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Three-photon yield of positron annihilation in Cab-O-Sil (SiO_2)†

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Abstract. Three-photon yields of positron annihilation in Cab-O-Sil (SiO_2 powder) samples, where the grains have different surface areas, have been measured using the valley-to-peak technique. Correlations among the three-photon yield, the intensity of the longest-lived component in the lifetime spectrum, and the surface area have been discussed.

1. Introduction

The strong dependence of positron annihilation lifetime spectra in fine powders was interpreted in terms of the diffusion of positronium (Ps) atoms from within the powder grains into interstices between the grains [1]. A relation (see reference [1]) between the intensity, I_2, of the bulk lifetime component attributed to ortho-Ps annihilating inside the grains and the Ps diffusion constant, D, was both theoretically predicted and experimentally tested [1, 2]. Also, an effort was made to establish a universal curve relating the intensity, I_3, of the longest-lived component (attributed to ortho-Ps annihilating in the space between grains) and the surface area, S, of the grains [2]. Recently, results of lifetime measurements on positron annihilation in Cab-O-Sil (SiO_2 powder) samples of different surface areas were reported [3]. The results showed that the equation of I_2 versus D can only be considered to be an approximation and the empirical relation of I_3 versus S was not a universal equation. The data also indicated that I_3 included both the 2-γ and the 3-γ annihilation. Present experiments measured the 3-γ yields of positron annihilation in the same samples using the valley-to-peak technique. Correlations among the 3-γ yield, I_3, and S have been established.

2. Experimental procedure

The samples of Cab-O-Sil (SiO_2 powder) were kindly supplied by Cabot Corporation, Tuscola, Illinois, USA. These powders are fine particles with spherical shape. The true density of the aggregate is 2.20 g cm^{-3} and its bulk density in the standard grade is

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approximately 0.032 g cm^{-3} [4]. This implies that the free volume per particle is about 69 times larger than the particle volume. Seven different grades of different surface areas were used for the present experiments.

The valley-to-peak method has been used by many researchers to determine the relative yield of 3-γ to 2-γ annihilation [5–8]. Let

I_v = the counts of photons in the energy region lower than 511 KeV (valley)

I_p = the counts of the 511 KeV photons (peak)

P_3 = the yield of 3-γ annihilation

P_2 = the yield of 2-γ annihilation

E_3 = the average detection efficiency of the detector for 3-γ annihilation

E_2 = the average detection efficiency of the detector for 2-γ annihilation

F_3 = the ratio of 3-γ annihilation photons in the 'valley' region to those in the whole spectrum

F_2 = the ratio of 2-γ annihilation photons in the 'peak' region to those in the whole spectrum

then

$$I_v/I_p = 3P_3E_3F_3/2P_2E_2F_2. \quad (1)$$

The factor $\frac{3}{2}$ is the ratio of photons emitted from 3-γ to 2-γ annihilation.

Since part of 2-γ annihilating photons contribute to the 'valley' region due to Compton effect, a correction must be made to equation (1). The correction is KI_p, where K is the ratio of 2-γ photons in the 'valley' region to 2-γ photons in the 'peak' region of a reference aluminium (Al) sample. Therefore,

$$(I_v - KI_p)/I_p = 3P_3E_3F_3/2P_2E_2F_2 \quad (2)$$

or

$$P_3/P_2 = (I_v - KI_p)/\tfrac{3}{2}(F_3/F_2)(E_3/E_2)I_p.$$

Positron annihilation energy spectra for seven kinds of SiO_2 powders both in vacuum and in air were obtained using a 7.6 cm diameter × 7.6 cm long NaI (Tl) detector. The 340–450 keV portion (channel interval 20–33) of the spectrum was chosen for the 'valley' region and the 450–578 KeV portion (channel interval 34–46) was chosen for the 'peak' region. For an aluminum sample, the ratio, K, of I_v to I_p was 0.058. The detection efficiencies E_2 and E_3 were estimated to be 18.5% and 22.0% respectively from the manual for the detector. The value of F_3 was determined to be 0.345 using the energy spectrum for 3-γ annihilation of positrons [9]. The value for F_2 was assumed to be 1.

3. Results and discussion

Figure 1 shows a set of positron annihilation energy spectra for Al and various SiO_2 powders in vacuum. Curve A is for Al. Curves B, C and D are for SiO_2 of surface areas 90, 200 and 380 (m^2 g^{-1}), respectively. The 511 KeV peaks have been normalised to be the same height. Figure 2 shows the corresponding spectra in air. It is evident that there is more 3-γ for SiO_2 in vacuum than in air since the 'valley' of the energy spectrum in vacuum is higher than the corresponding 'valley' in air.

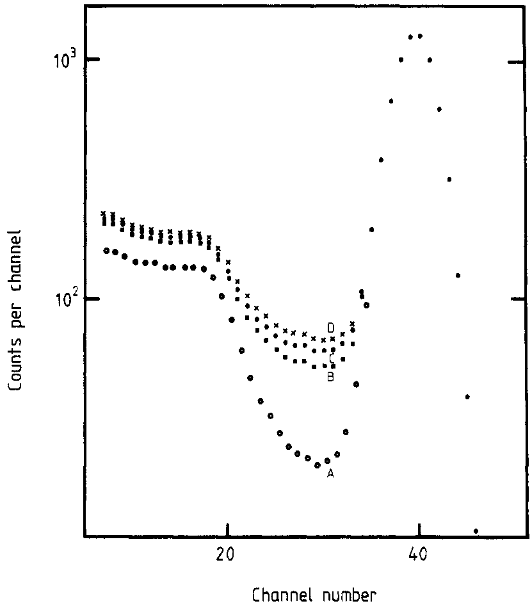

Figure 1. Positron annihilation energy spectra for Al (curve A) and for SiO_2 grains having surface areas 90 $m^2\,g^{-1}$ (curve B), 200 $m^2\,g^{-1}$ (curve C) and 380 $m^2\,g^{-1}$ (curve D) in a vacuum.

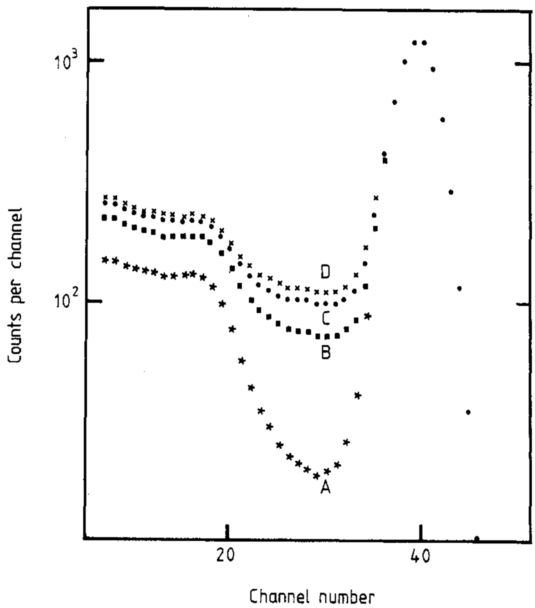

Figure 2. Positron annihilation energy spectra for Al (curve A) and for SiO_2 grains having surface areas 90 $m^2\,g^{-1}$ (curve B), 200 $m^2\,g^{-1}$ (curve C) and 380 $m^2\,g^{-1}$ (curve D) in air.

Values of I_v/I_p obtained from these measurements were used to calculate P_3/P_2 using equation (2). The relative 3-γ yields $I_{3\gamma} = P_3/(P_2 + P_3)$ were then obtained. Results of I_v/I_p and $I_{3\gamma}$ for various SiO_2 samples are listed in table 1 together with the lifetime τ_3, and the intensity, I_3, of the longest-lived component in the lifetime measurements. The value of I_v/I_p for Al was 0.058. The percentage errors for S and $I_{3\gamma}$ were about 10%, and for τ_3 and I_3 were about 5%.

The relative 3-γ yield, $I_{3\gamma}$, increases as the particle size decreases (surface area increases). This verifies the previous explanation that more ortho-Ps will diffuse into the free space for smaller size particle. Corresponding values of $I_{3\gamma}$ are smaller in air. This

Table 1. Positron annihilation parameters in SiO_2.

Samples	Vacuum				Air			
S ($m^2 g^{-1}$)	I_v/I_p	$I_{3\gamma}$ (%)	τ_3 (ns)	I_3 (%)	I_v/I_p	$I_{3\gamma}$ (%)	τ_3 (ns)	I_3 (%)
90	0.157	13.9	137	5.14	0.117	8.8	70.5	5.91
130	0.177	16.2	137	5.98	0.125	9.8	72.4	7.12
160	0.187	17.4	138	6.22	0.126	10.0	74.1	8.07
200	0.203	19.1	138	7.91	0.133	10.9	73.7	8.92
250	0.217	20.6	136	8.97	0.134	11.0	75.8	9.31
325	0.227	21.6	129	9.88	0.136	11.3	76.2	9.95
380	0.230	21.9	129	10.70	0.148	12.7	75.2	10.60

may be attributed to quenching reactions of ortho-Ps either by 'pick-off' annihilation or more probably by 'conversion' due to the oxygen in air [10]. This confirms results from lifetime measurements [3].

Values of $I_{3\gamma}$ are larger than I_3 obtained from lifetime measurements. This may be explained by the fact that I_3 does not include all self-annihilation of ortho-Ps [3]. The energy-selection window for the stopping signal (annihilation photons) was set at the upper 50% of the Compton spectrum of the 511 KeV γs. This will not give the real ratio of 2-γ to 3-γ events due to different energy distributions for 2-γ and 3-γ annihilations. Furthermore, the energy dependence of the efficiency of the detector used (5 cm diameter ×2.5 cm long Pilot U detector) may also alter the real ratio of 2-γ to 3-γ annihilations. Therefore, the measured I_3 does not include all self-annihilation of ortho-Ps in free space. Let this unmeasured portion be I_4; then $I_{3\gamma}$ should be equal to $I_3 + I_4$. I_4 is more significant in a vacuum when there are more 3-γ annihilations. This can explain the results in air, where the differences between $I_{3\gamma}$ and I_3 are smaller.

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