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Calculation of Coincidence Summing Correction Factors for an HPGe detector using GEANT4

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ABSTRACT

The aim of this paper was to calculate the True Coincidence Summing Correction Factors (TSCFs) for an HPGe coaxial detector in order to correct the summing effect as a result of the presence of ^{88}Y and ^{60}Co in a multigamma source used to obtain a calibration efficiency curve. Results were obtained for three volumetric sources using the Monte Carlo toolkit, GEANT4. The first part of this paper deals with modeling the detector in order to obtain a simulated full energy peak efficiency curve. A quantitative comparison between the measured and simulated values was made across the entire energy range under study. The True Summing Correction Factors were calculated for ^{88}Y and ^{60}Co using the full peak efficiencies obtained with GEANT4. This methodology was subsequently applied to ^{134}Cs, and presented a complex decay scheme.

1. Introduction

Gamma spectroscopy High-Purity Germanium (HPGe) detectors are generally used to analyze the activity of radionuclides in environmental samples, and have the advantage of not requiring laborious sample preparation. The goodness of results obtained using gamma-ray spectrometry depends on the efficiency calibration of the detector system. For this reason, to obtain an accurate activity measurement, good knowledge of the detector and the efficiency response (solid angle, source matrix, geometry, etc) is required. In general, efficiency calibration of HPGe detectors is performed using multi-line gamma-ray standards that cover the energy range under study. Unfortunately, these standards often contain some radionuclides that exhibit coincidence or cascade summing when measured in close proximity to the detector. Thus, the fitted efficiency curve obtained with these radionuclides is different from that obtained with monoenergetic radionuclides.

True summing coincidence takes place when two or more photons, which are emitted in cascade from an excited nucleus, are detected simultaneously within the resolving time of the detector. As a result, the detector cannot distinguish between the two interactions and treats them as a single event adding together the summed energy of both interactions. These effects of coincidence produce the so-called summing-in and summing-out effects. As a consequence, coincidence losses from full energy peaks and enhancement of sum peaks occur. In recent years, various methods to calculate True Coincidence Summing Correction Factors (TSCFs) have been proposed. Andreev et al. (1972) were the first to propose a numerical method to solve this problem. Later, TSCF calculation was addressed by Monte Carlo simulations and analytical approaches (Décombaz et al., 1992). When using a point source, coincidence summing can be calculated accurately by theoretical expression (Rizzo and Tormachio, 2010). Nevertheless, in the presence of a volumetric source this becomes difficult.

This work centered on the calculation of TSCFs for an HPGe detector using the Monte Carlo toolkit, GEANT4 (Agostinelli et al., 2003; Allison et al., 2006). A quantitative comparison using statistical methods was performed in order to validate the use of the GEANT4 toolkit in calculating full energy peaks (FEP). Finally, TSCF calculations were made according to the methodology proposed by Hurtado in 2004 (Hurtado et al., 2004).

2. Methodology

To date, a variety of approaches have been used to calculate

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TSCFs; in the following a short overview of these is given: KORSUM (Debertin and Schotzig, 1979) uses the Andreev recursive formula; CSCOR (Sinkko and Aaltonen, 1985) computes γ-ray summing with the method proposed by Debertin and Schotzig; the ETNA software was developed at the Laboratoire National Henri Becquerel and uses a numerical method, based on the Andreev and McCallum principle (Piton et al., 2000). Conversely, TRUECOINC (Sudar, 2002) uses a combinatorial method and a predefined database to calculate TSCFs. In this paper, results were obtained using experimental data and the Monte Carlo code, PENELOPE (Salvat et al., 2007). GESPECOR (Sima and Arnold, 2000) analytically computes the correction factors and uses the efficiency value simulated by Monte Carlo.

Nowadays, Monte Carlo methods are widely used to calculate detector efficiency and Coincidence Summing Correction Factors. Décombaz et al. (1992) were the first to propose a statistical approach based on Monte Carlo to calculate TSCFs. Later, other authors proposed different approaches to solving this problem, as reported by García-Talavera et al. (2001), García-Torano et al. (2005), Dziri et al. (2012), and Sima and Arnold (2000).

3. Measurements

Experimental measurements were taken using an HPGe detector. This detector was an ORTEC GMX series HPGe coaxial detector system with a relative efficiency of 40% at 1.33 MeV, a nominal resolution of 0.76 keV and 2 keV at 5.9 keV and 1.332 MeV, respectively, and a crystal volume of 201 cm^3, as specified in the manufacturer's data sheet. Table 1 summarizes the components and dimensions provided by the manufacturer. Data was recorded using a multi-channel analyzer with 8192 channels. The measurements were taken using a multigamma-ray standard solution containing the following radionuclides: ^{241}Am, ^{109}Cd, ^{57}Co, ^{139}Ce, ^{51}Cr, ^{113}Sn, ^{85}Sr, ^{137}Cs, ^{54}Mn, ^{88}Y, ^{65}Zn, and ^{60}Co. The characteristics of these radionuclides are shown in Table 2, and reveal that ^{60}Co and ^{88}Y presented true summing coincidence.

Gamma ray spectra were analyzed using GammaVision Software (ORTEC industries) and FEP efficiency, ε, for a given photon energy obtained from the following equation:

$$\varepsilon = \frac{N}{tAP_\gamma} \quad (1)$$

where N represented the number of net counts in the peak, calculated using the total summation method, A was the source activity, t was the counting time and Pγ was the photon emission probability. The dead time was below 1%, due to the low activity concentration of samples.

The experimental detector efficiency was determined for three different matrices: a 100 ml and a 15 ml Petri beaker filled with an aqueous solution containing the multigamma standard and a 100 ml Petri beaker filled with sea sand (major component SiO_2) where the multigamma source was homogeneously mixed. These matrices were placed on the center ring just above the detector window. For convenience, these matrices were labeled as PGAQ, PPAQ, and PGSI.

Table 1
Technical dimensions of the ORTEC GMX detector provided by the manufacturer.

Component	Dimension (mm)
Crystal diameter	60.0
Crystal length	71.1
Hole diameter	9.0
Hole depth	63.1
Be window	0.5

Table 2
Composition of the multigamma standard used for the efficiency calibration.

Source	E (keV)	I (%)	TCS
^{241}Am	59.5	35.90	None
^{109}Cd	88.0	3.61	None
^{57}Co	122.0	85.59	γ-γ
^{139}Ce	165.8	79.95	γ-X
^{51}Cr	320.0	9.83	γ-X
^{113}Sn	391.7	64.16	None
^{85}Sr	514.0	98.30	γ-X
^{137}Cs	661.7	85.21	None
^{54}Mn	834.0	99.97	None
^{88}Y	898.0	93.70	γ-γ, γ-X
^{65}Zn	1115.5	50.74	None
^{60}Co	1173.0	99.90	γ-γ
^{60}Co	1332.5	99.98	γ-γ
^{88}Y	1836.0	99.35	γ-γ, γ-X

4. Monte Carlo simulations

Simulations were performed with version 9.6 of the Geant4 toolkit, using CLHEP libraries 2.1.3.1. Geant4 is a toolkit written in C++ that can simulate the transport of particles through matter. It can aid the characterization of experiments as it enables particle properties to be examined at any location in the simulation model, thus facilitating the extraction of information about physics interactions and the energy deposited in the detector volume.

The data library used in this paper was G4EMLOW version 6.32 (Chauvie et al., 2001, 2004), which contains data files for low energy electromagnetic processes (Amako et al., 2005). Some electromagnetic processes require a threshold below which no secondary particles are generated. Because of this requirement, thresholds for gammas, electrons and positrons need to be defined by the user. These thresholds should be defined as a distance or range cut-off, which is internally converted to energy for individual materials.

The physics activated are the following: Auger electron production, Compton and Rayleigh scattering, pair production and the photoelectric effect for photons. Ionization processes and Bremsstrahlung for secondary particles are also activated.

To simulate the decay of the different radionuclides, the Radioactive Decay Module (RDM) was used in the context of the Geant4 toolkit. The RDM was originally developed by Truscott (2002) and simulates radioactive decays by sampling secondary particles on a per-decay basis. Validation and verification of this code was made by Hauf et al (2013). The RDM generates all the possible decay paths of a particular radionuclide using the branching ratios obtained from the Evaluated Nuclear Structure Datafile (ENSDF) library (http://www.nndc.bnl.gov/ensdf).

The output of Geant4 includes the events registered in the detector (kind of particle and interaction and energy deposited in the detector volume).

In the context of this work, the HPGe detector was modeled using the manufacturer's description and included the main components (germanium crystal, dead layer, inner hole, beryllium window, and aluminum holder). The dead layer thickness was taken as 75 μm via a comparison between calculated and experimental efficiency values at low photon energies. The geometry of the detector has several uncertainties (active crystal dimensions, dead layer thickness, inner hole dimensions, etc.) that can affect the efficiency. In a previous work (Gallardo et al., 2015), geometry uncertainties and its effects in the efficiency were studied.

The detector resolution was taken into account using a Gaussian broadening function. The width of the distribution has been chosen according to the real resolution of the detector. Photons were uniformly distributed in the source volume and isotropically emitted.

Once the simulation had been completed, the energy deposited from the primary and secondary particles was registered in the detector volume and sorted in a histogram using the data analysis toolkit ROOT (Brun and Rademakers, 1997). Fig. 1 shows the simulated spectra obtained for ^{88}Y and the presence of the summation peak at 2734 keV, as a result of summing energies at 898 keV and 1836 keV.

The FEP-simulated efficiency was calculated as the ratio of the photons recorded at the energy photopeak and the number of the simulated photons. Fig. 2 (a, b, c) shows both experimental (red) and simulated (black) FEP efficiencies for PGAQ, PGSI and PPAQ geometries, respectively. The relative errors of experimental measurements were about 2%, whilst the simulated relative errors were lower than 1%.

Comparisons between measured and simulated FEP efficiencies were made by calculating the ratio between them and are shown in Table 3. The ratio shows good agreement between simulated and measured efficiencies for each of the geometries. A statistical test was performed to quantitatively evaluate comparisons between measured and simulated efficiencies for the set of energies under study. The non-parametric Wilcoxon signed-rank test was used to compare simulated and measured efficiencies which were dependent samples paired by the respective energy. The p-value of the Z statistic obtained in the test for each of the geometries, PGAQ, PGSI and PPAQ, was greater than 0.05 ($(Z = -1.68, p = 0.093)$, $(Z = -1.923, p = 0.054)$, $(Z = -0.14, p = 0.889)$, respectively). These results show that there were no statistically significant differences between measured and simulated efficiencies for the set of energies at a 95% confidence level for each of the geometries. Therefore, the goodness of the geometrical detector model was accepted.

On the other hand, the ratio distribution was checked via the Shapiro-Wilk test for each of the geometries. A normal distribution of these deviations is to be expected in the absence of systematic uncertainties. The p-values obtained confirmed this normality at a 95% confidence level ($p = 0.978$, $p = 0.141$, $p = 0.147$ for PGAQ, PGSI and PPAQ geometries, respectively). Moreover, the mean deviation was less than 2% for each of the geometries under study.

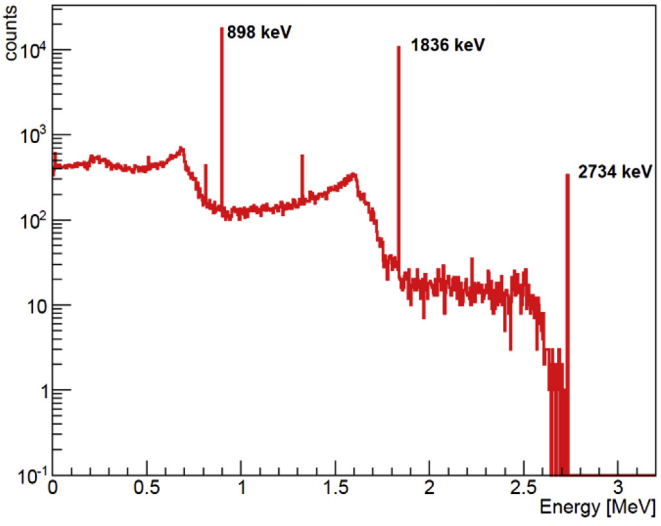

Fig. 1. Simulation spectra of ^{88}Y obtained using the GEANT4 toolkit.

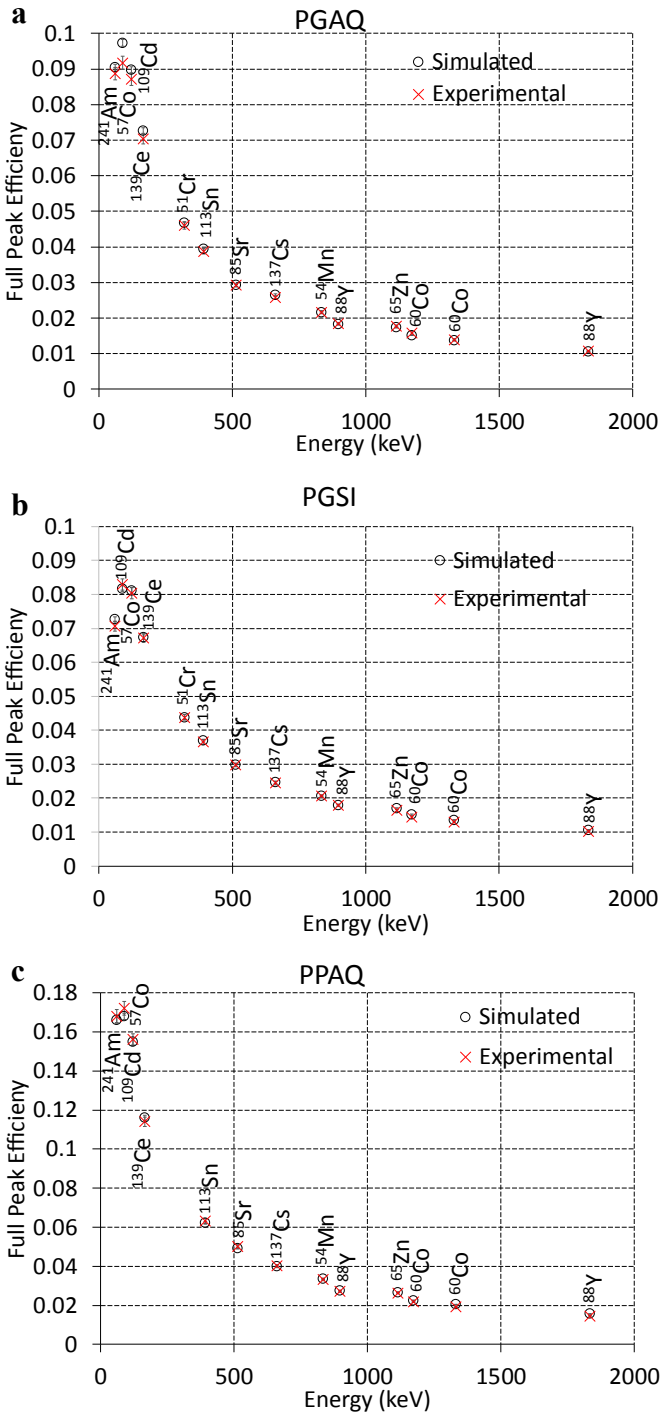

Fig. 2. a. Simulated (black) and experimental (red) full energy peak efficiencies as a function of gamma energy for the PGAQ. 1-sigma error bars included. b. Simulated (black) and experimental (red) full energy peak efficiencies as a function of gamma energy for the PGSI. 1-sigma error bars included. c. Simulated (black) and experimental (red) full energy peak efficiencies as a function of gamma energy for the PPAQ. 1-sigma error bars included. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

These results demonstrated the validity of the model for calculating TSCFs for these geometries.

5. Summing correction factor calculation

According to the standard used, the presence of ^{88}Y and ^{60}Co had

Table 3
Measured and simulated efficiencies for PGAQ, PGSI and PPAQ, respectively.

E (keV)	Experimental	GEANT	Ratio (Exp/GEANT)
59.50	0.0904	0.0887	1.019
88.00	0.0973	0.0917	1.060
122.08	0.0897	0.0872	1.029
165.85	0.0727	0.0704	1.034
320.08	0.0467	0.0460	1.015
391.69	0.0394	0.0388	1.016
513.14	0.0292	0.0293	0.997
661.66	0.0265	0.0257	1.030
834.83	0.0216	0.0216	1.000
898.00	0.0183	0.0184	0.993
1115.52	0.0174	0.0177	0.983
1173.20	0.0152	0.0158	0.960
1332.50	0.0138	0.0139	0.990
1836.00	0.0105	0.0107	0.980
59.50	0.0707	0.0726	0.973
88.00	0.0829	0.0817	1.013
122.08	0.0804	0.0812	0.990
165.85	0.0672	0.0674	0.996
320.08	0.0438	0.0437	1.003
391.69	0.0366	0.0369	0.990
513.14	0.0298	0.0298	0.999
661.66	0.0245	0.0246	0.995
834.83	0.02065	0.0206	1.002
898.00	0.0179	0.0179	0.999
1115.52	0.01635	0.0170	0.960
1173.20	0.0144	0.0152	0.950
1332.50	0.013	0.0135	0.960
1836.00	0.01024	0.0106	0.970
59.50	0.168	0.1662	1.014
88.00	0.172	0.168	1.020
122.08	0.156	0.155	1.005
165.85	0.114	0.116	0.987
391.69	0.0631	0.0623	1.013
513.14	0.0501	0.0493	1.015
661.66	0.0402	0.0399	1.006
838.83	0.0332	0.0335	0.988
898.00	0.0271	0.0276	0.980
1115.52	0.0264	0.0267	0.986
1173.20	0.0219	0.0224	0.975
1332.50	0.0193	0.0204	0.945
1836.00	0.0146	0.0156	0.940

Table 4
TSCFs obtained for all matrices.

Geometry	Radionuclide	E (keV)	TSCF
PGAQ	^{88}Y	898	1.099 (±0.011)
		1836	1.130 (±0.015)
	^{60}Co	1173	1.078 (±0.012)
		1332.5	1.100 (±0.013)
PGSI	^{88}Y	898	1.106 (±0.011)
		1836	1.110 (±0.015)
	^{60}Co	1173	1.094 (±0.012)
		1332.5	1.110 (±0.013)
PPAQ	^{88}Y	898	1.140 (±0.095)
		1836	1.160 (±0.013)
	^{60}Co	1173	1.150 (±0.011)
		1332.5	1.1350 (±0.011)

an important effect on summing coincidence that had to be taken into account in order to obtain proper fitting of the efficiency curve.

The TSCFs associated with the emissions of a given radionuclide were calculated as the ratio between the photo-peak areas at energies of interest obtained for the monoenergetic sources, and the photo-peak areas obtained when the complete radionuclide disintegration was simulated (Hurtado et al., 2004; Quintana and Montes, 2013).

A statistical test was applied to evaluate the influence of the solid angle and the absorber present in the container on TSCFs. The u statistic (Brookes et al., 1979) was calculated to compare two values (V_1, V_2) and their uncertainty (u_1, u_2), according to the following equation:

$$u = \frac{|V_1 - V_2|}{\sqrt{u_1^2 + u_2^2}} \sim N(0,1) \quad (2)$$

Table 5 shows the u statistics and the p-value results from the tests calculated for each energy value when comparing different absorbers (PGAQ-PGSI) and different solid angles (PGAQ-PPAQ). The p-values obtained from comparing different absorbers were greater than 0.05 for all energies, whilst the p-values obtained from comparing different solid angles were less than 0.05 (except at an energy of 1836 keV). These results show how TSCFs, for these energies, depended on the solid angle and not on the absorber present in the container, at a 95% confidence level.

Once the TSCFs had been calculated for ^{88}Y and ^{60}Co, the FEP efficiency ε was obtained for each photon energy according to Eq. (3).

$$\varepsilon = \frac{N}{tAP_\gamma} TSCF \quad (3)$$

These efficiencies were used to correct the experimental calibration curve, avoiding the efficiency underestimation in the energy range between 898 keV and 1836 keV.

Using the same methodology, the TSCFs were calculated for ^{134}Cs. This isotope presented a more complex decay scheme. The most intense photons are shown in Table 6.

Table 7 shows the TSCF values and uncertainties (in brackets) obtained for ^{134}Cs.

To verify the goodness of the TSCFs obtained using this methodology, a ^{134}Cs activity concentration in a water sample provided by the IAEA (World-Wide Open Proficiency test IAEA-TEL-2014-03) was calculated. The uncorrected and corrected results were 18.2 ± 1.0 Bq kg^{-1} and 20.5 ± 1.2 Bq kg^{-1} (k = 2), respectively. The reference value given by IAEA was 21.4 ± 0.4 Bq kg^{-1}. This shows an important improvement in the results. In fact, the relative difference between the calculated and the reference activity dropped from 15% to 4% when this correction was applied.

6. Conclusions

GEANT4 has been evaluated as a tool for producing simulated data sets which can be used for TSCFs calculation of HPGE detectors. The accuracy of GEANT4 simulations is heavily dependent on the modeled detector geometry. Characterizing a detector is difficult, especially if its technical characteristics are not well known. In this frame, dead layer thickness is of particular importance. The GEANT4 Radioactive Decay Module can simulate a complete decay path of the nuclide under study. A quantitative comparison using statistical methods shows that there were no

Table 5
Results of u statistic (p-value).

Radionuclide	E (keV)	PGAQ-PGSI	PGAQ-PPAQ
^{88}Y	898.0	0.44 (0.326)	2.20 (0.014)
	1836.0	0.94 (0.173)	1.51 (0.065)
^{60}Co	1173.0	0.94 (0.173)	4.42 (0.0000048)
	1332.5	0.54 (0.293)	2.06 (0.011)

The TSCF values and uncertainties (in brackets) calculated for all matrices are shown in Table 4.

Table 6
^{134}Cs Gamma radiation.

E (keV)	Iγ (%)
563.2	8.38
569.3	15.39
604.7	97.60
795.8	85.50
801.9	8.70

Table 7
TSCFs obtained for ^{134}Cs.

Geometry	E (keV)	TSCFs
PGAQ	563.2	1.093 (±0.011)
	569.3	1.148 (±0.009)
	604.7	1.127 (±0.006)
	795.8	1.140 (±0.007)
	801.9	1.200 (±0.014)
PGSI	563.2	1.098 (±0.012)
	569.3	1.180 (±0.010)
	604.7	1.145 (±0.006)
	795.8	1.160 (±0.007)
	801.9	1.205 (±0.015)
PPAQ	563.2	1.106 (±0.009)
	569.3	1.215 (±0.008)
	604.7	1.178 (±0.005)
	795.8	1.244 (±0.006)
	801.9	1.265 (±0.013)

statistically significant differences between measured and simulated efficiencies at a 95% confidence level for the energy range analyzed. Moreover, deviations were distributed for each geometry with a mean deviation of less than 2%. These results confirm the absence of systematic error and the good agreement between simulated and measured efficiencies for each of the geometries.

Using the TSCF evaluation methodology, TSCFs were obtained for ^{88}Y and ^{60}Co to correct the experimental calibration curve, thus avoiding the efficiency underestimation in the energy range between 898 keV and 1836 keV. Using the same methodology, TSCFs were calculated for ^{134}Cs.

The authors plan to use the presented method for calculating TSCFs for a selection of natural isotopes in the future.

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