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Monte Carlo method for gamma spectrometry based on GEANT4 toolkit: Efficiency calibration of BE6530 detector

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ABSTRACT

The combination of gamma-ray spectrometry, the development of related Monte Carlo method and the GEANT4 (GEometry ANd Tracking) toolkit have been developed for gamma spectrometry simulation. The main objective was to validate simulation models of broad energy germanium (BEGe) detector geometry built in our laboratory (BE6530 model). Monte Carlo simulation of the geometry of BE6530 detector for efficiency calibration was carried out with GEANT4 toolkit. The simulated efficiencies curves using MC were compared with experimental results. Measurement uncertainties for both simulation and experimental estimations of the efficiency were assessed in order to see whether the consequences of the realistic measurement fall inside adequate cut-off points. The validation of the simulation was carried out by experimentally estimating the activity concentration in a reference sample and the comparison showed good correlation between experimental and simulation. Therefore, from the outcomes of this study, it can be concluded that Monte-Carlo simulation is a helpful, reasonable option that additionally gives more prominent adaptability, greater flexibility, precision and accuracy, and gained time when determining the detector response and efficiency in routine of environmental radioactivity monitoring.

1. Introduction

The Monte Carlo (MC) numerical methods are the simulation statistical methods where the statistical simulation represents any method which utilises the sequences of aleatory numbers to realize the simulation (Hranitzky and Stadtmann, 2007; Mora et al., 1999; Mainegra-Hing et al., 2003). Nowadays, the Monte Carlo simulation is used in different fields of activity from the simulation of some complex physical phenomena such as the radiation transport in the earthly atmosphere and the subnuclear processes which occur at high energy to the simulation of a video game. A Monte Carlo calculation thus comprises of running an extensive number of particle occasions until some worthy statistical uncertainty of the coveted computed amount has been reached. Gamma-ray spectrometry is a non-destructive technique used for quantitative and qualitative analysis of gamma-ray emitter radionuclide in the sample. This analysis is performed through calibration of the detector, counting and measuring the individual photo-peak emitted from different elements present in the sample (Guembou et al., 2017a, 2017b). Simulation based on GEANT4 toolkit provides more information on the new technical of gamma-ray spectrometry. Photons can interact by means of the photoelectric effect, Compton scattering, pair production or Rayleigh scattering, and the probability of each interaction type is energy dependent as describe by many authors such as Baro et al. Biggs and Lighthill, Born, Butcher and Messel, Ford and Nelson, Gavrila, Grichine et al. Heitler, Hubbell et al. Kenneth, Messel and Crawford, Rossi, Berger (Baro et al., 1994; Biggs and Lighthill, 1990; Born, 1969; Butcher and Messel, 1960; Ford and Nelson, 1985; Gavrila, 1959; Grichine et al., 1994; Heitler, 1957; Hubbell et al., 1980; Hubbelet al, 1975; Kenneth Krane, 1987; Messel and Crawford, 1970; Penelope -ode System, 2001; Rossi, 1952; Berger et al., 2010).

To describe the behavior of the system, the physical processes were directly simulated with one condition: the system must be described by the probability density function. The Monte Carlo simulation can continue through the generation of aleatory values from the probability density function if this function is known. In Gamma spectrometry, Monte Carlo simulation is considered as a new method to assess

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environmental radioactivity because the results of this analysis is known as an average of the observed numbers (Mainegra-Hing et al., 2003; Wang, 1996; ISO, 1999). In addition, this method limits the high cost and risk related to the utilization of radioactive sources for calibration of the detectors and is an important task nowadays that favours the development of MC methods. When succeeded, the geometry constructed by MC simulation can be very helpful in the case of efficiency calibration of HPGe. Studies performed worldwide have proved the validation of this method. As a result, many laboratories found that computed values obtained with MC simulation significantly deviated to the experimental values. This interest results from the difficulties encountered during experimental calibration of the detectors.

Many authors used MCNP and MCNPX in the past decades for calculation of detection efficiency of the gamma detector. Akkurt presented results of MCNPX simulation obtained for NaI(Tl) detector system and compared them with the experimental results (Akkurt et al., 2015). A good agreement was found between calculation and experiment. In addition, simulation of high purity germanium detector efficiency as well as geometry based with GEANT4 code conducted by some authors found this method very useful alternative that also provides greater flexibility when determining the detector response and efficiency during a measurement (Akkurt et al., 2015; Breier et al., 2017; Britton et al., 2012, 2013a, 2013b, 2014a; Chagren et al., 2017; Crespi et al., 2008; Li et al., 2015; Hurtadoa et al., 2004a; Hansman et al., 2015; Nikolic et al., 2014a, 2014b, 2015; McNamara et al., 2012; Dababneh et al., 2014; Saraiva et al., 2016; Sivers et al., 2014). Britton (Britton et al., 2013a) developed a full GEANT4 model for the simulation of a broad-energy germanium detector. The results of simulation as compared with the experiment were found to be 3% accurate with confidence level of 95% for energies between 30.00 and 3000.00 keV. Gang Li performed study on efficiency corrections in determining the ^{137}Cs inventory of environmental soil samples by using relative measurement method and GEANT4 simulations (Li et al., 2015). Hurtadoa used GEANT4 toolkit in order to simulate Ge detectors in low-level gamma spectrometry and the comparison with experimental value were found to be within 10% confidence limit (Hurtadoa et al., 2004a). Chagren applied the GEANT4 code of CERN to calculate the peak efficiency in High Pure Germanium (HPGe) gamma spectrometry (Chagren et al., 2017). Jelena Nikolic performed research on the calibration of three HPGe detectors (Closed coaxial - p type Canberra with Al entry window, closed reversible coaxial - n type Canberra with Be entry window and Closed coaxial - p type Canberra with Be entry window) using GEANT4 toolkit and EFFTRAN software for efficiency transfer (Nikolic et al., 2014a). Saed Dababneh utilized GEANT4 to accomplish crucial corrections, in close geometry, for self-absorption and true coincidence summing in gamma-ray spectrometry of environmental samples (Dababneh et al., 2014). Nikolic applied GEANT4 simulation to calibrate two HPGe detectors, to measure liquid and soil-like samples in barrel shaped geometry (Nikolic et al., 2015). These researchers have proved the accuracy and the precision of the GEANT4 toolkit in the calibration of the gamma-ray spectrometry detectors because the overall comparison between simulation and experiment were in the majority in accord of 10% limit. The difference is partially due to different detector parameters that vary date to date and can be more optimise during simulation than experimentation. Therefore, more attention has to be given on the simulation of the different detectors model to improve scientific collaboration between different teams' work, friability of the results and also facilitate the implication on researchers in the countries under development to perform studies on environmental radioactivity without any cost of buying multi-gamma radioactive sources for calibration. None or very few attentions have been given on the simulation of broad energy germanium detector, especially the BE6530 model. Nevertheless, LabSOCS and ISOCS calibration software have been developed based on MC algorithm, to facilitate the calibration and these can only be done on characterised detector (Mirion Technology, 2013).

The aim of the present study is to apply GEANT4 MC Simulation for the efficiency calibration of BEGe 6530 gamma detector model. The intention was to improve the calibration process of Broad Energy Germanium detector, for measurement of environmental samples in cylindrical geometry. In addition, a full GEANT4 model of a broad-energy BEGe detector was presented with different views of the geometry construction. The efficiency obtained with GEANT4 was compared with experimental results. Comparison between experimental and simulated values was estimated, and the objective was to see whether the results obtained in the present study falls in agreement. The demonstration of the capability and robustness of a GEANT4 model on predicting detection efficiencies for the broad-energy germanium spectrometer (BEGe 6530) used in our laboratory was highlighted.

2. Methods and computation

2.1. Overview of Monte Carlo method

Monte Carlo techniques are computational calculation method in view of random number to get results. In the 1940s, Monte Carlo technique was initially presented for the nuclear weapon extend, Manhattan Project. Nowadays, Monte Carlo algorithms are broadly used as a part of many fields. With the creating of computer techniques, Monte Carlo (MC) techniques turn out to be increasingly well-known in many fields of uses to figure or solve issues which can be not really comprehended with classic analytic techniques. For a complete record of the MC techniques, the publications of Butcher and Messel, Messel and Crawford, or Ford and Nelson can be consulted (Biggs and Lighthill, 1990; Ford and Nelson, 1985; Messel and Crawford, 1970). In the event that we wish to sample x in the interval $[x_1, x_2]$ from the appropriation $f(x)$ and the normalised probability density function can be expressed as:

$$f(x) = \sum_{i=1}^{n} N_i f_i(x) g_i(x) \tag{1}$$

where $N_i > 0$, $f_i(x)$ are normalised density functions on $[x_1, x_2]$, and $0 \leq g_i(x) \leq 1$.

Most numerical packages with the capacity to include random number generation are also able to generate random numbers with other probability distributions, for example, Poisson or Gaussian distributions. The probability distribution function $p(x)$ is considered in this situation, with which the goal is to produce random numbers which take over this distribution function. The Geant4 code used in this study was built in accord with the interaction of gamma rays with matter based on cross section of Photoelectric effect, Compton backscattering, pair production and Rayleigh effect. Many authors, such as Biggs, Born, Butcher, Ford, Gavrila, Grichine, Heitle and Hubbell, developed the process of the passage of photon through matter (Hranitzky and Stadtmann, 2007; Mora et al., 1999; Baro et al., 1994; Biggs and Lighthill, 1990; Born, 1969; Butcher and Messel, 1960; Ford and Nelson, 1985; Gavrila, 1959; Grichine et al., 1994; Heitler, 1957). The probability of non-interaction of a photon is:

$$\mathrm{p(x)} = \mu.\mathrm{e}^{-\mu x} \tag{2}$$

2.2. Experimental

High resolution gamma-ray spectrometry is an effective method to determine activity concentration of radionuclides in the samples (Barros and Pecequilo, 2014; Conti Salinaset al, 2013). The detector under investigation is a commercial p-type BEGe detector (BE6530 Model) produced by Canberra. Major descriptions and specifications of the detector used are presented in Table 1. A schematic view of the detector configuration is shown in Fig. 1. The data acquisition systems in this work involve a charge-sensitive pre-amplifier, an integrated digital signal analyser and the Genie-2000 software versus 3.2.

Table 1
Specifications of HPGe detector used in the laboratory for experimental study (BE6530).

Descriptions	Detector
Detector type (Canberra)	BE6530
Detector geometry	Plan
Detector active area-facing window (mm^2)	6500
Active diameter (mm)	91.5
Thickness (mm)	31.5
Distance from window (outside) (mm)	5.0
Window thickness (mm)	0.6
Aluminium endcap distance from window (mm)	8.0
Window material	Carbon epoxy
Relative efficiency at 1332.5 of ^{60}Co (%)	60
Full Width Half Maximum (FWHM) Resolution (keV) at 5.9 KeV	0.478
Full Width Half Maximum (FWHM) Resolution (keV) at 122 KeV	0.695
Full Width Half Maximum (FWHM) Resolution (keV) at 1332.5 KeV	1.785
Depletion voltage	(+)4000
Recommended bias voltage Vdc	(+)4500
Time constant (μs)	4
Cryostat description	Vertical dipstick
Peak shape (FWTM/FWHM) for ^{60}Co	1.88
Cooling system	Electric
Recommended bias voltage	+4500 V

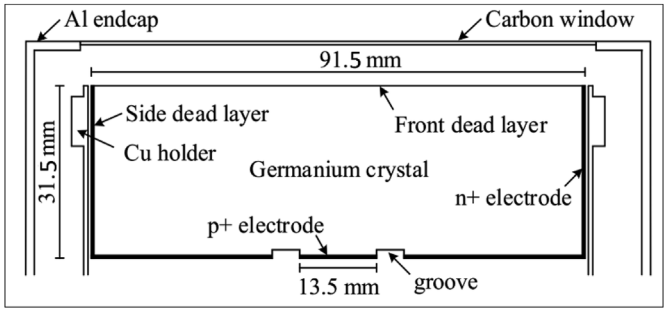

Fig. 1. Schematic view of the BEGe detector (BE6530) with real dimension.

The detector utilises a carbon epoxy window to ensure the protection of the crystal of the detector and to limit the attenuation of low energy photons. This decreases the response to photons below 10 keV when contrasted with an Aluminium window (which lessens the response to photons under 30 keV) or a Beryllium window (which can allow photons of energy ≥ 2 keV), that is significantly more robust. Other composers of detector protection are Aluminium endcap, lithium dead layer, lead shielding, boron layer and copper used in this case to reduce background and to reduce the effect of X-rays emitted by the lead shielding system.

The knowledge of the detection geometry is essential in the subsequent calculations (Hranitzky and Stadtmann, 2007; Grichine et al., 1994). For this reason, some information was available only on a special request to the Canberra if it has an influence on the simulation result. For example, real dimension and geometry of the germanium crystal can be acquired by the collaborating with Canberra that provide more specific details or through the image of the detector obtained by a scanner. Experimentally, the efficiency ε for a given photon energy is assessed throughout the following formula:

$$\varepsilon = \frac{N_c}{A\ t\ P_\gamma}\ C_i \tag{3}$$

Where N_c is the number of total counts in the peak, T is the detection time, A is the nuclide activity and P is the photon emission. C_i contains correction factors due to dead time, radionuclide decay and coincidence summing corrections. Calibration of equipment was done for both energy and efficiency. The uncertainty of the efficiency calculation (Δε) was calculated using the following equation:

$$\frac{\Delta\varepsilon}{\varepsilon} = \sqrt{\left(\frac{\Delta N}{N}\right)^2 + \left(\frac{\Delta P_\gamma}{P_\gamma}\right)^2 + \left(\frac{\Delta A}{A}\right)^2 + \left(\frac{\Delta M}{M}\right)^2} \tag{4}$$

Where ΔN is the count rate uncertainty, ΔP_γ the emission probability uncertainty found in the nuclear data tables, ΔA the relative uncertainty of the radioactive source provided by the manufacturer and ΔM the weighing uncertainty, introduced in the process of secondary standard material production. Point sources of ^{241}Am, ^{152}Eu (for energy calibration), ^{137}Cs, ^{109}Cd, ^{88}Y, ^{65}Zn, ^{60}Co, ^{57}Co and ^{54}Mn were used. To record at least the highest counts for each full-energy peak and minimize the statistical counting error, the counting time of gamma point sources were adjusted.

Validation of the experimental efficiency calibration was done through technical cooperation with the Canberra laboratory. The calibration efficiency curve was plotted in dual mode with cross-over energy to avoid error due to extrapolation. The influence of the decay scheme of nuclides, the sample geometry and composition, the detector features on the specific activity of radionuclides in the sample, and the cascade summing effect was automatically corrected using the peak to total (P/T) curve (Ababneh and Eyadeh, 2015).

2.3. GEANT4 toolkit for Monte Carlo simulation

Monte Carlo techniques are computational calculations in view of random sampling to get results. In the 1940s, Monte Carlo technique was initially presented for the nuclear weapon and used firstly in the Manhattan Project (Badash, 2011). The use of Monte Carlo method in the field of Nuclear applications was completely presented by several scientists (Butcher and Messel, 1960; Ford and Nelson, 1985; Messel and Crawford, 1970; Berger, 1963). GEANT4, a publicly available Monte-Carlo toolkit developed at CERN, was used to perform accurate simulations of particle propagation through an interaction with matter (Agostinelli et al., 2003; Geant4 Collaboration, 2016; Allison et al., 2016). Unlike the other computational Monte Carlo codes such as EGS, MCNP, MCNPX, FLUKA, GATE, PENELOPE, SRIM … etc, GEANT4 is not an executable program but rather a set of C++ class libraries for running, tracking, stepping and collecting information … etc.

Information about each particle can be obtained at both the pre-step and post-step points, including the energy deposited per step, the type of particle, number of secondary's, position, trajectory etc. By combining the information across all steps, information for each event can be obtained, and therefore used to create useful outputs, such as energy spectra. GEANT4 provides a visualisation system, which is based on graphics libraries, such as OpenGL and Qt and some other plotting applications, such as HepRApp or DAWN (Geant4 Collaboration, 2016; Allison et al., 2016; Zhang, 2011). G4UserRunAction; G4UserEventAction; G4UserStackingAction; G4UserTrackingAction; G4UserSteppingAction … etc. Were used when editing the code (Agostinelli et al., 2003; Geant4 Collaboration, 2016; Allison et al., 2016; Zhang, 2011; Britton, 2014). All these classes must be well known when using GEANT4 for geometries and efficiencies simulation. GEANT4 was implemented in respect to gamma-ray spectrometry requirements to build our code for simulation of BEGe 6530 efficiency.

2.4. Simulation

The GEANT4 simulation starts with a cycle that begins with the generation of a gamma-beam photon from the source (the source can be the standard source provided by IAEA or produced in our laboratory) and after that, these are trailed by the tracking of photons in different areas of modelled geometry. The tracking of a solitary photon is ceased when it leaves the volume of interest also called the active volume of the detector or when the vitality of photon progresses toward becoming

lower than predetermined limit esteem cut-off energy. This cycle, additionally called sampling, is repeated many times keeping in mind the end goal to diminish the measurable fluctuation in the parameters of interest beneath than recommended limits. In our sampling, inspecting of more than 10^9 photons was completed for every information point for good statistical measurements. To enhance the productivity of reproduction, source biasing procedure was utilized. Modelled geometry had been built taking into account the real parameters provided by Canberra in the technical manual of the broad energy germanium detector.

The version 10.2 of the GEANT4 (Geometry and Tracking fourth generation) Monte Carlo code, released in December 2015 was used in the simulations built in this work (Agostinelli et al., 2003; Geant4 Collaboration, 2016; Allison et al., 2016). It offers a broad selection of approved physical models to reproduce the interaction of radiation with matter and contains a comprehensive dataset of material properties (such as density, isotopic composition, photon-electron stopping power). The description of interaction of gamma-rays through the matter is very important during simulation. GEANT4 provide different classes of physics list and the author have to select and implement his own class. The Low-energy Electromagnetic Package of GEANT4 was selected for performing simulation in this work (Lechner et al., 2009). All types of relevant interactions of photons and electrons/positrons with matter are considered, utilising low-energy data packages available in GEANT4 database (dataset G4EMLOW6, model G4EmLivermorePhysics); inside this package, two optional physics, the “Livermore” model and the “Penelope” model, were implemented. The intention was to adapt the code to the environment of our laboratory. G4AnalysisManager was used to create and fill histogram (the corresponding spectrum) by defining a 1D histogram for the energy deposited per event for the sensitive detector. The histogram plotting was activated using UI commands available in GEANT4 guide (Geant4 Collaboration, 2016). Root with Root browser was used to generate root files and spectra plotting. The obtained spectra were then compared to the experimental one to see how to improve the code and how the simulation feels in accord with experiment.

The energy deposition in the sensitive volume had been histogrammed and spread with an energy-dependent peak shape function to copy the behavior of the signal processing electronics. Finally, the spectrum was written out in a file previously defined as a pair of an ASCII. In all simulations, 10^9 initial photons were generated in 4π that took presently about 2–6 h on an Intel i5 personal computer, in 64-bit Linux environment (Scientific Linux). The simulation runs brings common peak areas of around 5000 to 40,000 counts, thus their statistical precisions were similar to the experimental. During the simulation, the history of each individual primary particle comprises its emission by the source, its interaction with the detector and surrounding materials, production as well as transport of secondary particles, and track until the point when the photon escapes or undergoes a photoelectric interaction in the crystal, depositing all of its energy. Particle energy of 0.5 keV is set as the cut-off for the simulation (Geant4 Collaboration, 2016).

In broad energy germanium detector models, the active volume size and the dead layer thickness are key parameters for obtaining accurate simulation results. It is thus important to take into account this parameter for modelling detection systems. A point-like source of ^{241}Am was used to measure the front and side dead layer thicknesses, but the simulation was performed repeatedly and the thickness value of the corresponding dead layer in the detector model was changed every time, and along these lines the reliance of the detection efficiency on the dead layer thickness was acquired. This reliance relationship was fitted with an exponential function, and the genuine dead layer thickness was controlled by interpolating the acquired function to the experimentally acquired detection efficiency (Budjáš et al., 2009). The active volume of the detector was checked with a collimated ^{133}Ba point-like source. The fundamental factors that contributed to the simulation were characteristic dimension of the detector: the diameter and length of the crystal, the diameter and length of the crystal cavity, top and side dead layer, end cap diameter, window thickness and window to crystal gap. In order to limit the discrepancy amongst simulated and measured esteems, buletization, dead layer and window to crystal gap were varied in the simulation.

The introduction of a simple variance reduction scheme in view of directional bias improves the simulation. The goal of this technique was to simulate only primary photons that were emitted from the sample towards the detector active volume. The implementation in the developed GEANT4 code an algorithm described in details by Hurtado et al. simplified the simulation (Hurtadoa et al., 2004b). Sample dimensions were likewise measured with care, and sample volumes were built in the simulation code accordingly. Primary photons were created in the sample volumes with uniformly random positions and momentum directions in full space (4π) as described in GEANT4 user's manual (Geant4 Collaboration, 2016). Output files of each run were created via UI commands. Filled output files were exported and saved into an Excel sheet. The comparison between simulated output files and experimental result was done to see whether the simulated efficiency fall with experimental one. Simulated efficiency was also applied for realistic measurement to validate the Geant4 model built in this study. The results obtained in this research project are important and will now be presented.

3. Result and discussion

The present work is devoted to the simulation of a broad energy germanium detector and its response. The attention had been given on the reproduction of the geometry by the GEANT4 code and the simulation of the efficiency response of the BE6530. This type of detector has a short and planar shape to maximise efficiency. At this point the geometry and matrix were identical, so that there was only a distinction in the action of the two materials, the spectrum permitting one to perform direct efficiency calibration. That is a requirement in gamma-ray spectrometry: the calibration source and the sample must be in the same geometry and similar matrix (type, composition and density). As to the issue of coincidence summing, no corrections have to be applied if a natural sample is measured with respect to a standard of the same radionuclide, in a similar geometry. That is why attention was given to geometry construction using GEANT4 in this work. In the present work, the BEGe detector geometry (model BE6530) was built firstly and then, the simulation of its response done by using different radionuclides at different energy levels.

3.1. Result of geometry construction

The preliminary simulation of detector geometry began on the Germanium detector crystal and the lead shielding. Another part of the detection system was implemented consequently and progressively in the word volume which is the lead shielding in the code built in this research study. The physical volume, the mother volume and sensitive volume were implemented in the word volume accordingly using GEANT4 routines as describe in GEANT4 user manual (Geant4 Collaboration, 2016). The sample was easily simulated as a real sample with point like source and approximately 120 cm^3 cylindrical volume sample (5 cm diameter and 7.6 cm height).

As presented in Figs. 2 and 3, real dimension geometry can take more resource for first-time construction, but it is the best solution because all other used will just take into account some modifications. Detectors might be obviously characterised by restrictive programming in various circumstances. Monte-Carlo simulation is a useful while determining the detector response and efficiency during an environmental measurement (Guembou et al., 2016, 2017a, 2017b). When considering the sample in the geometry, it was plotted in three dimensions and surface coloration to optimise visualisation. The Qt and Open OGL

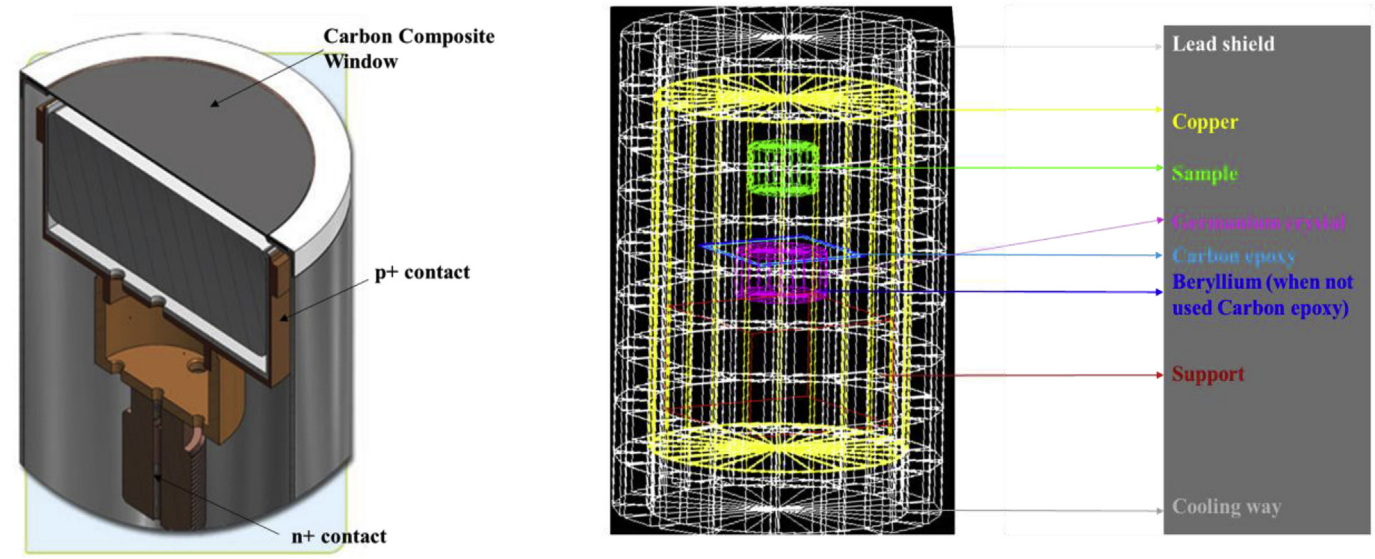

Fig. 2. Geometry construction of the Broad Energy Germanium detector (the BE6530 model). The geometry of the system features 10 cm lead thickness (presented here with white colour) and is jacketed by a 9.5 mm steel outer housing. The graded liner consists of a thin layer of 1 mm thickness and a copper layer (the yellow part of the geometry) of 1.5 mm thickness. There is no exposed lead in this model with or without the door closed. The inside dimensions are 28 cm diameter by 41 cm high approximately. The carbon epoxy end-cap was used and is shown in the geometry by blue colour. Only carbon epoxy or beryllium can be used, not two of them. The sample is the green part and the Germanium detector is the purple part of the geometry construction. Geometry provided by Canberra at left and simulated in the present work at right. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

visualisations were used when drawing the different part of the detector geometries. The crystal of Germanium detector is shown in the centre with purple colour. The sample cylinder is shown in green colour. Drawing geometry in this way is more comfortable and permitted easy modification and adaptation in the C++ code. The detector is defined in the centre. The surrounding lead shield and liners are also simulated, with yellow and white corresponding to lead. Some details are missing and are not visible, from the electrodes and other background

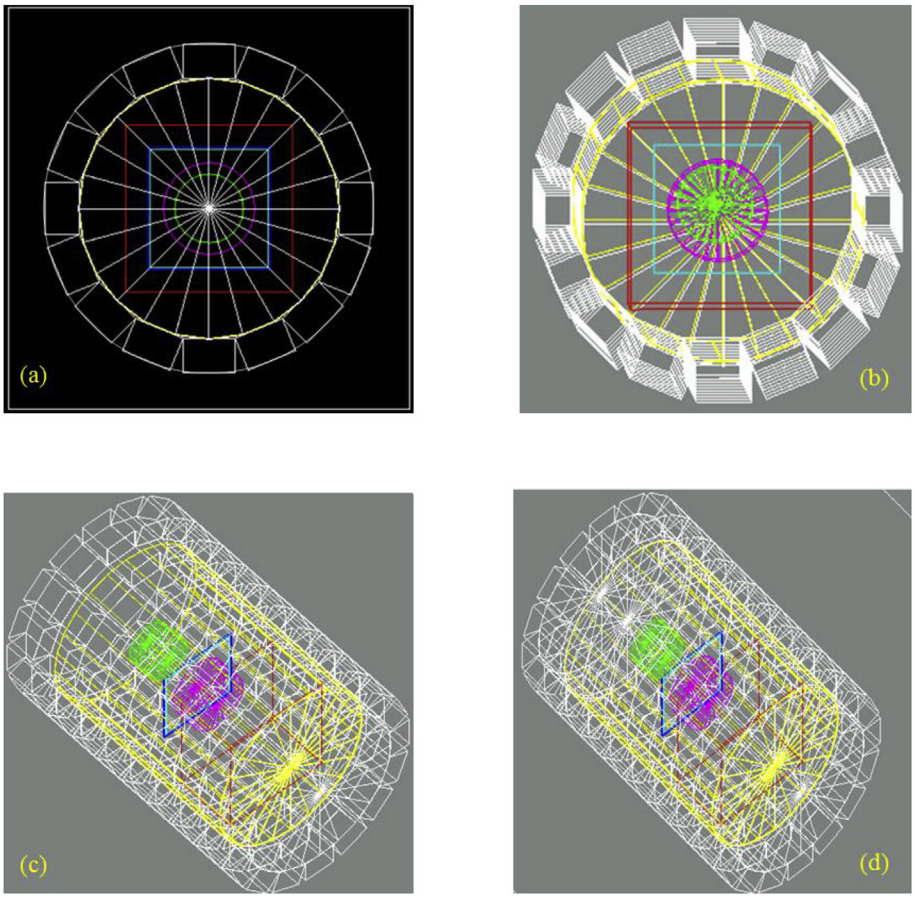

Fig. 3. Different 2D and 3D views of the geometry construction: (a) top view in two dimensions - planar representation. (b) Top view - representation in three dimensions. (c) Reel view of the geometry with rotation. The lead bulk is open. (d) The lead bulk is closed and the system is ready for an acquisition. (c and d) View without the lid and with it respectively.

Table 2
Simulated elements in the geometry of the detection system. Mat. Name for material name; NComp for number of chemical component.

Simple material (Elements)					
Mat. Name	Element	NComp	Density (g.cm^{-3})	I(eV)	Geant4 command name
Beryllium	Be (Z = 4)	1	1.848	63.7	G4_Be (Input window materials)
Support	Al (Z = 13)	1	2.699	166	G4_Al (Aluminum detector protection)
Germanium crystal (> 99.9%)	Ge (Z = 32)	1	5.323	350	G4_Ge (detector)
Copper (Shield mat.)	Cu (Z = 29)	1	8.960	322	G4_Cu (copper shield material)
Tin	Sn (Z = 50)	1	7.31	488	G4_Sn (tin shield material)
Tungsten	W (Z = 74)	1	19.3	727	G4_W (tungsten shield)
Lead (Shield mat.)	Pb (Z = 82)	1	11.35	823	G4_Pb (lead shield: the primary material for shielding)
Compounds					
Carbon Epoxy	H (1), C (6), O (8), Cl (17)	4	1.6	/	New G4Material (Molecule definition: $C_{21}H_{25}ClO_5$)
Word Volume (default)	Air	4	0.00120479	85.7	Base Mat: G4_AIR Mass fraction: C (0.000124), N (0.755268), O (0.231781), Ar (0.012827)
Cooling way	stainless steel + LN_2		7.85	/	Mass fraction (%): C(0.08), Mn(2.0), P(0.45), S(0.3), Si(0.75), Cr (18), Ni(8), N(10), Fe(71)
Polyethylene terephthalate (PET)	Sample container	3	1.38	/	New G4Material [Molecule definition: $\mathbf{(C_{10}H_8O_4)_5}$]

Sample SS1 elemental composition in % (New G4Material: Compound)									
SiO_2	Al_2O_3	K_2O	Fe_2O_3	Na_2O	TiO_2	CaO	MgO	MnO	P_2O_5
91.0	4.9	2.0	0.9	0.4	0.2	0.3	0.01	0.01	0.01

protection in this representation. Different parts of the simulated equipment and sample were presented in Table 2.

The geometry of similar detector was provided by Britton (Britton et al., 2013b, 2014b). As presented above, it is more reliable to simulate the geometry with rings for best viewed of different components. A regularly disregarded part of BEGe detector sensitivity is the sample geometry. For a given sample size the sample ought to be appropriated to limit the distance between the sample volume and the detector itself. The sample size ought to be as large as practicable for greater sensitivity. A perspective to consider is the absorption of γ-rays inside the sample material itself, and the impact of the dimensions of the sample compartment on this. GEANT4 allows complex geometry construction.

3.2. Efficiency simulation

To know which approach yields better result, well known activity sources were used as unknown samples. This comparative study was based on analysing measured spectra and the calculation of the radionuclide activity concentration values. The calculation was based on two steeps: step first to calculate the activity using experimental efficiency curve by applying self-absorption and true coincidence summing correction operate through Genie 2000 cascade summing correction. Second step to calculate of the activity using gamma efficiency from GEANT4 MC Code. The Canberra Broad Energy Germanium (BE6530 model) detector covers the energy range of 3 keV to 3 MeV like no other. For this reason, the efficiency simulation required the optimization of the GEANT4 code used.

Data were collected for approximately 7200–21,600 s, and tests were carried out with multiple sources to validate the model. The distance from the carbon epoxy window to the crystal was revised down during simulation to 4.7 mm, and the dead layers remained unchanged from the manufacturer's specifications. The optimised parameters of the real detector were used on an analogue simulation of the response of the used detector. Comparison of the acquired spectrum with Genie 2000 and simulated spectrum were performed as seen in Fig. 4. The spectra presented were from three samples consisting of a ^{137}Cs, ^{152}Eu and ^{57}Co point sources located at the reference geometry, and the agreement of the spectral shapes was astounding for the whole energy range. This outcome demonstrates the GEANT4 capacities for reproducing gamma ray spectrometers through MC simulation.

Fig. 4 show Broad Energy Germanium spectra for single and complex sources simulated utilising the GEANT4 Monte Carlo toolkit, with the geometry parameters enhanced. There is not significant difference between the experimental spectra and the simulated one. A sand NORM sample named SS1 from IAEA WW open proficiency test has likewise been tested using a GEANT4 based proficiency test, permitting wealth assessments to be ascertained for a range of radionuclides present in the analysed sample. The simulation permits significantly more adaptability than the present restrictive proprietary software, and this approved model would now be able to be utilized to portray the detector response for an assortment of complex geometries and source arrangements.

The simulated efficiency using MC method based on GEANT4 toolkit was provided and is in good agreement with our expectations. As shown in Fig. 5, the comparison between simulated and experimental efficiency present concordance at low energies and slightly discrepancies at high energies. Both calculated and measured values were then fitted utilising the following fitting function (Osae et al., 1999):

$$\ln \varepsilon = \sum_{i=1}^{n} a_i . (\ln E_\gamma)^i \quad (5)$$

where ε is efficiency, E_γ is energy given in keV and a_i are fitting coefficients.

It was set up that, for the detector used, the best calibration bend is accomplished by utilising i = 6. The fitting is used to obtain a calibration curve that would be utilized as a part of any realistic estimation. Adjustment bends for both experimental and simulated efficiency for geometry considered in this paper are shown in Fig. 4. Gotten results can be considered as a relatively good agreement between experimental values obtained directly with the calibration sources and values obtained utilising simulated efficiencies. The upgrades can be made considering the geometry of the detector and chemical composition, volume and mass of the samples. By fluctuating these parameters, recreated results should approach the genuine esteem.

This result shows the GEANT4 capabilities for simulating gamma ray spectrometer response. Comparison between experimental and simulated calibration curve shows the evidence and importance of MC method in environmental nuclear physics, especially in gamma-ray spectrometry. The comparison shows good agreement between experimental and simulated results except in the case of high energy range for the multigamma source. In fact, it was observed that the observed discrepancies were probably due to true coincidence summing

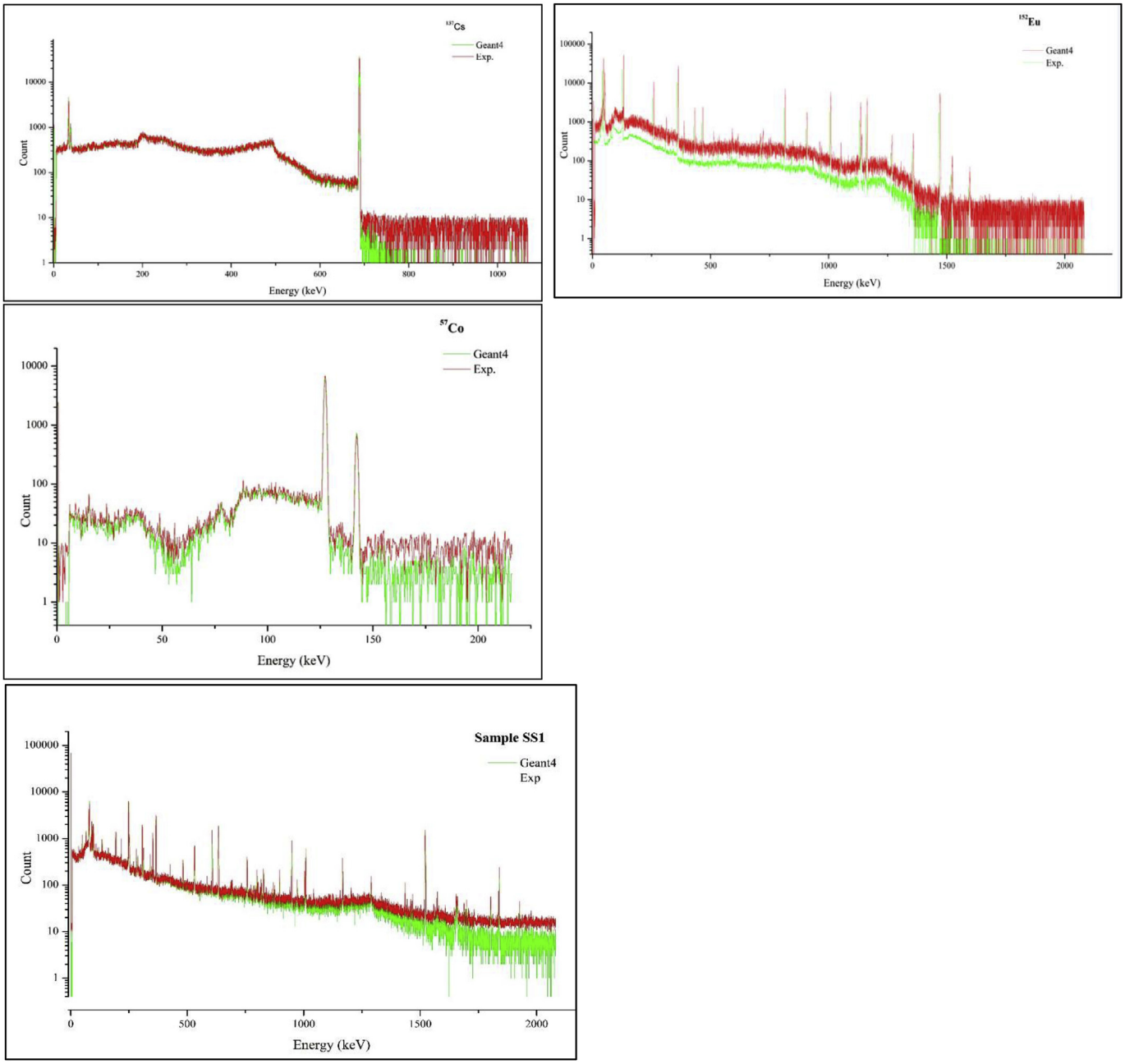

Fig. 4. The energy spectra obtained experimentally and from the simulation using the full GEANT4 decay library, with a pure ^{137}Cs, ^{152}Eu and ^{57}Co source in the half convenient geometry. The events were generated at a randomised coordinate within the source geometry, and sum peaks where two or more photons have 'summed out' could be clearly seen on the plotted graphs. The last graph is for a reference sample named SS1 prepared and labelled in collaboration with the IAEA.

of ^{60}Co and ^{88}Y. This discrepancy is probably due to the true coincidence summing caused by ^{60}Co and ^{88}Y emission present in the sample. Concerning environmental radioactivity assessment, the interest of high energy range is limited: the primordial radionuclides emitted low gamma rays and the ^{4}K is considered as the highest with gamma at around 1460 keV. For this reason, the agreement between experimental data and simulated one was found as a good result for Broad Energy Germanium (especially BEGe6530) detectors which were made for the environmental purpose. Results obtained using GEANT4 toolkit were useful and enabled us to calibrate broad energy germanium detector without radioactive sources.

The simulation using 120 ml sample in cylindrical geometry demonstrated the best efficiency (Fig. 5d and e). It is important to notice that the 120 ml geometry is set as a basic and recommended geometry in our laboratory and is being used at 90% for environmental sample analysis. This can be explained by the statistical counting during experiment and simulation. The solid angular parameter can contribute as well as it is big. But at low energies, it is observed a very low split on the two graphs (4d and 4e). Since the geometry of the detector built with GEANT4 was not as complete as the experimental detector, we can assume that this discrepancy is to be expected as reported by Park et al.: the secondary photon emitted after first interaction (Park et al., 2003; Allisonet al, 2006). In addition, the self-absorption of the sample influences the result at low energy. Since the dead layer thickness extraordinary influences the results, in the case of the detectors especially the type studied in his work, the dead layer thickness can change significantly from the one characterised by the producer. Since the thickness of the dead layer was one of the parameters that were improved in the simulation to deliver the best agreement with the test ones and that optimization did not diminish the error, some different

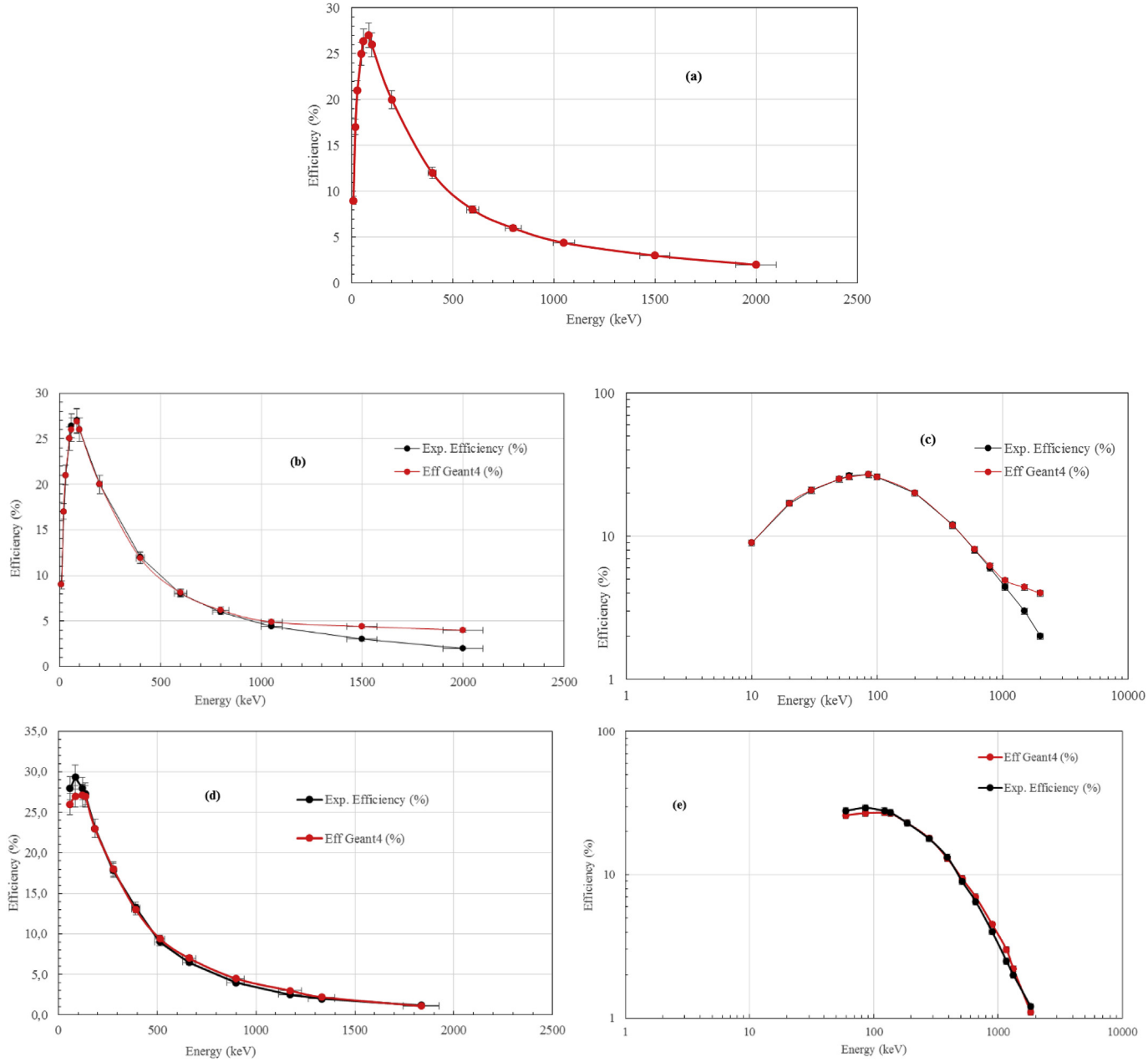

Fig. 5. Simulated and experimental calibration curve for BEGe detector (BE6530 model). Red curve represents results obtained by GEANT4 and black represent experimental results. (a) ^{152}Eu first sample; (b and c) are results from second sample of multigamma source containing ^{241}Am, ^{152}Eu, ^{137}Cs, ^{109}Cd, ^{60,} ^{57}Co radioisotopes; (d and e) measurement geometry is indicated in 120 ml cylindrical container of PET containing 320 Bq of soil sample (with the following chemical composition of material in percentage: $SiO_2 = 91 \pm 5$; $Al_2O_3 = 4.9 \pm 0.4$; $K_2O = 2.0 \pm 0.1$; $Fe_2O_3 = 0.9 \pm 0.1$; $TiO_2 = 0.2 \pm 0$; $Na_2O = 0.4 \pm 0$; $MgO = 0.01 \pm 0$; $CaO = 0.3 \pm 0$; $MnO = 0.01 \pm 0$; $P_2O_5 = 0.01 \pm 0$). $(C_{10}H_8O_4)_n$ is the chemical formula of PET used with n = 5 in GEANT4 code. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

remedies must be made (Nikolic et al., 2014b, 2015).

The estimated value of efficiency relied on the geometry of the sample – thickness, density, and separation distance from the detector. For the detectors utilized as a part of gamma spectrometry analysis, efficiency changes altogether as a function of these parameters. In this way, each counting geometry requires an efficiency alignment, utilising a well-known standard in the same geometry which includes multiple gamma-ray energies: the ^{152}Eu source can only be used for energy calibration. The estimation of the counting efficiency was controlled by factors such as: attenuation of photons inside the source because of absorption in the sample material; attenuation of photons in the canning material over the entrance face of the detector; the fraction of the photons discharged by the source that hits the sensitive volume of the detector; and the fraction of the photons hitting the detector that adds their contribution to the full-energy peak (Nikolic et al., 2014b; Allisonet al, 2006; Joković et al., 2009).

It is experimentally not easy to ascertain the efficiency esteems at various energy values for all the geometric plans that were utilized as a part of research in the laboratory. It is, along these lines, standard practice to get ready calibration samples for test and access the efficiency of the detector empirically. Similarity between the calibration samples and the field samples (that are to be analysed) in every respect must be respected. It includes the following characteristics: matrix composition, physical form, and dimensions. It must be conceivable later to align the field samples in the very same position with respect to the detector used for the experiment. They should likewise contain

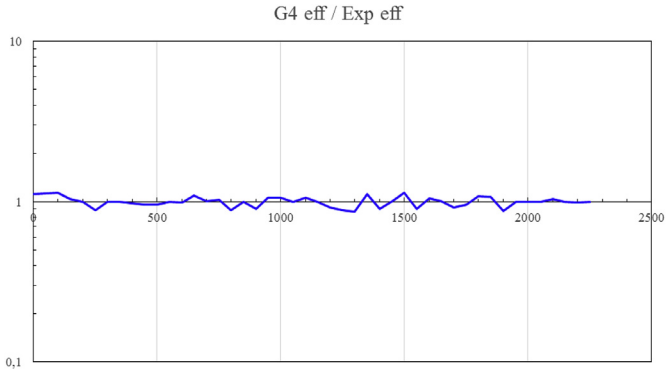

Fig. 6. Rapport of simulate and experimental efficiencies depending on gamma energy.

various nuclides to cover the energy range that is of interest. What's more, their activity estimation must be known precisely, and traceable. They must be mechanically strong (to forestall contamination of a detector) and chemically stable (to dispose segregation of the active component from the matrix).

It is ordinarily unrealistic to buy calibration standards that fulfil every one of these necessities, and the investigator is constrained to set it up in the laboratory. It requires a great deal of advancement, and regularly the examiner must bargain in view of reasonable issues. The issue can be streamlined to some degree if a predetermined number of geometric arrangements are characterised with the goal that the number of calibration samples can decrease. The following game-plan ought to be to enhance the meaning of the detector geometry and perform another realistic measurement utilising more appropriate reference materials and geometries (Nikolic et al., 2014b; Krmar et al., 2013; McNamara et al., 2012; Akkurt et al., 2014). The upgrades can be made considering the geometry of the detector and chemical composition and thickness of the sample. By varying these parameters in the code or algorithm using in GEANT4 toolkit, obtained simulated results should approach the genuine value with sophisticated sample geometry and real dimension of the detector.

3.3. Novel scientific result

The observed mean discrepancy lesser than ± 2% between the simulated and experimental efficiency (Fig. 6). Can be due to an inadequate knowledge of the real detector geometry and to imperfect charge collection in detector crystal used in our simulation. Fig. 5 proved that both experimental and simulated values of efficiency are too closed and that the results of the simulation can be properly used for calibration of the spectrometer. It is generally too difficult to obtain both closed experimental and simulated result less than 2% confidence. The Monte Carlo simulated efficiency based on Geant4 toolkit would be more suitable in the case of environmental radioactivity monitoring. Additionally, the simulations can aid in determining the best operation mode of the Broad Energy Germanium detector for a particular energy range.

The comparison between simulated and experimental values obtained in this study is given in Table 3. It can be seen that all radionuclides presented good result when comparing both experimental and simulated values to reference values. Therefore, it is possible to assume that the simulated efficiency with GEANT4 can be used with calibrating the detector for analysis. The ^{152}Eu is the largest value in term of deviation between experiment and computation, but the ratio is about 95%, a good agreement.

4. Conclusion

The present study was devoted to the simulation of the HPGe detector (BE6530 model) and its response. In this work, the Monte Carlo methods were effectively used for the simulation of the HPGe detector. The introduction of a basic variance reduction scheme in view of directional bias to simulate only primary photon emitted from the sample in the direction towards the detector active volume facilitated the work and permits gain time. The simulation of different sample's geometries and importation of the simulated calibration files in Genie (2000) reduced difficulties encountered during the calibration process. In addition, it is possible to construct the real dimension of our sample geometry when well known. The real geometry of BE6530 detector has been built and its response performed. 2D and 3D geometries were constructed using Qt and OGL visualisations. Rings were used for geometry construction due to the best views that it provides.

The multigamma sources used in our laboratory were used for computational calibration. Comparison between experimental and simulated calibration curve shows good fitting for low gamma energies and a slightly differences for high gamma energies. Discrepancies were probably due to true coincidence summing of ^{60}Co. It was observed that MC method based on GEANT4 can be used in the gamma spectrometry measurement as well as analytical method. In addition, it was found that this method is a valuable, inexpensive and gain time alternative that additionally provides more prominent adaptability while determining the detector response and efficiency during a measurement.

While new capabilities are as of now being produced to address the issues of investigation at the high energy, nuclear physics and gamma spectroscopy especially, unmistakably the expanding utilization of GEANT4 in other areas will likewise lead to new toolkit advancements. It is planned to simulate and provide more reliable geometries response, with all components from protection to electronics and their contribution to the low background, to the gamma spectrometry research in the near future.

Table 3
Simulated and experimental values obtained with the mix gamma standard source ABLA-17 as reported on the sample at the laboratory.

Nuclide	Energy (keV)	Emission prob	Activity concentration (Bq/kg)					Sim/Exp ratio
			Experimental	%Unc	Simulated	%Unc	Target values	
Am-241	59.54	0.36	418.20	2.60	398.00	2.23	420	0.95
Cd-109	88.03	0.04	1510.80	6.20	1495.68	6.21	1550	0.99
Co-57	122.06	0.86	43.30	1.50	44.78	1.53	45	1.03
Cs-137	661.66	0.85	370.10	1.90	367.37	1.62	380	0.99
Mn-54	834.84	1.00	190.30	1.40	193.28	1.29	200	1.02
Y-88	898.04	0.94	139.80	1.90	143.83	1.58	140	1.01
	1836.20	0.99	139.80	1.90	143.83	1.58	150	1.01
Zn-65	1115.55	0.51	329.90	1.90	329.06	1.80	335	1.00
Co-60	1173.24	1.00	398.20	1.50	410.32	1.77	400	1.03
	1332.50	1.00	398.20	1.50	410.32	1.77	420	1.03

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.jenvrad.2018.03.015.

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