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Calibration of the High Purity Germanium Gamma-Ray Spectrometer in CERT, ABU Zaria, Nigeria

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Abstract

Gamma-ray spectrometry is a very powerful tool for radioactivity measurements. The gamma-ray spectrometer laboratory in Centre for Energy Research and Training (CERT), Ahmadu Bello University, Zaria, Nigeria is accredited to perform measurements of radioactive content of samples collected from the environment, food chain or industrial products with the aid of a high resolution HPGe detector. For accurate gamma-ray spectrometry, certain measurements were considered; the efficiency of the detector was performed experimentally against energies within the range of 59.50 keV (^{241}Am) to 2204.50 keV (^{226}Ra) for the respective geometries of 1 - 6 cm. The sustained solid angle relations with respect to the inverse square of sample geometries from 1 - 6 cm were evaluated. Another main point of this work was focused on the efficiency at geometry of 5 cm with respect to the three selected energies: 661.60 keV (^{137}Cs), 1173.2 keV (^{60}Co) and 1332 keV (^{60}Co) for the main axis, ten degree off main axis, forty five degree off main axis and ninety degree off the detector main axis. In order to verify optimum geometries in our laboratory for both short lived and long lived radionuclides analyses, the evaluation of efficiencies for the respective energies: 1173.2 keV (^{60}Co), 1332.5 keV (^{60}Co), 1764 keV (^{226}Ra) and 2294 keV (^{226}Ra) were plotted against geometries of 1 to 6 cm from the detector end cap along the main axis.

Keywords

Gamma-Ray Spectrometry, Solid Angle, Efficiency, Radioactive Measurements

1. Introduction

Gamma-ray spectrometry using High Purity Germanium (HPGe) detectors has been an essential and principal

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spectroscopy technique in almost all radioactivity measurements laboratories worldwide [1] [2]. Its major advantages are being non-destructive, multi-elements analysis, simplified regarding sample preparation, *i.e*. mostly no need for any chemical separation processes, and its applicability for all types of samples, etc. [3]. In accredited laboratories, the gamma-ray spectrometry method is used to perform both qualitative and quantitative radioactivity analysis, for solid, liquid and gaseous samples [4] [5]. A typical gamma-ray spectrometry system consists of a detector with a shielding container—mainly leads to reduce the background interferences, high voltage power supply, electronics for signal processing (preamplifier, amplifier, multichannel analyzer), computer and dedicated software. The spectrometric system records, stores and pro- cesses the gamma-ray spectrum of the analyzed sample, using validated computer software packages [4] [6]. A proper energy calibration is needed to identify the energy of the gamma ray emissions from the spectrum, *i.e*. the gamma-ray emitter radionuclides contained by the sample (qualitative analysis). The quantitative analysis, *i.e*. the activity (expressed in Becquerel, SI units) and its standard uncertainty determination—for each radionuclide present in the sample, requires a full-energy peak (FEP) efficiency calibration [7] [8]. For efficiency calibrations and effective activity evaluation, various radioactive standard sources with certified activity are necessary. There are many technical and methodological aspects to be taken into account for an optimal radioactivity analysis which includes: careful choice of the detector (type, geometry, window) and the radioactive standard sources used for calibration; sample geometry, matrix, position relative to the detector; passive or active detector shielding; various corrections: background, dead time, geometry (sample different from the standard), deconvolution of overlapped spectrum peaks, true coincidence summing [1] [9]. The high resolution detectors (HPGe) are suitable for samples containing many radionuclides (*e.g*. from the natural radioactive series), when the gamma- ray spectrum presents a large number of peaks to be resolved. These detectors are expensive and must be kept at very low temperatures (in liquid nitrogen) for a correct functioning [10].

Source-detector geometry is an important factor in bringing out the major characteristics of HPGe detectors. Going by the ANSI/IEEE standard, 1996, a geometry of 25 cm is usually chosen to evaluate values of relative efficiency, peak-to-Compton ratio at a particular photon-peak is 1332.5 keV of ^{60}Co. In practice, utilization of germanium detectors has progressed to such a degree that IEEE standards for specifying the performance of these detectors are no longer adequate to either predict their efficiency for all geometries or given spectral energies for specific experimental situations [8]. Germanium detectors are now being used in more varied applications with varying requirements [11]. The specification of the relative efficiency and peak-to-Compton ratio for a ^{60}Co point source at 25 cm distance cannot be applied on a universal case as this efficiency data does not predict how a detector will perform at low or high energies except for the photon energy of 1332.5 keV and 25 cm geometry [12].

The aim of our work is to shed more light on the significance of experimental evaluation of efficiency against energies within the range of 59.50 keV (^{241}Am) to 2204.50 keV (^{226}Ra) for the respective geometries of 1 - 6 cm; sustained solid angle relations with respect to the inverse square of sample geometries from 1 - 6 cm; efficiency with respect to the three selected energies; 661.60 keV (^{137}Cs), and 1173.2 keV; 1332 keV (^{60}Co) with a constant radius of 5 cm from the main axis, ten degree off main axis, forty five degree off main axis and ninety degree off the detector main axis. This will enable us to develop measurement positions for both short lived and long lived samples.

2. Theoretical Overview

The precise determination of the activity concentration of each radionuclide requires the determination of full energy efficiency calibration for a given geometry. Therefore, a detection efficiency curve, known as efficiency calibration, over the energy region of interest must be established precisely in advance. The detection efficiency at certain gamma-ray energy and sample geometry is given by:

$$\varepsilon(E,n)=\frac{C(E,n)}{f(E,n)At_C} \tag{1}$$

where $C(E, n)$ = net photo-peak count of gamma-ray transition with energy E of n-radionuclide,

t_c = counting time, sec.,

$f(E, n)$ = branching ratio, number of photon with energy E per hundred disintegration of n-radionuclide,

A = activity concentration in Bq of n-radionuclide.

The detection efficiency curve depends not only on a detection system but also on both the sample shape and matrix [5]. The detection of efficiency curve experimentally can be performed using standard samples that contain a set of radionuclides with known activities and cover the gamma-ray energy range of interest (usually from 59.54 to 2204.5 keV). Standard or reference samples should have the closest specifications, regarding geometry and matrices (apparently density and composition), to the analyzed samples [4] [13] [14]. The accurate determination of the photo-peak efficiency curve for a given sample matrix represent the main challenge in gamma-ray spectrometry [15]. Practically, the samples geometry (shape and sample-detector geometry) can be easily reproduced using the established technique [16].

Gamma rays (photons) are generally emitted equally in all direction thereby covering a solid angle for a point source positioned at geometry "*d*", defined as [16]

$$\text{Solid angle } \Omega(d) = 2\pi\left[1-\left(1+\frac{R_D^2}{d^2}\right)^{-\frac{1}{2}}\right] \tag{2}$$

where $\Omega(d)$ = effective solid angle, d = specific geometry, and R_D = radius of the detector end cap.

The buildup of full energy peak efficiency ε is governed by the proportion of the intercepted space by the detector active area (A) which is given as:

$$\text{Detector active area } (A) = 2\pi d^2\left[1-\left(\frac{d^2}{d^2+R_D^2}\right)^{\frac{1}{2}}\right] \tag{3}$$

The experimental efficiency curve referred to as reference efficiency curve is related to the solid angle with respect to geometries as:

$$\varepsilon(d_j) = \left[\frac{C(E,n)}{f(E,n)At_C}\right]_{d_i} \times \left[1-\left(1+\frac{R_D^2}{d^2}\right)\right]_{d_j} \times \left[1-\left(1+\frac{R_D^2}{d^2}\right)\right]_{d_i}^{-1} \tag{4}$$

where $\varepsilon(d_j)$ = efficiency at j^{th} position

All symbols retain their meanings; where i = 1, 2, 3,⋯ and j = 1, 2, 3,⋯

2.1. Experimental Work

Five point gamma-ray emitter sources: 59.5 keV of ^{241}Am; 241.9 keV, 295.2 keV, 351.9 keV, 609.3 keV, 1274.5 keV and 2204.5 keV of ^{226}Ra; 1173.2 keV and 1332.5 keV of ^{60}Co; 661.6 keV of ^{137}Cs; 121.8 keV, 244.7 keV, 344.2 keV, 778.92 keV, 964.11 keV, 1112.07 keV and 1407.9 keV of ^{152}Eu obtained from the published work of [17] were utilized to perform solid angle transmission measurements with respect to efficiency and some basic calibrations parameters. Each of the five point sources were placed on the top of the detector and counted for 3600 second at each of the geometries (1 - 6 cm) in which a low acceptable dead time and acceptable low statistical error were observed and the result of efficiencies against energies transitions were plotted in **Figure 1** and **Figure 2**. Two of the point gamma-ray emitter sources: ^{60}Co and ^{137}Cs were used to investigate efficiency distribution along varying angles in a circular path of radius 5 cm. Each of the two point sources were placed at main axis, ten degree of the main axis, forty five degree of the main axis and ninety degree of the main axis and counted for 3600 seconds and the results were shown in **Figure 3**.

After accumulating sufficient counts (3600 seconds) by an multi-channels analyzer (MCA) for each of the peaks with respect to the sources, the MAESTRO emulation software program was used to obtained the net full peak (background counted for 3600 seconds was subtracted) counts for each photon of interest with gamma-ray emission probability of 13% and above [11]. The activity of each sources were normalized to the measurement date before obtaining the full energy peak efficiency ε for a particular sample-to-detector geometry.

2.2. Gamma Ray Spectrometry

Gamma-ray spectrometer with an extended range electrode germanium detector with a (ORTEC©) Model

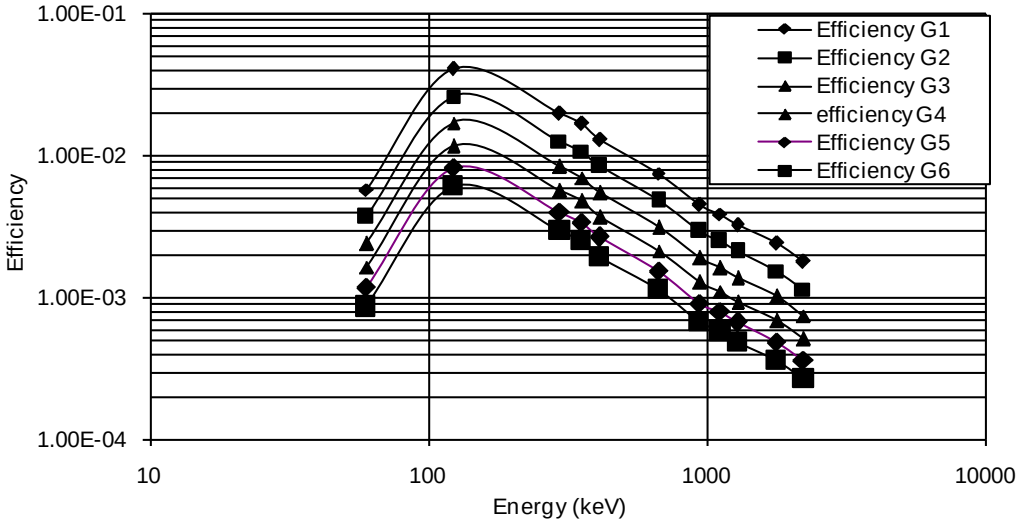

Figure 1. Efficiencies generated against energies distributions.

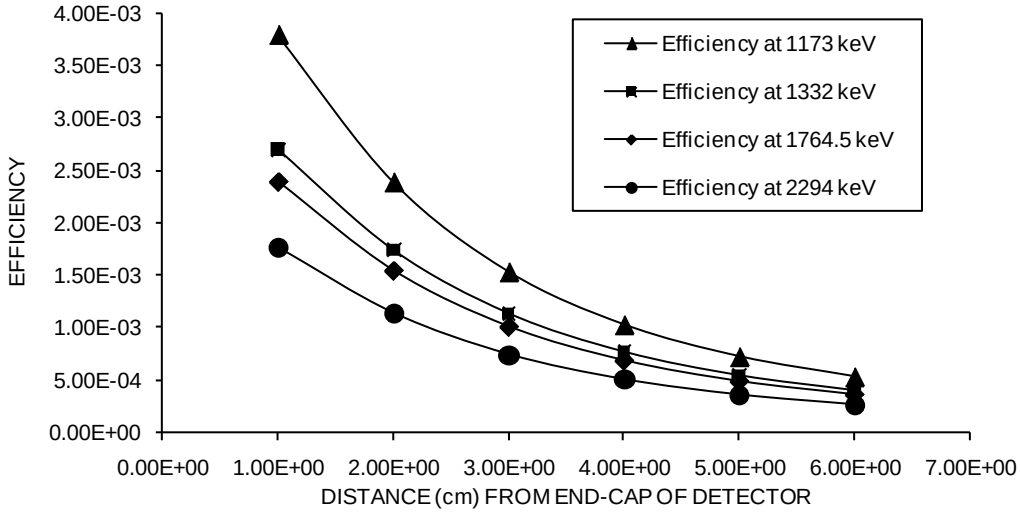

Figure 2. Efficiency distribution against geometries.

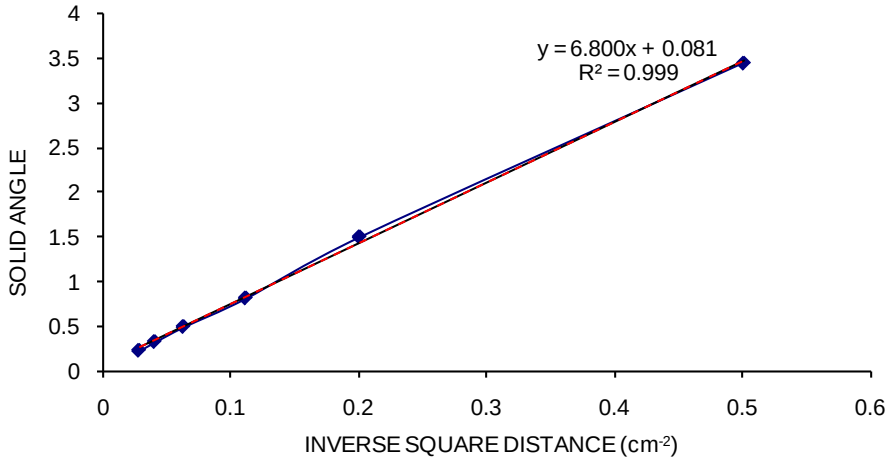

Figure 3. Inverse square distances against sustain solid angle generated.

number "GEM-30195 was used. The HPGe detector has a relative efficiency of 43.4% and full width at half maximum (FWHM) of 1.80 keV for ^{60}Co gamma energy transition at 1332.5 keV and amplifier time constant of 6 μs. It is connected to a computerized MCA where gamma spectrum analysis is performed using the MAESTRO emulation software program. Other performance specifications provided in the quality assurance data sheet are listed in **Table 1**.

3. Results and Discussions

Gamma-ray spectrometry based on high purity germanium detectors is a very powerful tool that has a very wide range of applications in radiation measurement, generally and specially in environmental radioactivity measurement in different bulk samples. Many studies have been focused on the accurate detection efficiency calibration using different, experimental and analytical techniques [9] [16] [18]. However, the optimum sample-detector geometry arrangement could be the main source of noticeable uncertainty, especially for the samples with high apparent densities and different chemical compositions from that of the standard samples used for efficiency calibration. The variation of efficiencies against energies and the geometries are shown in **Figure 1** and **Figure 2**.

In order to verify optimum geometries in our laboratory for both short lived and long lived radionuclides analysis, the evaluation of efficiencies for the respective energies: 1173.2 keV (^{60}Co), 1332.5 keV (^{60}Co), 1764 keV (^{226}Ra) and 2294 keV (^{226}Ra) were plotted against geometries of 1 to 6 cm from the detector end cap along the main axis as shown in **Figure 2**. This figure shows an almost stable efficiency data around the 5 cm regions and as we move toward lower geometries of 4 cm to 1 cm these data deviate significantly. At 1 cm, the separation of the energies was very significant and was adopted for long lived radionucliudes analysis while 5 cm was adopted for short lived radionuclides measurements.

The experimental derived efficiency data (values) and the instant effective solid angle data for 1 cm to 25 cm geometries have been generated and published elsewhere by [16]. However, the sustained solid angle relations with respect to the inverse square of sample geometries from 1 - 6 cm were evaluated and shown in **Figure 3**.

From **Figure 3**, it shows that the sustained solid angle is proportional to the inverse of the distances from the detector end cap. This implies that the closer the geometry, the better the area of gamma-ray coverage and detection by semi conductor detector. This further confirmed our choice of 1 cm and 5 cm geometries adopted in our laboratory. A work by Ahmed (2013) studied parameters needed for calibration of a GEM-30195 (39-P21439A) coaxial HPGe detector at the Nigeria Research Reactor-1 (NIRR-1) laboratory with a view to obtain the best geometry that would be adopted for routine analytical work and ensuring quality control addressed the detector's resolution, peak-to-Compton ratio and efficiency at four different geometries (1 cm, 3 cm, 5 cm and 7 cm) and 5 cm geometry was found to be the best. Since the range of 4000 keV energy was of interest at NIRR-1 laboratory, an extrapolation was done to obtain the tail end of the efficiency curve to capture elements like calcium.

The effect of geometry change on the detector's resolution and efficiency was also established by [9]. For quality assurance, seven standard reference materials (SRM) of different matrix obtained from NIST (Coal Fly Ash, Tomato Leaves and Total Diet) and IAEA (Soil 7, Lake Sediment, Cabbage and Lichen) were measured at 1 cm and 5 cm and the results obtained compared favorably with the certified values. This work also stressed the need for the NIRR-1 laboratory to obtain ^{22}Na, ^{54}Mn, ^{57}Co and ^{88}Y sealed sources for future detector parameter measurement, efficiency calibration and quality assurance work. However, it has been shown that geometry change has no effect on the detector's resolution but it does on its efficiency.

Table 1. The specifications of GEM-30195.

	Warranted	Measured	Amplifier Time Constant
Resolution (FWHM) at 1.33 MeV, ^{60}Co	1.95 keV	1.80 keV	6 μs
Peak-to-Compton Ratio, ^{60}Co	54	70.7	6 μs
Relative Efficiency at 1.33 MeV, ^{60}Co	30%	43.4%	6 μs
Peak Shape (FWTM/FWHM), ^{60}Co	1.98	1.88	6 μs
Peak Shape (FWFM/FWHM), ^{60}Co	2.98	2.51	6 μs

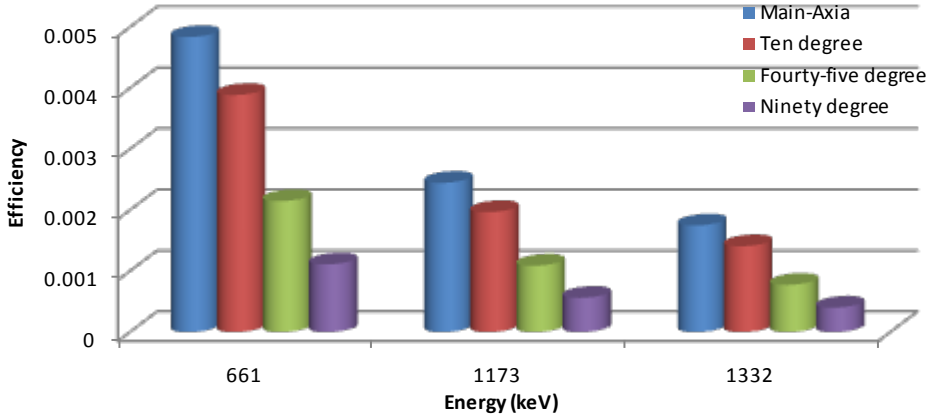

Figure 4. Relation of efficiencies distribution against sources placed at main to ninety degree.

Another main point of this work was focused on the efficiency at geometry of 5 cm with respect to the three selected energies: 661.60 keV (^{137}Cs), 1173.2 keV and 1332 keV (^{60}Co) for the main axis, ten degree off main axis, forty five degree off main axis and ninety degree off the detector main axis and the results were plotted in **Figure 4**.

As clearly observed, the efficiency distribution for the two point gamma-ray sources with respect to the energy transmissions: 661 keV (^{137}Cs) and 1173.2 keV (^{60}Co), 1332 keV (^{60}Co) indicated that counting at the main axis from the end cap gave high efficiency values compared to placing the points sources or any analytical samples away from the main axis. The lowest was observed when place perpendicularly to the main axis. However, all the placements points showed the same pattern of efficiency distribution.

4. Conclusion

The suggested procedure introduced in this work is an innovative, reliable and straightforward method to overcome the errors generally produced due to the difference in samples geometry analysis. It also minimizes the measurement errors and establishes the adopted measuring geometries in the instrumental neutron activation analysis (INAA) in laboratory in CERT, ABU, Zaria. This work also stresses the need of not shifting the samples for analysis away from the detector main axis. This work could be adopted within the laboratories where they encounter incoming wide varieties of point-like samples for analysis. Finally, this work basically reveals that efficiency curve does not depend only on the detection crystal but also on the solid angle area generated by the detector active area.

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