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Simulation of the response of a PIPS detector using GEANT4 code

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A B S T R A C T

The main objective of this work is to simulate a PIPS (*Passivated Implanted Planar Silicon*) detector response by Monte Carlo method and its validation with experimental results. Specifically, we have calculated via simulation the counting efficiency and the energy resolution of a PIPS detector. In order to do this, we have developed a Geant4 application that includes the most relevant physics processes, the geometry, composition and radioactive content of the sample, and the geometry of the PIPS detector and the vacuum chamber. However, some parameters involved in the Monte Carlo simulation are unknown (detector dead layer or sample thickness), and it was mandatory to estimate them through comparisons between experimental and calculated detector responses in order to obtain an accurate simulation of the PIPS detector.

To show the validity of the simulated results, we present a comparison of the simulated alpha particle energy spectrum with the experimental one from a uranium dioxide pellet.

1. Introduction

Alpha-particle spectrometry is a widely used analytical method, for example in isotopic determination, nuclear decay data measurements as emission probabilities, surveys of environmental radioactivity, or nuclear waste management [1]. Due to the continuous energy loss of alpha-particles along their interaction with matter, the application of this method is based on obtaining thin sources after a radiochemical separation and a deposition process, such as electrodeposition or micro-precipitation. However, there is currently an interest in non-destructive analysis of thick samples such as radioactive hot particles [2–4] or aerosol samples [5,6]. These thick sources undergo a degradation of the energy resolution which affects the quality of alpha spectra containing several overlapping peaks. It is nevertheless possible to employ Monte Carlo simulations to reproduce the shape of the experimental energy spectrum of thick sources and extract valuable information about the nuclide composition, their activity or the characterization of the source.

Many Monte Carlo codes simulate the interactions of alpha particles with matter. The well-known SRIM/TRIM programe [7] is a straightforward implementation for ion tracking in matter, but it can only be applied to simple slab geometries. More specific alpha-spectroscopic simulation codes provide easier implementations and faster results than the general-purpose ones. The code AlfaMC has been recently developed to simulate the transport of alpha particles [8] through the use of simple physical models in complex geometries. Another specific alpha simulation code known as Advanced Alpha-spectrometric SImulation (AASI) was designed to simulate alpha-particle energy spectra [9] with some simplifications in calculations as well (i.e. alpha particles are not tracked in the active volume of the detector). On the other hand, general-purpose Monte Carlo codes, such as MCNPX [10] or GEANT4 [11] are very powerful codes providing a list of capabilities such as handling of complex geometries, visualization tools, and physics models for interactions and transport of many types of particle. The main drawback of these general-purpose Monte Carlo codes is their high computational burden, resulting in very slow simulations in certain situations. Additionally, GEANT4 is also a very complex toolkit written in C++ with a steep and long learning curve.

But even if the best simulation algorithms and physics models are used, the validity of the results depends on the information about the detector setup obtained from the manufacturer. However, such information is not usually sufficient enough to build a realistic model of the experimental setup [1].

In our case, we have used GEANT4 toolkit to simulate the response of a PIPS detector trying to mimic its energy resolution and detection efficiency. In order to do so, it is necessary to determine several parameters such as the active area, source thickness, or the dimensions

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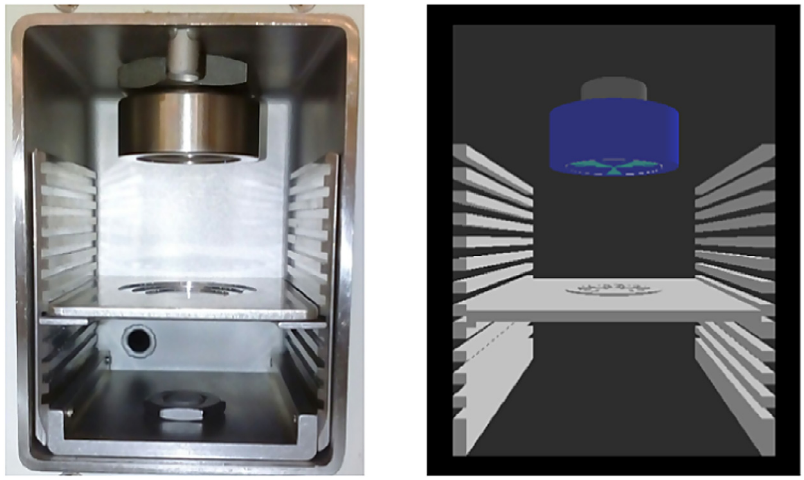

Fig. 1. PIPS detector setup (left) and the corresponding simulated geometry with GEANT4 visualized with OpenGL (right).

of the counting chamber where source and detector are placed. Also, it is important to know as accurately as possible the thickness of the detector dead layer because it plays an important role in the obtained results [12]. The values of these parameters were varied in our Monte Carlo simulations in order to fit the experimental values. Finally, the Monte Carlo simulation was validated by using those optimized parameters into the calculation of the alpha particle energy spectrum of a thick uranium dioxide pellet.

2. Materials and methods

2.1. Materials

The measuring equipment used in the laboratory at CITIUS (*Centro de Investigaciones Tecnológicas e Innovación de la Universidad de Sevilla*) is a fully automated and integrated alpha spectroscopic system (Alpha Analyst, Canberra) consisting of 12 vacuum chambers in which the vacuum can reach 0.022 Torr and each of them hosting a Passivated Implanted Planar Silicon (PIPS) detector. The PIPS detector (model A-450) characteristics provided by the manufacturer are an active area of 450 mm^2, a nominal thickness equivalent to 300 μm, and a front dead layer thickness less than 50 nm. The source is placed in front of the detector which has a sliding source support to adjust the source-to-detector distance (see Fig. 1).

We used two different radioactive sources. First, a ^{241}Am standard source electroplated on a stainless-steel disc containing 173.9 ± 3.5 Bq from CIEMAT (Spain) (s/n: FRC-2014-00360) was used to determine the energy resolution and the counting efficiency for the PIPS detector. Second, we used a natural source of uranium dioxide provided by CIEMAT. This source consists of a cylinder with radius 5.31 mm and height 4.40 mm, with a density of 10.34 g/cm^3 and a weight of 4.05 ± 0.02 g. The U activity was calculated from the uranium amount content, assuming the U is natural and secular equilibrium is fully established down its decay chain (99.284% ^{238}U, 0.711% ^{235}U and 0.005% ^{235}U). The activity obtained was 99.1 ± 0.7 kBq.

The homogeneity and active area of the sources was obtained through the imaging plate technique (FLA-5100, Fuji Film Co.) available in the *Servicio General de Investigación de Biología* (CITIUS). The sources were positioned directly onto the surface of the imaging plate. Information in the imaging plate was read out after 1 day using a reading system with a palette of 16 bits and a spatial resolution of 25 μm. The image was analysed using ImageJ software [13].

2.2. Simulations

Our Monte Carlo simulations have been carried out using the GEANT4 toolkit, (version 10.1.1) [11,14,15]. GEANT4 is an open-source toolkit for High Energy Physics (HEP) experiments using Object-Oriented paradigm and C+ + programming language. GEANT4 is not only for HEP but also for medical applications, space science and cosmic-rays physics. A large degree of flexibility and functionality are available for geometrical models, primary particle generation, physics processes and visualization and analysis algorithms.

The whole PIPS detector assembly and materials have been simulated with GEANT4, as shown in Fig. 1. The dimensions of the silicon detector, entrance window, vacuum chamber with tray and slots, stainless steel source substrate and vacuum (0.022 Torr air) were provided by the manufacturer or, in some cases, physically measured using a digital vernier calliper.

The spatial distribution of the radioactive sources was modelled through the General Particle Source (GPS) module which allows to model complex source geometries with macro-driven commands. This module allows us to define the primary particle and establish its position distribution within the source, angle distribution and energy distribution. In this work, the angular distribution was set to be isotropic, and the energy distribution and source position is shown in detail below.

As for the physics models considered in our simulations (usually named "physics list" in the GEANT4 context), we considered the Standard Electromagnetic option 3 that simulates ionization, bremsstrahlung, gamma conversion and other electromagnetic interactions of gamma, electrons, and charged particles with energies from 1 keV up to 10 PeV was included [14]. The production cuts were set to 1 nm.

Regarding the primary particles of the source, the G4RadioactiveDecay module was used to generate them [16]. G4RadioactiveDecay generates all the possible decay paths of a particular radionuclide using the branching ratios based on data from the Evaluated Nuclear Structure Data File (ENSDF). In particular, ^{241}Am alpha decays to its daughter isotope ^{237}Np. The nuclear de-excitation of ^{237}Np produces either gamma or conversion electrons (CE) emission. G4RadioactiveDecay computes the emission of these particles by reading the theoretical CE probabilities included in Photon Evaporation database. In a previous paper [16] different nuclear data sets for the de-excitation of ^{237}Np were shown. In this way several simulations of the nuclear decay of ^{237}Np were carried out by changing the CE probability data for the L_1, L_2, L_3 and N+ shells from experimental measurements of conversion electron spectroscopy [17,18] or from

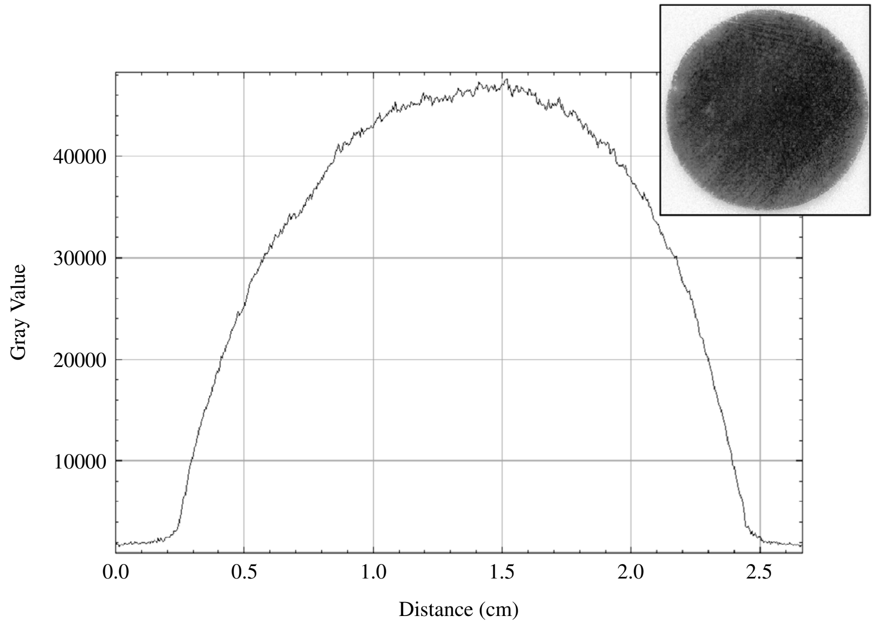

Fig. 2. Autoradiography and active area of the ^{241}Am standard source.

Table 1
Electron conversion probabilities for L_1, L_2, L_3 and N+ shells of the nuclear de-excitation of ^{237}Np using different databases.

Database	L_1	L_2	L_3	N+
PhotonEvaporation 3.2	0.1329	0.2638	0.3453	0.06413
DeVol [17] y BriCC [18]	0.2496	0.2292	0.0845	0.06331
NUCLEIDE [19]	0.84	0.84	0.84	0.06331

NUCLEIDE database [19], into the official Geant4 Photon Evaporation database (see Table 1).

Finally, the GEANT4 simulation histogram containing the calculated spectrum obtained was thus processed by an algorithm written in C++ in the ROOT analysis framework [20].

3. Results and discussion

It is of the utmost importance to know accurately the characteristics of the ^{241}Am standard source and the detector setup in order to mimic the experimental response as closely as possible through the GEANT4 simulation.

First, in order to accurately model the experimental pulse height distribution of the energy deposited by the charged particles, the statistical variation in the detector signal must be considered for the simulations. This was achieved by using the well-known experimental model of energy resolution described by Knoll [13]. The energy resolution of a detector (measured as the Full Width Half Maximum, FWHM) can be modelled by the square sum of three terms [21]:

$$\mathrm{FWHM}^2 = \mathrm{FWHM}^2_{\mathrm{e\text{–}h}} + \mathrm{FWHM}^2_{\mathrm{noise}} + \mathrm{FWHM}^2_{\mathrm{ion\text{–}exc}}.$$

The first term is inherent to the detector material; it is given by the expression $\mathrm{FWHM}_{\mathrm{e\text{–}h}} \approx 2.35\sqrt{\mathrm{FE}\varepsilon}$, where F is the Fano factor, E is the energy of the incident charged particle, and ε is the required energy to create an electron–hole pair in Si (0.1161 ± 0.0001 eV [22]). The final value obtained for this term was 3.606 ± 0.001 keV. The second term describes the electronic noise as a constant value added to every pulse processed by the electronic chain. This term was measured using a built-in pulser included in each chamber, obtaining a value of 18.1 ± 1.4 keV. The third term is due to the fact that the charged particles lose their energy through ionization and excitation processes with Si electrons [23,24]. The remaining energy is used in collisions with silicon atoms following a not Gaussian distribution. This ionization–excitation factor was calculated by GEANT4 simulation, using the non-ionizing energy-loss (NIEL) computation functionality of GEANT4 (available since v9.1), obtaining a value of 9.2 ± 1.1 keV. Finally, the calculated detector resolution was 20.6 ± 2.5 keV, deviating 3.5% from the experimental resolution. The final simulated spectrum was obtained by convolving with the calculated detector resolution through a ROOT algorithm.

The next step was the characterization of the ^{241}Am standard source, specifically its active area and thickness dimensions. The diameter of the active area was estimated at 2.2 ± 0.1 cm through the use of the imaging plate technique and ImageJ software (see Fig. 2). Furthermore, the autoradiograph showed that the radioactivity was uniformly distributed over the surface of the stainless-steel disc.

Finally, the last step of the characterization process consisted of determining the thickness of the detector dead layer, and the ^{241}Am source thickness. The detector dead layer was estimated to be less than 50 nm by the manufacturer. As for the ^{241}Am source thickness, it estimation was obtained through two measurements. In the first one, the source was measured parallel to the PIPS detector surface at a source-to-detector distance of 30.0 cm, and in the second, the source was rotated an angle of 46.4°with respect to the horizontal. In this way, the alpha particles have undergone more energy losses inside the source resulting in a displacement of any alpha peak towards the low energy spectrum zone. The experimental displacement of the peak maxima when rotating the detector was about 24 keV [25].

Next, the values of dead layer and source thickness were varied in GEANT4 simulations till the agreement between experimental and simulated spectra was reach for both angles (see Figs. 3 and 4). The simulated displacement of the peak maxima when rotating the detector was about 20 keV optimizing the values for dead layer and source thickness to 40 nm and 50 nm respectively. These optimized values being comparable with results found in the literature [26–28].

Once the characteristics of the ^{241}Am standard source and the detector setup had been adjusted, the efficiency for different source-to-detector distances was computed using those previous optimized values. The MC simulated spectrum with a source-to-detector distance of 7.2 cm closer to the detector (see Fig. 5) showed slight discrepancy with the experimental spectrum. The experimental and simulated FWHM are 19.9 keV and 20.5 keV respectively. Additionally, as a measure of the

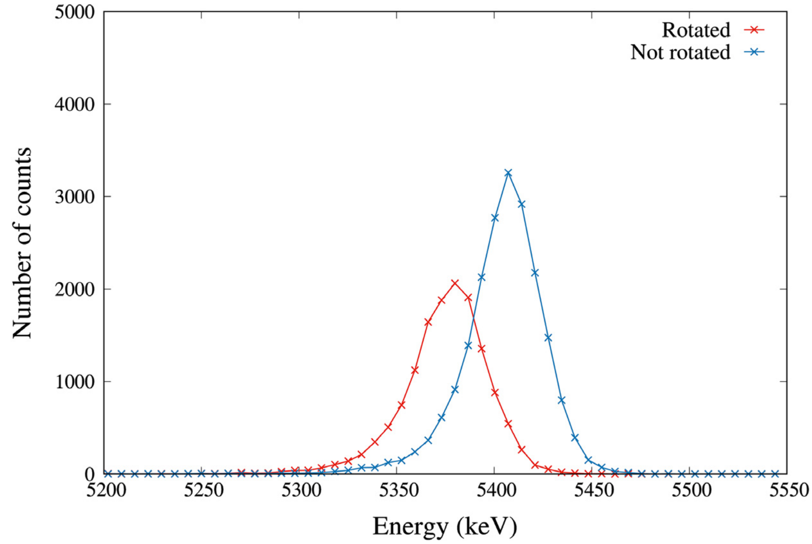

Fig. 3. Experimental spectra of non-rotated (blue) and 46.4 deg rotated (red) ^{241}Am standard source at a source-to-detector distance of 30.0 cm. The energy shift of the peak maxima is about 24 keV. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

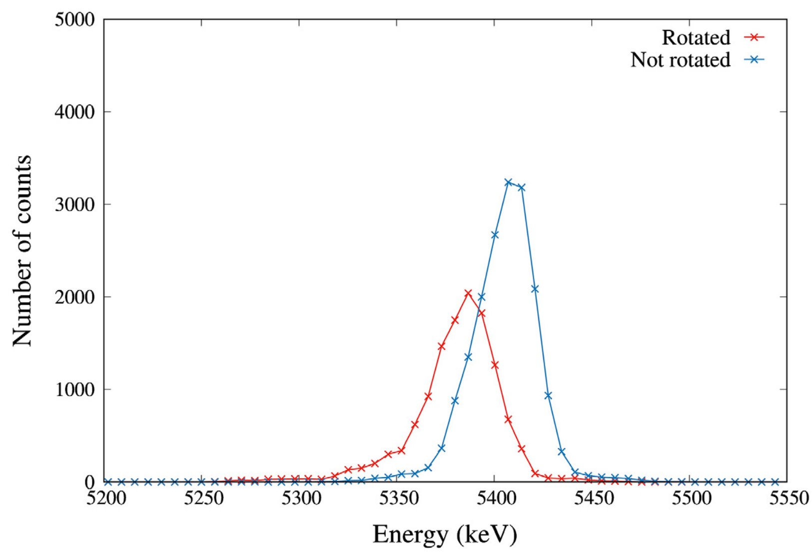

Fig. 4. Simulated spectra of non-rotated (blue) and 46.4 deg rotated (red) ^{241}Am standard source at a source-to-detector distance of 30.0 cm. The energy shift of the peak maxima is about 20 keV. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

level of accuracy of the GEANT4 simulation under consideration, a chi-square test was performed in order to compare against experimental data by following the next expression [29]:

$$\chi^2 = \frac{1}{\nu} \sum_i \frac{\left(y_i^{\text{exp}} - y_i^{\text{sim}}\right)^2}{\sigma_{i,\text{exp}}^2 + \sigma_{i,\text{sim}}^2}$$

where ν is the number of degrees of freedom, y_i^{exp} and y_i^{sim} are the counts of the ith bin for the experimental and simulated spectrum, respectively, and σ its associated uncertainty. A chi-square value of 6.63 was obtained during the comparison of the experimental and simulated spectra shown in Fig. 5.

One of the possible reason for the slight discrepancies could be caused by inaccurate CE coefficients included in GEANT4 PhotonEvaporation database [30]. The GEANT4 database of the ^{241}Am decay was modified by including the CE probabilities for the L_1, L_2, L_3 and N+ shells from experimental measurements of conversion electron spectroscopy [17,18] and from NUCLEIDE database [19], into the Photon Evaporation database as stated in previous section. However, no drastic positive change for a better chi-square value has occurred (see Table 2).

Table 2
Chi-square test for ^{241}Am standard source simulation using different databases.

Database	χ^2
PhotonEvaporation3.2	8.50
DeVol [17] et al. y BriCC [18]	7.68
NUCLEIDE [19]	6.63

Additionally, in order to check the GEANT4 simulation performance and accuracy, the counting efficiency was calculated for ^{241}Am standard source located at different source-detector distances. A good agreement between calculated and experimental efficiencies was obtained (see Table 3).

Finally, a validation of the proposed GEANT4 simulation was carried out. Non-destructive direct alpha spectrometry was performed over the uranium natural source (see Section 2.1. for a further description). On the other hand, the dimensions and composition of the uranium source were used as input variables for GEANT4 simulation. The experimental

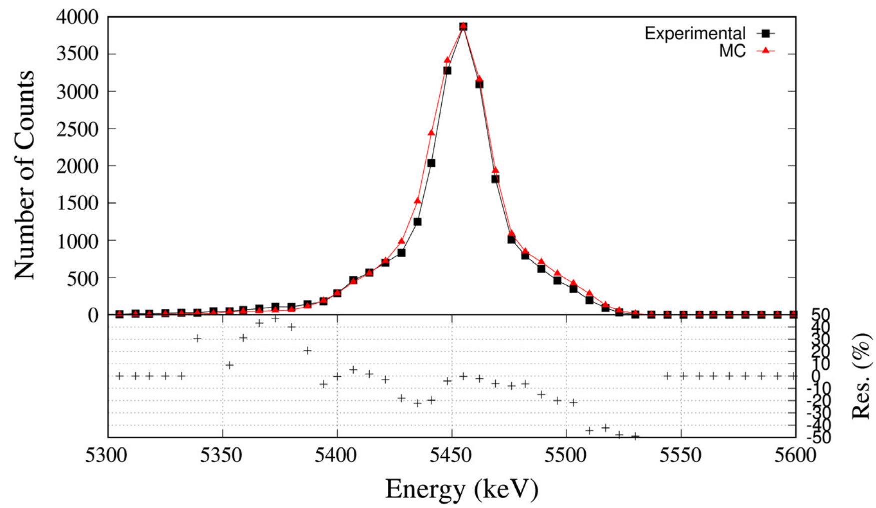

Fig. 5. Simulated (red) and experimental (black) spectra for the ^{241}Am standard source located at a source-to-detector distance of 7.2 cm. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

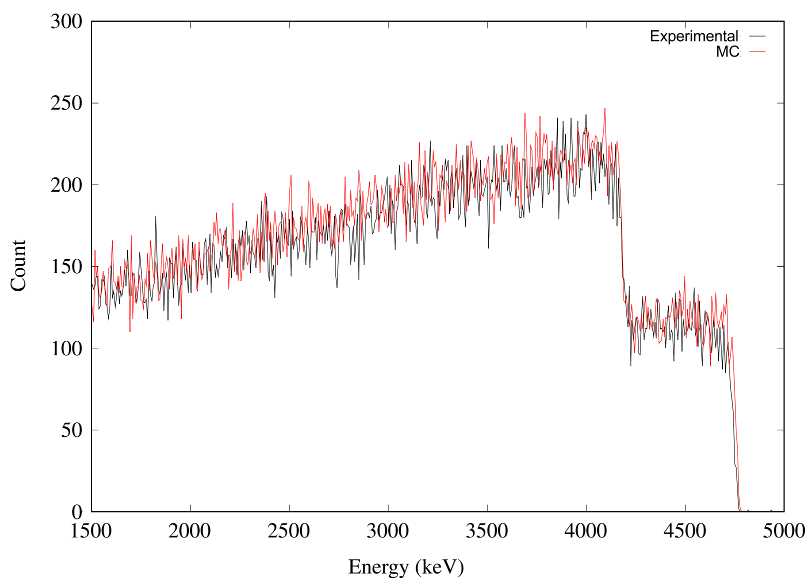

Fig. 6. Simulated (red) and experimental (black) spectra for a natural uranium source. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Table 3
Experimental (ε_{exp}) and calculated efficiencies (ε_{exp}) for different source-to-detector distances.

Source-detector distance (mm)	ε_{exp}	ε_{MC}	Deviation (%)
7.2 ± 0.1	0.220 ± 0.002	0.227 ± 0.008	3.2
18.0 ± 0.1	0.0720 ± 0.0003	0.0715 ± 0.0007	0.7
30.0 ± 0.1	0.0331 ± 0.0008	0.0326 ± 0.0005	1.5

and simulated spectra are shown in Fig. 6. It can be seen that there was a good agreement between both of them obtaining a chi-square of 1.16. Additionally, the detection efficiency was calculated using the simulated spectrum, and together with the number of total counts in the energy region, a uranium activity of 93.3 ± 0.4 kBq was estimated, with a relative deviation of 5.9% from the experimental activity.

4. Conclusions

We have simulated the response of a PIPS detector to alpha-emitter sources with the GEANT4 toolkit. Different factors affecting the accuracy of the simulation were examined, such as a modelling of the energy resolution (FWHM), the characterization of the source (active area and thickness), or the optimization of the detector dead layer.

Experimental spectra of a natural uranium source were compared with the predictions given by the proposed simulation, showing a good agreement. This validation opens up new perspectives for GEANT4 applications in direct alpha spectrometry without radiochemical treatment of the sample (hot particles characterization), metrology through the study of coincidence summing between emitted alpha particles and electrons, or the corrections for backscattering and self-absorption, among other areas.

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