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Efficiency computation for gamma-ray spectrometry assessment of samples with intrinsic inhomogeneity

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ABSTRACT

The full energy peak efficiency for inhomogeneous samples, comprising a number of components with different activity and matrix is evaluated by Monte Carlo simulation. The distribution of the values of the efficiency due to the effects of inhomogeneity is constructed. In the particular case when one of the components is a highly active and highly attenuating medium, then the efficiency and its uncertainty at low energies is sensitive to the properties of this component, including the dimensions of the grains.

1. Introduction

Obviously, the efficiency of gamma-ray detectors for measurement of a volume sample depends on the distribution of the radioactivity and of the matrix in the sample. In common applications it is considered that the samples are homogeneous with respect to matrix and that the activity is uniformly distributed. However, in some cases the activity distribution is not uniform, due to the action of specific physical phenomena, and the evaluation of the efficiency should properly take into account this fact. For example, efficiency corrections due to non-uniform activity distribution resulting from radon diffusion or neutron flux distribution in activation of large volume samples have been reported (Overwater and Hoogenboon, 1994; Sima, 1996, 2000; Carconi et al., 2012; Ott et al., 2014;). In other cases bulk samples (e.g. environmental media) possess an intrinsic inhomogeneity, comprising of components of different types, with specific granulometry (Savva et al., 2016; Caridi et al., 2016). In such cases the activity and the matrix distributions have a random character. It seems that a detailed study of the effects of this type of inhomogeneity on the efficiency is still lacking.

In this work we consider the assessment of the radioactivity content of an environmental medium such as soil or sediment, by gamma-ray spectrometry. We are especially interested in the effects on the efficiency and its uncertainty of the intrinsic inhomogeneity of the sample. The model adopted for describing the inhomogeneity of the environmental medium is the same as introduced in a previous paper (Sima and Lépy, 2016). The main features of the model are: the environmental medium is considered composed from a number of building blocks of equal dimensions, each being homogeneous with respect to matrix and with activity uniformly distributed. Each building block is of a specific type, i.e. it belongs to a class defined by its activity and matrix. The probability p_i of a block to belong to class i is given. With these parameters the statistical properties associated with the inhomogeneity of the environmental factor are completely defined. Thus in this model the inhomogeneity is characterized first by a scale, given by the dimensions of the building blocks, second by the activity and matrix of each class and third by the probability of a block to belong to a specific class.

In typical measurements, the activity of the environmental medium is assessed as follows. One or more samples are collected, prepared and measured. The activity of each sample is computed using the efficiency of the specific sample-detector configuration. Finally, the activities of the measured samples are combined for estimating the activity of the environmental medium. For a proper evaluation of the uncertainty of these results, several issues should be considered. First, the samples are random probes extracted from the parent population (i.e. the statistical population describing the environmental medium). Therefore, each sample has a specific composition with respect to the number of the building blocks of each type; then, in each sample there is a particular spatial distribution of various building blocks. The detection efficiency depends on these factors. The lack of knowledge about these factors should be reflected in the uncertainty of the efficiency, propagated further to the uncertainty of the activity of the environmental medium. In this paper the above subjects are studied in the framework established by the Supplement 1 to the Guide to the expression of the uncertainty in measurement (JCGM 101, 2008). In this approach, the value of each factor contributing to the quantity of interest is sampled from a corresponding distribution with a Monte Carlo procedure; using the set of sampled values of all the relevant factors, a value of the quantity of interest is computed. By repeating the procedure many

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times, the distribution of the values of the quantity of interest is constructed. Finally, this is summarized by the computation of the best estimate (taken as the expectation value of the distribution), the standard uncertainty (taken as the standard deviation) and specific coverage intervals.

2. Principle of the computation

The principle of the method for the computation of the efficiency for a sample with intrinsic inhomogeneity is the following. It is assumed that the individual components are known (activity A_i, matrix m_i and probability p_i). The dimensions of the building blocks, as well as of the samples to be measured are also known; then the total number N_t of the building blocks included in the sample to be measured is specified, $N_t = V_s/V_O$, where V_s is the volume of the sample and V_O the volume of the building block. In a first step, Monte Carlo simulation is used for randomly sampling, with probability p_i, the type of each of the N_t blocks, obtaining thus n_i blocks of type i, $i = 1, 2, \ldots k$, where k is the total number of classes (i.e. component types). Evidently, the sum of the numbers n_i over all types of components equals N_t. In this step not only the types of the blocks are sampled, but also their position inside the sample. In the second step, the efficiency for the particular distribution of components (thus, of the activity and matrix) is computed using a special extension of the GESPECOR software (Sima et al., 2001). Next another sample is drawn from the same parent distribution and the efficiency is again evaluated using GESPECOR; the total activity may be different from one sample to another, but in the calculation of the efficiency this fact is properly taken into account. The procedure is repeated many times and the individual efficiency values are used for the construction of the corresponding distribution.

3. Results and discussion

In this work the procedure is applied in the case of a soil sample which includes grains of pitchblende. The radioactive elements of interest are the members of the uranium-radium natural decay series. The elementary components of the sample considered are common soil, pitchblende and air. The composition of the soil fraction, of density 1.2 g cm^{-3}, is O (0.558), Si (0.316), Al (0.071), Fe (0.032), C (0.012) and H (0.011), where in parentheses the fraction by weight is given; pitchblende is considered composed from UO_2, of density 10.8 g cm^{-3}; standard air composition, with density 0.0012 g cm^{-3}, is adopted. It is assumed that the activity of air is negligible, and the ratio of the activity of one grain of pitchblende to one grain of soil is 1750. Note that pitchblende grains have a much higher specific activity and self-attenuation. Several sizes of the building blocks are considered, as well as several values of the probability of each component. It is assumed that individual samples (geometry: cube of dimensions 5 cm × 5 cm × 2 cm) are extracted from the parent distribution and measured in a close geometry with an n-type HPGe detector of 47% relative efficiency. For each sample the full energy peak efficiency for the energies E = 46.539 keV (^{210}Pb), 92.38 keV (^{234}Th), 186.211 keV (^{226}Ra) and 1001.44 keV ($^{234\,m}$Pa) emitted by nuclides from the U-Ra natural decay chain is evaluated and finally, after simulating a large number of samples, the distribution of the values of the efficiency is constructed.

A first example refers to the case of a soil characterized by: soil fraction $p_1 = 0.799$, pitchblende fraction $p_2 = 0.001$, air fraction $p_3 = 0.20$. The building blocks (grains) have cubic geometry, of dimension 0.5 mm, thus the total number of grains in the sample equals 400,000. Due to the much higher activity of pitchblende grains, the fraction of the photons emitted from pitchblende is very important.

A number of 5000 samples simulating the probes to be measured were prepared by Monte Carlo simulation. Despite the fact that the samples are extracted from the same parent population, the number of grains of pitchblende is a random variable from sample to sample, distributed over the range 330–470; this variable has a binomial distribution with $N_t = 400{,}000$ and $p = 0.001$, thus expected value equal to $N_t \cdot p = 400$ and $\sigma^2 = N_t \cdot p \cdot (1-p) \approx 400$. For a fixed number of pitchblende grains, their location within the sample is also a random variable.

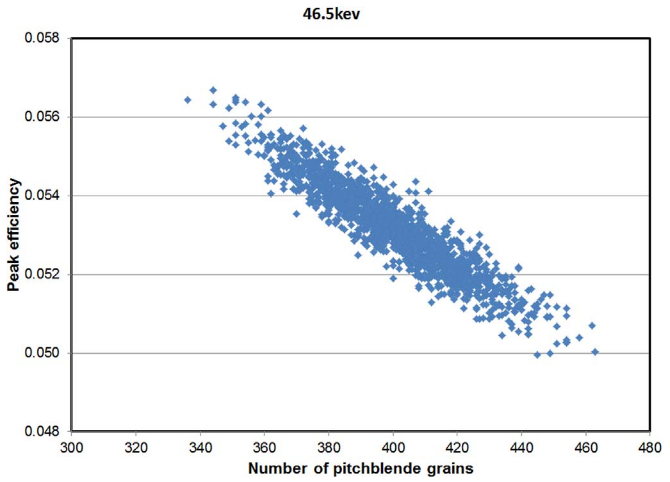

Fig. 1. Distribution of the values of the full energy peak efficiency for the energy $E = 46.5$ keV in function of the number of pitchblende grains; soil fraction $p_1 = 0.799$, pitchblende fraction $p_2 = 0.001$, air fraction $p_3 = 0.2$, block dimension $d = 0.5$ mm. Best estimate of the efficiency 0.053, standard deviation 0.001.

In Figs. 1–4 the distribution of the Monte Carlo calculated efficiency for this set of samples is displayed for E = 46 keV, 92 keV, 186 keV and 1001 keV, as a function of the number of pitchblende grains. For a given number of pitchblende grains the efficiency values are distributed in a specific range; this distribution is due to the various spatial locations of the grains. At E = 46 keV an interesting feature is evident on Fig. 1. The efficiency decreases with increasing number of pitchblende grains. This dependence can be understood in the following way. The fraction of 46 keV photons emitted by the nuclei incorporated in the pitchblende grains with respect to the total number of 46 keV photons emitted by all the nuclei from the sample increases with increasing number of pitchblende grains. Due to composition and density, the absorption of the 46 keV photons in the pitchblende grains is much higher than in the soil grains. If the fraction of the photons emitted by the nuclei from pitchblende grains increases, this means that the fraction of photons which are more attenuated also increases; correspondingly the probability of detection per photon emitted by the decay of nuclei from the sample decreases. In the case of higher energies, the attenuation in the pitchblende grains of dimension

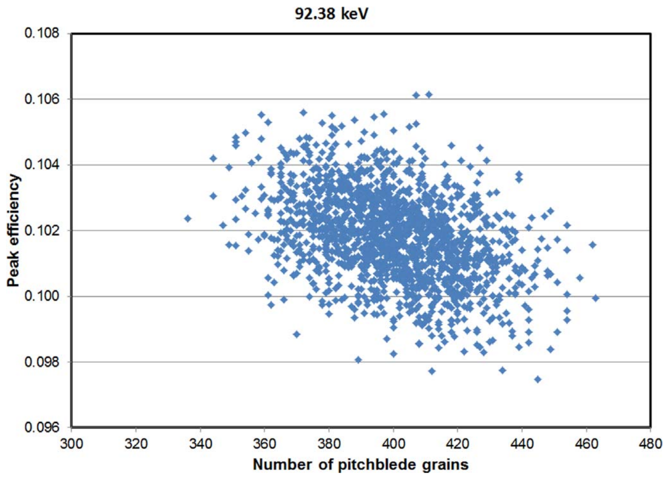

Fig. 2. Same as Fig. 1 for the energy $E = 92.38$ keV. Best estimate of the efficiency 0.102, standard deviation 0.001.