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Original Article

True coincidence summing correction factor for point source geometry with PHITS

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ABSTRACT

In this study, it has been shown that the true coincidence summing correction factor can be obtained for the first time using the PHITS Monte Carlo program. Determining this correction factor using different methods and tools in each laboratory to increase the possibility of achieving high-efficiency measurement conditions is still popular in gamma-ray spectrometry. By using ^{133}Ba, ^{152}Eu, ^{134}Cs, and ^{60}Co point sources, the true coincidence summing factor was investigated in both near and far counting geometries for 15 different energy values. GESPECOR software was used to validate the results obtained with PHITS. A remarkable agreement was obtained between PHITS and GESPECOR, with a discrepancy of less than 3%. With this study, a new tool has been proposed to obtain the true coincidence summing factor, which is one of the significant correction factors investigated/calculated in gamma-ray spectrometric studies.

1. Introduction

Some sources used/studied in gamma-ray spectrometry contain radionuclides that emit photons in two or more cascades. Radionuclides with more than one gamma-ray emitting complex decay scheme can be artificial radionuclides such as ^{152}Eu, ^{134}Cs, ^{133}Ba, ^{60}Co, ^{88}Y, as well as natural radionuclides such as ^{214}Pb, ^{208}Tl, ^{228}Ac, ^{214}Bi, and ^{234}Pa from the decay products of ^{238}U and ^{232}Th. The resolution time of the HPGe detector system is longer than the time intervals of two or more photons emitted in a cascading transition [1,2]. When these photons are emitted from the same nucleus and detected by the detector at the same time interval, the effect called true coincidence summing (TCS) occurs [3]. This effect, which causes count losses or gains in the peaks, should be considered especially at low source-to-detector distance [4]. In calculating the activity concentration of the radionuclides in the samples, the full energy peak efficiency (FEPE) should be determined for the energies of interest [5]. The TCS is one of the important correction factors to consider when calculating full energy peak efficiency. Studies on the complex procedures applied in the evaluation of this effect for high-precision results, how and with which method it will be determined are still continuing in many laboratories and working groups around the world [6]. In TCS calculations, different methods such as analytical approaches and semi-empirical methods mostly Monte Carlo (MC) simulation programs are used. Almost all methods are given in the intercomparison made by Lepy et al., in 2010 for point source geometry and in 2012 for volume sources and in which many laboratories participated [7,8]. These methods used; semi-empirical method, simplified method and Monte Carlo simulation method (specially dedicated codes such as ETNA [9,10], GESPECOR [11–14], KORSUM [15], TRUECOINC [16], EFFTRAN [1] and full Monte Carlo simulation code such as GEANT4 [17]). The TCS correction factor can be obtained directly with specifically dedicated MC programs such as GESPECOR and EFFTRAN, as well as indirectly in software and programs such as LabSOCS, ETNA, and TRUECOINC. For example, TCS corrections in the ETNA method; as calculated using full energy peak and total efficiency values obtained by MC simulation such as MCNP and PENELOPE, or using experimental efficiency values. In the study by Sima et al., in 2020, the internal consistency of results submitted by 21 teams was examined for coincidence summing correction factor calculation for extended sources [6]. It has been reported that most of the 33 sets of TCS values passed the test, but the results obtained using the quasi-point source approximation did not. The quasi-point source approximation for the evaluation of coincidence summing corrections assumes that integrals of products of efficiencies over the volume of the source are equal with the products of efficiencies separately integrated over the volume of the source. Because these integrals are difficult to evaluate, some codes use a quasi-point source approximation even in the case of volume sources; this is a much simpler solution, requiring either the use of experimental efficiencies, or the computation (e.g. by MC simulation) of a small number of efficiencies.

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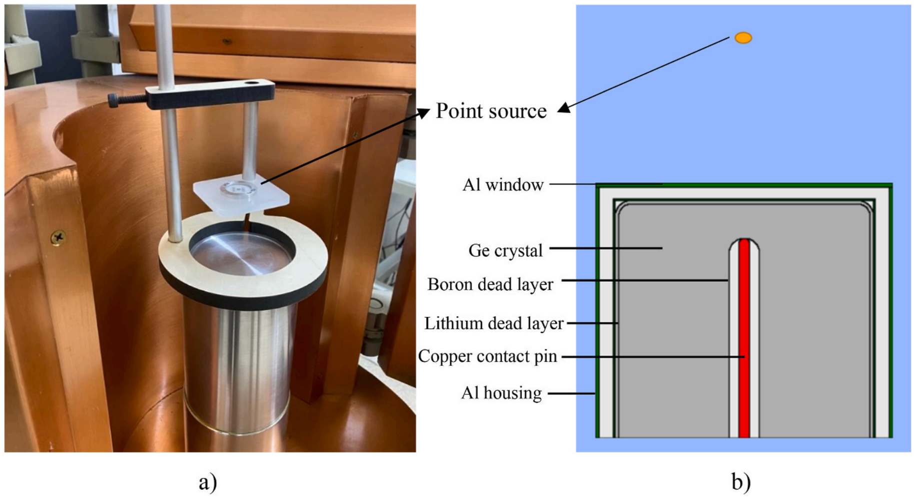

Fig. 1. a) The experimental setup and, b) Model with the PHITS code.

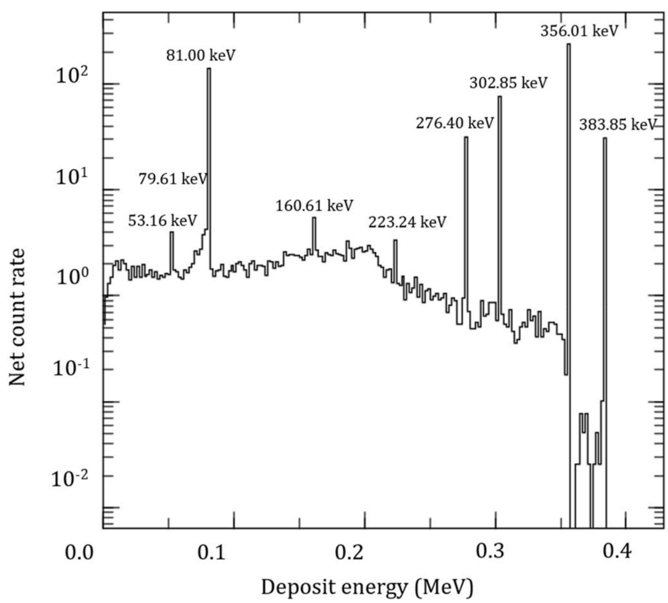

Fig. 2. Simulated spectrum to ^{133}Ba obtained with PHITS.

The TCS effect mainly depends on the decay scheme of a radionuclide and the detector-source geometry, i.e. the solid angle [18, 19]. The present study shows that the PHITS MC code can be used to obtain the true coincidence summing correction factor. This effect, which is significantly affected by the detector-to-source geometry, was investigated at three different distances, 5 cm, 10 cm and 12.5 cm. To validate the PHITS program, which was used for the first time for this purpose, TCS factors from PHITS were compared with findings from the GESPECOR program [12].

2. Materials and methods

2.1. Detector and point sources

p-type coaxial HPGe detector system from the GEM series (Ortec GEM150P4) with 1.5 mm Al window and a relative efficiency of 150% was used for efficiency measurements. The performance specifications of the detector are as follows: energy resolution (FWHM) is 0.89 keV at 122.1 keV (^{57}Co) and 2.11 keV at 1332.5 keV (^{60}Co), peak-to-Compton ratio for 1332.5 keV (^{60}Co) is 90:1. The physical properties of the detector are: the diameter of the crystal is 94.8 mm and the length is 87.2 mm; the diameter of the hole is 11.2 mm and the length is 73.4 mm; and the end cap-to-crystal distance is 5 mm. The dead layer value of 1.29 mm was used instead of the 0.7 mm value given by the manufacturer, which is the result of comparing the experimental and simulated full energy peak efficiencies [20]. In the study, the full energy peak efficiency determined by Monte Carlo simulation methods is given with the

Table 1
TCS correction factors were calculated by PHITS and GESPECOR, and the relative differences (Δ) between the two programs in percentage.

Nuclide	Energy (keV)	5 cm			10 cm			12.5 cm		
		PHITS	GESPECOR	Δ (%)	PHITS	GESPECOR	Δ (%)	PHITS	GESPECOR	Δ (%)
^{133}Ba	81	0.9453	0.9248	2.2	0.9881	0.9668	2.2	1.0074	0.9788	2.8
	302.85	0.9945	0.9798	1.5	0.9960	0.9921	0.4	1.0248	0.9950	2.9
	356.01	0.9786	0.9825	0.4	0.9895	0.9932	0.4	0.9912	0.9956	0.4
	383.85	1.0345	1.0416	0.7	1.0195	1.0162	0.3	1.0124	1.0102	0.2
^{152}Eu	121.78	0.9437	0.9260	1.9	0.9896	0.9635	2.6	1.0011	0.9763	2.5
	344.28	0.9659	0.9485	1.8	0.9801	0.9754	0.5	0.9968	0.9841	1.3
	778.91	0.9563	0.9307	2.7	0.9655	0.9691	0.4	0.9715	0.9803	0.9
	964.08	0.9624	0.9545	0.8	0.9879	0.9810	0.7	0.9915	0.9879	0.4
	1112.07	0.9766	0.9776	0.1	0.9842	0.9907	0.7	1.0023	0.9945	0.8
	1408.01	0.9831	0.9679	1.5	0.9914	0.9872	0.4	1.0111	0.9921	1.9
^{134}Cs	569.33	0.8926	0.8677	2.8	0.9312	0.9356	0.5	0.9439	0.9585	1.5
	604.72	0.9356	0.9169	2.0	0.9734	0.9598	1.4	0.9837	0.9740	1.0
	795.86	0.9343	0.9183	1.7	0.9693	0.9614	0.8	0.9827	0.9751	0.8
^{60}Co	1173.23	0.9365	0.9385	0.2	0.9738	0.9685	0.5	0.9827	0.9795	0.3
	1332.49	0.9388	0.9372	0.2	0.9707	0.9683	0.2	0.9825	0.9793	0.3

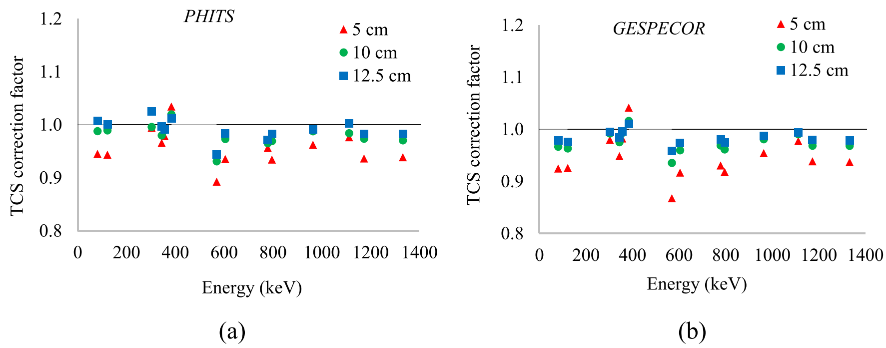

Fig. 3. TCS correction factors obtained with a) PHITS, b) GESPECOR for 5 cm, 10 cm, and 12.5 cm source-to-detector distances.

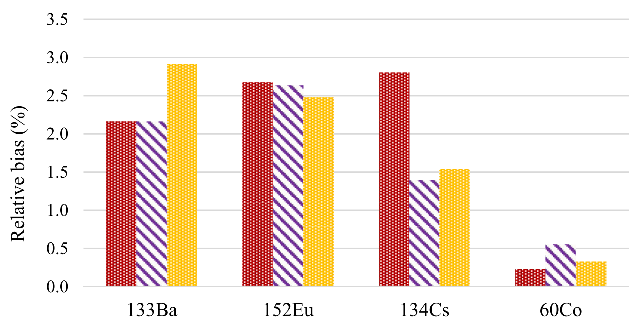

Fig. 4. Relative differences distribution between the data obtained using PHITS and GESPECOR for the four radionuclides investigated.

expression simulated. The detector is operated by the Ortec DSPEC jr 2.0 based on a digital signal processor. The DSPEC jr 2.0 is controlled by the Gamma Vision spectroscopy software [21]. ^{60}Co (14.75 ± 0.15 kBq), ^{133}Ba (21.57 ± 0.22 kBq), ^{134}Cs (54.0 ± 0.4 kBq), ^{152}Eu (54.5 ± 0.8 kBq) reference point sources purchased from PTB (Physikalisch-Technische Bundesanstalt) were used in both efficiency calculations and simulations. The reference dates of ^{133}Ba, ^{134}Cs and ^{152}Eu are May 1, 2012; the reference date of the ^{60}Co point source was June 1, 2013. The active diameter of the point sources is 5 mm, and an aluminum ring of 30 mm diameter and 4 mm thickness surrounds the source capsules. The counting times of the point sources were adjusted to continue until at least 20 000 counts were collected to minimize the statistical counting uncertainty for each peak. Since all point sources used in the measurements are multi-nuclide gamma sources, they have a true coincidence summing effect.

The full energy peak efficiency (FEPE), $\varepsilon(E_i)$, for a given photon energy has been obtained from the following expression:

$$\varepsilon(E_i) : \frac{N_p(E_i)/t}{A \bullet f_\gamma(E_i)} \Big/ TCS \quad (1)$$

where $N_p(E_i)/t$ represents the net count rate, A is the activity of the source at the measurement date, $f_\gamma(E_i)$ is the probability of gamma-ray emission, and TCS is the true coincidence summing correction factor. The experimental efficiency was measured at 5 cm, where the TCS effect is greater (Fig. 1). The calculated values by applying TCS corrections were then compared with the results of the Monte Carlo simulation.

2.2. TCS calculations with PHITS

PHITS (Particle and Heavy Ion Transport code System) is a Fortran code that performs transport and collision simulations of almost all particles (electron, photon, neutron, proton, ions, etc.) over a wide energy range (10^{-4} eV to 1 TeV) using the Monte Carlo method. All contents of PHITS (source files, binary, data libraries, graphic utility etc.) are fully integrated in one package [22]. The requirement for high-quality decay scheme data for calculation of coincidence summing

Table 2
Experimental and simulated efficiency values at 5 cm source-to-detector distance.

Radionuclide	Energy (keV)	Experiment			Simulated efficiency[d] (Uncertainty %)
		Efficiency[a] (Uncertainty %)	TCS factor[b]	Corrected efficiency[c]	
^{133}Ba	81.00	0.04079 (2.3)	0.9453	0.04315	0.04411 (0.15)
^{152}Eu	121.78	0.05869 (1.8)	0.9437	0.06219	0.06280 (0.13)
^{133}Ba	302.85	0.04684 (1.5)	0.9945	0.04710	0.04713 (0.15)
^{152}Eu	344.28	0.04210 (1.8)	0.9659	0.04359	0.04374 (0.15)
^{133}Ba	356.01	0.04155 (1.3)	0.9786	0.04246	0.04287 (0.15)
^{133}Ba	383.85	0.04225 (1.8)	1.0345	0.04084	0.04098 (0.16)
^{134}Cs	569.33	0.02861 (2.6)	0.8926	0.03205	0.03255 (0.18)
^{134}Cs	604.72	0.02933 (1.4)	0.9356	0.03135	0.03146 (0.18)
^{152}Eu	778.91	0.02587 (1.8)	0.9563	0.02705	0.02727 (0.19)
^{134}Cs	795.86	0.02503 (1.5)	0.9343	0.02680	0.02692 (0.19)
^{152}Eu	964.08	0.02315 (1.8)	0.9624	0.02405	0.02415 (0.20)
^{152}Eu	1112.07	0.02170 (1.9)	0.9766	0.02222	0.02224 (0.21)
^{60}Co	1173.23	0.02006 (1.4)	0.9365	0.02143	0.02156 (0.22)
^{60}Co	1332.49	0.01864 (1.4)	0.9388	0.01985	0.01999 (0.22)
^{152}Eu	1408.01	0.01884 (1.8)	0.9831	0.01917	0.01932 (0.23)

[a] Experimental FEPE values calculated without TCS correction factor.
[b] TCS correction factors obtained from PHITS.
[c] Experimental FEPE values calculated with TCS correction factor.
[d] Simulated FEPE values calculated with PHITS.

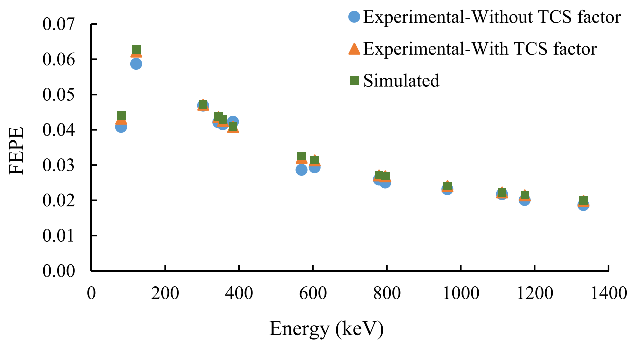

Fig. 5. Experimental curves without and with TCS correction factor, and efficiency curves from simulation.

correction factors is also met by the latest version of ENDF (Evaluated Nuclear Data File) and JEFF (Joint Evaluated Fission and Fusion), the nuclear data libraries used in PHITS. To obtain the efficiency value in PHITS, the T-deposit tally, which gives the energy distribution in a specific region of the detector, is defined as s-type = 9 and s-type = 1, depending on whether the source is point or volumetric in the [source] section. In the calculation of the TCS effect in radionuclides where two or more radiations are emitted simultaneously, the <source> parameter is used to simulate the detector response. Multiple sources with different source definitions, such as different particle types, geometries, or energy distributions, can be specified using the <source> multi-source parameter. Each resource definition begins with <source> = number, which specifies the relative weight of such a resource.

Each simulation considers the full cascade of events following the radioactive decay of each radionuclide emitting more than one gamma-ray. For example, the graph of the net count rate according to the energy of the gamma-ray peaks emitted from the ^{133}Ba radionuclide is seen as a spectrum (Fig. 2). Energy bin width was set to 0.2 keV. Simulated spectra of other ^{152}Eu, ^{134}Cs and ^{60}Co radionuclides are similarly obtained in PHITS. As can be seen, the peaks of ^{133}Ba such as 160.61 keV and 223.24 keV, which are not preferred in the analysis because they have low gamma emission probability (<1%), are also seen in the spectrum.

The following formula was used to calculate the true coincidence summing correction factor:

$$TCS(E_i) = \frac{FEPE\ (without\ TCS(E_i))}{FEPE\ (with\ TCS(E_i))} \tag{2}$$

$TCS(E_i)$: The true coincidence summing correction factor for the E_i gamma-ray energy

$FEPE\ (without\ TCS(E_i))$: The FEPE for the E_i gamma-ray energy without taking summation effects into account

$FEPE\ (with\ TCS(E_i))$: The FEPE for the E_i gamma-ray energy taking summation effects into account

2.3. TCS calculations with GESPECOR

GESPECOR is a Monte Carlo-based software developed for calculating efficiency, self-absorption effects, and true coincidence summing effects in gamma-ray spectrometry [12]. Since it can calculate coincidence summing effects with cascade gamma photons, coincidence losses, K_α, K_β, and multiple X-rays, it is reliably and widely used to obtain true coincidence summing correction factors [13,23]. All nuclides for which the decay scheme is available in GESPECOR are included in the KORDATEN file with data from DDEP (Decay Data Evaluation Project). DDEP uses Nucléide-Lara, developed for alpha and gamma-ray spectrometry users, to obtain nuclear data such as half-life, decay modes, branching ratios, the energies and intensities of the various emissions [24]. Since GESPECOR is a special-purpose Monte Carlo program, after all parameters of the detector, material, and measurement geometry are entered into the relevant sections, "Coincidence" is selected from the menu and directly gives the TCS value at the energies of interest.

3. Results and discussion

The TCS factors of 15 full energy peaks of ^{133}Ba (81 keV, 302.85 keV, 356.01 keV, 383.85 keV), ^{152}Eu (121.78 keV, 344.28 keV, 778.91 keV, 964.08 keV, 1112.07 keV, 1408.01 keV), ^{134}Cs (569.33 keV, 604.72 keV, 795.86 keV), and ^{60}Co (1173.23 keV, 1332.49 keV) in PHITS were calculated by Equation (2). Results from PHITS for 5 cm, 10, and 12.5 cm distances were compared with data from the GESPECOR program for the same distances (Table 1). As seen from Fig. 3a and b, correction factor values are more dominant in both PHITS and GESPECOR at close counting geometry 5 cm than 10 cm and 12.5 cm.

Comparisons of the corresponding values are given in Table 1, examined in terms of relative differences. The relative difference (Δ) for each gamma-ray energy was calculated with the following equation:

$$RB(\%) = \frac{|TCS_{PHITS} - TCS_{GESPECOR}|}{TCS_{PHITS}} \times 100 \tag{3}$$

TCS_{PHITS} and $TCS_{GESPECOR}$ are true coincidence summing correction factors calculated by PHITS and GESPECOR MC simulation programs, respectively. The results in Table 1 showed good agreement with the relative bias between the TCS values calculated by the PHITS and the GESPECOR results, ranging from 0.1% to 2.9% (Fig. 4).

Experimental and simulated efficiency values were also calculated at a low source-to-detector distance of 5 cm. The change in FEPE caused by the more effective TCS factor at this distance is given in Table 2. The greatest effect of the TCS factor on FEPE is the difference of approximately 11% at 569.33 keV (^{134}Cs) with a value of 0.8926, as seen in Fig. 5.

4. Conclusions

In this study, true coincidence summing correction factors for gamma emissions of ^{133}Ba, ^{152}Eu, ^{134}Cs, and ^{60}Co were calculated at three different distances with the PHITS simulation code. The results verified with the GESPECOR code, and a good agreement was obtained. The maximum relative bias between the two codes was 2.9%, 2.7%, 2.8%, and 0.5% for the ^{133}Ba, ^{152}Eu, ^{134}Cs, and ^{60}Co sources, respectively. These acceptable differences between the two codes can be explained by the use of different approaches, approximations, and libraries by the programs. The findings in this work prove that the PHITS Monte Carlo simulation program can be used reliably in obtaining the true coincidence summing correction factor.

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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