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Effect of oxygen impurities on positronium formed in voids of vanadium

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Abstract. To clarify the effect of oxygen impurity on positronium (Ps) formed in radiation-induced voids, measurements of the angular correlation of annihilation radiation have been made on vanadium samples with controlled oxygen impurity contents up to 1600 at.ppm after irradiation with 1×10^{20} fast neutrons cm^{-2} at about 430 °C. It has been shown that contamination of voids with oxygen impurities, presumably on the void surface, leads to Ps formation.

Since the first discovery of trapping of positrons in voids, positrons have been assumed to be trapped at the void surface (Brandt and Dupasquier 1983). However, a remarkable, narrow, component with high intensity has been observed in the angular correlation of annihilation radiation (ACAR) curve for niobium and vanadium (Kuramoto *et al* 1981, Hasegawa *et al* 1983, 1985) and aluminium (Jensen *et al* 1985, 1988). This narrow component is assumed to be due to positronium (Ps) formed in the void and is found to be sensitively affected by heating after irradiation as well as by doping with impurities such as gaseous elements in vanadium and niobium. Thus it is very important to study the effect of impurities on Ps formed in vanadium samples with controlled doping. It is also interesting to compare slow-positron behaviour in the voids with that on the clean surface, which was extensively studied recently (Lynn 1983, Mills 1983, Schultz and Lynn 1988). In order to understand the physical properties of material containing voids, it is very important to study the properties of the void interior (such as surface segregation) for which we do not have experimental tools.

Vanadium samples with oxygen content up to 1600 at.ppm were prepared as follows. Firstly, to remove the impurity oxygen atoms, a zirconium-foil wrapping technique (Abe *et al* 1976, Yoshinari *et al* 1981) was used; vanadium sheets wrapped with a zirconium foil were sealed in an evacuated quartz tube and then heated at 1100 °C for 2 h. During this heat treatment, the oxygen impurity content was reduced to the less than 10 at.ppm, which was the detectable limit of the internal friction method (Yoshinari *et al* 1981). The oxygen content was controlled by annealing the samples at 1120 °C in an evacuated quartz tube; on changing the heating time from 5 to 90 min, the oxygen content was varied from 210 to 1600 at.ppm. Subsequently the oxygen-doped samples were homogenised at 900 °C for 24 h. The oxygen content was determined from the height of the Snoek peak

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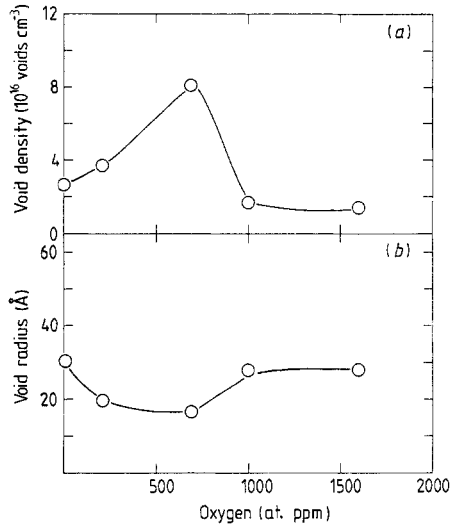


Figure 1. Effect of the oxygen doping on (a) the void density ρ_v and (b) the void radius R_v , obtained with an electron microscope.

in the internal friction spectra. The samples thus obtained were irradiated in the Japan Materials Testing Reactor to a fluence of 1×10^{20} fast neutrons cm^{-2} (at an energy greater than 1 MeV) at about 430 °C for 20 days. A long-slit apparatus with geometrical resolution of 0.63 mrad in FWHM (Hasegawa *et al* 1974) was used to obtain the ACAR curves.

Transmission electron microscopy observation has revealed voids of 20–30 Å radius and density of about 10^{16} cm^{-3} as shown in figure 1. The void density increases with increasing oxygen content up to 690 at.ppm, while the void radius decreases with increasing oxygen content in this concentration range. This result is consistent with previous electron microscopy observations (Bressers and van Witzenburg 1979). However, it has been found that the void radius increases but the density decreases for an oxygen content higher than 690 at.ppm.

Figure 2 shows ACAR curves for the vanadium samples with various oxygen contents. Remarkable narrowing of the ACAR curve caused by irradiation is observed in every sample, suggesting positron trapping in the voids. The narrowing of the ACAR curve becomes more evident for samples with higher oxygen contents; narrow components with a FWHM of about 1 mrad are clearly seen at the top portion of the curves for 210–1600 at.ppm O but not for the undoped sample. These ACAR curves for the doped samples can be decomposed into the sum of three Gaussians by least-squares fitting as shown by the broken and chain curves in figure 2, where the FWHM widths of narrow (N), intermediate (M) and broad (B) components are about 1 mrad, 5 mrad and 11 mrad, respectively. We have observed that the widths of the M and B components are independent of the oxygen content. We assume that the M component is due to the annihilation of surface state positrons, pick-off Ps and possibly physisorbed Ps. The B component is ascribed to the annihilation of positrons in the bulk matrix except the voids. The N component is due to self-annihilation of free Ps confined in the voids (Hasegawa *et al* 1985); the motion of the centre of mass of free Ps formed in voids at finite temperatures is given fairly well by a simple model in which the Ps atom is confined

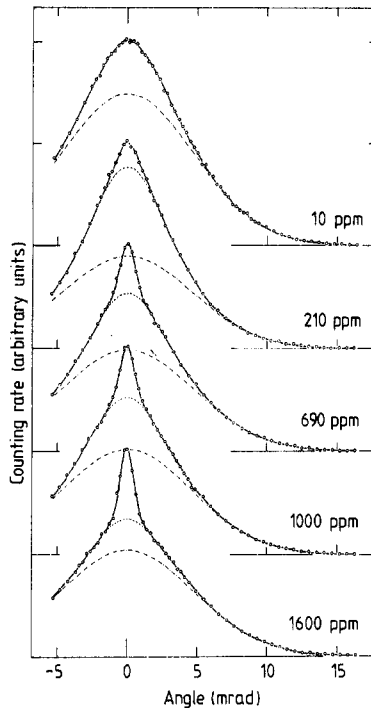


Figure 2. The ACAR curves for the irradiated samples: — — —, - - -, show the Gaussian decomposition.

by a spherical infinite well potential with the radius of the void and is populated in the higher-energy levels according to Boltzmann statistics.

It should be noted that the N component is recognised only in the curves for the samples with an oxygen content higher than 210 at.ppm, although all samples contain voids of nearly the same radius and comparable density as that for figure 1. These results suggest that the existence of oxygen atoms is responsible for the appearance of the N component.

In figure 2 we notice that the N component increases with increasing oxygen content. The intensity I_N of the N component and the intensity I_M of the M component are plotted in figure 3(a). I_N increases gradually with increasing oxygen content. I_M increases rapidly at lower oxygen contents and this is followed by a reduction on further increase in oxygen content. Figure 3(b) shows the total void component $I_V = I_N + I_M$ which comes from the total trapped positrons, i.e. surface positrons and Ps in the voids. I_V is not simply related to the oxygen content but presumably reflects positron dynamics and diffusion into the voids. An important Ps parameter is the relative intensity of the N component to I_V : $i_N = I_N/I_V$. The intensity i_N gives the ratio of singlet self-annihilation of Ps to the total trapped positrons in the voids. It depends only on the interior state of the voids and not on bulk properties such as densities of voids and dislocation. The remarkable result in figure 3(b) is that i_N increases linearly with increasing oxygen content, although I_V has the complex relation stated above. This confirms definitely that the oxygen impurity atoms, which presumably are located on the void surface, lead to Ps formation. Our other study (Hasegawa *et al* 1988) has showed that i_V increases with increasing irradiation dose as well as increasing heating time at higher temperature after irradiation. Both

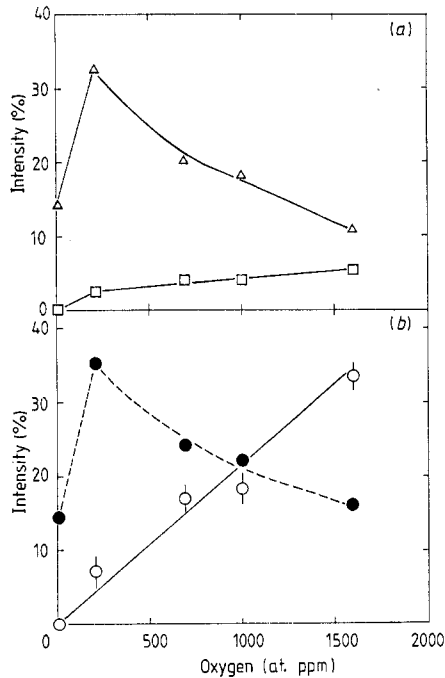


Figure 3. Effect of the oxygen doping on (a) the intensity of I_N , the N component (□) and the intensity I_M of the M component (△) and (b) the intensity I_v of the total void (●) and the relative intensity ($i_N (= I_N/I_v)$) of the narrow component (○).

procedures cause the contamination by the oxygen impurity on the internal surface of the void.

Unfortunately we cannot determine the coverage of oxygen atoms on the void surface because we do not have an experimental tool for it. However, the coverage is assumed to be less than monolayer from simple estimation. The fraction of atoms on the void surface in a sample containing voids of 30 Å radius and of $1 \times 10^{16} \text{ cm}^{-3}$ density is evaluated to be 1×10^{-4} . It will be impossible for all the oxygen impurity atoms to segregate on the void surface. Consequently it is reasonable to suppose that only a small fraction of the impurity oxygen atoms can be adsorbed on the surface. It should be noted that Ps formation is sensitively affected by the slight contamination such as submonolayer coverage on the void surface.

A possible way of explaining the enhancement of Ps formation by the contamination of void surface is to assume that the oxygen coverage leads to a decrease in the energy depth of the surface bound state for positrons or to a reduction in the work function of electrons with which the positrons form the Ps atoms. An effective positron energy level for Ps in open space is given by $-(6.8 \text{ eV} - \Phi_-)$ where 6.8 eV is the Ps binding energy and Φ_- is the electron work function (Ishii 1984a, b). In the case of voids, we have additional energy shifts due to the confinement, which can be estimated from the simple model of the infinite spherical potential; the ground-state energy level for the void with a 30 Å radius is evaluated to be about 20 meV higher than that for free space. This confinement effect will not cause a large difference in the energy level of Ps in the void relative to the surface bound state. Thus our finding corresponds well to recent slow-positron studies on the effect of oxygen in which the oxygen coverage on a clean surface

enhances Ps escaping from the surface (Lynn 1983, Mills 1983, Schultz *et al* 1985). Detailed study of close correlation between slow positrons at a clean surface and Ps formation in voids will be required in order to understand the physical process of the Ps-oxygen interaction.

In conclusion, Ps formation in the voids has been found to be attributed to the oxygen impurity atoms, presumably on the submonolayer coverage of the void surface. These results on Ps in the voids indicate that the Ps technique will give us useful information on radiation-induced voids.

Acknowledgments

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