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Optimization of Counting Time for Measurement of Ultra-low-level ^{238}U and ^{232}Th by 80% Relative Efficiency HPGe Detector

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Abstract: It has been observed that for quantitative measurement of ultra-low-level ^{238}U and ^{232}Th in natural samples, the relative standard deviation (RSD) between the activity values of different photopeaks of the daughter nuclides in the corresponding series is random and large in low counting time and after a certain counting time, the randomness of RSD is minimized. Depending on RSD and relative counting error (RCE), it was observed that the minimum counting time for measurement of ultra-low-level ^{238}U and ^{232}Th by a HPGe detector of 80% relative efficiency is ~60000 s. Different IAEA standards were used for this experiment.

Keywords: Ultra-low-level natural radioactivity, Optimization of counting time, Relative standard deviation.

1 Introduction

Exposure to natural radiation generally comes from the primordial radionuclides of ^{238}U, ^{232}Th, the decay products in these decay series and ^{40}K along with the contribution from cosmic radiation. The three existing decay series of ^{238}U, ^{235}U, ^{232}Th and omnipresent ^{40}K altogether symbolizes the natural radioactivity. Natural Radioactivity is present in any geological materials but concentration varies depending on the geographical location [1].

To avoid self-attenuation, usually sample size for natural samples like soils, sediments, etc., are kept between 40-60 g when measured by a HPGe detector (except Marinelli beakers). Usually, 1-2 Bq of activity is present in these experimental samples, which is ultimately expressed in Bq kg^{-1}. Hence, extreme care has to be taken for determination of such minuscule amount of activity.

There are numbers of reports available in literature, which dealt with various parameters like efficiency, resolution and background interference of gamma-ray detectors, which ultimately affect the sensitivity of the detectors [2,3,4]. These parameters are also well documented in standard text books [5,6]. However, in case of ultra-low-level natural radioactivity measurement many more interferences complex the spectrum. For example, more than 200 photopeaks of different intensities are present in ^{238}U, ^{232}Th series. It is difficult to choose particular photo-peaks for reliable end result. Efficiency calibration with standard point sources does not work well due to large difference in the geometry between standard and samples. For last few years we are working towards setting up a standard protocol for natural radioactivity measurement in environmental samples. We have addressed important parameters in ultra-low-level ^{238}U, ^{232}Th measurement like selection of appropriate photopeak, proper efficiency calibration, sample measurement time, etc. [7-14]. Finally, the come out of these papers were satisfactorily applied to evaluate first base level natural radioactivity measurement data in world's largest mangrove ecosystem, Indian Sundarbans [15-16].

Literature search reveals that different research groups have chosen different counting times starting from 5000 s to 150000 s for ultra-low-level natural radioactivity measurement without any valid rationale. To address this issue, we have demonstrated by rigorous statistical analysis that apart from relative counting error, (RCE), ($RCE = \frac{Counting\ error}{mean\ value} \times 100$), relative standard deviation, (RSD) ($RSD = \frac{Standard\ deviation\ of\ the\ activity\ values\ under\ different\ photopeaks}{mean\ value} \times 100$) also plays an important role in uncertainty estimation for ultra-low-level ^{238}U - ^{232}Th measurement.

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2 Experimental Section

2.1 Standard and sample preparation

U and Th standards were prepared from RGU-1 (Uranium ore) and RGTh-1 (Thorium ore) reference materials (RMs) procured from International Atomic Energy Agency (IAEA). RGU and RGTh contain recommended amount of ~4940 Bq kg^{-1} ^{238}U and 3250 Bq kg^{-1} ^{232}Th respectively. Hence, to prepare low-activity standards for the present experiment, appropriate amounts of RGU and RGTh were mixed with silica gel in 75 mm petri-plates to attain similar sample and standard geometry. For example, 0.6 g RGU mixed in silica gel corresponds to 2.964 Bq ^{238}U activity. Similarly, 1 g RGTh corresponds to 3.249 Bq ^{232}Th activity. Later, the prepared standards were kept for more than 30 d to reach the secular equilibrium. Irish sea sediment (IAEA-385) was considered as the test experimental sample. 50 g of this fine-powdered reference material (with certified radioisotopic concentrations) was sealed in 75 mm petri-plate and kept aside for 30 d to obtain secular equilibrium. Later, the experimental sample (IAEA-385) was compared with the standard samples (RGU, RGTh); activities were calculated for ^{238}U, ^{232}Th and finally matched with the known certified values.

2.2 Instrumentation

A HPGe detector having 80% relative efficiency and resolution of 1.65 keV at 1.33 MeV energy was procured from CANBERA and has recently been installed at Saha Institute of Nuclear Physics (SINP), Kolkata, India. The detector is well shielded with LS (Laboratory lead shield) series. The detector is connected with Canberra DSA-LX analyzer, having an integrated 16k multichannel analyzer. Energy calibration was done using the single point-source standards of ^{152}Eu, ^{137}Cs, ^{133}Ba and ^{60}Co. The prepared standards were measured over a range of counting times viz., 5000 s, 15000 s, 25000 s, 40000 s, 50000 s, 60000 s, 75000 s, 85000 s and 100000 s. Long background count of 200000 s was taken, which was accordingly stripped from the various sample spectra. ^{226}Ra and ^{232}Th activities in the samples were calculated using the photopeaks of 295.22, 351.93, 609.31 keV for ^{226}Ra and photopeaks of 338.32, 583.19, 911.20 keV for ^{232}Th [7].

3 Results and Discussion

Activities of ^{226}Ra and ^{232}Th in IAEA-385 RM were calculated using comparator method [9]. ^{226}Ra activity in IAEA-385 Irish soil sample was obtained considering standard IAEA-RGU of 2.964 Bq activity, whereas that of ^{232}Th activity was calculated using IAEA-RGTh standard of 3.249 Bq activity. Fig. 1 and 2 shows the graphical representation of RSD and RCE at different counting times.

Fig. 1 and 2 clearly demonstrate that the randomness of RSD values was minimized after 60000 s for both ^{226}Ra and ^{232}Th, and a regular trend of RSD values were also observed. After 60000 s, also RSD values became closer to the RCE values. Therefore, Figure 1 and 2 vouch that minimum counting time in 80% relative efficiency detector with heavy shielding would be 60000 s.

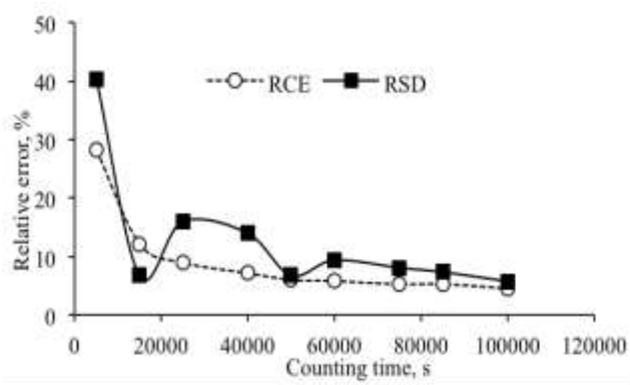

Fig.1: Variation of RCE and RSD with counting times for sample IAEA-385 RM for ^{226}Ra activity.

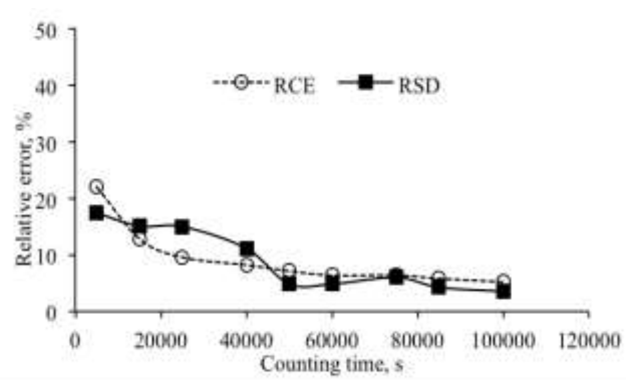

Fig. 2: Variation of RCE and RSD with counting times for sample IAEA-385 RM for ^{232}Th activity.

The observed activities for ^{226}Ra and ^{232}Th in different counting times have been tabulated in Table 1 and 2. It can be concluded from these two tables that ~ 60000 s counting time would be affordable compromise for ultra-low-level natural radioactivity measurement by a 80% relative efficiency detector.

The MDA (Minimum Detectable Activity) of the 80% HPGe detector was calculated using Currie's formula [17-18] and tabulated in the Table 3 for different counting time.

MDA= $\frac{4.653 \times \sigma NB + 2.71}{\in \times P\gamma \times T}$, where σN_B = error of the background count rate, € = efficiency of the counting channel, P_γ = intensity of the photopeak, T = counting time

It is observed that MDA decreased with increase in counting time. However, after 60000 s it became more or less constant, which also supports the statement that minimum counting time in 80% detector should be around 60000 s.

Table 1: Observed ^{226}Ra activities and errors of IAEA-385 Irish soil sample at different counting times.

Counting time, s	Calculated activity+ of ^{226}Ra, Bq	Observed activity of ^{226}Ra, Bq	RCE %	RSD %	RB%*
5000	1.095	0.60	28.2	40.2	-45.2
15000		0.92	12.2	6.8	-15.9
25000		0.92	9.0	16.0	-15.9
40000		0.86	7.2	14.0	-21.5
50000		0.90	6.0	6.8	-17.8
60000		0.96	5.8	9.4	-12.3
75000		0.91	5.3	8.1	-16.9
85000		0.81	5.3	7.4	-26.0
100000		0.83	4.6	5.8	-24.2

+ calculated from certified value

Table 2: Observed ^{232}Th activities and errors of IAEA-385 Irish soil sample at different counting times.

Counting time, s	Calculated activity+ of ^{232}Th, Bq	Observed activity of ^{232}Th, Bq	RCE %	RSD %	RB%
5000	1.685	1.52	22.1	17.4	-9.8
15000		1.54	12.8	15.1	-8.6
25000		1.78	9.6	15.0	+5.6
40000		1.48	8.2	11.1	-12.2
50000		1.51	7.1	4.8	-10.4
60000		1.61	6.4	4.8	-4.5
75000		1.58	6.4	6.0	-6.2
85000		1.53	5.8	4.2	-9.2
100000		1.55	5.2	3.5	-8.0

+ calculated from certified value

*Relative Bias, RB = $\frac{Value(measured) - Value(certified)}{Value\ (certified)} \times 100$

Table 3: MDA values in Bq for ^{226}Ra and ^{232}Th at different counting times

Counting time, s	MDA ^{226}Ra, Bq	MDA ^{232}Th, Bq
5000	0.72	0.36
15000	0.41	0.16
25000	0.36	0.12
40000	0.21	0.14
50000	0.21	0.14
60000	0.15	0.10
75000	0.13	0.09
85000	0.13	0.10
100000	0.13	0.09

4 Conclusions

Ultra-low-level NORM measurement requires much attention at every step for obtaining consistent result. In majority, environmental samples contain minuscule amount of ^{226}Ra (^{238}U) and ^{232}Th activity. This paper confirms that the corollary made in our earlier work [8] that the minimum counting time would be the time required when RCE and RSD values come closer is general in nature and not dependent on the system. However, the particular value of minimum counting time needs to be determined for every system. It has been found that for HPGe detector with relative efficiency of 80% the minimum counting time for measurement of ^{226}Ra (^{238}U) and ^{232}Th is 60000 s

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