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Revision of nuclear data of ^{235}U and ^{226}Ra for the 186-keV gamma-ray peak for the determination of activity in environmental samples

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Abstract In this work, the ratio contribution of ^{226}Ra and ^{235}U in γ spectra with the latest update nuclear data will be revised. The results showed that in the total count rate of the 186 keV peak consists of 57.2% of ^{226}Ra (186.2 keV) and 42.8% of ^{235}U (185.7 keV) with the existence of equilibrium. These calculations were used for determining mass activity of ^{238}U for the direct and fast measurement using only the 186 keV peak and verifying the ^{238}U–^{226}Ra secular equilibrium. An equilibrium method was measured secular equilibrium of decay chain ^{238}U (by the product daughter ^{214}Bi-609.3 keV) that was used for this study. The maximum relative deviation was less than 4.0% between both methods proved the direct method was reliable and can be applied to determine the mass activity of ^{238}U in the radioactive equilibrium samples.

Keywords Direct method · Equilibrium method · Secular equilibrium · Gamma spectrometry

Introduction

Natural uranium consists of three isotopes ^{238}U (99.2745%), ^{235}U (0.72%) and ^{234}U (0.0055%). It is conceivable that uranium within samples that originate from nuclear industry in various fields such as nuclear energy, nuclear weapons, nuclear medicine, etc. The production and using uranium have to comply with the safety radiation. The monitoring ratio activity ^{235}U–^{238}U was used to warn a leakage of uranium into the environment.

The γ spectrometry using HPGe detector has been demonstrated as a principal quantitative analysis technique for measuring of the radioactive environmental samples at many laboratories in the world. The activity of ^{235}U is determined directly from the peak of 185.7 keV energy (γ intensity 57%). The other peaks such as 143.8 keV (10.94%), 163.4 keV (5.08%) and 205.3 keV (5.02%) are not commonly used to determine the activity of ^{235}U because their intensity γ lower than intensity of peak 185.7 keV and the counting rates due to the peaks are often below the detection limits of the HPGe detector [1]. The activity of ^{238}U calculates based on the activities of ^{214}Pb (295.2, 351.9 keV), ^{214}Bi (609.3, 1120.3, 1764.5 keV) after waiting a time deposition sample about 30 days (achieving secular equilibrium of the decay chain ^{238}U) [2]. This method is only successful if the container sample seals off the laboratory for at least 30 days so that no radon (^{222}Rn, the half-life 3.825 days) escape to avoid disequilibrium problems between ^{226}Ra and ^{214}Pb, ^{214}Bi, its respective progenies [3]. The disadvantages of it require the prepared complex of manufacturing sample and take a long waiting time. Another method is direct using the peaks such as 63.3 keV (^{234}Th, 3.75%) and 186.2 keV (^{226}Ra, 3.555%). The contribution of X-ray from lead and the self-absorption of a sample would influence the result activity of ^{238}U at the peak 63.3 keV (^{234}Th) [4, 5]. Therefore, the peak 186.2 keV (^{226}Ra) was chosen to calculate activity of ^{238}U using the direct method.

When the environmental samples reached a secular equilibrium of the decay chain ^{238}U, the activity of ^{226}Ra is considered the equal activity of ^{238}U. These samples had a

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containing both ^{226}Ra (186.2 keV, 3.555%) and ^{235}U (185.7 keV, 57%) of count rate at the region 186 keV. Ebaid et al. determined the ratio of contribution ^{226}Ra and ^{235}U with correction factors of 58.3 and 41.7%, respectively [6]. Gilmore evaluated this ratio include 57.09% (^{226}Ra) and 42.91% (^{235}U) at the region 186 keV [7]. Hung et al. used the RGU standard with correction factors of 57.09% to correct the ^{226}Ra value and built the standard efficiency curve for HPGe detector in the energy range from 46.5 to 2204.2 keV [8]. In this work, we calculated the ratio contribution of ^{226}Ra and ^{235}U at the region 186 keV with the latest nuclear data. These results are used to perform the standard efficiency curve for HPGe detector and determined mass activity of ^{235}U and ^{238}U in geological samples using the direct method. In addition, the equilibrium method of the decay chain ^{238}U was also applied to determine mass activity of ^{235}U (185.7 keV) and mass activity of ^{238}U calculated through the daughter such as ^{214}Pb (351.9 keV) and ^{214}Bi (609.3, 1764.5 keV). The relative deviation was less than 4.0% for both methods.

Materials and methods

Theoretical work

In the γ ray spectrum, the total counts of 186 keV peak will include counts of ^{226}Ra (186.2 keV, 3.555%) and ^{235}U (185.7 keV, 57%). We have:

$$N_{\text{total}} = N_{^{226}\text{Ra}} + N_{^{235}\text{U}}. \quad (1)$$

The formula calculates radioactivity:

$$A = \frac{N}{I_\gamma \times \varepsilon \times m \times t}, \quad (2)$$

where A (Bq kg^{-1}) is a mass activity, m (kg) is dry mass, I_γ and ε are intensity γ ray and efficiency detector in energy E (keV), t (s) is an acquisition time, respectively.

We assume the analysis sample does not get rich and not exhaust uranium (in a state radioactive equilibrium). The dry mass sample will include 0.72% mass of ^{235}U and 99.2745% mass of ^{238}U [7]. If the sampling area had not changed geochemical, we considered the activity of ^{226}Ra equal the activity of ^{238}U (the half-life of ^{226}Ra is 1600 years, very small compared with the half-life of ^{238}U), the ratio count rate is:

$$\frac{N_{\text{total}}}{N_{^{226}\text{Ra}}} = \frac{0.72 \times T_{^{238}\text{U}} \times M_{^{238}\text{U}} \times \varepsilon_{^{235}\text{U}}^{185.7} \times I_{^{235}\text{U}}^{185.7}}{99.2745 \times T_{^{235}\text{U}} \times M_{^{235}\text{U}} \times \varepsilon_{^{226}\text{Ra}}^{186.2} \times I_{^{226}\text{Ra}}^{186.2}} + 1. \quad (3)$$

Due to the efficiency of HPGe detector at the 186.2 keV peak equal with the efficiency at the peak 185.7 keV ($\varepsilon_{^{235}\text{U}}^{185.7} = \varepsilon_{^{226}\text{Ra}}^{186.2}$), γ intensity of ^{226}Ra (186.2 keV) and ^{235}U (185.7 keV) are $I_{^{226}\text{Ra}}^{186.2} = (3.555 \pm 0.019)\%$ and $I_{^{235}\text{U}}^{185.7} = (57.0 \pm 0.3)\%$, respectively, the half-life $T_{^{235}\text{U}} = 7.04 \times 10^8$ years, $T_{^{238}\text{U}} = 4.468 \times 10^9$ years [9]. The ratio of the contribution for ^{226}Ra (186.2 keV) and ^{235}U (185.7 keV) show that in the region 186 keV consists of 57.2% ^{226}Ra and 42.8% ^{235}U using Eqs. (4) and (5), respectively.

$$N_{^{226}\text{Ra}} = 0.572 \times N_{\text{total}}, \quad (4)$$

$$N_{^{235}\text{U}} = 0.428 \times N_{\text{total}}. \quad (5)$$

Direct method

When the samples reached a secular equilibrium of the decay chain ^{238}U and a state radioactive equilibrium, we can calculate the mass activity (Bq kg^{-1}) of ^{235}U and mass activity of ^{238}U using Eqs. (6) and (7), respectively:

$$A_{^{235}\text{U}} = \frac{0.428 \times N_{\text{total}}}{\varepsilon_{^{235}\text{U}}^{185.7} \times I_{^{235}\text{U}}^{185.7} \times m \times t}, \quad (6)$$

$$A_{^{238}\text{U}} = \frac{0.572 \times N_{\text{total}}}{\varepsilon_{^{238}\text{U}}^{186.2} \times I_{^{238}\text{U}}^{186.2} \times m \times t}, \quad (7)$$

where N_{total} is counts of measured sample minus background, the relative uncertainties of mass activity were calculated following the propagation of uncertainty [10].

Equilibrium method

The samples were sieved and filled into the cylindrical container and then sealed off the laboratory at least 30 days so that no radon escape to avoid disequilibrium problems between ^{226}Ra and its respective progenies (^{214}Pb and ^{214}Bi). We have:

$$A_{^{226}\text{Ra}} = A_{^{214}\text{Bi}} \Rightarrow N_{^{226}\text{Ra}} = \varepsilon_{^{238}\text{U}}^{186.2} \times I_{^{238}\text{U}}^{186.2} \times t \times A_{^{214}\text{Bi}}. \quad (8)$$

The mass activity of ^{214}Bi (609.3 keV, 45.49%):

$$A_{^{214}\text{Bi}} = \frac{N_{^{214}\text{Bi}}}{\varepsilon_{^{214}\text{Bi}}^{609.3} \times I_{^{214}\text{Bi}}^{609.3} \times m \times t}. \quad (9)$$

From Eqs. (1), (2) and (8), the mass activity ^{235}U (Bq kg^{-1}) calculated to follow Eq. (10) [11], the relative uncertainties of mass activity were calculated to the law of propagation of uncertainty [10]:

$$A_{^{235}\text{U}} = \frac{N_{\text{total}}}{\varepsilon_{^{235}\text{U}}^{185.7} \times I_{^{235}\text{U}}^{185.7} \times m \times t} - \frac{I_{^{238}\text{U}}^{186.2}}{I_{^{235}\text{U}}^{185.7}} \times A_{^{214}\text{Bi}}. \quad (10)$$

The decay chain ^{238}U includes 14 series of radioactive isotopes other and the mean of mass activity can be calculated by Eq. (11), where n for number isotopes, A_i and u_i is mass activity and direct uncertainty of ith isotopes, respectively.

Table 1 The information of the standard and samples

Samples	RGU	M1	M2	M3	M4
Mass (g)	130	140	132	136	132
Density (g cm^{-3})	1.55	1.68	1.57	1.63	1.57

$$\bar{A} = \frac{\sum_{i=1}^{n} \frac{A_i}{u_i^2}}{\sum_{i=1}^{n} \frac{1}{u_i^2}}. \quad (11)$$

The direct uncertainty of mean mass activity:

$$\bar{u} = \sqrt{\frac{1}{\sum_{i=1}^{n} \frac{1}{u_i^2}}}. \quad (12)$$

Experimental data

The experimental measurement was performed with a coaxial p-type HPGe, supplied by Canberra, Inc., USA, crystal diameter 62.2 mm, crystal length 50.1 mm, the relative efficiency 35% and the energy resolution (FWHM) at 1332 keV (^{60}Co) is 2.0 keV, which is using the Lynx® based on advanced digital signal processing techniques, the spectra were recorded using Genie-2K software with 32,748 channels. The detectors are surrounded by a cylindrical low-background passive shielding made of 100 mm thickness of lead, and inside had copper and tin with thickness 1.6 and 1.0 mm, respectively. The samples are closed in a cylindrical container with external diameter 75 mm, filled to height 20 mm, the thickness of wall 2 mm and have a dry mass such as Table 1.

The efficiency calibration is experimentally determined by RGU standard (mass activity 4940 ± 30 Bq kg^{-1}) at encap of the detector, which is used to perform the efficiency calibration curve for HPGe detector in the energy range from 46.5 to 2447.9 keV. The acquisition time is 86,400 s for background and samples. The peaks and the overlapping peaks are processed using Colegram software [12]. The samples sealed off the laboratory for at least 30 days so that no radon (^{222}Rn, the half-life 3.825 days) escape to avoid disequilibrium problems between ^{226}Ra and its respective progenies (^{214}Pb and ^{214}Bi).

Table 2 The full energy peak efficiency for RGU source standard

E (keV)	Experimental efficiency	Uncertainty (%)	Fitted efficiency	Uncertainty (%)	RD (%)
46.5	0.014607	1.24	0.014595	1.40	0.08
63.3	0.046051	2.24	0.046456	2.28	0.87
186.2	0.070195	0.83	0.069027	0.95	1.69
241.9	0.056981	0.71	0.058349	0.77	2.34
295.2	0.049304	0.65	0.049706	0.75	0.81
351.9	0.043238	0.65	0.042292	0.78	2.24
609.3	0.023566	0.75	0.024059	0.79	2.05
768.4	0.019553	0.81	0.019289	0.78	1.37
934.1	0.016400	0.91	0.016509	0.77	0.66
1120.0	0.014848	0.70	0.014736	0.76	0.76
1238.0	0.013737	0.81	0.014010	0.78	1.95
1764.5	0.012135	0.76	0.012011	0.89	1.04
2204.2	0.009907	0.98	0.010155	0.91	2.44
2447.9	0.008977	1.37	0.008828	1.28	1.69

RD is relative deviation $RD = \frac{|\varepsilon_{Exp} - \varepsilon_{fitted}|}{\varepsilon_{Exp}} \times 100\%$

Table 3 Mass activities of ^{238}U (Bq kg^{-1}) for the samples

Samples	Mass activity (Bq kg^{-1})					
	Direct method		Equilibrium method			
	^{234}Th-63.3 keV	^{226}Ra-186.2 keV	^{214}Pb-351.9 keV	^{214}Bi-609.3 keV	^{214}Bi-1764.5 keV	Mean values
M1	107.84 ± 3.34	102.36 ± 1.42	102.10 ± 1.10	99.71 ± 1.19	107.03 ± 2.39	101.62 ± 0.76
M2	37.77 ± 1.63	36.49 ± 0.94	36.32 ± 0.54	35.38 ± 0.61	36.38 ± 1.36	35.94 ± 0.39
M3	283.71 ± 7.47	277.00 ± 3.67	289.78 ± 2.61	277.58 ± 2.69	285.70 ± 4.44	284.15 ± 1.73
M4	139.43 ± 4.14	132.56 ± 2.13	137.71 ± 1.40	132.06 ± 1.49	135.03 ± 2.81	135.05 ± 0.96

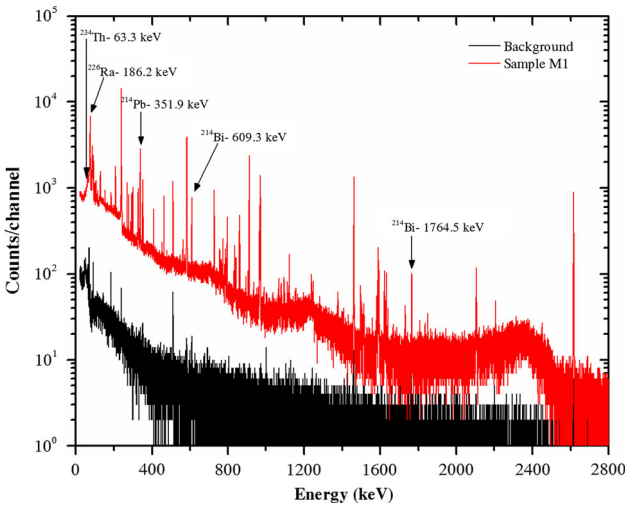

Fig. 1 Sample M1 and background spectra resulting from 86,400 s livetime

Table 4 Mass activities of the radionuclide ^{235}U for the samples

Samples	Mass activity (Bq kg^{-1})		RD (%)
	Direct method	Equilibrium method	
M1	4.93 ± 0.07	4.77 ± 0.07	3.2
M2	1.77 ± 0.05	1.70 ± 0.05	3.8
M3	12.86 ± 0.17	12.91 ± 0.17	0.4
M4	6.20 ± 0.10	6.18 ± 0.10	0.4

The full energy peak efficiency was determined at the 14 values of ^{210}Pb (46.5 keV), ^{234}Th (63.3 keV), ^{226}Ra (186.2 keV), ^{214}Pb and ^{214}Bi (in equilibrium with its parent ^{238}U). We used to correction factor 0.572 to correct count of ^{226}Ra at the region 186 keV. The experimental efficiency curve were fitted by the ACORES software [11]. The relative deviation between the values from experimental efficiency values and efficiency calibration curve was less than 2.5% (see Table 2).

Results and discussion

The mass activity of ^{238}U (Bq kg^{-1}) was calculated based on the activities ^{234}Th (63.3 keV), ^{214}Pb (351.9 keV) and ^{214}Bi (609.3, 1764.5 keV). The maximum relative deviation between the mass activity of ^{238}U at 63.3 keV (^{234}Th) and the mean of mass activity is less than 6.0%. Equation (7) was used for determining mass activity of ^{226}Ra (186.2 keV). The maximum relative bias between the mass activity of ^{238}U calculated by the direct method using the 186.2 keV peak (^{226}Ra) and the mean of mass activity of ^{238}U is less than 3.0% (see Table 3).

That shows the samples M1–M4 achieved secular equilibrium between ^{238}U and its respective progenies (^{214}Pb, ^{214}Bi). The radioactivity of the ^{226}Ra calculated for the samples ranging from 36.49 ± 0.94 (Bq kg^{-1}) to 277.00 ± 3.67 (Bq kg^{-1}). It is mass activities of the sample in this study are higher than the activity concentration of ^{226}Ra for rock samples [13], and soil sample [14] in the Ramanagara and Tumkur districts, Karnataka, India. Figure 1 shows to compare between background and M1 sample.

In the other hand, the direct method [Eq. (6)] and the equilibrium method [Eq. (10)] are used to determine the mass activity of ^{235}U for the samples. Table 4 presents a good agreement between both methods was less than 4.0%.

Conclusions

The revision of nuclear data shows that the 186 keV peak has contributed count of ^{226}Ra and ^{235}U about 57.2 and 42.8%, respectively. A direct analysis method with correction factor 0.572 for ^{226}Ra value and 0.428 for the ^{235}U value can be determined mass activity of ^{226}Ra and ^{235}U in the samples with the existence of equilibrium. In this paper, the radioactivities of the ^{238}U and ^{235}U can be calculated, which is a good agreement with both methods. Moreover, the advantage of the direct method can be applied after preparation sample, which does not need for the secular equilibrium between ^{226}Ra and its progenies (^{214}Pb and ^{214}Bi).

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