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Modeling of a HPGe well detector using PENELOPE for the calculation of full energy peak efficiencies for environmental samples

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ABSTRACT

When determining the activity concentration of radionuclides using gamma-ray spectrometry the Full Energy Peak Efficiency (FEPE) for the energies of interest must be known. Determination of the FEPE can be made by means of either experimental calibration or theoretical–mathematical methods, such as Monte Carlo simulations. Given the difficulties related to experimental calibration and improvements in the capabilities of modern computers, Monte Carlo simulation is an increasingly widely used alternative, but requires an accurate model of the detector. The purpose of this work is to generate and validate a computational model, based on Monte Carlo simulation, of an HPGe well detector that permits the performance of FEPE calculations with appropriate precision and accuracy for the measurement of environmental samples. To achieve this, an optimization methodology is applied to the model that minimizes the differences between a set of computational FEPEs and a set of experimental FEPEs used as a benchmark. The resulting optimized model is used to calculate computational FEPEs for 25 different samples with different reference materials and sample heights, which are measured by means of the well detector modeled here. To validate the optimized model, the abovementioned computational FEPEs are used during the calibration of the corresponding spectra, to enable the subsequent comparison of the results of the analyses with the expected values. The measured activities differ from the reference values by less than 10% in most cases and are compatible with these considering the uncertainties involved, thus confirming the validity of the model.

1. Introduction

Gamma-ray spectrometry is a technique commonly used for the identification and quantification of a wide range of radionuclides by means of their direct gamma emissions or those of their progeny. HPGe detectors are currently the most widely used gamma-ray detectors due to their various advantages, of which high energy resolution is the most important; this permits high-precision during the identification of the peaks. For measurement of the radioactive content of environmental samples of low volume and activity in laboratory, HPGe well detectors are the best option due to their high detection efficiency, as the counting geometry approaches 4π [1]; this implies improvements in both the uncertainty of the results and the Minimum Detectable Activity. More specifically, the use of this type of detector is recommended when samples of only a few grams of organic matter are to be analyzed for multidisciplinary studies, including radiological techniques [2,3]. These aspects motivated us to acquire and setup a spectrometry system of this type.

When the activity concentration of radionuclides in a sample is intended to be quantified by gamma-ray spectrometry, the Full Energy Peak Efficiency (FEPE) for the energies of interest must be determined. FEPE determination can be made either by means of experimental calibration, or by theoretical–mathematical methods such as Monte Carlo simulations. The experimental efficiency calibration [4–8] requires the preparation of standard sources using reference materials with known activities and with the same measuring geometry as the sample to be measured. Materials with very similar densities and chemical compositions need to be used during the experimental calibration if self-attenuation corrections are to be avoided, especially at low energies [1]. Moreover, when environmental samples are studied, the preparation of standard sources that are very similar may be a process of considerable complexity, involving appreciable economic and time costs.

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The Monte Carlo method has been applied in the calculation of FEPEs for several decades [9,10] and is now an increasingly widely used alternative [11–16] in view of the difficulties related to experimental calibration and the enhanced capabilities of modern computers. However, a computational model of a detector with a similar response to the real one must be utilized in order to obtain reliable FEPEs by direct Monte Carlo simulation. For this reason, many recent publications have focused on the modeling of gamma-ray detectors [17–20], also known as detector characterization, including two papers by the current authors [21,22], in which automatic methodologies for modeling HPGe detectors were developed, and which were validated using two real coaxial detectors in a third publication [23].

In the first of these papers [21], the characterization methodology involves the use of the Differential Evolution (DE) algorithm [24] for the optimization of a detector model built using the PENELOPE Monte Carlo simulation code [25]. This optimization starts from a sufficiently accurate set of reference FEPEs obtained for a specific sample-detector arrangement and sample material. In the current paper, this methodology is applied to an HPGe well detector used for the measurement of environmental samples. The computational model of the detector should allow the calculation of FEPEs with acceptable precision and sufficient accuracy for efficiency calibration with this type of sample, irrespective of whether the dimensions considered in the model are an exact match for the real dimensions of the detector.

2. Material and methods

2.1. Instrumentation

In this work, a Canberra HPGe well detector (model GCW4023) is used. This detector has a relative efficiency of 40%, a nominal FWHM of 1.2 keV at 122 keV and 2.0 keV at 1.33 MeV and a peak-to-Compton ratio of 60.8:1 at 1.33 MeV. The detector is connected to a DSA-1000 Canberra multichannel analyzer and is located within an iron shield with a thickness of 15 cm. The acquisition and analysis of the gamma-ray spectra is performed using the Genie 2000 software package [26].

A Radon Scout detector from SARAD GmbH [27] was been used to determine the escape of ^{222}Rn from samples of one of the reference materials used in this work, due to its relatively high activity concentration of ^{226}Ra and exhalation rate (see Section 2.4). This detector and the samples were placed within an accumulation radon-tight chamber, made of methacrylate with an internal volume of 0.185 ± 0.006 m^3 and walls 10 mm thick [28].

2.2. Samples of reference materials used

Reference materials IAEA RGU-1, IAEA RGTh-1, IAEA RGK-1, IAEA 447 and IAEA 448 were used for the generation and validation of the model of detector. The first three of these are meant for the experimental calibration of gamma-ray spectrometers for potassium, uranium and thorium [29]. IAEA 448 [30–32] and IAEA 447 [32–34] are environmental samples with certified activity for several radionuclides of interest to environmental radioactivity laboratories. Both the chemical composition and the reference activity concentrations were obtained from the documents referenced above.

Five samples were prepared for each of these reference materials by placing the materials directly into the beakers with different sample heights (10, 20, 30, 40 and 51 mm) and densities ranging from 1 to 2 g/cm^3. All the samples were prepared in cylindrical polypropylene beakers with an internal diameter of 11.5 mm, an internal height of 51 mm and a thickness of 1 mm. In the case of materials containing ^{226}Ra, once shut and sealed, the beakers remained undisturbed for at least four weeks (approximately seven times the half-life of ^{222}Rn) before performing each spectrum acquisition, in order to attain radioactive equilibrium between ^{226}Ra and ^{222}Rn. In the specific case of the IAEA 448, we carried out experimental checks that, even sealing with aluminum tape (as we perform routinely in the laboratory), a non-negligible escape of radon was noticed. The procedure used to correct the results for this effect is explained in Section 2.4.

Table 1
Experimental FEPEs used as a reference during the optimization process of the computational model of the detector (sample height = 51 mm).

Radionuclide	En (keV)	$FEPE$	Uncertainty (1σ)
^{210}Pb	46.5	0.272	0.003
^{234}Th	63.3	0.350	0.008
^{235}U + ^{223}Ra	143.8	0.365	0.009
^{236}Ra + ^{235}U	186.2	0.294	0.003
^{214}Pb	295.2	0.1948	0.0014
^{214}Pb	351.9	0.1652	0.0011
^{234m}Pa	1001.0	0.0659	0.0025
^{214}Bi	1764.5	0.0392	0.0003

2.3. Reference FEPEs

Calibration of the detector in terms of energy and resolution was performed using a standard point source of ^{22}Na and ^{155}Eu, provided with the detector by Canberra. As a result, we obtained the corresponding calibration functions (energy in keV): $energy = 0.3787 + 2.1212 \cdot centroid$ and $FWHM = 0.9672 + 0.0254 \cdot \sqrt{energy}$. The experimental FEPEs used as a reference during the optimization process for the computational model of the detector were obtained by analyzing a RGU-1 sample with a height of 51 mm (i.e. the beaker completely filled). Fig. 1 shows the corresponding spectrum acquired. Table 1 shows the gamma lines chosen, the radionuclides emitting these and the corresponding experimental FEPEs with their uncertainties. These eight gamma emissions were selected because they are virtually free of coincidence summing effects and of considerable interferences with other peaks of the spectrum and are sufficiently intense in order that the peaks stand out sufficiently from the continuum. A sample height of 51 mm was chosen to obtain the reference FEPEs, since this height gives the highest counting rate and therefore the lowest counting uncertainty; in addition, with the beaker completely filled, potential uncertainties in the sample height are eliminated.

The experimental FEPEs and uncertainties are determined using Eqs. (1) and (2), where C represents the net counts observed in the relevant full energy peak (subtracting both the Compton Continuum and the laboratory background, using GENIE2000 [26]), $[Ac]$ the activity concentration of the relevant radionuclide, P_γ the corresponding probability of the gamma emission, t the acquisition time, m the sample mass, and $u_C, u_{[Ac]}, u_{P_\gamma}, u_m$ are the corresponding uncertainties. The values of P_γ and the uncertainties used in this paper were obtained from the NationalNuclearDataCenter [35].

$$FEPE = \frac{C}{[Ac] \cdot P_\gamma \cdot t \cdot m} \tag{1}$$

$$u_{FEPE} = FEPE \sqrt{\left(\frac{u_C}{C}\right)^2 + \left(\frac{u_{[Ac]}}{[Ac]}\right)^2 + \left(\frac{u_{P_\gamma}}{P_\gamma}\right)^2 + \left(\frac{u_m}{m}\right)^2} \tag{2}$$

2.4. Determination of the escape of radon for the case of IAEA 448

The reference material IAEA 448 contains relatively high concentrations of ^{226}Ra (19 kBq/kg), which, together with the high exhalation rate of this material, means that the escape of radon from the beaker is a considerable source of error when measuring the ^{226}Ra concentration through the gamma emissions corresponding to ^{222}Rn progeny (^{214}Pb and ^{214}Bi) [30]. For this reason, a correction factor was determined for the escape of radon when measuring this material.

To determine this correction factor, 20 beakers, identical to those used for gamma-ray analysis, were completely filled with IAEA 448 with a total mass of 81.2 ± 0.1 g. These 20 samples were placed within the accumulation chamber described in Section 2.1, with the Radon Scout [27], in order to measure the ^{222}Rn concentration (activity per unit volume) accumulated in the chamber over a period of one month, during which time the detector recorded the ^{222}Rn concentration every three hours. The recorded values were fitted to the curve shown