

Signature Verification of $^{235}\text{U}/^{238}\text{U}$ Activity Ratio for Ores and Natural Samples Using γ -Ray Spectrometry and a Derived Equation

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Abstract—The activity ratio of uranium in unknown samples and materials is a crucial signature tool for verification, assay, and tracking of their origin. This paper deals with using γ -ray spectrometric analysis and a new derived formula to calculate the $^{235}\text{U}/^{238}\text{U}$ activity ratio. Reference IAEA materials and a highly efficient calibrated HPGe spectrometer were used for the verification and validation of this equation and the results of assay. The calculated specific activities and $^{235}\text{U}/^{238}\text{U}$ activity ratios show very good agreement with the declared reference values, 0.046, with differences less than 2%. Natural origin samples were also investigated and assayed with the maximum deviation in the range 1.3–2.9%. Any potential deviation from the reference natural value, 0.046, in uranium samples could be precisely detected, identified, assessed, and quantified using the suggested system and equation.

Keywords: equation derivation, U isotopic ratio, natural origin samples, γ -ray spectrometry

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INTRODUCTION

Uranium is a terrestrial naturally occurring radioactive material which always exists in rocks and soils in low quantities [1]. Uranium is found in the earth's crust with an average content of 3 ppm. Natural uranium (NU) is a mixture of three radioactive isotopes: ^{238}U (99.27% abundance), ^{235}U (0.72% abundance), and ^{234}U (0.0054% abundance) [2]. Enrichment processes are widely practiced to produce enriched uranium (EU) of activity ratio higher than 0.046, used for the production of nuclear energy. On the other hand, depleted uranium (DU) with the activity ratio lower than 0.046 is also produced as a by-product of this enrichment processes. Accordingly, monitoring the isotopic $^{235}\text{U}/^{238}\text{U}$ ratio is a very important step associated with any sample that contains uranium [2]. Moreover, the U along with Pb isotope measurements can be used to identify and ensure the geological age of minerals and rocks [3, 4]. The detection of illicit trafficking of nuclear materials,

including “nuclear smuggling,” is necessary in order to determine the possible origin of the nuclear material. Investigations are carried out by laboratories capable of handling and analyzing nuclear material of unknown origin [5]. During the past few years, as the international illicit traffic of radioactive/fissionable materials has increased, it has become imperative to have the capability to apply fast and reliable methods for determining the uranium enrichment percentage [6].

This study is mainly focused on verifying and validating the $^{235}\text{U}/^{238}\text{U}$ activity ratios for natural origin samples as a signature identification tool using γ -ray spectrometry and derived equation.

THEORETICAL DERIVATION OF THE EQUATION AND CALCULATIONS

The activity of an isotope can be calculated from the following relation [7–9]:

$$A = N\lambda, \quad (1)$$

where A is the activity, N is the number of atoms present, and λ is the decay constant. So, the activity ratio can be expressed as follows:

$$\frac{A_{235}}{A_{238}} = \frac{N_{235} \lambda_{235}}{N_{238} \lambda_{238}}. \quad (2)$$

The decay constant is $\lambda = \ln 2/T_{1/2}$, where $T_{1/2}$ is the half-life of the isotope; thus,

$$\frac{A_{235}}{A_{238}} = \frac{N_{235} T_{1/2(238)}}{N_{238} T_{1/2(235)}}. \quad (3)$$

The uranium enrichment percentage (E , %) is defined as follows:

$$E = \frac{N_{235}}{N_{238} + N_{235} + N_{234}}. \quad (4)$$

In the case of natural or depleted uranium, the N_{235} and N_{234} values are relatively low in comparison to N_{238} and can be neglected in Eq. (4); thus,

$$E = \frac{N_{235}}{N_{238}}. \quad (5)$$

By substituting Eq. (5) in Eq. (3), we obtain

$$\frac{A_{235}}{A_{238}} = E \frac{T_{1/2(238)}}{T_{1/2(235)}}. \quad (6)$$

The ^{235}U percentage in natural uranium is 0.72% [10]. The ^{235}U half-life is $T_{1/2(235)} = 7.04 \times 10^8$ years, and that of ^{238}U is 4.468×10^9 years [10, 11].

By substituting these values into Eq. (6), we obtain

$$\frac{A_{235}}{A_{238}} = \frac{0.72}{100} \times \frac{4.468 \times 10^9}{7.04 \times 10^8}. \quad (7)$$

Finally, the $^{235}\text{U}/^{238}\text{U}$ activity ratio of can be formulated as

$$\frac{A_{235}}{A_{238}} = 0.0457. \quad (8)$$

This ratio is almost the same as the certified activity ratio of uranium (0.046) in any natural origin samples with about 0.65% relative difference to this value (0.0457).

EXPERIMENTAL

The radiometric measurement using γ -ray spectrometry is considered to be an essential technique for the analysis of uranium in ores and rock samples. This is attributed to

several reasons: (i) it is both simple and cheap for routine analysis, (ii) the samples are measured directly without any chemical or other treatments, (iii) this technique is essential in studying the geochemical prospecting and evaluation of the radioactive mineral deposits, (iv) it has high resolution and progressively improved performance, and (v) it is considered to be the easiest and cleanest method for measuring U concentrations [10, 12].

HPGe Detector Characteristics

A hand-held radioisotope identifier system (TRANS-SPEC) was used in this study for the rapid identification of radioisotopes. The detector consists of a p -type HPGe detector with 40% efficiency. The energy resolution of the detector was found to be ~ 1.32 and 2.2 at 122 and 1332.5 keV gamma lines, respectively. Control of the acquisition is achieved using a built-in digital electronic system (DigiDart). A Hymatic Stirling cycle cooler system is designed for continuous operation. The count rate and position (geometry) of sample relative to HPGe detectors were fixed during the measurements of the samples.

γ -Ray Spectrometer Energy and Efficiency Calibration

Before the spectrum analysis, both the energy and efficiency calibration of the measuring system were performed using ^{60}Co , ^{137}Cs , ^{152}Eu , and ^{133}Ba point sources. These radioactive isotopes have well-known activities and are routinely used for verification and validation of calibration results. The point sources were positioned on the top at the source-to-detector distance of 25 cm. The counting time was 1800 s (30 min) to obtain high counts with good statistics. In this study, both energy and efficiency calibration of the HPGe system were performed to ensure the accuracy of the detector and, consequently, of the spectrum analysis.

Energy calibration. The energy calibration of the HPGe detector system was performed in the range 53.16–1408.08 keV to obtain a relationship between the channel number of the peak position in the spectrum and the corresponding γ -ray energy. The linear relationship between the channel number and energy is plotted in Fig. 1. This relationship can be described by linear Eq. (9):

$$\text{Energy (keV)} = 0.3455 \times (\text{channel}) + 0.0266. \quad (9)$$

The radionuclides in reference material and natural samples can then be identified based on their energies. Efficiency calibration. The absolute photopeak efficiency

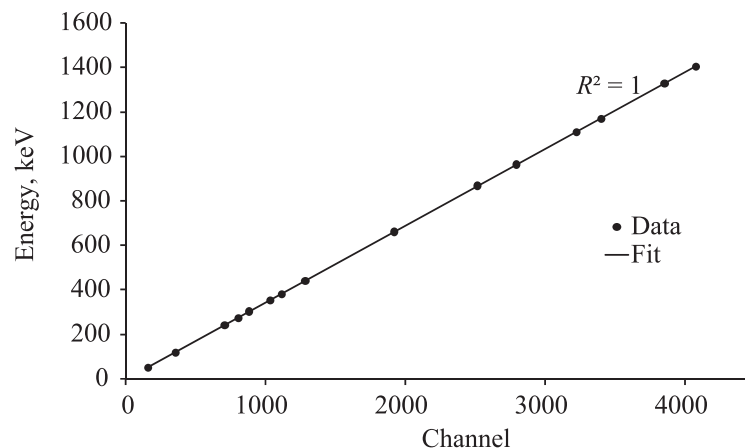


Fig. 1. Energy calibration curve of HPGe spectrometer.

ξ_{abs} (%), which depends on the source–detector geometry and the gamma energy, was measured and calculated using the following equation [13]:

$$\xi_{\text{abs}} = \frac{C_t}{N_\gamma} \times 100, \quad (10)$$

where C_t is the total number of counts recorded per unit time, and N_γ is the number of gamma quanta emitted by the source per unit time; it can be calculated using the equation [14]

$$N_\gamma = D_S I_\gamma(E_\gamma), \quad (11)$$

where D_S is the certified activity or the disintegration rate of the source, and $I_\gamma(E_\gamma)$ is the branching ratio or the emission probability per disintegration.

Due to the different geometry and density for the reference material and natural samples, the relative efficiency curves established for different sets of samples were used as reference efficiency curves in Angel-3 software together with the characteristics of the detector crystal, source container, and source–detector geometry [15–18].

The above-mentioned steps are considered to generate different efficiency curves for the same detector but for different containers and geometries.

Using the Excel sheet, the efficiency curves of the detector were plotted vs. different photopeak energies for a point source and densities of 1.6 and 2.5 g/cm³ (Fig. 2).

Sample Preparation and Readiness for Assay

Two sets of samples were taken. The first set included IAEA-RGU-1 and IAEA-RGTh-1 reference materials

(323 g) prepared on behalf of IAEA by the Canada Centre for Mineral and Energy Technology. The samples were prepared by dilution of BL-5 uranium ore (7.09% U) and OKA-2 thorium ore (2.89% Th, 219 μg U/g) with floated silica powder of similar grain size distribution. No evidence for between-bottles inhomogeneity was detected after mixing and bottling. BL-5 has been certified for uranium; the ^{226}Ra and ^{210}Pb measurements confirmed that it was in radioactive equilibrium. The agreement between radiometric and chemical measurements of thorium and uranium in OKA-2 showed that both series were in radioactive equilibrium [19].

The second set included two groups of natural samples. The first group consisted of seven environmental phosphate samples collected from Aswan (coded PS1–PS7). The second group consisted of seven black sand samples collected from the beach of Rosetta–Damietta (coded BS1–BS7). All the natural samples were dried at 100°C and then kept in a plastic cup for a period of more than 28 days. The samples were then counted three times for 24 h using the considered γ -ray spectrometry system.

RESULTS AND DISCUSSION

The γ -transitions emitted from the samples lie in a wide range of energies from approximately 50 keV to 3 MeV, covering the naturally occurring radioactive materials (^{238}U , ^{235}U , ^{232}Th decay series and ^{40}K). The ^{238}U specific activity was determined from γ -rays emitted by its progenies: ^{234}Th (determined from the 63 keV photopeak, $I_\gamma = 0.27$) [20] and $^{234\text{m}}\text{Pa}$ (determined from the 1001 keV photopeak, $I_\gamma = 1.021$, which is the most

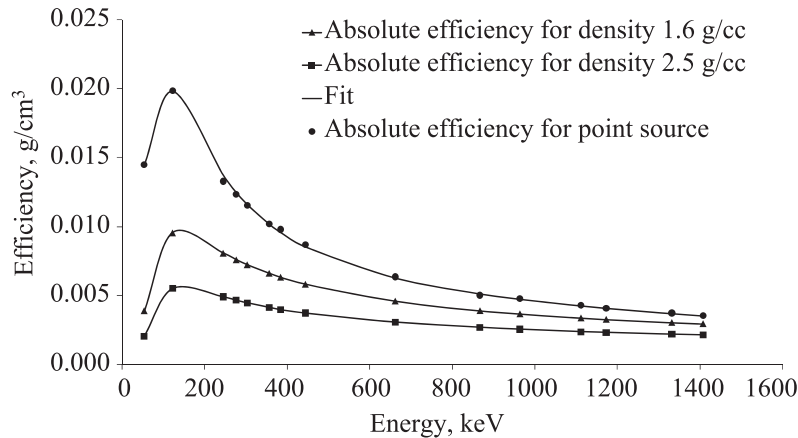


Fig. 2. Relative efficiency for different geometries and densities of samples.

suitable peak for the analytical purposes) [21]. Further decay leads to ^{226}Ra and then to radon. Afterwards, the average of the specific activity of ^{214}Pb and ^{214}Bi , which can be measured using 295.2 ($I_\gamma = 18.414$) and 351.9 keV ($I_\gamma = 35.60$) lines for ^{214}Pb and the 609.3 keV ($I_\gamma = 45.49$) line for ^{214}Bi , can then be taken into consideration [22, 23]. Furthermore, a theoretical calculation of the ^{226}Ra specific activity can be performed by assuming that the entire 186.21 keV ($I_\gamma = 3.555$) peak is due to ^{226}Ra . Then, the following correction factor can be used to correct the ^{226}Ra value [14, 24]:

$$\left(\text{Corrected}^{226}\text{Ra}\right) = 0.5709 \left(\text{Apparent}^{226}\text{Ra}\right). \quad (12)$$

The ^{235}U activity can be estimated at the gamma energy of 185.72 keV [24, 25]:

$$\left(\text{Estimated}^{235}\text{U}\right) = 0.02662 \left(\text{Apparent}^{226}\text{Ra}\right). \quad (13)$$

Also, the γ -energies of ^{235}U (143.7, 163.3 and 205.3 keV) can be used for the activity calculations [22, 26]. Both ^{40}K (1460.83 keV) and ^{232}Th series (583, 911, 968 keV) [27] were observed in the spectra of the analyzed samples before treatment. However, it should be noted that their activities are not considered in this study. The specific activities (A) of the samples are given in Bq/kg for ^{238}U and ^{235}U (A_{238} and A_{235}). They are estimated by substituting the corresponding count rate of the γ -lines in Eq. (14) [28]:

$$A = \frac{C_{\text{net}}}{\xi_{\text{abs}} I_\gamma (E_\gamma) M}, \quad (14)$$

where M is the mass of the measured sample (kg), ξ_{abs} is the absolute detector efficiency at specific energy E_γ with branching ratio $I_\gamma (E_\gamma)$, and C_{net} is the net count

rate in a given photopeak region (ROI) subtracting the background, which is corrected using Eq. (15) [29, 30]:

$$C_{\text{net}} = \frac{C}{1 - (C\tau)}, \quad (15)$$

where C is the measured count rate in the peak of interest in the sample spectrum [s^{-1}], and τ is the analog-to-digital converter (ADC) dead time [s].

For all the measurements, the sample to detector distance for the assayed samples is optimized to keep the dead time correction below 2%.

The Gamma Vision software determines the total uncertainty σ_t of activity by summing in quadrature the individual uncertainties from various analysis components as in Eq. (16):

$$\sigma_t = \sqrt{\sigma_{\text{count}}^2 + \sigma_{\text{nor}}^2 + \sigma_{\text{rsum}}^2 + \sigma_{\text{abs}}^2 + \sigma_{\text{nuc}}^2 + \sigma_{\text{eff}}^2 + \sigma_{\text{geo}}^2 + \frac{\sigma_{\text{uni}}^2}{3}}, \quad (16)$$

where σ_{count} is the counting uncertainty estimate, σ_{nor} is the additional normally distributed uncertainty estimate such as balance resolution, σ_{rsum} is the random summing uncertainty estimate, σ_{abs} is the absorption uncertainty estimate, σ_{nuc} is the nuclide uncertainty estimate, σ_{eff} is the efficiency uncertainty estimate, σ_{geo} is the geometry uncertainty estimate, and σ_{uni} is the uniformly distributed uncertainty estimate such as sample mass determination.

Validation of the Derived Activity Ratio Equation Using IAEA References

For the IAEA-RGU-1 samples, the specific activity of ^{238}U was measured and found to be 4869.6 Bq/kg, and the specific activity of ^{235}U was measured as 229 Bq/kg.

Table 1. Measured and certified specific activity (Bq/kg) and $^{235}\text{U}/^{238}\text{U}$ ratios of the IAEA reference materials

Sample	^{238}U specific activity, Bq/kg		^{235}U specific activity, Bq/kg		Measured $^{235}\text{U}/^{238}\text{U}$ ratio	Certified $^{235}\text{U}/^{238}\text{U}$ ratio	Deviation, %
	measured	certified value	measured	certified value			
IAEA-RGU-1	48670 ± 150	4910–4970	229 ± 5	226–230	0.0470	0.046	1.7
IAEA-RGTh-1	79 ± 3	72–84	3.7 ± 0.11	3.3–3.9	0.0468		1.4
Average					0.0469		1.55

Table 2. Specific activity of ^{238}U daughters in phosphate and black sand samples

Sample	Average specific activity (Bq/kg) for ^{238}U daughters						Ratios of ^{234}Th , $^{234\text{m}}\text{Pa}$, and ^{226}Ra to average of ^{214}Pb and ^{214}Bi			
	^{234}Th	$^{234\text{m}}\text{Pa}$	^{226}Ra (calculated)	^{214}Pb	^{214}Bi	average of ^{214}Pb and ^{214}Bi	^{234}Th : average	$^{234\text{m}}\text{Pa}$: average	^{226}Ra : average	
Phosphate	PS1	389 ± 18	396 ± 11	401 ± 17	395 ± 11	379 ± 10	387 ± 10	1.01	1.02	1.04
	PS2	461 ± 27	449 ± 14	439 ± 18	476 ± 13	459 ± 12	467 ± 12	0.99	0.96	0.94
	PS3	356 ± 17	370 ± 16	360 ± 15	373 ± 10	357 ± 9	365 ± 10	0.98	1.01	0.99
	PS4	359 ± 24	373 ± 16	376 ± 16	370 ± 12	358 ± 9	364 ± 10	0.99	1.02	1.03
	PS5	383 ± 18	398 ± 26	412 ± 18	397 ± 6	379 ± 10	388 ± 8	0.99	1.03	1.06
	PS6	387 ± 24	335 ± 16	381 ± 16	394 ± 12	377 ± 10	385 ± 11	1.01	0.87	0.99
	SP7	398 ± 23	386 ± 17	398 ± 16	395 ± 10	380 ± 10	387 ± 10	1.03	0.99	1.02
Average	391 ± 22	387 ± 17	395 ± 17	400 ± 11	384 ± 10	392 ± 10	1.00	0.99	1.01	
Blacksand	BS1	463 ± 29	588 ± 21	522 ± 16	464 ± 8	479 ± 20	472 ± 15	0.98	1.25	1.11
	BS2	469 ± 27	443 ± 20	520 ± 18	482 ± 8	496 ± 20	489 ± 15	0.96	0.91	1.06
	BS3	467 ± 28	472 ± 22	482 ± 14	481 ± 8	469 ± 19	475 ± 15	0.98	0.99	1.02
	BS4	461 ± 28	494 ± 22	484 ± 18	476 ± 8	454 ± 19	465 ± 14	0.99	1.06	1.04
	BS5	455 ± 28	497 ± 22	498 ± 12	482 ± 8	463 ± 19	473 ± 15	0.96	1.05	1.05
	BS6	464 ± 27	507 ± 22	492 ± 17	464 ± 8	480 ± 20	472 ± 15	0.98	1.07	1.04
	BS7	459 ± 27	446 ± 21	497 ± 9	455 ± 8	479 ± 20	467 ± 15	0.98	0.95	1.06
Average	492 ± 28	499 ± 21	472 ± 15	474 ± 8	473 ± 19	492 ± 15	0.98	1.01	1.06	

The resulting $^{235}\text{U}/^{238}\text{U}$ ratios on the average are 0.0470 with the deviation of about 1.7 %. For the IAEA-RGTh-1 samples, the calculated specific activity of ^{238}U was 79 Bq/kg, and the specific activity of ^{235}U was found to be 3.7 Bq/kg. The resulting average $^{235}\text{U}/^{238}\text{U}$ ratio is 0.0468 with the deviation of only 1.4%. All the measured and calculated specific activities are in very acceptable agreement with the certified value of 0.46, as shown in Table 1.

Verification of Activities and U Ratio of the Natural Samples

The specific activities of ^{235}U and ^{238}U have been measured in two different groups of the natural samples, and their uranium isotopic contents and ratios were determined.

The average specific activity of phosphate samples, based on ^{234}Th , $^{234\text{m}}\text{Pa}$, ^{226}Ra , and average of ^{214}Pb and ^{214}Bi (^{238}U daughters), was found to be 391 ± 22, 387 ± 17, 395 ± 17, and 392 ± 10, respectively; that is, the ratios of ^{234}Th , $^{234\text{m}}\text{Pa}$ and ^{226}Ra to the average of ^{214}Pb and ^{214}Bi are 1.00 : 1, 0.99 : 1, and 1.01 : 1, respectively. The average specific activity of black sand samples, based on ^{234}Th , $^{234\text{m}}\text{Pa}$, ^{226}Ra , and average of ^{214}Pb and ^{214}Bi , was found to be 492 ± 28, 499 ± 21, 472.27 ± 15, 474 ± 8, and 473 ± 19, respectively; that is, the ratios of ^{234}Th , $^{234\text{m}}\text{Pa}$, and ^{226}Ra to the average of ^{214}Pb and ^{214}Bi are 0.98 : 1, 1.01 : 1, and 1.06 : 1, respectively (Table 2).

The ^{235}U specific activity in phosphate and black sand samples ranges from 16.8 to 20.6 Bq/kg with the average value of 19.5 Bq/kg and from 20.8 to 22.6 Bq/kg

Table 3. ^{235}U specific activity measured for different γ -lines

Sample		^{235}U specific activity, Bq/kg, based on indicated for indicated γ -line, keV				
		185.71 (calculated)	143.76	163.33	205.31	average
Phosphate	PS1	18.02 ± 0.17	17.9 ± 0.9	17.3 ± 0.5	18.2 ± 0.3	17.9 ± 0.5
	PS2	20.6 ± 1.0	19.8 ± 0.3	19.3 ± 1.3	22.7 ± 0.8	20.6 ± 0.9
	PS3	16.3 ± 0.5	16.60 ± 0.19	17.00 ± 0.20	17.3 ± 0.5	16.8 ± 0.4
	PS4	15.3 ± 1.5	16.8 ± 1.0	16.2 ± 0.6	18.9 ± 2.4	16.8 ± 1.5
	PS5	18.0 ± 0.5	17.77 ± 0.25	18.4 ± 0.9	18.5 ± 1.0	18.2 ± 0.7
	PS6	18.6 ± 1.2	17.7 ± 1.1	17.1 ± 0.3	17.9 ± 1.2	17.8 ± 1.0
	PS7	18.4 ± 1.2	17.7 ± 1.4	18.7 ± 1.6	18.0 ± 1.1	18.2 ± 1.3
Black sand	BS1	20.50 ± 0.21	20.9 ± 0.6	20.4 ± 0.4	19.9 ± 0.4	20.8 ± 0.4
	BS2	18.7 ± 2.4	24 ± 3	21.32 ± 0.08	21.14 ± 0.25	21.4 ± 2.1
	BS3	19.5 ± 0.8	23 ± 3	22.5 ± 2.4	22 ± 3	21.3 ± 2.3
	BS4	20.1 ± 1.6	22.3 ± 0.58	22.4 ± 0.6	20.7 ± 1.1	21.4 ± 1.0
	BS5	19 ± 3	25 ± 3	24.4 ± 2.0	21.7 ± 0.8	22.6 ± 2.2
	BS6	20.63 ± 0.14	21.2 ± 0.7	19.3 ± 1.1	22.6 ± 2.3	20.9 ± 1.4
	BS7	18.1 ± 2.4	21.30 ± 0.18	26 ± 5	20.9 ± 0.6	21 ± 3

with the average value of 21.6 Bq/kg, respectively (Table 3).

For the phosphate samples, the specific activity of ^{238}U varied from 365 to 467 Bq/kg with an average value of 412 Bq/kg. Thus, the average $^{235}\text{U}/^{238}\text{U}$ activity ratio is 0.0460 with a deviation of about 1.30% in maximum.

For black sand samples, the specific activity of ^{238}U varied between 465 and 489 Bq/kg with an average value of 473 Bq/kg. Thus, the average $^{235}\text{U}/^{238}\text{U}$ activity ratio is 0.0452 with the deviation of about 2.70% in maximum. The results are given in Table 4.

These values show a slight variation in the natural uranium percentage of the analyzed samples with an

Table 4. Calculated specific activity (Bq/kg) and $^{235}\text{U}/^{238}\text{U}$ activity ratio in the natural samples

Sample		^{238}U specific activity, Bq/kg	^{235}U specific activity, Bq/kg	Measured $^{235}\text{U}/^{238}\text{U}$ ratio	Certified $^{235}\text{U}/^{238}\text{U}$ ratio	Deviation, %
Phosphate	PS1	387 ± 10	17.9 ± 0.5	0.0462	0.0460	0.38
	PS2	467 ± 12	20.6 ± 0.9	0.0441		4.24
	PS3	365 ± 10	16.8 ± 0.4	0.0460		0.02
	PS4	364 ± 10	16.8 ± 1.5	0.0460		0.09
	PS5	388 ± 8	18.2 ± 0.7	0.0468		1.74
	PS6	385 ± 11	17.8 ± 1.0	0.0462		0.50
	PS7	387 ± 10	18.2 ± 1.3	0.0470		2.15
	Average	392 ± 10	18.9 ± 1.0	0.0460	1.30	
Black sand	BS1	472 ± 15	20.8 ± 0.4	0.0441	4.15	
	BS2	489 ± 15	21.4 ± 2.1	0.0437	4.93	
	BS3	475 ± 15	21.3 ± 2.3	0.0449	2.35	
	BS4	465 ± 14	21.4 ± 1.0	0.0460	0.10	
	BS5	473 ± 15	22.6 ± 2.2	0.0477	3.71	
	BS6	472 ± 15	20.9 ± 1.4	0.0444	3.58	
	BS7	467 ± 15	21 ± 3	0.0467	0.08	
	Average	492 ± 15	21.6 ± 1.9	0.0452	2.70	
Total average		440 ± 8	20.40 ± 1.52	0.0455		2.00

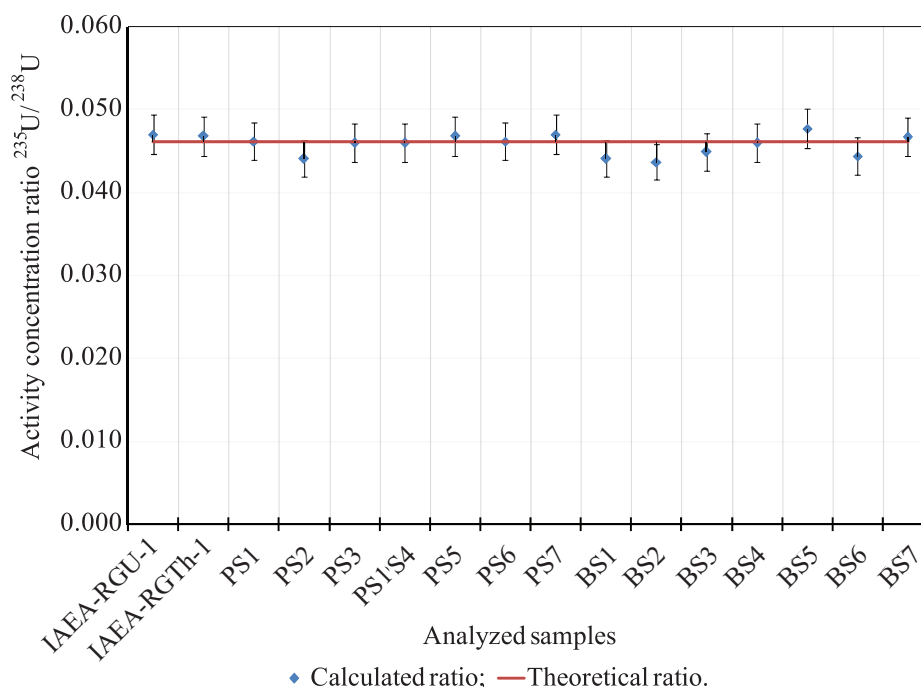


Fig. 3. $^{235}\text{U}/^{238}\text{U}$ activity ratios for the verified reference material and natural samples.

average value within the normal signature value, 0.72%. This acceptable variation could be attributed to the change in the detector efficiency at different gamma transitions of ^{235}U (143.7, 163.3, 185.7, and 205.3 keV) and ^{238}U (63.766, 185.99, 295.2, 609.3, and 1001 keV). The calculated $^{235}\text{U}/^{238}\text{U}$ activity ratios for the reference IAEA materials and natural samples are depicted against the certified value, 0.046, as shown in Fig. 3. The deviation between the measured and calculated/derived activity ratio values is low (<2%).

Thus, there is good agreement between the theoretical calculation [Eq. (15)] and the certificate values. The average difference does not exceed 1.6% for the reference IAEA materials and about 2.0% for the natural origin samples.

CONCLUSION

The investigation of origin and history of unknown samples by back-tracing approach is an effective tool for identifying undeclared radiological and nuclear activities. This paper introduces a developed mathematical formula for the calculation and verification of $^{235}\text{U}/^{238}\text{U}$ activity ratio based on efficient γ -spectrometric analysis measurements. This equation was validated by using reference IAEA materials. The results show deviation of less than 2% from the certified values. The verification

of uranium isotopes and activity ratios of natural samples was also carried out with average differences from 1.3 to 2.7%. It may be concluded that the abnormal and deviated activities from the natural origin and signature can be followed and verified by our derived equation and accurate γ -ray spectrometric investigation.

CONFLICTS OF INTEREST

The authors declare that they have no conflicts of interest.

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