STEPS TOWARD OPTIMIZING HPGe DETECTOR EFFICIENCY IN THE CONTEXT OF LOW-LEVEL RADIOACTIVITY DETECTION

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Abstract. Different empirical and theoretical methods are used to precisely determine radionuclide activity concentrations. This study used Monte Carlo simulation MCNP5 code and EFFTRAN software (Efficiency Transfer) to determine the HPGe detector Full-Energy Peak (FEP) efficiency. A set of point sources (^{133}Ba, ^{152}Eu, ^{137}Cs, ^{60}Co, and ^{22}Na) fixed on the top of the standard geometry plastic container were measured in order to obtain the calibration curve. Because of the importance of chemical composition parameters, the detector behavior due to different matrices was investigated. Experimental verification of the calibration was obtained using IAEA-TEL-2021-03 quality control water sample spiked with ^{152}Eu and ^{133}Ba, and the results were compared with reported results. A set of Certified Standard Reference samples were used for method validation. The obtained results were compared with the experimental results. The comparison clarified the advantages and disadvantages of both methods and their precision to demonstrate and suitable method for matrix types.

Key words: radioactivity, Gamma spectrometry, HPGe efficiency calibration, EFFTRAN, Monte Carlo.

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1. INTRODUCTION

For natural and man-made radionuclides identification in environmental monitoring, High Purity Germanium (HPGe) detector is used because of its high energy resolution [1]. Some parameters are required to get high accuracy and high precision results and the Full Energy Peak Efficiency (FEPE) is a crucial measure [2]. It is possible to compute the relative, semi-absolute, or absolute efficiency of the FEPE calibration. Despite being the most accurate, the relative approach needs samples with similar matrices (physical and chemical). Several semi-empirical techniques have also been developed for FEPE determination. The detector

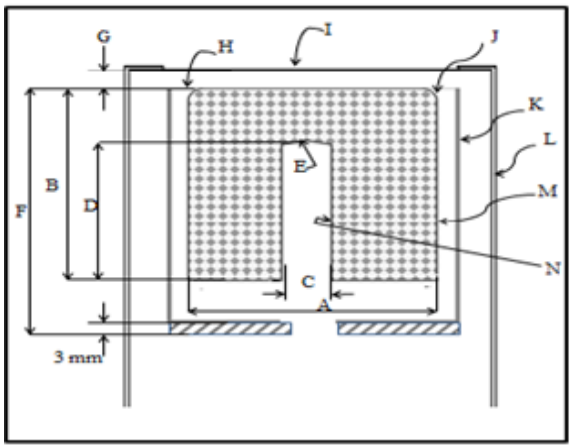
configuration and the properties of the tested samples affect how precisely FEPE is calibrated. Due to the lack of comparable standards, Monte Carlo (MC) simulations, such as MCNP, could be used to perform an accurate calibration process [3–5]. Monte Carlo simulation detectors were widely used as numerical alternatives to empirical and experimental methods [6]. It is occasionally used to generate the spectra and extract the desired efficiency [7]. There are many software packages dedicated to determine the FEPE, such as Angle [8, 9], Lab-SOCS [10], ETNA [11], EFFTRAN [12], MEFFTRAN [13], GESPECOR [14], GEANT4 [15].

In this study, the FEPE of the HPGe detector was determined using both MCNP-5 simulation and the efficiency transfer software EFFTRAN. Results were compared with the experimental results. The detector characteristics were presented for simulations. The accuracy and precision of both methods were also evaluated.

2. MATERIALS AND METHODS

2.1. DETECTOR SPECIFICATIONS

The HPGe (N-type) gamma-spectrometer was shielded by four mm Pb, 1 mm Cd, and 1 mm Cu and has 30% relative efficiency. The detector performance specifications are FWHM resolution of 1.9 keV both at 1.33 MeV ^{60}Co, and the peak-to-Compton ratio for ^{60}Co is 52:1. The dead layer is equal to 5 mm. The applied bias voltage is 3500 V. In this study, a pre-amplifier (Model A257N) and the Genie-2000 software make up the data-gathering system. The supplier has provided manufacturer's data for detector components are shown in Fig. 1.



A- Crystal Diameter	55.8 mm
B- Crystal Length	68.5 mm
C- Hole Diameter	9.1 mm
D- Hole Depth	59.8 mm
G- Space	4 mm space
I -Be	0.5 mm
K - Al	0.8 mm
L -Al	1mm
M- Ge/B DEAD LAYER	0.3 micron
N- Ge/Li DEAD LAYER	700 micron

Fig. 1 – The HPGe detector parameters given by the manufacturer.

2.2. EXPERIMENTAL EFFICIENCY CALIBRATION

A set of calibration sources (^{133}Ba, ^{152}Eu, ^{137}Cs, ^{60}Co, and ^{22}Na) with known activity were measured to determine the detector's relative efficiency using the following equation:

$$\varepsilon(E) = \frac{N}{A\ P\ t\ W} \tag{1}$$

where: N is the number of net counts in each energy line,

t – the counting time,

P is the emission probability at a given energy E,

A is the activity of each certified point source, and

W is the weight of the sample.

The efficiency calibration curve was generated and normalized to an absolute efficiency curve for a certain geometrical configuration (cylindrical container with 100cc) to cover gamma energies ranging from 121.78 keV to 1764.0 keV.

The Minimum Detectable Activity (MDA) calculations are based on the following equation:

$$\mathrm{MDA}\left[\frac{\mathrm{Bq}}{\mathrm{kg}}\right] = \frac{2.71+4.66\sqrt{B}}{T_S\ \varepsilon\ I_\gamma\ W} \tag{2}$$

where: B denotes the background counts, T_S is the counting time expressed in seconds, I_γ denotes the gamma emission probability, and ε is the detector's absolute efficiency at the given gamma energy.

The relative combined uncertainty measurement was calculated according to the equation:

$$\mathrm{u}(\varepsilon) = \sqrt{(\delta\mathrm{A})^2 + (\delta\mathrm{N})^2 + (\delta\mathrm{C(E)})^2} \tag{3}$$

where: δA represents the relative uncertainty of the total activity in the standard,

δN is the relative counting uncertainty, and

$\delta C(E)$ is the relative uncertainty introduced *via* coincidence correction factors.

The amount of error associated with the measurement of time t and mass of the standard, m, was extremely small. At a level of confidence of 2 s, the relative total measurement uncertainty, denoted by $u(\varepsilon)$, did not exceed 5% for any of the energies [16, 17].

2.3. MATHEMATICAL EFFICIENCY CALIBRATION

I. MCNP-5 CODE

MCNP-5 code was used to simulate and create a model for the HPGe detector taking into consideration the parameters provided by the manufacturer. Detector specification, such as the detector-dimensions, its Al-cap, Al-holder and the distance from the detector crystal to the front of a detector cap, was used to construct MCNP-5 input file. The sample was prepared in plastic container with the 3.2 cm radius, 0.2 cm base thickness and the sample height 2.5 cm. The output from the simulation is called tally. Pulse height distribution tallies are defined by tally type (given a number 1, 2, 4, 5, 6, 7, 8). The pulse height tally F8 with corresponding energy bin card E8 was used to estimate detector efficiency at gamma lines with energies. The number of histories for each setup was defined as $N = 1 \times 10^7$ (run 4 min for each energy peak) and the number of simulated histories was determined so as to keep the uncertainties in Monte Carlo calculations always better than 0.2% [3]. An MC input file was constructed for the response function and parameter characterization. Simulations were carried out for a number of different discrete values within the range of 121.78 to 1764 keV. Because the photons enter the crystal through its top face, the attenuation of photons caused by the aluminum layer that surrounds the detector laterally was not taken into consideration. The FEPE was verified using the Reference Standard Water Sample IAEA-TEL-2021-03-qc1.

Figure 2 shows the characteristics of the simulated HPGe detector drawn by MCNP-5 for standard geometrical samples.

II. EFFTRAN

The Belgian Nuclear Research Centre created the efficiency transfer program EFFTRAN to determine efficiency transfer for cylindrical samples [12]. On the basis of the presumption that the calibration source would be measured with known activity, it is utilized to calculate the detector efficiency for measuring samples [18]. Some factors must be taken into consideration in order to determine the detector efficiency, including the activity of the calibration source, the measured sample (diameter, filling height, matrix composition, container thickness, etc.), and the geometrical characteristics of the detector crystal (crystal, housing, and composition, active and inactive layers, etc.) [19, 20].

In EFFTRAN, this is accomplished by characterizing the detector, the standard, and the sample geometry (geometry of the calibration), with the measurement of the standard providing the input of the efficiencies at various gamma-ray energies; the efficiencies for the sample geometry are determined as output from EFFTRAN.

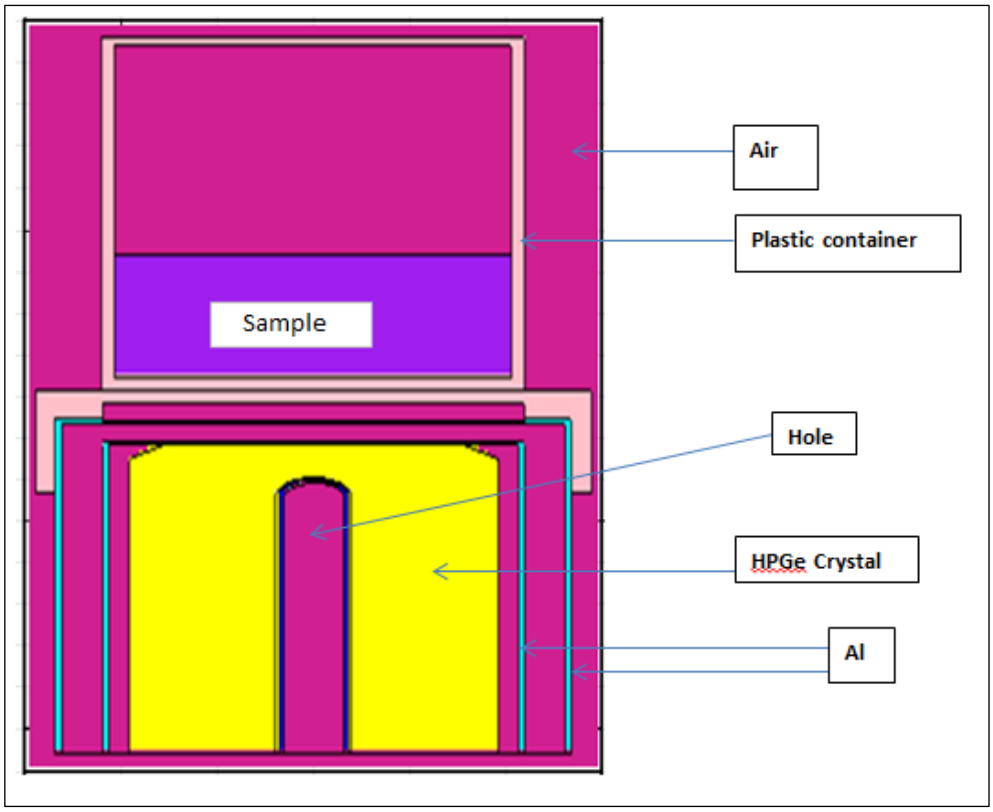


Fig. 2 – MCNP-5 simulated model for HPGe detector.

III. METHOD VERIFICATION AND VALIDATION

For precise determination of the detector efficiency, the calibration was verified using Certified Reference Materials provided by the International Atomic Energy Agency (IAEA-TEL-2021-03). Through the participation of the CLERMIT laboratory in Proficiency Tests (PT) organized annually by IAEA, as a member of the ALMERA network (Analytical Laboratories for the Measurement of Environmental Radioactivity), the methods were validated using several IAEA Certified Reference Materials: IAEA-TEL2018-S04-Soil, IAEA-TEL-2017-04–Milk powder, IAEA-TEL-2021-05-Water, IAEA-RGU-1-Ore, and IAEA-373-Grass.

The precision *P* of the methods is calculated for each radionuclide according to the following formula:

$$P = \sqrt{\left(\frac{\text{Unc IAEA reported}}{\text{Value IAEA reported}}\right)^2 + \left(\frac{\text{Unc target}}{\text{Value target}}\right)^2} \times 100\% \qquad (4)$$

based on the uncertainties (Unc) and the results (Value) found in the IAEA approved certificate and calculated for each target nuclide. The *P* value is compared to the values of Maximum Acceptable Relative Bias (MARB) presented in Table 1 [21]; if $P \leq$ MARB, the reported results will be "Accepted" for precision.

Table 1

Reference Certified Activity Concentration and Maximum Acceptable Relative Bias (MARB)

IAEA-TEL-2021-05 WATER			
Radionuclide	Activity corrected to 14/12/2021 [Bq/kg]	Uncertainty [Bq/kg]	MARB [%]
Cs-134	14.04	0.63	30
Cs-137	25.48	1.14	25
IAEA-TEL-2017-04 MILK POWDER			
Radionuclide	Activity corrected to 24/01/2022 [Bq/kg]	Uncertainty [Bq/kg]	MARB [%]
Ba-133	97.85	5	15
Cs-137	87.569	5	15
IAEA-TEL2018-S04-SOIL SAMPLE			
Radionuclide	Activity corrected to 5/5/2021 [Bq/kg]	Uncertainty [Bq/kg]	MARB [%]
U-238 series	25	1.7	20
K-40	374	15	20
Cs-134	37.83	1.6	20
Cs-137	60.23	1.2	20
Co-60	92.62	2.7	20
Ba-133	45.9	0.9	20

3. RESULTS AND DISCUSSION

The gamma spectra of the standard point sources (^{133}Ba, ^{137}Cs, ^{22}Na, ^{60}Co, and ^{152}Eu) were analyzed, and the total count for each gamma energy considered was determined. The experimental relative photo peak efficiency curve was plotted, and a relative efficiency curve was normalized to the absolute volume efficiency curve using a KCl-solution [22].

The efficiency curves for experimental and simulated empirical results based on MCNP-5 and EFFTRAN have been shown in Fig. 3. Certain deviations were noticed between the results obtained by empirical methods (MCNP-5 and EFFTRAN) and the experimental method. The difference between the experimental-efficiency

and the empirical-method based on MCNP-5 (Exp/MCNP-5) was 0.88%, while the difference between experimental efficiency and the empirical method based on EFFTRAN transfer programs (Exp/ EFFTRAN) was 0.94%. From the data obtained, some insufficient parameters affect the systematic change in the detector efficiency. First, the simulated efficiencies were performed based on point source geometry with the detector dimensions. Secondly, the detector geometry (radius and length), dead layer thickness and chemical composition, and inhomogeneity of the matrix materials (composition for reference materials) may significantly affect the detector efficiency.

The experimental errors have been determined by taking into account the counting statistics, the detection efficiency, and the uncertainties in the certified activities – the uncertainty components combined according to ordinary error propagation law.

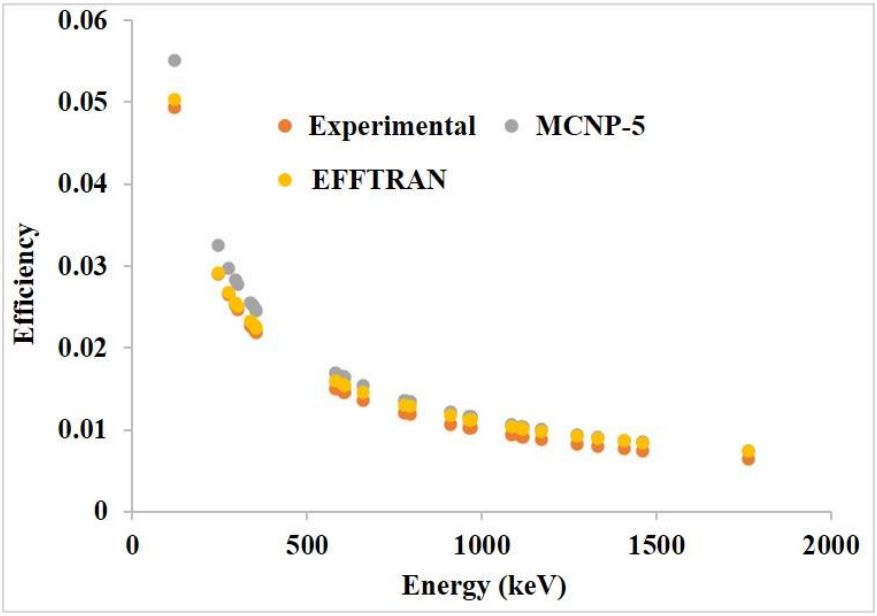


Fig. 3 – The experimental and simulated efficiency based on MCNP-5 simulation and EFFTRAN.

For method verification, IAEA-TEL-2021-03-qc1 water sample with certified activities was measured many times (5 times repetition), then the mean activity concentrations were determined for ^{133}Ba and ^{152}Eu as obtained in Table 2 and Fig. 4. The results obtained by MCNP-5 agree well with that given in the certificate in the whole energy range. This may be attributed to a more detailed description of the detector, surrounding materials, sample matrix, and the container used. However, in EFFTRAN, the main dimensions of the container and source are used only.

Table 2

Results of water sample (IAEA-TEL-2021-03) using experimental and empirical (MCNP-5 and EFFTRAN) methods

Nuclide	Activity concentration (Bq/kg)			
	Reference Activity	Experimental Activity	MCNP-5 Activity	EFFTRAN Activity
Ba-133	55.145±0.4	56.5±2.8	55.2±2.7	56.4±2.8
Eu-152	110.22±0.3.5	111.3±5.6	110.6±5.5	111.2±5.5

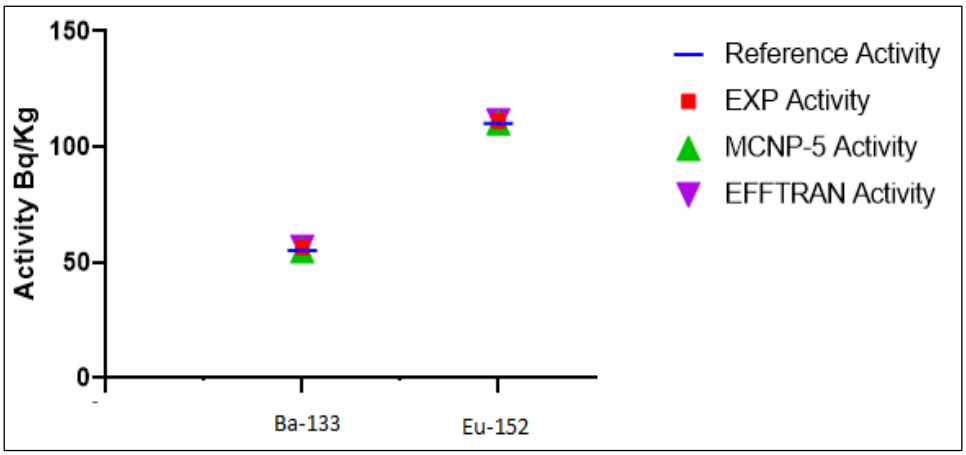


Fig. 4 – The experimental and simulated efficiency based on MCNP-5 simulation and EFFTRAN.

3.1. METHOD VALIDATION

Using the efficiency values that were obtained, the activity concentrations of several certified reference samples that contained a variety of matrices were determined. The IAEA's evaluation for the proficiency tests (PT) allowed for a relative bias of up to%MARB for each of the individual analytes that were specified by the provider of the PT. A warning is given to the result that was reported [13] if the total combined relative uncertainty of a certified and reported activity is higher than the%MARB threshold (Table 3). The outcomes of this study demonstrated that the validity of the efficiency estimations achieved using EFFTRAN and MCNP-5 software. The majority of the results agree within around 10% of the uncertainties, which is satisfactory for the measurements of the environmental samples (Table 3).

Table 3

Results of some certified reference samples using experimental (EXP.) and empirical (MCNP-5 and EFFTRAN) methods

Radio-nuclide	Activity (Bq/kg), IAEA-TEL-2021-05 WATER						
	Certified activity corrected to 14/12/2021	EXP. Activity	P%	MCNP-5 Activity	P%	EFFTRAN Activity	P%
Cs-134	14.04±0.63	13.6±0.67	6.66	14.38±0.7	6.620	14.05±0.7	6.705
Cs-137	25.48±1.14	25.6±1.28	6.709	25.8±1.129	6.223	25.7±1.28	6.695
	Activity (Bq/kg), IAEA-TEL-2017-04 MILK POWDER						
	Activity corrected to 24/01/2022	EXP. Activity	P%	MCNP-5 Activity	P%	EFFTRAN Activity	P%
Ba-133	97.85±5	97.9±4.89	6.179	96.9±4.8	6.146	97.7±4.9	6.196464
Cs-137	87.569±5	87.6±4.4	7.137	88.3±4.4	7.109	86.3±4.3	7.109252
	Activity (Bq/kg), IAEA-326-SOIL						
	Reference Activity	EXP. Activity	P%	MCNP-5 Activity	P%	EFFTRAN Activity	P%
U-238 series	32.6±2.1	31.3±1.5	8.029	32.7±1.6	8.09	33±1.6	8.06
Th-232 series	39.4±1.8	39.8±2	6.79	40.6±2.05	6.92	40.8±2.1	6.88
K-40	580±9	588.9±29.5	5.24	576±28.8	5.23	572.3±28.6	5.23
	Activity (Bq/kg), IAEA-RGU –ORE						
	Reference Activity	EXP. Activity	P%	MCNP-5 Activity	P%	EFFTRAN Activity	P%
U-238 series	4940±30	5008.2±250	5.028	4833±241.6	5.03	4924.4±246.2	5.036
	Activity (Bq/kg), IAEA-373 –GRASS						
	Reference Activity	EXP. Activity	P%	MCNP-5 Activity	P%	EFFTRAN Activity	P%
Cs-137	6171.9±110	6235.6±312	5.312	5.3115	5.302	6144.8±307	5.305
K-40	432±11	440±22.6	5.733	5.733	6.223	428.4±21.4	5.61

4. CONCLUSIONS

This study aims to calculate the efficiencies of HPGe detectors utilizing MCNP-5 simulation and EFFTRAN efficiency transfer software. The results that were acquired contrasted with the outcomes of the experiment. Both the MCNP-5 simulation and the transfer of efficiency from a point source to a bulk source using different matrices of our measuring system were successful. Both the definition of the detector geometry and the chemical make-up of the samples are likely to be

considered the primary contributors to the disparities. Because of the reliability and exactness of both approaches, we will be able to choose and select the approach that is most suited to the various circumstances that arise on a regular basis. When performing calculations on the efficiency of the transfer, calibration sources that have a matrix that is comparable to that of the real samples should be utilized as a starting point. Additionally, in order to arrive at a computation of the specific activity of radionuclides in samples using gamma spectrometry that is as exact as possible, it is required to take into consideration two key factors. These characteristics have a direct impact on the actual coincidences that occur and the amount of gamma radiation that is absorbed by the tested sample.

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