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Determination of mass attenuation coefficient by numerical absorption calibration with Monte-Carlo simulations at 59.54 keV

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ABSTRACT

This study presents a numerical method in order to determine the mass attenuation coefficient of a sample with an unknown chemical composition at low energy. It is compared with two experimental methods: a graphic method and a transmission method. The method proposes to realise a numerical absorption calibration curve to process experimental results. Demineralised water with known mass attenuation coefficient ($0.2066\ \mathrm{cm^2\,g^{-1}}$ at 59.54 keV) is chosen to confirm the method. $0.1964 \pm 0.0350\ \mathrm{cm^2\,g^{-1}}$ is the average value determined by the numerical method, that is to say less than 5% relative deviation compared to more than 47% for the experimental methods.

1. Introduction

Geochronologic methods with natural radioisotopes, such as ^{210}Pb, ^{7}Be and anthropogenic ones (^{137}Cs), make the determination of geologic characteristics possible; sedimentation speed and rate, bioturbation and sediment's age [1]. In order to quantify the gamma photons emitted by these isotopes, the commonly used method is the gamma spectrometry. The quantification of ^{210}Pb is often used for environmental sample dating [2–4]. This isotope originates through the decay of ^{238}U which is found in most soil. ^{226}Ra is a transitional daughter which produces gaseous ^{222}Rn. A proportion of ^{222}Rn decays to ^{210}Pb within the soil (called the supported ^{210}Pb, $^{210}\mathrm{Pb}_{supp}$) which is in equilibrium with the parent ^{226}Ra. Another proportion of gaseous ^{222}Rn decays into the atmosphere. The produced ^{210}Pb (called the excess ^{210}Pb, $^{210}\mathrm{Pb}_{ex}$) is quickly deposited as fallout and is not in equilibrium with its parent. The quantification of ^{210}Pb by gamma spectrometry is realised through its emitted gamma rays at low energy (46.5 keV). The measure of this photoelectric peak gathers both parts of ^{210}Pb together, but sample dating is carried out by quantification of excess ^{210}Pb. The amount of $^{210}\mathrm{Pb}_{ex}$ is determined by subtracting $^{210}\mathrm{Pb}_{supp}$, which is in equilibrium with ^{226}Ra, from the ^{210}Pb total activity. The counting at such a low energy as 46.5 keV requires a broad energy or N type detector. However the photons are affected by self-absorption phenomena. These effects prevent some photons from reaching the detector and induce an underestimated quantification [5]. Studying sediment activities involves calibrating the detector to find the efficiency curve for the considered geometry. The standard sample needs to have the same geometry, density and chemical composition as the studied sample, in order to make sure that the degree of self-absorption is the same between both samples. The Chrono-Environment Laboratory uses the IAEA-447 moss-soil certified reference material [6]. This standard can take the desired geometry, but the exact composition is unknown. Moreover, chemical composition and density are not similar, due to the large range of soil's composition and the difficulty to prepare a sample with the same settlement.

The methods to overcome the self-absorption difference are:

1. apply a self-absorption correction between standard and studied sample [7,8],
2. perform a calibration of the efficiency with numerical calculation.

In the second method, the knowledge of the chemical composition is essential. This work proposes the determination of the mass attenuation coefficient at low energy. This coefficient is linked to the chemical composition through the following equation:

$$\mu_m = \Sigma(\mu_{mi} \cdot \omega_i) \tag{1}$$

where μ_{mi} is the mass attenuation coefficient of element i and ω_i is the mass percentage of element i. μ_m represents the attenuation of

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the photon beam, at a given energy, in matter. The determination of this coefficient makes it possible to define a virtual chemical composition which has the same attenuation characteristics as the studied sample. In other words, the sample and the virtual sample have the same degree of self-absorption (for a given density). The simulation can be carried out with the new virtual composition for lower energy to realise the efficiency curve. This work determines the mass attenuation coefficient of demineralised water at 59.54 keV. The results are compared with the theoretical value.

2. Materials, device and simulation

2.1. Materials

The used detector in this study is a planar BEGe detector (Broad Energy Germanium, model BE3825). It was manufactured by Canberra in 2014. The nominal efficiency is higher than 28%, and its resolution is 1.72 keV at 1332 keV. All spectra are analysed using the Genie 2000 software (Canberra) [9]. The dimensions and materials of this detector provided by manufacturers are shown in Fig. 1. The germanium crystal is surrounded by a low-background copper holder kept into a low-background aluminium end-cap in vacuum. The entrance window is made up of carbon-epoxy material. The dead layers are not included in Fig. 1. The front and the lateral dead layer are given respectively as 0.4 µm and 0.6 mm.

For measurements, a radioactive point source of ^{241}Am was used. It is sealed into thin plastic sheets and the dimensions of the active zone are 0.11 mm thickness and 3 mm diameter. The activity was 38,750 Bq when the measurements were carried out. ^{241}Am emits gamma-ray at 59.54 keV (35.9%) and enables to study the attenuation at low energy. The point source is collimated by a lead collimator in order to obtain a photon beam perpendicular to the detector window. The dimensions of the collimator are 5.05 cm × 7.63 cm, the hole's diameter is 0.19 cm and the height is 3.28 cm.

The studied sample is water. The theoretical total mass attenuation coefficient of pure water (H_2O) is 0.2066 $cm^2\ g^{-1}$ at 59.54 keV [10]. Demineralised water was chosen in order to be closer to this attenuation coefficient, removing elements which could change this value. The density of the solution was determined by weighting the known water volume in a 25 cm^3 volumetric flask. In the laboratory's temperature condition, the density of water is $0.9889 \pm 0.0079\ g\ cm^{-3}$. The water solution was contained in a SG 50 standardised bottle.

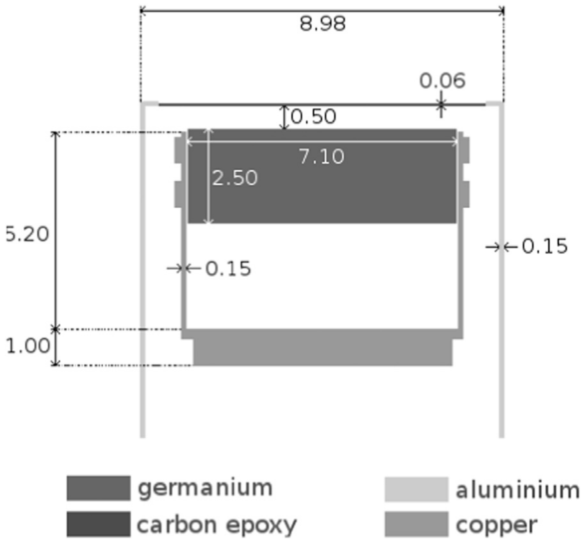

Fig. 1. Geometry of the planar detector. All dimensions are given in centimetres.

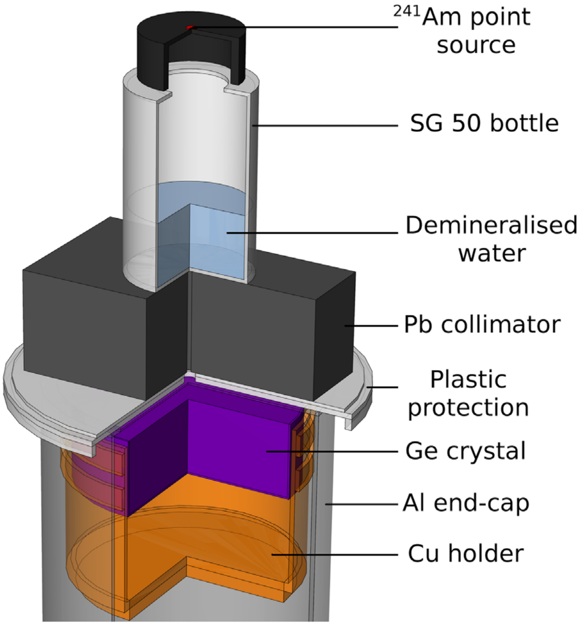

Fig. 2. Sectional view of the experimental device as implemented in SimpleGeo 4.3 [11].

2.2. Experimental device

The experimental device is shown in Fig. 2. In this configuration, all the photons which reach the germanium crystal have crossed the same thickness of the sample due to the position of the lead collimator. A plastic protection is inserted between the lead collimator and the carbon-epoxy window to protect the top of the detector from the collimator's mass. There is a hole (diameter 0.5 cm) in its centre to avoid absorption in this material. This device was used for the three tested methods in this work.

2.3. Monte-Carlo simulation

The MCNP6 (Monte-Carlo N-Particle) code [12] was used for modelling the BEGe detector and the device of the study (Section 2.2). The model was established based on the dimensions provided by the manufacturer (Fig. 1). The most suitable tally for modelling detector response is the F8 tally which provides the energy distribution of pulses created by gamma photons in the detector.

Each run used 10^9 histories and the uncertainties on the number of counts are about 1.8% maximum for the photopeak including the region of interest at 59.54 keV.

The compatibility between the simulated results and the experimental ones (for the same device and the same sample) is very important in order to validate the detector model, and it must be checked first. If these results are different, a correction factor must be found. The correction factor f can be expressed as:

$$f = \frac{N(\exp)}{N(\mathrm{MCNP6})}. \quad (2)$$

To define f, an experiment with the device (Section 2.2) was carried out with an empty SG 50 bottle for an active time of 5 h.