

Experimental determination of the HPGe spectrometer efficiency curve

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Abstract

A review of the calibration procedures of the semiconductor HPGe spectrometer is presented in this paper. Calibration standards were prepared using the standardized radioactive solution of the common monoenergetic radionuclides mixture and standardized multigamma ^{152}Eu solution. The matrix materials were spiked using the activated carbon, and homogenized mechanically or by suspension in $\text{C}_2\text{H}_5\text{OH}$. Experimentally determined efficiency curves were compared with those obtained by Monte Carlo simulation. © 2008 Elsevier Ltd. All rights reserved.

1. Introduction

Activity determination of gamma-ray-emitting radionuclides in environmental samples is a difficult task, in most cases, due to the low level of activity concentration. The main problem involved with low-level activity measurements is insufficient counting statistics. This could be avoided by increasing the volume of the sample and/or by performing close geometry measurements. However, in close geometry measurements, true coincidence summing effects for radionuclides with a complex decay scheme have to be taken into account.

To accurately determine activity concentrations, correct and reliable determination of the efficiency curve is of great importance. Detection efficiency is a complex function of the energy, spectrometer characteristics, measurements geometry, volume and density of the sample, etc. In order to calibrate the spectrometer properly, calibration standards should be prepared with matrices with similar chemical composition and density, similar concentration for the relevant radionuclides, and in the same geometry and counting configuration as the real samples (IAEA, 1989). Although several theoretical efficiency calibration methods are in use, it is recommended that calibration should be experimentally determined for environmental measurements.

Comparison of techniques for the preparation of various calibration standards with different matrices are presented

in this paper. The matrix materials are placed in cylindrical PVC containers (100 ml). They are measured in contact geometry and obtained curves $\varepsilon(E)$ are compared.

2. Preparation of calibration standards

In the Laboratory for Radionuclide Metrology at Vinča Institute (LRMV), the activity concentrations in environmental samples are routinely measured. The standardized solution of a common mixture of gamma-emitting radionuclides purchased at the Czech Metrological Institute (CMI, 2005), and standardized multigamma ^{152}Eu solution, purchased at LMRI, were used to prepare standards for efficiency and energy calibration of gamma-ray spectrometer in accordance with the IAEA recommendation (IAEA, 1989).

The first step in dealing with the standard multigamma solution is dilution to the adequate specific activity. Dilution was done by adding a carrier solution prepared with $\text{SnCl}_2 \cdot 2\text{H}_2\text{O}$, CdI_2 , $\text{Ce}(\text{SO}_4)_2 \cdot 4\text{H}_2\text{O}$, HgCl_2 , $\text{SrCl}_2 \cdot 6\text{H}_2\text{O}$, CsCl , $\text{CoCl}_2 \cdot 6\text{H}_2\text{O}$ and YCl_3 . This radioactive solution was homogeneously mixed in the matrix materials.

In order to cover the wide range of densities of the samples, the calibration standards were prepared with the following matrices: mineralized grass ($\rho = 0.09 \text{ g cm}^{-3}$), powdered milk (0.47 g cm^{-3}), surface soil (0.75 g cm^{-3}), water (1.00 g cm^{-3}) and sand (1.07 g cm^{-3}). The samples of grass and surface soil were collected in the vicinity of Belgrade site; powdered milk and sand were sampled from

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commercially available sources and tap water samples were taken from the Belgrade plumbing system. Grass, soil and sand were first dried at 105 °C for 24 h. Grass was additionally ashed for 15 h at 430 °C. Bulk samples of all matrix materials, prepared as described above, were used both for standards and blanks preparation.

Prepared materials of the spiked standards were placed in 100 ml PVC cylindrical boxes ($d = 66$ mm; $h = 35$ mm)—the mostly used geometry (except of the water—120 ml bottles) for routine measurements of environmental samples in LRMV. Two techniques for spiked standards homogenization were applied. Details of the applied procedures will be explained by reference to the soil standard.

2.1. Preparation of soil standards

Soil samples were pretreated mechanically. The root, mat, vegetation and stones were removed. Prepared soil material was then spiked separately with standardized solutions of ^{152}Eu , natural thorium and radionuclides mixture, applying different homogenization methods.

2.1.1. ^{152}Eu

Soil material was contaminated with 200 μl of standardized solution of ^{152}Eu . The sample was diluted in $\text{C}_2\text{H}_5\text{OH}$. After stirring on a magnetic stirrer, the homogenized liquid phase was dried under an infra-red lamp. A homogeneity test was performed by counting aliquots of contaminated soil on a well-type NaI(Tl) scintillation detector. The nonhomogeneity determined was less than 6%.

2.1.2. ^{152}Eu (activated carbon)

In this approach, the soil material was contaminated applying the procedure given by Taskaeva et al. (1996). According to the mentioned procedure, 150 mg of powered activated carbon was used as the activity carrier. Two hundred microliters of standardized ^{152}Eu solution was added to the carbon, and placed in a plastic spoon, by pipetting. After drying at room temperature the carbon was transferred to the polyethylene bottle and the soil sample was homogenized. A certain amount of carbon, which remains on the spoon was removed with a small portion of the soil matrix. The activity of the used spoon was measured on an NaI(Tl) scintillation counter to check the remains of the activity. The obtained results show that the loss of activity is negligible ($<0.01\%$), i.e. that the carbon banded all ions—carrier of the activity. After the homogenization, the homogeneity test was performed in the same manner as mentioned in the previous section. Nonhomogeneity was less than 2%.

2.1.3. Radionuclide mixture (activated carbon)

Soil matrix was spiked, applying the procedure given by Taskaeva et al. (1996), with a certified mixture of monoenergetic gamma emitters (^{241}Am , ^{109}Cd , ^{139}Ce , ^{57}Co , ^{60}Co ,

^{137}Cs , ^{203}Hg , ^{113}Sn , ^{85}Sr and ^{88}Y), purchased at the Czech Metrological Institute (CMI, 2005). Mixed nuclide standard solution is a preferable choice because it covers a wide energy range, and allows determining the efficiency curve from 30 to 1900 keV. The corrections for summing effects are made only for ^{60}Co and ^{88}Y . Further procedure of homogenization and homogeneity test was performed as in the previous section. Nonhomogeneity was less than 6.4%.

2.1.4. Thorium salt

In order to determine the suitability of the available standardized materials, we spiked soil with p.a. thorium salt $\text{Th}(\text{NO}_3)_4 \cdot 5\text{H}_2\text{O}$ (Merck). Since the salt was extracted from the natural ore at the beginning of 20th century, it is supposed that ^{232}Th is in equilibrium with its progenies. This assumption was confirmed by alpha spectroscopic measurements (Djurašević et al., 2007). Specific activity was evaluated using the nuclear data for ^{232}Th series. In evaluation of the specific activity, weight fraction of thorium in salt and natural abundance of thorium isotopes were taken into account. The calculated specific activity of ^{232}Th in salt was $(1652 \pm 53) \text{ Bq g}^{-1}$. An amount of 5.17 g was added directly to the soil material. The further procedure of homogenization and homogeneity test was performed as in previous sections. Nonhomogeneity was less than 5%.

2.2. Preparation of water standards

2.2.1. Spiking with ^{152}Eu

For the preparation of the water standard with ^{152}Eu , the standardized solution was added directly to the tap water, in the cylindrical PVC bottle of 120 ml ($d = 50$ mm, $h = 60$ mm). Homogenization was performed by the simple method of mechanical stirring.

2.2.2. Water spiked with radionuclide mixture

To make a standard spiked with mixed radionuclide, we used the carrier solution made by dissolving adequate salts in 4 M HCl (Taskaeva et al., 1996). Activity was added directly to a carrier solution placed in the PVC bottle of 120 ml. Homogenization was performed by the simple method of mechanical stirring.

2.3. Preparation of miscellaneous standards

The routine measurements of environmental samples indicate the needs for preparing standards for various matrices. In that purpose, beside the ground soil and water standards, the calibration standards for the mineralized grass, powdered milk and sand matrices were also prepared. These standards were prepared by applying the procedure described in Section 2.1.3.

2.4. Blank preparation

Since the used matrix materials have intrinsic activity, the blank sources should be prepared and subtracted from standards spectra used for determination of the efficiency curve. For all matrix materials, blank samples were prepared in the same manner as the reference standard materials and measured in the same geometry as the calibration samples. Their recorded spectra were subtracted as a background from the corresponding spectra used for efficiency calibration.

3. Monte Carlo simulation using FOTELP

Software package FOTELP (Ilić, 2002) has been developed to simulate the transport of photons, electrons and positrons by Monte Carlo method for numerical experiments in dosimetry, radiation shielding, radiotherapy and for evaluating the efficiency of detectors and counters.

The software package version FOTELP-PEN5 for the simulation of HPGe detector response was used. It includes PENGEO5 geometry package from PENELOPE code (Salvat et al., 2001). The input file for PENGEO5 describes geometry characteristics of the different material zones. The physical characteristics of the detector are described by means of six main dimensions: crystal diameter (49 mm), crystal length (49 mm), thickness and diameter of the aluminum window (1 and 75 mm, respectively), end-cap-to-crystal distance (5 mm) and thickness of the inactive Ge layer (1 mm). The efficiency calibration curves were calculated for the soil and water standards for previously described geometries.

4. Measurement of the efficiency calibration curves

The radiological analysis of environmental samples, in LRMV, is routinely performed by means of the Schlumberger coaxial HPGe spectrometer (resolution: 2.1 keV; relative efficiency: 15%, at 1332.5 keV), including the digital spectrum analyzer, Canberra DSA 1000. For the purpose of efficiency calibration, measurements of the prepared standards were performed in close geometry. These experimental spectra were recorded and processed using the Canberra's Genie 2000 software; net areas of the peaks were corrected for the background (blank sample), dead time and coincidence summing effect, applying the calculation method of Debertain and Schötzig (1990).

Three soil standards spiked by different procedures were prepared. After the analysis of recorded spectra and evaluation of the obtained data for efficiency at given energies, calibration curves were obtained by fitting. The analytical expression of obtained efficiency curves is $\varepsilon = e^{-P(\ln E)}$, where ε is detection efficiency, E is energy, $P(\ln E)$ is a polynomial function of the fifth order.

Fig. 1 shows that experimentally determined efficiencies, (a), (b) and (c) are in reasonable agreement with each other, and with simulated values of efficiency in the overall

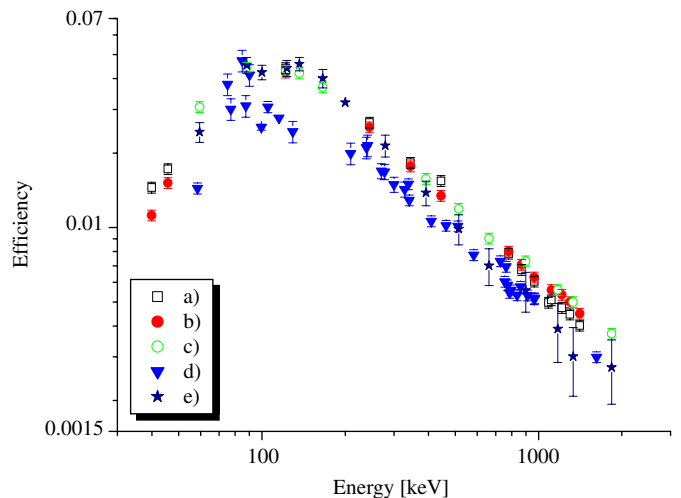


Fig. 1. Efficiency curves for soil standards spiked with (a) ^{152}Eu (alcohol); (b) ^{152}Eu (activated carbon); (c) radionuclide mixture; (d) thorium; (e) Monte Carlo simulation. Uncertainties are quoted with the coverage factor $k = 2$.

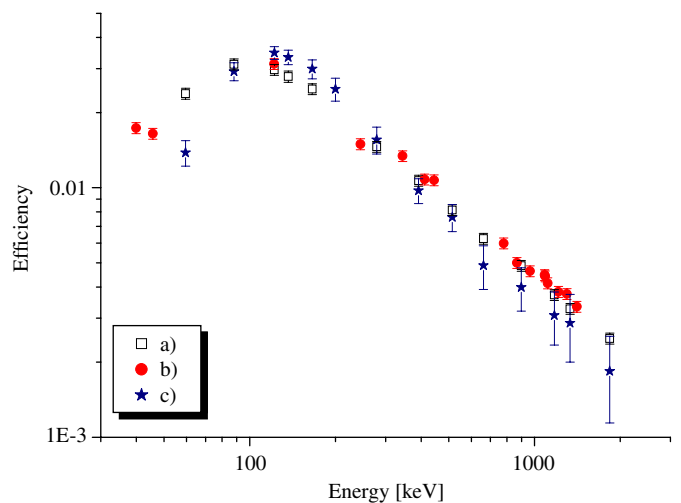


Fig. 2. Efficiency curves for water standards: (a) spiked with monoenergetic radionuclide mixture; (b) spiked with ^{152}Eu (c) Monte Carlo simulation. Uncertainties are quoted with the coverage factor $k = 2$.

energy range. On the other hand, efficiencies (d) are shifted to the left and cannot be used for reliable radiological analysis, and the reason for this is the hygroscopy of the used thorium salt, i.e. the questionable determination of the referent specific activity.

Analysis of water samples is also important in environmental surveys. In order to calibrate the detection system, two water standards were prepared, the first one with the radionuclide mixture, the second one with ^{152}Eu . Comparison of the two experimentally obtained and simulated efficiency values for water matrix is presented on Fig. 2. The calculated curves show systematical deviations from the experimental ones, but not more than 10%. The quoted uncertainties are estimated by the Monte Carlo calculation. In our opinion this discrepancy is caused

by the insufficiently precise knowledge of the detector parameters (detector dead layer and window thickness).

Obtained efficiency curves show that evaluation of the efficiency is most difficult in the energy range from 30 to 200 keV, the knee position is questionable, but all curves are more or less consistent in the high-energy region.

5. Conclusion

Spiked standards and blank samples of surface soil, water, sand, powdered milk and grass were prepared and measured. Efficiency curves for measurement of PVC boxes 100 and 120 ml in contact with the detector were determined and compared. Those geometries need a small amount of sample and are routinely used in our laboratory.

The curves are in good agreement except in the low-energy region where the standard spiked with mixed solution gave better results. Since measurements were performed in contact geometry, coincidence summing corrections along with a background subtraction were made. Efficiency curves were also determined by Monte Carlo simulation. It must be pointed out that the discrepancies between calculated and experimental efficiencies in low-energy region (< 60 keV) are more pronounced. The reason for these discrepancies lies in the fact that the calculated efficiency for very-low energies is very sensitive to the thicknesses of the germanium dead layer and of the detector entrance window, which strongly attenuate low-energy photons (Jurado et al., 2003). Therefore, it was necessary to optimize the thickness of the Ge inactive layer in order to bring into accordance the experimental and calculated efficiencies for very-low energies.

Acknowledgments

The Software package FOTELP for the Monte Carlo simulation used in this article was developed and improved by Dr. Radovan Ilić. The authors wish to express their gratitude for his support and useful discussions.

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