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MONTE CARLO METHOD

Validation of gamma-ray true coincidence summing effects modeled by the Monte Carlo code MCNP-CP

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When radionuclides decay by cascading photons, the accuracy of the measured nuclide activity may be affected by true coincidence summing effects. The effects can be quantified by Monte Carlo simulations that can handle correlated γ- and X-ray emissions from a radionuclide. Analysis techniques are also available commercially to correct for the effects due to cascading γ-rays. The MCNP-CP code was used to compute the effects in high purity germanium detectors for several commonly used nuclides and geometries and the results were compared to measurements and an analysis technique. Excellent agreement in true coincidence summing corrections predicted by MCNP-CP and the analysis technique was obtained. In addition, the X-ray true coincidence summing effects were evaluated.

Introduction

Radioactive decay of a nuclide to the ground state of its product often occurs through the emission of multiple photons (γ- and X-rays) and electrons. When two or more photons from the same decay are emitted within a short time interval, and are detected within the resolving time of the γ-ray spectrometer, the photons are said to be in true coincidence.

As a result, the measured energy deposit is subject to true coincidence summing (TCS), or cascade summing, hence, the full energy peak (FEP) counts of a γ-ray of interest can be lost (e.g., summing-out) or gained (e.g., summing-in). The magnitude of the TCS effects is a function of the nuclear decay schemes and the measurement efficiency which is also related to source-geometry. Measurements made in highly efficient geometries (e.g., larger detector, closer source, etc.) are affected the most. The probability of summing-out depends on the total efficiencies of the geometry at the photon energies of interest, and the probability of summing-in depends on the FEP efficiencies.[1] Unless the measurement matches the calibration conditions or unless otherwise corrected for TCS effects during spectroscopic analysis, the apparent activity based on the FEP is not a true report of the radionuclide activity.

Canberra's Genie-2000 spectroscopy software (released version 3.1) provides an analytic technique to calculate and apply the correction due to γ-ray TCS effects.[2–4] To determine total efficiency, the cascade summing correction (CSC) algorithm uses a point-source peak-to-total (P/T) efficiency calibration and a mathematically calculated FEP efficiency. Validation analysis of the technique using a comprehensive set of high purity germanium (HPGe) detector characterization data with both cascade and non-cascade nuclides have been reported previously.[5] It has been shown to provide reliable and accurate corrections for a range of problems of practical interest. However, the algorithm in version 3.1 does not take into consideration of the TCS effects between decay γ- and X-rays or annihilation photons. Consequently, previous validation measurement using low energy detectors required absorbing material to adequately attenuate the X-rays.

Efforts are underway to enhance the present CSC algorithm to include corrections for TCS effects due to correlated X-rays and the 511 keV annihilation photons. Effectiveness of the CSC algorithm on handling cascading γ-rays is here further verified and the magnitude of the X-ray TCS effects is evaluated. To this end, we take the advantage of the recent advancement in the software development that has extended the MCNP Version 4c[6] code to perform calculations with correlated nuclear source particles, known as MCNP-CP.[7] The tool allows us to separately evaluate the TCS effects for the analytic γ-rays due to correlated γ- and X-rays/511 keV. Using MCNP-CP, we have computed the TCS effects for a representative set of HPGe detectors for several nuclides and laboratory geometries commonly used to calibrate assay systems and also of interest as unknowns in quantitative measurement such as in activation studies.[8] The TCS effects predicted by MCNP-CP were compared with measurements and with those obtained using the CSC algorithm in the Genie-2000 version 3.1.

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Experimental

The study used archived measurement data from 8 HPGe detectors. Five were different types of coaxial detectors and three were broad energy (BEGe) detectors. Among the five coaxials, two were thin window detectors (XtRa) sensitive to very low energy photons, two were standard p-type coaxial detectors (Coax), and one was a reverse electrode (REGe) or n-type detector. The relative efficiencies of the detectors ranged from 11% to 150%. Each of these detectors has been fully ISOCS/LabSOCS[9] characterized to allow accurate calculation of FEP efficiencies using geometry templates that can be tailored to specific measurement geometries. The measurements consisted of four calibrated voluminous γ-ray source standards measured at five source–detector geometries. These include a near (3 mm from detector end cap) and far (101 mm from detector end cap) filter paper (0.1 mm thick disk with active dia. of 48 mm), a 20 mL liquid scintillation vial (25 mm dia.×51 mm) on detector end cap, a 400 mL cylindrical beaker (76 mm dia.×76 mm) on detector end cap, and a 2.8 L Marinelli beaker. Each voluminous source contains the following nuclides: ^{241}Am (59 keV), ^{109}Cd (88 keV), ^{57}Co (122 keV), ^{139}Ce (166 keV), ^{113}Sn (392 keV), ^{137}Cs (662 keV), ^{54}Mn (835 keV), ^{65}Zn (1115 keV), ^{60}Co (1173, 1332 keV), and ^{88}Y (898, 1836 keV). For the purpose of this study, ^{60}Co, ^{88}Y, and ^{139}Ce were the nuclides of interest with cascade γ-rays, and ^{57}Co (negligible TCS effects), ^{54}Mn, and ^{65}Zn are non-cascading nuclides with energies conveniently close to those of the cascading nuclides.

Since the original purpose of the characterization measurements was not primarily for studying TCS effects, source-based P/T calibrations were not performed. Furthermore, the detectors were no longer available. Therefore, Monte Carlo methods were used to create the P/T calibration curves. Using the validated MCNP detector model, four monoenergetic γ-ray sources with the geometries described earlier were modeled for each detector. It has previously been shown[10] that it is sufficiently accurate to use the MCNP based P/T calibration when applying the CSC algorithm. Together with the FEP efficiencies, CSC algorithm calculates the CSC factors due to correlated decay γ-rays.

For each voluminous source and measurement geometry, MCNP-CP was used to compute the FEP efficiency. The nuclide decay scheme information was taken from the Evaluated Nuclear Structure Data File (ENSDF)[11] available at National Nuclear Data Center. For ^{60}Co, TCS is between decay γ-rays and the FEP efficiencies for 1173 and 1332 keV were computed. In the first simulation, FEP efficiencies (ε_1) were computed without correlation between the two photons, i.e., the 1173 and 1332 keV γ-rays were tracked through the problem geometry in a separate history disregarding the timing of their emission. In the second simulation, the two γ-rays were considered correlated, and the FEP efficiencies (ε_2) were computed by tracking both γ-rays in the same history. The efficiency ratio at each energy:

$$\mathrm{COI}_\gamma = \varepsilon_2/\varepsilon_1$$

provides the CSC factor for each γ-ray. For ^{88}Y, three sets of FEP efficiencies at 898 and 1836 keV were computed. The first two sets of efficiencies were computed the same way as for ^{60}Co, which only involves the decay γ-rays. The third efficiency (ε_3) computation involved not only the correlated decay γ-rays, but also the correlated X-rays. The efficiency ratios at each energy:

$$\mathrm{COI}_\gamma = \varepsilon_2/\varepsilon_1 \text{ and } \mathrm{COI}_{\gamma+x} = \varepsilon_3/\varepsilon_1$$

provide the CSC factors due to correlated decay γ-rays only and due to both correlated decay γ- and X-rays. For ^{139}Ce, the TCS is only between 166 keV γ-ray and EC X-rays. Consequently only two efficiencies at 166 keV are computed: ε_1 is the same as for ^{60}Co, and ε_3 is the same as for ^{88}Y. The efficiency ratio:

$$\mathrm{COI}_{\gamma+x} = \varepsilon_3/\varepsilon_1$$

provides the CSC factor for the 166 keV due to the presence of the EC X-rays.

Results and discussion

The validity of the MCNP-CP, Genie-2000 CSC algorithm, and the contribution of the correlated X-rays to TCS effects were assessed by analyzing the measurement and the computational results. For each detector and geometry, the ratio of measured to certificate activities and its standard deviation were analyzed. The measured activity was obtained based on the measured FEP yield of the nuclide of interest and the ISOCS efficiency calibration of the measurement geometry. The ISOCS efficiency calibration has accounted for voluminous source self-attenuation. The raw measured/certificate activity ratio is defined as:

$$R_0 = \frac{C_{net}}{A \cdot y \cdot \varepsilon_{Isocs}} \tag{1}$$

where C_{net} is the rate loss corrected net count rate at γ-ray energy of interest, A is the certificate activity, y is the γ-ray yield, and ε_{Isocs} is the ISOCS efficiency. The measured/certificate activity ratio corrected by CSC factor from CSC algorithm ($\mathrm{COI}_\gamma^{\mathrm{CSC}}$) is defined as:

$$R_1 = \frac{R_0}{\mathrm{COI}_\gamma^{\mathrm{CSC}}} \tag{2}$$

Deviation in R_1 from unity for detectors insensitive to low energy X-rays is an indicator of the inaccuracy of the CSC algorithm. The ratio between the CSC factors from CSC algorithm ($\mathrm{COI}_\gamma^{\mathrm{CSC}}$) and that given by MCNP-CP with correlated decay γ-rays only ($\mathrm{COI}_\gamma^{\mathrm{MCNP-CP}}$) is defined as:

$$R_2 = \frac{\mathrm{COI}_\gamma^{\mathrm{CSC}}}{\mathrm{COI}_\gamma^{\mathrm{MCNP-CP}}} \tag{3}$$

where R_2 is expected to be unity within simulation uncertainty for all detectors if both methods accounted for the γ-ray TCS effects properly. The measured/certificate activity ratio corrected by CSC factor from MCNP-CP with correlated decay γ- and X-rays ($\mathrm{COI}_{\gamma+x}^{\mathrm{MCNP-CP}}$) is defined as:

$$R_3 = \frac{R_0}{\mathrm{COI}_{\gamma+\mathrm{x}}^{\mathrm{MCNP-CP}}} \tag{4}$$

where R_3 is also expected to be unity for all detectors if MCNP-CP effectively compensate for the TCS effects due to correlated γ- and X-rays. The ratio between R_3 and R_1 is a measure of the magnitude of the TCS effects due to X-rays.

Deviation of the measured activity from the certificate activity for nuclides with non-cascading γ-rays (e.g., ^{57}Co, ^{56}Mn, and ^{65}Zn) occurring at a similar energy region (122, 835, and 1115 keV) as the cascading γ-rays is a direct measure of the absolute accuracy of the detector response characterization. Other sources of systematic uncertainty include the certificate activity and source position uncertainties. The dead time ranges from 1–17% depending on the measurement geometry and is corrected by a reference pulser. Sources of statistical uncertainty include the measurement statistics and simulation precision. The standard deviation is the quadratic sum of the statistical and systematic uncertainties. As a result, the ratios along with their standard deviations for the three cascading nuclides are given in Table 1 for the near filter paper geometry for detector No. 1, and are graphed in Figs 1 and 2 for all measurement geometries and detectors. Of the five geometries available, the efficiencies were the highest (lowest) for the near (far) filter paper geometry. A maximum of 2–3% TCS effects is observed for the least efficient far filter paper geometry (Fig. 1).

It is evident from Fig. 2 that for all standard coaxial detectors (Nos 6, 7, and 8) at all measurement geometries, R_1 overlaps unity within 1–2 standard deviations for ^{60}Co and ^{88}Y. The standard coaxial detectors have a typical front dead layer of 0.5 mm Ge, thus more than 99.9% of 15 keV X-rays from ^{88}Y are attenuated. These detectors are rather insensitive to low energy photons and the TCS effects which is typically around 15–20% is due to high energy decay γ-rays of these nuclides. The result confirms the validation tests done by RUSS et al.[4]

A greater bias for ^{88}Y was observed when analyzing results from detectors without a significant dead layer at the front of the detector (Nos 1 through 5). Such low energy detectors without absorber can also detect the 15 keV and 37 keV X-rays that are in true coincidence with the γ-rays in ^{88}Y and ^{139}Ce. Table 1 and Fig. 2 show that for low energy detectors, R_1 is around 90% for ^{88}Y for near filter paper geometry, a significant deviation from unity compared to the measurement uncertainty. This is primarily due to TCS effects between decay γ- and X-rays which is not corrected by the present CSC algorithm. Consequently, when MCNP-CP takes the X-ray TCS effects into account, R_3 becomes unity within measurement uncertainty as is evident for the near filter paper geometry.

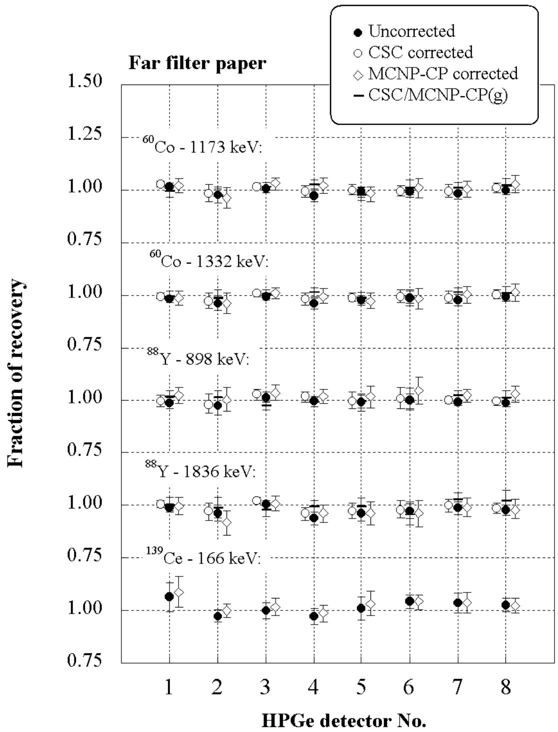

Fig. 1. CSC factors for far filter paper geometry. Dataset from top to bottom of each panel is for γ-rays of ^{60}Co (1173, 1332 keV), ^{88}Y (898, 1836 keV), and ^{139}Ce (166 keV), respectively. The four ratios are labeled as: "Uncorrected" (R_0), "CSC Corrected" (R_1), "CSC/MCNP-CP(g)" (R_2), and "MCNP-CP Corrected" (R_3)

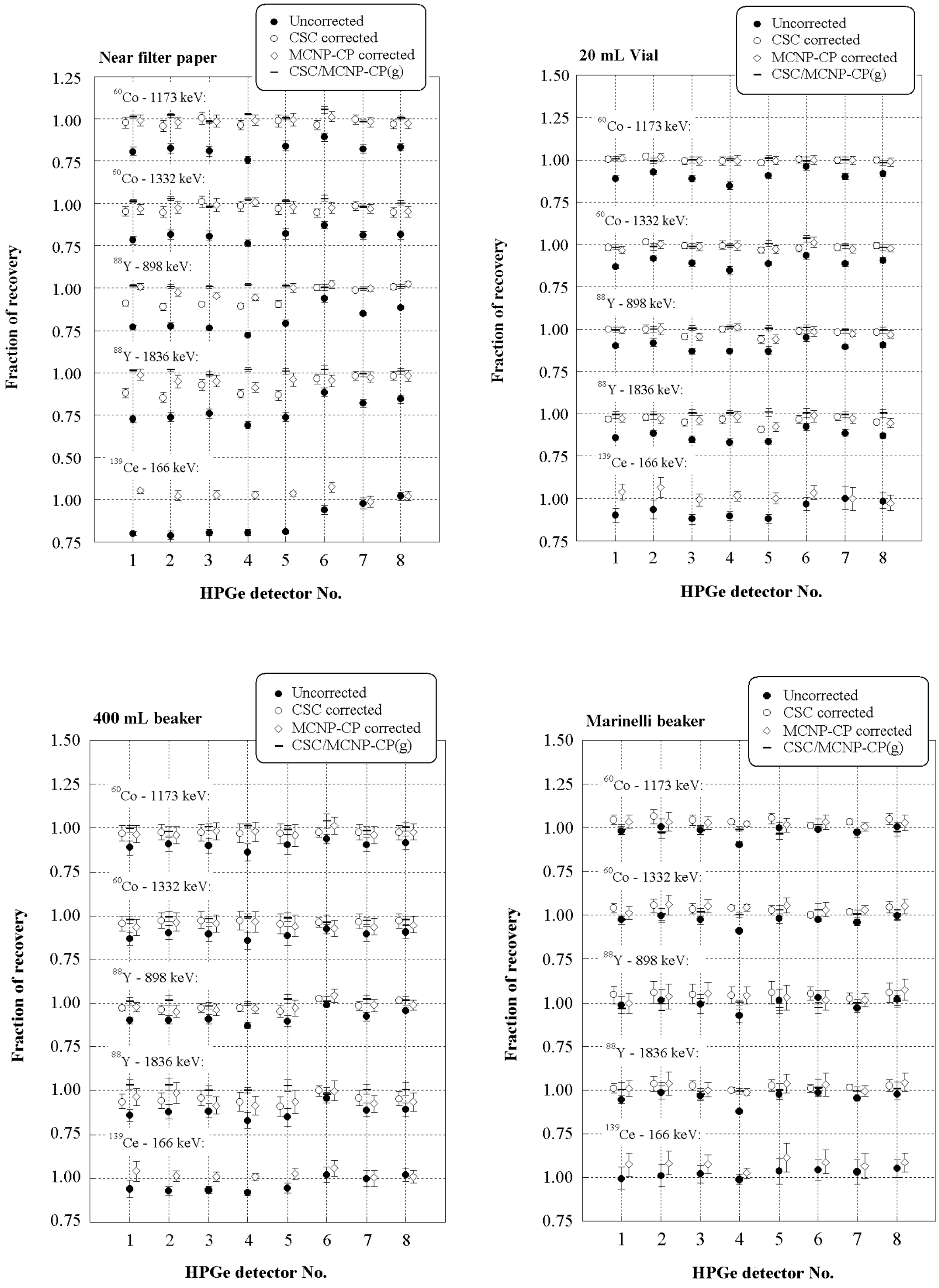

Fig. 2. CSC factors for near filter paper, near 20 mL liquid scintillation vial, near 400 mL beaker, and 2.8-L Marinelli beaker geometries. Dataset from top to bottom of each panel is for γ-rays of ^{60}Co (1173, 1332 keV), ^{88}Y (898, 1836 keV), and ^{139}Ce (166 keV), respectively. The four ratios are labeled as: "Uncorrected" (R_0), "CSC Corrected" (R_1), "CSC/MCNP-CP(g)" (R_2), and "MCNP-CP Corrected" (R_3)

Table 1. The measured to true certificate activity ratios for each γ-ray of the three nuclides of interest for detector No. 1 using near filter paper geometry

Nuclide	^{60}Co		^{88}Y		^{139}Ce
Energy, keV	1173	1332	898	1836	166
R_0	0.808 ± 0.025	0.783 ± 0.024	0.767 ± 0.012	0.725 ± 0.022	0.799 ± 0.009
R_1	0.979 ± 0.030	0.953 ± 0.029	0.908 ± 0.015	0.879 ± 0.027	–
R_2	1.011 ± 0.010	1.015 ± 0.010	1.011 ± 0.010	1.016 ± 0.014	–
R_3	0.989 ± 0.032	0.966 ± 0.031	1.010 ± 0.019	0.988 ± 0.033	1.053 ± 0.013

The ratio between R_3 and R_1 is a measure of the TCS effects due to correlated X-rays. Depending on nuclide and geometry, the TCS effects due to correlated X-rays are comparable to that of γ-rays, and as large as 20% for the cases studied.

Figure 2 also shows that R_2 equals unity to within one standard deviation for nearly all nuclides, detectors, and geometries. This is a strong indication of excellent agreement in CSC factors given by the CSC algorithm and MCNP-CP for the case of decay schemes only having correlated γ-rays.

Conclusions

It has been shown that for HPGe detectors insensitive to low energy X-rays, the Genie-2000 CSC algorithm is an effective and accurate method of compensating for γ-ray TCS effects in cascading nuclide activities. Excellent agreement has been found in CSC factors given by the CSC algorithm and MCNP-CP. Measuring cascading nuclides with correlated X-rays using low energy detectors, the errors due to TCS effects of up to ~20% are typically reduced to 10% or less after the correction. The accuracy of the TCS effects predicted by MCNP-CP due to correlated γ- and X-rays is verified by the measurement results.

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