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Self-shielding corrections in cylindrical samples in gamma spectrometry with germanium well-type detectors

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ABSTRACT

Self-shielding in spatially distributed samples can be very significant when detecting photons below 100 keV. A correction method has been developed for cylindrical samples, typically used in measurements of natural radioactivity with well-type detectors, as is the example of sediment dating. The method calculates the probability of photons to escape the sample, using Monte Carlo techniques with a program written in C. The effects of self-shielding on the angular distribution of photons have been indirectly analyzed as a function of sample geometry, given a fixed detector geometry. The results given by the program have also been used to provide a straightforward way of calculating the self-shielding factors, with uncertainties. Required inputs are the attenuation coefficient of the sample, its radius, and its height. A procedure has also been specified in order to make good use of all the information. To check on the method, measurements of reference material of known activity have been compared with calculated values, obtaining very satisfactory results.

1. Introduction

Germanium well-type detectors are a good choice when low gamma activity samples need to be measured with good energy resolution. The detector's cylindrical cavity gives them high efficiency even at high energies. For this reason they are commonly used when dating sedimentary cores by means of the ^{210}Pb or ^{137}Cs techniques, where the contents of these radioisotopes are rather low.

Since the material under study is not transparent to gamma radiation, the photon flux escaping from the sample is altered in magnitude and in spacial and angular distributions. These effects are considerable especially under 100 keV, where absorption coefficients are relatively high; therefore, a correction to the detection efficiency becomes necessary.

Several methods have been proposed for self-absorption corrections or for studying self-absorption phenomena. Many authors address the problem of sample self-shielding for coaxial detectors. For these detectors, the approach of Vesic and Anicin [1] consists of finding an equivalence between the cylindrical sample and an ideal linear source by means of an effective linear attenuation coefficient. The proposed procedure is self-contained and requires the corrections to be performed both for the reference material used in the determination of detection efficiency, and for the unknown sample activity. Cutshall et al. [2], Battiston et al. [3] and Galloway [4] have discussed experimental methods for self-absorption corrections combining transmission measurements with a linear source – or an equivalent attenuation distance – model. These approaches are commonly used in the field of sediment dating, but not applicable to measurements in well-type detectors since geometry and photon distribution are essentially different. Hussain et al. [5] presented an experimental technique called 'geometrically normalized', based on performing a one time standard "efficiency vs. sample mass" curve for a certain detector; this approach yields accurate results only when mass absorption coefficients of both the unknown and standard samples are similar. Capponi et al. [6] have already used Monte Carlo techniques, taking into account self-absorption, to estimate overall detection efficiency for extended sources; however, contrary to our work, the referred study was performed for cylindrical detectors. Appleby et al. [7] and Appleby and Piliposian [8] address the problem of self-absorption in well-type detectors and provide well supported semi-empirical formulae.

Although, as the literature shows, the problem under study has in general been addressed before, our aim was to find a very simple, fast, and "low cost" method of self-shielding correction for well-type detectors. Our method is based on Monte Carlo calculations that do not require to perform many previous measurements on the material properties of each sample.

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However, using very few and easy to obtain parameters the method delivers self-shielding factors that are close to experimental values.

In this work we study the self-shielding phenomena in cylindrical sources to be measured in well-type detectors by means of a simple Monte Carlo calculation routine. We analyze the influence of the radius and height of the sample. As a result of this analysis we provide a simple way to calculate values of self-shielding correction factors for samples compatible in size with commercially available detectors, as well as simple formulae for calculating these correction factors for different geometrical parameters. Finally we propose a procedure for calibration of a detector and self-shielding corrections in measurements of samples of unknown activity in the field of low activity gamma counting and, in particular, sediment dating. Additionally, we evaluate the validity of the correction factors through the measurement of samples of reference material of known activity.

2. Self-shielding factors

2.1. Description of the problem

In the chain of events leading from the emission of a gamma-ray by the material under study to its detection as a full-energy-peak count we shall distinguish three separate events in our formulation:

- escaping from the sample material,
- reaching the detector crystal,
- producing a full-energy-peak count.

We first define the sample as having a cylindrical shape with radius R and height H, and assume that the photons originated in this sample are monoenergetic, have a uniform spatial distribution, and are emitted isotropically.

Let P_x represent the probability that a photon from the sample produces the event x, where x can take the value s for escaping the sample, l for reaching the crystal and f for producing a full-energy-peak count.

The efficiency of detection P_f (probability of producing a full-energy-peak count) will be affected by the absorption in the sample, which depends strongly on μ_s, the linear attenuation coefficient of the sample. Therefore, $P_f = P_f(\mu_s,R,H)$.

In the ideal case that the sample is totally transparent to its own radiation, that is $\mu_s = 0$, the efficiency for this sample, with the same geometry setup, would be $P_f = P_f(0,R,H)$.

Calling f_a the sample self-shielding factor, we can write, as in the work by Sima and Dovlete [9]

$$f_a(\mu_s,R,H) = \frac{P_f(\mu_s,R,H)}{P_f(0,R,H)}. \tag{1}$$

Of course, the ideal sample with no self-absorption is just that, ideal, and we cannot make an experimental use of it. However, if we measure, with the same geometrical setup, a sample of known activity (standard sample), we can write for that standard sample in particular:

$$f_a(\mu_{st},R,H) = \frac{P_f(\mu_{st},R,H)}{P_f(0,R,H)}. \tag{2}$$

From Eqs. (1) and (2)

$$P_f(\mu_s,R,H) = \frac{f_a(\mu_s,R,H)}{f_a(\mu_{st},R,H)} P_f(\mu_{st},R,H). \tag{3}$$

If one wants to estimate P_f by calculation it is necessary to correctly evaluate the complicated phenomena taking place when a photon enters the detector crystal and to know exactly the internal geometry of this crystal. We will not tackle this problem here. Instead we see that a simple mathematical manipulation allows us to write the efficiency as

$$P_f = \underbrace{\frac{P_f}{P_s}}_{P_{f/s}} P_s = \underbrace{\frac{P_f}{P_l}}_{P_{f/l}} \underbrace{\frac{P_l}{P_s}}_{P_{l/s}} P_s. \tag{4}$$

The introduction of the conditional probabilities $P_{x/y}$ allows us to evaluate what fraction of photons is lost in each stage of the journey from the source to the crystal before photons are detected as a full-energy-peak count. Eq. (1) can then be reformulated as

$$f_a(\mu_s,R,H) = \frac{P_{f/l}(\mu_s,R,H)}{P_{f/l}(0,R,H)} \frac{P_{l/s}(\mu_s,R,H)}{P_{l/s}(0,R,H)} \frac{P_s(\mu_s,R,H)}{P_s(0,R,H)}. \tag{5}$$

A favorable situation would be that one where the conditional probabilities $P_{x/y}$ do not depend strongly on μ_s. In general, the conditional probabilities could depend strongly on the sample properties regardless of the events x and y, as they are ratios between the integral probabilities P_x and P_y. This dependence is physically based on the fact that each attenuation stage affects the global angular distribution of photons entering the next stage, thus leading to different probability density functions of path lengths in each material.

It will be assumed at this point that at low energies $P_{f/l}$ is independent of μ_s. This is true because Ge is quite opaque to low energy photons; therefore, the alterations in the distribution of photon path lines on the crystal surface due to the absorption in the sample and detector casing will not appreciably affect the fraction of photons detected.

The same assumption is not made for $P_{l/s}$. Absorption in materials between source and detector does not reach saturation for high values of path length to mean free path ratio (the term *saturation* refers to the fact that $P_{abs.} \sim 1-e^{-l/\Lambda}$, where l represents the photon's path length and Λ their mean free path); therefore, changes in the angular distribution of photons could generate variations in this probability. Thus, constancy of $P_{l/s}$ will not be assumed but rather investigated.

One of the objectives of this work is – through Monte Carlo calculations – to determine the values of P_s and P_l in order to study their dependence on geometry, and that of $P_{l/s}$ on μ_s. If after this analysis, $P_{l/s}$ could, as $P_{f/l}$, be assumed independent of μ_s, the mathematical expression for the correction factor would be

$$f_a(\mu_s,R,H) = P_s(\mu_s,R,H) \tag{6}$$

given that $P_s(0,R,H) = 1$ by definition. In that case, the final objective is to provide a simple systematic way of calculating P_s (that is, self-shielding correction factors) for a given triad (μ_s,R,H).

2.2. Monte Carlo implementation

The calculation of probabilities by means of Monte Carlo techniques requires a rather explicit description of the geometry of the problem. It is necessary to specify the geometry and material properties of the samples, vials, and detector parts.

Fig. 1 shows a diagram of the geometry used in the calculations presented in this work. The measurements to which this correction method is addressed are performed with cylindrical samples, consisting of sedimentary powder or a fluid inside a plastic vial, positioned vertically in a well-type detector. The problem presents cylindrical symmetry. The detector has a 0.5 mm thick aluminum casing, with the germanium crystal inside. Separation between the crystal and casing is assumed to be approximately 3 mm. The surface dead layer of the detector has been taken into account, assuming a thickness of 0.7 mm.