A calibration transmission method to determine the gamma-ray linear attenuation coefficient without a collimator

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HIGHLIGHTS

- The gamma-ray linear attenuation coefficient of a compound is determined using wide-beam collimation-free transmission measurements, and calibration curves.
- An experimental procedure is presented and applied to test samples.

ARTICLE INFO

ABSTRACT

It is shown that the gamma-ray linear attenuation coefficient of a sample with unknown chemical composition can be determined through a systematic calibration of the correlation between the linear attenuation coefficient, gamma-ray energy and the relative degree of attenuation. For calibration, H_2O, MnO_2, $NaCl$, Na_2CO_3 and $(NH_4)_2SO_4$ were used as reference materials. Point-like gamma-ray sources with modest activity of approximately 37 kBq, along with an HPGe detector, were used in the measurements. A semi-empirical formula was derived to calculate the linear attenuation coefficients as a function of the relative count rate and the gamma-ray energy. The method was applied to the determination of the linear attenuation coefficients for K_2CrO_4 and SiO_2 test samples in the same setup used in calibration. The experimental result agreed well with the ones calculated by elementary data.

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1. Introduction

Self-attenuation in gamma-ray spectrometry of radioactive voluminous samples affects the determination of measured radioactivity, especially for nuclides emitting low energy gamma-rays. The attenuation coefficient of a sample needs to be known to correct for self-attenuation. The attenuation coefficient is typically determined using the ratio of transmitted and incident gamma-ray intensities with well-collimated sources and detectors. However, this method requires either an intense gamma-ray source or a long counting period source, to compensate for the loss in photon counting due to the collimation process. If the chemical composition of a sample is known, the linear attenuation coefficient can be also calculated using elementary data, provided by a library such as XCOM (Hubell, 1982; Berger et al., 2010). However, measuring a sample's composition is tedious and costly. A more useful convenient method to determine the linear attenuation coefficient is desirable. Therefore, uncollimated transmission methods were

introduced and applied to determine the linear attenuation coefficient or directly the self-attenuation correction factor (Cutshall et al., 1983; Sima and Arnold, 2002). The following approximate mathematical model to determine the degree of attenuation without collimation was established and validated for H_2O, MnO_2, $AMP((NH_4)_3[PO_4Mo_{12}O_{36}] \cdot 3H_2O)$ and KCl by Byun and Lee (2005):

$$\frac{I_m}{I_0} = \frac{\int_\theta e^{-\mu_l \, t(\theta)} \left(1 - e^{-\mu_p \, l(\theta)}\right) d\theta}{\int_\theta \left(1 - e^{-\mu_p \, l(\theta)}\right) d\theta} \tag{1}$$

where I_m is the transmitted photon's intensity through a material, I_0 is the incident photon's intensity, μ_l and μ_p are respectively, the linear attenuation coefficients of the test sample and a Ge detector crystal, and $t(\theta)$ and $l(\theta)$ are photon path lengths through the test sample and detector crystal, respectively. However, the mathematical modeling method given in Eq. (1), in addition to being approximate, requires complicated calculations with knowledge of detector geometry and material.

This study aims at finding a simple and effective experimental method to determine the linear attenuation coefficient for samples with unknown matrix material without any chemical composition

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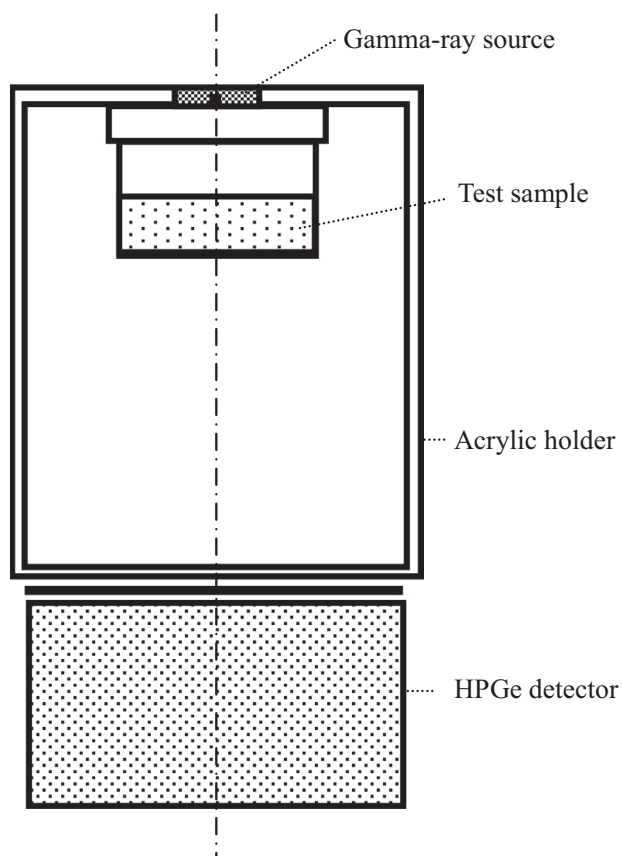   *E-mail address:* k975bji@kins.re.kr (J.-I. Byun).

Fig. 1. Experimental arrangement (not to scale).

analysis or collimating systems. This paper presents the experimental method and its application.

2. Materials and method

In order to determine the linear attenuation coefficient, μ_l, of an unknown sample, systematic calibration of the measurement system needs to be performed using reference materials. In this study, H_2O, MnO_2, NaCl, Na_2CO_3 and $(NH_4)_2SO_4$ were utilized as reference materials. The solid forms of these materials were grinded to particle size of less than 250 μm. Then, they were dried and filled to a level of 20 mm in a plastic cylindrical bottle with a diameter of 50 mm and a height of 50 mm. Source radiation was made incident on the flat surface of the cylinder. Apparent densities of H_2O, MnO_2, NaCl, Na_2CO_3 and $(NH_4)_2SO_4$ were measured and found to be 1000 kg/m³, 2916 kg/m³, 1386 kg/m³, 1471 kg/m³ and 1205 kg/m³, respectively. The gamma-ray standard sources used in this study were: ^{241}Am (59.5 keV), ^{133}Ba (276.4, 302.9 and 356.0 keV), ^{60}Co (1173.2 and 1332.5 keV), ^{137}Cs (661.7 keV), ^{152}Eu (121.8, 244.7, 344.3, 411.1, 444.0, 778.9, 964.1, 1112.1 and 1408 keV), ^{22}Na (1274.5 keV) and ^{210}Pb (46.5 keV), with an activity volume of ∅1 × 1 mm³. Each source had a radioactivity of 37 kBq, which is typical of calibration sources enclosed in an acrylic capsule with the diameter of 24.5 mm and a height of 3.2 mm. The distance from the center of the source to the external front face of the detector was set to 224 mm, in order to avoid the cascade summing effect. The transmitted gamma-rays were measured using a Canberra series BEGe (Broad Energy Germanium) detector, with relative efficiency of 60%. The diameter and length of the detector crystal are 91.6 mm and 32.2 mm, respectively. Each gamma-ray source was placed on the central axis of the front face of the detector end cap. The reference materials, gamma-ray sources and detector were arranged as shown in Fig. 1. For each of the seven standard gamma-ray sources, the count rare with an empty bottle, I_0, and reference materials, I_m, was measured over a counting period from 5600 s to 53000 s (depending on the sample and source) to provide ≤1% relative standard deviation for the energy of interest. The μ_l of reference materials was calculated using the XCOM program. Then, the linear attenuation coefficients for different gamma-ray energies and materials were plotted versus the ratio of count rates with and without the reference materials. The count rate ratios were plotted versus the gamma-ray energy. A semi-empirical formula was then derived from two fit-curves, to calculate the linear attenuation coefficients as a function of the count rate ratio and gamma-ray energy.

The validation of this method was carried out for K_2CrO_4 and SiO_2 test samples with the same geometric configuration as the calibrations samples. The apparent densities of K_2CrO_4 and SiO_2 filled in a bottle were 1652 kg/m³ and 1507 kg/m³, respectively. The gamma-ray measurements for these test samples were performed in the same manner as for the systematic calibration.

3. Results and discussion

As shown in Fig. 2, μ_l for H_2O, $(NH_4)_2SO_4$, NaCl, Na_2CO_3 and MnO_2, is plotted versus $\ln(I_m/I_0)$, and fitted with a straight line. The fitting results conform well to the linear fits over the broad ratio (I_m/I_0) range, with a correlation coefficient, $R^2 \geq 0.9975$ in all cases. In order to obtain a more accurate systematic calibration curve over a wide range of I_m/I_0, μ_l for all reference material was plotted together and fitted by linear least squares method, as shown in "All" of Fig. 2. The fitting formula for μ_l was

$$\mu_l = y_0 + a \cdot \ln(I_m/I_0) \tag{2}$$

The fit-parameters were assigned as follows: $y_0 = -8.504 \times 10^{-4}$ cm⁻¹, $a = -5.118 \times 10^{-1}$ cm⁻¹ in this study. The availability of this calibration line enables the determination of μ_l for a sample of unknown composition using its measured I_m/I_0 ratio.

The method was tested using for two samples of the K_2CrO_4 and SiO_2, fitted within the same size of container as used in the calibration. Fig. 3 shows the I_m/I_0 ratio recorded experimentally at different energies. These ratio measurements were fitted with a 6th degree polynomial versus energy.

$$f_R = \sum_{i=0}^{6} a_i \cdot \log(E)^i \tag{3}$$

where E is energy and a_i is a i-th fit-parameter. Then, the value of f_R at a given E can used in place of I_m/I_0 in Eq. (2) to obtain μ_l (cm⁻¹) of the sample at the designated energy. Fig. 4 shows μ_l obtained using these procedures for the two samples. Since the composition and density of these two samples were known, their attenuation μ_l values were obtained using XCOM (Berger et al., 2010). As Fig. 4 and Table 1 show, the experimentally obtained results agree well with those calculated using XCOM and the uncertainties were obtained by combining standard uncertainty for the measured gamma-ray count and standard fitting uncertainty. The relative standard counting statistical uncertainty was below 1% for all measurements in the energy of interest and the standard uncertainty ranges in the curve fit at energy of I_m/I_0 for K_2CrO_4 and SiO_2 were 0.5–9.1% and 0.5–1.2%, respectively. It should be kept in mind that the fit-parameters used in this work depend on the given experimental conditions, such as the arrangement of the calibration point-like source, detector and test sample as well as geometry, matrix and density of test samples. Therefore, to utilize this method to determine μ_l for an unknown sample, the sample