Measurements of natural radioactivity in soil using an array of cerium doped lanthanum bromide scintillator detectors

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Abstract. An array of eight 2" $\times$ 2" $LaBr_3$:Ce scintillator detectors linked to a XIA PIXIE-16 Digital Signal Processing data-acquisition system was used to measure KCl, U ore, Th ore, and soil reference samples placed in the centre (17.5 cm from each detector) of the array (with all detectors lying in the horizontal plane) for 12 hours per sample. The absolute full-energy peak gamma-ray detection efficiencies of each detector was determined. The weighted average absolute full-energy peak detection efficiencies were found to be $3.21 \times 10^{-2} \pm 8.96 \times 10^{-4}$ %, $2.57 \times 10^{-2} \pm 8.79 \times 10^{-4}$ %, $1.87 \times 10^{-2} \pm 6.34 \times 10^{-4}$ % at 1461 keV (^{40}K), 1764 keV (^{238}U) and 2615 keV (^{232}Th) respectively. For the soil reference sample, the weighted average activity concentrations for ^{40}K and ^{232}Th series radionuclides were consistent with zero to within measurement uncertainties. For the same sample the activity concentration of ^{238}U series radionuclides was consistent with zero when an interval of $\pm 2\sigma$ about the mean value is considered.

1. Introduction

The efficiency of $LaBr_3$:Ce detectors is 1.2 - 1.7 times better at detecting gamma-rays than NaI:Tl detectors above 350 keV and have an energy resolution of 2.5 - 3.0 % at the 662 keV gamma-line of Cs-137, compared to 6 - 7 % for NaI:Tl detectors for 3.8 cm by 3.8 cm (1.5" $\times$ 1.5") detectors [1, 2]. The detector crystal has other advantages such as a high scintillation light output with a fast decay time [3] which make it to be an excellent detector choice for high count rate scenarios, and is capable of performing well with count rates up to 500 kHz[5]. The light output of $LaBr_3$:Ce crystals displays improved temperature stability compared to traditional NaI:Tl scintillators, varying by less than 1% between 0°C and +55°C [1, 6].

There are challenges associated with using $LaBr_3$:Ce detectors. First, $LaBr_3$:Ce is itself radioactive, due to the presence of radioactive ^{138}La [1, 2]. La-138 accounts for 0.09% of naturally occurring lanthanum and produces two gamma-rays: a 788.7 keV from beta decay to ^{138}Ce, and a 1435.8 keV from electron capture to ^{138}Ba, and X-rays: 32.2 keV ($K_{\alpha 1}$), 31.8 keV ($K_{\alpha 2}$), 36.4-37.3 keV (K_{β}), 3.9-5.9 keV (L) from β^+ decay and 95.7 keV from β^- decay. The activity concentration of ^{138}La in the scintillator material determined from the sum peak at 1468 keV is 12.9 Bq/kg [1]. As of now, it is still unclear whether this internal radioactivity acts

as a fundamental limitation for low-level activity measurements of natural occuring radioactive materials (NORM).

2. Materials and methods

An array of eight 2" × 2" in. $LaBr_3$:Ce scintillator detectors linked to a XIA PIXIE-16 Digital Signal Processing data-acquisition system was used as shown in Fig. 1. ^{137}Cs and ^{22}Na were used for energy calibration. The KCl, U and Th ore, soil reference and background samples were counted for 12 hours each. Each of these samples was placed in 1.0 L Marinelli beaker and placed 17.5 cm from each detector for measurements. The background measurement was done using an empty Marinelli beaker. Further information about the reference sample is given in Table 1. The absolute full-energy peak (FEP) detection efficiencies of the detectors were evaluated using Eqn 1 and the activity concentration determined using Eqn. 2 [7]:

$$\varepsilon = \frac{N_c}{A \times m \times t \times P_\gamma} \tag{1}$$

$$A = \frac{N_c}{\varepsilon \times m \times t \times P_\gamma} \tag{2}$$

where N_c is the full-energy peak count, t is the counting time in seconds, A is the activity concentration in Bq/kg, m is mass of the sample in kg and P_γ is the probability of emission of the particular gamma-ray being measured.

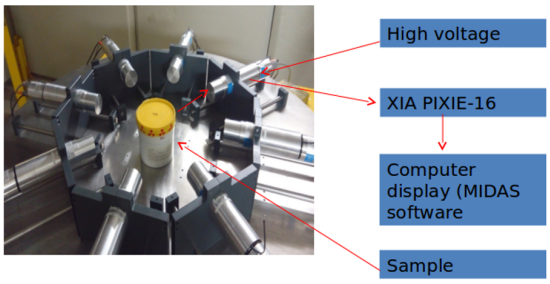

Figure 1. Experimental setup showing an array of 2" × 2" $LaBr_3$:Ce detectors and reference sample placed in the centre.

Table 1. Reference sample information

Sample	Mass (kg)	Activity (Bq/kg)	Supplier	IAEA code
`Potassium Chloride`	1.29080	16259 ± 172	IAEA	RGK-1
`Uranium Ore`	1.40912	4940 ± 99	IAEA	RGTh-1
`Thorium Ore`	1.36494	3248 ± 17	IAEA	RGU-1
`Soil`	1.50263	Vary for each radionuclide (see Table 2)	IAEA	IAEA-375

3. Results and discussion

The absolute full-energy peak (FEP) detection efficiency determined at 1461 keV (^{40}K), 1764 keV (^{238}U) and 2615 keV (^{232}Th) are shown in Fig. 2. This efficiencies were measured using the potassium chloride; uranium ore and thorium ore samples respectively (see Table1). The variation in the efficiency values between detectors could be due to slight shifts in the sample

position toward a certain detector and/or scattering. The weighted average absolute full-energy peak (FEP) detection efficiency as a function of gamma-ray energy is shown in Fig. 3. The experimental points exhibit a power-law behaviour which was fitted by the following equation:

$$\varepsilon = a\left(\frac{E}{E_0}\right)^b$$

where ε is the full-energy peak efficiency; E is gamma-ray energy in keV; E_0 is an arbitrary energy to make the argument dimensionless [here, $E_0 = 1$ keV]; and a, b are the coefficients extracted by a least-squares fit to the experimental data.

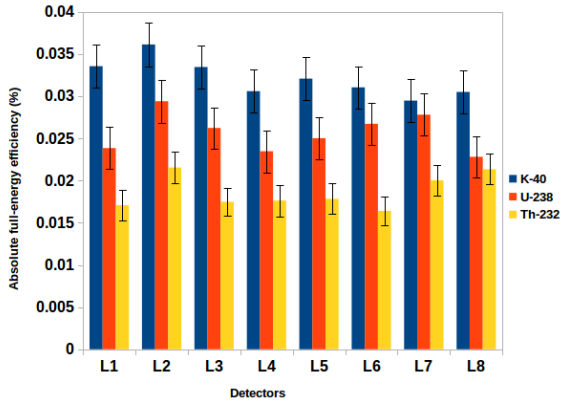

Figure 2. The absolute full-energy peak (FEP) detection efficiency for the eight detectors.

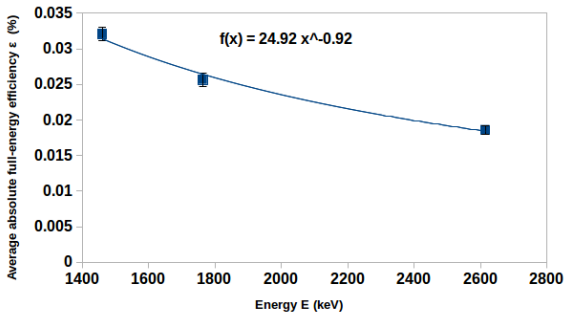

Figure 3. Average absolute full-energy peak (FEP) detection efficiency as a function of energy.

Table 2 gives the activity concentration of ^{40}K, ^{238}U and ^{232}Th series radionuclides measured in the soil reference sample using the detection efficiencies discussed above. For the soil reference sample, the weighted average activity concentrations for ^{40}K and ^{232}Th series radionuclides were consistent with zero to within measurement uncertainties. For the same sample the activity concentration of ^{238}U series radionuclides was consistent with zero when an interval of $\pm 2\sigma$ about the mean value is considered. For the measurement setup and conditions described here the minimum detectable activty (MDA) of ^{40}K; ^{238}U and ^{232}Th series radionuclides are 2391 Bq/kg; 524 Bq/kg and 369 Bq/kg, respectively. This explains the activity concentration results for the soil reference sample found in this study.

4. Conclusion

This paper presented the absolute (FEP) detection efficiencies of $LaBr_3$:Ce scintillators placed at a distance 17.5 cm away for volume samples (Marinelli geometry); for 1461 keV (^{40}K), 1764 keV (^{238}U) and 2615 keV (^{232}Th). These efficiencies were used to calculate the activity concentration of ^{40}K, ^{238}U and ^{232}Th series radionuclides in the soil reference sample. The soil activity concentration were found to be below the MDA for the setup used in this study.
In order to address this problem we are now investigating ways to reduce the influence of room background on our measurements by making use of passive shielding.

Table 2. Activity concentration of ^{40}K, ^{238}U, ^{232}Th in soil reference sample

Detector	K-40 (Bq/kg)	U-238 (Bq/kg)	Th-232 (Bq/kg)
L1	1079 ± 1681	257 ± 703	623 ± 474
L2	1248 ± 1594	570 ± 583	-34 ± 400
L3	-195 ± 1670	-230 ± 625	-4 ± 443
L4	-2458 ± 1866	134 ± 707	-168 ± 485
L5	-912 ± 1753	200 ± 658	141 ± 456
L6	-3366 ± 1851	912 ± 632	-1299 ± -536
L7	1035 ± 1892	244 ± 597	171 ± 407
L8	-2546 ± 1854	528 ± 715	234 ± 386
Average	-633 ± 623	332 ± 289	14 ± 156
Expected range	$417.0 - 432.0$*	$19.0 - 29.8$*	$19.2 - 21.9$*
Expected mean	424.0*	24.4*	20.5*

* IAEA/AL/075 report on the intercomparison run (IAEA-375)

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