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Accurate computation of coincidence summing corrections in low level gamma-ray spectrometry

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Abstract

The GESPECOR (Germanium Spectrometry Correction factors) software, previously developed for computing the self-attenuation and coincidence summing corrections, was applied to the computation of the coincidence summing correction factors for a well-type and two coaxial HPGe detectors. Cylindrical samples as well as Marinelli beaker samples were considered. The computed values are in good agreement with carefully measured values. A detailed study of the uncertainties assigned to the results was carried out. The analysis shows that the procedures used in GESPECOR are reliable and provide results with a well defined accuracy. © 2000 Elsevier Science Ltd. All rights reserved.

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1. Introduction

The problem of coincidence summing corrections in gamma-ray spectrometry has been the subject of important papers for almost 30 years (Debertin and Helmer, 1988). However, due to the availability and more extensive use of high efficiency detectors (including well-type) and due to the more stringent quality assurance criteria that are now required, the interest in this field has been revived in recent years.

In this paper we present our recent work on the application of the Monte Carlo method to the computation of coincidence summing corrections. The previously developed algorithms (Sima, 1996; Sima and Arnold, 1996; Sima and Dovlete, 1997) were extended and improved. A special subroutine and the KORDATEN data file (Debertin and Schötzig, 1979) were added for an automatic preparation of the complex decay data required for the calculation of coincidence summing. The computational routines, complemented by user friendly interfaces, were integrated in a dedicated software, called GESPECOR (Germanium Spectrometry Correction factors). GESPECOR is currently running on IBM-compatible PC computers.

2. Theory

A detailed presentation of the subject may be found in the book by Debertin and Helmer (1988). In what follows, only essential features related to our approach will be outlined.

Consider a source of volume V, containing the

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nuclide X with a total activity A, uniformly distributed within V. It is assumed that the de-excitation following the nuclear decay takes place through a series of cascades in which several photons γ_i (energy E_i) are emitted in a time interval shorter than the resolving time of the spectrometer. Let p_i, p_{ij}, $p_{ij,k}$... be the emission probability of the γ_i photon, of the pair (γ_i, γ_j), of the triplet $(\gamma_i, \gamma_j, \gamma_k)$ ... in one decay. In the absence of coincidence summing effects, the count rate N_i in the peak at the energy E_i would be:

$$N_i = \frac{A}{V} p_i \int_V \varepsilon(E_i \vec{r})\, \mathrm{d}V = A p_i \varepsilon(E_i; V) \tag{1}$$

Here $\varepsilon(E_i; V)$ is the full energy peak (FEP) efficiency for the extended source, and $\varepsilon(E_i, \vec{r})$ is the FEP efficiency for an elementary source located at $\vec{r}$

The rate N^m of *summing out* events (coincidence losses) from the γ_i peak due to coincidences with γ_j is:

$$N^m_{i;j} = \frac{A}{V} p_{i,j} \int_V \varepsilon(E_i, \vec{r}) \varepsilon_T(E_j, \vec{r})\, \mathrm{d}V \tag{2}$$

where $\varepsilon_T(E_j, \vec{r})$ is the total detection efficiency for an elementary source of photons with energy E_j located at $\vec{r}$. The total detection efficiency for the complete source $\varepsilon_T(E_j; V)$ is given by:

$$\varepsilon_T(E_j; V) = \frac{1}{V} \int_V \varepsilon_T(E_j, \vec{r})\, \mathrm{d}V \tag{3}$$

Terms involving the effect of coincidences between two photons (eg. of the type $N^m_{i;j}$) are called first order correction terms. The losses from the peak at energy E_i should be evaluated by summing the terms $N^m_{i;j}$, $N^m_{i;k}$, ... for all the photons γ_j, γ_k, ... which may be emitted simultaneously with the γ_i photon. The second order terms, e.g. $N^m_{i;j,k}$ resulting from the simultaneous detection of three photons, are counted both in $N^m_{i;j}$ and in $N^m_{i;k}$ and therefore should be deducted from the first order terms

$$N^m_{i;j,k} = \frac{A}{V} p_{i,j,k} \int_V \varepsilon(E_i, \vec{r}) \cdot \varepsilon_T(E_j, \vec{r}) \cdot \varepsilon_T(E_k, \vec{r})\, \mathrm{d}V \tag{4}$$

The total coincidence losses from the peak at energy E_i should be computed by evaluating to all higher orders the appropriate terms produced by any radiation which may be emitted together with the γ_i photon.

The *coincidence summing in* effect is produced when two (or more than two) photons γ_p, γ_q are totally absorbed in the detector simultaneously. In the first order, the additional count rate in the peak at the energy $E_p + E_q$ due to this type of summing will be:

$$N_{p+q} = \frac{A}{V} p_{p,q} \int_V \varepsilon(E_p, \vec{r}) \cdot \varepsilon(E_q, \vec{r})\, \mathrm{d}V \tag{5}$$

This count rate should be corrected further due to coincidence losses of the type $N^m_{p+q;j}$

Finally, the actual count rate in the peak at energy E_i in the presence of coincidence summing corrections is given by:

$$N^c_i = \left(N_i - N^m_{i;j} - N^m_{i;k} - \cdots + N^m_{i;j,k} + \cdots\right) + \left(N_{p+q} - N^m_{p+q;j} - \cdots\right) + \cdots \tag{6}$$

The coincidence summing correction factor $F_c(E_i)$ for the peak at the energy E_i is defined by the ratio:

$$F_c(E_i) = \frac{N^c_i}{N_i} = \frac{N^c_i}{A \cdot p_i \cdot \varepsilon(E_i; V)} \tag{7}$$

In the case of pure sum peaks $p_i = 0$ and $N_i = 0$ and the correction factor is defined by:

$$F_c(E_s) = \frac{N^c(E_s)}{A \cdot \varepsilon(E_s; V)} \tag{8}$$

where E_s is the energy of the sum peak. Eq. (7) can still be applied, with the convention that p_i is set equal to 1.

For the evaluation of F_c the emission probabilities of various sets of photons should be combined with the appropriate integrals of products of efficiencies.

3. Calculation of efficiencies

In GESPECOR the Monte Carlo method is applied for the computation of the efficiencies $\varepsilon(E, \vec{r})$ and $\varepsilon_T(E, \vec{r})$ and for the evaluation of the integrals. Powerful variance reduction techniques are used to increase

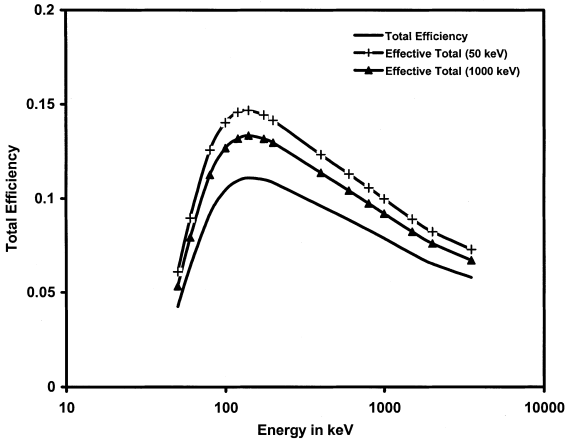

Fig. 1. Total and effective total efficiency for a water source contained in a 1 l Marinelli beaker, measured with a 50% relative efficiency HPGe detector. In the case of the effective total efficiency the energy of the peak is given in parentheses.

the efficiency of the computation. Secondary radiations, such as bremsstrahlung photons or X-rays excited in the shield or in the matrix of the source (Arnold and Sima, 2000) are included in the computation of $\varepsilon_T(E, \vec{r})$. The detailed composition of the materials is taken into account. The assumption of cylindrical symmetry of the experimental arrangement is applied; however, point sources placed off the symmetry axis are allowed.

In Fig. 1 the effective total efficiency $\varepsilon_T^{\text{eff}}(E; E_i)$ which is required for the computation of coincidence losses from the peak at the energy E_i due to coincidences with a photon with energy E is represented together with $\varepsilon_T(E; V)$ for the volume source. The effective total efficiency is defined by:

$$\varepsilon_T^{\text{eff}}(E, E_i) = \frac{\int_V \varepsilon(E_i, \vec{r})\varepsilon_T(E, \vec{r})\, dV}{\int_V \varepsilon(E_i, \vec{r})\, dV} \quad (9)$$

A Marinelli beaker with a volume of 1 l, filled with water, and an HPGe detector with a 50% efficiency was considered. The effective total efficiency depends on the value of E_i and it is always higher than $\varepsilon_T(E; V)$. This is due to the fact that the points closer to the detector, for which $\varepsilon_T(E, \vec{r})$ is higher, are more heavily weighted in Eq. (2) [by the presence of $\varepsilon(E_i, \vec{r})$] than in Eq. (3). The additional weight results both from solid angle variation and from self-attenuation effects.

4. Calculation of joint emission probabilities

For complex decay schemes the computation of the emission probabilities p_{ij}, $p_{ij,k}$, ... is very difficult. In GESPECOR, a special subroutine called INPEN is included for the computation of these probabilities for an arbitrary decay scheme. The decay scheme data for approx. 100 nuclides are read from the KORDATEN data file (Debertin and Schötzig, 1979). They are: nuclide name, decay type, mean energy of KX rays, fluorescence yield ω, level feeding probability $b(l)$ by parent decay, K-capture probability $P_K(l)$ (in EC decay), and for each gamma transition, the initial (i) and final (j) levels, the energy, the photon emission probability $w(i, j)$, total $\alpha(i, j)$ and K conversion coefficients $\alpha_K(i, j)$.

INPEN is started by giving the name of the nuclide, the energy E of the peak and an energy search interval ΔE. After selecting in the KORDATEN file the transition corresponding to the energy E all possible radiations (de-excitation photons, X-rays from EC decay or IC transitions, annihilation photons from β^+ decay) which might be emitted together with the energy E are listed and the appropriate probabilities p_i, p_{ij}, $p_{ij,k}$, ... are computed. Then all possible sum peak combinations contributing to the same peak are selected and the associated probabilities are computed and stored for input in the Monte Carlo programs.

5. Results

Very accurate experimental values of coincidence summing correction factors have been measured at PTB for several detectors and extended sources. These values were obtained by applying Eq. (7) in the measurement of sources with known activity.

The values of F_c corresponding to the measurements carried out with three detectors have also been computed by GESPECOR and compared with the experimental data.

The highest coincidence summing effects are present in the measurements carried out with a 350 cm^3 well type HPGe detector. For example, in the case of the measurement of a cylindrical water source with a radius of 0.4 cm and a height of 3 cm, the count rate in the 276 keV peak of ^{133}Ba is reduced by a factor of 20; on the other extreme, the count rate in the 401 keV peak of ^{75}Se is increased by a factor of about 8. Second order and higher order terms have an essential contribution to the total effect. This can be easily seen in Fig. 2, displaying the spectrum of a ^{22}Na source. Without coincidence summing effects only the peaks at 1275 and 511 keV would be present, with a count rate approx. 6 and 7.5 times higher than measured. In a computation based only on first order corrections, the peak at 2297 keV (2297 = 1275 + 511 + 511) would have been absent and the number of counts in the 1275 keV peak would have been negative!

The results computed by GESPECOR for nuclides

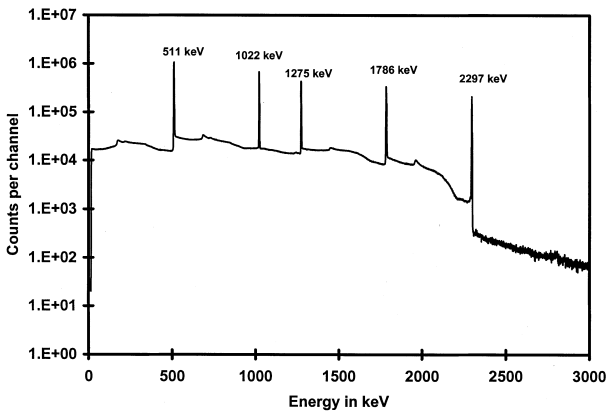

Fig. 2. The spectrum of a ^{22}Na source measured in a well-type detector geometry.

like ^{60}Co and ^{88}Y are in agreement within 1.5% with the measured values. For nuclides with more complex decay schemes, such as ^{125}Sb, the agreement is better than 5% (for the peak at 636 keV, besides the correction for the photon with energy 635.9 keV, resulting from the transition between the levels with energy 671.41 and 35.49 keV, two other contributions should be added: (a) a pure sum peak with energy 636.03 keV, resulting from the cascade transition from the 636.03 keV level to the 35.49 keV level and from this level to the ground state; (b) a sum peak between the 606.64 keV photon and the KX rays. These contributions are properly suggested by INPEN). For even more complex schemes, with important contributions from higher order coincidence summing terms, the discrepancy may reach 10–12% for some peaks, which have very low values of F_c. In Fig. 3, several measured and computed values of the apparent efficiency for the water source with a filling height of 3 cm are presented. The apparent efficiency is defined as

$$\varepsilon^{\overline{\text{app}}}(E_i) = \frac{N_i}{p_i A} \tag{10}$$

From Eq. (7) it is seen that the apparent efficiency divided by the value of $F_c(E_i)$ should be equal to the efficiency from the calibration curve. The corrected apparent efficiency, computed in this way, is also displayed in Fig. 3, together with the efficiency curve obtained by a measurement with mono-gamma nuclides.

The GESPECOR computations also reproduce the dependence of F_c values on the filling height (Sima and Arnold, 1996) and the effects of coincidence summing with matrix X-rays (Arnold and Sima, 2000).

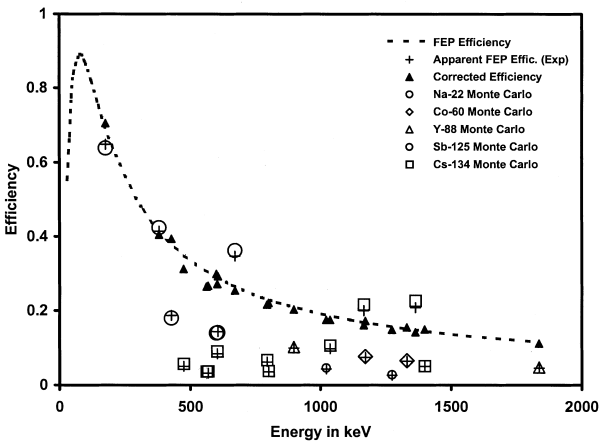

Fig. 3. The experimental apparent FEB efficiency (+) compared with the compound apparent efficiency (open symbols) in the case of the well type detector. Also the corrected efficiency (full triangles) based on the computed values of F_c compared with the experimental efficiency obtained from measurements with mono-gamma nuclides (- - -).

The comparison between the computed and the experimental values was also carried out for a p type coaxial HPGe detector with 50% relative efficiency. In the case of the measurements of a 25 cm^3 water source, the coincidence summing correction factors for the measured nuclides range from about 0.7 to 2.2. The discrepancy between the computations and the measurement is lower than in the case of the well type detector measurements. In the case of a 1 l Marinelli beaker, the F_c values are between 0.7 and 1.6. The highest discrepancy between the computed and the measured values is about 5%.

Similar results have been obtained for other measuring geometries and for an n type HPGe detector with the relative efficiency equal to 25%.

6. Evaluation of uncertainties

The following sources of uncertainty in the computed values of F_c have been taken into account: detector data; measurement geometry; sample composition and density; nuclear decay scheme data. In the case of decay scheme data, the uncertainty is known from the literature. In other cases the uncertainty is not so well defined and reasonable values were established. For example, the uncertainty in the detector parameters was obtained by comparing the computed values of ε with the measured values and taking into account the sensitivity of ε to changes in the parameters; in the case of the well detector, also selected values of F_c were included in the comparison.

Using the uncertainty of different input parameters and the computed sensitivity of F_c to the changes in the values of the parameters, the uncertainty assigned to F_c was evaluated.

6.1. Nuclear decay scheme data

For complex decay schemes the uncertainty of decay data is propagated in a complicated way to the values of F_c, through the probabilities p_i, p_{ij}, $p_{ij,k}$, ... The primary decay scheme data which are used in INPEN for the evaluation of these probabilities are: $b(l)$, $p(i,j)$, $\alpha(i,j)$, $\alpha_K(i,j)$, $P_K(l)$ and ω. The uncertainty of F_c associated with these data was evaluated as follows. First, a set of primary decay data was obtained, by sampling for each parameter a random value from a normal distribution with the mean equal to the recommended value, and the standard deviation σ equal to the uncertainty, read from the Nucléide software (Bé et al., 1996). Each parameter was independently sampled, neglecting the correlation between different parameters, because the covariance matrix was not

Table 1
Partial contributions and the final relative uncertainty (in %) assigned to F_c

Experimental set-up	Contribution	563 keV	1401 keV	276 keV
50% coaxial detector (Source: 25 cm^3; Matrix: water)				
	Detector data	1.23	2.90	1.02
	Geometry data	0.27	1.20	0.15
	Matrix data	0.003	0.01	0.002
	Nuclear data	0.05	0.10	0.50
	Computation	1.00	1.00	1.00
	Total	1.61	3.30	1.52
50% coaxial detector (Source: Marinelli 1 l; Matrix: water)				
	Detector data	0.92	2.90	0.74
	Geometry data	0.05	0.05	0.01
	Matrix data	0.007	0.02	0.005
	Nuclear data	0.03	0.10	0.30
	Computation	1.00	1.00	1.00
	Total	1.36	3.08	1.28
Well-type detector (Source: 3 cm height; Matrix: water)				
	Detector data	1.50	0.63	0.92
	Geometry data	0.006	0.003	0.01
	Matrix data	0.001	0.001	0.001
	Nuclear data	0.50	0.20	12.80
	Computation	1.00	1.00	1.00
	Total	1.87	1.20	12.90

available. However, part of this correlation was introduced later by a re-normalisation of the values which should obey constraints (e.g. the sum of $b(l)$ over l, the sum of de-excitation probabilities of each level). Then the corresponding value of F_c was computed. The procedure was repeated many times and the spread of the values of F_c was evaluated. This spread was entirely due to the spread in the decay scheme data, as a correlated sampling technique was used for the simulation of the radiation transport.

The uncertainties are higher in the case of the well type detector, both because the coincidence summing effects are bigger and because the higher order terms (which are more sensitive to the uncertainties in the decay scheme data) are more important. For example, in the case of the 276 keV peak of ^{133}Ba, the uncertainty is 12.8%. The value of F_c is around 0.05 in this case.

6.2. Final uncertainty

The final uncertainty of each computed value of the coincidence summing correction factor F_c was obtained by combining the above-mentioned sources of uncertainty. Additionally, an estimated contribution of 1% resulting from the approximations applied in the theoretical computations was included. In Table 1, the final uncertainty is presented for three representative cases: a pure sum peak (1401 keV), a peak with important summing out effects from coincidences with medium energy photons (563 keV) and a peak with important coincidences with low energy photons (276 keV). The estimated uncertainty is close to the discrepancy between the measured and the computed values of F_c.

7. Conclusions

In this work the GESPECOR software was applied for the computation of the coincidence summing correction factors for a well-type and two coaxial HPGe detectors. A detailed analysis of the uncertainty of the computed values of F_c was carried out. The comparison with the experimental data proves that the computation of F_c is basically correct and that the procedure for the evaluation of uncertainties incorporates the most important contributions.

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References

Arnold, D., Sima, O., 2000. Coincidence-summing in gamma-ray spectrometry by excitation of matrix X-rays. Appl. Radiat. Isot. 52, 725–732.

Debertin, K., Schötzig, U., 1979. Coincidence summing corrections in Ge(Li)-spectrometry at low source-to-detector distances. Nucl. Instrum. and Meth. 158, 471–477.

Debertin, K., Helmer, R.G., 1988. Gamma- and X-ray Spectrometry with Semiconductor Detectors. North-Holland, Amsterdam.

Bé, M.M., Duchemin, B., Lamé, J., 1996. An interactive database for decay data. Nucl. Instrum. and Meth. A369, 523–526.

Sima, O., 1996. Applications of Monte Carlo calculations to gamma-spectrometric measurements of environmental samples. Appl. Radiat. Isot. 47, 919–923.

Sima, O., Arnold, D., 1996. Self-attenuation and coincidence-summing corrections calculated by Monte Carlo simulations for gamma-spectrometric measurements with well-type germanium detector. Appl. Radiat. Isot. 47, 889–893.

Sima, O., Dovlete, C., 1997. Matrix effects in the activity measurement of environmental samples: implementation of specific corrections in a gamma-ray analysis program. Appl. Radiat. Isot. 48, 59–69.