

Effect of positron source irradiation on positronium annihilation in fine powdered alumina



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ABSTRACT

Positron lifetime and Doppler broadening of positron annihilation radiation were measured as a function of time to study the irradiation effect by ²²Na positron source in fine powdered alumina. The γ -Al₂O₃ samples were put in a vacuum chamber with a pressure of about 10⁻⁶ Torr and were cooled down to 10 K by a closed-cycle helium gas refrigerator. The irradiation of γ -Al₂O₃ samples by positron source was taken for a duration of about two days immediately after the sample was cooled down. After that, the sample was subjected to a warm up process from 10 K to 300 K with a step of 10 K. Positron lifetime and Doppler broadening spectra were measured simultaneously during these processes. Two long lifetime components corresponding to ortho-positronium annihilation were observed. A significant shortening of these long lifetime components and a large increase in S parameter is observed during irradiation. It is supposed that positron source irradiation creates a large number of paramagnetic centers on the surface of the γ -Al₂O₃ grains, which induce spin conversion quenching of positronium. The irradiation induced paramagnetic centers are unstable above 70 K and are nearly annealed out when the temperature rises to 190 K. After warming up of the sample to room temperature, the positron lifetime spectrum is identical to that before irradiation. It was also found that after irradiation, a medium long lifetime component of about 5 ns appears, of which the intensity increases with increasing irradiation time. This may be originated from the formation of the surface o-Ps state. This surface o-Ps state is also inhibited at elevated temperatures. Our results indicate that positronium is a very sensitive probe for the surface defects in porous materials.

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1. Introduction

Positron annihilation in fine powdered materials is an old but still very interesting subject [1–4]. In high electron density environment positron will annihilate soon after being injected into solids, or diffuse to the surface, which is the case of pure metal [5–7]. Unlike the situation in pure metal, when positron is injected into fine powdered insulator oxides such as Al₂O₃, SiO₂ and MgO, it can show very long-lifetime components and high probability of three gamma annihilation due to the formation of positronium [2,4]. Positrons emitted from ²²Na source will lose kinetic energy very rapidly through ionization and scattering and eventually get thermalized within few ps. After this process of thermalization, positrons start to diffuse randomly with a typical diffusion length of about hundred nm. Generally the grain size of materials is much larger than the mean diffusion length of positrons, which means that positron could hardly escape from the grain to the surface. So

the positron can only stay inside the grain either in the interstitial site or get trapped into vacancy defects and annihilate with an electron nearby. In case of fine powdered materials the grain size can be smaller than the mean diffusion length of positron so that some of the bulk positrons get chances to diffuse to the surface of the grains to form surface state positrons or to capture electrons to form the hydrogen-like meta-stable positronium (Ps) atom. In the following part of this paper we will demonstrate how important is this Ps atom at low temperature.

Positronium, the bond state of a positron and an electron, can exist in either singlet state ($S=0$) or triplet state ($S=1$) which is called *para*-positronium (*p*-Ps) and *ortho*-positronium (*o*-Ps), respectively. Different decay modes take part in the annihilation of Ps in vacuum according to their spin states: *p*-Ps tends to annihilate into two gamma rays while *o*-Ps mainly annihilates into three gamma rays. In vacuum *o*-Ps has a lifetime of 142 ns while *p*-Ps has lifetime of only 125 ps. After formation of Ps, *o*-Ps lives more than thousand times longer than *p*-Ps thus has enough chances to continuously collide with grains to lose its kinetic energy. Once emitted into the open space between the grains, the Ps atoms

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can never penetrate back into grains because of the negative Ps work function. So *o*-Ps only interacts with the grain surface before annihilation. Once the positronium is localized in open space of materials, its annihilation (especially that of *o*-Ps) is strongly dependent on the structure and chemical environment of the open space where the positronium is localized. The *o*-Ps lifetime will be largely reduced to a few ns due to its pick-off annihilation with the surrounding electrons. The smaller the open space, the shorter will be the *o*-Ps lifetime. On the other hand, positronium can also interact with some chemically active components which may exist on the wall of free space, and both the lifetime and formation probability of *o*-Ps will be affected. These properties make Ps the good probe for pore structure of materials.

In conventional positron annihilation experiments, the positron emitted from ^{22}Na source has a wide energy distribution with maximum energy of 545 keV and average energy of about 220 keV. After the energetic positrons are injected into materials they may produce large amount of electron–hole pairs [8–10]. On the other hand, the ^{22}Na source will also emit 1.28 MeV γ -rays, and these γ -rays have similar irradiation effects. In fine powdered materials the irradiation induced electrons or holes may migrate to the surface of the grains and form surface defects where positron may be trapped as surface positron state or surface positronium state. Interactions between Ps atoms and irradiation-induced defects on the grain surfaces of powdered Al_2O_3 and α - SiO_2 were reported by Dauwe et al. [11] and Saito et al. [12,13] respectively. Both work reported that the long lifetime of *o*-Ps atoms and its intensity decreased which they attributed to the spin conversion quenching of positronium through collision with the paramagnetic centers on the grain surface induced by the irradiation of the energetic positrons. Dauwe et al. attribute the surface paramagnetic centers in fine powdered Al_2O_3 to Al^{3+} interstitials which migrate to the surfaces of the grains at low temperature. Saito et al. pointed out that the observed surface paramagnetic centers are unstable at room temperature and are likely due to non-bridging oxygen–hole center. However the detailed thermal stability of these paramagnetic centers is not available.

In this paper we performed both positron lifetime and Doppler broadening measurements for fine powdered Al_2O_3 at low temperatures as a function of time to study the irradiation effect and the temperature dependence of irradiation induced defects. Clear evidence of spin conversion of positronium due to the irradiation induced surface defects is observed, and the surface defects are unstable above 190 K. Our results indicate that Ps atom is a sensitive probe to investigate the irradiation effect induced by positron source.

2. Experimental

Commercially available γ - Al_2O_3 nanopowder (grain size ~ 20 nm, purity $> 99.96\%$) were compressed into pellets of 15 mm diameter and 1 mm thickness under a static pressure of 6 MPa for 5 min. The samples were dried at 110°C for 2 h before any measurements to remove the water.

Positron annihilation lifetime and Doppler broadening apparatus used in this study has been described elsewhere [14]. A $15\ \mu\text{Ci}$ ^{22}Na source was used as positron source. For all the positron experiments the samples were evacuated to 10^{-6} Torr and were cooled by a closed-cycle helium gas refrigerator. A Lakeshore model 331 temperature controller was used to stabilize temperature at low temperature with accuracy of 0.1 K. The γ - Al_2O_3 samples were cooled down from room temperature to 10 K in 45 min. The irradiation of γ - Al_2O_3 samples was taken at 10 K for about two days immediately after been cooled to low temperature. After that the samples were heated from 10 K to 300 K with a step of 10 K, and the

positron lifetime and Doppler broadening spectra were measured at each temperature step.

3. Results and discussion

3.1. Irradiation effect

The measured positron lifetime spectra before and after irradiation for about two days at 10 K are shown in Fig. 1. The main features of these spectra are the appearance of a long tail after the peak, which suggests existence of ultra long lifetime components. This long lifetime components is obviously due to *o*-Ps annihilation in the free space which might be the interspace between grains. It is unambiguous that after positron source irradiation at low temperature for two days, a more slant tail instead of the long flat tail appeared and the shoulder after peak became wider. This indicates that the irradiation effect is obvious even a $15\ \mu\text{Ci}$ weak source was used. On the contrary, positron lifetime spectrum measured at 300 K showed no time dependence.

All the positron lifetime spectra were analyzed using PATFIT program by fitting the statistical data with a sum of several exponential functions [15]. The average counts before the lifetime peak were selected as the background to be subtracted before analysis. Since a kapton foil sealed ^{22}Na positron source was used in our experiment, the source correction contains a 380 ps component with relative intensity 10% and a 2.09 ns component with relative intensity 1.05% [6]. Commonly the lifetime spectrum of γ - Al_2O_3 samples consists of four components: [16,17] two long and two short. The shortest component τ_1 has a value of (200 ± 4) ps while the second short component $\tau_2 = (478 \pm 7)$ ps at low temperatures. Free positron state in the γ - Al_2O_3 grains and the weak *p*-Ps state contribute τ_1 together. Meanwhile τ_2 should be carefully treated because this positron surface state may be mixed up with positron defect state and source component. The two long lifetime components τ_3 (8.55 ± 0.44 ns) and τ_4 (83.31 ± 0.98 ns) are obviously due to *o*-Ps annihilation in small voids and large pores, respectively. The longest lifetime τ_4 is responsible for the long tail appeared in the lifetime spectrum as shown in Fig. 1.

With elongated measurement time, the two short lifetime components show no change. So we will pay more attention to the long lifetime components. Variations of τ_3 and τ_4 with irradiation time are shown in Fig. 2 and the corresponding variations of intensities I_3 and I_4 are shown in Fig. 3. A significantly shortening of τ_4 can be observed as a function of irradiation time, and the shorter *o*-Ps lifetime τ_3 shows similar but a weaker decrease. The intensity I_4 also shows a significant decrease, while the variation of I_3 is opposite, which increases with increasing time. There must be a quenching

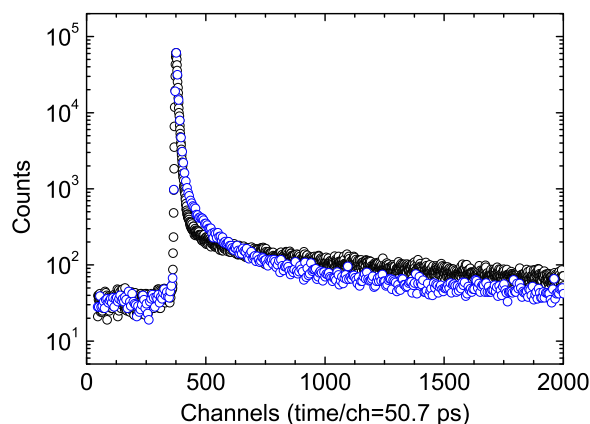


Fig. 1. The raw lifetime spectra of γ - Al_2O_3 measured at low temperatures before and after irradiation.

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