Research Article

Journal of Electrical Electronics Engineering

Towards the Reliability Check of Complementary Methods for the Precise Estimation of Peak Efficiency of a Long Run Used Germanium Detector in Low Activity Environmental Measurements

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Submitted: 19 Aug 2022; **Accepted:** 26 Aug 2022; **Published:** 13 Sep 2022

Citation: *Margret M*, Chandrasekaran S, Srinivas CV, Venkatraman B. (2022). Towards the Reliability Check of Complementary Methods for the Precise Estimation of Peak Efficiency of a Long Run Used Germanium Detector in Low Activity Environmental Measurements. J Electrical Electron Eng, 1(1), 31-38.*

Abstract
The gamma-ray spectrometry using long run used hyper pure germanium (HPGe) detector needs a stringent full energy peak (FEP) efficiency calibration for environmental radioactivity measurements. The FEP efficiency calibration was carried out using the complementary methods like analytical estimations and Monte Carlo (MC) simulations. In our work, for the analytical estimations, the relative deviation of the theoretical results with that of experimental FEP efficiency was found to be less than 6%. However, in the case of MC methodologies, an overall estimated standard deviation of 5% could be achievable only with the optimized geometry of the long run used germanium detector for low-activity environmental measurements.

Key Words: Gamma ray Spectrometry, HPGe, Photons, Efficiency curve

Introduction

The details about radiation levels and radionuclide distribution in the environment caught attention worldwide for assessing the effects of radiation exposure [1]. The naturally occurring radionuclides, such as Th-232, Ra-226 and as well their daughter progenies and K-40 emit gamma radiations thereby contributing significantly to enhance the natural background radiation level in the environment [2]. For a wide energy ranges, gamma-ray spectrometry using hyper purity germanium detector (HPGe) is the ideal technique for detecting and quantifying radionuclides [3]. The reason is that the vast majority of radionuclides emit gamma rays with very well defined and characteristic energy. In order to acquire good quality results, exact knowledge of detector full energy peak (FEP) efficiency relevant to the particular measurement conditions is essential [4]. The detection efficiency is the percentage of radiation detected by a given detector to the total yield emitted from the source. The worldwide procedure for carrying out the efficiency calibration is by means of radioactive sources of equivalent geometry, chemical composition and density as of the measured sample. Prior to the measurements, the activity of the source and their emission probability over the region of interest should be known. The advantage of using standard radioactive source is that it contains the details of activity of the radionuclides associated with its uncertainty, reference date, purity, chemical composition, mass or volume, emission probability for all modes of decay and half-life values. To obtain the response function (FEP efficiency calibration) of the detector, radioactive sources generally used are monoenergetic (e.g. Am-242, Cr-51, Cd-109, Hg-203, Zn-65, Cs-137) or multi gamma ray point sources (e.g. Eu-152 and Ba-133). Although using these sources can yield superior results, they have their own limitations that can lead to inaccuracies in the final FEPE calculation for the following reasons: (i) Some of the sources like Cr-51 ($t_{1/2}$-27.7 d) Hg-203 ($t_{1/2}$-46.6 d) are not very long lived and they need to be replaced periodically (ii) As multi-gamma sources produce gamma rays in cascade, there may be a significant coincidence summation effect for close sample-to-detector distances, resulting in erroneousness efficiency calibration [5]. Hence, alternative methods are required for obtaining the FEP efficiency values of the detector used for analysis. The complimentary method namely theoretical estimation could be the proper choice, but deployment of many sources to cover from low to high-energy regions can result in huge uncertainty when interpolating using fitted curves. Hence, there lies a restriction of using multiple sources, wherein radioactive source having wide energy range coverage should be used to eliminate the erroneous results while obtaining the efficiency curve. To overcome all the mentioned scantiness, the other complementary method namely Monte Carlo (MC) modelling is becoming increasingly desirable. The conventional usage of the modeling is limited due to the requirement of substantial computer time per energy and inadequacy of accurate detector geometry from the manufacturers [6]. However, Monte Carlo simulations have become more reliable now-a-days due to the latest improvements in computerization [7]. They can be used to obtain the FEP efficiency with great accuracy at any energy, provided the detector model is appropriate. The quality of the detector modeling is generally checked by comparing the measured experimental efficiency with that obtained from simulations [8].

In this paper, an experimental determination of the full energy peak efficiency of a long run used HPGe detector using IAEA standard source has been explained. A new fit function has been formulated to calculate theoretically the FEP efficiencies of an HPGe detector within 200 keV-2700 keV energy range. Using the theoretical efficiency, the activity of certified reference material IAEA-375(soil) has been validated. In addition, the MC modelling has been developed for the same HPGe detector and the FEP efficiency is calculated with appropriate modifications in the detector geometry for the mentioned energy range. The results obtained from the theoretical and MC simulated is compared with the datas obtained through experimental determinations for our in-house built gamma spectrometry system designed for the low-radioactivity measurements.

Materials and Methods

Measurement System

The gamma ray spectrometry system consists of coaxial p-type HPGe detector attached to a 16K multichannel analyzer is used for spectral measurements. The detector when it had been procured has a resolution (FWHM) of 1.8 keV at 1332 keV Co-60, but now the resolution has slightly deteriorated due to its ageing. The peak-to-Compton ratio and peak shape (FWTM/FWHM) is 60:1 and 2 respectively both for Co-60. To obtain the low background required for environmental applications, the detector and the pre-amplifier are enclosed in a 10cm thick lead shield. The contribution of the lead characteristic X-rays are reduced with the lining of 3mm cadmium and 1.5mm copper. A pulse height analysis system having a signal processor transforms pulses, records and stores in a computer-based MCA. The Wilkinson type analog to digital converter (clock rate of 100 MHz) is used to convert analog to digital signals and the datas are stored in 8192 sequential channels [9-11]. The FEP efficiency calibration has been carried out using Reference Gamma (RG) standard sources in 250ml geometry. The counting time is four hours to establish the region of interest (ROIs) on the spectrum of the detector. For the same measurement time, the background spectrum has also been collected. The spectral analysis is conducted up to 2.7 MeV energy.

Measurement Procedure

Energy Calibration

For energy calibration, the pulse height scale should be calibrated in terms of energy to identify the photo-peaks present in the gamma ray spectrum. This is generally carried out using standard sources like Cs-137 and Co-60 as these sources are having well specified energies in the desired range of interest. The relationship between energy and the channel number is given as follows

$$E = D*C+F \qquad \text{......... (1)}$$

Where E the gamma energy (keV), C the channel number corresponding to the energy E, D and F are calibration constants.

FEP Efficiency Calibration

The efficiency calibration establishes the inter-relationship between the counts that is under a photo-peak and the gamma standard source activity [12]. The photo-peak efficiency of the detector is determined using IAEA Reference Gamma standard Uranium (RGU), Reference Gamma standard Thorium (RGTh) and Reference Gamma standard Potassium (RGK). The RGU is mounted on the detector and it is counted for the prescribed time mentioned already. The photo peak counts for the energies of 295 keV, 352 keV, 609.3 keV, 1120.2 keV and 1764.49 keV for a pre-defined energy window width is noted. The net peak counts are obtained from subtracting the background counts for the same region of interest. The FEP efficiency (ε) can be obtained in terms of gamma-ray energies using RGU as

$$\varepsilon = \frac{N}{A*I*t}\ \% \qquad \text{......... (2)}$$

Where N is the counts for particular gamma energy, A is the radioactivity of radionuclide of interest in reference material in Bq, I is the emission probability (yield) in %, t is the counting time in sec. The combined standard uncertainty, Uε/ε of the FEP efficiency (ε) is calculated as per equation (3), where k is the coverage factor, $(U_N/N)^2$, $(U_A/A)^2$, $(U_I/I)^2$, $(U_t/t)^2$ and $(U_M/M)^2$ are the measurement of uncertainties for the peak net area measurement, activity, gamma yield, counting time and mass of the sample. The same set of procedures are repeated for RGTh for the gamma energies 209.2 keV, 238.63 keV, 911.2 keV and 2614.5 keV and as well as for energy of 1461 keV of RGK.

$$\frac{U\varepsilon}{\varepsilon} = k*\sqrt{\left(\frac{U_N}{N}\right)^2+\left(\frac{U_A}{A}\right)^2+\left(\frac{U_I}{I}\right)^2+\left(\frac{U_t}{t}\right)^2+\left(\frac{U_M}{M}\right)^2} \qquad \text{......... (3)}$$

Computational Modelling

The experimental setup has been modeled using the MCNP4B Code and is a general-purpose code that is being used for neutron, photon, electron or coupled photon/electron/ neutron transport [13]. A comprehensive physical treatment including the incoherent scattering with form factors, the photoelectric effect with X-ray fluorescence and pair production are included in the energy range between 1 and 2000 keV. This code is unique in that it follows the path of each photon until the energy is expended in the detector. This code includes extensive cross-section libraries for simulating photon transport across a wide energy range and for variety of materials. The input file for MC code should be defined precisely to get the accurate results. The information of the file comprises about the characteristic of geometry materials specification, choice of cross-section, the features of the photon source and the tallies. MC simulation takes a long time to achieve statistically meaningful results in FEP efficiency estimates due to the isotropic nature of the source and the limited solid angle of the detector. Hence, a variance reduction technique has to be adopted to redirect the photons to the detector thereby reducing the computing time. Source biasing is the only variance reduction methodology allowed with F8 tallies having energy bins and it has been implemented in the modelling to increase the efficiency.

Figure 1 shows a vertical 2D layout of the detector geometry and the cylindrical source used in MCNP modelling. The simulated model includes Germanium crystal, inner void cavity, dead layer, lithium contact, aluminum end cap and a radioactive source on contact with the detector. The detector and the source modelling have been made in accordance with the manufacturer's design so as to maintain the dimensions and materials of the various components as defined by the manufacturer. In the simulation, the modelling of the detector's crystal is crucial because it is the region where the gamma energy deposition is accounted. The material of the crystal and the dead layer is assumed germanium. An important parameter that has been taken care in the detector model is the thickness of the dead layer, since this parameter has a larger influence on the results of calibration simulations [14]. The parameter has been calculated to match the experimental data using suitable method and is not within the scope of this work. The F8 (Pulse Height Distribution) tally i.e. energy deposited in the detector has been used to model the detector. The spectrum peaks obtained in the experiment are generally in the form of Gaussian distribution. The MC code, on the other hand, does not model the physical process that leads to spectral broadening [15]. To account for the realistic model of the detector, the energy broadening option must been included while simulating the spectra, taking into account of FWHM (Full Width at Half Maximum) from the calibrated spectrum. The Gaussian broadening for the tallied energy is,

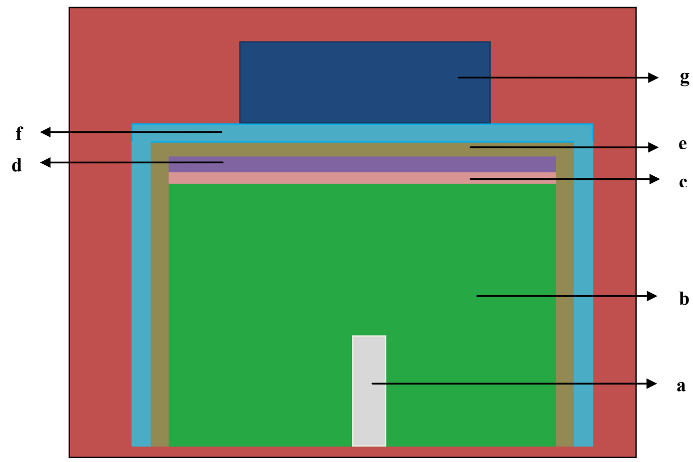
$$g(E) = D * \exp\left[-\left(\frac{E - E_o}{W} \right)^2 \right] \quad \text{……… (4)}$$

Where D is a normalization constant, Eo and E are the un-broadened and broadened energy and W is the width of the Gaussian curve.

$$W = \frac{FWHM}{2.35482} \quad \text{……. (5)}$$

However, full width half maximum is mathematically defined [17-19] as

$$FWHM = p + q * \sqrt{E + rE^2} \quad \text{……. (6)}$$



(a) Inactive core (b) Germanium crystal (c) Dead layer (d) Lithium contact (e) Air (f) Aluminium end cap (g) Radioactive source

Figure 1: Layout for detector set-up in MCNP Modelling (Not to scale)

Where *E* is the incident gamma energy and p, q, and r are the parameters with the units MeV, $MeV^{1/2}$, and MeV^{-1} that are obtained by fitting Eq. (6) with the experimental FWHM versus energy. Volumetric radioactive sources emitting multiple gamma rays at various energies have also been modeled to simulate the gamma ray spectrum of the HPGe detector. When the history of a photon has been completely followed and its response to the detector is assessed, a count will be logged in a bin according to its energy. The simulated results are typically normalized per photon emitted by the source. New source photons are generated at random until a predetermined number of histories have been tracked, after which the simulation is terminated. The total number of histories should be substantial to achieve minimum error in the calculated efficiency. In our work, a maximum relative error of 0.04 has been observed for 4E+08 source particles.

Results and Discussion

When HPGe detector is employed for low-level radioactivity measurements, two procedures should be adopted for calibrating the detector: energy calibration and efficiency calibration. These calibrations enable the radioisotopes to be appropriately identified and their activity could be quantified in the matrix under study.

Energy Calibration

The graph shown in the fig 2 represents the energy calibration for the HPGe gamma spectrometer used for the experiment. If E is the energy associated to channel number Ch, then the spectrometer is been energy calibrated using the equation (1). The typical coefficients values obtained from the fit are the offset coefficient, F = 0.39 and the slope, D = 5.71. The plot for energy versus channel number is a linear fit (with a coefficient of regression of 1) and from the curve, the channel at which the gamma energy peaks appear could be identified from the graph.

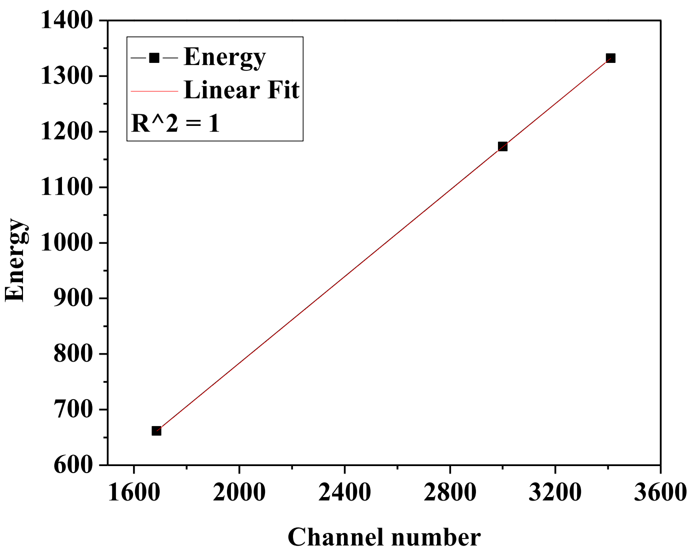


Figure 2: Energy calibration curve of HPGe gamma detector using the energy calibration standards Cs-137 and Co-60

FEP Efficiency calibration

Measured and calculated FEP efficiency

The experimental efficiency calibration of an HPGe detector is performed by using standard sources containing known activities of radionuclides. Sixty peaks are identified for the selected radionuclides (RGU, RGTh & RGK) and only 10 prominent peaks, which are having almost higher yield, covering the energy range from 200 keV to 2700 keV, are taken for the efficient calibration of the system. The FEP efficiency values are calculated using the equation (2) and are displayed in table 1.

Fig 3 shows the FEP efficiency curve for the HPGe detector as a function of gamma energies from the standard source. It is seen from the graph, that at low energy (209.2 keV), the efficiency is found to be "the maximum" where the gamma ray deposits all its energy to the detector. However, as the energy increases, the efficiency decreases monotonically due to less interaction within the detector and as well as the absorption between the components of the source and the detector. It is also evident that for photon energies above 200 keV, the efficiency calibration is distinctive for the source-to-detector geometry in our experimental set-up. There is only a slight variation (<1%) in the counts registered due to positioning, composition and density change in the source. A single efficiency calibration curve is sufficient for the existing sample-to-detector geometry. Since only a certain number of calibrated efficiency points are taken for the whole energy range, a fitting procedure is required to generate a continuous FEP efficiency function. A single function with logarithmic power series has been used and the following fitted equation (eq-7) could be used to adequately represent the experimental points for the entire cited energy range.

Table 1: FEP Efficiency (%) for the energy range 200 keV - 2700 keV

IAEA Standards	Isotopes	Parent source	Energy(keV)	FEP Efficiency (%)
RGTh	^{228}Ac	^{232}Th	209.2	2.6±0.26
RGTh	^{212}Pb	^{232}Th	238.63	2.398±0.239
RGU	^{214}Pb	^{238}U	295.0	1.901±0.190
RGU	^{214}Pb	^{238}U	352	1.761±0.176
RGU	^{214}Bi	^{238}U	609.3	1.183±0.118
RGTh	^{228}Ac	^{232}Th	911.2	1.089±0.108
RGU	^{214}Bi	^{238}U	1120.2	0.827±0.083
RGK	^{40}K	-	1461	0.611±0.061
RGU	^{214}Bi	^{238}U	1764.49	0.620±0.062
RGTh	^{208}Tl	^{232}Th	2614.5	0.519±0.052

$$\varepsilon = \frac{A1 + \frac{A2}{A3} * \ln(E) + \frac{A4}{E} * \ln(E^2) + \frac{A5}{E} * \ln(E^3)}{E^{0.75}} \% \qquad \text{.......} (7)$$

Where ε is the FEP efficiency, E is the gamma ray energy in keV and A1 – A5 are the function fitting parameters. The unique feature of this calibrated curve is that, it has maximum parameters to hold the shape of the curve, yet within the available calibrating points. Even though the shape of the curve is in near proximity to the experimentally obtained measurements, the said function fits the data with linear regression having reduced χ^2 = 0.99 (Fig 3). Moreover, the individual uncertainties of the deviations from the fitted curve (cal- exp)/cal are good in agreement with a maximum relative deviation of 6%. In the residual plot (Fig 4), the residues bounce randomly around the zero line and no specific pattern is observed, which indicates a decent fit to the efficiency data under consideration.

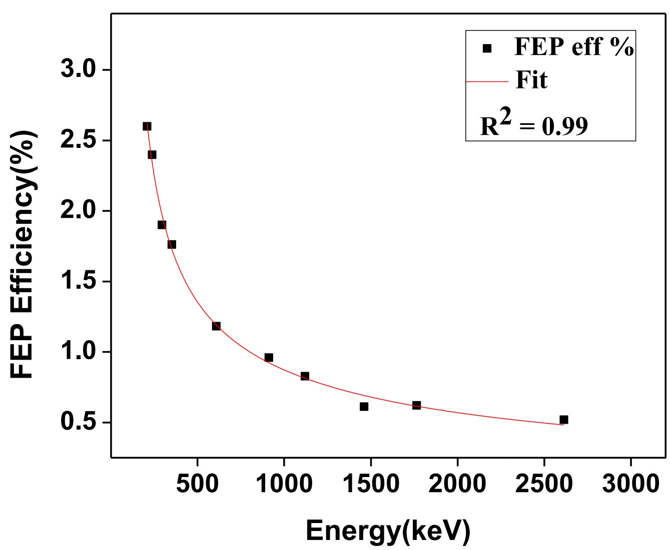


Figure 3: FEP efficiency (%) for the detector against gamma energy

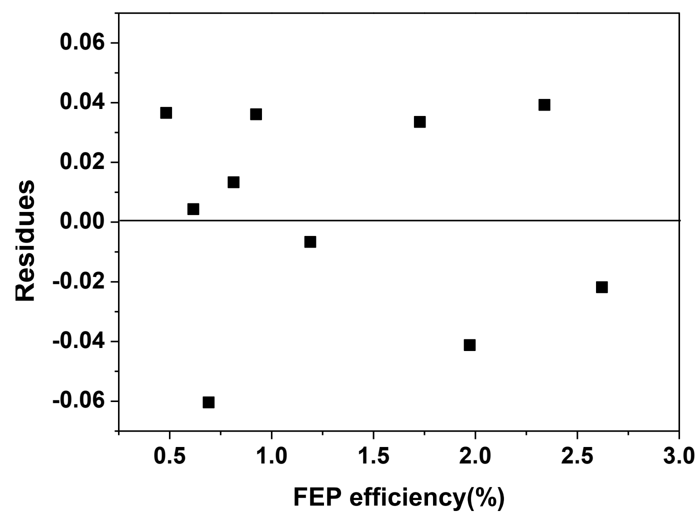


Figure 4: Residual graph obtained from the differences between calculated values and the experimentally obtained

The correlation of the data obtained from the fitting function has been compared with the experimental values, by plotting efficiency ratios versus their respective energies (Fig 5). The results reveal that, at higher energies, the efficiency ratios do not stay constant at unity, but instead fluctuate at energy level, with a maximum and minimum at 1461 keV and 2614.5 keV respectively. These fluctuations are due to the characteristic features of the germanium detectors and these variations account for the partial differences between the experimental and calculated FEP efficiency values [20]. Though the continuous fitting function shows uncertainties and performs inferior than the individual calibrations, it can be a better alternative to describe the detector property characterization for a wide energy range concerning environmental studies.

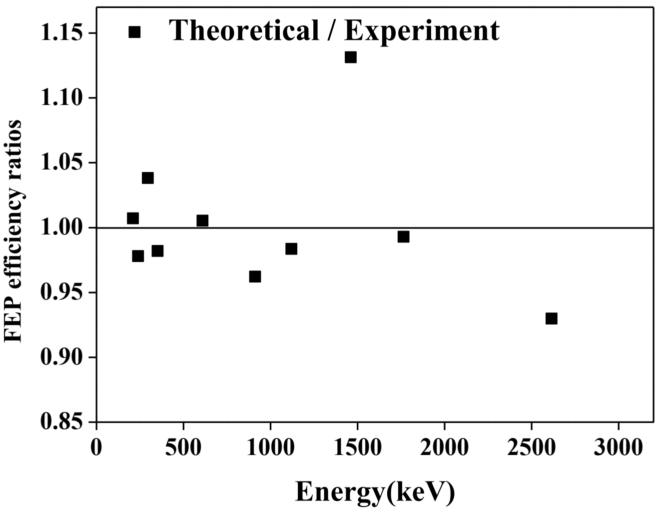


Figure 5: FEP efficiency ratios for the investigated energy range

Validation of theoretical FEP efficiency

When the gamma spectrometer has been calibrated with the theoretical FEP efficiency values, the Cs-137 activity of IAEA 375 standard can be determined using the continuous efficiency function. The procedure is followed as described in previous section with the same geometry. The activity (Bq/kg) of the Cs-137 gamma energy has been calculated by re-writing the equation (2) and the efficiency value (ε) has been obtained from the fitted curve of fig (3). The validated results are tabulated in table-2 and the calculated activity of the Cs-137 is found to have an uncertainty of 6%.

Table 2: Activity Values calculated from the fitted function for Cs-137 of IAEA 375 standard

IAEA Standards	Isotope	Energy (keV)	Theoretical Efficiency (%)	Calculated Activity (Bq/kg)	Uncertainty (%)
IAEA 375	^{137}Cs	661.6	1.1664	3055±152	6

MC Calculated FEP Efficiency

In an attempt to evaluate the FEP efficiency values of HPGe detector used for environmental low activity measurements in our laboratory, the detector has been modeled with MCNP code to validate the simulated response function for the energy range 200 keV to 2700 keV.

For the reference gamma standard sources, the graph (Fig 6) displays the comparisons between experimental full energy peak efficiencies and direct MC computed efficiencies of the detector. As seen in the graph, for the active volume of the crystal in the detector, the detector FEP efficiency is maximum while considering the cross-section of low energy photons [21]. On the other hand, as the energy increases along the abscissa, the values of the FEP efficiency calculated with MC falls and it follows the same trend as that of the experimentally calculated value.

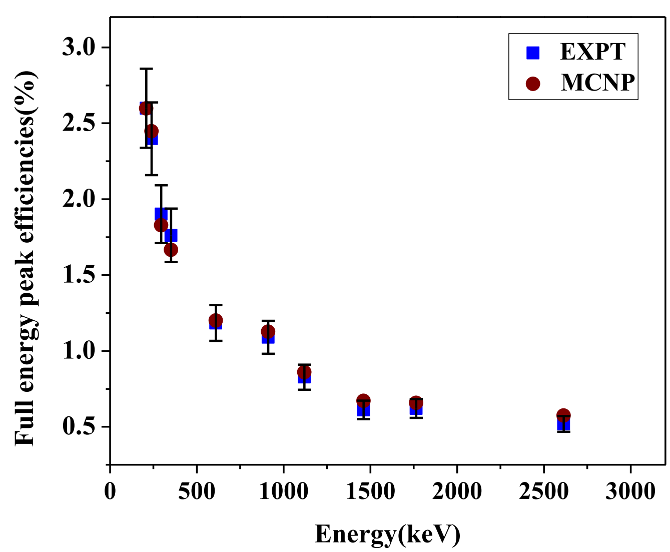


Figure 6: Measured and computed (MCNP) full energy photo-peak efficiency

The effectiveness of the optimized models can be assessed using the relative deviation. Fig 7 shows the relative deviation between the experimental FEP efficiencies and MC computed for the complete energy range. In the comparative graph, most of the deviations account for the uncertainties are closer to the uncertainties used in the calculations of the experimental FEP efficiencies. The results obtained by the simulations yielded maximum deviations not greater than ±10% with the estimated standard deviations less than 5% between simulation and of the measurement. This reveals a fair indication of the simulated detector model [22].

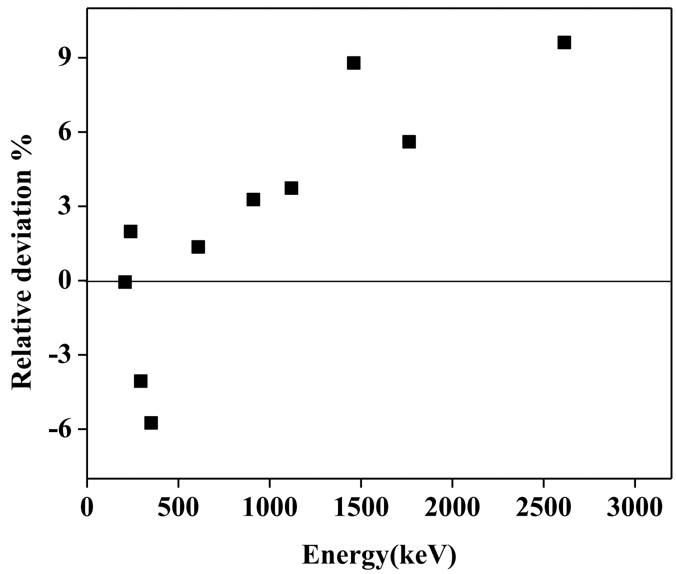


Figure 7: Relative deviation (%) between the MC calculated values and experimentally measured FEP efficiencies

A quantitative evaluation of the variation of the FEP efficiency is shown in Fig 8, considering the ratio between the MC

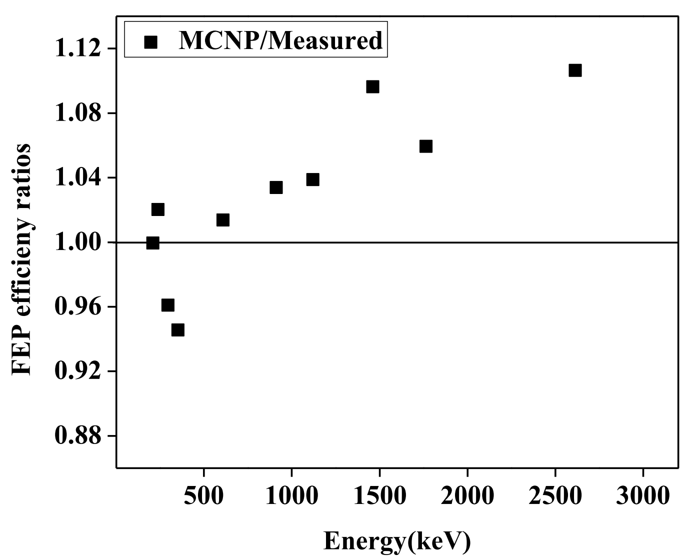


Figure 8: MCNP to Experimental Efficiency ratios Vs gamma energy values

and experimental FEP efficiency values against the gamma peak energy. Even though there is a fluctuation in the studied energy ranges, the average ratio for all the energies is found to be 1.01, which is closer to unity with an average uncertainty of less than 5%. This also indicates the implausible simulation of the detector model using MC code.

Conclusion

The method presented for the construction of the FEP efficiency curve enables limitless accuracy depending on the quality of the data acquired and the corrective curve employed. A third-order logarithmic power series closely related to five-parameters appears to be adequate to represent all the experimental data and be able to hand round to define FEP efficiency curve for gamma spectrometry method. The key benefit is that a single calibration curve employing a continuous non-linear energy dependent function covers the entire energies and the function fitting parameters are calculated by straightforward mathematical functions. The results obtained based on experimental and theoretical efficiency seems to be in good agreement with a maximum deviation of less than ±6% with an expanded uncertainty of corresponding to a confidence level of 95%. This new fit function could undoubtedly used to compute the absolute FEP efficiency for any unknown gamma energy with the cited experimental set-up for environmental investigations. The integrity of MC simulation for calculating the FEP efficiency of germanium detector has been confirmed by comparing simulating results to experimental measurements over a range of energies. There exists a disagreement in the MC calculated efficiencies than the experimental efficiencies and is merely due to the changes in the detector geometrical parameters. When the geometrical parameters like dead layer thickness has been incorporated in the modelling, the observed variations are comparable with the estimated standard deviations of measurement and of modelling that range less than 5% except for slightly larger uncertainties at higher energies. It has been found out the average FEP efficiency ratio of the simulation results to measurements for 10 photon energies between 200 keV to 2700 keV is 1.01 with the ratios at individual energies ranging between 0.94 and 1.09 for the volume sources under consideration. This work emphasize that the theoretical method for obtaining FEP efficiency of a long – run HPGe detector does not require any modifications with reference to experimental determinations, but the results obtained by the MC methodologies requires suitable amendments for the said measurement geometry thereby aiding for the faster analysis of low activity environmental soil samples.

Acknowledgements

The authors thank the colleagues of RESG, SQRMG for their support.

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