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Original Article

Coincidence summing correction for a voluminous ^{152}Eu source

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ABSTRACT

A code is developed to correct for the coincidence summing effect in detecting a voluminous gamma source, and this code is applied to a ^{152}Eu standard source as a test case. The source is 1000 mL of liquid in a cylindrical shape. To calculate the coincidence summing effect, the cylindrical source is considered as 10(radial) × 8(height) sectional sources. For each sectional source, the peak efficiency and total efficiency are obtained by Monte Carlo simulation at each energy for 10 energies between 50 keV and 2000 keV. The efficiencies of each sector are then expressed as polynomials of gamma energy. To calculate the correction coefficients for the coincidence summing effect, the KORSUM code is used after modification. The magnitudes of correction are 4%–17% for the standard ^{152}Eu source measured in this study. The relative deviation of 4.7% before the coincidence correction is reduced to 0.8% after the correction is applied to the efficiency based on the measured gamma line. Hence, this study has shown that a new method has been developed that is applicable for correcting the coincidence effect in a voluminous source, and the method is applied to the measured data of a standard ^{152}Eu cylinder source.

1. Introduction

When a gamma source with coincidently emitted multiple gamma rays is detected by using a single HPGe detector, coincidence summing effects are involved. The measured counts are required for a correction of the coincidence summing (CS) effect, typically the count loss effect, this problem was reported as early as the 1980s by ICRM (International Committee for Radionuclide Metrology) [1]. The CS correction is large and required when the source-to-detector distance is small or a large detector is used, which causes the full energy peak efficiency (peak efficiency) to be high. For example, the detection conditions that necessarily require the CS correction are measuring an environmental sample or nuclear material that emits a number of gamma rays during decay. Therefore, the CS correction must be considered to determine the activities when counting a volume source of large size.

The CS correction for measuring a point source was discussed earlier by several authors [2,3]. Recently, a study on CS correction reported that decomposition of the correction magnitude is possible by the order of the size when the related equations are re-expressed in matrix form [4,5]. For CS correction in volumetric sources, the study of Debertin [6] was the initiation, and then, their developments were followed by a calculation based on Monte Carlo simulation [7] and a new code GESPECOR [8,9]. The software package Genie 2000 of Canberra Inc. performs the CS correction for a point or a volume source by an efficiency calibration software (ISOCS/LabSOCS) [10]. In the code TRUECOINC, the volumetric source is considered as a point source and the correction factor is calculated by matrix formulation [11]. Other methods to find CS correction of volumetric source are based on using the efficiency transfer code EFFTRAN [12] or the ETNA [13]. Recently, Novković et al. have reported on their study of coincidence summing corrections for point and volume ^{152}Eu sources [14]. A proper treatment of this topic and the intercomparison of performance between different methods and codes are beyond the scope of the present study. There is a report on this topic based on a work of international intercomparison of methods [15]. In that study for the volume source, a large fluctuation was shown in the value of the CS correction factors calculated with different codes [16]. In result, the intercomparison has shown the CS factors are varied in different methods but it had led no conclusion about the accuracy of the CS correction. Hence, these studies indicate that the specific code or a certain computational method that produces the most accurate CS correction factor (CF) cannot be confirmed. In addition, the time to obtain the CS correction and the usage convenience are important

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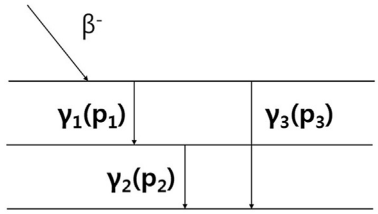

Fig. 1. A schematic level diagram showing three γ rays emitted after a beta decay.

aspects from a practical point of view, but this information was not given in the previous studies.

The code KORSUM [6] requires an input set of efficiencies and P/T ratios for 9 energies at 70 positions in a cylinder source. Since the code KORSUM is not applicable for the volumetric source in this study, a modified KORSUM code is developed such that a routine is provided to calculate the CS-CF for a volumetric source. The routine can address cylinder sources, Marinelli-type sources and arbitrary volumetric sources once the set of efficiencies for partial sectors is provided. In this study, a set of peak and total efficiencies is obtained for 80 sectional volumes at each of 10 gamma energies by MCNP simulations. The efficiencies are input to KORSUM, and each set is fit by a function of the gamma energy. The CS-CF values are calculated for the partial volume and finally combined as the CS-CF value for the full volume source. In the experiment, a ^{152}Eu standard source containing 1000 mL of liquid in a cylindrical shape is measured by an n-type HPGe with a relative efficiency of 45%. The source-to-detector distance is 0 cm. Since the source activity is known, a set of peak efficiencies for ^{152}Eu gamma rays is obtained, and the efficiencies before and after applying the CS-CF are compared.

2. Theoretical discussion

The coincidence summing effect is involved in detection of a nuclide simultaneously emitting two or more gamma rays. When the three gamma rays emitted from a point source with the three energy levels given in Fig. 1 are measured, the corresponding correction factors are given by:

$$C_1 = \frac{1}{1 - P_2 \cdot \varepsilon_{T2}} \tag{1}$$

$$C_2 = \frac{1}{1 - P_1 \cdot \varepsilon_{T1}} \tag{2}$$

$$C_3 = \frac{1}{1 + \frac{P_1 P_2}{P_3} \cdot \frac{\varepsilon_{P1} \cdot \varepsilon_{P2}}{\varepsilon_{P3}}} \tag{3}$$

where C_i is the correction factor applied to γ_i counts and is defined as the ratio (T_i/S_i) of true counts (T_i), which is the counts free from the coincidence effect, and the observed counts (S_i). ε_{pi} is the peak efficiency, ε_{Ti} is the total efficiency, and p_i is the emission probability of γ_i. The equations for a general scheme of decay are given in Refs. [2,3].

For the case of a volumetric source, the peak efficiency and the total efficiency in Eqs. (1)–(3) are a function of the source position **r** in the volume. Hence, the CS-CF is given by an expression involving integration over the source volume, assuming the density of the voluminous source is constant [6]:

$$\frac{1}{C_1} = 1 - P_2 \frac{\int \varepsilon_{p1}(\vec{r})\varepsilon_{T2}(\vec{r})dV}{\int \varepsilon_{p1}(\vec{r})dV} \tag{4}$$

$$\frac{1}{C_2} = 1 - P_1 \frac{\int \varepsilon_{p2}(\vec{r})\varepsilon_{T1}(\vec{r})dV}{\int \varepsilon_{p2}(\vec{r})dV} \tag{5}$$

$$\frac{1}{C_3} = 1 + \frac{P_1 P_2}{P_3} \cdot \int \varepsilon_{P1}(\vec{r}) \cdot \varepsilon_{P2}(\vec{r})dV \Big/ \int \varepsilon_{P3}(\vec{r})dV \tag{6}$$

In detail, since the peak efficiency and the total efficiency in Eqs. (4)–(6) are functions of (r,z) and γ energy (E_γ), it is never a simple task to prepare the functions $\varepsilon_p(r,z,E_\gamma)$, $\varepsilon_T(r,z,E_\gamma)$ and integrate them over the source volume. In this study, we try an approximate solution in which the CF is obtained as a sum of partial volume's value where the source volume is comprised of N-sectional volumes of small size. The partial volume is considered a uniform source in the description for Monte Carlo simulation of gamma emission and detection. By obtaining the CFs of all the sectors, the CFs for the overall volume are given by the following argument.

When the measured intensity is s_k, the true intensity t_k, and the CS-CF C_k for the given gamma-ray emitted from the k-th sector, the corresponding quantities for the whole volume are denoted by S_V, T_V, and C_V, respectively. By denoting the location of the sector, r_k, they are related by

$$s(\overrightarrow{r_k}) = t\frac{(\overrightarrow{r_k})}{C_k} \tag{7}$$

Since the CF for the whole volume is given as

$$C_V = \frac{T_V}{S_V} = \sum_{k=1}^{\infty} t(\overrightarrow{r_k})\Delta V_k \Big/ \sum_{k=1}^{\infty} s(\overrightarrow{r_k})\Delta V_k \,, \tag{8}$$

then, by combining with Eq. (7), we obtain

$$\frac{1}{C_V} = \sum_{k=1}^{\infty} s(\overrightarrow{r_k})\Delta V_k \Big/ \sum_{k=1}^{\infty} t(\overrightarrow{r_k})\Delta V_k \cong \sum_{k=1}^{N} [t(\overrightarrow{r_k}) / C_k]\Delta V_k \Big/ \sum_{k=1}^{N} t(\overrightarrow{r_k})\Delta V_k \tag{9}$$

Since the true intensity of the k-th sector is $t(\overrightarrow{r_k}) \propto p\varepsilon_p(\overrightarrow{r_k})$ (p: emission probability), the CF for the whole volume is given by

$$\frac{1}{C_V} \cong \sum_{k=1}^{N}[t(\overrightarrow{r_k})/C_k]\Delta V_k \Bigg/ \sum_{k=1}^{N} t(\overrightarrow{r_k})\Delta V_k = \sum_{k=1}^{N}[\varepsilon_p(\overrightarrow{r_k})/C_k]\Delta V_k \Bigg/ \sum_{k=1}^{N}\varepsilon_p(\overrightarrow{r_k})\Delta V_k \quad (10)$$

The KORSUM [6] code was written in Fortran-4. In this study, the code is rewritten in MATLAB [17] by translating the algorithm. A routine is also developed and added to KORSUM to calculate the volumetric CF by Eq. (10). To distinguish this from the original KORSUM, the code is referred to as KORSUM-Mat. In KORSUM-Mat, the treatment of the volumetric source has been revised. The peak efficiency and the total efficiency are given as polynomials of gamma energy obtained by fitting to the input data. The present method has the merit that the correction factor C_k is obtained for all k sectors, and then, the values are combined to give the factor of the whole volume by weighting (ΔV_k). Hence, the coefficient can be obtained for a volume of arbitrary size once the corresponding peak and total efficiencies are given as the input. This feature is an improvement from KORSUM, which can calculate only the cases of a point source and a cylindrical source where the efficiencies are represented by a fit to a set of those at 70 positions and for 9 energies. The present method can calculate CFs, by using Eq. (10), for the case of an arbitrary number of sectors having different volumes.

3. Experiment

To assess the effect and accuracy of the CS-CF calculation, a comparison is made for the peak efficiencies without CS correction and those with correction. The peak efficiencies without CS correction are obtained by measuring a standard ^{152}Eu volumetric source by an n-type HPGe (ORTEC) detector whose specification is shown in Table 1.

The ^{152}Eu volumetric source is prepared and standardized by the Korea Research Institute of Standards and Science (KRISS). The activity of the source is 86.2 ± 1.5 kBq (Date: 2013 May 1). The source is distributed uniformly inside a cylinder (100 mmφ, 149 mmH) filled with 0.1 M HCl and has an active volume of 1000 ml. The geometry of the source-detector is coaxial, and the distance of the source to the detector is 0 cm. A lead shield surrounds the detector to reduce the background from the room floor and wall.

The spectrum was acquired for 80000 s. The peak analysis proceeds for gamma rays with an emission probability of 1% or more [18], and the obtained spectrum is analyzed by HyperGam [19] for peak fitting and area determination. Each peak area is converted to the peak efficiency by using the source activity and the gamma emission probability [18].

4. Coincidence summing correction

Two sets of input are required for KORSUM-Mat: one for the decay scheme of the source nucleus and the other for the efficiencies of the detector. The input for the decay scheme includes the type of decay, the levels and the transitions between the levels, the transition energy, the gamma emission probability and the internal conversion coefficient for the transition, etc. The level and decay data are taken from a recent database [18]. Each decay scheme is prepared for the ^{152}Eu decay branch of EC+β^+ and β^- decay. X_K radiation is also considered.

The detector's information includes a set of peak efficiencies, total efficiencies at a number of gamma energies and the volume fraction of the source sector. These data are repeated for all sectors of the volume source. To prepare the set of peak and total efficiencies for the given sector within the cylinder, a number of Monte Carlo (MCNP5) [20] simulations were performed. The geometry of the simulation is the same as that of measurement - coaxial and a 0 cm distance between the cylinder source and the detector. The geometry given in the MCNP simulation is sketched in Fig. 2. In the simulation, the source volume is partitioned into 80 sectors, which are 10 equi-divisions in the radial direction and 8 equi-divisions in height. In each run, only one of the sectors is set as the mono-energetic gamma source, and the run is repeated for each of 10 energies in the range 50–2000 keV. An overall 800 runs are performed for the history of 10^9 - 10^{10} particles that are generated uniformly in the simulated source sector, and the number of histories increased in proportion to the volume of the sector. Hence, the energy spectrum absorbed in the detector crystal is obtained from each run. From a simple counting of the spectrum, the peak efficiency and the P/T ratio are obtained for the gamma energy. The

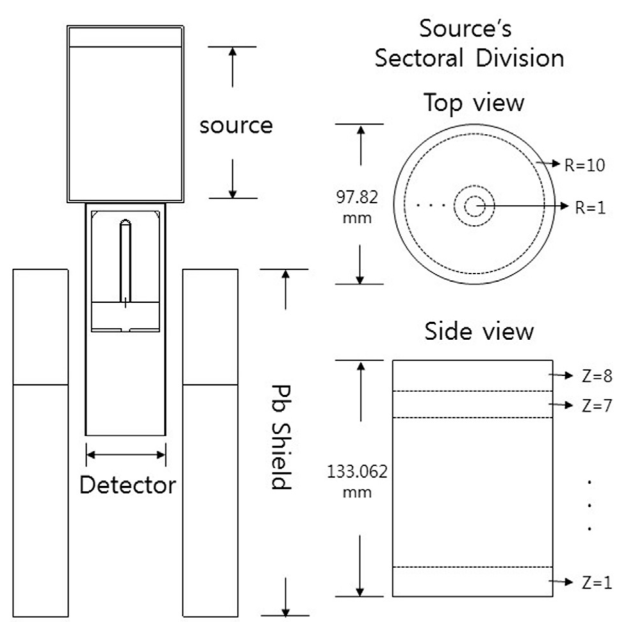

Fig. 2. The geometry used in the experiment and simulated by the code MCNP5.

Table 1
Specification of the HPGe detector used in this work.

Detector	Relative efficiency [%]	type	Crystal Diameter [mm]	Crystal Length [mm]	Window thickness [mm]
HPGe	45	n-type	58.1	79	0.5(Be)

calculated peak efficiencies for the sectors are summed to give the efficiencies of the overall volume and are fitted as a polynomial function of gamma energy. The calculated peak efficiencies are then normalized to the corresponding measured efficiencies at the energies of ^{152}Eu γ rays. Then, the same normalization factor is applied to the calculated peak efficiency for each source sector. The total efficiency is obtained by using the normalized peak efficiency and the P/T ratio. They are also calculated for each of the energies and the sectors in this work. Fig. 3 shows the calculated total efficiency and peak efficiency of several sectors after normalization is applied, where R denotes the radial indices 1–10 for the 10 equi-partitions in the radial direction (r) and Z the height indices 1–8 for the equi-partitions in the axial direction (z).

5. Results and discussion

In this study, the CS-CFs of detected γ rays emitted from a 1000 ml cylinder source of ^{152}Eu are obtained for the 14 ^{152}Eu γ rays of different energy by a sum of those for the individual sectors. The

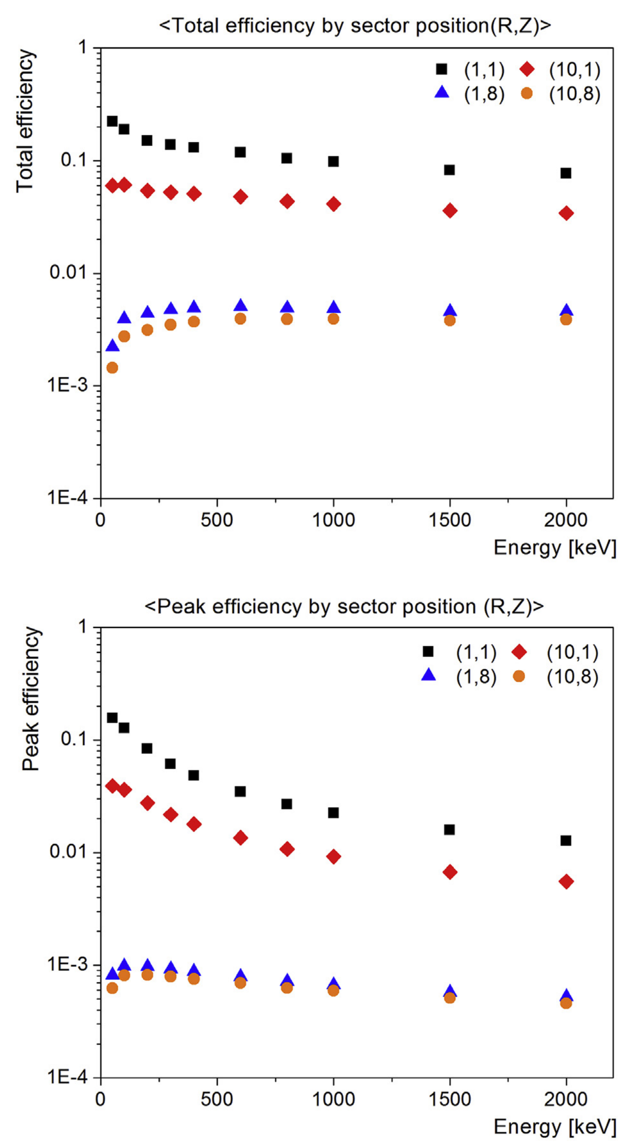

Fig. 3. Total efficiency (top) and peak efficiency (bottom) for some sectors calculated by MCNP5 simulation. In sector positions (R,Z) indices, R is numbered 1–10 in the radial direction and Z is numbered 1–8 in the height direction.

Table 2
Correction factor for ^{152}Eu gamma rays measured in this study.

Decay mode	Energy (keV)	Correction factor
EC(β+)	121.78	1.1235
	244.7	1.1674
	443.97	1.1375
	867.38	1.17
	964.082	1.1154
	1085.84	1.036
	1112.08	1.0977
	1212.95	1.0553
	1408.01	1.101
β-	344.28	1.0377
	411.12	1.0961
	778.91	1.0551
	1089.74	1.0491
	1299.15	1.0758

CS-CF is based on the peak and the total efficiency set obtained from a number of Monte Carlo simulations by MCNP code. All the CS-CFs for the sectors are summed to give the CS-CF for the counting of the corresponding γ ray. Table 2 lists the calculated correction factors for the ^{152}Eu γ rays measured in this study. The magnitude of them is in the range of 4%–17%.

By applying the CS-CFs to the measured peak areas, or equivalently to the measured efficiencies, the peak efficiency free from the coincidence effect (or the true peak efficiency) is obtained. Fig. 4 shows a comparison of the measured peak efficiency and the true peak efficiency for the ^{152}Eu γ rays. Fig. 5 shows the relative deviation, which is given by the residual data from the fit curve. The set of efficiencies without CS correction shows a relative deviation of 4.7% from the fit curve over the gamma energies, while the deviation is improved to 0.8% after the correction is applied for the CS effect. Therefore, the correction performed in this study is proven effective and useful. Although the simulation of γ rays is a tedious, time-consuming process, it must be performed once for each combination of the volumetric source and the detector under study, for example, as a part of detector characterization. Automating the continuous Monte Carlo simulation will save some processing time.

6. Conclusion

A general method is developed to obtain CS-CFs for a volumetric γ source. In this approach, the source volume is divided into a number of sectors. By considering the source sectors successively

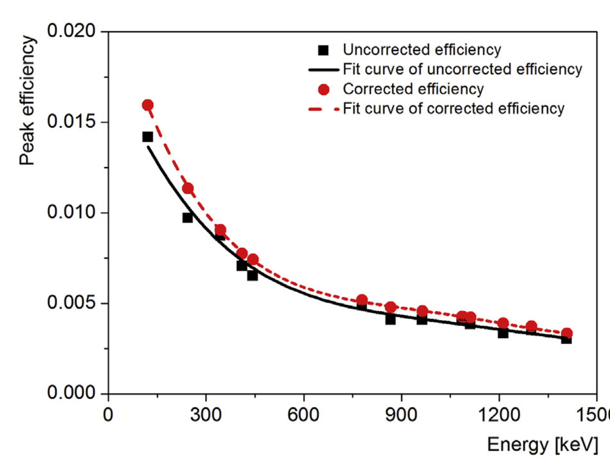

Fig. 4. Comparison of ^{152}Eu peak efficiency calibration curve before and after applying CS-CF.

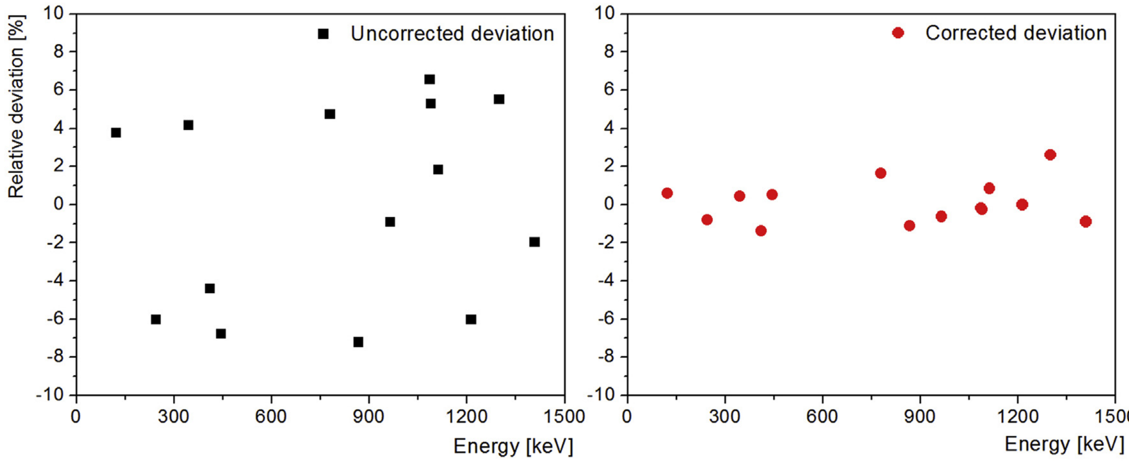

Fig. 5. Relative deviation of the data points from the fitted curve (left: uncorrected vs. right: corrected). Relative deviation [%] means the residual divided by the peak efficiency and multiplied by 100.

through Monte Carlo simulations by MCNP, the whole set of peak and total efficiencies is obtained in a range of 50–2000 keV and for the configuration of the volume source and the detector used in this study. The source considered in this study is a 1000 ml standard cylindrical source containing ^{152}Eu dissolved in 0.1 M HCl, which is measured using an HPGe of 45% efficiency. By a measurement of the source at zero distance, the 14 γ peaks are analyzed in the spectrum to give peak efficiency in the range of 122–1408 keV. The corresponding CS-CFs are calculated using the modified code KORSUM-Mat with the input set of efficiencies prepared by a number of MCNP simulations. In the simulation, the source cylinder is divided into 10(r) $\times$ 8(z) partitions, and each partition is given as the gamma source in a successive run of MCNP.

When the CS-CFs are applied, a new set of corrected and consistent efficiencies are obtained for the ^{152}Eu γ lines. Comparing the measured efficiency obtained from the experiment with the corrected efficiency curve using the CS-CFs, the absolute value of the relative deviation, which accounts for the degree of agreement with the fit curve, is greatly reduced. This result shows that the calculated CS-CFs are effective in producing a consistent set of efficiencies after correction for the coincidence summing effect.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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