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In-flight annihilation correction for 511 keV photon spectrometry

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Abstract. Spectrometry based on the 511 keV annihilation photopeak requires the positron source to be surrounded by a suitable absorber material. While this method is well established in the literature, correction for in-flight annihilation losses from the 511 keV photopeak is often found to be neglected. Application to volume sources, such as typically employed in a radionuclide production environment for yield determination and quality control (QC) purposes, is presented. Source strengths obtained by analyzing the 511 keV photopeak are compared with values obtained from characteristic γ-lines for a selection of non-pure positron emitters. Better overall agreement is obtained when in-flight annihilation loss corrections are explicitly performed.

1. Introduction

An important requirement in the spectrometry of positron emitters based on the 511 keV photopeak is complete annihilation within the materials that constitute the source. Such spectrometry is often performed in cases where a particular radionuclide has significant β^+ emissions but lacks other characteristic γ-lines suitable for quantitative radioactivity assessment. In excitation function measurements utilizing the stacked-foil technique, for example, it is customary to sandwich each activated foil between sufficiently thick absorber discs when the 511 keV photopeak has to be assayed accurately. Various absorber materials have been exploited, e.g. Plexiglass (Kawade et al. [1]), Cu (Hess et al. [2], Kettern et al. [3], Qaim et al. [4]), Pb (Lépy et al. [5]), to name but a few. In one of the earlier investigations that utilized such absorbers, Kantele and Valkonen [6] warned against pair production induced by high-energy γ-rays in the absorber material. Under such conditions the use of a low-Z stopping (or absorber) material is preferred. In their work, Plexiglass, Cu, Cd and Pb were investigated for use as absorber materials.

While the use of absorbers seems to be well established, the same is not true concerning corrections for in-flight annihilation losses from the 511 keV photopeak. One finds that some authors make the necessary corrections, others do not make any explicit corrections but acknowledge these losses by introducing an additional uncertainty, while some authors simply neglect it altogether. In the case of radionuclides with relatively low β^+ end-point energies, neglecting these corrections is generally not serious but with increasing end-point energy the in-flight annihilation losses become increasingly significant.

The main aims of this paper are as follows: (1) to briefly revisit the reasons for in-flight annihilation losses; (2) to present a method for performing 511 keV spectrometry suitable for any radionuclide production counting room equipped with a good HPGe detector; (3) to apply the method to volume sources (e.g. by taking a small sample of a product solution for γ-ray assessment, which is standard practice for determining the production yield and normally required for quality control (QC) purposes [7]); and (4) to present in-flight annihilation probabilities suitable for correcting the 511 keV photopeak area, for a number of different positron emitters. Here we selected the so-called non-conventional or non-pure positron emitters, namely radionuclides that have measurable characteristic γ-ray emissions in addition to positron emissions: ^{22}Na, ^{64}Cu, ^{65}Zn, ^{66}Ga, ^{68}Ga and ^{82}Rb. This made it possible to compare yield measurements based on the 511 keV annihilation radiation directly with expectations based on the characteristic γ-rays. For a selection of absorber materials, a simplified scheme is also presented by means of which estimates of in-flight annihilation losses can easily be performed. The absorber materials investigated are water, Plexiglass, borosilicate glass, Cu and Pb.

2. In-flight annihilation losses

The majority of electron-positron annihilations that take place before the positron is slowed down to thermal energies give rise to photons with energies significantly different from 511 keV. Consequently, the probability for exclusion from the 511 keV photopeak is, to a good approximation, the same as the in-flight annihilation probability. In-flight annihilation corrections were derived using a similar approach to that of Lépy et al. [5]. As some aspects differ, the method used in the present work is discussed in some detail. In-flight annihilation resulting in the emission of one quantum or two quanta are taken into account. The sub-percent contribution of three-quantum annihilation is neglected. The cross section for

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one-quantum annihilation (OQA) of a positron with total energy $\mathcal{E}$ with an atomic K-shell electron is given by

$$\sigma_{OQA}(\mathcal{E}, Z) = \left[\frac{2\pi\alpha^4 Z^5 r_0^2}{(\mathcal{E}+1)^2\sqrt{\mathcal{E}^2-1}}\right] \times \left[\mathcal{E}^2 + \frac{2}{3}\mathcal{E} + \frac{4}{3} - \frac{\mathcal{E}+2}{\sqrt{\mathcal{E}^2-1}}\ln(\mathcal{E}+\sqrt{\mathcal{E}^2-1})\right], \quad (1)$$

where α is the fine-structure constant, Z is the atomic number of the atom in the absorber medium and r_0 is the classical electron radius (2.818×10^{-13} cm). The total positron energy has to be given in units of the electron rest mass:

$$\mathcal{E} = \mathcal{E}(E) = 1 + \frac{E}{m_0c^2}, \quad (2)$$

where E is the kinetic energy of the positron and m_0 its rest mass ($m_0c^2 = 511$ keV). According to Bethe [8], only very tightly bound electrons will significantly contribute to one-quantum annihilation (the nucleus being the third body in the kinematics) therefore shells other than the atomic K-shells can be neglected. In contrast, two-quantum annihilation (TQA) occurs readily with electrons from all shells. The cross section of a TQA event is given by

$$\sigma_{TQA}(\mathcal{E}) = \left[\frac{\pi r_0^2}{(\mathcal{E}^2-1)(\mathcal{E}+1)}\right] \times \left[(\mathcal{E}^2+4\mathcal{E}+1)\ln(\mathcal{E}+\sqrt{\mathcal{E}^2-1}) - (\mathcal{E}+3)\sqrt{\mathcal{E}^2-1}\right]. \quad (3)$$

The in-flight annihilation probability of a positron emitted with an energy E' and traversing a single-element absorber material with atomic number Z, is given by

$$P(0, E', Z) = \frac{N_A}{A}\rho \int_0^{E'} \left[2\sigma_{OQA}(\mathcal{E}, Z) + Z\sigma_{TQA}(\mathcal{E})\right] \times \left[1 - P(E, E', Z)\right]\left(-\frac{dE}{dx}\right)^{-1} dE, \quad (4)$$

where N_A is Avogadro's number, ρ is the density of the absorber medium, A is the atomic mass of the absorber medium, and $-dE/dx$ is the positron stopping power in the absorber (in units of MeV/cm). The first two parameters of function P are the limits of integration. The factor $[1 - P(E, E', Z)]$ in the integrand is a survival probability; it is the probability that the positron survived the slowing down from a higher energy E' to a lower energy E. This factor is necessary as a positron that annihilated at a higher energy cannot reappear at a lower energy. Equation (4) is a recurrence relation which can easily be solved numerically because it has a very simple initial condition, namely $P(E', E', Z) = 0$. By applying Simpson's rule, a set of terms for the integrand is first calculated by neglecting the survival probability. The summation of terms is then done in the order of highest energy towards lower energies, multiplying each successive term with the appropriate positron survival probability prior to performing the addition.

In the case of compounds, Eq. (4) can still be applied by using values for ρ and $-dE/dx$ that pertain to the compound material and the value of the molecular mass for A. Separate calculations have to be performed for each atomic species, which result in a set of partial probabilities. The total in-flight annihilation probability is then given by a linear combination of the partial probabilities with multiplication factors taken as the number of atoms of each particular atomic species as per the particular composition stoichiometry of the compound material. In this work, the composition stoichiometry of Plexiglass was taken as $(C_5O_2H_8)_n$ and for borosilicate glass (Pyrex) a typical composition was approximated as $(Si_7B_2Na_1O_17)_n$. A word of caution: The paper by Lépy et al. [5] contains an error in the expression of the OQA cross section (an incorrect sign) but the correct form is given in older works by Azuelos and Kitching [9] as well as the original paper by Bethe [8]. Stopping powers were obtained from the Nucleonica Nuclear Science Portal [10].

Table 1. In-flight annihilation probability as a function of positron emission energy for a selection of absorber materials.

E'	In-flight annihilation probability (%)				
(keV)	Water	Plexiglass	Borosilicate glass	Copper	Lead
100	2.480E-1	2.459E-1	2.710E-1	3.245E-1	4.218E-1
200	6.147E-1	6.108E-1	6.684E-1	7.858E-1	1.007E+0
300	1.007E+0	1.002E+0	1.093E+0	1.271E+0	1.621E+0
400	1.400E+0	1.395E+0	1.517E+0	1.749E+0	2.224E+0
500	1.782E+0	1.778E+0	1.930E+0	2.209E+0	2.799E+0
750	2.668E+0	2.670E+0	2.888E+0	3.254E+0	4.090E+0
1000	3.452E+0	3.463E+0	3.736E+0	4.155E+0	5.178E+0
1250	4.145E+0	4.167E+0	4.485E+0	4.933E+0	6.100E+0
1500	4.762E+0	4.797E+0	5.153E+0	5.613E+0	6.888E+0
1750	5.317E+0	5.366E+0	5.754E+0	6.214E+0	7.571E+0
2000	5.819E+0	5.882E+0	6.298E+0	6.749E+0	8.170E+0
2500	6.699E+0	6.792E+0	7.253E+0	7.668E+0	9.171E+0
3000	7.451E+0	7.574E+0	8.070E+0	8.433E+0	9.980E+0
3500	8.106E+0	8.258E+0	8.782E+0	9.085E+0	1.065E+1
4000	8.687E+0	8.868E+0	9.414E+0	9.651E+0	1.122E+1
4500	9.207E+0	9.416E+0	9.981E+0	1.015E+1	1.171E+1
5000	9.679E+0	9.916E+0	1.049E+1	1.059E+1	1.213E+1

In-flight annihilation probabilities calculated according to Eq. (4) are shown for some absorber materials in Fig. 1 and representative point-data are listed in Table 1. It is evident that the in-flight annihilation probability of positrons is very similar for water and Plexiglass, and only marginally higher in borosilicate glass.

The correction factor for losses from the 511 keV annihilation peak is given by

$$P_\beta = 1 + \int_0^{E_{max}} S(E)P(0, E, Z)dE, \quad (5)$$

where $S(E)$ is the normalized total β^+ spectrum of the radionuclide and E_{max} is the largest end-point energy of all the contributing β^+ transitions. The integral represents a weighted average over the energy range of the continuous positron emission spectrum. All allowed transitions that contribute more than 0.1% to the total β^+ spectrum were included in the calculations. Spectral data were obtained from ICRP Publication 107 [11] and/or references therein.

Table 2 compares in-flight annihilation probabilities for several positron emitting radionuclides as obtained by averaging over the full β^+ spectrum (i.e. the second

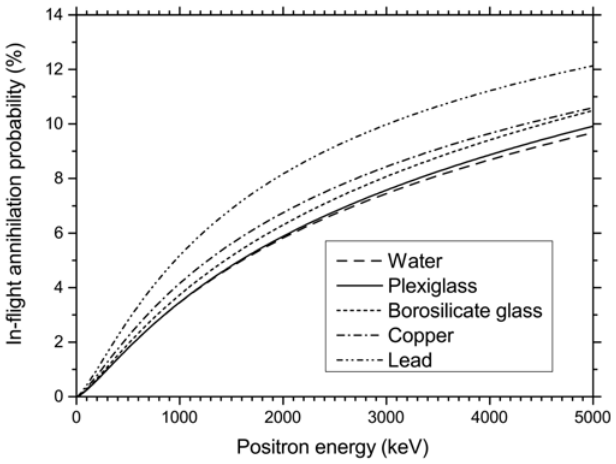

Figure 1. In-flight annihilation probability versus positron emission energy for a selection of absorber materials, as indicated.

term of Eq. (5)) with values estimated from the average β^+ energies of these radionuclides (i.e. the quantities given by $P(0, \overline{E}_\beta, Z)$ from Eq. (4)). The reasonably close agreement suggests that estimates can be made using the latter method, although the former is more precise.

3. Photon spectrometry

Sources of ^{22}Na, ^{64}Cu, ^{65}Zn, ^{66}Ga, ^{68}Ga and ^{82}Rb were prepared at iThemba LABS and counted in front of an HPGe detector to obtain their respective γ-ray emission spectra. The ^{68}Ga and ^{82}Rb sources were obtained from ^{68}Ge/^{68}Ga and ^{82}Sr/^{82}Rb generators, respectively, and had excellent radionuclidic purity. A relatively strong ^{64}Cu source initially contained some ^{61}Cu contamination, however, by the time the measurements were performed the contaminant had decayed to a negligible level. The ^{22}Na source was prepared from a stock solution that contained no measurable γ-emitting radio-contaminants. The ^{66}Ga source contained some ^{67}Ga, however, the latter radionuclide is not a positron emitter and did not affect the measurements. The ^{65}Zn was recovered together with cold Zn target material used for the production of ^{67}Ga at iThemba LABS. While the resulting ^{65}Zn product was not carrier-free, it nevertheless provided a radionuclidically pure source. All source solutions (in nominally 0.1 M HCl) were sealed in 10 mL borosilicate glass vials. These were standard serum vials with an outer diameter of 21.4 mm and a wall thickness of 1.2 mm. Each vial fitted snugly inside a cylindrical Plexiglass absorber, as shown in Fig. 2. For this work, six absorbers were prepared with wall thicknesses of 2, 4, 6, 8, 10 and 12 mm, respectively. The higher the maximum β^+ end-point energy, the thicker the absorber thickness required to stop all positrons. Radiation transport calculations were performed using the code MCNPX [12] to determine the best choice of degrader for each radionuclide investigated, i.e. the smallest Plexiglass wall thickness that would stop positrons that escaped from the source solution and borosilicate vial. Space limitations preclude presenting the MCNPX modeling results here but these will be published separately.

A single HPGe detector with a relative efficiency of 12% and a FWHM resolution of 1.8 keV at 1.33 MeV, connected to a 16 k channel Silena EMCA 2000 analyzer system, was used for all the measurements. The calibrations were performed using standard sources of ^{152}Eu and ^{133}Ba, traceable to the BIPM. These calibration sources consisted of standardized solutions, similarly sealed in identical borosilicate glass vials. The source-detector distance was 1.2 m, large enough to neglect coincidence summing effects. The most recent evaluated decay data used are summarized in Table 3.

As only activity ratios were investigated (obtained for each radionuclide from the 511 keV line and a characteristic γ-line as per Table 3) only the uncertainties associated with a relative efficiency calibration curve needed to be

Table 2. Calculated in-flight annihilation probabilities in Plexiglass for selected non-pure positron emitters.

Nuclide	$\overline{E}_\beta$ (keV)[a]	E_β^{max} (keV)[b]	In-flight annihilation probability (%)	
			$P(0, \overline{E}_\beta, Z)$	spectrum averaged
^{22}Na	216.62	546.44	0.675	0.737
^{64}Cu	278.21	653.03	0.916	0.974
^{65}Zn	142.5	330.1	0.395	0.430
^{66}Ga	1750.0	4153.0	5.366	4.812
^{68}Ga	823.5	1899.1	2.913	2.889
^{82}Rb	1479.0	3381.0	4.747	4.246

[a] Average β^+ energy.
[b] End-point energy of dominant β^+ transition.

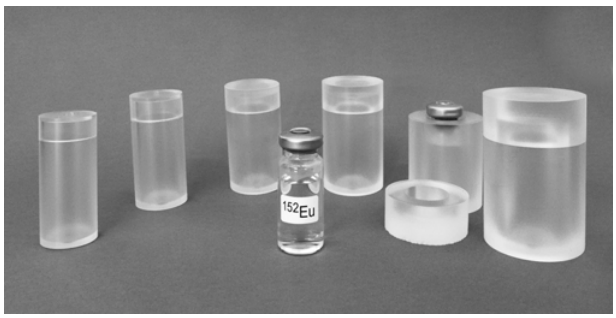

Figure 2. Plexiglass absorbers of various thicknesses (see text) and some source vials. Each absorber has a removable lid.

Table 3. Photon emission probabilities (%) and other relevant decay data of selected positron emitters.

Nuclide	$T_{1/2}$	E_{photon} (keV)	Photons per 100 disintegrations	Source
^{22}Na	2.602 a	511[a]	180.76 ± 0.04	NuDat [13]
		1274.54	99.940 ± 0.014	"
^{64}Cu	12.70 h	511[a]	35.04 ± 0.30	DDEP [14]
		1345.77	0.4748 ± 0.0034	"
^{65}Zn	244.01 d	511[a]	2.842 ± 0.014	NuDat [13]
		1115.54	50.04 ± 0.10	"
^{66}Ga	9.49 h	511[a]	114.0 ± 8.0	NuDat [13]
		1039.23	37.0 ± 2.0	"
^{68}Ga	67.83 m	511[a]	177.8 ± 0.8	DDEP [14]
		1077.34	3.235 ± 0.030	"
^{82}Rb	1.265 m	511[a]	190.9 ± 0.6	DDEP [14]
		776.52	15.02 ± 0.19	"

[a] All annihilation photons – predominantly 511 keV.

Table 4. Activity ratio (A_γ/A_{511}) for a selection of non-pure positron emitters.

Radionuclide	Ratio A_γ/A_{511}	
	No correction	With correction
^{22}Na	1.022 ± 0.016	1.015 ± 0.016
^{64}Cu	1.006 ± 0.019	0.996 ± 0.019
^{65}Zn	1.011 ± 0.016	1.007 ± 0.016
^{66}Ga	1.051 ± 0.078	1.003 ± 0.074
^{68}Ga	1.025 ± 0.019	0.996 ± 0.019
^{82}Rb	1.065 ± 0.023	1.022 ± 0.022

taken into consideration. Uncertainties in the decay data (see Table 3) as well as counting statistics were also taken into account. The attenuation of photons by the Plexiglass absorbers were quantified experimentally for all the relevant γ-lines, introducing uncertainties less than 0.5%.

4. Results and discussion

The ratios of activities determined via the 511 keV annihilation photopeak and selected photopeaks of characteristic γ-lines are summarized in Table 4, both with the in-flight annihilation losses neglected and with corrections for these losses explicitly applied. As the in-flight annihilation probabilities in water, Plexiglass and borosilicate glass differ only slightly (see Fig. 1), corrections were based on the calculations for Plexiglass only. An overall improvement is evident when the corrections are performed. Within the experimental uncertainties, the activity ratios are all consistent with unity. This is taken as ample proof that precision spectrometry based on the 511 keV annihilation photopeak is feasible for such volume sources, provided that suitable absorbers are used and in-flight annihilation loss corrections are made when the β^+ end-point energies become relatively large (> 1 MeV).

5. Summary and conclusion

An experimental procedure is described for precision spectrometry based on the 511 keV photopeak of β^+ emitting radionuclides. For this purpose, Plexiglass absorbers with thicknesses ranging between 2 and 12 mm have been employed. Source solutions sealed in standard 10 mL borosilicate glass serum vials fit snugly inside these absorbers. It was shown that explicit corrections for in-flight annihilation losses result in an overall improvement in the agreement between yield measurements based on the 511 keV annihilation photopeak and values determined from other selected characteristic γ-lines.

In-flight annihilation probabilities obtained by averaging over the full β^+ spectrum were found to be in satisfactory agreement with values evaluated at the average β^+ energies of the investigated radionuclides. This suggests that estimates can be made using the latter (simpler) method, although the former is expected to be more precise.

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