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Correction factors determination in large samples gamma assay using its own multi-gamma lines spectrum

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ABSTRACT

An easy and simple method for gamma assay of large multi-gamma lines samples was introduced in this work. This method performs the assay using point source calibration. The correction factors for volume and self-attenuation are experimentally deduced from the spectra of different thicknesses samples utilizing the following two simple well known facts: large and small samples of the same homogenous material have identical specific activities; the self-attenuation of gamma line decreases as its energy increases. The method was successfully applied to naturally occurring radioactive material (NORM) large samples. This method does not require complicated mathematical procedures. Neither sample matrix data nor detector unit composition is needed.

1. Introduction

There are two major reasons for assaying large portions of materials. Large sample achieves better representativeness of the material and improves the assay sensitivity as the sample signal is enhanced compared to the background. Gamma spectrometric measurement of large samples of different types is a very common practice. Many environmental samples cannot be measured as point sources, due to their low specific activities, which require measurement of large samples. The large samples in general differ in their composition, dimensions and density from the standard calibration sources. Thus, in general it is not possible to use the calibration curves resulted by using standard sources to assay large samples. This problem could be solved by preparation of large reference samples, which requires additional experimental or computational procedures and include some approximations (Abbas et al., 2006; Jodlowski, 2006). This solution is generally time consuming, troublesome, complicated and does not give accurate result (Quindo et al., 2006).

Theoretical and numerical methods of determining the gamma-ray peak efficiency for a cylindrical source, based on a modified expression for point sources was derived (Aguiar et al., 2006; Haase et al., 1995; Overwater et al., 1993; Korun and Martincic, 1992). A Monte Carlo simulation was used also to determine volume efficiency (Saegusa et al., 2004). This approach demand high simulation skills, which is rare in analytical laboratories. A mathematical relation between the peak efficiencies of disks sources and that of point sources were proposed (Helmer, 1983). All these methods solve the problem using either lengthy and complicated mathematical calculation or numerical models. These methods also require information about the detector unit and the sample composition. Thus the need for a new solution is quite serious. In this work a new method is introduced. This method is completely experimental and does not require complicated mathematical procedures. Neither sample matrix data nor detector unit composition is needed.

2. Experimental

The possibility of large sample gamma assay using point source calibration will be investigated. The multi-gamma lines spectrum of the assayed material forms the main prerequisite of this method. This prerequisite is fulfilled by naturally occurring radioactive material (NORM) samples, whose radioisotopes emit gamma spectrum of many gamma lines. These gamma lines cover wide range of energies. Thus the work will be performed using NORM samples. The NORM material was obtained from collected scales, which was removed from oil pipes in Der-Alzor (Syria) fields in August 2005. The material was dried, powdered and homogenised to prepar the cylindrical samples in polyethylene capsules of fixed diameter (45.5 mm) and variable heights. The gamma spectra of the large cylindrical samples were measured experimentally by HPGe coaxial detector based gamma spectrometer. The coaxial condition for detector and sample is fulfilled in the experimental setup.

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If the radioisotope decay during acquisition is negligible, which is true in our measurements, the activity of radioisotope i is calculated using the measured count rate of its j gamma line by

$$A_{ij} = \frac{Att_j}{B_{ij}\varepsilon_{tot\,j}} cps_j \quad (1)$$

where Att_j is the self-attenuation correction factor for j-th gamma line; B_{ij} the branching ratio; $\varepsilon_{tot\,j}$ the total detection efficiency of the measuring setup and cps_j the measured count rate.

The dependence of the activity value on the self-attenuation correction factor and total efficiency is obvious in this relation. To use the standard (point) source efficiency calibration for large cylindrical samples, the total efficiency formula was modified to account for the volume of the large cylindrical samples. Thus the total efficiency is expressed as a product of three factors

$$\varepsilon_{tot\,j} = \varepsilon_{\mathrm{int}\,j}\varepsilon_d\varepsilon_v \quad (2)$$

where $\varepsilon_{\mathrm{int}\,j}$ is the detection unit (detector and its housing) intrinsic efficiency for the j-th gamma line; ε_d the geometric efficiency factor for standard source and ε_v the volume efficiency factor for volume cylindrical samples.

The intrinsic efficiency $\varepsilon_{\mathrm{int}\,j}$ is the probability that a gamma ray that enters the detector unit will interact and give a pulse in the full-energy peak. The geometric efficiency ε_d is the fraction of emitted photons that are intercepted by the detector. The values of $\varepsilon_{\mathrm{int}\,j}$ and ε_d were determined experimentally by traditional calibration using standard ^{152}Eu source for different detector–source spacings. The volume efficiency ε_v is the fraction of photons emitted in the solid angle subtended by the detector and virtual standard source surface located in the sample base center. The factor (ε_v) accounts for the differences in volume between the measured large sample and the standard source. It is obvious that ε_v depends on three variables, which are the sample radius and height and the detector–sample spacing. This factor is equal to unity for standard source. To determine this factor experimentally for specific detector–sample spacing it was assumed that

$$\varepsilon_v = \varepsilon_r\varepsilon_h \quad (3)$$

where ε_r and ε_h are factors account for the radii and heights differences between the standard source and large sample.

According to this equation and the abovementioned definitions, it is obvious that ε_r and ε_h will be unity for standard source and very thin sample, respectively.

By substituting this relation in relation (1) and rearranging we got

$$A_{ij} = \left(\frac{1}{\varepsilon_{\mathrm{int}\,j}\varepsilon_d}\right)\left(\frac{1}{\varepsilon_r}\right)\left(\frac{Att_j}{\varepsilon_h}\right)\left(\frac{cps_j}{B_{ij}}\right) \quad (4)$$

This relation shows that the measured large sample activity is a multiplication of four factors. The first factor terms are simply the standard source calibration parameters ($\varepsilon_{\mathrm{int}\,j}$, ε_d). The fourth factor terms are either measured easily (cps_j) or tabulated in references (B_{ij}) (JEF- PC Version 2). The second and third ($1/\varepsilon_r$, Att_j/ε_h) factors need to be determined. Determination of these factors using self-radiation of large samples forms the main problem of this work. This problem will be solved for NORM samples using its multi-gamma lines radiation, which cover wide range of energies (Fig. 1).

The radioisotope measured activities using any of its gamma lines are identical. So, if $j1$ and $j2$ are the low and high energy gamma lines emitted by the concerned isotope or another in secular equilibrium with it, then the measured activities using these lines are equal. Thus

$$A_{ij1} = A_{ij2} \quad (5)$$

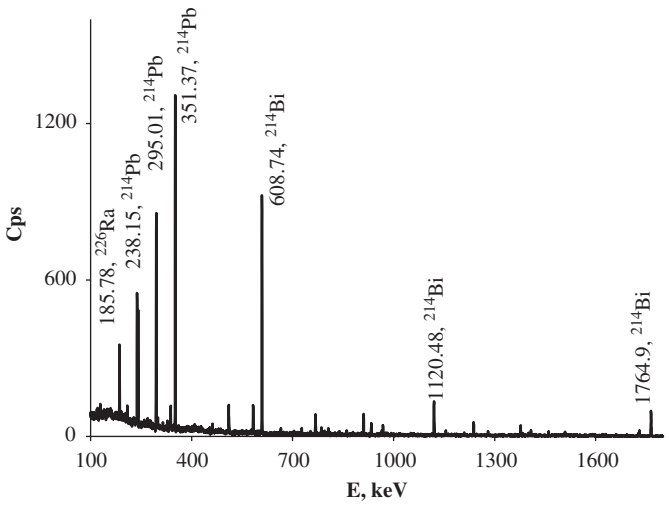

Fig. 1. Gamma spectrum of NORM sample.

The line of less energy will be attenuated more than that of higher energy in the measured sample, so Att_j value for $j1$ will be higher than that of $j2$. The Att_{j1}/Att_{j2} value can be expressed quantitatively by substituting relation (4) in (5) and rearrangement to get

$$\left(\frac{Att_{j1}}{Att_{j2}}\right) = \left(\frac{\varepsilon_{\mathrm{int}\,j1}}{\varepsilon_{\mathrm{int}\,j2}}\frac{B_{ij1}}{B_{ij2}}\right)\left(\frac{cps_{j2}}{cps_{j1}}\right) \quad (6)$$

This ratio will be more than unity in general and it will be equal to unity when the attenuation for both gamma lines is negligible. This idea will be used in this work as a criterion to decide when the attenuation can be neglected.

The specific activity per radioisotope (i), which is measured using its j-th gamma line, (α_{ij}) is defined as the activity of unit mass. Thus it can be expressed using relation (1) as

$$\alpha_{ij} = Att_j cps_j/(mB_{ij}\varepsilon_{tot\,j}) \quad (7)$$

The specific activities ratio for two cylindrical samples ($k1$, $k2$) prepared from one homogeneous NORM material can be calculated using relation (7) as

$$\frac{\alpha_{ij\,k2}}{\alpha_{ij\,k1}} = \left(\frac{Att_j}{B_{ij}\varepsilon_{tot\,j}}\frac{cps_j}{m}\right)_{k2} \Big/ \left(\frac{Att_j}{B_{ij}\varepsilon_{tot\,j}}\frac{cps_j}{m}\right)_{k1} \quad (8)$$

Noting that both samples were:

- prepared from the same material, so the ratio of their specific activities will be equal to one and
- measured using the same gamma line at the same source–detector spacing, so B_{ij}, $\varepsilon_{\mathrm{int}\,j}$, and ε_d will be identical for both.

According to these reasons and to the relation (4), the relation (8) can be rewritten as

$$1 = \left(\frac{1}{\varepsilon_r}\frac{Att_j}{\varepsilon_h}\frac{cps_j}{m}\right)_{k2} \Big/ \left(\frac{1}{\varepsilon_r}\frac{Att_j}{\varepsilon_h}\frac{cps_j}{m}\right)_{k1} \quad (9)$$

This relation will be used to determine ε_h and ε_r values for the prepared large samples as will be seen in the following sections.

3. Results

3.1. Experimental determination of ε_r

To determine ε_r, three very thin NORM samples of different radii [7 (standard disc source radius), 13, 24 mm] were prepared from the same homogenous NORM material. The two gamma