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Determination of full-energy peak and total efficiency functions for Ge-detectors by iteration calculation using a mixed radionuclide source

H. Ishizu[a,*], T. Yamada[b]

[a] Japan Radioisotope Association, 28-45, Honkomagome 2, Bunkyo-ku, Tokyo 113-8941, Japan
[b] Atomic Energy Research Institute, Kindai University, 3-4-1, Kowakae, Higashiosaka City, Osaka 577-8502, Japan

HIGHLIGHTS

- Peak and total efficiencies of a well-type and a p-type Ge detector were determined.
- Efficiency functions were estimated by not only normal peaks but also sum peaks.
- The iteration was performed using the "Solver" add-in in Excel™.
- Not only point source but also volumetric source was examined.
- Squared-to-linear curve was applied to the experiment of volumetric sources.

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ABSTRACT

Empirical calibrations of full-energy peak and total efficiency curves for well-type and coaxial p-type germanium detectors were carried out using several combinations of practical nuclides. As a result, reliable efficiency functions of both types of detectors were successfully obtained using a home-made program coded with Visual Basic for Applications (VBA) and the "Solver" add-in in Excel™. We also examined the applicability of our advanced program to volumetric sources using squared-to-linear curve technique.

1. Introduction

True coincidence summing arises in case of nuclides which emit cascade gamma-rays within the resolving time of a spectrometer system, especially when the source is positioned close to the detector (Marinelli geometry or well-type detector). In such a case, not only the full-energy peak efficiency but also the total efficiency is required to correct coincidence summing effects. In the conventional approach, single photon nuclides are preferably used to determine these two efficiency functions. However, nuclides with summing effects such as ^{60}Co or ^{88}Y are also needed to obtain the peak efficiency curve ranging from 80 keV to 2000 keV, resulting in a significant error of the peak efficiency function in case of inaccurate summing corrections. In addition, since there are few summing-free nuclides available, especially in the high energy range, it is difficult to obtain the total efficiencies accurately by the direct empirical approach. Instead of such a conventional technique, Monte-Carlo simulation was successfully adapted by several authors (Lépy et al., 2010). However it might be not easy to reproduce a real geometry of the detector (e.g. dead layer thickness and its aging variation, exact dimensions of the detector and housing, impurities of constitutional materials) or to replicate drift with time.

As another approach, Blaauw (1993) developed a direct calibration technique of full-energy peak and total efficiency curves using the multi-gamma emitting nuclide ^{82}Br. In our previous study, these two functions for a well-type germanium detector were successfully determined using ^{75}Se and ^{134}Cs sources instead of the short-lived ^{82}Br (Ishizu and Yamada, 2017), though very large coincidence summing effects were found in the measurement. However, by using these two nuclides, it is difficult to determine efficiency functions accurately in the energy range above 1.3 MeV. Also, it might not be easy to adapt this method straightforwardly to germanium detector calibration in the case of extended sources. To deal with this case, Blaauw and Gelsema (2003) also developed the squared-to-linear curve technique in order to expand the applicability of his technique to volumetric sources. Subsequently, the determination of the squared-to-linear curve, so-called LS-curve, using Monte Carlo simulation has been studied (Vidmar and Korun, 2006; Vidmar et al., 2007, 2011; Vidmar and Kanisch, 2010). Blaauw and Gelsema (2003) also suggested that the empirical calibration

* Corresponding author.
E-mail address: ishizu-hidetake@jrias.or.jp (H. Ishizu).

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method with LS-curve was applicable to calibration using mixed nuclide reference sources with a suitable combination of nuclides. However, which nuclides are suitable choices in the method is still unclear.

This problem was studied in the present work. We tested mixed point sources with several combinations of nuclides not only for a well-type germanium detector but also for a coaxial p-type one. All measurements were carried out using an X-ray attenuation filter made of brass or copper in order to avoid complicated calculations due to coincidence summing with X-rays. Unknown parameters, namely the coefficients of the three curves (full-energy peak efficiency, peak-to-total ratio and squared-to-linear), were calculated using the iteration algorithm based on least squares technique as shown in our previous study (Ishizu and Yamada, 2017). The iterations for directly calculating the coefficients of the efficiency curves were carried out with a home-made program coded with Visual Basic for Applications (VBA) and the"Solver" add-in in Excel™. Trial determinations of efficiency functions of a p-type coaxial germanium detector for a series of cylindrical sources were also examined in order to adapt this method to more practical uses.

2. Theoretical background

In case that coincidence summing effects with X-rays are negligible, peak areas of all possible peaks for a beta decay nuclide can be expressed according to the following equations.

$$N_E^{\mathrm{calc}} = \sum_N \left\langle N_N \sum_C \left[k_C \prod_{\substack{S(i,j)=1 \\ (i,j)\in P_C}} \left(\frac{1}{1+\alpha_{i,j}^{\mathrm{T}}} p_{i,j} \varepsilon_{i,j}^{\mathrm{p}} \right) \prod_{\substack{S(i,j)=0 \\ (i,j)\in P_C}} \left\{ p_{i,j} \left(1 - \frac{1}{1+\alpha_{i,j}^{\mathrm{T}}} \varepsilon_{i,j}^{\mathrm{t}} \right) \right\} \right] \right\rangle \tag{1}$$

Where N_E^{calc} is the calculated peak area corresponding to the full-energy peak of energy E, N_N the number of decays of the nuclide N which can contribute to the peak of energy E during the measurement time, C a definite decay path in the decay of nuclide N (up to the ground state of the decay product) which can contribute to the peak of energy E, P_C the set of all transitions (i,j) along the decay path C, k_C the probability of the decay of the parent nucleus to the highest level of the decay path C, $p_{i,j}$ the branching ratio corresponding to the transition from the i-th to the j-th state, $\varepsilon_{i,j}^{\mathrm{p}}$ the full-energy peak efficiency for the photon emitted in the transition (i,j), $\varepsilon_{i,j}^{\mathrm{t}}$ the total efficiency for the transition (i,j), $\alpha_{i,j}^{\mathrm{T}}$ the total internal conversion coefficient for the transition (i,j), $S(i,j)=1$ if the gamma-ray corresponding to the (i,j) transition belonging to the decay path C can contribute to the peak of interest, $S(i,j)=0$ in all the other cases of transitions belonging to the decay path C. $\sum_N$ means summation over all nuclides N which can contribute to the peak of energy E, and $\sum_C$ summation over all the decay paths C which can contribute to the peak of energy E.

The present calculation was carried out to minimize M described in Eq. (2). The minimization was done by iteration calculations using our home-made program.

$$M = \sum \frac{(N_E^{\mathrm{meas}} - N_E^{\mathrm{calc}})^2}{N_E^{\mathrm{calc}}} \tag{2}$$

Where N_E^{meas} is the measured peak area corresponding to the full-energy peak of energy E.

In the present work, we used the multi-gamma nuclides for the calibration, whereas the coincidence-free nuclides were used for the validation. Nuclear decay data from the Decay Data Evaluation Project website (http://www.nucleide.org/DDEP_WG/DDEPdata.htm) were used in this work. Even peaks of single photon emitters can contribute to the calculation of the efficiency curves using the present approach. The energy dependence of the functions of interest was the same approximation as in our previous study (2017):

$$\ln\{\varepsilon_{\mathrm{p}}(E)\} = c_1 \ln(E) + c_2 \qquad \text{for } E > 200\mathrm{keV} \tag{3}$$

$$\ln\{\varepsilon_{\mathrm{p}}(E)\} = c_3\{\ln(E)\}^2 + c_4 \ln(E) + c_5 \qquad \text{for } E < 200\mathrm{keV} \tag{4}$$

$$R_{\mathrm{pt}}(E) = \frac{\varepsilon_{\mathrm{t}}(E)}{\varepsilon_{\mathrm{p}}(E)} = c_6 E + c_7 \tag{5}$$

Where $\varepsilon_{\mathrm{p}}(E)$ is the full-energy peak efficiency, $\varepsilon_{\mathrm{t}}(E)$ is the total efficiency, $R_{\mathrm{pt}}(E)$ is the peak-to-total ratio, and $c_1 \sim c_7$ are unknown coefficients.

In order to calculate the coefficients of the squared-to-linear curve (required in the case of volume sources), an advanced version of our home-made program was coded with Excel™ VBA and the "Solver" add-in. In all the terms including products of two or more efficiencies in Eq. (1), each efficiency was replaced by the product of the efficiency and the corresponding squared-to-linear value $l(E)$. For example, the product of two efficiencies $\varepsilon(E_1)\varepsilon(E_2)$ was replaced by $l(E_1)\varepsilon(E_1)l(E_2)\varepsilon(E_2)$. In this study, in accordance with Blaauw and Gelsema's approach (2003), the following approximation function for the squared-to-linear curves $l(E)$ was used:

$$l(E) = c_8 + c_9 \ln(E)^{-c_{10}} \tag{6}$$

Where $c_8 \sim c_{10}$ are unknown coefficients.

In our previous study (Ishizu and Yamada, 2017), it was demonstrated that ^{75}Se and ^{134}Cs are successfully adapted to calculating coefficients of full-energy peak efficiency and peak-to-total ratio functions ranging from 66 keV to 1365 keV based on the empirical approach developed by Blaauw (1993). However, ^{75}Se is not convenient for the practical long-term use, due to its half-life of 120 days. In addition, due to its chemical toxicity, special care on source preparation procedure might be required. In the present study, ^{133}Ba was used to determine the efficiency functions in the low energy range. Though ^{133}Ba is useful to determine efficiencies of germanium detectors in the low energy range (e.g. Yoshizawa et al., 1983; Novković et al., 2009), the number of gamma-rays emitted per decay is not sufficient to obtain a reliable peak efficiency function using the present empirical calibration approach. Indeed, the gamma-rays of ^{133}Ba, having the energy below 200 keV are: 53.2 keV ($p_\gamma = 0.0214$), 79.6 keV ($p_\gamma = 0.0263$), 81.0 keV ($p_\gamma = 0.3331$) and 160.6 keV ($p_\gamma = 0.00638$). Since 79.6 keV and 81.0 keV have nearly the same energies, it is difficult to separate these two peaks due to the energy resolution of germanium detectors. Thus, only the data for 3 peaks are available below 200 keV. In addition, the 53.2 keV and 160.6 keV gamma-ray peaks have small areas as compared with other peaks and might not contribute significantly to the calculation of peak efficiencies, resulting in the divergence in the iteration calculation of the efficiency coefficients. To overcome this difficulty, ^{57}Co was also employed as a supplemental nuclide. There is a cascade between 14.4 keV and 122 keV lines, but due to the specific decay scheme and the presence of the filter, the coincidence with the low-energy line is not significant.

3. Experiments

3.1. Source preparation

We prepared eleven reference solutions containing ^{109}Cd, ^{57}Co, ^{139}Ce, ^{51}Cr, ^{85}Se, ^{137}Cs, ^{54}Mn, ^{60}Co, ^{88}Y, ^{133}Ba and ^{134}Cs. Radionuclidic purities of these solutions are better than 99%. Activity concentrations of these solutions were calibrated using a high pressurized ionization chamber which is traceable to National Metrology Institute of Japan (NMIJ). Then, these solutions were mixed and each activity concentration was determined according to the respective dilution factor. In order to prepare a point source with a known deposited activity,