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Intercomparison of full energy peak efficiency curves for an HPGe detector using MCNP6 and GEANT4

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ABSTRACT

This work is focused on studying the capabilities of different Monte Carlo tools to complement the Full Energy Peak Efficiency (FEPE) calibration procedures of the Environmental Radioactivity Laboratory of the Universitat Politècnica de València, UPV. In this frame, detailed models of a High Purity Germanium detector have been implemented using MCNP6 and GEANT4. Accordingly, an inter-comparison with experimental values ensures the model validation and improves the analysis of the physics phenomena involved. The validation of the models is determined by a quantitative comparison between the simulated and the measured efficiencies over the energy range under study (59.54–1836.01 keV). The results show discrepancies between both Monte Carlo tools for ^{139}Ce, ^{88}Y and ^{60}Co as GEANT4 is able to simulate the coincidence summing effect of these radionuclides.

1. Introduction

High Purity Germanium (HPGe) detectors are widely used in gamma-ray spectrometry for the determination of radionuclides and their activity in environmental samples. To obtain accurate measurements, a detailed characterization of the efficiency response is required. In this frame, computational techniques can be applied joint to experimental procedures to obtain the efficiency calibration curve of a detector system. The Full Energy Peak Efficiency (FEPE) can be performed using both Monte Carlo and deterministic-approach codes. In the literature, several codes and tools based on the Monte Carlo method can be found: GESPECOR (Sima et al., 2001), MCNP (Briesmeister, 1997), GEANT (Brun et al., 1986), PENELOPE (Salvat et al., 2003), EFFTRAN (Vidmar, 2005), EGS4 (Nelson et al., 1985), CYLTRAN (Halbleib and Mehlhorn, 1986) and FLUKA (Ferrari et al., 2005). Deterministic-approach codes can be also suitable for this purpose: ETNA (Piton et al., 2000) and LABSOCS (Bronson, 2003). The comparison between simulation and experimental efficiency curves, normally present discrepancies that can be attributed to the modelization of the geometry or other aspects like the dead layer thickness of the germanium crystal (García-Talavera et al., 2000; Jurado-Vargas and Guerra, 2006). Therefore, it is mandatory to carry out a realistic geometric characterization considering the data provided by the manufacturer.

This work is focused on studying the capabilities of two widely accepted Monte Carlo tools, MCNP6 and GEANT4 (Ródenas et al., 2000; Hurtado et al., 2004), to complement the experimental calibration procedures of the Environmental Radioactivity Laboratory of the Universitat Politècnica de València (UPV). Accordingly, an inter-comparison between models ensures the validation of the model and improves the analysis of the physics phenomena involved. In this context, the main difference between both codes is that GEANT4 simulates the radioactive decay of a radionuclide by using the Radioactive Decay Module (RDM) (Hurtado et al., 2009). Moreover, the RDM allows simulating the Coincidence Summing (CS) effect present in the experimental measurements (Hauf et al., 2013). This phenomenon takes places when, in close geometries, a radionuclide emits photons in cascade reaching the detector within the resolution time. Consequently, the efficiency of those radionuclides and their activity could be affected.

2. Materials and methods

2.1. Experimental set-up

The experimental set-up consists of a gamma spectrometer with an HPGe detector (ORTEC GMX series) and a multi-channel analyzer with 8192 channels. The system has a relative efficiency of 40% at 1.33 MeV and a nominal resolution of 0.76 keV and 2 keV at 5.9 keV and 1.332 MeV, respectively. Table 1 describes the geometry features of the detector provided by the manufacturer.

The experimental measurements have been performed using a multigamma-ray standard source containing the following

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Table 1
Geometric features of the detector (manufacturer values).

Parameter	Nominal values (mm)
Ge crystal diameter	60
Ge crystal total length	71.1
Inner core depth	63.1
Inner core radius	4.5
External Dead Layer	0.0003
Inner core Dead layer	0.7
Be windows distance	4.0
Be windows thickness	0.5
Al/Mylar thickness	0.03/0.03
Al cup thickness	1.0
Al holder thickness	0.8

Table 2
Composition and density of sea sand and zirconium sand.

	Sea sand	Zr sand
Silicon dioxide	86.57%	57.33%
Calcium	11.48%	–
Aluminum	1.95%	–
Zirconium	–	42.67%
density	1.65 g/cm^3	3.5 g/cm^3

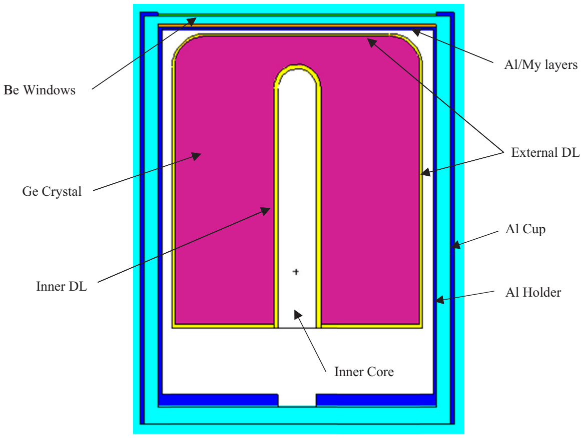

Fig. 1. Detector model. The scheme is not to scale.

Table 3
Optimized geometric parameter.

Parameter	Manufacturer values (mm)
Hole radius	7.5
Top Ge dead layer	0.045
Edges Ge dead layer	0.025
Side Ge dead layer	0.130
Bottom Ge dead layer	0.130
Inner Ge dead layer	1.7
Window distance	4.0

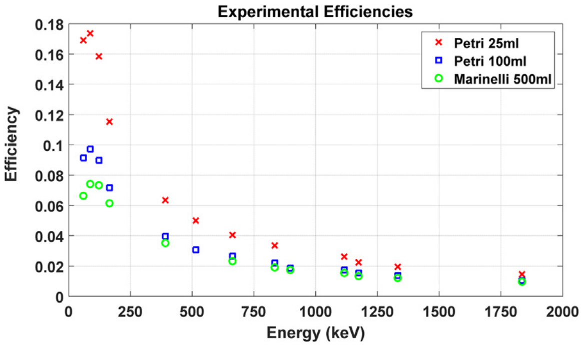

Fig. 2. Experimental efficiencies (water matrix).

radionuclides: ^{241}Am, ^{109}Cd, ^{57}Co, ^{139}Ce, ^{113}Sn, ^{85}Sr, ^{137}Cs, ^{54}Mn, ^{88}Y, ^{65}Zn, and ^{60}Co. Gamma-ray spectra has been analyzed using Gamma Vision Software [GammaVision V5.10, ORTEC] and the Full Energy Peak Efficiency (FEPE), ε, for a given photon energy has been obtained from the following expression:

$$\varepsilon = \frac{N}{t\bullet A\bullet P_\gamma} \tag{1}$$

where N is the number of net counts in the peak, A is the source activity, t the counting time and Pγ is the photon emission probability.

The experimental detector efficiency is determined for different five samples: 15 ml 25 ml Petri box filled with water (PS25W), 100 ml Petri boxes filled with water, sea sand and zirconium sand (PS100W, PS100SS, PS100ZrS) and 500 ml Marinelli beaker filled with water (MS500W). The composition of the sands (Table 2) has been analyzed by electronic microscopy. In each sample, the multigamma standard source has been homogeneously distributed. The maximum relative errors of the experimental measurements were about 2.4%.

2.2. Monte Carlo models

MCNP6 (Monte Carlo N-Particles Transport Code) (MCNP6TM Monte Carlo team, 2013) is a Monte Carlo transport code system for coupled neutron, photon and electron, with all the corresponding cross-section data to transport calculation. The F8 tally for photons and electrons has been used to collect the deposited energy in the active crystal (Pulse Height Distribution, PHD) per emitted gamma particle. This tally provides the energy distribution of the pulses created in the active germanium crystal. The energy resolution has been simulated using the Gaussian Energy Broadening (GEB) card, obtaining a realistic spectrum performance. The GEB card characterizes the energy resolution using the following expression:

$$FWHM\,(E) = a + b\bullet\sqrt{E + c\bullet E^2} \tag{2}$$

where E is the energy of the photon, a = 0.00088692 MeV, b = 0.00033971 MeV$^{1/2}$ and c = 5.0257 MeV^{-1} [MCNP6 User's Manual, 2013]. These constants are determined by mathematical regression from the experimental data. This option, along with the GEB card, provides a gamma spectrum comparable to the experimental one as the relation channel-energy and energy-resolution is the same. The efficiency calculation has been performed applying the same method as the experimental measurements by importing the MCNP6 output into Gamma Vision.

The Geant4 toolkit (Agostinelli et al., 2003) presents an object-oriented programing that allows choosing among a wide range of physical processes or implementing them according to the experiment needs. The physics processes activated in the detector model of this work are the following: Auger electron production, Compton and Rayleigh scattering, pair production, photoelectric effect for photons, ionization processes and Bremsstrahlung for secondary particles. To simulate the coincidence summing effect and the decay of the different radionuclides, the Radioactive Decay Module (RDM, G4RadioactiveDecay class) was used (Truscott, 2002). The RDM simulates radioactive decays by sampling secondary particles on a per-decay basis, using branching ratios from the Evaluated Nuclear Structure Datafile (ENSDF).

The data output has been distributed into 8192 channels from 0 to 2040 keV simulating the multi-channel analyzer used in the experimental set-up and considering the energy calibration obtained with Gamma Vision from the experimental measurements. A variance reduction method consisting of a cut-off for secondary particles with energies below 10 keV (MCNP6) and a mean free path below 10 µm

Table 4
Simulated to experimental efficiency ratios (**) MCNP6 & GEANT4; PS25W, PS100W; MS500W.

Radionuclide	E (keV)	PS25W			PS100W			MS500W		
		Exp. Efficiency	MCNP6 ratio	GEANT4 ratio	Exp. Efficiency	MCNP6 ratio	GEANT4 ratio	Exp. Efficiency	MCNP6 ratio	GEANT4 ratio
^{241}Am	59.5	0.1692	0.99	0.99	0.0915	0.99	0.98	0.0662	1.02	1.02
^{109}Cd	88.0	0.1735	1.00	0.99	0.0972	0.99	0.98	0.0740	1.06	1.06
^{57}Co	122.1	0.1584	1.01	1.01	0.0899	1.02	1.01	0.0733	1.05	1.07
^{139}Ce	165.9	0.1151	1.18	1.05	0.0717	1.13	1.03	0.0711	1.16	1.04
^{113}Sn	391.7	0.0633	0.99	0.99	0.0396	1.01	0.99	0.0352	1.01	1.03
^{85}Sr	514.0	0.0499	0.98	0.98	0.0306	1.04	1.02	-*	–	–
^{137}Cs	661.7	0.0405	0.97	0.97	0.0264	0.95	0.97	0.0231	1.01	1.02
^{54}Mn	834.8	0.0333	0.97	0.97	0.0219	0.97	0.96	0.0192	1.02	1.03
^{88}Y	898.0	-*	–	–	0.0185	1.08	0.97	0.0174	1.06	0.97
^{65}Zn	1115.5	0.0261	0.99	0.98	0.0174	0.98	0.98	0.0154	1.02	1.03
^{60}Co	1173.2	0.0225	1.09	0.95	0.0154	1.07	0.96	0.0134	1.09	1.03
^{60}Co	1332.5	0.0196	1.14	0.98	0.0137	1.07	0.97	0.0121	1.11	1.04
^{88}Y	1836.0	0.0145	1.15	0.99	0.0106	1.07	0.96	0.0098	1.07	0.98

* ^{88}Y (PS25W) and ^{85}Sr (MS500W) present high uncertainties in the experimental measurements. Therefore, the FEPE experimental calibration has been performed without considering these efficiencies.

Table 5
Simulated-to-efficiency ratios for ^{139}Ce, ^{88}Y and ^{60}Co; PS100W; MCNP6 & GEANT4 (without RDM).

Radionuclide	E (keV)	MCNP6	GEANT4
^{139}Ce	165.9	1.13	1.12
^{88}Y	898.0	1.08	1.08
^{60}Co	1173.2	1.07	1.06
^{60}Co	1332.5	1.07	1.08
^{88}Y	1836.0	1.07	1.07

(GEANT4) has been applied. The number of histories in each simulation has been established in 20 million to achieve statistical errors lower than the 1.5%.

Both Monte Carlo tools have been used to analyze the detector efficiency response of the system. Parameters such as the distance between the detector window and the germanium crystal, the dead layer thickness or the crystal volume, among others, are of relevance in the efficiency calibration and must be characterized (Chham et al., 2015). A model of the detector was performed in a previous work (Giubrone et al., 2016), using GEANT4 to simulate the efficiency calibration curves for the PS25W, PS100W and PS100SS samples using the manufacturer's parameters. In that model, the crystal shape and the inner core were simplified to a perfect cylinder, increasing the active detection volume and thus, overestimating the efficiencies. The external dead layer surrounding the crystal was taken as constant with a value of 75 µm and without differencing the top from the lateral or the upper edges zones. The inner core dead layer was kept at 700 µm (manufacturer's value).

In this work an optimized model of the detector has been implemented using MCNP6 and GEANT4 for all the samples mentioned in Section 2.1. The geometry of the germanium crystal is now modeled considering the curvature of the edges as well as that of the inner core, obtaining a more realistic model (Fig. 1). The external dead layer has been characterized studying the efficiency of the ^{241}Am (59.5 keV) due to its low penetration and dividing the crystal surface into three different zones: top, upper edges and side/bottom. The characterization of the inner core dead layer as well as the crystal volume has been performed using mainly medium and high energies, from ^{113}Sn (391.7 keV) to ^{65}Zn (1115.5 keV). The window-to-crystal distance, as it affects the solid angle between the sample and the detector, has required all the energy range under study. The final optimized parameters are shown in Table 3.

The main difference between both MC tools regarding this work is the fact that MCNP6 code simulates the gamma particles emissions without considering the coincidence summing effect. Therefore,

Table 6
Simulated to Experimental efficiency ratios (**) MCNP6 & GEANT4; PS100SS, PS100ZrS.

Radionuclide	E (keV)	PS100SS			PS100ZrS		
		Exp. Efficiency	MCNP6 ratio	GEANT4 ratio	Exp. Efficiency	MCNP6 ratio	GEANT4 ratio
^{241}Am	59.5	0.0715	1.02	1.00	0.0089	1.04	1.01
^{109}Cd	88.0	0.0831	1.03	1.02	-*	–	–
^{57}Co	122.1	0.0800	1.04	1.04	0.0382	1.05	1.05
^{139}Ce	165.9	0.0682	1.15	1.04	0.0454	1.06	1.04
^{113}Sn	391.7	0.0364	1.05	1.03	0.0291	1.07	1.06
^{85}Sr	514.0	0.0297	0.97	1.00	0.0257	0.99	0.98
^{137}Cs	661.7	0.0247	0.99	0.98	0.0212	0.99	0.99
^{54}Mn	834.8	0.0205	1.00	0.99	0.0183	0.98	0.98
^{88}Y	898.0	0.0178	1.09	0.97	0.0158	1.08	0.96
^{65}Zn	1115.5	0.0164	1.02	1.00	0.0147	1.00	0.99
^{60}Co	1173.2	0.0143	1.11	1.00	0.0131	1.08	0.97
^{60}Co	1332.5	0.0130	1.10	0.98	0.0118	1.09	0.97
^{88}Y	1836.0	0.0102	1.08	0.97	0.0095	1.07	0.95

* The Zr sand, is a mineral sample containing ^{238}U and ^{232}Th. X-rays from Bismuth, from both decay series, with an energy of 87.35 keV overlaps gamma particles at 88.0 keV (^{109}Cd) being impossible to difference the contribution of each one without avoiding high uncertainty. Therefore, the experimental efficiency calibration has been performed without considering the efficiency of the ^{109}Cd.

simulated and experimental efficiencies cannot be compared for those radionuclides with CS effect (^{139}Ce, ^{88}Y and ^{60}Co) and it is not possible to characterize the detector for high energies (1173.2 – 1836.0 keV) using only MCNP6.

3. Results

Fig. 2 shows the experimental efficiencies for the three geometries with the multi-gamma standard source in a water matrix. The MS500W has the lowest efficiency as it contains a more dispersed source than the other two geometries. At high energies, efficiencies for MS500W and PS100W are almost overlapped. On the other hand, the PS25W represents the opposite case, with the highest efficiencies being more likely to a point source.

Table 4 summarizes the simulated to experimental efficiency ratios obtained with MCNP6 and GEANT4 for the PS25W, PS100W and MS500W samples. The simulated-to-experimental efficiency ratios are within the acceptance range (± 5%) except for ^{109}Cd and ^{57}Co in the MS500W sample. For the radionuclides with the CS effect, ^{139}Ce, ^{88}Y and ^{60}Co, MCNP6 overestimates the efficiency obtaining ratios out of bounds. In order to achieve realistic and comparable results for these radionuclides, the use of GEANT4 is required as it can simulate this phenomenon activating the G4RadioactiveDecay class. If the RDM is deactivated, the efficiencies obtained for these radionuclides are comparable with those calculated by MCNP6 (Table 5).

Table 6 summarizes the results for each sand sample. PS100SS results are in good agreement with the experimental efficiencies for all the energy range, taking into account the limitation of MCNP6 for the CS effect. PS100ZrS presents similar results than the sea sand matrix except for ^{113}Sn in both codes (ratios of 1.07 and 1.06). The ratio obtained with MCNP6 for the ^{139}Ce (165.9 keV) is considerably lower (1.06) in the Zr sand sample and almost the same than the one obtained with GEANT4 (1.04). This result implies that the CS effect is probably attenuated due to the high density of this sand (3.5 g/cm^3), increasing the auto absorption effect for low energies.

4. Conclusions

MCNP6 code and GEANT4 toolkit have been used to obtain the FEPE calibration of an HPGe detector for environmental radioactivity measurements. Simulations of different geometries and matrices have been performed to validate the model by two complementary methods: comparison between simulated and experimental efficiencies and comparing the simulated-to-efficiency ratios obtained with both MC programs.

The simulated to experimental efficiency ratios obtained with MCNP6 and GEANT4 for each sample are within the acceptance range of ± 5% from 59.5 to 1115.52 keV except for ^{109}Cd and ^{57}Co in MS500W sample and ^{113}Sn in PS100ZrS sample. Both tools are reliable to the FEPE calibration in that energy range for the HPGe detector and the multi-gamma standard source used in this particular work. However, from 1173.2 to 1836.0 keV (^{60}Co and ^{88}Y), as these radionuclides have coincidence summing effect, only GEANT4 is able to properly obtaining the efficiency curve as it simulates this effect using the RDM.

On the other hand, comparing MCNP6 and GEANT4 efficiencies, both MC programs show almost exactly ratios for monoenergetic radionuclides in each sample regardless the geometry or the matrix. Moreover, results show that the resolution, modeled only in MCNP6 simulations, has barely any effect on the efficiency values. These results contribute to validate the optimized detector model not only because of the good agreement with the experimental data but between both MC tools.

The particular case of ^{139}Ce in the PS100ZrS shows a ratio of 1.06 and 1.04 for MCNP6 and GEANT4, respectively. This result indicates that, probably, the summing-out effect for this radionuclide (X-ray-gamma) is attenuated due to the high density of the sand.

The use of realistic Monte Carlo HPGe detectors models will lead to replace experimental measures by simulations with the consequent waste reduction and the simplification of working procedures in the laboratory.

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