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Contents lists available at ScienceDirect

Nuclear Instruments and Methods in Physics Research A

journal homepage: www.elsevier.com/locate/nima

A practical experimental approach for the determination of gamma-emitting radionuclides in environmental samples

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ARTICLE INFO

Article history:

Received 1 March 2014

Received in revised form

23 May 2014

Accepted 9 June 2014

Available online 18 June 2014

Keywords:

Efficiency calibration

Gamma detector

Radionuclide

Self-absorption correction

Proficiency test

ABSTRACT

The work presents a practical and cost saving approach for the experimental efficiency calibration of a coaxial n-type high-purity germanium detector (HPGe), with the intended use in routine laboratory measurements of all gamma emitting radionuclides in different environmental volumetric samples. The calibration was performed using the ^{238}U and ^{232}Th natural radionuclide chains in secular equilibrium, and additionally KCl salt of analytical purity, covering the energy range between 26 and 1461 keV. The performance evaluation in the worldwide proficiency test (PT) organized by the International Atomic Energy Agency in 2013 confirmed the reliability of the analytical measurement results provided by this experimental approach for both natural and artificial radionuclides. Yet, further consideration is needed in the low energy range regarding the effect of the sample composition on the self-absorption and geometry corrections.

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1. Introduction

Gamma-ray spectrometry is a widely used, non-destructive measurement technique for the analysis of different gamma-emitting radioisotopes in environmental samples. Standard point sources having single energy emissions or volumetric sources of mixed radionuclides, obtained through primary methods and commercially available are commonly employed for efficiency calibration of the gamma detectors. An example of multinuclide standard solution would consist of artificial radionuclides such as ^{241}Am , ^{109}Cd , ^{57}Co , ^{139}Ce , ^{203}Hg , ^{113}Sn , ^{85}Sr , ^{137}Cs , ^{60}Co and ^{88}Y , with energy lines in a wide spectra region (59–1836 keV). The great advantage of using this mixed-radionuclide standard source is that no correction must be performed for interfering radionuclides and nor for the background. Hence, a disadvantage in the short half-lives of several nuclides must be recognized, thus making the source useful over the whole spectra region only for a short period of a few months, being also very costly for the radioanalytical

laboratory. The half-life of any mixture like this is given by the half-life of its shortest-lived member.

Another example of multigamma standard would include ^{125}Sb , ^{154}Eu and ^{155}Eu , covering the energy range between 27 and 1596 keV and having half-lives beyond 2.5 years [1]. The disadvantage of this mixture is the many cascade transitions of Eu for which high coincidence summing corrections are needed, giving additional uncertainties in the massic activity results.

The use of a radioactive chain source in secular equilibrium can be seen as a practical approach for the efficiency calibration of a gamma system, as its half-life is given by the longest-lived parent. U and Th are the only very long-lived radionuclides with natural radioactive chains. Yet, they have a disadvantage in the lower specific activities, leading to the necessity of physically larger volume sources.

This study presents the experimental efficiency calibration of the HPGe detector from Cluj-Napoca laboratory, using RGU-1 and RGTh-1 certified materials provided by the International Atomic Energy Agency (IAEA). These materials were prepared in 1987 in a different manner than the previous ones, being considered to form a set of three certified sources (including RGK-1) to be used for the calibration of laboratory gamma-ray spectrometers for U, Th and K measurements in geological samples [2]. They were prepared using natural U and Th ore, respectively, diluted with floated silica powder, and contain ^{238}U and ^{232}Th natural radionuclide chains in secular equilibrium. Instead of RGK-1, our calibration method included KCl salt crystals of analytical purity, provided by Merks.

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Disequilibrium state in the decay chain ^{226}Ra – ^{210}Pb , caused by the gaseous ^{222}Rn escaping from the source container after a long time period (> 20 years) since the preparation and enclosing of the material, was taken into account.

The calibration procedure is presented with the aim to be used within the laboratory for routine activity measurements of gamma emitting radionuclides of unknown energies. It has the advantage in allowing the determination of any gamma-emitting radionuclide in different liquid and solid environmental samples, even if not present in the calibration source. The proposed efficiency calibration procedure and the laboratory performance in radio-analytical measurements was verified through participation in the *IAEA-TEL-2012-03 worldwide open proficiency test on the determination of radionuclides in water, hay and soil*, organized by the IAEA Laboratories in 2013. The final results and corrective actions are presented further on.

2. Materials and methods

2.1. Gamma-ray spectrometer

An Ortec n-type coaxial detector (GMX30P4-ST) with Be window, 34% relative efficiency and a resolution of 1.92 keV at 1.33 MeV line of ^{60}Co has been employed for the gamma measurements. It is connected to a Digital Multichannel Analyser and mounted on mechanical cooling system. The gamma-ray spectrometric facility also incorporate passive background shielding, using a non-contaminated old lead tower of 8 cm thickness with additional Cu shield inside the tower, of 3 mm. Gamma-spectra were analysed with dedicated commercial software for gamma-ray spectrometry, namely Maestro-32.

The detector resolution and energy calibration is periodically verified for stability using a water-equivalent ^{152}Eu source [3] and a ^{241}Am point source, certified by the Horia Hulubei National Institute of Research & Development for Physics and Nuclear Engineering Bucharest (Romania).

2.2. Calibration sources

Three samples containing RGU-1, RGTh-1 and KCl were prepared and measured with the n-type Ortec detector. The calibration sources were encapsulated in aluminium containers (80.5 mm diameter, 0.22 mm bottom thickness, 1.00 mm side wall thickness and 26.5 mm filling height) which do not allow radon diffusion [4]. After properly sealed with glue and electrical band, the calibration samples were kept for minimum four weeks before measuring to ensure that equilibrium between ^{222}Rn and ^{226}Ra is reached. The container geometry was selected with a diameter close to the end cap diameter of the detector.

The measurement time was set to 24 h for RGU-1 and RGTh-1 and to 4 h for KCl, in order to reach meaningful statistics for each full energy peak which meant to be used for efficiency calibration. This measurement time gave a dead time correction of 0.5%. The spectra were collected separately and afterwards analysed and plotted into the same file to cover the whole energy range.

RGU-1 contains U-ore with 400 ppm of U, less than 20 ppm of K and less than 1 ppm of Th and RGTh-1 contains 800 ppm of Th, 6.3 ppm of U and K (0.02%) [2]. The specific activity concentration (Bq g^{-1}) of ^{238}U , ^{235}U and ^{232}Th and their natural abundance were used to derive the radionuclides activity (Bq). Thus, the specific activity of ^{238}U was found to be $4.9399 \pm 0.0250 \text{ Bq g}^{-1}$, $0.2303 \pm 0.0012 \text{ Bq g}^{-1}$ for ^{235}U and $3.2536 \pm 0.0650 \text{ Bq g}^{-1}$ for ^{232}Th . In the case of ^{40}K , its specific activity was determined considering the elemental weights for KCl and ^{40}K natural abundance. It corresponds to a ^{40}K specific activity of $2.645 \times 10^5 \text{ Bq kg}^{-1}$. All nuclear

data used in the gamma-spectrometric analysis were extracted from Monographie BIPM-5 [5].

2.3. Reference materials and proficiency test samples

The internal validation of the efficiency calibration curve was performed with three IAEA reference materials: IAEA-447 soil-moss [6], IAEA-375 soil [7], IAEA-372 hay [8] enclosed in cylindrical plastic containers of 90 mL (75.98 mm external diameter, 0.82 mm side and bottom wall and 29.00 mm height) and measured for more than 24 h. The selected materials included different radionuclides and matrices, in order to cover a wide energy range.

Within the worldwide proficiency test (PT), each participating laboratory received five homogenized samples of different matrices (3 water bottles, one soil box and one hay box) for the analysis of the radionuclide content. The water was spiked with unknown anthropogenic radionuclides and with ^{226}Ra to simulate a natural background. One water sample contained known radioisotopes and massic activities, and was provided for quality control purpose. The hay sample contained hay collected in Austria, shortly after the Chernobyl accident, spiked with ^{134}Cs and 'naturally contaminated' with ^{137}Cs . Soil sample contained unknown natural and anthropogenic radionuclides and was collected from Jeju Island, Republic of Korea. All results were to be reported in Bq kg^{-1} , on a dry-weight basis. The samples were prepared in 90 mL cylindrical plastic geometries and measured for 24 h, apart from the soil sample which was measured for 72 h in order to achieve statistically meaningful data. All samples were measured under the same conditions as those under which the system has been calibrated.

The final results were given as massic activity \pm combined standard uncertainty (Bq kg^{-1} , dry mass), 95% confidence interval. The uncertainty budget followed the error propagation low and included the following components: sample and calibration source counting statistics, calibration source activity, background correction, photopeak efficiency fitting, self-attenuation and geometry correction and coincidence summing correction. ^{210}Pb was determined from its low gamma-energy at 46.5 keV, the 1460.8 keV energy line was considered for ^{40}K estimation and ^{228}Ac was determined from its 911.19 keV gamma-line. ^{208}Tl and ^{212}Pb were determined from the 583.19 keV and 238.63 keV gamma energies, respectively. ^{137}Cs was calculated from the 661.6 keV gamma-emission of $^{137\text{m}}\text{Ba}$ and ^{241}Am was estimated from its 59.54 keV line. No correction has been made for ingrowth from ^{241}Pu . Two or three gamma-energies were analysed for ^{134}Cs (604.72 keV, 795.86 keV), ^{60}Co (1173.23 keV and 1332.49 keV) and ^{152}Eu (344.28 keV, 778.9 keV and 1408.01 keV) and the resulted massic activities were reported as arithmetic mean.

2.4. Corrections performed for massic activity determination

Decay-corrections were applied to calibration sources and samples to be measured with the reference time given in their Certificates or with the one requested by the PT convener. For background corrections, an empty geometry was measured in the same conditions for about 72 h, and the average background net counts of two intercalated spectra were subtracted from the net peak area of the source.

Corrections for geometry and self-attenuation, true coincidence summing corrections for calibration sources (for ^{227}Th and ^{228}Ac) and samples (for ^{152}Eu , ^{134}Cs , ^{60}Co , ^{208}Tl and ^{228}Ac) were also considered. The summing correction factors were derived using the available code EFFTRAN [9], while for the efficiency transfer the ETNA code [10] was employed. The use of these codes makes necessary to have a detailed description of the geometric intrinsic characteristics of the detector, which were given by the supplier

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