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Section A

A new summing-correction method for gamma-efficiency calibration with multi-gamma-ray radionuclides

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Abstract

Coincidence-summing effects play an important role in HPGe spectrometry at low source-to-detector distances (usual arrangements when environmental samples have to be measured). Although these corrections are not important for environmental samples (less than 5%), they can be significant in the efficiency calibration with multi-gamma-ray radioisotopes as they have to be measured in the same geometry. In this paper we propose a new method for determining summing corrections which does not require other monoenergetic radioisotopes. Thus, a HPGe-detector-efficiency calibration can be performed with radionuclides emitting gamma rays in cascade, such as ^{152}Eu or ^{226}Ra. The method has been successfully validated.

Keywords: Coincidence-summing effects; Gamma spectrometry; Efficiency calibration; Radioisotopes; Environmental samples

1. Introduction

In gamma-ray spectrometry a good knowledge of the full-energy-peak efficiency is required in order to determine the emission rates of gamma rays accurately. A possible problem in the efficiency calibration or in the use of the gamma-ray spectrometer arises from the simultaneous detection of other radiations occurring in cascade with the gamma-ray being measured. Of course, no correction needs to be applied if a sample is measured relative to a standard of the same radionuclide, but this is not always possible.

In employing any source that emits more than one radiation in coincidence, some care must be taken to ensure that measured peak intensities are not affected by coincidence-summing effects. This problem is particularly serious for those sources that involve many cascade gamma rays, such as ^{226}Ra or ^{152}Eu.

Since summing-effects depend on the square of the detector solid angle [1] whereas the simple peaks vary linearly, the relative effect of summing can be reduced by reducing the solid angle. However, when we measure low-level-counting systems, as environmental samples, it is necessary to reduce the counting time by increasing the detector efficiency (high-source volumes and short detector-source distances). As a consequence, the coincidence-summing corrections can reach values of about 5% in such environmental samples for normal size detectors [2] and, what is more significant, coincidence effects can also appear in the efficiency measurements, since they must be carried out in the same measuring conditions as the activity determinations.

A way to avoid summing corrections is to use monoenergetic sources, but some other errors may arise, e.g. due to sample preparation. Besides, this process is too complex and time consuming. Another possibility is to use a mixed gamma-ray standard source. However, this is not always possible as some radionuclides in this source decay very fast. It is much easier to use sources like ^{152}Eu or ^{226}Ra with many gamma-ray emissions in the energy range of 100 keV to 2 MeV and half-lives long enough so that laboratories can always have them available.

In this paper we show that it is possible to use ^{152}Eu or ^{226}Ra as calibration gamma sources in high-efficiency arrangements by means of a semi-empirical method to determine summing corrections.

2. Method

2.1. History

The summing corrections can be computed by means of the formulae given by Andreev et al. [3] and rewritten by McCallum and Coote [4] or by using the tables

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of Schima [5]. For these corrections, the full-energy-peak efficiency and the absolute total efficiency of the detector are required. However, these curves are difficult to obtain experimentally as coincidence-summing interferes with their experimental determination. The problem becomes more important for extended sources for which the usual formulae [3–5] are no longer valid [6]. So, calculating coincidence-summing corrections in the measuring conditions required by environmental samples (high-source volumes and short detector-source distances) is not an easy task. In 1995 Quintana et al. [2] proposed an empirical method to determine these summing corrections. It is based on the comparison, for a sample of the same activity, of the measured areas from the same gamma-ray emissions in two different geometries: one which will be of interest (M-geometry) and another, a reference geometry, where a "point-like" source is placed at a large distance from the detector (P-geometry). So, the summing correction for an energy E_i can be expressed as

$$C_i = \frac{M_i^P}{M_i^M} C_G(E_i), \tag{1}$$

where M^P is the count rate of the full-energy peak for a geometry in which no summing correction is needed (the "point-like" source is far away from the detector) and M^M is the count rate in the measuring geometry. $C_G(E_i)$ is an energy-dependent function that can be written as a ratio between full-energy-peak efficiencies, i.e.,

$$C_G(E_i) = \frac{\varepsilon_M(E_i)}{\varepsilon_P(E_i)}. \tag{2}$$

Of course, $C_G(E_i)$ has to be determined for energies without summing-correction problems. Quintana et al. [2] calculated this function empirically for some monoenergetic sources. However, preparing a "point-like" source of the same activity as our sample in addition to monoenergetic sources in both geometries (M-and P-) for measuring $C_G(E_i)$ is tedious and complicated, especially when the M-geometry is considered for environmental samples. We have developed a method in which no additional sources are needed.

2.2. Experimental part

The measurements were perfomed with an integral preamplifier-CANBERRA GC2020-7500SL p-type coaxial HPGe detector system. The useful energy range of this detector is from 50 keV to more than 10 MeV. The peak/Compton ratio is 46 for the 1333 keV ^{60}Co photon. It has 50.5 mm diameter, 46.5 mm length and the distance window-detector front is 5 mm. The relative efficiency to a 3 in × 3 in INa(Tl) detector is 20% and resolutions at 122 and 1332 keV of 1.1 and 2 keV, respectively, are given.

The spectrometer is shielded by a 10 cm cubic geometry box of low-background lead (smelted more than 500 years ago), with an inner 1 mm Cd + 1 mm Cu layer.

The detector is connected to a standard set up: CANBERRA Model 2020 Amplifier and a PC-based 8 K multichannel analyzer.

Water as well as sea-sediment matrix samples spiked with ^{152}Eu were prepared in polyethylene cylindrical containers of 64 mm inner diameter filled to 55 mm height. The spectrometric measurements were performed in three different geometries, all axially symmetric: (1) geometry *a*, 3 mm of distance between the bottom of the cylindrical container and the window detector; (2) geometry *b*, 116 mm of distance, and (3) geometry *c*, 192 mm of distance.

2.3. Our method

To calibrate the efficiency of environmental extended sources in cylindrical geometry, at least one water matrix spiked with a radioactive source is required [7]. The easiest way to do this would be to spike it with a multi-gamma ray source whose half life is long enough so that most laboratories can supply it when it is required (for example, ^{152}Eu). However, a water- or sea-sediment matrix sample spiked with ^{152}Eu produces summing coincidences in many HPGe detectors for high-efficiency geometries. It is difficult and tedious to produce "point-like" sources with the same ^{152}Eu activity as our water matrix in order to apply Eq. (1) for correcting such effects. However, it is very easy to place our water matrix at a larger distance from the detector so that in this Reference geometry (R-geometry) no summing corrections are required. Therefore, Eq. (1) can be written as

$$C_i = \frac{M_i^R}{M_i^M} C_G(E_i), \tag{3}$$

where M^R and M^M are the count rates in the E_i-full-energy peak for the cylindrical matrix in geometries R and M, respectively, and with

$$C_G(E_i) = \frac{\varepsilon_M(E_i)}{\varepsilon_R(E_i)}. \tag{4}$$

To calculate $C_G(E_i)$ empirically it would be necessary to produce samples spiked with monoenergetic sources [2] and to place them in geometry R or M. This is heavy and time consuming. To avoid this, $C_G(E_i)$ can be computed numerically, but as it is much easier to calculate total efficiencies than full-energy-peak efficiencies [8], Eq. (4) can be written in terms of total efficiencies. This is possible as the ratio of full-energy-peak efficiency to total efficiency, $\varepsilon/\varepsilon_t$, is independent, for every energy, of both the number of counts and the geometry of the arrangement and is called "intrinsic characteristic" [9], of the detector. Therefore, Eq. (4) can be written as

$$C_G(E_i) = \frac{\varepsilon_t^M(E_i)}{\varepsilon_t^R(E_i)}. \tag{5}$$

The total efficiency can be calculated with a program based on that developed by Haase et al. [8] and programmed in FORTRAN 77. The mass-attenuation coefficients are taken from Ref. [10] for the water matrix and measured using the transmission method, [7,11], for the sediment matrix. This does not imply additional work since the transmissions are required for calculating an efficiency-calibration function which depends on the energy and the density of the matrix [7,11].

3. Results

Firstly, we have made a couple of water matrix samples in cylindrical geometry spiked with the same activity of ^{152}Eu. Such samples were placed in three different positions (geometries *a*, *b* and *c*, as described above) and were counted long enough to ensure that statistical uncertainties were less than 1%.

The functions $C_G(E_i)$ were then calculated numerically for the ratio between the geometries *a* and *c* as well as *b* and *c*. They can be seen in Figs. 1 and 2.

It is a well-known fact that for a good total efficiency calculation it is necessary to know the exact active volume of the detector since every small uncertainty in the parameters of this could lead to large differences in the efficiency. According to our calculations, a change of 10 mm in the detector diameter leads to a change of 30% in the total efficiency. However, the function $C_G(E_i)$ varies only by 2%, as numerator and denominator changes in the same way so that errors are compensated. Therefore, we can conclude that even if the active volume is not exactly that which we have considered in our calculations, the function $C_G(E_i)$ is still valid.

On the other hand, the shape of the $C_G(E_i)$ curves is coherent. Remembering that $C_G(E_i)$ is a ratio between efficiencies in close and far positions, it is normal that the efficiency ratio increases with decreasing energy as the effective solid angle subtended by the crystal is the larger, the lower the energy, and as the degree of variation with energy is the larger, the smaller the source distance [6].

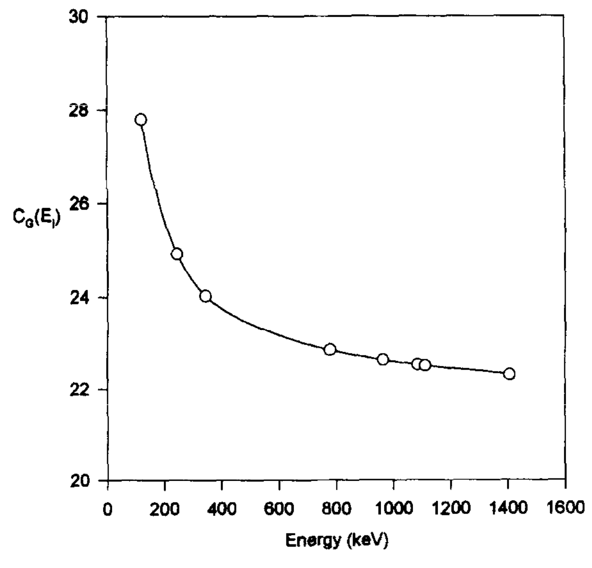

Fig. 1. Ratio of the total efficiencies (or full-energy-peak efficiencies) in geometries *a* and *c*.

Finally, for 8 emissions from ^{152}Eu, we calculate the summing corrections in the geometries *a* and *b*, by means of Eq. (3), assuming that no correction is needed in geometry *c* as the sample is far enough from the detector (Table 1). The results were the same for the two water samples.

It is easy to see from Table 1 that geometry *b* does not require summing corrections as they are less than 2%. This fact seems to confirm our method, as geometry *b* is far enough from the detector. However, a more stringent check is necessary, and for this reason, a water-matrix sample spiked with a mixed source (^{241}Am, ^{109}Cd, ^{57}Co, ^{139}Ce, ^{203}Hg, ^{113}Sn, ^{137}Cs, ^{88}Y and ^{60}Co) was elaborated and placed in geometry *a* for calculating experimentally the efficiency-calibration function. Most radioisotopes in

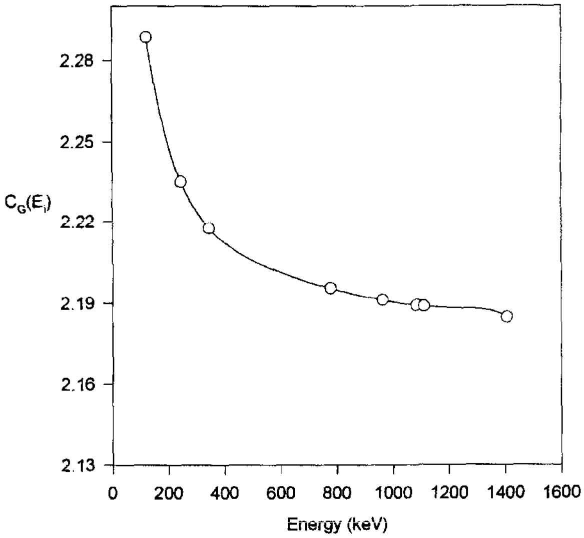

Fig. 2. Ratio of the total efficiencies (or full-energy-peak efficiencies) in geometries *b* and *c*.

Table 1
Summing corrections for a cylindrical water matrix spiked with ^{152}Eu, in the arrangements in geometries *a* and *b*

Energy (keV)	C_i^a	C_i^b
121.78	1.340	0.980
244.70	1.377	1.003
344.28	1.239	0.985
778.92	1.277	1.004
964.06	1.282	1.004
1085.84	1.190	1.020
1112.09	1.277	1.007
1408.02	1.265	1.007

Table 2
Validation of the method for a water matrix

Energy (keV)	Ef_w	C_i	$\mathrm{Ef}^\mathrm{c} = \mathrm{Ef}_\mathrm{w} \times C_\mathrm{i}$	Ef_M
121.78	0.0304(8)	1.340(7)	0.0408(13)	0.036(7)
244.7	0.0202(6)	1.377(17)	0.0278(11)	0.023(5)
344.28	0.0162(4)	1.239(9)	0.0201(7)	0.018(4)
411.13	0.0126(2)	1.38(4)	0.0174(11)	0.016(3)
443.97	0.0119(5)	1.34(4)	0.0159(11)	0.015(3)
778.92	0.0076(2)	1.277(18)	0.0098(4)	0.0095(19)
867.39	0.00634(18)	1.41(4)	0.0089(5)	0.0087(17)
964.06	0.00627(18)	1.282(18)	0.0080(3)	0.0079(16)
1085.84	0.00596(16)	1.19(2)	0.0071(3)	0.0071(14)
1112.09	0.00559(15)	1.277(19)	0.0071(3)	0.0070(14)
1212.09	0.00495(16)	1.33(9)	0.0066(6)	0.0064(13)
1408.02	0.00469(13)	1.265(16)	0.0059(2)	0.0056(11)

Note: Ef_w is the efficiency of the cylindrical water matrix spiked with ^{152}Eu in geometry *a*, without summing corrections, C_i is the summing correction calculated according to our method, and Ef^c is the corrected efficiency for the matrix spiked with ^{152}Eu. Ef^c can be compared to Ef_M, obtained with the efficiency function for monoenergetic emissions.

Table 3
Same as Table 2, but with the sediment matrix instead of water. Here, Ef_s is the efficiency of the cylindrical sediment matrix spiked with ^{152}Eu and Ef_M is the efficiency function for monoenergetic sources in geometry *a*, with a density of the sediment of 1.64 g/cm^3

Energy (keV)	Ef_s	C_i	$\mathrm{Ef}^\mathrm{c} = \mathrm{Ef}_\mathrm{s} \times C_\mathrm{i}$	Ef_M
121.78	0.0236(6)	1.320(4)	0.0312(9)	0.030(6)
344.28	0.0132(4)	1.240(4)	0.0164(5)	0.016(3)
443.97	0.0099(4)	1.30(2)	0.0129(7)	0.013(3)
778.92	0.00638(18)	1.272(8)	0.0081(3)	0.0083(17)
964.06	0.00542(15)	1.282(8)	0.0069(2)	0.0069(14)
1299.15	0.00421(13)	1.32(3)	0.0055(3)	0.0053(11)
1408.02	0.00408(11)	1.255(7)	0.00512(17)	0.0049(10)

Table 4
Same as Tables 2 and 3, but with a different sediment matrix (IAEA-SOIL6)

Energy (keV)	Ef_s	C_i	$\mathrm{Ef}^\mathrm{c} = \mathrm{Ef}_\mathrm{s} \times C_\mathrm{i}$	Ef_M
295.207	0.014(2)	1.45(2)	0.020(3)	0.022(2)
351.925	0.012(2)	1.37(2)	0.016(3)	0.019(2)

such mixed sources are monoenergetic so that no summing corrections are needed, except for ^{88}Y (898 and 1836 keV) and ^{60}Co (1173 and 1332 keV). To calculate the efficiency at 898 and 1173 keV, we extrapolated the curve obtained with the monoenergetic emissions. Then we calculated the summing corrections by comparing the extrapolated efficiencies with the experimental ones and applied the same corrections to 1836 and 1332 keV, respectively, in order to have high-energy points for the curve.

In Table 2 the efficiency of the water sample spiked with ^{152}Eu (corrected by coincidence-summing) and the efficiency function obtained with the sample spiked with the mixed source are compared. Both efficiencies are the same as considering the uncertainties, thus confirming our method.

We have also calculated the summing corrections, according to our method, for a sea-sediment matrix sample spiked with ^{152}Eu in geometry *a* and we have compared the summing-corrected efficiency with the function obtained for the same matrix spiked with a mixed source. The results, in Table 3, again validate our method.

Finally, we have checked the method with a sea-sediment sample spiked with ^{226}Ra (IAEA-SOIL6) used in intercalibration exercises. For two emissions that do not interfere with the background radiation, the summing corrections and efficiencies were calculated. Comparing the efficiency (corrected by coincidence summing) with the calibration function obtained for monoenergetic emissions (product of a correction factor and a water-matrix efficiency [7]), the results are within their respective uncertainties (Table 4).

4. Conclusions

When environmental samples are measured, the counting time can be reduced by increasing the detector efficiency (high-source volumes and short detector-source distances). As a consequence, coincidence effects can appear in the efficiency measurements since they must be carried out in the same measuring geometry.

In this paper, a new method has been developed to evaluate summing corrections which are applied to calibrate an HPGe detector when radionuclides emitting gamma rays in cascade are used. The method does not need additional monoenergetic radionuclides although it had to be validated by employing samples spiked with monoenergetic radionuclides. The summing correction can be calculated according to Eq. (3) in which M^{R} and M^{M} , independent of energy, are the count rates for a source that includes many cascading gamma rays in two different geometries: one in which we are interested (M-geometry) and another where no summing corrections are needed as the sample is far enough from the detector. The function $C_{\mathrm{G}}(E_{\mathrm{i}})$ in Eq. (3) is easily calculated with a simple FORTRAN program. This method can be very useful as it is much easier to calibrate with a source like ^{152}Eu or ^{226}Ra, with many gamma-ray emissions, than to use a number of monoenergetic sources.

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