Applied Radiation and Isotopes 67 (2009) 160–163

Contents lists available at ScienceDirect

Applied Radiation and Isotopes

journal homepage: www.elsevier.com/locate/apradiso

Application of the sum-peak method to activity standardizations of extended ^{60}Co sources

T. Vidmar [a,*], K. Kossert [b], O.J. Nähle [b], O. Ott [b]

[a] Jožef Stefan Institute, Jamova cesta 39, SI-1000 Ljubljana, Slovenia
[b] Physikalisch-Technische Bundesanstalt (PTB), Department 6.1, Bundesallee 100, D-38116 Braunschweig, Germany

ARTICLE INFO

Article history:
Received 2 May 2008
Received in revised form
23 June 2008
Accepted 8 August 2008

PACS:
29.30.Kv
07.05.Tp

Keywords:
Gamma-ray spectrometry
Activity standardization
Sum-peak method
Extended sources
Angular correlations
Monte Carlo simulations

ABSTRACT

The sum-peak method was successfully applied to the determination of the activity of extended ^{60}Co sources measured on a HPGe detector. Monte Carlo simulations were used to account for the effects of the spatial variation of the efficiency across the sample volume and for the angular correlations between the emitted gamma rays. The determined activities agree with the reference values within a range of 1.0%.

1. Introduction

The sum-peak method for activity standardization by means of a single NaI or HPGe detector was introduced in a series of papers by Brinkman and his collaborators in the 1960s (Brinkman et al., 1963; Brinkman and Aten, 1963, 1965) and has been used extensively ever since (Debertin and Helmer, 1988). Most commonly, it is applied to point sources measured in a close geometry, which not only improves the statistics of the sum-peak area, but also eliminates or reduces the need to take into account the angular correlation effects.

It is, nevertheless, desirable to be able to standardize also extended sources by means of the sum-peak method. An example may be measurements aimed at the half-life determination of very long-lived radionuclides, e.g. ^{176}Lu (Gehrke et al., 1990), which exhibit an appropriate decay scheme for the sum-peak method application, but may require a substantial amount of material to be measured in order to obtain good statistics.

For an extended source, however, the effects of the spatial variations of the full-energy peak and the total efficiency across the sample's volume cannot be ignored. They make the original formulae by Brinkman invalid, as demonstrated by Sutherland and Buchanan (1967), who analyzed measurements of extended sources of ^{125}I of various sizes.

That is why we decided to apply a correction factor to these formulae, determined with the help of Monte Carlo calculations. The advantage of this approach is that the experimental procedure and the formulae remain, apart from the factor itself, in their usual form and that good accuracy can be obtained. In addition, Monte Carlo simulations can take any angular correlation effect into account directly.

2. Method

The approach presented in this paper applies to ^{60}Co, which emits two coincident gamma rays. A simplified decay scheme was adopted with all the less abundant gamma rays neglected. The emission probability of the first gamma ray with the energy E_1 cancels out in the final result and is therefore not considered in the derivation below, which is a consequence of the assumption that the second gamma ray with energy E_2 always follows the first one, with negligible probability of internal conversion. A sum peak appears in the spectrum at the energy $E_1 + E_2$. Angular

* Corresponding author. Tel.: +386 1 477 3325; fax: +386 1 477 3151.
E-mail address: Tim.Vidmar@ijs.si (T. Vidmar).

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doi:10.1016/j.apradiso.2008.08.007

correlations between the directions of emission of the two gamma rays can be described by the correlation function $W(\Omega)$, which gives the probability $\mathrm{d}P(\Omega) = W(\Omega)\,\mathrm{d}\Omega$ that the second gamma ray is emitted into the solid angle $\mathrm{d}\Omega$ relative to the first one. Since this probability distribution is independent of the azimuthal angle, it can be given as a function of the polar angle θ only (Siegbahn, 1955). ^{60}Co decays in a two-step ($4^+ \to 2^+ \to 0^+$) cascade, for which the correlation function can be computed and has also been measured (Siegbahn, 1955):

$$W(\theta) = 1 + 0.1020P_2(\cos\theta) + 0.0091P_4(\cos\theta). \quad (1)$$

Here, $P_i(x)$ denotes the Legendre polynomial of the order i.

For a point source, the count rate $\dot{T}$ in the entire spectrum and the count rates $\dot{N}_1, \dot{N}_2$ and $\dot{N}_{12}$ in the peaks corresponding to the energies E_1, E_2 and the sum-peak energy, respectively, are related to the activity of the source A, the full-energy-peak efficiencies ε_1 and ε_2 for the two gamma-ray lines and the corresponding total efficiencies η_1 and η_2 through the following set of equations:

$$\begin{aligned}
\dot{N}_1 &= A\varepsilon_1(1 - w_{\mathrm{total}}\eta_2),\\
\dot{N}_2 &= A\varepsilon_2(1 - w_{\mathrm{total}}\eta_1),\\
\dot{N}_{12} &= Aw_{\mathrm{peak}}\varepsilon_1\varepsilon_2,\\
\dot{T} &= A(\eta_1 + \eta_2 - w_{\mathrm{total}}\eta_1\eta_2).
\end{aligned} \quad (2)$$

The factors w_{total} and w_{peak} take the angular correlation into account. They are not necessarily the same for partial or total energy deposition, as pointed out by Kim et al. (2003).

If we are dealing with an extended sample, the above equations are no longer valid. However, an extended source can be considered as a collection of point sources:

$$\begin{aligned}
\dot{N}_1 &= \int_V a(\vec{r})\varepsilon_1(\vec{r})[1 - w_{\mathrm{total}}(\vec{r})\eta_2(r)]\,\mathrm{d}^3\vec{r},\\
\dot{N}_2 &= \int_V a(\vec{r})\varepsilon_2(\vec{r})[1 - w_{\mathrm{total}}(r)\eta_1(r)]\,\mathrm{d}^3 r,\\
\dot{N}_{12} &= \int_V a(\vec{r})w_{\mathrm{peak}}(\vec{r})\varepsilon_1(\vec{r})\varepsilon_2(\vec{r})\,\mathrm{d}^3 r,\\
\dot{T} &= \int_V a(\vec{r})[\eta_1(\vec{r}) + \eta_2(\vec{r}) - w_{\mathrm{total}}(\vec{r})\eta_1(\vec{r})\eta_2(\vec{r})]\,\mathrm{d}^3\vec{r}.
\end{aligned} \quad (3)$$

All quantities now depend on the position $\vec{r}$ inside the sample and the activity concentration $a(\vec{r})$ has been introduced. If, however, we can consider the activity to be distributed homogeneously in the sample, Eqs. (3) can be simplified to

$$\dot{N}_1 = c_1 A, \quad \dot{N}_2 = c_2 A, \quad \dot{N}_{12} = c_{12} A, \quad \dot{T} = c_t A. \quad (4)$$

Here c_1, c_2, c_{12} and c_T are constants for a given sample size, position and composition and for a given detector on which the sample is measured, and A is the activity of the extended sample as a whole.

From this set of equations it follows that

$$A = \frac{c_{12}}{c_1 c_2 + c_{12} c_t}(\dot{T} + \dot{N}_1\dot{N}_2/\dot{N}_{12}) = C(\dot{T} + \dot{N}_1\dot{N}_2/\dot{N}_{12}), \quad (5)$$

where a new constant C has been defined. Up to this constant, the equation is the same as the one derived for a point source by Brinkman et al. (1963). According to this equation, no efficiencies need to be known to determine the activity of the source, which is the main advantage of the sum-peak method.

The correction factor C is a function of the efficiencies $\varepsilon_1, \varepsilon_2, \eta_1$ and η_2. On the other hand, it is a constant when we are dealing with a fixed geometry and fixed sample characteristics. In our approach, the correction factor C is determined by means of Monte Carlo simulations, as will be explained in the next section. This approach has the advantage that the easily measurable quantities $\dot{T}, \dot{N}_1, \dot{N}_2$ and $\dot{N}_{12}$ yield the desired result, i.e. the activity of the source.

3. Monte Carlo simulations

In order to determine the correction factor accounting for the extended nature of the sources, Monte Carlo simulations were run with the GEANT3 package (Brun et al., 1987). Most of the parameters of the detector model were taken directly from the manufacturer's data sheet. Since some data were not available in the sheet, some parameters, such as the diameter of the end-cap, had to be measured or estimated. The data were used without any optimization, and are listed in Table 1. The shielding comprising layers of lead, copper and plastic was also taken into account in the simulation. The source was modelled by sampling the geometrical dimensions of the standard ampoules of the Physikalisch-Technicshe Bundesanstalt (PTB), which were used for the measurements. The sealed ampoules are cylindrical in shape, made of glass and have an outer diameter of 15.2 mm. The height without the sealing part is about 40 mm and the thickness of the wall and bottom is 0.5 mm. The composition and density of its glass material were taken from the manufacturer's data.

Five hundred million events were simulated for each different sample geometry. A uniform distribution of the random origin of the events was assumed within the sample volume. For each simulated event two gamma rays were emitted, the first one with the energy E_1 and the second one with the energy E_2. The direction of the first gamma ray was chosen randomly from a uniform distribution spanning the full 4π solid angle. The direction of the second gamma ray was then determined according to Eq. (1), applying the Monte Carlo rejection method (Press et al., 1992).

For each sample geometry, the simulation was run twice, once with the angular correlations taken into account, and once ignoring them. In both cases, the correction factor C required in Eq. (5) was determined as

$$C = N/(T + N_1N_2/N_{12}), \quad (6)$$

where N is the number of simulated events and N_1, N_2 and N_{12} are the areas of the peaks at the energies E_1, E_2 and $E_1 + E_2$, respectively. These values were determined from the simulated spectra and so was the total number of counts in the spectrum, T. The analysis of the spectra was straightforward since the perfect resolution of the simulated spectrum made the peaks appear in a single channel. It is to be noted that Eq. (6) corresponds to Eq. (5) when the count rates are multiplied by the measurement time.

Table 1
The parameters of the detector model used in the Monte Carlo simulations

Parameter	Value A in mm	Value B in mm	$(C_B - C_A)/C_A$ (%)
Crystal diameter	58.8	58.0	0.1
Crystal length	78.0	76.0	−0.09
End-cap diameter	70.0	80.0	0.05
End-cap thickness	1.0	2.0	−0.40
Top dead layer	3.0E−4	3.0E−4	–
Side dead layer	3.0E−4	3.0E−4	–
Window thickness	0.5	0.5	–
Window-crystal distance	3.0	4.0	−0.2
Well length	65.0	60.0	−0.02
Well diameter	10.0	8.0	0.14
Crystal rounding	0.0	5.0	0.05
Sample-window distance	14.0	16.0	−0.09
Square root of quadratic sum of components			0.50

The values denoted by A were taken from the manufacturer's data sheet. Some of them were then changed one by one to the values denoted by B to assess the reliability and uncertainty of the results obtained through simulation. The corresponding values of the correction factor are denoted by C_B and C_A. They refer to Ampoule B (see Table 2). The end-cap material was in all cases aluminum and the window material was beryllium.