotopic activity ratio, the multigroup gamma-ray method (MGA method) was used [2,6]. The method is to measure basically the intensity of two or more peaks from gamma-ray of similar energy but from different isotopes. Then the activity ratio of two different (1 and 2) can be expressed as follows:

$$\frac{A_1}{A_2} = \frac{n_1.Br_2.\Omega_2.\varepsilon_2.\tau_2}{n_2.Br_1.\Omega_1.\varepsilon_1.\tau_1} \quad (5)$$

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 isotopes 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 and are the same for both γ_1 and γ_2 and cancels out; 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 detertor 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 formula (5) becomes:

$$\frac{A_1}{A_2} = \frac{n_1.Br_2}{n_2.Br_1} = \frac{n_1/Br_1}{n_2/Br_2} = \frac{n_1/Br_1}{f(E)} \quad (6)$$

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 ray [5,7].

2.3. Self - absorption correction

In this paper, isotopic activity ratios were determined from the low energy photons. The count-rate of low energy photons used for determination of uranium enrichment is highly affected by the intense self- absorption of the photons inside sample. It is also important to mention that the self

absorption of photons inside sample [7,8]. To correct the self- absorption of the photons inside sample, the values of the count rate to the total mass of the sample were standardized in unit mass as ratio, K(m), of the count rate to the total used mass of the sample (m) [6,8]. The data were fitted with the function:

$$K(m) = K_0 \frac{1-\exp(-am)}{am} \quad (7)$$

The parameter K_0 is the value where the curve crosses the vertical axis and physically it represents the net count rate of 1 g of the sample corrected for self absorption. The parameter (a) provides information about the matrix of the sample and its density. A non- linear least-squares fit yields the values K_0 and a were determined. Count rate correted for self absorption for mg of sample, n_{cor}, is:

$$n_{cor} = \frac{n_{mea}}{(1-e^{-am})/am} \quad (8)$$

where n_{mea} is count rate of photo peak were determined by experiment; n_{cor} is count rate corrected for self absorption, which were used calculated the activity ratios.

3. Experimental and results

The sample investigated was in the form of oxide (U_3O_8) with highly-enriched uranium. This sample 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.

3.1. Measuring the activity of ^{234}U, ^{235}U, ^{238}U

The U_3O_8 powder was placed within a thin, closed polyethylene cylinder of 2.9 cm inner diameter. The sample was measured at 10 cm distance from the detector. The gamma spectrum was taken by 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 being recorded until the statistical error of the 120.9 keV line dropped below 1%. The gamma spectra were measured and analyzed by using the GammaVision and Genie2000 program. In order to correct for self - absorption of gamma rays in the sample, a series of measurements were performed for the samples with 0.5, 2.0, 5.0 and 10 g of reference material U_3O_8 powder. The samples were measured at 2cm distance from the detector cap. After measurements, the gamma spectra were processed to identify isotopes (gamma energy/branching ratio-Br) and get net area of gamma peaks (S). Correction factor ($\frac{1-e^{-am}}{am}$) for self absorption effect in sample were determined based on the net area of gamma peaks. The obtained results were given in Table 1. These data will be used to determine the activity ratios A_{u234}/A_{u235}, A_{u238}/A_{u235},

Table 1. The experimental results in region of gamma energy below 300 keV for 5.g sample with the measuring time 10922.4 s

Gamma ray energy (keV)/ Br (%)	S (counts)	n_{mea} (cps)	$\frac{1-e^{-am}}{am}$	n_{cor} (cps)	n/Br (cps)
58.75/(0.462)	11808 ± 109	1.081	0.295	3.66 ± 0.01	793.0 ± 16.0
63.29/(3.7)	29596 ± 172	2.71	0.309	8.7 ± 0.1	235 ± 14
120.99/(0.04)	30967 ± 176	2.835	0.388	7.31 ± 0.02	18267 ± 110
143.76/(10.96)	414927 ± 644	37.989	0.512	74.20 ± 0.06	677.0 ± 54.2
163.35/(5.08)	207936 ± 456	19.038	0.591	32.21 ± 0.04	634.1 ± 38.1
185.75/(57.2)	2291665 ± 1514	209.815	0.661	317.42 ± 0.14	554.9 ± 44.4
205.3 /(5.08)	188249 ± 109	17.234	0.704	24.48 ± 0.04	488.7 ± 34.2

3.2. Determination of the activity ratio A_{u234}/A_{u235}

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:

$$Au_{234}/Au_{235} = \frac{n(120.9)/Br(120.9)}{f(120.9)}. \quad (8)$$

In this case, the function f(E) is obtained by fitting a second order polynomial to relative efficiencies at 143.8, 163.3, 185.7, 205.3 keV peaks of ^{235}U (Fig 1).

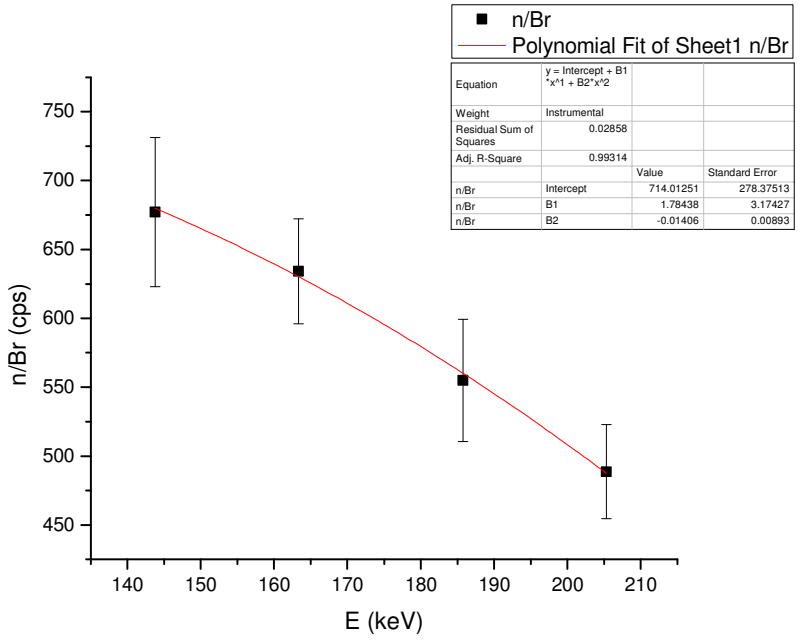

Fig. 1. The relative efficiency curve using the peaks of U^{235}

$f(E) = a + b_1E + b_2E^2$ with the value of R^2 is 0.983.

Function f(E) is derived as follows:

$$f(E) = -0.0141.E^2 + 1.7844E + 714. \quad (9)$$

where E is energy of gamma-ray in keV.

From formula (9) we obtained the value of f(120.9) = 724.2 cps. From data in Table 1 and formula (8) we obtained value of activity ratio: $\frac{A_{u234}}{A_{u235}} \approx 25.20 \pm 2.1\ (\mathrm{Bq/Bq})$.

3.3. Determination of activity ratio A_{U238}/A_{U235}

Because of ^{231}Th has a short half- life (25.52 h) and therefore it is practically always in equilibrium within a reasonable time, we have: $\frac{A_{Th234}}{A_{Th231}} = \frac{A_{U238}}{A_{U235}}$. The activity of ^{234}Th was determined from 63.29 keV peak of ^{234}Th and the activity of ^{231}Th was determined from 58.57 keV peak of ^{231}Th. For that reason the efficiency of planar detector for the peaks in the 20-100 keV energy region is similar, from formula (6) activity ratio A_{U238}/A_{U234} is calculated by the following equation:

$$\frac{A_{U238}}{A_{U235}} = \frac{n_{63.27}}{n_{58.57}} . \frac{Br_{58.47}}{Br_{63.27}} = \frac{n_{63.27} / Br_{63.27}}{n_{58.57} / Br_{58.57}} \quad (10)$$

From data in Table 1 and formula (10) we obtained value of activity ratio:

$\frac{A_{U238}}{A_{U235}} \approx 0.297 \pm 0.028\ (\mathrm{Bq/Bq})$.

3.4. Determination of the uranium enrichment of material

By experiment, the activity ratios were obtained as 25.20 ± 2.1 (Bq/Bq) for A_{U234}/A_{u235} and 0.296 ± 0.028 (Bq/Bq) for A_{u238}/A_{u235}. Uranium enrichment of investigated sample was determined based on the activity ratio A_{U234}/A_{u235} and A_{u238}/A_{u235} and using formula (4).

The obtained result of uranium enrichment: q_{235} = (34.4 ± 3.1) %.

The main sources of the errors are statistical error, photopeak area determination, gamma ray self absorption, fitting procedure and nuclear data used.

4. Conclusion

In this work, the gamma-spectrometric technique was applied to determine precisely the uranium enrichment of highly enriched material up to 36%. This method doses not require the use of standard samples nor the knowledge of the detector absolute efficiency. It is also applicable for samples with any arbitrary geometrical shape. The uranium enrichment of investigated sample was calculated from the activity ratio ^{234}U/^{235}U and ^{238}U/^{235}U. The intrinsic efficiency calibration was used in determining

activities $^{234}U/^{235}U$ and $^{238}U/^{235}U$. The result obtained is in good agreement with estimated value (36%) from IAEA.

This paper is completed with finalcial support from Protect QG.TD. 12-02 of VNU.

References

[1] A Luca, Experimental Determination of the Uranium Enrichment Ratio, Rom. Journ. Phys, Vol. 53, No. 1-2, P35 -39, Bucharest, 2008.

[2] Haluk YÜcel, The applicability of MGA method for depleted and natural uranium isotopic analysis in the presence of actinides, Applied Radiation and Isotopies 65 (2007) 1269-1280.

[3] M.H. Nassef, W.EL Mowafi, and M.S.EL Tahawy, Non destructive assay for 235U determination in reference material of uranium oxide, Jornal of nuclear and Radiation Physics, Vol.4 No.2, 2009, pp 65-73.

[4] Y.Y. Ebaid, Use of gamma-ray spectrometry for uranium isotopic analysis in environmental samples, Rom. Journ. Phys. Vol.55,Nos 1-2 P69-74, Bucharest,2010.

[5] H. Yucel, H.Dikmen, Uranium enrichment measurements using the intensity ratios of self- fluresence X-ray-92* keV gamma ray in UXK_{α} spectral region, Talanta 78 (2009) pp 410-417.

[6] C.T. Nguyen, J. Zsigrai, Gamma-spectrometric uranium age-dating using intrisic efficiency calibration, Nucl. Instr. And Meth. B 243 (2006) 187.

[7] D.J. Karangelos anf et all, Determination of depleted uranium in environmental samples by gamma-spectroscopic techniques. Jour. of Environnmental Radioactivity 76 (2004) 294-310

[8] Tam.Ng.C and et al, Characterization of uranium-bearing malerial by passive non- destructive gamma spectrometry, Procce. of the 7th Confere. On Nucl. And Part. Phys. 11-15 Nov. 2009, Sham El- Sheikh, Egypt, 413-423..

[9] Delynn Clark, U235: A gamma ray analysis code for uranium isotopic determination, Lawrence Livermore National Laboratory, 1996

[10] E.Browne, R.B. Firestone, Table of Radioactive Isotopes, Wiley, New York, 1986.