

INVESTIGATION OF CERTAIN PERFORMANCE CHARACTERISTICS OF A THALLIUM ACTIVATED SODIUM IODIDE SCINTILLATION DETECTOR

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Abstract: An investigation and quantification of certain performance characteristics of a flat type 3" x 3" thallium activated sodium iodide scintillation detector was carried out. Certain performance characteristics like the operating voltage, the energy calibration, the photomultiplier tube (PMT) gain, the detector resolution and detector efficiency of the detector were determined and discussed. A quality assurance programme was carried out in laboratories of North Eastern Hill University, Shillong, India and Bhabha Atomic Research Centre (BARC), Mumbai, India to validate our analytical method for determination of radio-nuclides in environmental samples. The quality of the data generated by cross-checking method assures that the NaI(Tl) detector which is used for our study is efficient and trustworthy whereby data generated by it can be considered authentic and of acceptable quality.

Keywords: Performance characteristics; NaI(Tl) scintillation detector; quality assurance

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1. Introduction:

A spectrometer is an instrument that provides information about both the energy and intensity of the radiation emitted from a radioactive source. A gamma – ray spectrometer using the NaI(Tl) scintillation detector is employed for our study. The most prominent property of the NaI(Tl) is its excellent light yield. As the gamma particles emitted during the decay of specific radioactive elements have very well defined energies, the measurement of specific energy gammas indicates what radioactive nuclei are decaying. The characteristic features of the spectrum produced can be interpreted by an understanding of the underlying physics. The Compton continuum arises from events which result in a partial deposition of the incident photon energy in the detector (i.e. from Compton scattering) and the Compton edge corresponds to the maximum energy that can be transferred to a recoil electron in a single Compton scatter event. A backscatter peak is often seen in gamma-ray spectra and is caused by gamma rays from the source that has first interacted by Compton scattering in one of the materials surrounding the detector. A final feature is the presence of characteristic X-ray peaks. The incident photon may interact with the material via the photoelectric effect and subsequently be absorbed by the detector [1].

The basic set up of a gamma-ray spectrometer besides the thallium doped sodium iodide detector consists of a photomultiplier tube (PMT) and PMT 'base', a pre-amplifier, an amplifier and a multi channel analyser (MCA). As the gain of the setup is a strong function of the inter-dynode potential (or the applied voltage), the resolution is expected to vary with the applied voltage. Thus, the determination of the optimum operating voltage has to be taken into consideration before any measurement is made [2].

Prior to any measurements taken, it is therefore important to quantify certain performance characteristics of the detector chosen. The most essential requirement for the estimation of gamma emitting radio-nuclides in any sample is the exact identification of the spectrum produced by the detector system and the estimation of its activity. The energy calibration establishes an exact correspondence between the channel number and gamma energy while the efficiency calibration determines the activity of a radionuclide vis-a-vis the standard used in the efficiency calibration.

In our study, the investigation of the performance characteristics of a 3" x 3" flat type NaI(Tl) scintillation detector used for our study is carried out experimentally with a good number of gamma sources. The



experimental details for determining a few factors related to the operating characteristics of the detector and a brief discussion are presented.

- 2. Experimental procedure:
- 2.1. Operating voltage:

In setting up a nuclear counting measurement, it is often desirable to establish an operating point that will provide maximum stability over long periods of time. It is often possible to vary the gain or amplification provided for the charge produced in radiation interactions. This variation can be accomplished by varying the amplification factor of a linear amplifier between the detector and counting circuit or in many cases more directly by changing the applied voltage to the detector itself [1].

To obtain an optimum operating voltage, the method described in the manual provided by the manufacturer of the gamma-ray spectrometer is followed [3]. Using a ¹³⁷Cs gamma source, the gamma-ray spectrum is acquired. The experiment is repeated for varying operating voltage values and the corresponding plots of the count rate versus base line voltage (or LLD) are also done. The resolution is calculated in each case. Finally, a plot of the resolution as a function of the applied voltage is drawn from which the optimum operating is determined. To allow the PMT to stabilise, care has been taken by allowing an elapse time of thirty minutes for each increase in voltage. This warm-up period is allowed so as to minimize the variation in output voltage and ambient temperature due to voltage shifts in the PMT.

2.2. Energy calibration:

A desirable feature of a good spectroscopy system is that there should be a linear relationship between the magnitude of the output pulse and the photon energy deposited in the detector [1]. The electric pulses extracted from the PMT are amplified by the amplifier which are then fed to a multi channel analyser (MCA) and are then registered by the computer and sorted out in the form of a histogram according to their amplitude. As the amplitude is proportional to the gamma energy, the histogram generated represents the energy distribution of the detected quanta.

The incoming pulses are read by an analog-to-digital converter, which makes a classification i.e. sorts the pulses into different boxes (channels) according to their pulse height. The channels are numbered according to increasing pulse height and the channel number is thus proportional to the gamma energy. The energy calibration is also used to determine the resolution and location of 'regions of interests' (ROI). Energy calibration for our gamma-ray spectrometer was carried out using standard caesium (¹³⁷Cs) and cobalt (⁶⁰Co) gamma reference sources.

2.3. PMT gain:

The photomultiplier tube (PMT) accomplishes the task of converting the light output of a scintillation pulse into a corresponding electrical signal. It convert light signals that typically consist of no more than a few hundred photons into a usable current pulse without adding large amount of random noise to the signal. As it is known that the overall gain of a PMT is a sensitive function of the applied voltage, it is therefore very important that sources of high voltage be well regulated and free of ripples. Any changes in the tube gain results in the drift of the operating voltage and can, if sufficiently large, deteriorate the energy resolution of the scintillation detector [1].

Using a ¹³⁷Cs source, the high voltage (HV) is adjusted until the 662 keV full energy peak is just visible on the lowest channel of the MCA. The channel number (C) along with its corresponding voltage (V) are recorded for increasing values of voltage until the full energy peak passes beyond the end of the MCA window. A graph of \log_{10} C against \log_{10} V is plotted to determine the dependence of the position of the peak channel with HV for the PMT.

2.4. Detector resolution:

The peak resulting from the photo-peak is the distinguishing characteristics of all spectra. The width of this peak is a measure of the energy resolution of the detector [2]. This width reflects the fact that a large amount of

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fluctuation was recorded from pulse to pulse even though the same energy was deposited in the detector for each event. If the amount of these fluctuations can be made smaller, the width of the corresponding distribution will also become smaller and the peak will approach a sharp spike or a mathematical delta function [1].

The energy resolution of a scintillation spectrometer is a measure of the ability to distinguish the presence of two gamma-rays closely spaced in energy. Since the essential information is contained in the photo-peak, the practical measure of the resolution is the width of the photo-peak. The most important contributor to peak broadening is the intrinsic resolution of the detector itself. This places an absolute limit on the attainable resolution since it is a fundamental property of the detector [1].

The full energy peak for any type of detector can be approximated as a Gaussian curve, where the main contributor to peak broadening is the statistical fluctuations in the number of charge carriers produced in the detector for a given deposited energy. These fluctuations could be characterised by Poisson statistics where the resolution R is related to the peak gamma-ray energy as [1]:

 $R \alpha E^{-1/2}$ (1) The energy resolution R of the detector is investigated for each of the peaks collected from the radioactive sources of ²²Na, ⁴⁰K, ⁶⁰Co, ¹³⁷Cs, ²³²Th and ²³⁸U, and is calculated by using the following formula

$$R = \frac{FWHM}{Ho} \times 100\%$$
(2)

where H_o is the peak centroid channel number.

A graph of $log_{10}R$ as a function of $log_{10}E$ is plotted and is compared with equation (1).

2.5. Detector efficiency:

All radiation detectors will, in principle, give rise to an output pulse for each quantum of radiation that interacts within its active volume. Uncharged radiation such as gamma-rays or neutrons must first undergo a significant interaction in the detector before radiation is possible. As these radiations can travel large distances between interactions, detectors' efficiency is thereby reduced. Hence, a precise figure for the detector efficiency is necessary in order to relate the number of pulses counted to the number of neutrons or photons incident on the detector [1].

To calculate the efficiency values for specific energies of the detector employed, IAEA standard samples RGK-1, RGU-1, RGTh-1 with known activity and gamma ray abundance were used. Each standard sample was counted for 50,000 seconds for five times to minimise the statistical error.

2.6. Quality assurance of measurement:

The validity of the analytical methods for determination of radio-nuclides in environmental samples (soil, water, sediments, food and vegetables) was done through quality assurance programmes. The quality control of the data can be carried out through analysis of standard reference samples, replicate analysis, cross-method checks and inter-comparison studies. The purpose of assurance programme for radio-nuclide measurement is to reduce errors and ensure that the results obtained are of acceptable quality. Thus, two concepts are involved in quality assurance programme: quality control aimed at controlling the errors in measurement and quality assessments to verify that the measurements are made within acceptable error limits.

Standard reference samples provided by International Atomic Energy Agency (IAEA), Vienna were analysed for determination of the activity concentration of the primordial radio-nuclides.

Inter-comparison studies for the estimation of the primordial radio-nuclides in a few soil samples was carried out in the Department of Physics (DOP), North Eastern Hill University (NEHU), Shillong and the Environmental Assessment Division (EAD), Bhabha Atomic Research Centre (BARC), Mumbai. Gamma spectrometry was used in the analytical technique for the estimation of radio-nuclides in both the laboratories.

The quality of the data generated during the study was also assured by cross – method checking. In this method, the same soil sample was analysed using two different analytical techniques. The analysis of ²²⁶Ra and ²³²Th activity concentration in soil samples was carried out using gamma – ray spectrometry technique in our laboratory, Department of Physics (DOP), NEHU, Shillong and neutron – activation analysis (NAA) at EAD laboratory, BARC, Mumbai.



3. Results and discussions:

3.1. Operating voltage:

Figures 1(a) to 1(e) depict the plot of the count rate versus the base line voltage (or LLD) for different operating voltage values. The corresponding resolution value is also shown. A plot of the resolution as a function of the operating voltage values is also depicted in Figure 1(f). It can be seen from Figure 1(f) that the optimum (best) resolution is obtained at 625 V and hence this value is used as the best operating for the detector in our study.



Figure 1(a): ¹³⁷Cs photo-peak spectrum at 550V.



Figure 1(b): ¹³⁷Cs photo-peak spectrum at 600V.





Figure 1(c): ¹³⁷Cs photo-peak spectrum at 625V.



Figure 1(d): ¹³⁷Cs photo-peak spectrum at 650V.



Figure 1(e): ¹³⁷Cs photo-peak spectrum at 700V.





Figure 1(f): Applied voltage versus resolution characteristics.

3.2. Energy calibration:

A plot of the typical spectrum obtained for energy calibration is shown in Figure 2(a).



Figure 2(a): Typical spectrum obtained for energy calibration.

Figure 2(b) depicts the plot of the channel number as a function of the gamma-ray peak energy. The energy calibration was carried out using the second order polynomial fit. The values of the parameters were found out to be: $a = -1.14 \times 10^{-4}$; b = 3.21 and c = 24.7. With 'a' being extremely small, the curve very nearly matches with a linear curve. This linear relationship between the peak channel number and incident gamma-ray energy shows that the quantity of energy which is deposited in the detector by the incident radiation is directly proportional to the output pulse.





Figure 2(b): Energy calibration curve obtained for the gamma-ray spectrometer.

3.3. PMT gain:

Figure 3 shows the plot of $log_{10}C$ against $log_{10}V$ for a ¹³⁷Cs source. The 662 keV full energy peak became just visible on the MCA at 470 V. As can be seen from Figure 3, a linear relationship between the channel number and applied voltage is observed. This gain is due to electron multiplication in the PMT and is proportional to the high voltage V applied to the dynode by a factor of V^n where n varies between 6 and 10 depending on the dynode material [1]. The position of the photo-peak on a particular channel number depends on the PMT gain M, which in turn is proportional to Vⁿ where V is the applied voltage ie

$$e. M = kV^n \tag{3}$$



Figure 3: Dependence of photomultiplier tube gain on high voltage.

Linear Regression for Data of Figure 3 $\mathbf{Y} = \mathbf{A} + \mathbf{B} * \mathbf{X}$



| Parameter | Value Error | |
|---------------------|--------------|---------|
| A -25.93 B 10.09 | 1.24 0.45 | |
| R SD N | Р | |
| 0.99 0.0 | 8 8 | <0.0001 |

On using the logarithm of both the channel number of peak centroid and the high voltage, the gradient is the factor n. This gradient was found to be 10.1 ± 0.4

3.4. Detector Resolution:

A graph of $\log_{10}R$ as a function of $\log_{10}E$ is plotted and is compared with equation (1). Table 1 gives the calculated values of the resolution (in %) for the different energy values of the radioisotopes used.

| Energy (keV) | Isotopes | Resolution (%) |
|--------------|-------------------|----------------|
| 511 | ²² Na | 9.9 |
| 662 | ¹³⁷ Cs | 8.5 |
| 1170 | ⁶⁰ Co | 6.4 |
| 1280 | ²² Na | 6.1 |
| 1330 | ⁶⁰ Co | 5.9 |
| 1460 | ⁴⁰ K | 5.5 |
| 1764 | ²²⁶ Ra | 5.3 |
| 2614 | ²³² Th | 3.9 |

Table 1: Resolution values for different gamma energies.

Figure 4 represents the resolution of the detector as a function of gamma-ray energy. Resolution values ranged between 9.9 % and 3.9% for gamma energies of 511 keV to 2614 keV, indicating that as the incident photon energy increases, the NaI(Tl) detector is able to distinguish the peaks of two radiations with energies that lie close to each other.



Figure 4: Energy resolution of the NaI(Tl) detector.

Linear Regression for Data of Figure 4: Y = A + B * XParameter Value Error A 2.51 0.08



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| В | -0.56 | 0.03 | | |
|-------|-------|---------|----|--|
| R | SD | N | Р | |
| -0.99 | 0.015 | 8<0.000 |)1 | |

The gradient and intercept were found out to be -0.56 ± 0.03 and 2.51 ± 0.08 respectively with a correlation value of R = -0.99. It can be concluded that the detector resolution is in good agreement with equation (1). There are a number of potential sources of fluctuations in the response of a given detector resulting in a poor energy resolution of the NaI(Tl) detector. These include any drift of the operating characteristics of the detector during the course of measurements, sources of random noises within the detector and instrumentation system, and statistical noise arising from the discrete nature of the measured signal itself. In a wide category of detector applications, the statistical noise represents the dominant source of fluctuations in the signal and thus sets an important limit on detector performance. This statistical noise arises from the fact that the charge generated within the detector by a quantum of radiation is not a continuous variable but instead represents a discrete number of charge carriers [1].

3.5. Detector efficiency:

The detector efficiencies calculated for the three radio-nuclides taken for our study were found out to be: i) 4.8% for 1460.8 keV ⁴⁰K peak ii) 3.5 % for 1764.5 keV ²²⁶Ra peak iii) 1.3 % for 2614.5 keV ²³²Th peak

A plot of the efficiency value as a function of the gamma-ray energy is shown in Figure 5. The figure depicts that the efficiency values decrease as a function of increasing gamma-ray energy. One of the factors for this decrease is because the higher energy photons spend less time in the vicinity of the detector material and are therefore able to pass straight through the material with less attenuation interaction of any kind. The counting efficiency is also strongly influenced by the sample geometry and tends to increase as the sample thickness increases.



Figure 5: Photo-peak efficiency of NaI(Tl) detector.

3.6. Quality assurance of measurement:

The observed concentrations in soil (IAEA - 375), soil (IAEA - 326) and marine sediments (IAEA - 315) along with certified values are summarised in Table 3. It can be seen from Table 7.1 that the observed value of the primordial radio-nuclides concentration measured by our gamma-ray spectrometer system varied within 10% of the certified value.



| Reference sample | Radio-nuclide | Certified value | Observed value | Accuracy (%) |
|------------------|-------------------|-----------------|----------------|--------------|
| | | $(Bq.kg^{-1})$ | $(Bq.kg^{-1})$ | |
| Soil | 40 K | 424.0 | 403.6 | 95.2 |
| IAEA – 375 | ²²⁶ Ra | 24.4 | 22.1 | 90.6 |
| | ²³² Th | 20.5 | 18.8 | 91.7 |
| Soil | ⁴⁰ K | 580.0 | 561.7 | 96.8 |
| IAEA – 326 | ²²⁶ Ra | 29.4 | 27.3 | 92.9 |
| | ²³² Th | 39.4 | 35.9 | 91.1 |
| Sediment | ⁴⁰ K | 297.0 | 281.5 | 94.8 |
| IAEA – 315 | ²²⁶ Ra | 17.6 | 16.3 | 92.6 |
| | ²³² Th | 25.6 | 23.6 | 92.2 |

Table 3: Activity concentration of ⁴⁰K, ²²⁶Ra and ²³²Th in IAEA standard reference samples.

The inter-comparison study are summarised in Table 4. It is evident from Table 4, that the variation in the measurements from both the laboratories is marginal.

| | | Activity (Bq.kg ⁻¹) | | |
|-------------|------------|---------------------------------|-------------------|-------------------|
| Soil sample | Laboratory | 40 K | ²²⁶ Ra | ²³² Th |
| S-1 | BARC | 314 ± 12 | 36 ± 11 | 68 ± 6.2 |
| | NEHU | 332 ± 11 | 39 ± 10 | 60 ± 9 |
| S-2 | BARC | 601 ± 15 | 47.1 ± 6 | 102 ± 9 |
| | NEHU | 648 ± 15 | 43 ± 7 | 105 ± 7 |
| S-3 | BARC | 227 ± 10 | 41 ± 6 | 166 ± 8 |
| | NEHU | 246 ± 9 | 49 ± 8 | 181 ± 9 |
| S-4 | BARC | 342 ± 8.1 | 980 ± 24.1 | 399 ± 20.2 |
| | NEHU | 323 ± 14 | 900 ± 17 | 345 ± 14 |

Table 4: Inter-comparison study for soil samples between laboratories of NEHU and BARC.

The results obtained by both the techniques are shown in Table 5. It is seen that the difference in measurements between the two techniques is within the tolerable limit.

Table 5: Comparison of ²²⁶Ra and ²³²Th activities in soil samples using gamma-ray spectrometer and neutron-activation analysis.

| | 226 Ra (Bq.kg ⁻¹) | | 232 Th (Bq.kg ⁻¹) | |
|-------------|------------------------------------|----------------|------------------------------------|----------------|
| Soil Sample | Gamma spectrometer | NAA* technique | Gamma spectrometer | NAA* technique |
| S-1 | 39 | 37.1 | 60 | 56.1 |
| S-2 | 43 | 46.3 | 105 | 101.9 |
| S-3 | 49 | 45.5 | 181 | 172.5 |
| S-4 | 900 | 905 | 345 | 347.2 |

* NAA – Neutron Activation Analysis

4. Conclusions:

A series of experiments with a purpose of investigating certain performance characteristics of the NaI(Tl) scintillation detector used for our study were carried out. Hence, prior to any measurements taken, certain performance characteristics like the operating voltage, the energy calibration, the photomultiplier tube (PMT) gain, the detector resolution and detector efficiency of the detector were determined and discussed. Further, the quality assurance programmes carried out in laboratories of the two mentioned departments validate our analytical method for determination of radio-nuclides in environmental samples. The quality of the data generated by cross-checking method assures that the NaI(Tl) detector which is used for our study is efficient and trustworthy whereby data generated by it can be considered authentic and of acceptable quality.

However, it is to be noted that owing to the poor resolution of the scintillation crystal, the analysis of any complex spectra is inadequate as the detector will not be able to distinguish between radiations with energies



close to each other. For a system with better resolution, knowledge about how the peak efficiency varies with gamma-ray energy can be utilised for the study of sources with unknown content and activity.

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