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Fast self-attenuation determination of low energy gamma lines

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HIGHLIGHTS

- Gamma spectrometric measurement of low energy gamma lines is required in many fields.
- A fast and simple method for the selfattenuation correction of 46.5 keV (^{210}Pb) 63.3 keV (^{238}U) has been developed.
- The method depends on counts ratio – selfattenuation correlation.
- Transmission technique overestimates selfattenuation correction.

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ABSTRACT

Linear correlation between self-attenuation factor of 46.5 keV (^{210}Pb) and the 1764 keV, 46.5 counts ratio has been developed in this work using triple superphosphate fertilizer samples. Similar correlation has been also developed for 63.3 keV (^{238}U). This correlation offers simple, fast, and accurate technique for self-attenuation determination of low energy gamma lines. Utilization of 46.5 keV in the ratio has remarkably improved the technique sensitivity in comparison with other work, which used similar concept. The obtained results were used to assess the validity of transmission technique.

1. Introduction

Gamma spectrometric measurement of low energy gamma lines is required in many fields. ^{210}Pb measurement is needed in contamination studies, which presents a diverse range of challenges in many industries such as phosphate, oil and gas (Al-Masri et al., 2014; Al-Masri and Haddad, 2012). It is also required in sediment dating (Zaborska et al., 2007). However, ^{210}Pb emits low-energy gamma photons (E) of 46.5 keV, which generally requires self-attenuation correction factor (F). Similarly, the 63.3 keV photon is widely used for ^{238}U (^{234}Th) measurement in environmental samples. The 63.3 keV photon has high F (Huy and Luyen, 2004; Lenka et al., 2009). F depends mainly on E, density, elemental composition and sample geometry. Transmission (T) technique is widely used to determine F (Al-Masri et al., 2013; San Miguel et al., 2005). However, T technique requires additional standard sources and procedures, which consume time and efforts. Moreover, the accuracy of T technique for F estimation was assessed; it was found that, T technique overestimates F (Jodłowski et al., 2014; Jodłowski, 2016). The counts ratio - self-attenuation correlation (*R*-F) technique was successfully used to determine F of E in the range from 186 keV up to 1764 keV (Haddad and Suman, 2006; Haddad and Albiyat, 2009; Haddad and Bouch, 2011; Haddad, 2014). This technique was used also to determine F of 46.5 keV, where the activity ratios of 911, 209 keV pair and 609, 295 keV pair were utilized (Długosz-Lisiecka and Bem, 2013). The 911 keV and 129 keV intensity ratio was applied for F of 63 keV in typical building materials and soil samples (Długosz-Lisiecka and Martyna, 2015). However, the used gamma lines (911, 209, 609, 295, 129 keV) have energies much higher than 46.5 and 63 keV. Thus, their sensitivities to the changes in the sample, such as height, density, and effective atomic number, are less than the corresponding value of 46.5 and 63.3 keV. Therefore, the activity ratios of these gamma lines might not represent 46.5 and 63.3 keV attenuation accurately.

^{214}Bi radionuclide is characteristic of samples containing naturally occurring radioactive material (NORM). Its 1764.49 keV (^{214}Bi) gamma line has interesting properties (coincidence-free, interference-free, reasonable branching ratio of 0.15) (Yucel et al., 2010a, 2010b). Therefore, this line was selected in this work to

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study the correlation of cps1764/cps46 and cps1764/cps63 ratios with F of 46.5 keV and 63.3 keV in triple superphosphate fertilizer (Fer). F values were determined in the sample matrix and the obtained results has been compared to estimate the accuracy of T technique.

2. Theory

2.1. Counts ratio – self-attenuation correlation technique

The activity of radionuclide (i) in cylindrical sample is determined using the measured count rate of its gamma line (j) by (Haddad, 2014):

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

where: ε_{intj} – detection unit intrinsic efficiency for j-th gamma line; ε_d – detector-sample spacing factor; ε_r and ε_h – are factors account for the radii, heights differences between the standard source and cylindrical sample respectively; F_{cj} and F_j – are true coincidence and self-attenuation corrections for j-th gamma line respectively; B_{ij} – branching ratio; cps_j – the measured count rate.

The activity ratio of two isotopes i1, i2 in the sample is calculated using their gamma lines j1 and j2 by (1) as:

$$\frac{A_{i2j2}}{A_{i1j1}} = \frac{F_{cj2}}{F_{cj1}}\frac{F_{j2}}{F_{j1}}\frac{cps_{j2}}{cps_{j1}}\frac{\varepsilon_{\mathrm{int}\,j1}B_{i1j1}}{\varepsilon_{\mathrm{int}\,j2}B_{i2j2}} \quad (2)$$

Rearranging Eq. (2) and denoting $cps_{j2}/cps_{j1}$by $R_{j2,j1}$ gives:

$$R_{j2,j1} = \frac{cps_{j2}}{cps_{j1}} = \left[\left(\frac{A_{i2j2}}{A_{i1j1}}\right)/\left(\frac{\varepsilon_{\mathrm{int}\,j1}B_{i1j1}}{\varepsilon_{\mathrm{int}\,j2}B_{i2j2}}\right)\right]\frac{F_{cj1}}{F_{cj2}}\frac{F_{j1}}{F_{j2}} \quad (3)$$

This relation was applied in this work for j2 constant value of 1764 keV. The $R_{j2,j1}$ values were determined in the samples for j1 = 46.5, 63.3 and 609 keV. The F_c values for these gamma lines 46.5, 63.3, 609 and 1764 keV are 1.00, 10.00, 0.81 and 0.99 respectively (Yucel et al., 2010a, 2010b). The samples were prepared from the same material and measured in the same experimental setup and geometry. Thus, the $(A_{i2j2}, A_{i1j1}, \varepsilon_{\mathrm{int}\,j1}, \varepsilon_{\mathrm{int}\,j2}, B_{i1j1}, B_{i2j2})$ values are constant and consequently, Eq. (3) is reduced to:

$$R_{j2,j1} = K\frac{F_{j1}}{F_{j2}} \quad (4)$$

where: K – constant.

The constant K includes the (A_{i2j2}/A_{i1j1}) ratio. Therefore, Eq. (4) is valid regardless of the radioactivity disequilibrium between i2 and i1 radionuclides. The 1765 keV is a high-energy gamma line, therefore its self-attenuation is negligible(as will be confirmed experimentally), i.e, $F_{j2} = 1$ then Eq. (4) is reduced and reformulated to:

$$R_{j2,j1} = KF_{j1} \quad (5)$$

This Eq. shows linear correlation between $R_{j2,j1}$ and F_{j1}, thus theoretical base for $R_{j2,j1}$-F_{j1} linear correlation technique has been formulated.

2.2. Self-attenuation factor determination by transmission technique

Gamma spectrometric measurement in laboratories is performed generally either in near counting geometry, or in far counting geometry. In the near counting geometry the dimensions of both sample and detector are comparable with the sample – detector spacing. Whereas, in the far counting geometry the dimensions of both sample and detector are negligible compared to the sample – detector spacing. In the far counting geometry, F_j in a sample is determined by T technique using mass attenuation coefficient of the sample (μ_j). It is possible to determine μ value for a particular material through calculation and computational simulation, but it is not practical to do this for environmental samples due to the need to ascertain the chemical composition of each sample. Instead, it is usually more useful to determine μ_j by sample T measurement. Therefore, this technique for F_j determination is known by T technique and the obtained value is denoted by $F_j(T)$. The $F_j(T)$ in cylindrical sample of height (h) in the far counting geometryis calculated by (Reilly et al., 1991):

$$F_j(T) = \frac{\mu_j h}{1 - \exp(-\mu_j h)} \quad (6)$$

where: μ_j – the mass attenuation coefficient of the material for the j-th gamma line.

Eq. (6) was derived for the far counting geometry, so it is valid for this case only. However, it is widely used as an approximation in the near field geometry.

3. Experimental

3.1. Samples preparation

The developed theoretical formulation was validated experimentally using cylindrical samples of triple superphosphate fertilizer (Fer). The material was dried, powdered, homogenized, and sieved using 500-μm mesh. Eight cylindrical samples of fixed diameter (34 mm) and variable heights (3 – 38) were prepared in polyethylene capsules (Table 1).

3.2. Gamma spectrometric measurement

Samples measurement was performed using gamma spectrometry (Eurysis systems, Lenglshiem) using high resolution (FWHM equals to 2 keV at 1.33 MeV), high relative efficiency (80%) and low background shield HPGe detectors; INTERWINNER 0.4: spectroscopy software. Efficiency calibration was performed using reference samples (IAEA/RGU-1 & IAEA RGTH-1) provided by the International Atomic Energy Agency. The prepared samples were measured on the detector cap. The gamma lines of 46.54 keV (^{210}Pb), 63.3 keV(^{238}U), and (609 keV, 1764.49 keV) (^{214}Bi) were measured. All measurements in this work were performed in the same conditions. The counting statistic, whose uncertainty did not generally exceeds 3%, was the main source of uncertainty. Thus, the overall uncertainty was about 5%.

3.3. μ_j measurement

Standard point sources of ^{152}Eu, ^{241}Amand ^{210}Pb were positioned above a sample, which is located on the detector, and the number of counts in the full energy peak is measured. The intensity of gamma rays at a specific energy is easily taken from the peak counts for that energy in a gamma ray spectrum. Therefore, it is possible to measure the counts of each gamma line emitted by a radioactive point source as it has been attenuated through the sample of interest. In order to determine the initial non-attenuated intensity of the point source, it is necessary to obtain a

Table 1
Masses (g) of the triple superphosphate fertilizer in the samples.

h (mm)	3	8	13	18	23	28	33	38
m (g)	14.1	37.7	61.3	84.9	108.5	132.0	155.7	179.3