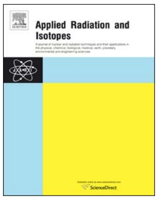
Applied Radiation and Isotopes 135 (2018) 131–134

Contents lists available at ScienceDirect

Applied Radiation and Isotopes

journal homepage: www.elsevier.com/locate/apradiso



Correction of coincidence summing effects for add-back mode measurements with a 4π clover detector using experimental total efficiency

Takahiro Yamashita[a], Hiroki Kamada[a], Yasuaki Kojima[b], Michihiro Shibata[b,*]

[a] Department of Materials, Physics and Energy Engineering, Nagoya University, Nagoya 464-8603, Japan
[b] Radioisotope Research Center, Nagoya University, Nagoya 464-8602, Japan

HIGHLIGHTS

- Standard sources were measured with 4-π clover Ge detector with add-back mode.
- Peak and total efficiency in add-back mode were determined between 0.03 and 2.7 MeV.
- Correction for coincidence summing was performed with measurements and simulation.

ARTICLE INFO

Keywords:
4π clover detector
Full energy peak efficiency
Total efficiency
Add-back mode
Standard source measurement
Monte Carlo calculation
Coincidence summing correction
GEANT4

ABSTRACT

We demonstrated coincidence summing corrections for the measured spectra of multi γ-ray emitters with the add-back mode of a 4π clover detector with an almost 98% solid angle condition using a Monte Carlo calculation based on nuclear decay data. The total and peak efficiencies were determined by Monte Carlo simulation code GEANT4 so that the experimental efficiencies measured mono/quasi-mono energetic γ-ray sources of ^{109}Cd, ^{139}Ce, ^{137}Cs, ^{54}Mn, ^{57,} ^{60}Co and ^{88}Y may be reproduced well. Under a large solid angle condition, (i.e., a large coincidence summing condition), the corrected peak efficiencies, deduced from ^{134}Cs and ^{152,154}Eu measured, were in agreement within 5% with peak efficiencies in the absence of coincidence summing. The coincidence summing effects were corrected properly according to the decay scheme information, using experimental and simulated values of the total efficiency. We thus demonstrated the effectiveness of measurement with the add-back mode of the detector for γ-ray spectroscopy.

A through-hole-type clover detector (4π clover detector) has four large volume n-type Ge crystals and a through hole in the center of the detector. Specifications for the diameter and length of each Ge crystal are 80 mm and 90 mm, respectively. The solid angle of point source subtended by each crystal is approximately 24.5%. In previous research, we determined full-energy peak efficiency for the detectors in singles-mode measurements (i.e., each Ge crystal working independently) with an uncertainty of about 3% by processing the coincidence summing correction with standard source measurements together with the Monte Carlo simulation code GEANT4 (Agostinelli et al., 2003). The detector also works as a large volume true-well-type detector in combination with an event-by-event mode data acquisition system, namely an add-back mode. This has very large detection efficiency, and is thus expected to determine γ-ray intensities of rare radioisotopes far from the β-stability line. Nevertheless, a large amount of coincidence-summing effects require proper correction.

In this paper, using the same approach as for the singles mode (Shima et al., 2014)—that is, source measurements together with the Monte Carlo simulation code GEANT4—we evaluated the coincidence summing correction between 0.03 and 2.7 MeV for the add-back mode. At first, experimental total efficiencies $\varepsilon_{t:exp}$ were determined with mono-energetic γ-ray sources and experimental full energy peak efficiencies $\varepsilon_{p:exp}$ were determined with quasi-mono γ-ray sources, which have relatively simple decay schemes, in addition to the mono-energy γ-ray sources. The $\varepsilon_{t:exp}$ and $\varepsilon_{p:exp}$ in the energy range from 0.03 to 2.7 MeV were then reproduced with $\varepsilon_{t:sim}$ and $\varepsilon_{p:sim}$ simulated by the Monte Carlo simulation code GEANT4, respectively, by adjusting the crystal dimensions. Next, the measured peak counts of the spectra of multi-en-

---

* Corresponding author.
E-mail address: i45329a@nucc.cc.nagoya-u.ac.jp (M. Shibata).

https://doi.org/10.1016/j.apradiso.2018.01.024

ergy γ-ray emitters ^{134}Cs, ^{152}Eu and ^{154}Eu with complicated decay schemes were then corrected by means of the developed coincidence summing correction program based on each decay scheme (Shima et al., 2014). Finally, the corrected ε_p, that is the corrected peak counts divided by the emission probabilities, were compared with $\varepsilon_{p:sim}$. The ratio of the corrected ε_p to the $\varepsilon_{p:sim}$ shows the practical uncertainties of coincidence summing correction for the radioisotopes; that is, the probable uncertainties of determining γ-ray intensities for unstable nuclei far off the β-stability line generally measured with the add-back mode of the detector.

The geometrical configuration of the measurements has been described elsewhere (Shima et al., 2014). Radioactive point sources, with activities of the order of a few kBq (approximate uncertainties of 2%) were set at the center of the through-hole sandwiched with a pair of 7-mm-diameter plastic β-ray absorbers.

Each output from the pre-amplifier of the four Ge crystals was independently put into a VME-based digital-signal-processing data acquisition system APV8008 (Techno AP Co.) and information about pulse and time was registered in event-by-event mode. The add-back spectra were produced by off-line sorting by setting a coincidence time of 600 ns between the crystals. This coincidence time shows the best peak-to-total ratios for the mono γ-ray sources under the counting rate of a few thousands counts per second.

The $\varepsilon_{t:exp}$ and $\varepsilon_{p:exp}$ were obtained from the measured counts divided by the γ-ray emission probabilities of nuclear data included in recommended data from the Laboratoire National Henri Becquerel (2013). The $\varepsilon_{t:exp}$ s were determined with the mono-energy γ-ray sources ^{109}Cd, ^{137}Cs, and ^{54}Mn and were reproduced by the Monte Carlo simulation GEANT4. The $\varepsilon_{p:exp}$ was determined between 34 keV and 2734 keV using mixed γ-ray sources containing ^{109}Cd, ^{57}Co, ^{139}Ce, ^{137}Cs, ^{54}Mn, ^{60}Co and ^{88}Y. The $\varepsilon_{p:exp}$ for ^{109}Cd, ^{137}Cs and Ba KX-ray, and ^{54}Mn did not need correction, written by $\varepsilon^{mono}_{p:exp}$ here. On the other hand, the $\varepsilon_{p:exp}$ s for quasi-mono γ-ray sources ^{139}Ce, ^{57}Co, ^{60}Co and ^{88}Y were in the presence of coincidence summing, written by $\varepsilon^{CS}_{p:exp}$ here. Those were corrected and deduced the $\varepsilon^{mono}_{p:exp}$ s as follows. In the case of ^{57}Co, the $\varepsilon^{CS}_{p:exp}$ s for the 122 and 136 keV γ-rays were corrected using the $\varepsilon_{t:sim}$ of the 14 keV γ-ray. In the case of ^{139}Ce, the $\varepsilon^{CS}_{p:exp}$ for 166 keV γ-ray was corrected using $\varepsilon_{t:sim}$ of the KX-rays of the daughter nucleus associated with electron capture. After the correction, the deduced $\varepsilon^{mono}_{p:exp}$ s of them agreed with $\varepsilon^{mono}_{p:sim}$ s. Similarly, the γ-rays of ^{60}Co and ^{88}Y, which have simple cascade relations, were corrected analytically using the $\varepsilon_{t:sim}$. By correcting the $\varepsilon^{CS}_{p:exp}$ s using the $\varepsilon_{t:sim}$, the deduced $\varepsilon^{mono}_{p:exp}$ s of the 1173 and 1332 keV γ-rays of ^{60}Co, and the 898, 1836 and 2734 keV γ-rays of ^{88}Y were in agreement with the $\varepsilon^{mono}_{p:sim}$ s. The $\varepsilon^{CS}_{p:exp}$ of the 2734 keV γ-ray is two orders of magnitude larger than the $\varepsilon^{mono}_{p:sim}$.

The deduced $\varepsilon^{mono}_{p:exp}$ and $\varepsilon_{t:exp}$ were reproduced with by the Monte Carlo simulation GEANT4. In the geometrical configuration used in the GEANT4 simulation, crystal size, dead layer and shielding materials of the detector were taken into account. It was not possible to reproduce well the values of $\varepsilon^{mono}_{p:exp}$ and $\varepsilon_{t:exp}$ with the same detector geometries. Therefore different detector geometries, optimized separately for $\varepsilon^{mono}_{p:exp}$ and $\varepsilon_{t:exp}$ by adjusting the crystal dimensions, were adopted (Table 1). The reason for the differences between the best geometries of the detector for the simulated values is not clear, but it is possible that there were unidentified materials in the detector. The $\varepsilon^{mono}_{p:exp}$ and $\varepsilon_{t:exp}$ are shown in Fig. 1**(a)** together with the $\varepsilon^{mono}_{p:sim}$ and $\varepsilon_{t:sim}$, and the ratio to the $\varepsilon^{mono}_{p:sim}$ and $\varepsilon_{t:sim}$ by GEANT4 are shown in **Fig. (b)**. The results were summarized in Table 2. The uncertainties of the $\varepsilon^{mono}_{p:sim}$ and $\varepsilon_{t:sim}$ were evaluated to be 2% from Fig. 1**(b)**.

Table 1
Nominal crystal geometry and adopted one for Monte Carlo simulation GEANT4.

		Crystal dimension (mm)		
		Diameter	Length	Dead layer
Specification sheet		80	90	–
Monte Carlo	$\varepsilon_{p:sim}$	80	76.3	0.01
	$\varepsilon_{t:sim}$	80	87.8	0.01

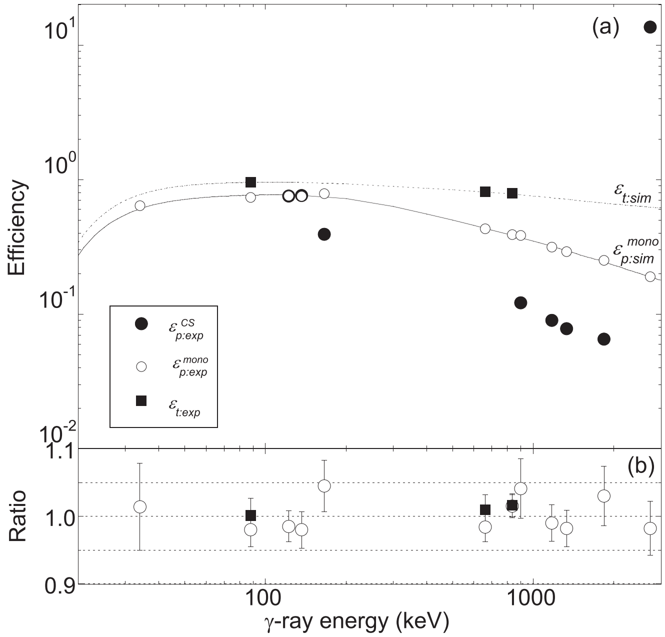


Fig. 1. The corrected full energy peak efficiency, total efficiency measured with mono/quasi-mono energy γ-ray sources, and the simulated peak and total efficiencies for the clover detector in the add-back mode (a) and the ratios of the corrected efficiencies to the efficiencies in the absence of the coincidence summing simulated by GEANT4 (b). In fig (a), the open circles (○) and the closed circles (•) indicate the peak efficiency corrected for the coincidence summing and that in the presence of coincidence summing for ^{57}Co, ^{141}Ce, ^{60}Co and ^{88}Y, respectively. The closed squares (■) indicate the total efficacies by ^{109}Cd, ^{137}Cs, and ^{54}Mn. The solid and dotted curves represent the full energy peak efficiency and the total efficiency with simulated by the Monte Carlo code GEANT4, respectively.

Using the $\varepsilon^{mono}_{p:sim}$ and $\varepsilon_{t:sim}$, the corrections of coincidence summing were applied to the measured spectra of multi-energy γ-ray emitters ^{134}Cs and $^{152,\ 154}$Eu. These are nuclides having complicated decay schemes, and they are thus good examples of unstable nuclides, on which we can demonstrate appropriately the quality of the correction for coincidence summing effects. The coincidence summing correction based on the decay scheme information of each nuclide was carried out using the Monte Carlo calculation as described elsewhere (Shima et al., 2014) using the level structure from the data file (Laboratoire National Henri Becquerel, NUCLEIDE Database, 2013), including the β-branching ratio, emission probability of γ-ray, total and K internal conversion coefficients. We describe the method briefly. The decay path from an excited level to the ground state in the decay scheme of each nucleus was randomly sampled using the Monte Carlo simulation, and the events of full and partial photon energy deposition in the detector were also randomly sampled on the basis of ε_p and ε_t, respectively. A number of 10^7 events were simulated. According to the efficiencies and the decay scheme, energy deposition events were simulated without (one γ-ray) or with (cascading γ-rays) coincidence summing. The ratio of the number of events in which the total energy of the γ_i photon was deposited in the detector in the two cases gives the correction factor for coincidence summing f_{γ_i}. The results were summarized in Table 3. In Fig. 2**(a)**, the closed and open symbols indicate the $\varepsilon^{CS}_{p:exp}$ and $\varepsilon^{mono}_{p:exp}$ of ^{134}Cs and $^{152,\ 154}$Eu, respectively. The degree of corrections was very large; nevertheless, the discrepancies between $\varepsilon^{mono}_{p:exp}$ and $\varepsilon^{mono}_{p:sim}$ were within 5% agreement, as shown in Fig. 2**(b)**.

As summarized in Table 4, the uncertainty of the peak efficiency (σ_{ε_p}) was mainly caused by the 2% uncertain intensities of single-emitters ($\sigma_{s.e.}$), and the uncertainty caused by the deviation of the