RAD³ PROCEEDINGS

CALIBRATION OF HPGE DETECTORS FOR ENVIRONMENTAL SAMPLES USING GEANT4 SIMULATION

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Abstract. Determination of full energy peak efficiency is one of the most important tasks that have to be performed before gamma spectrometry. Calibration of the measurement system for measuring environmental samples poses a special challenge to the laboratory. Many different approaches to this task have been developed and examined. One of the most detailed is GEANT4, a Monte Carlo simulation toolkit developed for wide variety of applications. The aim of this paper is to apply GEANT4 simulation for calibration of three HPGe detectors, for measurement of aerosol, plant and coal-like environmental samples. The detectors were modeled using the certificate provided by the manufacturer. The samples chosen for the simulation were secondary reference materials produced in the Laboratory for the purpose of experimental calibration. The efficiencies obtained using a simulation were compared with experimental results, in order to evaluate the trueness of the results. Measurement uncertainties for both simulation and experimental values were estimated in order to see whether the results of the realistic measurement fall within acceptable limits.

Key words: HPGe, calibration, GEANT4

1. INTRODUCTION

One of the main problems in quantitative gamma-ray spectroscopy is the determination of detection efficiency for different energies, different source-detector geometries and different composition of voluminous sources. Experimental approach utilizes standard sources with composition, density and geometry as close to the samples of interest, as possible. The direct measurement of different calibration sources containing isolated γ-ray emitters within the energy range of interest, and their subsequent fitting to a parametric function, yields the best results. However, when the energy interval is broad, it requires a large number of primary standards, implying a high financial cost, a long counting time and much work in preparing samples. This problem is especially pronounced when environmental samples are of interest due to their diversities in composition and structure [1]. This problem has been partially solved by using procedures based on the Monte Carlo simulations, developed in order to complement the experimental calibration procedures used in gamma-ray measurements of environmental samples [1]. Monte Carlo methods are based on determining full energy peak (FEP) efficiency by simulating all relevant physical processes taking place along the path of a photon emitted by the source. Since no approximations are needed, there is no limitation on the source–detector configuration. The best known and the most sophisticated Monte Carlo simulation kit is the CERN GEANT4. Monte Carlo simulation, based on GEANT4 simulation package has been developed to obtain the response of germanium detectors, with the aim to reproduce experimental spectra of detectors in wide range of applications in gamma spectroscopy measurements [2].

The aim of this paper is to apply GEANT4 simulation for calibration of three HPGe detectors, for measurement of aerosol, plant and coal-like environmental samples. The detectors were modeled using the certificate provided by the manufacturer. The samples chosen for the simulation were secondary reference materials produced in the Laboratory for the purpose of experimental calibration. The efficiencies obtained using the simulation were compared with the experimental results, in order to evaluate the trueness of the results. Measurement uncertainties for both simulated and experimental values were estimated in order to see whether the results agree within the acceptable limits.

2. MATERIALS AND METHODS

The detectors considered for the Monte Carlo simulations were the ones commonly used in our

laboratory: two p-type detectors with relative efficiencies of 20% (named Detector 1) and 50% (named Detector 3), and one n-type detector with relative efficiency of 18% named Detector 2.

GEANT4 allows the description of an experimental setup represented by a structure of geometrical volumes filled by specified materials. In the simulation, germanium detectors, together with the whole detector assembly, are constructed in great detail, according to the manufacturer's data specifications, while the bulletisation and depth and diameter of the central cavity were estimated based on the known dimensions of other detectors produced by the same manufacturer [3, 4]. The characteristics of the detectors are presented in Table 1.

Sample dimensions are also measured with care, and sample volumes are constructed in the simulation code accordingly. Primary photons are generated in the sample volumes with uniformly random positions and momentum directions in full space (4π srad). Each photon undergoes interaction processes and deposits energy in the detector or it traverses the detector (or the whole system) without interaction. All types of relevant interactions of photons and electrons/positrons with matter are taken into account, using low-energy data packages (dataset G4EMLOW6.32, model G4EmLivermorePhysics); tracking of the particles is performed down to below 10 keV. Distribution of the photon energy deposited in the detector's active volume gives spectral response of the detector, i.e. simulated spectrum. From the simulated spectrum one can obtain the simulated FEP intensities, and from this, derive the simulated detector efficiency for a given energy.

In order to verify the results obtained by GEANT4, simulated efficiencies were compared to the experimental values. Experimental efficiency calibration is readily performed in the Laboratory for Environment and Radiation Protection of the Institute for Nuclear Sciences Vinca. For the purpose of calibrating detectors for the environmental samples, a set of secondary reference materials was produced in order to serve as calibration samples. These samples were produced by spiking the chosen matrices with the certified radioactive mixture solution ER X 9031-OL-426/12 issued by Czech Metrological Institute, Inspectorate for Ionizing Radiation. The radioactive solution contained following radionuclides: [241]Am, [109]Cd, [139]Ce, [57]Co, ^{60}Co, ^{137}Cs, [203]Hg, [113]Sn, [85]Sr and [88]Y, with the energies that span from 59keV to 1898keV with total activity of 1342 Bq at reference date 31.08.2012. Matrices and geometry of secondary reference materials were chosen in a manner that best mimics the realistic situation. These matrices were aerosol, charcoal and mineralized grass. Aerosol was placed in vial, mineralized plant in cylindrical container of 120 ml with 6g and 26g of the matrix, named grass 1 and grass 2 respectively and charcoal was placed in cylindrical containers of 120 and 250 ml, named charcoal 1 and charcoal 2 respectively.

The geometrical characteristics of the detectors were optimized by varying the parameters of the detector and conducting a simulation for water matrix [5]. After that, optimized parameters were used in the simulation.

The secondary reference materials were prepared by applying the procedure with activated carbon, as defined in [6]. The standardized solution had been diluted to the adequate specific activity by adding carrier solution. This radioactive solution was homogeneously mixed in the bulk matrix materials previously mechanically prepared. Detailed procedures were presented in [7].

3. RESULTS AND DISCUSSION

The measurement was conducted on all three detectors for the duration of 60000s. Background radiation was measured by placing non spiked sample of the same geometry and composition as the secondary reference material and background spectrum was subtracted. After analyzing the spectra, the efficiency at the given energy was calculated according to:

$$\varepsilon = \frac{N \cdot C(E)}{t \cdot P_\gamma \cdot A} \qquad (1)$$

where N represents the net count at the energy E, t is counting time, P_γ is emission probability, $C(E)$ is true coincidence correction factor and A is source activity on the given energy, with the decay correction.

Coincidence correction factors were calculated using EFTRAN software [8]. The corrections ranged from 0.1% to as much as 9% for higher energies. Coincidence was more prominent for Detector 2 due to its beryllium window and the fact that it is an n-type detector. For Detector 1 and Detector 3, corrections were needed only for energies of [88]Y and [60]Co.

Relative measurement uncertainty for the experimental values was calculated according to the following Equation:

$$u(\varepsilon) = \sqrt{(\delta A)^2 + (\delta N)^2 + (\delta M)^2 + (\delta P)^2} \quad (2)$$

For aerosol, combined measurement uncertainty was 3% and 5% for grass 1, grass 2, charcoal 1 and charcoal 2.

In case of GEANT4 simulation, combined relative uncertainty was estimated according to following consideration. The main variables that have been input into the simulation are 9 characteristic dimensions of the detector (crystal diameter and length, crystal cavity diameter and length, top and side dead layer, end cap diameter, window thickness and window to crystal gap) and 4 characteristics related to the sample (sample volume, sample and container material, namely density and chemical composition, and container to absorber gap). For these variables, except chemical composition of sample and container, the uncertainty can be estimated to be 1% for geometry of crystal and container to absorber gap and 10% for window thickness and window to crystal gap. In order to minimize the discrepancy between simulated and measured values, bulletization, dead layer and window to crystal gap were varied in the simulation, and the uncertainty was lowered to estimated 1%. The chemical composition of the container is well defined, but for the sample the situation is more complicated and poor knowledge of the chemical composition of aerosol and

grass can be the source of larger uncertainty. This is estimated to be 10%. Since the uncertainty of the simulation, $u_{simulation}$ is calculated according to

$$u_{simulation} = \sqrt{\sum (\delta x_i)^2} \qquad (3)$$

where δx_i represents relative uncertainty of the value x_i, overall combined uncertainty of the simulated results is estimated to be 5.3% for aerosol and grass and 6%
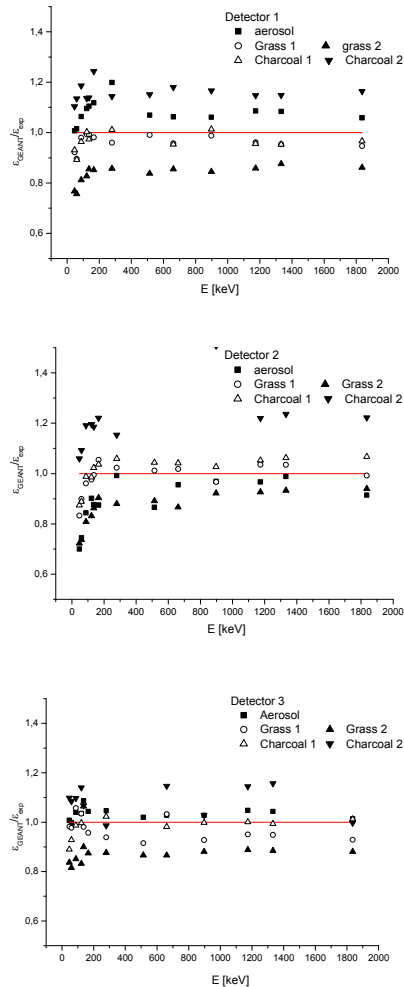

Figure 1. The relative disrepancies between simulated and experimental efficiencies.

for charcoal.

Simulation was conducted for the same secondary reference materials and efficiency was calculated using Equation (1).

The relative discrepancies between simulated and experimental values are presented in Figure 1 in form of $\varepsilon_{GEANT}/\varepsilon_{exp}$ ratio. As it can be seen from Figure 1, the discrepancies ranged from 0.7-19.9% for Detector 1, from 0.8-25.5% for Detector 2 and from 0.7-18.5% for Detector 3. Although the ranges are wide, the largest discrepancies are for low energies (46 and 59 keV), while for the rest of the spectrum, average discrepancy was 5-10%. Similar to all Monte Carlo

based simulations, [9, 10, 11], the largest discrepancies are for the lower energies. It should be noticed that GEANT4 simulation has great sensitivity to definition of the material through which the photon traverses, and that in Monte Carlo simulation, errors in definition of the matrix (such is the case for grass and aerosol) can influence the result for lower energies, (due to self-absorption correction factors) resulting in up to 15% discrepancy [5, 11]. This is especially noticeable in the case of Detector 1 and Detector 2 where the discrepancy at lower energies exceeds the measurement uncertainty. The homogeneity of the sample can also influence. After the preparation of the secondary reference material, the homogeneity was checked by measuring the random part of the spiked matrix. The measurement showed that the inhomogeneity of the samples was below 2% [6]. It was shown in [8, 9] that thickness and homogeneity of the detector dead layer, as well as the density of the detector material, also can have significant influence on the result of the simulation. Since both Detector 1 and Detector 2 are old, the homogeneity of dead layers is compromised resulting in larger discrepancies at lower energies. As it can be seen that is not the case for Detector 3 which is newer (2008.). It can be noticed that grass 1 shows better agreement with the experimental values than grass 2, although the only difference is the sample filling height. The results for grass 2 show unexplained bias for all three detectors, which require the repetition of both measurement and simulation. Discrepancies noticed for charcoal 2 in Detector 1 are due to uncertainties regarding the sample filling height of the matrix, which leads to erroneous selection of the attenuation coefficients. However, the other values show good agreement with experimental efficiencies. For the purpose of environmental samples measurement, this proves to be quite satisfactory, since it does not exceed measurement uncertainty of experimental values.

Table 1 Detector characteristics

		Detector 1	Detector 2	Detector 3
Geometry and type of detector		Closed coaxial - p type Canberra	Closed reverzibile coaxial - n type Canberra	Closed coaxial - p type Canberra
Relative efficiency		20 %	18 %	50 %
Resolution [keV]	on 122 keV	0,850	0,759	1.00
	on 1332 keV	1,8	1,69	1,9
Peak/Compton ratio		51:1	56.1:1	65:1
Crystal diameter [mm]		49,5	48	65
Crystal lenght [mm]		56,5	48,5	67

Crystal to window Distance [mm]	5,5	5	5
Entry window	Al	Be	Be
Central void (diameter x lenght) [mm]	10 x 40	10 x 40	10 x 55

4. CONCLUSION

This paper presents the results of efficiency calibration of three HPGe detectors using GEANT4 simulation. The efficiencies obtained by simulation were compared to the experimental values, obtained by measuring different secondary reference materials. The results showed good agreement with the experimental results, with the discrepancies ranging from <1% to about 20%. The largest discrepancies were noticed for low energies, for all investigated secondary reference materials. Also, the discrepancies are larger for grass 2 and charcoal 2 in case of p-type detectors. This suggests that the definition of the sample chemical composition and geometry have to be revised. Most of the results were within the uncertainty limits proving that simulated efficiencies can be utilized for measurement of the environmental samples.

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