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Coincidence summing corrections for point and volume ^{152}Eu sources

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HIGHLIGHTS

- The count rate equation system for ^{152}Eu was formed by DMM method.
- Coincidence summing corrections and peak and total efficiencies for point and volume sources were determined.
- Obtained results were compared with ones calculated by using the GESPECOR 4.2 software.

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ABSTRACT

In this article, the X-ray and gamma-ray coincidence summing effect in ^{152}Eu is studied. Coincidence summing corrections and peak and total efficiencies of point and volume sources were determined using the direct matrices multiplication (DMM) method. The theoretically evaluated peak count rates were found to be in good agreement with the experimentally obtained values. Validation was performed by comparing the calculated efficiency curves and the corresponding correction factors with the results obtained using GESPECOR 4.2 software.

1. Introduction

The radionuclide ^{152}Eu is widely used for energy and efficiency calibration of semiconductor HPGe spectrometers. The energy range of the emitted photons is broad, that is, 40–1769 keV, making this radionuclide suitable for the aforementioned purpose. ^{152}Eu disintegrates into ^{152}Sm by electron capture (72.1%) and positron emission (0.027%), and ^{152}Gd by beta minus emission (27.9%) with a half-life of 13.522 years (Vanin et al., 2004). Because the decay scheme of ^{152}Eu is very complex, the total number of gamma photon energies amounts to 132, in addition to K_α and K_β photons of ^{152}Sm, and ^{152}Gd. Thus, the spectrum analysis and the determination of efficiency curve are particularly complicated. Furthermore, coincidence summing, especially in a close-to-detector measurement geometry, requires accurate determination of correction factors (Debertin and Schötzig, 1979; Morel et al., 1983). Accurate determination of correction factors for ^{152}Eu point sources is an important topic that has been presented in many studies (Rizzo and Tomarchio 2010; Agarwal et al., 2011; Delgado et al., 2006; Arnold and Sima, 2004). Methods for the evaluation of coincidence summing correction factors for point sources were verified by the intercomparison performed by the Gamma Ray Spectrometry Working Group (GSWG) of the International Committee for Radionuclide Metrology (ICRM) (Lépy et al., 2010).

In some previously published studies, the authors described the direct matrices multiplication (DMM) method, which was experimentally checked with ^{152}Co, ^{133}Ba, and ^{75}Se point sources (Novković et al., 2007a, 2007b, 2012a, 2012b). Kanisch et al. (2009) compared the coincidence summing correction factors obtained by different methods and confirmed an excellent agreement between the DMM method and GESPECOR 4.2 software (2011) for the ^{133}Ba and ^{152}Eu point sources.

Calculating the coincidence summing correction factors and peak and total efficiencies of volume sources using Monte Carlo methods is very complex. Considerable effort has to be made to obtain accurate results. Some previously published articles deal with coincidence summing phenomenon in volume sources (Lee et al., 2008; Agarwal et al., 2011; Lian et al., 2005; Quintana and Montes, 2014; Vargas et al., 2014; Ramos-Lerate et al., 1997). In the intercomparison, recently organized by the GSWG, three volume sources filled with radioactive solution (^{152}Eu, ^{134}Cs) were considered, and the results obtained from different methods were compared: specifically dedicated codes (CCCC, CSCOR, ETNA, GESPECOR, KORSUM, and TRUECOINC), full Monte Carlo simulation

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(GEANT4), semi-empirical and simplified methods, and the numerical DMM method (Lépy et al., 2012). A total of 16 laboratories participated in the study and provided 23 series of results. Unfortunately, a large fluctuation of the results was observed, indicating the complexity of determining the volume source for coincidence summing correction factors. It is noteworthy that, despite the large discrepancies of many results, correction factors obtained by the DMM method showed satisfactory agreement with the mean values. Results of this intercomparison indicated that this method can be further improved for application to volume sources by finding better approximations (e.g. total-to-peak efficiency ratio) used in the numerical algorithms and consideration of angular correlation influence on the correction factors.

In this study, the DMM method was applied to ^{152}Eu point and volume sources (cylindrical geometry) to calculate the efficiency curve and the corresponding correction factors, and estimate the possibilities of application of the developed method to various matrices and geometries.

2. Theoretical approach to the coincidence summing of X-rays and gamma rays of ^{152}Eu

The theoretical method for obtaining the count rate equations was developed by Novković et al. (2007a) and successfully applied to ^{133}Ba (Novković et al., 2007b) and ^{75}Se (Novković et al., 2012a). This method was later named the Direct Matrices Multiplication method (Novković et al., 2012b).

The DMM method generates theoretical expressions for all peak count rates (single and sum peaks) in the spectrum as well as for the total count rate. Using this method, one can identify all possible decay paths and decay path outcomes, calculate all path outcome probabilities and the corresponding energy deposited in the detector, and determine theoretical expressions for count rates for each peak (excluding escape peaks) as well as total count rate, for cases in which the total and full-energy peak efficiencies and activity of the measured source are unknown. The unknown coefficients (decay scheme constants) characterize the measured radionuclide, which can be found in the literature (Novković et al., 2012b).

Because the decay scheme of ^{152}Eu is very complex, a detailed description of the application of the DMM method to this radionuclide would be difficult to describe, due to the large number of equations. Therefore, only the basic features will be provided.

2.1. Transition probability matrix for ^{152}Sm

The daughter nucleus ^{152}Sm has 19 excited levels, and the transition probability matrix formed using the nuclear decay data published by Vanin et al. (2004), has 21 rows and 21 columns, including the ground states of ^{152}Eu and ^{152}Sm. Elements of this matrix determine the transition probabilities between two levels. For example, if this matrix is denoted by **X**, then the transition probablities from ^{152}Eu to the ground state of ^{152}Sm in 1, 2, 3, 4, 5, and 6 steps are given by matrix elements $[\mathbf{X}]_{21,1}$, $[\mathbf{X^2}]_{21,1}$, $[\mathbf{X^3}]_{21,1}$, $[\mathbf{X^4}]_{21,1}$, $[\mathbf{X^5}]_{21,1}$, and $[\mathbf{X^6}]_{21,1}$ (elements in the first column of the 21st row), respectively (Table 1). The transition probability from

Table 1
Probabilities for the transition from the ground state of ^{152}Eu to the ground states of ^{152}Sm and ^{152}Gd in different number of steps.

Number of steps	1	2	3	4	5	6
Probability (^{152}Sm)	0.0005	0.1493	0.7013	0.1431	0.0061	0.000033
Probability (^{152}Gd)	0	0.2986	0.6351	0.0651	0.00012	0.0000014

Table 2
Number of decay paths for the transition from the ground state of ^{152}Eu to the ground states of ^{152}Sm and ^{152}Gd.

Number of steps	1	2	3	4	5	Total
Number of decay paths (^{152}Sm)	1	5	36	74	85	201
Number of decay paths (^{152}Gd)	0	6	20	27	17	70

the ground state of ^{152}Eu to that of ^{152}Sm in six steps is too small to be ignored. The probability of transition from ^{152}Eu to ^{152}Sm by positron emission was also ignored.

2.2. Transition probability matrix of ^{152}Gd

The transition probability matrix of ^{152}Gd has 17 rows and 17 columns, and the transition probabilities from the ground state of ^{152}Eu to that of ^{152}Gd are summarized in Table 1. The transition probability from the ground state of ^{152}Eu to that of ^{152}Gd in six steps is too small to be ignored.

2.3. Decay paths

The decay path is defined by the cascade transitions from the parent ground state to the daughter ground state. All possible decay paths were identified by a symbolic matrix **Y**, by replacing nonzero matrix element x_{ij} of the transition matrix with y_{ij}, and calculating $[\mathbf{Y^n}]_{21,1}$ ($n=1, 2, 3, 4, 5$) for ^{152}Sm and $[\mathbf{Y^n}]_{17,1}$ ($n=1, 2, 3, 4, 5$) for ^{152}Gd (see Novković et al. (2007a, 2007b)). The number of decay paths of ^{152}Eu to ^{152}Sm and ^{152}Gd are summarized in Table 2.

2.4. Decay path outcomes

In order to determine all decay path outcomes, the matrix element y_{ij} is replaced by a matrix row whose elements are probabilities of detection of K_α, K_β, and gamma photons, and nondetection of any photon:

$$y_{ij} = \left[p_{\alpha ij}, p_{\beta ij}, \gamma_{ij}, q_{ij}\right]. \tag{1}$$

The matrix row representing an electron capture with the transition to jth level of ^{152}Sm has three elements as

$$y_{21j} = \left[p_{\alpha 21j}, p_{\beta 21j}, q_{21j}\right]. \tag{2}$$

The matrix row representing beta decay of ^{152}Eu with the transition to jth level of ^{152}Gd has only one element as

$$y_{17j} = \left[q_{17j}\right]. \tag{3}$$

The matrix element y_{ij} is associated with the matrix row z_{ij}, whose elements contain energies deposited in the detector and correspond to the probabilities in y_{ij} as

$$z_{ij} = \left[E_\alpha, E_\beta, E_{\gamma ij}, 0\right], \tag{4}$$

for an electron capture with the transition to jth level of ^{152}Sm,

$$z_{21j} = \left[E_\alpha, E_\beta, 0\right], \tag{5}$$

and, for beta decay of ^{152}Eu with the transition to jth level of ^{152}Gd,

$$z_{17j} = [0]. \tag{6}$$

In the matrices presented above, E_α and E_β are the energies of K_α and K_β photons, respectively, $E_{\gamma ij}$ is the energy of gamma photon at $i \to j$ transition, and 0 corresponds to nondetection. In order to obtain decay path outcomes, probability matrices y_{ij} should be directly multiplied along their decay paths. For instance,