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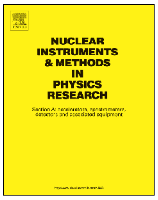

Validation of an efficiency calibration procedure for a coaxial n-type and a well-type HPGe detector used for the measurement of environmental radioactivity

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ABSTRACT

To obtain reliable measurements of the environmental radionuclide activity using HPGe (High Purity Germanium) detectors, the knowledge of the absolute peak efficiency is required. This work presents a practical procedure for efficiency calibration of a coaxial n-type and a well-type HPGe detector using experimental and Monte Carlo simulations methods. The method was performed in an energy range from 40 to 1460 keV and it can be used for both, solid and liquid environmental samples. The calibration was initially verified measuring several reference materials provided by the IAEA (International Atomic Energy Agency). Finally, through the participation in two Proficiency Tests organized by IAEA for the members of the ALMERA network (Analytical Laboratories for the Measurement of Environmental Radioactivity) the validity of the developed procedure was confirmed. The validation also showed that measurement of ^{226}Ra should be conducted using coaxial n-type HPGe detector in order to minimize the true coincidence summing effect.

1. Introduction

Many approaches have been proposed in the literature for efficiency calibration of HPGe detectors when environmental samples are measured [1–3]. Set of standard point sources having single energy emissions, standard solution of mixed radionuclides and Certified Reference Materials commercially available, are commonly employed. In most of these cases, the activity measured by this method has to be corrected for summation effects induced by photons emitted in coincidence and also for self-absorption when the measured sample has a different matrix (density or composition) than the source used in the calibration process. A direct calibration can be performed using standard radioactive sources of the same geometrical dimensions, density, and chemical composition, compared with the samples of interest. However, standards are not often available for all environmental matrices or for all radionuclides of interest.

In addition, theoretical and computational methods have also been employed for efficiency calibration, coincidence-summing and self-absorption corrections. In several studies have been used general and specific software based on Monte Carlo (MC) codes like Geant, MCNPX, EFFTRAN or DETEF [4–7]. A good agreement between experimental and calculated data can be reached using these codes; in addition the working time can be reduced considerably. In these cases, main limitations reside in the precise knowledge about the characteristics of the experimental geometry and sample compositions [8]. Generally, the efficiency values obtained experimentally and by MC simulation based on nominal values of the parameters supplied by the manufacturer show significant differences due to the inaccuracy in some critical parameter like the thickness of the dead layer or the active volume. The optimization of these parameters can result in a substantial decrease of the deviations between the experimental and calculated values [9,10]. However, even when precise geometrical data are available, it is necessary to refine the model by feeding it back with experimental results when accuracy is desired. This is because some parameters involved in the detector response cannot easily be assessed. They include the distribution of the electrical field in the crystal, its mounting and dimensions and properties of the dead layers [11,12].

One effective procedure to overcome these difficulties is to use an efficiency transfer function from reference geometry to other source configurations, using MC calculations and experimental measurements. This procedure consists of calculating the full energy peak efficiency (FEPE) by an energy dependent transfer

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factor, which is derived by comparing the direct calculated FEPEs with the source experimental values at a reference position. In the literature, different authors have reported differences below 5% after using the transfer function [13,14]; even when there was no adjustment of parameters of the detectors.

Independent of the chosen calibration method (experimental or computational), there is a group of limitations which will be required to overcome. However, the combination of these techniques can be a potential tool as a practical and cheap method for routine measurement purpose in many laboratories. The goal of this study is to develop a simple procedure for efficiency calibration of two different HPGe detectors, complementing experimental and MC simulation methods. The main advantages of this approach is that it can be applied to coaxial detectors as well as well detectors and it can be used for different environmental matrices.The method was verified measuring several IAEA reference materials and finally validated through the participation in two ALMERA Proficiency Tests organized by IAEA for the ALMERA network members.

2. Materials and methods

2.1. Detectors

Two high purity germanium (HPGe) detectors were used for experimental measurements. Detector 1 (D1) was a coaxial n-type detector (model NGC 3019, from DSG Detector Systems GmbH) with epoxy–carbon window and 31.5% of relative efficiency. It was coupled to an electronic chain, including a multichannel analyzer type TMCA. Detector 2 (D2) was a well-type HPGe detector (model EGPC100 P-15, from Canberra) with an absolute efficiency of 12.1% at 661 keV. The data acquisition system of this detector consists of a PSC822 preamplifier, Canberra amplifier model 7245 and electronic card MCA 5000 which includes a 7602 ADC with 8192 channels and InterFast multichannel analyzer. Both detectors are surrounded by a cylindrical low-background chamber made with the following elements from outside to the inner region: 240 mm of steel, 37 mm of lead, 1 mm of aluminum and 1 mm of copper. In both detectors the gamma spectra were recorded and analyzed using Winner™ 6.0 software. The detectors resolution and energy calibration is periodically verified for stability using a set of point sources (^{241}Am, ^{137}Cs, ^{60}Co and ^{226}Ra).

2.2. Monte Carlo simulation

The first step of the calibration procedure was to obtain the efficiency calibration curves for both detector (D1 and D2) by Monte Carlo simulation methods. It was employed the code MCNPX 2.6. The efficiency response for both detectors have been previously reported in the studies [6,15]. Nominal values of the parameters supplied by the manufacturer are described in those studies.

For detector D1 the Monte Carlo efficiency transfer method was applied using directly the manufacturer supplied data in all MC calculations. The efficiencies were calculated using the expression $\epsilon_x = \epsilon_{ref}\left(\epsilon_x^{MC}/\epsilon_{ref}^{MC}\right)$ where ϵ_x is the efficiency for a particular geometry and energy, ϵ_{ref} is the experimental efficiency for a reference case, and ϵ_{ref}^{MC} and ϵ_x^{MC} are calculated efficiencies (via Monte Carlo) for the reference case and the geometry of interest, respectively. The characteristics of the reference source and source-detector configuration are described in [6]. We used the detector model described in [6] to compute the efficiencies of the samples that were measured here (ϵ_x^{MC}) and we used the same values of ϵ_{ref} and ϵ_{ref}^{MC} measured and calculated in this work, respectively.

For detector D2 the Monte Carlo efficiency calculations were made directly. In this case, a tuning of some critical parameters of the detector was made. This tuning showed that the thickness of external dead layer (EDL) and distance between the Ge crystal and the Al end cap (DGA) are critical parameters and they were optimized. For the rest of parameters we used the nominal values.

We used the pulse-height tally (F8) per photons emitted from the source to compute the absolute efficiency and we generally obtained relative errors lower than 1% with a number of histories about 10^5–10^6 and 12–14 min of computational times for each energy. This computational time per energy allows us to build an efficiency calibration curve in approximately 4 h. Therefore, the proposed calibration method is good for practical application in everyday measurements. All MC calculations covered the energy range 40–1460 keV.

2.3. True coincidence summing corrections

The second step consisted of the determination of the coincidence summing correction factors (TCSs). For this purpose a simple experimental technique was applied. The corrections were estimated by measuring of a sample, containing the radionuclides of interest with summing effect and one single-emitter radionuclide as reference, in two detector geometries (near and far) [16,17]. The coincidence summing correction factor is defined by the relations $TCS = \left(\frac{R_r}{R_s}\right)_h / \left(\frac{R_r}{R_s}\right)_g$ where R_r is the count rates of the single-emitter radionuclide of reference and R_s is the count rates of the radionuclide to correct. The ratios $\left(\frac{R_r}{R_s}\right)_h$ and $\left(\frac{R_r}{R_s}\right)_g$ are calculated at height *h* from the detector where coincidence summing is negligible and over the detector (indicated by *g*), respectively. This method is simple to use and it is independent of the sample activity which contribute to minimize several uncertainties.

Water solution of unknown activity with the following radionuclides of interest: ^{134}Cs, ^{152}Eu, ^{60}Co and ^{137}Cs, was used. For the reference we used ^{137}Cs in the middle energy of the spectra (661.7 keV). For detector D1 the sample was measured at 10 cm from the end cap and over the detector while for detector D2 the sample was measurement out of the well (at 6 cm from the top of the well) and into the well of the detector. In order to calculate the corrections factors, the TCS expression was applied to the following energy lines: ^{134}Cs (604.7 and 795.7 keV), ^{152}Eu (344.3 keV) and ^{60}Co (1173.2 and 1332.5 keV). It should be noted that no correction was applied to ^{226}Ra and ^{232}Th because the available water source did not contain these radionuclides. All measurements were carried out for about 6–10 h ensuring a meaningful statistics in each source to detector configuration and in all cases the dead time corrections were intrinsically taken into account by the software Winner™.

2.4. Experimental verification

The experimental verification included the internal validation of MC efficiency curves and the participation in two ALMERA Proficiency Tests: IAEA-TEL-2013-04 and IAEA-TEL-2014-04 [18].

The validations of the efficiency calibration curves obtained by Monte Carlo simulation was performed with high purity KCl and several Certified Reference Materials (CRMs) provided by IAEA [19]: Uranium Ore IAEA-RGU-1, soil IAEA-375, IAEA-326 and IAEA-444, marine sediments IAEA-300 and IAEA-306, and grass IAEA-373. For detector D1 the samples were encapsulated in a cylindrical container of polystyrene of 1.8 mm thickness with an internal diameter of 72 mm and filling height of 20 mm. The measurement geometry of detector D2 was a cylindrical polystyrene vial of 1 mm thickness with an internal diameter of 10 mm and filling height of 45 mm. Both capsules do not allow radon diffusion. In the sample IAEA-RGU-1, the radionuclides ^{226}Ra and ^{210}Pb are in secular equilibrium. The specific activity of ^{40}K was determined considering the elemental weights for KCl and ^{40}K natural abundance.

The 2013 ALMERA proficiency test (PT) consisted of three water samples and one flour sample. The participating laboratories were

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