VNU Journal of Science: Mathematics – Physics, Vol. 33, No. 1 (2017) 48-52

Intrinsic Efficiency Calibration for Uranium Isotopic Analysis in Soil Samples

Bui Van Loat[1,*], Somsavath Leuangtakoun[1,2], Cao Dang Luu[1], Bui Thi Hong[1], Khong Nam Khang[1], Nguyen The Nghia[1], Nguyen Hong Ha[1], Vu Thi Kim Duyen[3]

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

[2]*National University of Laos, P.O.Box: 7322, Dongdok, Vientiane, Laos*

[3]*Centre for Env. Trea. Tech. Minisry of Defence, 282 Lac Long Quan, Hanoi, Vietnam*

Received 12 January 2017
Revised 28 February 2017; Accepted 20 March 2017

Abstract: In this work, we present the results of using the non-destructive gamma spectroscopic method for uranium isotopic analysis and checking the status of radioactive equilibrium between ^{238}U and ^{226}Ra in soil samples. In order to analyze uranium isotopes and check the status of equilibrium between ^{238}U and ^{226}Ra the activity ratios $^{235}U/^{238}U$ and $^{238}U/^{226}Ra$ were measured. The these activity ratios were determined based on the characteristic gamma rays and using intrinsic (relative) detection efficiency calibration method. The results obtained shows that this suggested analytical method can be used to determine the uranium concentration in the case of the samples having arbitrary shapes and it does not require the use of any reference materials.

Keywords: Secular equilibrium, gamma-spectrometry, intrinsic efficiency calibration, MGA method.

1. Introduction

The activity ratio $^{235}U/^{238}U$ and status of equilibrium between ^{238}U and ^{226}Ra is one of the important parameters for analyses of geological and soil samples [1, 2]. When ^{238}U and ^{226}Ra in secular equilibrium, the activity ratio $^{238}U/^{226}Ra$ is equal to one. However, the ^{238}U is sometime radioactive disequilibrium with ^{226}Ra, then the activity ratio $^{238}U/^{226}Ra$ will be different from one. As it is difficult to interpret disequilibrium by simply comparing radiometric and chemical assay values of uranium, analyses should be made of the activity ratios.

The gamma spectrometry were employed to determine the activity ratios $^{238}U/^{226}Ra$ and $^{235}U/^{238}U$ in the geological and soil samples [2]. The our purpose is to use a gamma-spectroscopy with HPGe detector and intrinsic efficiency calibration method for checking the status of radioactivity equilibrium between the radioactive isotopes in ^{238}U series and for determining the activity ratio $^{235}U/^{238}U$ in the

*Corresponding author. Tel.: 84-912865869
Email: loat.bv58@gmail.com

geological and soil samples. The intrinsic efficiency calibration method was developed performance-based method MGA [3 - 5]. The activity of ^{235}U was determined from 185.75 keV gamma peak of ^{235}U and the activity of ^{238}U was determined from 1001.03 keV peak of ^{234m}Pa in equilibrium with ^{238}U [2]. The activity of ^{226}Ra was determined from peaks of ^{214}Bi and ^{214}Pb, which in equilibrium with ^{226}Ra.

2. Methodology

2.1. Calculation of activity ratios ^{214}Pb / ^{214}Bi and ^{238}U/ ^{214}Bi

There are many gamma transitions of ^{214}Bi have high branching ratios and they have energy respectively: 609.31 keV (46.10%), 806.17 keV (1.22%), 1120 keV (15.10%), 1377.67 keV (4.00%), 1509.49 keV(2.11%), 1729.59 keV (2.29%), 1764.49 keV (15.40%) [6]. These peaks-will be used to build the intrinsic efficiency function. The activity of ^{214}Pb was determined from 785.96 keV peak of ^{214}Pb and the activity of ^{238}U was determined from 1001.03 keV peak of ^{234}Pa. Based on relative efficiency calibration [3, 4], the activity ratios $^{214}Pb/^{214}Bi$ and $^{238}U/^{214}Bi$ determined by the following equations:

$$\frac{A_{Pb^{214}}}{A_{Bi^{214}}} = \frac{n_{785.96} \ / \ Br_{785.96}}{f_1(785.96)} \tag{1}$$

$$\frac{A_{U^{238}}}{A_{Bi^{214}}} = \frac{n(1001.03) / Br_{1001.03}}{f_1(1001.03)} \tag{2}$$

where A_i are the activities of ^{214}Pb, ^{214}Bi and ^{238}U isotopes respectively; $n_{1785.8}$, $n_{1001.03}$ and $Br_{785.96}$, $Br_{1001.03}$ are the net count rates and branching ratios corresponding to 1785.8 keV and 1001.03 keV gamma peaks; $f_1(E)$ is the intrinsic efficiency function, which was built from gamma peaks of ^{214}Bi; $f_1(785.95)$ and $f_1(1001.03)$ are values of function $f_1(E)$ at energy of 785.96 keV and 1001.03 keV respectively.

2.2. Calculation of activity ratios ^{238}U / ^{226}Ra and ^{235}U/ ^{238}U in the RGU sample

The activity ratios $^{238}U/^{226}Ra$ and $^{235}U/^{238}U$ was determined according to the following equations:

$$\frac{A_{U^{238}}}{A_{Ra^{226}}} = \frac{n(1001.02) / Br_{1001.02}}{f_2(1001.02)} \tag{3}$$

$$\frac{A_{U^{235}}}{A_{U^{238}}} = \frac{A_{U^{235}}}{A_{Ra^{226}}} \cdot \left[\frac{A_{U^{238}}}{A_{Ra^{226}}} \right]^{-1} = \frac{n_{185.712} \ / \ Br_{185.712}}{f_2(185.712)} \left[\frac{n_{1001.02} \ / \ Br_{1001.02}}{f_2(1001.02} \right]^{-1} \tag{4}$$

where $f_2(E)$ is the intrinsic efficiency calibration function, which was built from gamma peaks of ^{214}Bi and ^{214}Pb; $f_2(1001.03)$ and $f_2(185.712)$ are values of function $f_2(E)$ at 1001.03 keV and 185.75 keV of respectively.

2.3. Calculation of count rate of 185.712 gamma ray of ^{235}U in the 186 keV –peak

The 185.712 keV peak of ^{235}U and 186.21 keV peak of ^{226}Ra were overlapping peaks. The total peak of 186 keV were formed. The count rate of the total 186 keV photopeak can be expressed as:

$$n_{186} = n_{185.712} + n_{186.21} \tag{5}$$

where: $n_{185.712}$ and $n_{186.21}$ is count rate due to 185.712 keV gamma ray of ^{235}U and 186.21 keV gamma ray of ^{226}Ra in total 186 keV peak respectively.

Net count rate of gamma ray of 186.21 keV emitted from ^{226}Ra is determined by the formula:

$$n_{186.21} = f_2(186.21)\times Br_{186.21} \quad (6)$$

where $f_2(186.21)$ is value of the intrinsic efficiency calibration function at energy of 186.21 keV; $Br_{186.21}$ is branching ratio of 186.21 keV gamma ray emitted from ^{226}Ra.

From equations (5) and (6), the count rate $n_{185.712}$ is determined by the following formula:

$$n_{185.712} = n_{186} - n_{186.21} = n_{186} - f(186.21).Br_{186.21} \quad (7)$$

Branching factor, $Br_{185.71}$, is taken from [6], the count rate of 185.712 keV peak of ^{235}U can be determined.

3. Experimental results and discussions

3.1. Sample measurements

The US1 radioactive source and IAEA-RGU1 uranium ore reference soil sample were measured for checking the status of radioactivity equilibrium between the radioactive isotopes in ^{238}U series and for determination of the activity ratio ^{235}U/^{238}U in soil samples. The gamma spectra of samples and US1 radioactive source were taken by low background gamma spectroscopy using the GEM 40P4 HPGe detector (ORTEC). The detection efficiency of the GEM 40P4 detector is 20% relative to a 3”×3” NaI(Tl) detector and FWHM of 1.85 keV at peak 1.332 MeV of ^{60}Co. The US1 source was measured with three different configurations: G1: the surface of the source parallel to the detector surface; G2: The surface of the source perpendicularly to the surface of the detector and G3: sources covered in 1.5 mm thick lead and parallel to the detector surface.

An amount of 121 gram of the IAEA-RGU1 reference soil sampl was placed in polyethylenebox with diameter of 6,7 cm and height of 2.2 cm. The time needed for establishing secular equilibrium between ^{226}Ra with ^{214}Bi and ^{214}Pb is about 4 weeks. 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 1001.03 keV of ^{234m}Pa dropped below 1.5%.

3.2. Checking the status of equilibrium between the radioactive isotopes in ^{238}U series

To check the status of equilibrium between the radioactive isotopes in ^{238}U series going to measure the activity ratios ^{214}Pb / ^{214}Bi and ^{238}U/ ^{214}Bi. These activity ratios were determined by the formulas (1) and (2). The function $f_1(E)$ is obtained by fitting a second order polynomial to relative efficiencies at 609.31 keV, 806.17 keV, 1120 keV, 1377.67 keV, 1509.49 keV, 1729.59 keV, 1764.49 keV peaks of ^{214}Bi (Fig.1). Table 1 shows the calculated activity ratios ^{214}Pb / ^{214}Bi and ^{238}U/ ^{214}Bi corresponding to three different configurations: G1, G_2, G_3.

From Fig. 1 and table 1 to see that: three different different measurement configuration, the relative efficiency calibration curves are different forms, however the results of the activity ratio ^{214}Pb / ^{214}Bi and ^{238}U/^{214}Bi determined by three measuring configurations is almost the same. In the US1 radioactive source the radioactive isotopes in ^{238}U series are in radioactive equilibrium status.

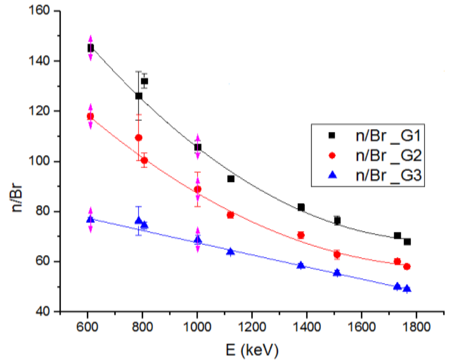

Fig 1. The relative efficiency curve is constructed based on gamma peaks of ^{214}Bi corresponding to three different configurations: G1,G_2,G_3.

Table1**.** The results determine the activity ratios ^{214}Pb/^{214}Bi and ^{238}U/^{214}Bi corresponding to three different configurations: G1, G_2, G_3.

The activity ratios	G1	G2	G3
^{214}Pb/^{214}Bi	1.02 ± 0.02	0.99 ± 0.03	1.02 ± 0.03
^{228}U/^{214}Bi	0.99 ± 0.02	1.02 ± 0.03	0.99 ± 0.03

3.3. Measuring radioactivity ratios ^{238}U/^{226}Ra and ^{235}U/^{226}Ra

The activity ratios ^{238}U / ^{226}Ra and ^{235}U/ ^{238}U were determined by the formulas (3) and (4). The function $f_2(E)$ is obtained by fitting a second order polynomial to relative efficiencies at 295.22 keV, 351.93 keV, 785.96 keV peaks of ^{214}Pb and 609.31 keV, 665.45 keV, 806.17 keV of ^{214}Bi (Fig. 2). is derived as follows:

$$f_2(E) = 186.99 - 0.13647 \times E + 0.0000361 \times E^2 \quad (7)$$

with R^2 = 0.9997, where E is the energy in keV.

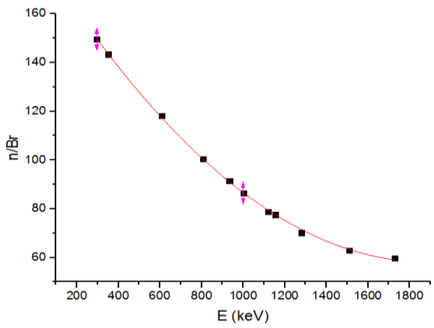

Fig 2. The relative efficiency curve is constructed based on gamma peaks of ^{214}Bi and ^{214}Pb.

The value of $f_2(1001.03)$ was calulated from equation (7) and found to be 86.59. From analysis of the gamma spectra of RGU1 sample, the ratio $n_{1001.03}/Br_{1001.03}$ was found to be 86.47. is: The *obtained value* of the activity ratios ^{238}U / ^{226}Ra is as *follows:*

$$\frac{A_{U^{238}}}{A_{Ra^{226}}} = \frac{n(1001.02)/Br_{1001.02}}{f_2(1001.02)} = 0,999 \simeq 1.$$

The results obtained shows that the RGU-1 sample contains ^{226}Ra in equilibrium with ^{238}U.

3.4. Determination of activity ratio $^{235}U/^{238}U$:

Value of $f_2(186.21)$ = 162.833 and $f_2(185.75)$ = 162.889. From analysis gamma spectrum of RGU1 sample we have: n_{186} = 10.06 (count/s). The $Br_{186.21}$ = 0.0356 [3], the count rate $n_{185.712}$ is determined by the following:

$$n_{185.712} = n_{186} - n_{186.21} = n_{186} - f(186.21).Br_{186.21} = 4,26(count/s).$$

The $Br_{186.75}$ = 0.572 [2, 6], the activity ratios^{235}U/ ^{238}U was determined according to the following equation:

$$\frac{A_{U^{235}}}{A_{U^{238}}} = \frac{n_{185.712}/Br_{185.712}}{f_2(185.712)}\left[\frac{n_{1001.02}/Br_{1001.02}}{f_2(1001.02}\right]^{-1} = \frac{4.26/0,572}{162.889}.\frac{86.59}{86.47} = 0,0457$$

The activity concentrations of ^{235}U and ^{238}U in RGU-1 sample are $(228 \pm 2)Bq/kg$ and $(4940 \pm 30)Bq/kg$ respectively [7]. The activity ratio ^{235}U/^{238}U in this sample is 0.0462.

The our result is in good agreement with estimated value from IAEA. The main sources of the uncertainties for the obtained results are due to statisticcal errors: 1.5%; the fitting relative efficiency curve 1.5%; the gamma branching ratio 1%.

4. Conclusion

In this work, the gamma-spectrometric technique was applied for uranium isotopic analysis and checking the status of equiblimum between the radioactive isotopes in ^{238}U series. The intrinsic efficiency calibration was used in determining the activity ratios ^{235}U/^{238}U and ^{238}U/^{226}Ra. 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.

References

[1] M. Omar, M.S. Hamzah and A.K. Wood - Radiacative disequilibirum and total activity concentration of norm waste- JNUCL & Rel.TECH , volume 5, No 2, December 2008, p 47-56.

[2] Y.Y. Ebaid- Use of gamma-ray spectrometry for uranium isotopic analysis in environmental samples. Rom.Journ.Phys., Vol 55 , Nos 1-2, P 69-74, Bucharest, 2010

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

[4] 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., Sham El- Sheikh, Egypt, 413-423, (11-15 Nov. 2009).

[5] B.V.Loat and et al, Enrichment determination of low – enriched by gamma spectroscopic method. Nuclear Science and Technology. Vol 4, No. 2 (2014) pp 01-06.

[6] http://www.nucleonica.net.

[7] https://nucleus.iaea.org/rpst/referenceproducts/referencematerials/radionuclides/IAEA-RGU-1.htm.