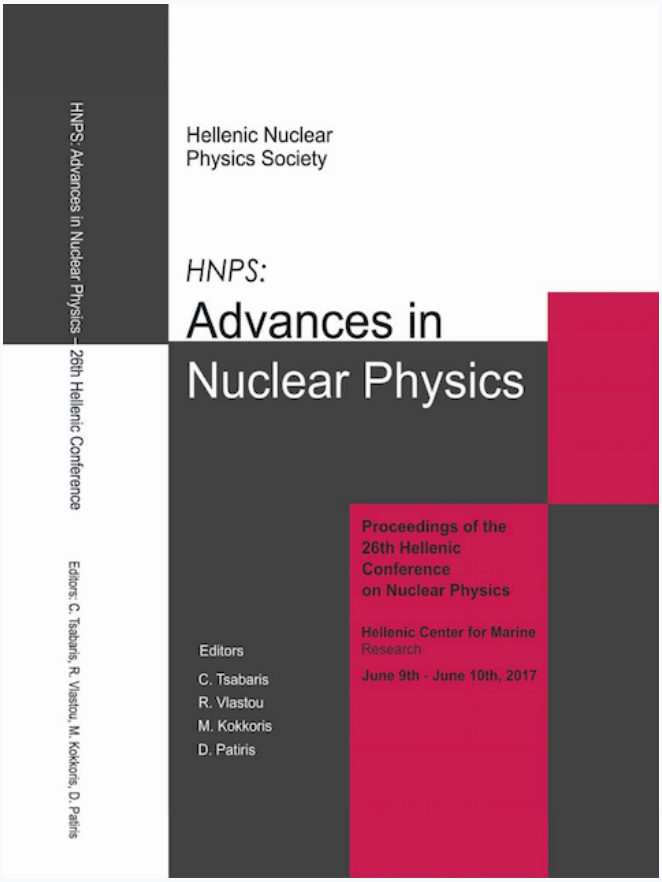
HNPS Advances in Nuclear Physics

Vol 25 (2017)

HNPS2017



Radioactivity measurements in granite samples using NaI(Tl) spectrometry

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doi: 10.12681/hnps.1962

Radioactivity measurements in granite samples using NaI(Tl) spectrometry

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Abstract In the present work, the possibility of exploiting more peaks in the NaI (Tl) spectra and overcome the peak convolution due to detector's low resolution has been investigated. The use of more than the typically utilized peaks in environmental radioactivity measurements may enhance the accuracy of the determination of the concentration of 238U and 232Th series and expand the usability of low resolution detectors (NaI) in environmental studies. Preliminary results from granite samples spectra, measured with a (3″x 3″) NaI (Tl) detector, are presented and compared with those obtained by means of high resolution gamma-ray spectrometry.

Keywords Gamma ray Spectroscopy, NaI(Tl), radioactivity, sample analysis, granite

INTRODUCTION

NaI (Tl) detectors are virtually the first solid-state detectors used for gamma-ray spectrometry. The main disadvantages of NaI detectors are the low resolution, the gain drift due to changes in the ambient temperature and the nonlinearity issue. A solution to the gain drift is to use a stabilizer, or collect spectra of short duration and sum them, using appropriate software. To minimize the nonlinearity problem, the applied voltage is set to the maximum recommended value.

In this work a methodology is examined to overcome the issue of low resolution. The methodology combines software technology [1] with nuclear data and simple equations in an attempt to de-convolute multiple peaks. Such a methodology may extend the usability of low and medium resolution detectors to a wide range of energies that appear in environmental radioactivity data.

The gamma-ray peaks that are typically utilized for quantitative analysis in environmental radioactivity measurements using NaI (Tl) detectors are 1460.8 keV of ^{40}K, 2614.5 keV of ^{208}Tl (^{232}Th decay series) and 1764.5 keV of ^{214}Bi (^{238}U decay series), respectively. On the other hand, using a high resolution HPGe detector more than 20 peaks potentially can be used.

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In this work the exploitation of the 609, 934,1120,1238,1280,1729,1764 and 1847 keV gamma rays of ^{214}Bi and 241.9 keV of ^{214}Pb for the ^{238}U activity determination as well as the 238.6 keV of ^{212}Pb and 911,964 ,969 keV of ^{228}Ac for the ^{232}Th activity determination are examined. Preliminary results from granite samples, measured with a (3″x 3″) NaI (Tl) detector and analyzed with the present methodology, are presented and compared with those obtained by a high-resolution gamma-ray detector.

MATERIAL AND METHODS

Experimental

Gamma spectrometry experimental set up

The gamma spectrometry system of PUAS, based on a 3"x3" NaI detector with 7% energy resolution at 662 keV, was used to record spectra in the γ-energy range 50 - 3000 keV. It was connected to a DigiBASE (Ortec) system. The MAESTRO multi-channel analyzer (MCA) was used for data acquisition, storage and display of the acquired gamma spectra.

Shielding: The detector was shielded with 5 cm lead to reduce the laboratory γ-background.

Spectral Stabilization: To stabilize the NaI spectral drift, the DigiBASE-E gain stabilizer was used, monitoring the centroid of 1461 keV peak in the present case. Furthermore, spectra were registered every 2 hours instead of a 24-hour total spectrum. Then, these partial spectra were recalibrated and added offline using SPECTRW software [1].

Samples

Samples of different commercial granites with surface 49 cm^2, 2 cm thickness and average weight 0.25 kg, without any scratches, were used for the measurements after appropriate cleaning.

Sample counting: Each sample was placed in contact with the detector in the shielded area and counted for 24 hours. Furthermore, a 24 h spectrum of the background radiation present at the laboratory was taken every two days under the same conditions with the granite samples.

Efficiency calibration curve: The detector efficiency curve (see [1]) was obtained using a) a multiple gamma ray emitting large volume source (^{241}Am, ^{137}Cs, ^{60}Co) and b) a ^{40}K source of similar dimensions to the granite samples. A $^{152\text{-}154}$Eu source was used for the low part of the spectrum. The sources were placed on top of the detector in the same geometry as the samples. All the appropriate corrections were taken into account (geometry, summing up corrections, self absorption [2]).

The spectrometric system of HCMR, based on an HPGe detector (64*85 mm) with relative efficiency 50% and 1.8 keV resolution at 1332 keV, was used for the test cases.

Methodology of analysis

The analysis of the net spectra (Original - BGR) was carried out with the SPECTRW [1]. A spectrum recorded with a high-resolution HPGe system was used as a reference for the test cases.

The methodology comprises of the following steps: In each energy region, photo-peaks from NaI for potential use are selected. The expected γ-lines in each "wider photo-peak" of the NaI spectrum and the isotopes to which they belong are noted. The efficiency coefficients, branching ratios and true summing correction factors for each line [2] are necessary in order to split the total area of the wide photo-peaks.

The starting point of the analysis is the most reliable photo peak in the NaI spectrum at 2614 keV that corresponds to ^{208}Tl, being free of any neighboring peaks. The total number of counts N_0 are modified using the detector efficiency, the decay branching ratio and the summing up correction factor through the relation:

$$N_{E_{2614}} = \frac{N_0 \, S_f}{\varepsilon_{E_{2614}} \, B_r} \qquad [1]$$

where S_f is the summing up correction factor, ε is the efficiency at the energy E=2614 keV and b_r is the decay branching ratio. With this modification, N_E represents the number of counts that would have been detected for 100% efficiency, 100% decay branching ratio and with no summing up effects. The same isotope has another peak at 583 keV, which is convoluted together with the 609 keV of the ^{214}Bi . The fitting procedure serves to evaluate the total number of counts.

The expected number of counts for the 583 keV γ-line can be determined from the modified number of counts of the 2614 keV. Using the relation

$$N_{E_{583}} = \frac{N_{E_{2614}} \, \varepsilon_{E_{583}} \, b_{r583}}{S_{f583}} \qquad [2]$$

the expected yield for the Eγ=583 keV peak is estimated. The rest of the total convoluted area is attributed to the 609 keV γ-ray of ^{214}Bi. The information thus obtained for the 609 keV of ^{214}Bi and the 2614 keV of ^{208}Tl can serve to have a first estimate of the ^{238}U/^{232}Th activity ratio, assuming radioactive equilibrium for the two decay chains. In general, the activity of an isotope is determined from each individual γ-ray from:

$$C_R = \frac{S_f \, N_{E_\gamma}}{mt\varepsilon_{E_\gamma} \, b_r} \qquad [3]$$

where C_R is the specific activity, m is the mass of the sample, t is the real collection time and the rest of the notation is as in equation [2].

A very small peak at 563.0 keV, not belonging to any of the observed isotopes, could not identify, either as sum-up spurious peak or as a single/double escape of any of the observed γ-lines. In any case, it is a very small contribution and can be ignored.

The next step is to divide the spectrum in 5 energy regions and analyze the composed photo-peaks contained, as described below. These regions are: a) 40 - 470 keV, b) 850 – 1000 keV, c) 1120-1380 keV, d) 1460 keV and e) 1729-1847 keV.

Region: 40 - 470 keV: The expected γ-Lines in the 239 keV composite peak (figure 1), the isotopes and Branching Ratios (%), are: [238.63, ^{212}Pb, 43.5], [241.0,^{224}Ra,4.05] and [241.9,^{214}Pb, 7.46].

The efficiency coefficients and summing-up factors are almost the same for these energies. The estimated mean ratio of ^{238}U vs ^{232}Th activities, based on the rest of lines is ~ 0.5. So the branching ratio of 241.91 line is modified (^{212}Pb and ^{224}Ra belong to the same chain - ^{232}Th - and assumed in radioactive equilibrium). Thus the total number of counts in 239 peak (before sum-up correction), distributed as follows: 238.6 keV ►85%, 241.0 keV ►8% and 241.9 keV ►7% .

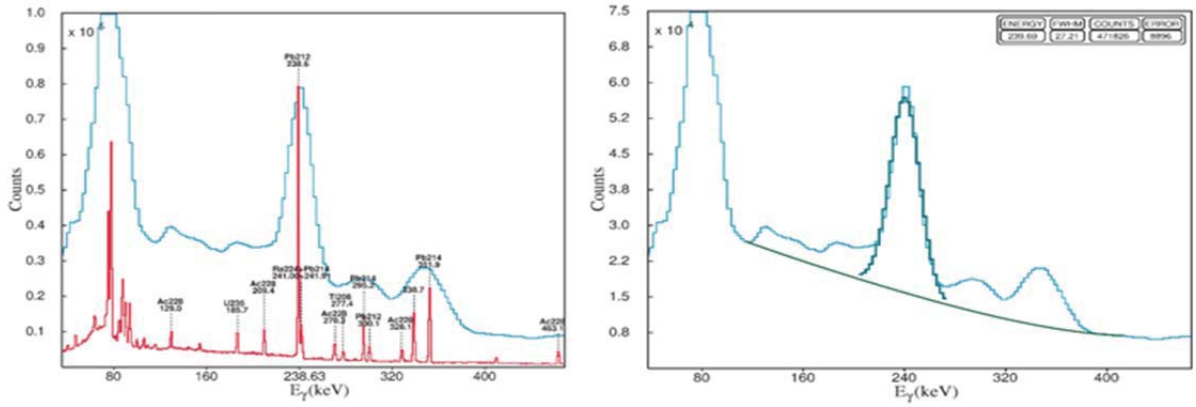

Fig. 1. The HPGe and NaI spectra in the region of 40-470 keV and the photo-peak evaluation (fitting)

Region: 850 – 1000 keV: The expected γ-lines in the 911-968 keV peak (figure 2) are 911.16, 964.64 and 968.97 keV of ^{228}Ac and a small contribution of ^{214}Bi (934.06 keV). From peak-analysis of other γ-lines, the mean activity of the ^{238}U chain (assuming equilibrium) is approximately half of that of the ^{232}Th decay chain. Therefore, the branching ratio of the 934.06 keV belonging to ^{214}Bi can assume half of its original value, normalized in this way to the ^{232}Th chain. Thus, the total number of counts in the composite peak can be distributed: 911.16 keV►55.5%, 934.06 keV►2.6%, 964.64 keV►10%, and 968.97 keV►31.9%.

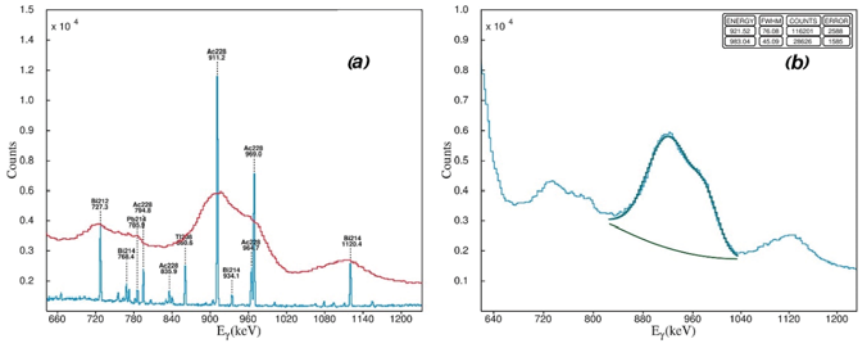

Fig. 2. a) The HPGe and NaI spectra in the region of 850-1000 keV, b) Deconvolution of the composite peak, evaluating the total number of counts.

Region: 1120-1380 keV: There are two major problems in this region. The first is the Compton-edge of the 1460.8 keV (K40) on which three composite photo-peaks are superimposed. The second is the existence of 3 small photo-peaks at 1065, 1079 and 1111 keV that cannot identify. The first problem will resolve by recording a ^{40}K spectrum and subtracting it after normalization in order to have the same number of counts. This will eliminate the "shoulder" of the Compton edge. The second problem will resolve with the help of an HPGe spectrum and/or the deconvolution of the composite peak in the NaI spectrum. Both gave an estimate of 64 ± 5 %.for the ratio of the 1120 keV photo-peak over the total area that includes all 6 peaks, (1065, 1078, 1084, 1111, 1120 and 1155 keV), Thus, the first composite peak can split into the 1120 keV area and all the rest. The deconvolution of the other two composite peaks is more straightforward.

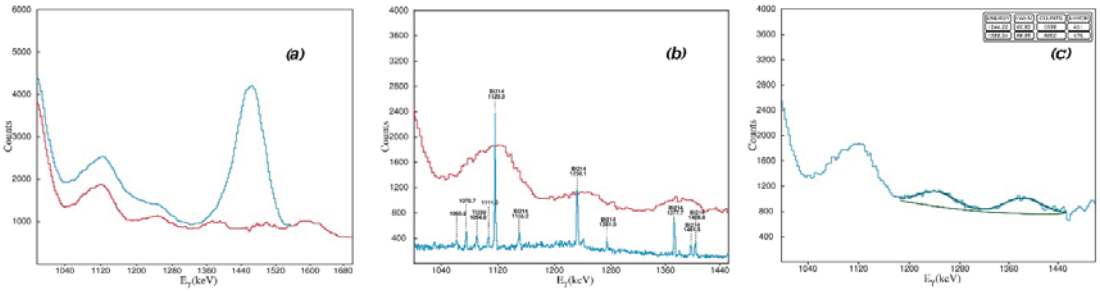

Fig. 3. a) Partial NaI spectrum before and after subtracting ^{40}K, b) The HPGe and NaI spectra in the region of 1120-1380 keV, c) Evaluation of the number of net counts in the 1238+1281 and 1378 peaks.

Apart from a very small un-identifiable peak at 1245 keV, the 1238+1281 photo-peak in the NaI spectrum is almost free of any "contaminants". It is only of the order of a few percent as compared to the 1238+1281 peak and in any case smaller than the experimental error in determining the area under the 1238+1281composite line. As for the 1378 composite peak, the extra three small contributions belong to the same isotope as the 1378 keV line and the total area can split using the branching ratios, the efficiency coefficients and the summing-up factors as described before.

Region: 1460 keV: The 1460.82 keV line is almost free of any "contaminating" peaks in the NaI spectrum (figure 4).

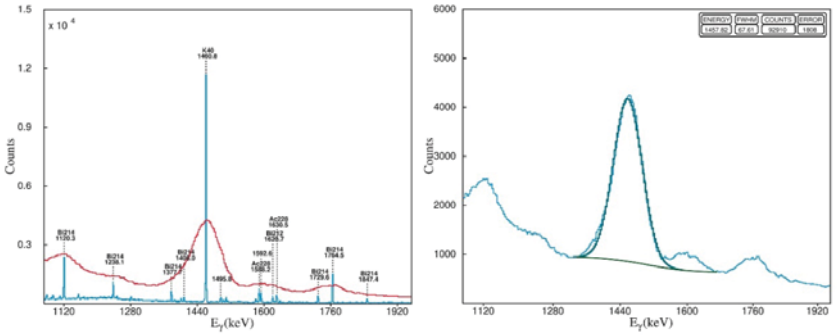

Fig. 4. The HPGe and NaI spectra in the region of 1460 keV and the evaluation of the 1460 photo-peak

Region: 1729-1847 keV: The 1729.6, 1764.5 and 1847.4 lines of ^{214}Bi appear as a composite peak in the NaI spectrum (figure 5).

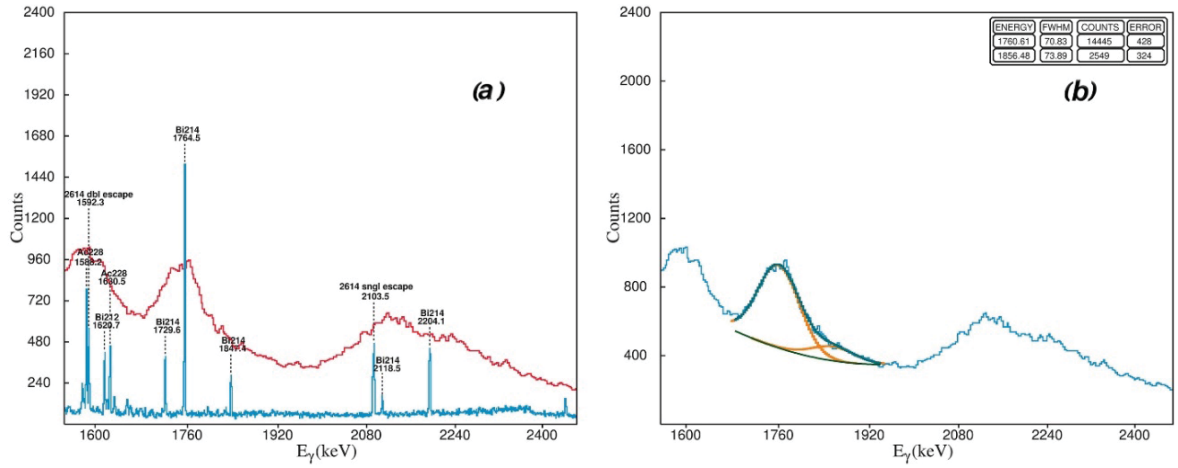

Fig. 5. a) The HPGe and NaI spectra containing the region of 1729-1847 keV, b) Deconvolution of the composite peak

After de-convolution to separate the first two from the third, the 1729.6+1764.5 composite can split into its components using as before the branching ratios, the efficiency coefficients and the summing-up factors.

RESULTS AND DISCUSSION

Results from the analysis of ten granite samples extracted using the described methodology, are presented in table 1. The weighted average of the specific activities (Bq/kg) of ^{232}Th and ^{238}U (assuming equilibrium in both chains) and the specific activity of ^{40}K accompanied with their relative uncertainty are shown.

The specific activities of all radioisotopes cover a wide range, 160-1580 Bq/kg for ^{40}K, 8-127 Bq/kg for ^{238}U and 9-250 Bq/kg for ^{232} Th. The combined uncertainties are of the order of 10 %.

The results for two sample cases were compared with those obtained by means of high resolution γ-ray spectrometry. In the HPGe activity measurements, the γ-emissions from 20 photo-peaks of ^{214}Bi and ^{214}Pb contributed to the weighted average for the ^{238}U series. For the ^{232}Th activity measurements, the photon emission of 20 photopeaks from ^{228}Ac, ^{212}Pb, ^{212}Bi and ^{208}Tl were used. The single photon emission at 1460 keV was used as a direct measure of the ^{40}K activity.

In table 2 results of the activities measured using NaI(Tl) and HPGe detectors are shown. The inter-comparison between the two spectrometry systems for two test cases, on the average, exhibited differences of the activity concentration: 2% for ^{40}K, 8% for the ^{232}Th and ^{238}U decay series while the overall statistical and systematic errors is of the order of 10%.

Sample	^{40}K Specific Activity	Relative uncertainty %	^{238}U Weighted average	Relative uncertainty %	^{232}Th Weighted average	Relative uncertainty %
Test Sample1	1137	10	111	4	256	5
Test Sample2	1277	10	10	9	89	5
Sample 3	1246	10	46	10	77	6
Sample 4	1038	10	127	6	9	11
Sample 5	1581	10	26	11	27	5
Sample 6	1431	10	113	6	252	4
Sample 7	734	10	23	9	61	5
Sample 8	1462	10	42	11	140	5
Sample 9	164	10	9	20	15	7
Sample 10	342	10	8	11	25	5

Table 1. Specific activities in ten samples (Bk/kg) obtained with a NaI spectrometer

	Specific activity (Bq/kg)					
	^{238}U		^{232}Th		^{40}K	
Sample	NaI(Tl)	HPGe	NaI(Tl)	HPGe	NaI(Tl)	HPGe
TS1	111 ± 5	128 ± 6	256 ± 12	269 ± 11	1137 ± 112	1120 ± 186
TS2	10 ± 1	10 ± 1	89 ± 5	87 ± 5	1317± 220	1277 ±125

Table 2. Specific activities measured using NaI(Tl) and HPGe detectors respectively

CONCLUSIONS

A new methodology for the analysis of environmental radioactivity spectra measured with a (3″x 3″) NaI (Tl) detector was investigated. In this methodology an extended part of the NaI spectrum was exploited instead of the usually utilized three energy peaks. Preliminary results of the activity concentrations of ^{40}K, ^{232}Th and ^{238}U in granite samples, measured with a (3″x 3″) NaI (Tl) detector and analyzed with the new methodology, were presented.

The comparison between the results extracted from the NaI and HPGe spectrometry systems for two test cases is very satisfactory. On the average, the exhibited differences of the activity concentration are: 2% for ^{40}K, 8% for the ^{232}Th and ^{238}U decay series, while the overall statistical and systematic uncertainties are of the order of 10%.

References

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