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FAST COINCIDENCE-SUMMING CORRECTION PROCEDURES FOR GAMMA SPECTROMETRIC MEASUREMENTS IN CLOSE GEOMETRIES

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Abstract. *The spectrometric analysis of a sample measured in "close geometry" can be affected by errors due to coincidence-summing effects that occur when two or more cascade photons are emitted within the resolution time of the spectrometric system. The probability that these effects occur depends primarily on the nuclide decay scheme, the sample-detector distance, and the intrinsic efficiency of the detector. The values of correction factors are obtained by using well known expressions which terms depend on decay data, Full-Energy-Peak Efficiency (FEPE) and Total Efficiency (TE) values. The experimental determination of FEPE and TE calibration curves performed by using single-γ emitter sources for each measurement geometry is a long and tedious task. Equally time-consuming is the use of a Monte Carlo (MC) simulation both for the validation of the detector model and the number of analyses to be carried out. In this work some faster and simpler procedures to evaluate true coincidence-summing correction factors are tested and the results are compared with the ones obtained through experimental FEPE and TE calibration curves as well as a MC simulation. For point sources a TE behaviour approximation (linear or a constant), the use of Total-to-Peak ratios and a Virtual Point Detector (VPD) approach are considered. For volume sources, a VPD efficiency transfer method by integration of elementary efficiencies over the whole source volume is used. The different approaches give very close results and differences are of the order of a few per cent.*

Keywords**:** *Coincidence-summing, Efficiency, Gamma-ray spectrometry, Monte Carlo, Virtual Point Detector.*

1. INTRODUCTION

Gamma-ray spectrometry with HPGe detectors is the most used technique to determine the radionuclide activities in various type of samples. For low specific activity samples, measurement geometries very close to the detector are needed to avoid significant loss of sensitivity of the system. The spectrometric analysis of a gamma-emitter sample measured in "close geometry" can be affected by errors due to coincidence-summing effects that occur when two or more cascade photons are emitted within the resolution time of the spectrometric system. The probability that these effects occur depends primarily on the nuclide decay scheme, the sample-detector distance, and the detector intrinsic efficiency. Coincidence-summing corrections of the photopeak areas are requested particularly if the gamma-ray spectrometric analysis is aimed to efficiency or activity determination.

The evaluation of correction factors is a complex task, especially for volume sources with various shape. Some equations for the calculation of the correction factors for point sources as function of Full-Energy-Peak Efficiency (FEPE) and Total efficiency (TE) were developed in previous works [1,2]. The number and values of the numerical coefficients depend on the decay scheme data of the radionuclide of interest. The needed efficiency values can be evaluated through spectrometric measurements of point single-γ emitter sources, i.e. radioactive sources that emit gamma rays of single energy. This type of sources is not always available and, furthermore, many spectrometric measurements are needed for each measurement geometry considered. All this makes the determination of FEPE and TE calibration trends a long and tedious task.

An alternative method to determine FEPE and TE calibration curves concerns use of a Monte Carlo (MC) simulation. However, detector modelling procedure may become not easy and time-consuming because of validation process. Furthermore, variation over time in detector characteristics must be considered, as for example the increase of dead layer thickness [3,4].

As suggested in [5] the computation procedures can be simplified and make faster with some suitable approximations on the determination of efficiency values, particularly TE. For this trend, it can be assumed a linear function (or also a constant) without significant variation on correction factor values in a given energy range.

A similar approach involves use of Total-to-Peak (T/P) ratios to determine the corresponding TE values, considering the invariability with measurement geometry of T/P ratio for any energy [6].

An interesting chance is to extend the concept of "Virtual Point Detector" (VPD) introduced by Notea [7] and Debertin and Helmer [8] to compute efficiency values related to a measurement geometry from a reference one. The validity of VPD concept was

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demonstrated also for off-axis point source [9], extended to absorbing media [10], disk sources [11], planar and semi-planar detectors [12], and applied to evaluate coincidence-summing correction factors [13].

The aim of this work is to verify the reliability of fast procedures to determine coincidence-summing corrections for point sources based both on TE or T/P approximation and on VPD approach.

As regards volume sources, the relations to compute coincidence summing correction factors for point sources cannot be used, as the effect of correction is related to cascade-emissions from each point of source. In this case, the VPD approach is very attractive because the point source equations can still be used for volume samples if point-source FEPE and TE values are replaced with the corresponding "*effective efficiencies*" as defined in [14]. This method is preferable or at least comparable with a MC simulation which require knowledge on the shape and composition of the detector in addition to that of the sample.

The simplified procedures lead to differences in the values of the correction factors of the order of a few percent, acceptable in comparison with the uncertainties associated to the measurement results.

2. Experimental

All the gamma-ray spectrometric measurements were performed by using a coaxial EG&G Ortec HPGe detector, mod. GEM 50195S, 60% relative efficiency and FWHM of 1.75 keV for the 1332 keV peak of ^{60}Co. The detector is housed within a complex box-shaped shielding composed from outside to inside of 9 cm thick polyethylene bricks, a 1 cm thick boric acid plates and 10 cm thick lead internally lined with 3 cm OFHC copper [15]. A valve junction box allows the flushing the measurement cavity with exhaust LN_2 in the dewar.

The electronic equipment associated with the detector consists of an ORTEC 672 amplifier and an ORTEC 919E EtherNIM multichannel Buffer connected into an Ethernet environment. For spectrometric data acquisition and analysis EG&G ORTEC Gamma Vision®-32 software was used [16]. FEPE and TE calibration parameters were determined through 4th order log-log polynomial fits of spectrometric data of point single γ-emitter sources whose activity values are adequate to "*close geometry*" measurements. The sources kit, provided by CEA, 9CH04-EGEA10, was composed of the following sources: ^{241}Am, ^{109}Cd, ^{57}Co, ^{139}Ce, ^{113}Sn, ^{137}Cs, ^{54}Mn, ^{65}Zn. In addition, to extend the energy range, was also used a source of ^{88}Y of the same type. The spectrometric measurements were carried out at various distance from the cap of the detector. The counting live times were chosen in order to reach at least 10^4 counts as photopeak area, so the counting uncertainties were less than 1 percent (1σ).

3. Coincidence-summing correction procedures

The radionuclide activity A of a sample can be determined through the relationship

$$A = \frac{n(E) \times C_s(E)}{T_c \times I(E) \times FEPE(E)} \quad (1)$$

where $n(E)$ is the photopeak area at energy E obtained in a live counting time T_c, $I(E)$ the emission intensity and $C_s(E)$ the coincidence-summing correction factor.

General equations to calculate $C_s(E)$ taking into account γ–γ and γ–X$_K$ coincidences are reported in [1,2,17,18]. The required decay data can be found in the most recent databases, e.g. [19].

For each gamma emitted by a radionuclide of interest, the correction equation was developed, the expected FEPE and TE values were determined experimentally or through MC simulation and finally the correction coefficient was calculated. This task turned out to be very onerous.

To make the procedure simpler and faster, various approximations based on the assumption that a variation on TE has a limited effect on the C_s value can be adopted. The simplifications considered in this work are listed below:

- A linear fit of TE values for energies more than 120 keV;
- calculation of TE values as the product of the FEPE ones by the pertinent T/P ratio value;
- a VPD approach for the evaluation of efficiencies in different positions starting from a reference one.

The reliability of the simplifications with reference to a given geometry is verified by comparing the obtained C_s values with the ones derived from FEPE and TE experimental measurements as well as MC simulations.

3.1. Experimental FEPE and TE calibration curves

The equation reported in [1,2] can be directly used for point sources by replacing FEPE and TE values assessed for the considered measurement geometry. To this aim, a series of spectrometric measurements of single-γ emitter sources were carried out to determine both FEPE and TE calibration trends for each measurement geometry. As an example, in Fig. 1 the trends obtained from spectrometric measurements of single-γ emitter point sources placed on the Ortec GEM 50195S detector cap are showed. The values at energies 122 and 136 keV of ^{57}Co can be considered both free from coincidences. Indeed, 122 keV photons emissions are in cascade with 14.4 keV photons but the last have a very low detection probability in a p-type HPGe detector due to the thickness of the cap. As far as TE is concerned, an intensity weighted average energy equal to 125 keV was assumed.

As regards TE at 1836 keV of ^{88}Y, the procedure suggested in [20] was followed by subtracting from the total spectrum counting the contribution of the 898 keV photons, evaluated by the calibration curve.

In this way, the efficiency values for the 1 cm geometry and the C_s values to compare with the ones computed through the approximations were obtained.

3.2. Monte Carlo simulation

MC simulation is an increasingly used technique for the evaluation of FEPE or TE efficiency curves due to the continuous increase in the computing power of modern workstations. However, detector model validation is a difficult task and several parameters must be considered. To overcome these difficulties, in a recent work [21] was suggested the use of an equivalent detector model having a fictitious dead-layer thickness to represent the effects of all detector characteristics variations. This allowed to perform a reliable MC simulation of a measurement geometry with PENELOPE 2018 code [22] obtaining FEPE and TE values necessary for Cs factors calculation to compare with the results of the simplified procedures.

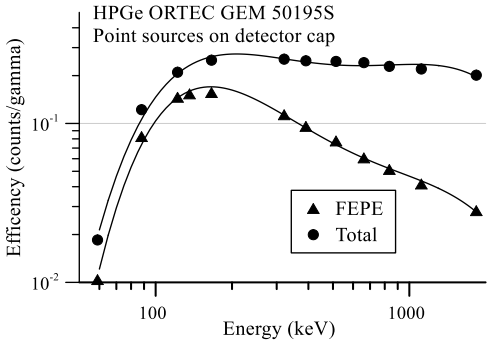

Figure 1. FEPE and Total efficiency calibration trends obtained through single-γ emitter measurements. Fitting function are log-log 4th order polynomials.

3.3. Total Efficiency approximation

The first proposed simplification is based on the observation that the total efficiency curve has a small slope (see Figure 1) in the energy range from approximately 120 keV up to 1836 keV. In this energy range, TE curve can be represented by a linear trend or, if the case, with a constant.

For lower energies, the equivalence of TE values with the FEPE ones can be assumed due to the high probability that an interaction of the photons in this energy range is photoelectric.

3.4. T/P ratio geometrical invariability

A second simplifying approach assumes a geometrical invariability of T/P ratio, as demonstrated in [6]. For single-γ emitter sources, T/P value can be computed as the ratio of the counting of the entire gamma-ray spectrum (extrapolated to zero energy) and the photopeak area value. The T/P values thus obtained can be fitted with a linear relationship to obtain a T/P(E) curve. In this way, for any energy value, the total efficiency T(E) is evaluated as the product of the FEPE(E) by the T/P(E) ratio derived from the calibration curve. In Fig. 2 experimental values of T/P ratio as a function of energy for various measurement geometries are reported. A linear fit is also highlighted as a calibration curve for T/P(E).

3.5. Virtual Point Detector approach

A third choice is to assume the detector as a "Virtual Point" at a distance h_o(E) from the detector cap.

In this way, all the physical quantities can be considered to vary with the square of distance between the point source and the VPD.

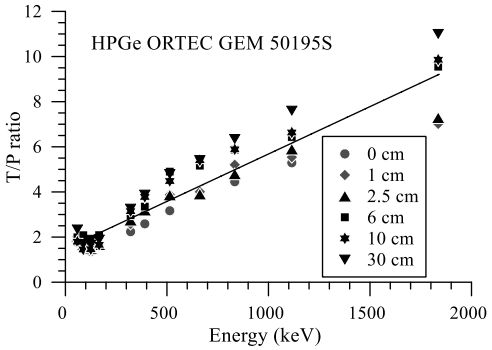

Figure 2. Behaviour of T/P ratio as a function of energy for some measurements geometries. A linear fit is representative of the T/P ratio variation vs energy.

With reference to the scheme of Figure 3, the efficiency value can be derived by using VPD method with the simple relation [9]

$$\varepsilon(E,r,h) = \varepsilon(E,0,0)\frac{(h_0(E))^2}{r^2+[h+(h_0(E)]^2} \qquad (2)$$

where $\varepsilon(E,r,h)$ is FEPE or TE value related to a source placed at r and h coordinates, $\varepsilon(E,0,0)$ the one measured on a reference position (in this case at the center of detector cap), $h_o(E)$ is the VPD distance from the detector cap which value depends on gamma energy.

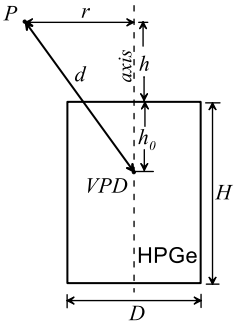

Figure 3. Scheme of a point source measurement geometry with a VPD approach.

For each energy, the value of $h_o(E)$ can be derived both from measurements along the detector axis or in radial direction. The following relations can be adopted for on axis and radial measurements [11,12]

$$h_0(E) = \frac{h}{\sqrt{\frac{\varepsilon(E,0,0)}{\varepsilon(E,0,h)}}-1} = \frac{h}{\sqrt{\frac{C(E,0,0)}{C(E,0,h)}}-1} = \frac{h}{R_X(h)} \qquad (3)$$

$$h_0(E) = \frac{r}{\sqrt{\frac{\varepsilon(E,0,0)}{\varepsilon(E,r,0)}}-1} = \frac{r}{\sqrt{\frac{C(E,0,0)}{C(E,r,0)}}-1} = \frac{r}{R_H(r)} \qquad (4)$$

Although, for each energy, at least two measurements are needed, it is useful to plot $R_H(r)$ or $R_X(h)$ trends as a function of distance r or h. The experimental points can be fitted with a linear relationship. The inverse of the slope represents the value of $h_0(E)$. The goodness of the last assessment affects directly the accuracy of determination of efficiency values. Figure 4 shows R_H and R_X trends for the 662 keV gamma emission of ^{137}Cs. The values of h_o evaluated through FEPE and TE counts are very close, and an average value can be assumed as the best value. In Figure 5 are showed the $h_0(E)$ data obtained for GEM50195S detector, fitted with a logarithmic interpolation curve in the range 59-1836 keV.

As a validation of the efficiency computation process, the FEPE and TE values evaluated with a VPD approach for a measurement geometry were compared with the corresponding experimental ones. For example, in Figure 6 a) is showed the trend of the experimental 662 keV radial FEPE and TE values compared with the ones calculated by VPD approximation. The percentage differences between the experimental and calculated efficiencies, showed in Figure 6 b), are at most 10% for FEPE and 11% for TE. Figure 6 c) shows the variation of T/P values for radial measurements of a ^{137}Cs source.

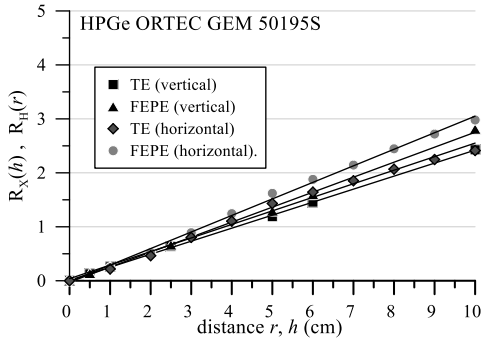

Figure 4. Linear trends of R_X and R_H as a function of distances r and h, respectively. Source: ^{137}Cs. The slope of the linear fit represents the h_o value.

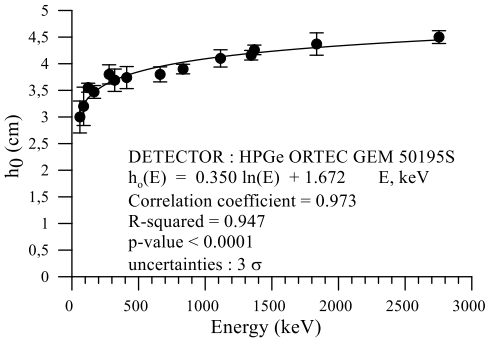

Figure 5. Behaviour of h_o values as a function of energy.

The same VDP approach can be used if an absorbing media is placed between a point source and the detector, introducing an attenuation coefficient μ(E) variable with the energy E and related to media composition [10].

The relation (2) is changed as

$$\varepsilon(E,r,h) = \varepsilon(E,0,0)\,\frac{(h_0\,(E))^2}{r^2+[h+(h_0\,(E)]^2}\; e^{-\mu(E)\,d_s} \quad (5)$$

where d_s is the photon path inside the absorbing media.

For volume sources, the same numerical equations [1,2] can be still employed replacing FEPE and TE values with the corresponding "*effective FEPE*" and "*effective TE*", defined as [14]

$$\varepsilon_P^{eff}(E_m,E_n,V) = \frac{\int \varepsilon_P(E_m,\vec{d})\,\varepsilon_P(E_n,\vec{d})\,dV}{\int \varepsilon_P(E_m+E_n,\vec{d})} \quad (6)$$

$$\varepsilon_T^{eff}(E_t,E_s,V) = \frac{\int \varepsilon_P(E_s,\vec{d})\,\varepsilon_T(E_t,\vec{d})\,dV}{\int \varepsilon_P(E_s,\vec{d}\,)} \quad (7)$$

where subscripts m, n, s and t identify the cascade gamma transitions, $\varepsilon_T(E,\vec{d})$ and $\varepsilon_P(E,\vec{d})$ are the elementary efficiencies associated to an elementary volume dV (considered as point source) whose gamma emissions are attenuated in the path length travelled by the photons in the sample. This approach is generally not valid because it was only demonstrated for the simple case with one gamma emission candidate for a coincidence besides the gamma emission under consideration. In general, efficiency values cannot be volume-averaged using the peak efficiency as a weight function. Different approaches were suggested, as for example the introduction of a "third curve" [23,24]. However, from the simplification point of view, the use of "*effective efficiencies*" seems a good approximation for the computation of C_s factors for extended sources. A comparison with the C_s factors determined directly with MC simulation, considered the most meaningful way of determining correction factors for extended sources, allows to validate this procedure.

In this work, the VDP approach was used to derive elementary efficiency values, taking into account self-absorption within the sample related to matrix composition and dimensions (as eq. (5)).

With reference to Figure 7, the sample matrix can be regarded as consisting of N elementary volumes, each with ΔV volume magnitude, and efficiency constant within. Then, the relations (6,7) can easily be transcribed into

$$\varepsilon_P^{eff}(E_m,E_n,V) = \frac{\sum_{k=1}^{N}\varepsilon_P(E_m,\vec{d_k})\,\varepsilon_P(E_n,\vec{d_k})\,\Delta V_k}{\sum_{k=1}^{N}\varepsilon_P(E_m+E_n,\vec{d_k})\Delta V_k} \quad (8)$$

$$\varepsilon_T^{eff}(E_t,E_s,V) = \frac{\sum_{k=1}^{N}\varepsilon_P(E_s,\vec{d_k})\,\varepsilon_T(E_t,\vec{d_k})\,\Delta V_k}{\sum_{k=1}^{N}\varepsilon_P(E_s,\vec{d_k})\Delta V_k} \quad (9)$$

4. Results and Discussion

In Table 1 are compared C_s factors computed for some gamma emissions by using FEPE and TE experimental values and results obtained with the above-described simplified methods with reference to 1 cm on-axis measurement geometry. Values computed adopting a MC simulation with a suitable equivalent model [21] are also reported as a comparison with the previous data.

The coefficients are rounded considering measurement errors. It can be noted that the values differ of about 1 percent. The adoption of the simplified methods above examined is justified by the fact that 10 percent variations in efficiencies lead to differences in the C_s values of the order of a few percent. This is confirmed by an analysis of the variations caused by artificial-created inaccuracies in the values of the total efficiencies which results are summarized in Table 2.

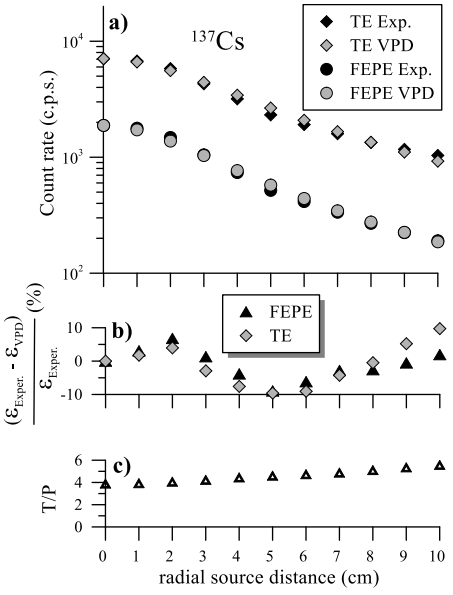

Figure 6. a) Comparison of experimental FEPE and TE values with VPD calculated ones. b) Percentage differences among the different efficiency evaluations. c) T/P ratio variation along radial axis. Detector: Ortec GEM 50195S. Geometry: 1 cm.

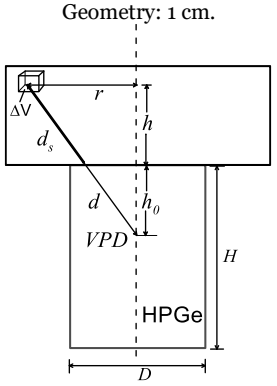

Figure 7. Scheme for the computation of effective efficiencies for a volume source.

The differences are not significant for the purposes of this work because changes of 15 percent in TE efficiency values involve errors in the determination of C_s values at most 3 percent.

As a volume source application example, a packet-sample geometry, consisting of a cellulose filter used for air particulate sampling and reduced to about 0.8cm × 6cm × 6cm package by means of a 15-ton press has been analyzed. The final obtained density results about 0.8 g cm^{-3}. The mass attenuation coefficients were determined by measuring the above mentioned 9CH04-EGEA10 kit sources at a distance of 1 cm with and without a packet-sample.

To compute effective efficiencies, bearing in mind the symmetry of sample measurement geometry, a quarter of the entire volume, composed by 144 elementary volumes each with 0.5 cm × 0.5 cm × 0.2 cm dimensions, was considered. In Figure 8 is represented the sample measurement geometry and in Table 3 are reported the C_s values computed for the same gamma emissions above considered. Table 3 shows the coefficients calculated with the VPD approach, the only method applied to volume sources, in comparison with Monte Carlo determinations. The differences are of the order of a few percent.

Table 1. A comparison between C_s factors computed with experimental FEPE and TE and the ones computed with linear TE approximations, or TE evaluated through T/P ratio, VPD approach and MC simulation. Measurement geometry: 1 cm. Detector: Ortec GEM 50195S.

Nuclide	Energy (keV)	I (%)	FEPE, TE Exp.	TE Approx.	T/P ratio	VPD Approx.	MC Simul.
^{60}Co	1173.2	99.85	1.17	1.16	1.18	1.18	1.18
	1332.5	99.98	1.17	1.16	1.18	1.18	1.18
^{152}Eu	121.8	28.41	1.18	1.18	1.18	1.18	1.19
	244.7	7.55	1.23	1.24	1.23	1.22	1.24
	344.3	26.59	1.12	1.12	1.11	1.12	1.13
	778.9	12.97	1.18	1.17	1.18	1.17	1.19
	964.1	14.50	1.09	1.10	1.10	1.08	1.10
	1112.1	13.41	1.06	1.06	1.06	1.07	1.07
	1408.0	20.85	1.07	1.08	1.08	1.07	1.08

Table 2. Analysis of variation of C_s values with an artificial increase of TE values.

Nuclide	Energy (keV)	I (%)	TE Experim.	+5%	+10%	+15%
^{60}Co	1173.2	99.85	1.17	1.18	1.19	1.20
	1332.5	99.98	1.17	1.18	1.19	1.20
^{152}Eu	121.8	28.41	1.18	1.19	1.20	1.22
	244.7	7.55	1.23	1.24	1.26	1.27
	344.3	26.59	1.12	1.13	1.13	1.14
	778.9	12.97	1.18	1.19	1.20	1.22
	964.1	14.50	1.09	1.10	1.11	1.12
	1112.1	13.41	1.06	1.06	1.07	1.07
	1408.0	20.85	1.07	1.08	1.08	1.09

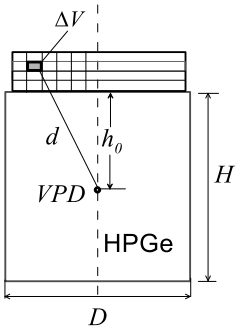

Figure 8. Application of VPD approach to determine effective efficiencies for a packet-sample geometry.

5. Conclusions

The simplified procedures here described makes it possible to quickly determine the coincidence-summing correction factors without using expensive experimental measurement activities, Monte Carlo simulations or other complex computation methods. However, it is needed to carry out some single-γ emitter source efficiency measurements and/or accurate assessment of h_0 values. The fast procedures are able to compute C_s values without significant differences with respect the precise formalism. The technique of assimilating the detector at a virtual point allows us to determine the correction coefficients with a good reliability. Possible inaccuracies in the assessment of efficiencies do not significantly affect the determination of correction values, as 10 percent variations on efficiencies change only a few percent the correction factors values. Furthermore, it was verified that VDP approximation is still valid for TE and it was confirmed the invariability of T/P ratio with the source position for energies more than 100 keV and distances not far above the detector size.

Table 3. Coincidence-summing correction factors for a packet-sample geometry. Detector: Ortec GEM50195S.

Nuclide	Energy (keV)	I (%)	VPD Approx.	MC
^{60}Co	1173.2	99.85	1,18	1,17
	1332.5	99.98	1,18	1,17
^{152}Eu	121.8	28.41	1,20	1,18
	244.7	7.55	1,24	1,22
	344.3	26.59	1,13	1,12
	778.9	12.97	1,16	1,15
	964.1	14.50	1,08	1,09
	1112.1	13.41	1,04	1,04
	1408.0	20.85	1,06	1,06

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