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A semi-empirical method for calculation of true coincidence corrections for the case of a close-in detection in γ-ray spectrometry

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Abstract In this paper, a semi-empirical method is proposed to determine true coincidence-summing (TCS) correction factors for high resolution γ-ray spectrometry. It needs the knowledge of both full energy peak (FEP) efficiency and total-to-peak (TTP) efficiency curves. The TTP efficiency curve is established from the measurements with a set of coincidence-free point sources. Whereas for a volume source, the coincidence-free FEP efficiency curve is obtained iteratively by using the peaks from almost the coincidence-free nuclides and those from the coincident nuclides in the mixed standard sources. Then the fitting parameters obtained for both TTP and FEP efficiency curves are combined in a freely-available TCS calculation program called *TrueCoinc*, which yields the TCS correction factors required for any nuclide. As an application, the TCS correction factors were determined for the particular peaks of ^{238}U, ^{226}Ra and ^{232}Th in the reference materials, measured in the case of a close-in detection geometry using a well-type Ge detector. The present TCS correction method can be applied without difficulty to all Ge detectors for any coincident nuclide.

Keywords True coincidence-summing · Total-to-peak ratio · Full energy peak efficiency · γ-Ray spectrometry · Well-type Ge detector

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Introduction

In the γ-ray spectrometry, it is a common practice to use both a high efficiency Ge detector and a close-in detection geometry with a large sample usually placed on the end-cap or a small sized-sample placed inside the well of the detector, thus resulting in the highest peak counting statistics for low level activity samples. The proximity to the detector gives to the lower minimum detectable activities (MDA) and reduces the counting time for an observed activity with a desired precision. However, this proximity also causes true coincidence-summing (TCS) effects, resulting in a biased activity value for many nuclides [1]. Therefore, the problem of TCS correction has been investigated for both "point" and "voluminous" sources by many works [2–12]. Especially, when the small samples with low activity are to be analyzed in well-type detectors, the main drawback for those measurements is the presence of high TCS effects in case of multi-photon emitting nuclides [13]. For example, ignoring TCS effects can lead to a typical error with a factor of 2 in the determination of ^{60}Co activity, which is a two-step cascading nuclide [14]. Similarly, the decay products of ^{232}Th, ^{226}Ra, ^{235}U and ^{238}U are frequently encountered in typical environmental measurements and most of them can have serious TCS effects due to their complex decay schemes with multi-cascading transitions [15]. Therefore, self-attenuation and TCS corrections can often be calculated by Monte Carlo modelling (MCNP) [14, 16, 17]. For instance, GESPECOR software was specifically developed for the corrections due to TCS effects and the corrections for self-absorption due to matrix effects by using MCNP calculation methods [18, 19]. The GESPECOR program as a post-analysis correction tool has been applied to all type of detector, including well types, and to all types of sample geometry [20]. Similarly, ISOCS/

LabSOCS software that uses MCNP modelling is a convenient tool for calibrating the detection efficiency as a function of energy for a wide variety of source geometries and activity distributions [21, 22]. However, when using an ISOCS/LabSOCS software, a MCNP modelling of the detector requires a Canberra Inc. characterization of the detector [23]. ISOCS is a useful tool, and fit-for-purpose if it is used for field survey work. The LabSOCS program is applied to a large number of source geometries used for γ-ray measurements. On the other hand, KAYZERO for Windows software uses the effective solid angles for both the point sources placed at a large (reference) distance from the detector where TCS effects are generally assumed to be negligible, and for the geometry of the actual sample, in order to convert the point source efficiency measured at a large (reference) distance from the detector to the efficiency for the actual sample distance [24]. This "classical" approach relating to the use of effective solid angles in the efficiency transfer has been further improved for the close-in detection measurements [25]. In addition, a method was developed to characterise a well-type detector with using a full-energy peak (FEP) and a peak-to-total (PTT) efficiency curves that were determined using coincident photons from a nuclide by Blaauw [26], who used a two-parameter function to describe the energy dependence of PTT ratio and extended it to volume sources. Thus TCS correction is reduced to knowing two efficiencies and decay scheme properties of the nuclides with cascade γ-rays. This method is implemented in Ortec Gamma Vision software, which includes an automatic calculation from either a set of single nuclide standards or a particular mixed standard. Then, the LS (linear-to-square) curve was used to calculate TCS factors with algorithms to point sources, and this useful method has recently been further developed for the extended samples by Vidmar et al. [12] assuming as if the sample were a point-source, if the LS curve introduced by Blaauw and Gelsema [9] is known and applied properly. Lastly, a simple method was developed for the determination of TCS corrections using MCNP method [27, 28].

In the present work, a semi-empirical TCS correction method is suggested which can be applied to all Ge detectors without difficulty in practice. But it needs to determine the total-to-peak ratios, $(\mathrm{TTP}) = \varepsilon_t/\varepsilon_p$ from the measurements of traditional coincidence-free point sources and the knowledge of an undisturbed, coincidence-free FEP efficiency curve, ε_p from the measurements of the well known mixed-standard sources. In determining the coincidence-free FEP efficiency for a volume source, mostly the coincidence-free nuclides but also a few coincident nuclides such as ^{60}Co and ^{88}Y are used. In case of ^{60}Co and ^{88}Y, an iterative procedure is applied to obtain a coincidence free-FEP efficiency curve for a volume source by using TrueCoinc program, developed by Sudàr [29]. The presented method was tested in the case of a close-in detection geometry which might be considered a worst case in view of TCS effects. For the application of the proposed TCS correction method, certified reference materials (CRMs) were measured in small tubes inside the well of the detector because they contain decay products of ^{235}U, ^{238}U, ^{226}Ra and ^{232}Th of which most of them have many multi-photon transitions causing to TCS effects.

Experimental

The experiments were performed with a well-type Ge detector (Canberra Inc., GCW 4023) with the well diameter of 16 mm and the well depth of 40 mm. Its crystal is a p-type HPGe with a 66.5 mm diameter and a 67 mm length. The detector has a measured relative efficiency of 44.8%, an energy resolution of 2.0 keV and a peak-to-Compton ratio of 60.8:1 at 1332.5 keV of ^{60}Co. A shield of 10 cm lead thickness graded with a 1 mm thick tin and 1.6 mm thick copper liners for reducing the 72–88 keV region Pb X rays was used. The shield was jacketed by a 9.5 mm steel outer housing with a top-opening lead cover (Canberra Model 747). The detector was centred in the shield to minimize scattered radiation from it. The floor of lead shield has a 11.4 cm diameter hole blocked by an annular lead plug in which accommodate only a dipstick cryostat, the detector cables and a hose of 3–5 mm outer diameter. In order to purge radon and thoron gas contamination around the sample, the nitrogen (N_2) gas with a flow rate of 1–2 L min^{-1}, supplied from a separate N_2 gas storage through the plastic tubing was flushed inside the shield. The detector was interfaced to a 16 K channels ADC conversion/MCA spectral memory analyzer (Canberra Multiport II) operating through a γ-ray spectroscopy software (Canberra Genie 2000). The amplifier was adjusted with a gain of 0.75 keV/channel to collect 4096 channel spectra.

The used point sources are obtained from Eckert & Ziegler Isotope Products Inc. They have the total uncertainties, generally less than $\pm 3\%$ at a 99% confidence level for the certified activities between 35 and 389 kBq, deposited in the capsules (25.4 mm outer diameter $\times$ 3.14 mm thick).

For the FEP calibration, the mixed-standard source spiked with sand (purchased from Isotope Products Inc.), containing the nuclides ^{210}Pb, ^{109}Cd, ^{57}Co, ^{123m}Te, ^{51}Cr, ^{113}Sn, ^{85}Sr, ^{137}Cs, ^{88}Y, ^{60}Co, and the standard (IAEA RGK-1) containing 44.8 wt% K were measured inside the well of the detector. The small polystyrene tubes of 1 mm thickness and a 1.4 cm internal diameter were filled in a 3.5 cm filling height. The powder CRMs and the mixed-standard were sealed tightly. The CRM samples were kept

for 6 months to attain radioactive equilibrium. Since the procedures for sample preparation, spectral interferences and self-absorption corrections have already been described in detail in the previous work of Yücel et al. [30], they were not repeated in this paper to keep space at minimum. The measurement periods set between 100,000 and 200,000 s were so high that statistical precisions on the FEP areas were achieved at the order of 2–3% for the spectra of the CRMs. Also, the dead time and count rate losses due to pulse pile-up were negligible for the CRMs for the present well geometry. Room background spectrum was also collected and then subtracted from the sample spectra. At least three separate measurements were performed for each sample.

Methodology

The present approach consists of the procedures for (1) TTP efficiency calibration, (2) FEP efficiency calibration, and (3) Calculation of TCS factors by using TrueCoinc program.

Total-to-peak efficiency calibration

In the present approach, the first step is to obtain a TTP efficiency curve as a function of energy. In the measurements, the γ-ray spectra were obtained from the "single" energy, almost coincidence-free point sources ^{241}Am (59.5 keV), ^{109}Cd (88.0 keV), ^{57}Co (122.1 keV), 661.6 keV (^{137}Cs) ^{54}Mn (834.8 keV) and ^{65}Zn (1115.5 keV). Since some of the point sources have high activities and their sizes are too big for accommodating them inside the well of the detector, these sources were placed at the entrance of the well and also these measurements were repeated at a given distance of 4.5 cm from the end-cap in order to compare with those obtained in the entrance of the well. The amplifier was set up in pile-up rejection circuitry on mode (PUR-ON) to recover to a large extent random coincidences (pile-up effects). The measurements were also carried out by using a 3.175 mm thick Al absorber (840 mg cm^{-2}) interposed between source and detector because of filtering out low energy X-rays especially at 88.0 keV energy of ^{109}Cd. Except for ^{109}Cd, however, the results for TTP ratios from other point sources did not vary much. This implies that γ-X ray coincidences can be negligible. In case an absorber was placed between the source and the detector, it was found that the absorber such as 2 mm thick Cu was sufficiently effective to diminish the effects of Pb KX-rays of 72–84 keV on the left shoulder side of the 88.0 keV γ-ray of ^{109}Cd, however it also has led to underestimation of the TTP ratios especially for low energy peaks, say, below 100 keV. This shows that the absorbers can be used between source and detector to attenuate the X-rays, however, the transmission fraction through a given thickness of any absorber can be varied.

In this procedure a background spectrum was always subtracted from each source spectrum striping channel by channel basis, i.e., source spectrum = original source spectrum $-$ (T_s/T_b)x background, where T_s and T_b are the measurement periods for the source and the background spectrum, respectively. Then, a cut-off channel (extrapolation to zero) $EZ_{cut\text{-}off}$ for each nuclide was defined at a channel number greater than that corresponding to highest X-ray peak energy. However, it is expressed by Lee et al. [31] that the cut-off limit of $\sim$20 keV is to be sufficient for the total efficiency calculation. In fact, the $EZ_{cut\text{-}off}$ of $\sim$20 keV is quite lower than that of the highest X-ray energy of 36.5 keV for ^{137}Cs source. Hence, as given in Table 1, a cut-off energy for a TTP calibration using the traditional sources can be varied between 20 and 40 keV in order to exclude almost all low energy X-rays from the

Table 1 Experimental and fitted total-to-peak ratios for a p-type well Ge detector

Nuclide	Energy (keV)	$EZ_{cut\text{-}off}$[a] (keV)	From present approach			From Genie 2000 software		
			Measured TTP ratio	Fitted TTP ratio[b]	% Deviation	Measured TTP ratio[a]	Fitted TTP ratio[c]	% Deviation
^{241}Am	59.54	32.6	2.064 ± 0.003	2.061	0.16	1.557 ± 0.001	1.571	−0.90
^{109}Cd	88.03	33.3	1.564 ± 0.011	1.637	−4.67	1.950 ± 0.013	1.437	26.32
^{57}Co	122.06	27.0	1.563 ± 0.003	1.562	0.06	1.444 ± 0.003	1.485	−2.83
^{137}Cs	661.66	41.6	4.016 ± 0.005	3.972	1.08	4.050 ± 0.005	4.082	−0.79
^{54}Mn	834.82	27.0	4.441 ± 0.011	4.697	−5.76	4.396 ± 0.012	4.790	−8.98
^{65}Zn	1115.55	27.2	5.676 ± 0.007	5.644	0.57	5.609 ± 0.006	5.716	−1.91

[a] However, $EZ_{cut\text{-}off}$ is chosen as 40 keV for all measured spectra used for PTT calibration in the Genie 2000 software, where PTT means peak-to-total ratio corresponding to the reciprocal of total-to-peak ratio TTP

[b] For TTP fit: $a_1 = 24.542$, $a_2 = -12.7045$, $a_3 = 2.15018$, $a_4 = -0.114428$, $R^2 = 0.9985$

[c] For TTP fit: $a_1 = 17.621$, $a_2 = -9.452$, $a_3 = 1.648$, $a_4 = -0.08883$, $R^2 = 0.9966$

evaluation, provided that γ-X coincidences are negligible in actual measurement conditions. Later, the $EZ_{cut\text{-}off}$ energy corresponding to $EZ_{cut\text{-}off}$ channel can be determined taking by average number of the counts in the "window" of four channels to left side and right side of the $EZ_{cut\text{-}off}$ channel as suggested by Venkataraman [32]. Finally, one can estimate the "total" number of the counts T as

$$T = \sum_{i=EZ_{cut-off}}^{i=R} C_i + C_{ave} \times EZ_{cut-off} \quad (1)$$

where C_i is the count at ith channel, R is the channel number corresponding to the right side of the full-energy peak, C_{ave} is the average count at the $EZ_{cut\text{-}off}$ channel, calculated from the total counts in the "window" dividing by nine channels. The FEP area, N_p can be obtained by standard procedures subtracting a linear or stepwise Compton continuum from the gross counts in the peak as done in any commercial spectrum analysis programs. The measured TTP ratios are then fitted to an exponential logarithmic function valid for a p-type Ge detector as follows:

$$TTP = \exp - [a_1 + a_2 \ln(E) + a_3 \ln^2(E) + a_4 \ln^3(E)] \quad (2)$$

The resulted TTP ratios are given in Table 1, together with the fitting parameters (a_1, a_2, a_3, a_4) and relative deviations from the experimental values.

Alternatively, a TTP ratio curve can also be obtained by use of the PTT calibration module of Genie 2000 software that has a well-defined algorithm for the PTT calibration [1]. The reciprocals of PTT ratios obtained by Genie 2000 program are also given in Table 1. However, in this automatic calculation, one must be careful while choosing the $EZ_{cut\text{-}off}$ channel as an input value because Genie 2000 program allows only one option to user for all different source spectra to be processed [33]. Hence in this work the $EZ_{cut\text{-}off}$ channel was chosen as 40 keV for the determination of PTT ratios through Genie 2000 program.

Additionally, a threshold of $\sim$18 keV was set by ADC discriminator to offset many X-rays in the low energy region of the γ-ray spectrum because a p-type Ge detector is used. Although there is a thin contact ($\sim$0.3 mm) inside the well and a thicker contact ($\sim$0.5 mm) outside the well of the present detector, the thicknesses of these materials transmitted through photons are assumed to be sufficient for neglecting γ-KX ray coincidences. For instance, the polystyrene sample tube, Al thickness inside the well and inside Ge dead layer serve as a filter for soft X- and γ-rays in the well region of Ge crystal. This assumption is also supported by the argument that γ-KX ray coincidences are not so important for coaxial p-type Ge detectors [1]. Hence γ-KX ray coincidences below 40 keV were not accounted for the present calculation of TCS factors for a p-type well Ge counting geometry, although TrueCoinc program is capable of calculating γ-X ray TCS correction factors.

Table 2 Experimental and fitted full-energy peak efficiencies for a small tube placed inside the well of a Ge detector

Nuclide	Energy (keV)	True coincidence-summing factor[a] F_{COI}	Full-energy peak efficiency				
			Apparent efficiency ε_p	First fit[b] ε_p	Deviation from first fit %	Final fit[c]	Deviation from final fit %
^{210}Pb	46.54	1.000 ± 0.004	0.5215 ± 0.0172	0.4877	6.47	0.4877	6.47
^{109}Cd	88.03	1.000 ± 0.003	0.6530 ± 0.0060	0.6599	−1.06	0.6599	−1.06
^{57}Co	122.06	0.999 ± 0.004	0.6294 ± 0.0056	0.5805	7.77	0.5805	7.77
^{123m}Te	158.97	0.999 ± 0.003	0.4746 ± 0.0046	0.4779	−0.69	0.4812	−1.39
^{113}Sn	391.69	1.000 ± 0.004	0.2348 ± 0.0022	0.2320	1.19	0.2325	1.00
^{137}Cs	661.66	1.000 ± 0.004	0.1457 ± 0.0012	0.1455	0.10	0.1480	−1.55
^{88}Y	898.03*	0.920 ± 0.006	0.0712 ± 0.0007	0.1074	−50.80	0.1095	0.67
^{60}Co	1173.23*	0.877 ± 0.003	0.0525 ± 0.0005	0.0811	−54.42	0.0850	−1.80
^{60}Co	1332.50*	0.855 ± 0.003	0.0455 ± 0.0004	0.0706	−55.11	0.0760	−1.39
^{40}K	1460.83	1.000 ± 0.004	0.0632 ± 0.0002	0.0637	−0.08	0.0685	−8.38
^{88}Y	1836.05*	0.807 ± 0.003	0.0333 ± 0.0003	0.0491	−47.41	0.0599	−1.10

[a] Since the star marked γ-lines have cascade summations, F_{COI} factors in this column are obtained after the first fit, however, their final fit ε_p values for these star marked γ-lines given in 7th column are determined by use of their final F_{COI} factors that are obtained after fourth fit, as given in Table 3

[b] For first ε_p fit in the low energy region—$46.5 < E < 392$: $b_1 = -42.29$, $b_2 = 24.76$, $b_3 = -4.758$, $b_4 = 0.2941$, $R^2 = 0.9918$, in the high energy region—$392 < E < 1460.8$: $c_1 = 0.2771$, $c_2 = 0.2876$, $c_3 = -0.09654$, $R^2 = 0.9999$

[c] For final ε_p fit in the low energy region—$46.5 < E < 392$: $d_1 = -42.29$, $d_2 = 24.76$, $d_3 = -4.758$, $d_4 = 0.2941$, $R^2 = 0.9918$, in the high energy region—$392 < E < 1836.1$: $e_1 = -43.91$, $e_2 = 20.85$, $e_3 = -3.28$, $e_4 = 0.1645$, $R^2 = 0.9995$

Full-energy peak efficiency calibration

The standard, well-known FEP efficiency calibration procedure starts with use of the coincidence-free ε_p values, obtained experimentally by using ^{210}Pb, ^{109}Cd, ^{57}Co, ^{123m}Te, ^{113}Sn, ^{137}Cs and ^{40}K. The apparent ε_p values were then approximated to a function in the form of Eq. 2 and the resulted curve called here as the "first fit", covering the energy range up 40 keV to $\sim$1.5 MeV which links two parts, low (46.5–392 keV) and high (392–1460.8 keV) energy regions. The results for apparent and fitted ε_p values are given in Table 2. But the ones from the apparent efficiencies for the γ-rays from the coincident nuclides such as ^{60}Co and ^{88}Y are not used in the first fit. Thus, the coefficients of the first fit obtained for both TTP and first FEP efficiency curves are combined in TrueCoinc program, which calculates TCS correction factors required for any coincident nuclide. It is worth noting that the γ-ray peaks used in initial fitting FEP efficiency curve are assumed to be almost undisturbed ε_p values, i.e., coincidence-free or slightly interfered peaks are used. In fact, as a novelty in this approach, these peaks are repeatedly used as the "reference" peaks in the subsequent fits in which their TCS correction factors are normally taken to be unity, $F_{COI} \cong 1$. If one uses the definition of the TCS correction factor [11], $\varepsilon'_p = \varepsilon_p / F_{COI}$ where ε'_p is the TCS corrected-FEP efficiency value and ε_p is the apparent efficiency, the calculated ε'_p values for each of the γ-rays at 1173.2 and 1332.5 keV of ^{60}Co and at 898.0 and 1836.1 keV of ^{88}Y are given in Table 3. After a few manual iterations were made using a spreadsheet data program, the resulted ε'_p values for all peaks were fitted to a function in the form of Eq. 2. The final fit has two linked parts, describing low (46.5–392 keV) and high (392–1836 keV) energy regions. Thus after few iterations, the final FEP efficiency curve can assumed to be coincidence-free, i.e., TCS corrected FEP efficiency for a close-in detection well geometry a few. The present iterative fitting procedure is repeated manually until no change in TCS factors for the interested γ-rays from ^{60}Co and ^{88}Y is observed. In our case, this procedure was stopped after four iterations. After fourth fitting, each of the final F_{COI} factors for the peaks of ^{60}Co and ^{88}Y approaches to almost a unity. This means that the TCS effects for these four peaks were fully corrected.

Calculation of true coincidence-summing correction

In order to compute TCS correction factors for the γ-rays from any coincident nuclide, a freely available and user-friendly TrueCoinc program was used. To this end, the fitting parameters obtained for both FEP and TTP calibration curves given in Tables 1 and 2 are introduced in the menu of TrueCoinc program. Additionally, a cut-off energy limit is predetermined to exclude γ-KX-ray coincidences for the low-energy region. Before the run, the database is selected for any nuclide. The decay scheme properties for the selected nuclide can be read either from an update ENDSF database, which is accessible online or can easily be read from a CD-ROM version of the Table of isotopes [34].

After the selection of a nuclide from the menu of TrueCoinc program, the TTP ratios are used for calculating total efficiency value ε_t together with the known ε_p value at a given energy. This can be accomplished by using the fitting coefficients, already fed into its menu. In the report page of TrueCoinc program, the resulted TCS factors F_{COI} for any nuclide are also displayed as a multiplicative factor consisting of the "summing-in" and "summing-out" factors, which are also calculated by the program.

Results and discussion

In the present method, a TTP ratio calibration was obtained by employing different cut-off energies $EZ_{cut\text{-}off}$. In addition, a TCS corrected-FEP efficiency curve for a volume source was obtained by using mixed-standard sources. Then, TCS correction factors are easily calculated with a

Table 3 True coincidence-summing correction factors for the γ-rays of ^{60}Co and ^{88}Y for a small tube source measured inside the well of a 44.8% relative efficiency Ge detector

Nuclide	Energy (keV)	Apparent FEP efficiency, ε_p	True coincidence-summing correction factors, F_{COI}			
			After 1st iteration	After 2nd iteration	After 3rd iteration	After 4th iteration
^{88}Y	898.03[a]	0.0712 ± 0.0007	0.920	0.663	0.767	0.646
^{60}Co	1173.23[a]	0.0525 ± 0.0005	0.877	0.568	0.737	0.629
^{60}Co	1332.50[a]	0.0455 ± 0.0004	0.855	0.534	0.706	0.607
^{88}Y	1836.05[a]	0.0333 ± 0.0003	0.807	0.498	0.625	0.562

FEP full-energy peak

[a] For these star marked γ-lines, F_{COI} factors did not change any longer after the 4th iteration, therefore, F_{COI} factors were used to obtain final fitted ε_p values already given in the 7th column of Table 2, called a final fit

freely available TrueCoinc program, based on ENDSF decay database. The function in the form of Eq. 2 was chosen in the present calibrations since it is generally suitable for p-type Ge detectors. However, it is possible to use another forms of the functions for the efficiency calibrations depending on type of the detector and the conditions [3].

The TTP ratios can be assumed to be an advantage for this type of description of total efficiency ε_t by Eq. 2, since TTP ratios are independent of a given measurement geometry and the shape of ε_t curve. Therefore, the use of TTP ratio curve can be adequate for the characterization of a Ge detector to estimate TCS factors, also for the voluminous samples [35]. The TTP ratios were determined experimentally at the entrance of the well of a detector and at a distance of 4.5 cm from the end-cap, however, they did not vary much for both measuring geometries. It can still be thought that there is a limitation for the present TTP calibration curve for its energy dependence. For instance, the useful energy range for such a TTP curve could not be extended from 1115.6 keV (^{65}Zn) to much higher energy values, say, 2.7 MeV, due to either lack of coincidence-free standard sources or avoiding the use of sum peaks of ^{60}Co and ^{88}Y to extend the efficiency calibration energy range up to 2.7 MeV [19]. This might lead to either underestimation or overestimation of TCS factors for the peaks in high energy region, from 1.5 up to 2.7 MeV. However, it has been shown in the closed-end coaxial Ge detectors that the total efficiency obtained by TTP ratios does not vary strongly with increasing energy and does not vary much with the source-detector distance [3, 36, 37].

Additionally, a rough comparison can be made amongst TCS correction factors as given in Table 4. For this, the F_{COI} factors were obtained for the samples of nearly equal volumes (5.4 and 5.5 cm^3) measured inside the wells of 218 and 300 cm^3 p-type Ge detector, respectively [14]. Although it is known that two detectors have different well sizes, sample volumes, dead layers and crystal volumes, thus leading to different TCS effects between two detectors, the F_{COI} factors obtained from a 218 cm^3 well Ge detector are generally smaller than those from a 300 cm^3 well Ge detector, as expected. This indicates that the present semi-empirical method works out well for the determination of the TCS factors by using TrueCoinc algorithm, on condition that the well-established FEP and TTP efficiency curves are used in it. The TCS correction factors required for the analysis of ^{226}Ra, ^{232}Th, ^{235}U and ^{238}U are given in Table 5, together with the necessary self-absorption factors for CRM samples. The resulted activities given in Table 6 were corrected for both self-absorption effects and the spectral interferences besides TCS corrections.

In the present analysis, the uncertainty sources on the experimental data are mainly due to the counting statistics, the γ-ray emission probabilities, the detection efficiency, weight determinations and the uncertainties in the certified activities quoted. They are combined using the usual law of propagation of uncertainty, adding an additional systematic uncertainties of 1.5–2% due to possible variations in sample heights. Standard deviations of three measurements for each CRM sample was calculated from the internal (pooled) variance. Thus, the overall measurement uncertainties on the resulted activities within $\pm 1\sigma$ confidence limits were

Table 4 Comparison of true coincidence-summing correction factors for two different well Ge detectors

Nuclide	Energy (keV)	True coincidence-summing correction factor[a], F_{COI}		
		For a 218 cm^3 well-type Ge detector[b]	For a 300 cm^3 well-type Ge detector[c]	
		Present method Experimental	Experimental[d]	MCNP calculated[d]
^{234}Th	63.29	1.000 ± 0.007	1.01 ± 0.01	1.00 ± 0.01
^{226}Ra	186.1	1.001 ± 0.004	0.95 ± 0.02	1.00 ± 0.02
^{214}Pb	295.21	0.982 ± 0.007	0.99 ± 0.02	0.98 ± 0.01
^{214}Pb	351.92	1.006 ± 0.008	0.97 ± 0.02	1.02 ± 0.01
^{214}Bi	609.31	2.011 ± 0.017	2.15 ± 0.04	1.91 ± 0.03
^{214}Bi	1120.28	2.053 ± 0.081	2.55 ± 0.05	2.21 ± 0.06
^{214}Bi	1238.11	2.066 ± 0.075	2.50 ± 0.07	1.99 ± 0.07
^{214}Bi	1764.49	0.990 ± 0.005	0.98 ± 0.02	1.01 ± 0.03

[a] These values are inverse of the mean values of F_{COI} factors given in the columns 4 and 5 in the Table 5

[b] For a 5.4 cm^3 sample measured inside the well of a 44.8% relative efficiency Ge detector (from Canberra Inc.) with the well depth of 40 mm and well diameter of 16 mm

[c] For a 3 cm^3 and 8 cm^3 samples measured inside the well of a 60% relative efficiency Ge detector (from Canberra Inc.) with the well depth of 54 mm and well diameter of 33 mm

[d] Literature values are taken from the paper of Laborie et al. [14]

Table 5 Calculated factors for true coincidence-summing and self-absorption effects for relevant γ-rays for a small tube placed inside the well of a 44.8% relative efficiency, p-type Ge detector

Nuclide	Energy[a] (keV)	γ-Ray emission probability	TCS factors using T/P ratios from the present method	TCS factors using T/P ratios from Genie 2000	Self-absorption factors[b], F_s			
		I_γ (%)	F_{COI}	F_{COI}	DL-1a	DH-1a	UTS-1	UTS-2
^{234}Th	62.86	0.0211	1.000 ± 0.003	1.000 ± 0.003	1.16	1.19	1.49	1.51
^{234}Th	63.29	4.8378	1.000 ± 0.005	1.000 ± 0.005	1.16	1.19	1.48	1.50
^{232}Th	63.81	0.023	0.998 ± 0.003	0.999 ± 0.003	1.16	1.19	1.47	1.49
^{231}Th	63.86	0.267	0.767 ± 0.003	0.775 ± 0.003	1.15	1.19	1.47	1.49
^{235}U	185.71	57.2	0.930 ± 0.005	0.933 ± 0.006	1.09	1.09	1.12	1.12
^{226}Ra	186.10	3.5	0.998 ± 0.003	0.999 ± 0.003	1.09	1.09	1.12	1.12
^{214}Pb	295.21	18.5	1.018 ± 0.005	1.018 ± 0.005	1.07	1.08	1.09	1.09
^{228}Ac	338.32	11.2518	0.852 ± 0.004	0.853 ± 0.004	1.07	1.07	1.09	1.08
^{214}Pb	351.92	35.8	0.994 ± 0.006	0.994 ± 0.006	1.07	1.07	1.08	1.08
^{208}Tl	583.19	30.58	0.456 ± 0.011	0.453 ± 0.011	1.06	1.06	1.07	1.07
^{214}Bi	609.31	44.7905	0.496 ± 0.003	0.509 ± 0.003	1.05	1.06	1.07	1.06
^{212}Bi	727.33	6.5789	0.809 ± 0.042	0.810 ± 0.042	1.05	1.06	1.06	1.06
^{228}Ac	911.20	26.6	0.857 ± 0.007	0.858 ± 0.007	1.04	1.05	1.06	1.05
^{228}Ac	968.97	16.1728	0.868 ± 0.004	0.869 ± 0.004	1.04	1.04	1.05	1.05
^{228}Ac	1000.69	0.00535	0.333 ± 0.001	0.332 ± 0.001	1.04	1.04	1.05	1.05
^{234m}Pa	1001.03	0.837	1.067 ± 0.004	1.067 ± 0.004	1.04	1.04	1.05	1.05
^{214}Bi	1120.28	14.7968	0.490 ± 0.009	0.484 ± 0.008	1.04	1.04	1.05	1.05
^{214}Bi	1238.11	5.8588	0.472 ± 0.009	0.496 ± 0.008	1.04	1.04	1.05	1.05
^{214}Bi	1764.49	15.3568	1.010 ± 0.004	1.010 ± 0.004	1.03	1.03	1.04	1.04

[a] This column indicates the analytical and interference peaks used for the determination of ^{226}Ra, ^{232}Th, ^{235}U and ^{238}U in CRMs

[b] These factors were calculated from the data taken from NIST XCOM database [38] for a small tube placed inside the well of a Ge detector

Table 6 Certified and measured specific activities of CRM samples placed inside the well of a 44.8% relative efficiency, p-type Ge detector

Samples[a]	^{238}U			^{232}Th			^{226}Ra		
	Certified activity A_{cert}	Measured activity A_{meas}	Relative deviation[d] %	Certified activity A_{cert}	Measured activity A_{meas}	Relative deviation[d] %	Certified activity A_{cert}	Measured activity A_{meas}	Relative deviation[d] %
DL-1a	1432 ± 37	1518 ± 134	−5.66	308 ± 16	339 ± 18	−0.32	1400 ± 40	1453 ± 20	−3.65
DH-1a	32462 ± 41	30167 ± 760	−5.55	3692 ± 123	3629 ± 48	1.73	31500 ± 1100	32376 ± 420	−2.71
UTS-1	[b]	906 ± 124	[c]	680 ± 16	626 ± 16	8.63	3670 ± 38	3328 ± 376	10.28
UTS-2	[b]	1014 ± 150	[c]	880 ± 18	793 ± 21	10.97	5600 ± 46	5515 ± 46	1.54

[a] These samples were obtained from by CANMET-Mining and Mineral Sciences Laboratories, Canada

[b] There is no quoted activity in its certificate

[c] There is no calculated or certified value

[d] The percentage relative deviation = $100 \times (A_{cert}/A_{meas} - 1)$, where A_{cert} is certified activity and A_{meas} is measured activity in Bq kg^{-1}, corrected for TCS, spectral interference and self-absorption effects

amounted up to 14.7%. It is seen in Table 6 that the percentage relative deviations from the certified activities varied between 1.5 and 11%, which are generally within confidence limits. This indicates that the measured activities generally agree with those certified activities. However, the results show that among the measured samples such as UTS-1 and UTS-2 might also have radioactive equilibrium breaks due to some chemical separation processes that might be undergone on these materials. It is understood that these relative biases between measurement precision and all systematic errors are discernable although CRMs have the complex gamma-ray spectra for the nuclides of natural decay series. The activity results given in Table 6 also indicate that the influence of the efficiency variations

depending on TCS corrections be carefully taken into account, especially when the samples are to be counted in a "close" measurement geometry condition.

Conclusions

A semi-empirical method has been described for the calculation of TCS correction factors. The method was tested on a p-type, well Ge detector using the reference materials. It requires the experimentally determined, coincidence-free TTP and FEP efficiency calibration curves. This requirement can be met by use of a set of standard point sources and the mixed standards, generally available in every laboratory. The suggested method works out for the well-type Ge detector in case of a close-in detection geometry. It has some distinctive features resulting in fairly accurate TCS correction factors. Therefore it can be a practical way for the calculation of TCS correction factors for any coincident nuclide, and it can also be applied without difficulty to all other Ge detectors for the volume samples.

However, it is worth noting that the above argument, i.e., "the approach without difficulty" is also true for other commercial programs which are, for example, the well known KAYZERO for Windows, GESPECOR, LabSOCS and Ortec GammaVision, etc. Additionally, it is worthwhile that there is no need for any source measurements to be performed for efficiency calibrations because these programs generally use MCNP modelling. Accordingly, the simplicity of our method does not hinder the advantages and their advanced features of the commercially available TCS programs above. Nevertheless, it is supposed that in practice the present method can still be employed as a cost-free tool by the γ-ray spectroscopy users when a commercial software for TCS correction is not available.

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References

1. Kolotov VP, Koskelo MJ (1998) J Radioanal Nucl Chem 223:95
2. Andreev DS, Erokhina KI, Zvonov VS, Lemberg IKh (1973) Izv Akad Nauk SSSR Ser Fiz 37:1609
3. Debertin K, Helmer RG (1988) Gamma- and X-ray Spectrometry with Semiconductor Detectors. Elsevier Science Publishers B.V., North-Holland, Amsterdam
4. Moens L, De Corte F, Simonits A, Xilei L, De Wispelaere A, De Donder J, Hoste J (1982) J Radioanal Nucl Chem 70:539
5. Korun M, Martincic R (1993) Nucl Instrum Methods Phys Res A 325:478
6. Kolotov VP, Atrashkevich VV, Gelsema SJ (1996) J Radioanal Nucl Chem 210:183
7. Sima O (2000) J Radioanal Nucl Chem 244:669
8. Korun M (2001) Nucl Instrum Methods Phys Res A 457:245
9. Blaauw M, Gelsema SJ (2003) Nucl Instrum Methods Phys Res A 505:311
10. Vidmar T, Korun M (2006) Nucl Instrum Methods Phys Res A 556:543
11. Arnold D, Sima O (2006) Appl Radiat Isot 64:1297
12. Vidmar T, Korun M, Vodenik B (2007) Appl Radiat Isot 65:243
13. Sima O (2000) Nucl Instrum Methods Phys Res A 450:98
14. Laborie J-M, Le Petit G, Abt D, Girard M (2000) Appl Radiat Isot 53:57
15. Garcia-Talavera M, Leadermann JP, Decombaz M, Daza MJ, Quintana B (2001) Appl Radiat Isot 54:769
16. Sima O, Arnold D (1996) Appl Radiat Isot 47:889
17. Wang TK, Hou IM, Tseng CL (1999) Nucl Instrum Methods Phys Res A 425:504
18. Sima O, Arnold D, Dovlete C (2001) J Radioanal Nucl Chem 248:359
19. Arnold D, Sima O (2004) Appl Radiat Isot 60:167
20. Gilmore GR (2008) Practical gamma-ray spectrometry, 2nd edn. Wiley, Chichester, England
21. Venkataraman R, Bronson F, Alrashkevich V, Young BM, Field M (1999) Nucl Instrum Methods Phys Res A 422:450
22. Zhu H, Venkataraman R, Mueller W, Lamontagne J, Bronson F, Morris K, Berlizov A (2009) Appl Radiat Isot 67:696
23. Russ W, Venkataraman R, Bronson F (2005) J Radioanal Nucl Chem 264:193
24. DSM Research, Kayzero for Windows User's Manual, Ver. 2, Geleen, NL, 2005
25. De Wispelaere A, De Corte F (2006) Nucl Instrum Methods Phys Res A 564:645
26. Blaauw M (1998) Nucl Instrum Methods Phys Res A 419:146
27. Dryák P, Kovar P, Suran J (2002) Appl Radiat Isot 56:111
28. Dryák P, Kovar P (2009) J Radioanal Nucl Chem 279:385
29. Sudár S (2002) "TRUECOINC", a software utility for calculation of the true coincidence correction, in the specialized software utilities for gamma spectrometry, IAEA-TECDOC-1275, pp 37–38, International Atomic Energy Agency, Vienna, ISSN 1011-4289
30. Yücel H, Solmaz AN, Köse E, Bor D (2009) Appl Radiat Isot. doi:10.1016/j.apradiso.2009.07.11 (Accepted paper)
31. Lee M, Park TS, Woo JK (2008) Appl Radiat Isot 66:799
32. Venkataraman R, Croft S, Russ WR (2005) J Radioanal Nucl Chem 66:183
33. Customization Tools Manual (2004) Canberra spectroscopy software Genie-2000, Ver. 3.0, Canberra Inc., Meriden, USA
34. Firestone RB, Shirley VS (2008) Table of isotopes, 8th ed. CD-ROM Ver. 1.0 (1996) or on the web page http://www.nndc.bnl.gov/nndc/ensdf/
35. Kolotov VP, Atrashkevich VV, Dogadkin NN (1999) Czech J Phys 49:423
36. Lepy M-C (2007) Nucl Instrum Methods Phys Res A 579:284
37. Ramos-Lerate I, Barrera M, Ligero RA, Casas-Ruiz M (1997) Nucl Instrum Methods Phys Res A 395:202
38. NIST, Xcom (2008) Photon Cross Sections Database on the web page http://physics.nist.gov/PhysRefData/Xcom/