Detection efficiency of NaI(Tl) detector based on the fabricated calibration of HPGe detector

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ABSTRACT

Determination of the full-energy peak efficiency of a detection system is significant in the activity calculation of the measured radioactive samples. In this work, we present an experimental method to determine the absolute efficiency calibration of a NaI(Tl) detector, considering that a standard source of interest is unavailable and by using the known specific activity of a standard sample measured using a HPGe detector. The efficiency of a gamma-ray spectrometer that consists of a coaxial HPGe detector is calculated using Canberra ISOCS/LabSOCS software and a fabricated calibration source. To validate our method, environmental samples (rock and soil samples) were analyzed on both the detectors. The obtained activity concentrations were checked by means of the IAEA proficiency test procedure. The performance criteria evaluation results were found to be ‘Acceptable’ for all the analytical determinations of all the radionuclides under study, except for the samples with low activity (<15 Bq kg\(^{-1}\) and <10 Bq kg\(^{-1}\) for \(^{226}\)Ra and \(^{232}\)Th, respectively). The precision of the low-activity samples was not considered ‘Acceptable’ as the results were slightly inaccurate; in this case, the results were considered as ‘Warning’ because the relative bias (RB) was less than the maximum acceptable bias (MAB).

1. Introduction

Gamma-ray spectroscopy is widely used in many laboratories across the world to determine and quantify natural and artificial radionuclides for radioactive measurements such as environmental sample analysis and activation experiments (Ortiz, 2015). Its major advantages include non-destructive testing, multi-elemental sample analysis, and non-chemical sample processing (Gilmore, 2008). This technique can be performed using different types of detectors, especially high-purity germanium (HPGe) detectors and thallium-activated sodium iodide NaI(Tl) detectors.

HPGe detectors require long counting times and sophisticated cooling systems and provide precise quantitative results in radioactivity measurements because of their high resolution. On the other hand, NaI(Tl) detectors work at room temperature and use large-area detectors that leads to a significant reduction in the measurement time; as a result, these detectors have a higher detection efficiency than HPGe detectors, making it a preferred option for radioactivity measurements (Cinelli, Tositti, Mostacci, & Bar, 2016).

In order to determine the activity for each radionuclide, it is important to know the absolute detection efficiency of the detector at each photon energy for a given efficiency calibration using known standard sources that have the exact geometrical dimensions, density and chemical composition of the sample under study. Most standard radioactive samples are not that easily available to every laboratory. In general, the efficiency calibration of NaI(Tl) detectors is usually performed using standard source. Such calibration methods are neither cost-effective nor easily accessible. Moreover, they are complicated to build with the required homogeneity and may prove to be complicated in terms of maintenance especially with regard to radiation protection (Chiozzi, De Felice, Fazio, Pasquale, & Verdoya, 2000). Furthermore, the geometrical dimensions (size, density, shape and elemental composition) of the sample of interest (SOI) and standard source (SSO) should be identical. In many cases, not all SSOs are identical to the SOIs under investigation.

The accuracy of the efficiency calibration of a detector determines the accuracy of radioactivity measurements. If the detector has a wide efficiency calibration energy range, the activity concentration of a large number of radionuclides can be determined. Therefore, efficiency measurement should be made with precision.

Some techniques for evaluating the efficiency calibration of a detector include the implementation of Monte Carlo simulation (MCS) (Ewa, Bodizs, Czifrus, & Molnar, 2001; Rodenas, Martinavarro, & Rius, 2000; Vidmar & Vodenik, 2010) or a combination of both MCS and genetic algorithms (Quang, Do, & Vo, 2012). In MCS, it is important to have detailed information about the configuration, dimensions, and constituent materials of the detector, which are usually different from the manufacturer’s specifications (Andreotti et al., 2014), especially in the case of...
old detectors that have dead layer formation, as noted in HPGe detectors (Chham et al., 2015; Rodenas et al., 2003).

In this study, we propose a technique to overcome the limitation of finding a standard source and inaccuracy of MCS in some cases to determine the absolute efficiency calibration of NaI(Tl) detector at Radiation Physics Laboratory (RPL), Physics Department, Faculty of Science, Assiut University, Egypt. An environmental sample that has similar geometrical dimensions and composition as that of the SOI is used as standard sample (SSA) to evaluate the absolute detection efficiency of the NaI(Tl) detector.

2. Materials and methods

2.1. The standard sample (SSA)

In this study, the physical dimensions, chemical composition, and material density of the highly radioactive SSAs are similar to the SOI under analysis and this homogeneity minimizes the deviation in the measured activity. In order to achieve greater homogeneity, the SSA and all the selected environmental samples were dried for 3 h at ~105°C and stored in cylindrical polyethylene containers (75 mm in diameter and 90 mm in height) and sieved through an 18-mesh screen. The SSAs were weighed and stored for a minimum period of 30 days to allow daughter products achieve radioactive equilibrium with their parent nucleotides $^{226}$Ra and $^{232}$Th.

2.2. NaI(Tl) scintillation detector

A gamma-ray spectrometer consisting of a 3”×3” NaI(Tl) scintillation detector coupled with a multichannel analyzer (Easy-MCA-8K, ORTEC) was used for the spectral measurements of naturally occurring radionuclides. Following were the specifications of the NaI(Tl) system: the resolution (FWHM) at 1.33 MeV $^{60}$Co was 60 keV, and the relative efficiency at 1.33 MeV $^{60}$Co was 7.5%. The detector was placed in the center of a two-layered chamber fabricated from stainless steel (10-mm thickness) and lead (30-mm thickness). This chamber shielded the detector from unwanted background radiation and minimized the contribution of scattered radiation from the shield. Subsequently, the sample was placed on the detector for a minimum of 10 h. The spectra were analyzed using the MAESTRO software program (Ver. 7.01, ORTEC, Inc.).

2.3. High-purity germanium detector (HPGe)

A gamma-ray spectrometer consisting of p-type, closed-end coaxial HPGe detector (GR4020 model, Canberra) coupled with a 16 K multichannel analyzer (Canberra Industries, Inc., USA) was used for spectral measurements. The detector had a vertical configuration and was cooled with liquid nitrogen. Following were the specifications of the HPGe detector: the resolutions (FWHM) at ≤2.00 keV and ≤0.925 keV were 1332 keV and 122.0 keV, respectively; the relative efficiency was 40%. To reduce the background radiation to <1%, the detector is housed inside a four-layered lead shield (Model 747E, Canberra) (Outer Jacket (3/8 in.), thick low carbon steel Bulk Shield (4 in.), and thick low background lead with Graded Lining (0.040 in.) tin and (0.062 in.) copper) (El-Gamal, Sefelnasr, & Salaheldin, 2019).

The system was calibrated for energy and efficiency. The energy calibration was carried out by acquiring spectra from SSOs of known energies such as $^{137}$Cs (662 keV) and $^{60}$Co (1332 and 1172 keV). The optimization of the geometric dimensions of the containers and improvement of the detection efficiency of HPGe detectors for radioactivity measurements was carried out for efficiency calibration. This was achieved using the Geometry Composer tool of ISOCS/Lab-ISOCS based on the fabricated calibration source using Monte-Carlo N-Particle (MCNP) simulation software instead of the SSO for each sample individually. The tool defines all geometry-related parameters, including sample dimensions, detector properties, and distances or shielding between the sample and the detector and even the densities. Samples were counted, and the counting time depended on the concentration of radionuclides in them.

The spectra were analyzed using the Canberra’s Genie 2000 software (Canberra Industries, Inc., USA). The radioactivity concentration of $^{226}$Ra was determined from the photopeaks of $^{214}$Pb (295.22 and 351.93 keV) and $^{214}$Bi (609.31, 1120.29, and 1764.49 keV); $^{232}$Th from $^{228}$Ac (911.2 and 968.97 keV), $^{212}$Pb (238.63 keV), and $^{208}$Ti (583.19 and 2614 keV); and $^{40}$K from $^{40}$K (1460.8 keV) (El-Gamal, Sidique, & El-Azab Farid, 2018).

3. Results and discussion

Environmental samples were used as SSAs to calibrate the efficiency of the NaI(Tl) detector. The activity concentrations of $^{226}$Ra, $^{232}$Th, and $^{40}$K of this sample obtained from the HPGe detector are 6421.90 ± 206.25, 2451.09 ± 96.51, and 285.50 ± 16.71 Bq kg$^{-1}$, respectively. Subsequently, the SSA was placed on the NaI(Tl) detector to estimate the net counts per second (cps) for peaks of interest. The absolute efficiency as a function of gamma-ray for the NaI(Tl) detector is calculated using Equation (1), and the values are listed in Table 1.

$$
\varepsilon(E_\gamma) = \frac{N_p}{I_p(E_\gamma) \times t_c \times A \times M}, \quad (1)
$$

where $N_p$ is the number of counts in a given peak area corrected for the background of the peak at energy $E_\gamma$ from the NaI(Tl) detector, $A$ is the specific activity of the SSA that is obtained from the HPGe detector; $I_p(E_\gamma)$ is the gamma-ray intensity per decay and therefore, $I_p(E_\gamma)$ is the abundance of energy at $E_\gamma$. $M$ is the mass of the SSA;
and \( t_c \) is the counting lifetime. The absolute efficiency values of NaI(Tl) are calculated (Table 1) using the radioactive concentration of \(^{226}\text{Ra}\) obtained from the photopeaks of \(^{214}\text{Pb}\) (295.22 and 351.93 keV) and the values are 0.095051 and 0.045937, respectively; and from \(^{214}\text{Bi}\) (609.31, 1120.29, and 1764.49 keV) and the values are 0.029546, 0.016689 and 0.011257, respectively. For \(^{232}\text{Th}\), the absolute efficiency is calculated from the photopeak of \(^{228}\text{Ac}\) (911.2 keV) and the value is 0.020305; from \(^{212}\text{Pb}\) (238.63 keV), it is 0.080115; and from \(^{208}\text{Ti}\) (2614 keV), it is 0.00771. For \(^{40}\text{K}\), the absolute efficiency is calculated from the photopeaks of \(^{40}\text{K}\) (1460.8 keV) and it is 0.013542.

The Full energy peak efficiency as a function of gamma-ray energy of a NaI(Tl) detector is shown in Figure 1(b). As shown in the figure, the efficiency curve (186.1–2614 keV) shows two regions indicating a difference in behavior due to attenuation and absorption processes. At low energies; the efficiency increases exponentially due to a marked decrease in the attenuation of the radioactive source, detector cap, or inner dead layer of the detector. The energy reaches the maximum value depending on the detector and source characteristics. Above a few 100 keV, the efficiency decreases gradually (Harb, Salahel, & Abbady, 2008).

The absolute detection efficiency is plotted as a function of the gamma-ray energy to obtain the full-energy peak efficiency curve for the HPGe detector (Figure 1(a)). As observed, the efficiency decreases drastically with an increase in the gamma-ray energy. The full-energy peak efficiency of the HPGe detector is considerably lower than that of the NaI(Tl) detector. This is because the NaI(Tl) detector used in this study is a large-area detector that can efficiently detect gamma radiation although it has a poor resolution.

### Table 1. Absolute detection efficiency of NaI(Tl) detector.

| Series or Isotopes | Isotopes | Energy | \( \eta_c \) (%) | Activity by (HPGe) | Net peak \( \eta_c \) | Efficiency of NaI(Tl) |
|--------------------|----------|--------|-----------------|---------------------|---------------------|-----------------------|
| \(^{226}\text{Ra}\) | Pb-214   | 186.1  | 0.0351          | 1122.394            | 1.4817              | 0.037611              |
|                    | Pb-214   | 295.22 | 0.1815          | 905.3897            | 15.67792            | 0.095051              |
|                    | Pb-214   | 351.93 | 0.351           | 1130.607            | 18.22975            | 0.045937              |
|                    | Bi-214   | 609.31 | 0.446           | 1070.692            | 14.10917            | 0.029546              |
|                    | Bi-214   | 1120.29| 0.147           | 1110.791            | 2.72506             | 0.016689              |
|                    | Bi-214   | 1764.49| 0.151           | 1245.374            | 2.11695             | 0.011257              |
|                    | Pb-212   | 238.63 | 0.433           | 355.6217            | 12.33645            | 0.080115              |
| \(^{232}\text{Th}\) | Ac-228   | 911.12 | 0.258           | 520.0503            | 2.724396            | 0.020305              |
|                    | Ti-208   | 2614   | 0.3564          | 367.5705            | 1.010052            | 0.00771               |
| \(^{40}\text{K}\)  | K-40     | 1460.83| 0.1067          | 55.76368            | 0.080574            | 0.013542              |

![Figure 1](image-url). Full energy peak efficiency as a function of gamma-energy for a typical, (A) HPGe detector, and (B) NaI(Tl) detector. The data points are fitted by different mathematical models as shown in the Figures.
Therefore, in comparison, the NaI(Tl) detector was found to be more efficient than the HPGe detector (Hossain, Sharip, & Viswanathan, 2012).

The mathematical models used in fitting the experimental data can be presented as the following:

- Fitting models for low energy region:
  - For HPGe M1 ⇒ efficiency = \( a \cdot (E - b)^c \)  
  - For NaI M1 ⇒ efficiency = \( \exp[a + b \cdot \ln(E) + c \cdot \ln(E^2)] \)  

- Fitting models for high energy region for HPGe and NaI detectors:
  - M2 ⇒ efficiency = \( \exp(a + b \cdot \ln(E)) \)  
  - M3 ⇒ efficiency = \( a \cdot (E - b)^c \)  
  - M4 ⇒ efficiency = \( a \cdot \exp\left(-\frac{E}{b}\right) + c \cdot \exp\left(-\frac{E}{d}\right) \)  

where \( a, b, c, \) and \( d \) are the fitting parameters of the used model. For the high energy region for the both HPGe and NaI detectors the models M2, M3, or M4 can be used with good matching.

The environmental samples investigated using both the detectors were evaluated by determining the procedure performance, following the approach adopted by the International Atomic Energy Agency (IAEA) in the recent intercomparison exercises and proficiency tests (Shakhashiro, & Mabit, 2009). The \( z_{score} \) is calculated from the activity concentration results obtained from the NaI(Tl) and HPGe detectors and the standard deviation as follows:

\[
z_{score} = \frac{A_{HPGe} - A_{NaI(Tl)}}{0.1 \times A_{HPGe}},
\]

where \( A_{HPGe} \) and \( A_{NaI(Tl)} \) are the activity concentrations obtained from the HPGe and NaI(Tl) detectors, respectively. The NaI(Tl) detector performance is considered as satisfactory if \( z_{score} \leq 2 \); debatable if \( 2 < z_{score} < 3 \); and unsatisfactory if \( z_{score} \geq 3 \).

The relative bias (RB) is calculated using the following equation:

\[
RB = 100\% \times \frac{|A_{HPGe} - A_{NaI(Tl)}|}{A_{HPGe}} \quad (8)
\]

The proficiency test results were evaluated against the acceptance criteria for trueness and precision and assigned the status ‘Acceptable, Warning, or Not Acceptable’ accordingly (Shakhashiro, & Fajgel, & Sansone, 2006).

In trueness evaluation, the results are considered Acceptable if the following condition is satisfied:

\[
|A_{HPGe} - A_{NaI(Tl)}| \leq 2.58 \times \sqrt{U^2_{HPGe} + U^2_{NaI(Tl)}} \quad (9)
\]

where \( U_{NaI(Tl)} \) and \( U_{HPGe} \) are the corresponding standard uncertainties and parameter of 2.58 for a level of probability at 99% to determine whether a result passes the proficiency test.

Precision (P) is calculated using the following equation:

\[
P = 100\% \times \sqrt{\left(\frac{U_{HPGe}}{A_{HPGe}}\right)^2 + \left(\frac{U_{NaI(Tl)}}{A_{NaI(Tl)}}\right)^2} \quad (10)
\]

The NaI(Tl) detector results are considered Acceptable for precision if the following condition is satisfied: \( P \leq \text{Limit of Acceptable Precision (LAP)} \).

In this study, for all the radionuclides, the LAP and maximum acceptable bias (MAB) were set to approximately 10%. The result should receive the Acceptable status in trueness and precision to be assigned the final status of Acceptable. In cases where either precisior or trueness is Not Acceptable, then the RB value is compared with the MAB value. If \( RB < \text{MAB} \), the result

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**Table 2.** Activity concentrations of \(^{226}\text{Ra},^{232}\text{Th},\) and \(^{40}\text{K} \) for environmental samples measured using NaI(Tl) and HPGe detectors.

| Sample Code | \(^{226}\text{Ra} (\text{Bq kg}^{-1})\) | \(^{232}\text{Th} (\text{Bq kg}^{-1})\) | \(^{40}\text{K} (\text{Bq kg}^{-1})\) |
|-------------|-------------------------------|-------------------------------|-------------------------------|
| HPGe        | NaI(Tl)                       | NaI(Tl)                       | NaI(Tl)                       | NaI(Tl)                       |
| S1          | 2219.3 ± 73.4                 | 2453.76 ± 95                  | 495.36 ± 16.7                 | 526.9 ± 50.06                 |
| S2          | 1896.18 ± 123                 | 1982.6 ± 122.7                | 704.27 ± 23.9                 | 716.65 ± 6.8                  |
| S3          | 2345.59 ± 83.6                | 2406.13 ± 174                 | 938.17 ± 36.1                 | 994.7 ± 84.9                  |
| S4          | 395.83 ± 22.74                | 395.83 ± 22.7                 | 46.43 ± 3.1                   | 50.21 ± 3.61                  |
| S5          | 197.41 ± 11.13                | 208.82 ± 15.84                | 161.3 ± 12.1                  | 164.65 ± 7.65                 |
| S6          | 110.78 ± 6.77                 | 115.49 ± 9.82                 | 128.38 ± 9.3                  | 126.51 ± 6.41                 |
| S7          | 56.42 ± 2.1                   | 50.89 ± 3.55                  | 87.13 ± 5.9                   | 74.41 ± 3.87                  |
| S8          | 35.43 ± 4.10                  | 31.95 ± 2.21                  | 44.41 ± 7.72                  | 37.93 ± 2.01                  |
| S9          | 25.91 ± 1.1                   | 23.37 ± 1.74                  | 36.24 ± 1.86                  | 30.95 ± 1.26                  |
| S10         | 14.375 ± 0.75                 | 15.7 ± 4.25                   | 8.8726 ± 0.5                  | 8.17 ± 1.25                   |
| S11         | 12.86 ± 0.553                 | 13.51 ± 2.88                  | 5.693 ± 0.28                  | 5.4 ± 0.83                    |
| S12         | 14.339 ± 0.72                 | 15.6 ± 3.65                   | 8.869 ± 0.52                  | 9.145 ± 1.513                 |
| S13         | 13.635 ± 0.801                | 14.7 ± 3.5                    | 7.499 ± 0.41                  | 7.801 ± 1.1                   |
| S14         | 14.081 ± 0.793                | 14.7 ± 2.2                    | 8.011 ± 0.64                  | 8.68 ± 0.78                   |
| Min         | 12.86 ± 0.553                 | 13.51 ± 2.88                  | 5.693 ± 0.28                  | 5.4 ± 0.85                    |
| Max         | 2345.59 ± 83.6                | 2453.76 ± 95                  | 938.17 ± 36.1                 | 994.7 ± 84.9                  |
| STDV        | 921.47                        | 947.77                        | 299.25                        | 315.04                        |
is considered as a Warning; on the other hand, if RB > MAB, the result is considered as Not Acceptable.

Table 2 lists the activity concentrations of $^{226}$Ra, $^{232}$Th, and $^{40}$K obtained using the HPGe and NaI(Tl) detectors. In the table, all the activity concentrations ranging from < 20 Bq kg$^{-1}$ to > 1000 Bq kg$^{-1}$ are listed. The maximum deviation of the $z_{score}$ is 0.958 for radionuclides of the samples; the HPGe versus the NaI(Tl) detectors values were in good agreement with it. In addition, the RB values were in good agreement for both the detectors. All the studied radionuclides passed the proficiency test of trueness and precision evaluation, except for Samples S10–S14 with regard to the precision of specific activity of $^{226}$Ra and $^{232}$Th. This is because of the uncertainty associated with the target values of these radionuclides that are found to be relatively high due to the low activity of these analytes, as shown in Table 2. The RB values are compared with the MAB for these samples, and the results are found to come under the Warning criterion. Therefore, the final score for all the analytical determinations from the proficiency test is the performance criteria that results for all the radionuclides in most of the samples are received as Acceptable and for few samples that are characterized by low activity concentrations, the results are obtained as Warning.

Figure 2. Elemental correlation for both the detectors for (a) $^{226}$Ra, (b) $^{232}$Th, and (c) $^{40}$K.
(<15 Bq kg\(^{-1}\) and <10 Bq kg\(^{-1}\) for \(^{226}\)Ra and \(^{232}\)Th, respectively; Table 2).

Figure 2(a–c) shows a good correlation between the activity concentrations of \(^{226}\)Ra, \(^{232}\)Th, and \(^{40}\)K as measured by both the detectors, where the correlation coefficient R is nearly unity in all the cases.

Ammir, Ahmed, and Hana (2014) conducted an efficiency calibration study of the NaI(Tl) detector for performing radioactivity measurements in soils from Ain Zalah oil field, Iraq, wherein the efficiency of the detector was measured using four different methods with point sources and natural radioactive samples combined with soil and Al\(_2\)O\(_3\). Our study proposed a method that determines the absolute efficiency calibration of a NaI(Tl) detector when radioactive sources with known energies are not available. The proposed method improved the overall cost-effectiveness of the detector system, overcame the limitation of finding a standard radioactive sample with exact geometric dimensions as the source samples under study, and resolved the inaccuracy of MCS to a significant extent (Chiozzi et al., 2000). Moreover, in this study, the NaI(Tl) detector was preferred to HPGe because the measurement time was found to have significantly reduced as it was a large-area detector and no sophisticated cooling equipment was required, thereby reducing the maintenance costs. The full-energy peak efficiency of the NaI(Tl) system was found to be higher than that of the HPGe detector even though it had a lower resolution.

From the 14 samples investigated in this study, only four samples that were characterized by low activity concentrations, and therefore, the uncertainty levels were considerably high for the target values of the respective nucleotides. Further studies are required in this regard to improve the accuracy of the analytical determinations.

4. Conclusions

This study presents a simple method to evaluate the full-energy peak efficiency of a NaI(Tl) detector over a wide energy range using the known specific activity of a standard sample obtained from a HPGe detector. The goal of this study is to examine the possibility of calibrating the NaI(Tl) detector using this technique in case a standard source is unavailable. For both the detectors, the obtained activity concentrations of the selected environmental samples show that this method works sufficiently well.

By using the proficiency test procedure, as supported by the International Atomic Energy Agency (IAEA), the analytical determinations are found to be ‘Acceptable’ for all the radionuclides, except for the samples characterized by low activity concentrations, which indicates a need to improve and ensure accurate measurement. The activity concentrations of \(^{226}\)Ra, \(^{232}\)Th, and \(^{40}\)K for some environmental samples are found to be higher than the recommended levels. Further investigation on more samples from this area is required to estimate the radiological hazards of these terrestrial radionuclides to the surrounding population.

Acknowledgments

The authors sincerely thank Prof. Dr. Abdelhady El-Kamel the head of nuclear physics group at Faculty of Science, Assiut University for his support during measuring, analysis and writing of this research article.

Disclosure statement

The authors declare no conflict of interest.

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