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Positron Annihilation in Neutron- and Electron-Irradiated Silica Glass

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Positron lifetime and angular correlation of annihilation radiation (ACAR) have been measured on fused and synthetic silica glass before and after irradiation with fast-neutrons up to $8x10^{18}$ n/cm^2 below 150°C or with 3 MeV electrons up to $1x10^{18}$e/cm^2 below 50°C. Before irradiation positron lifetime spectra show characteristic long-lifetimes due to ortho-positronium (o-Ps) formation and are well decomposed into 3 components with the time constants τ_i (i=1,2,3) and the relative intensities I_i ($I_1 + I_2 + I_3 = 1$). The τ_3 lifetime is about 1.7 ns, being nearly independent of the samples, while τ_2 is strongly dependent on the samples, ranging from 0.7 to 1.2 ns. Narrow peaks on the ACAR curves and the τ_3 lifetime give evidence of Ps formation in structural open-spaces (voids) with radius about 0.3nm. By neutron-irradiation the τ_2 lifetime has reduced to about 0.47 ns irrespective to the samples but its intensity (I_2) has increased with the dose. This component suggests positron trapping without Ps formation at irradiation-induced defects such as peroxy radicals. Annealing behavior of lifetime parameters is also presented and compared with previous ESR work.

KEYWORDS: Positron Annihilation, Positronium, Silica Glass, Irradiation Effects,Voids, Defects

1. Introduction

Study of irradiation damage in silica glass is related to important technological problem of optical fiber in nuclear power plants and long-term behavior of radioactive waste storage materials. It is also of particular interest because silica glass is a prototype for many glassy materials and its defect structure is of intrinsic scientific interest.

The positron annihilation technique is widely used in the studies of irradiation-induced voids in metals, especially for investigating the initial stage of void nucleation and the interior of voids [1-3]. Positronium (Ps), which is the bound state of a positron and an electron, has been known to be formed in irradiation-induced voids of Nb, V and Al when these voids are decorated with impurities [4-7]. In this case Ps provides useful information about the interior of the voids, such as impurity segregation and gaseous-molecule inclusion, through the Ps momentum distribution and Ps chemical reactions in the voids. Recently Ps formation has been observed in irradiation-induced voids in aluminum oxide single crystal and provide us with information about average radius and internal states of the voids [8].

A perfect silica glass is visualized as a continuous random network (CRN) of SiO_4 tetrahedron joined at the corners such that each silicon is bonded to four oxygens and each oxygen bridges between two silicons [9,10]. The resulting structure is rather open, as seen from its density of about 2.2 g/cm^3 much less than that of crystal quartz (2.65g/cm^3). Then positrons are likely trapped in structural open spaces (voids). Irradiation causes displacement of oxygen and silicon atoms from their bonding state. Because of its smaller displacement energy for oxygen atoms (about 10 eV) than that for silicon atoms (about 20 eV) [10], dominant defects are supposed to be oxygen vacancies (E' center) and their complementary members of Frenkel defects, oxygen-associated hole centers, such as nonbridging-oxygen hole centers (NBOHC) and peroxy radicals (POR). It is therefore of particular interest to investigate positron trapping and Ps formation in structural voids and irradiation-induced defects in silica glass.

2. Experimental

Specimens of about 8x8x1 mm^3 were cut from various kind of silica glass plates (Nippon Silica Glass Corp., Japan), as listed in Tab.1 together with their chemical analysis. The HRP glass is produced from natural quartz by electrical fusion and has a low OH-group content but a relatively high metallic-impurity content. The OX glass was produced from quartz powder by flame fusion. The ES and ED glass are synthetic silicas; ES is flame-hydrolyzed silica and contains 1200 ppm OH and 70ppm Cl, while ED was produced by the vapor phase axial deposition (VAD) [12] method and contains very small amount of metallic impurity. For reference, single crystal plates of quartz were also employed.

These were bombarded with fast neutrons up to a dose of $8.3x10^{18}$ n/cm^2 below 150°C in the Irradiation Facility of Hydraulic Rabbit II in Japan Materials Testing Reactor (JMTR). The fast neutron flux was $1.6x10^{13}$n/cm^{2}sec. The samples irradiated to $8x10^{17}$ were isochronally annealed for

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Table 1. Silica glass samples and their chemical analysis. Results of chemical analysis are from Nippon Silica Glass Co. and presented in unit of wt.ppm. Classification of silica glass (silica type, I, II and III) is according to its preparation method [11].

	Type	Al	Ca	Cu	Fe	Na	K	Li	Cl	OH
HRP	I	8.8	0.7	<0.01	0.2	0.1	0.2	0.05	0	10
OX	II	20	0.4	0.01	0.3	1.0	1.0	1.0	0	150
ES	III	0.1	0.1	0.01	0.05	0.05	0.05	0.05	70	1200
ED	VAD	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<1.0	100

Table 2. Positron lifetime parameters of the HRP silica glass irradiated with fast neutrons or 3MeV electrons.

Fluence(1/cm2)	τ_1 (ns)	τ_2 (ns)	I_2 (%)	τ_3 (ns)	I_3 (%)	I_2+I_3	variance
Unirrad.							
	0.1064	0.8783	20.22	1.6890	59.41	79.63	1.306
	±0.0019	±0.0454	±2.14	±0.0186	±2.26	±0.20	
Neutron							
8.6×10^{16}	0.1547	0.4728	38.59	1.5973	41.47	80.06	1.384
	±0.0050	±0.0104	±0.70	±0.0085	±0.44	±0.93	
8.6×10^{17}	0.1751	0.4712	67.09	1.6163	21.73	88.83	1.262
	±0.0129	±0.0070	±1.11	±0.0153	±0.41	±1.36	
8.3×10^{18}	0.1558	0.4712	81.71	1.7650	11.36	93.07	1.422
	±0.0149	±0.0040	±0.72	±0.0233	±0.27	±0.88	
Electron							
5×10^{17}	0.1303	0.5540	21.90	1.6220	57.31	79.21	1.521
	±0.0028	±0.0175	±0.43	±0.0075	±0.58	±0.44	
1×10^{18}	0.1334	0.4769	27.38	1.6173	52.84	80.22	1.452
	±0.0035	±0.0122	±0.45	±0.0066	±0.42	±0.61	

30 min to 900°C in air. Some samples were irradiated with 3 MeV electrons up to a dose of $1x10^{18}$ e/cm^2 below 50°C.

Positron lifetime measurements were carried out with use of a usual apparatus with time resolution of about 180 ps (FWHM). The lifetime spectra were analyzed with the PATFIT-88 programs [13]. The ACAR curves were measured using a conventional long-slit apparatus with geometrical resolution of 0.63 mrad (FWHM).

3. Results and Discussion

Figure 1 shows typical lifetime spectra for the unirradiated and irradiated samples of silica glass. Before irradiation lifetime spectrum shows characteristic a long-lifetime , usually longer than about 1ns, due to o-Ps pick-off annihilation. Ps exhibits two spin states which are called "ortho" (triplet) and "para" (singlet). In vacuo self-annihilation lifetimes for o-Ps and p-Ps are 142 ns and 0.125 ns respectively. In solids, however, this long lifetime of o-Ps is shortened to a few ns or less by pick-off and spin-conversion processes. After irradiation mean positron lifetime is shortened; the intensity of the long lifetime component decreases with neutron dose. For single crystal, on the other hand, the long lifetime component due to o-Ps pick-off annihilation is hardly observed and further mean lifetime increases by irradiation (fig.1(b), as usually observed in metals and semiconductors.

The lifetime spectra for silica glass are well decomposed into three lifetime components with the time constants τ_i and the relative intensities I_i (i=1,2,3) ($I_1 + I_2 + I_3 = 1$). Table 2 and Figure 2 present these lifetime parameters against fast-neutron dose. The lifetime τ_2 decreases from 0.88ns to 0.47ns by irradiation of $8.6x10^{16}$n/cm^2, but shows

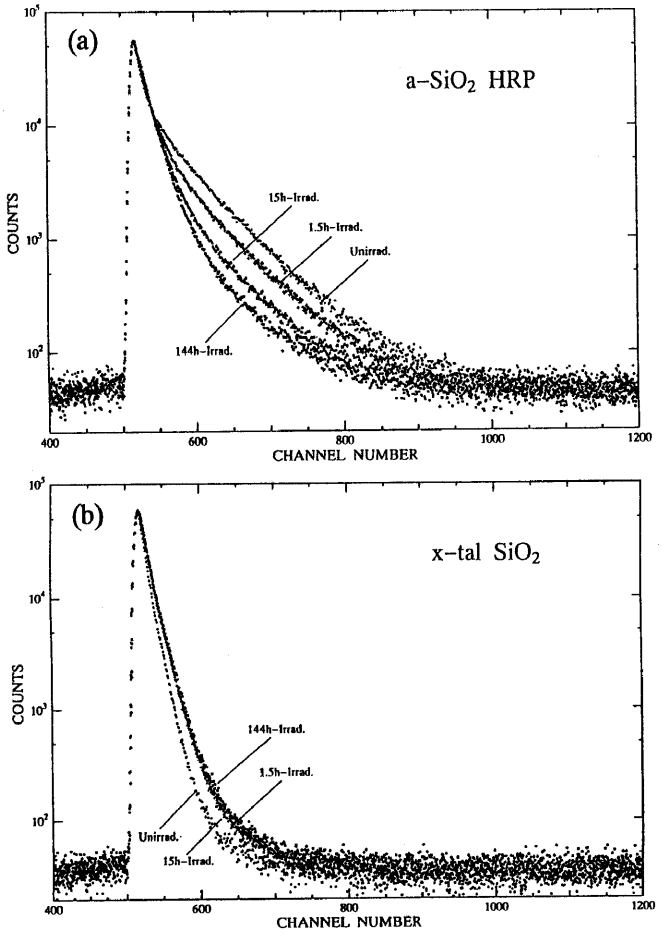

Fig. 1. Positron lifetime spectra for (a) the HRP silica glass, and (b) quartz single crystals. Fast-neutron doses are $8.6x10^{16}$n/cm^2(1.5h), $8.6x10^{17}$n/cm^2(15h), and $8.3x10^{18}$n/cm^2. The time scale is 25.62 ps/channel.

no changes for higher doses. This clearly gives evidence that positrons are trapped at irradiation-induced defects and annihilates there with the observed lifetime of about 0.47 ns. The intensity I_2 increases with fast-neutron dose and attains as high as about 80%, which shows accumulation of the irradiation-induced defects responsible for the lifetime of about 0.47ns. For the crystal sample τ_2 is seen before irradiation and increases slightly with fast-neutron dose (Fig.2(b)). Dannefaer et al. [14] have observed a lifetime of about 0.30 ns due to silicon vacancy or oxygen divacancy in single crystal quartz. Then in the crystal sample τ_2 is considered to be due to vacancies and vacancy-clusters. The slight increase by irradiation reflects agglomeration of irradiation-induced vacancies as expected in case of silicon [15]. Dannefaer et al. [14] measured lifetimes in synthetic fused silica glass and found a long lifetime component of 0.48 ns. Uedono and Tanigawa [17] also observed a long lifetime component of about 0.46 ns in silica glass. These two groups ascribed the long lifetime component to small voids as seen in metals and semiconductors. It should be noted that the long lifetime components very close to that of present study has been observed in unirradiated samples by these two research groups.

It has been found that defect centers in silica glass, usually observed by electron spin resonance(ESR) and optical absorption, are very sensitive to starting materials, preparation method and resulting impurities such as OH groups and metallic ones [9,10,16]. The lifetime parameters for before and after neutron irradiation of 8.6×10^{17} are displayed in Fig.3. Before irradiation τ_3 is found to be nearly independent of the silica samples, whereas τ_2 is strongly dependent on the silica samples, ranging from 0.7 to 1.2ns. The intensity I_2 is about 20% for HRP, OX and ED, and about 30% for ES. The intensity I_3 is very high; it reaches about 60% for HRP and OX, and about 50 % for ES and ED. The fact that τ_2 is sensitively changed with the samples while τ_2 is almost independent of the samples provide direct evidence of the existence of two kinds of positron (or Ps) states for the long lifetime, τ_2 and τ_3, components.

Firstly we consider a physical origin of the τ_3 component. We note the fact that τ_3 of about 1.7 ns is hardly dependent of the samples and further that Dannefaer et al [14] and Uedono and Tanigawa [16] have also observed nearly the same lifetime. Lifetime longer than about 1 ns is usually ascribed to pick-off annihilation of o-Ps. Then above results indicate that $\tau_3$components is due to pick-off annihilation of Ps in intrinsic open spaces in silica glass. To clarify this we have measured ACAR curves for silica glass and single crystals (Fig.4). For the crystal before irradiation a narrow peak at top portion of the ACAR curve is clearly seen and is evidence of self-annihilation of para-Ps (p-Ps) in a "Bloch" state in quartz crystal [18]. The width of the narrow peak, about 1 mrad (FWHM), is well explained by thermal (free) motion of "Bloch" Ps. ACAR curves for the unirradiated glass are very similar to those for synthetic zeolites; Ps is not in free motion but localized at the structural cages (voids) of zeolites [19]. In the same analysis applied to the silica glass we found that the ACAR curves were well decomposed into 2 Gaussians: the narrow components with FWHM of 3.8 $\pm$0.2 mrad and a relative intensity of about 20%, and the broad component with FWHM of 10.3 $\pm$0.3 mrad as shown by dashed lines in Fig.4. The width of the narrow component is much broader than that of the narrow peak seen in the quartz crystal. This is usually evidence of Ps formed in an open space with sub-nanometer size, where Ps is in zero-point motion and the narrow component reflects its momentum distribution of center-of-gravity motion [8,19]. In the previous work on

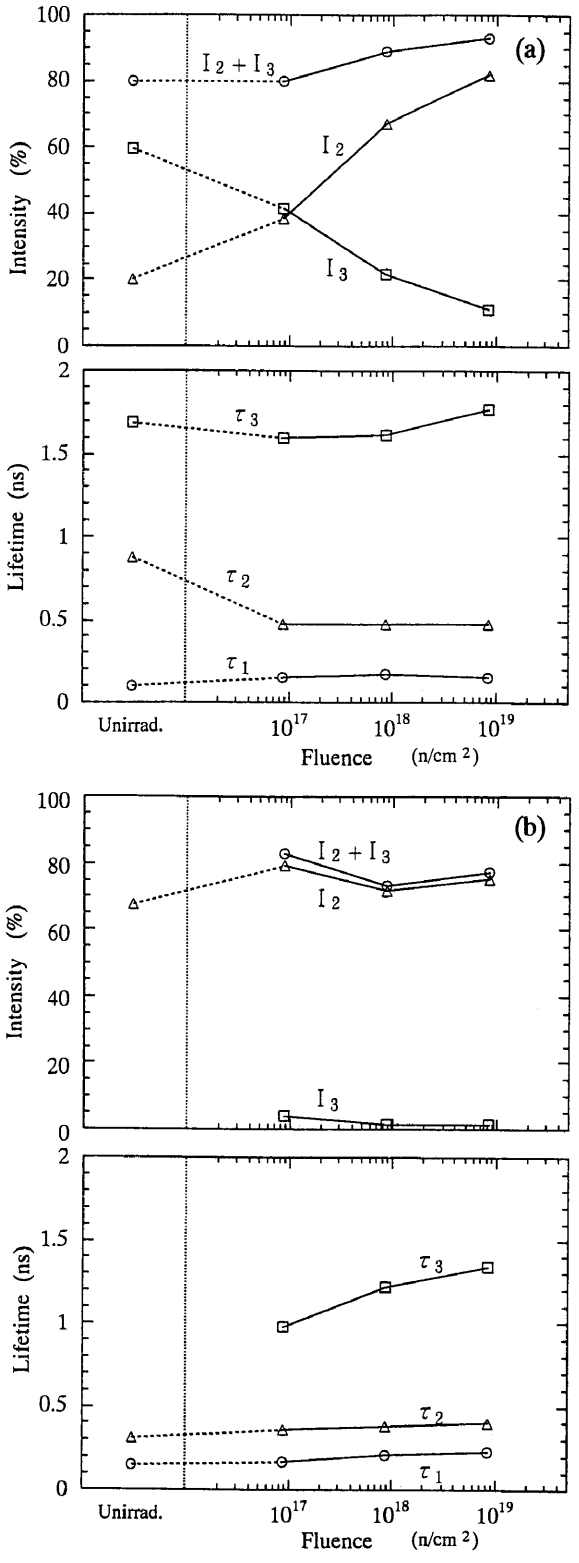

Fig. 2. Fast-neutron dose dependence of positron lifetimes and their relative intensities for (a) the HRP silica glass, and (b) quartz single crystal.

Ps formed in voids in zeolites [19] and in alumina single crystal [8], it has been shown that the Ps momentum distribution is given by a simple model in which Ps is confined in an infinitely deep spherical (IDS) square-well potential with the void radius.

The FWHM (mrad) of the momentum distribution of Ps in the ground state in the IDS square-well potential model is given by $1.66/R_V$(nm). Further the calculated momentum distribution has been shown to be very close to a Gaussian distribution function [8, 18]. The IDS model may be too simple for the present case. In a more realistic model, a free volume model [21,22], R_V is given by

$$R_V = R_0 - \triangle R, \quad (1)$$

and

$$R_0(\text{nm}) = 1.66(\text{nm}) / \Gamma\ (\text{mrad}), \quad (2)$$

where Γ is FWHM width of the narrow component, and $\triangle R$ is a measure of overlapping of Ps wave function with molecules on the void wall and usually assumed to be 0.166 nm. In this model Γ of 3.8 mrad yields 0.27 nm for the void radius. Further in the free volume model, a pick-off annihilation lifetime of o-Ps, τ_p(ns), is given by

$$\tau_p = 0.5\ [\ 1-(R_V/R_0) + (1/2\pi)\sin(2\pi R_V/R_0)]^{-1}. \quad (3)$$

Using this equation with the observed lifetime 1.7 ns, we get 0.26 nm for the void radius. It should be noted that the two independent experiments, namely the longer lifetime τ_3 and the narrow component width yield almost the same void radius. When Ps is formed in the voids, fractions of o-Ps (triplet) and p-Ps (singlet) are 3/4 and 1/4 respectively as expected from the spin-statistics. Usually in solids o-Ps undergoes quenching reaction, such as pick-off annihilation or spin-conversion with surrounding electrons, and decays with much shorter lifetime than that in vacuum (144ns). If almost all o-Ps decay by pick-off annihilation and p-Ps make self-annihilation in the voids, pick-off annihilation lifetime intensity (I_3) is about three times of the narrow component intensity (I_N) as seen, for example, in the HRP and OX samples; I_3 is about 60% and I_N is about 20%. It should be noted that the pick-off annihilation o-Ps gives rise to 2 photon annihilation and hence to the broad component of ACAR curves. These measurements provide direct evidence that Ps is localized in "intrinsic" voids with radius of about 0.3nm in silica glass and gives the τ_3 component and the narrow component. This picture is consistent with a silica glass structure in which an average-distance of silicon with its second nearest neighbor silicon is about 0.5nm and the bond length of Si-O is 0.16nm [23].

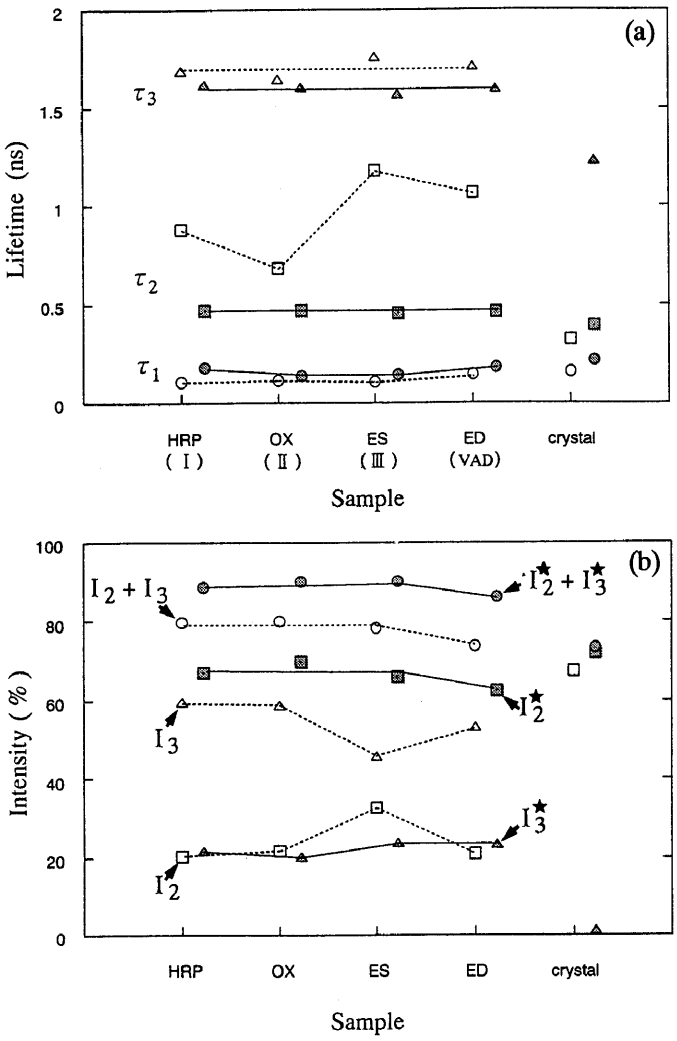

Fig. 3 Variation of the positron lifetime parameters in samples used. Open circles, squares and triangles stand for unirradiated samples, and dotted symbols for neutron-irradiated ones with a dose of $8.3\text{x}10^{17}\text{n/cm}^2$. The intensities I_i for the irradiated samples are asterisked. Results for the crystal are also displayed.

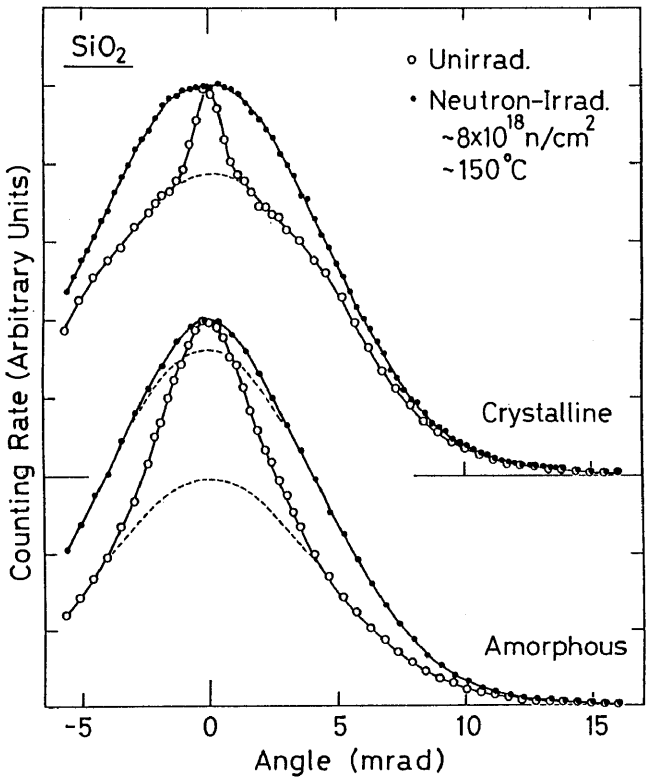

Fig. 4 ACAR curves for quartz crystals and silica glass before and after neutron-irradiation of $8.3\text{x}10^{18}$ n/cm^3. For silica glass dashed lines show the deconvolution into two Gaussian components by a least -square fitting.

These structure contains voids at possible higher-order rings such as 7-membered ring in the CRN networks and provides the voids for Ps formation. Slight decrease by irradiation may be most probably due to some modification of the networks and resulting effective shrinkage of the voids.

Next we consider the τ_2 component. As stated above, τ_2 is strongly dependent on the samples. This suggests that τ_2 is presumably due to positron trapping at impurities or defect centers contained in the silica glass. Seeing that I_3 is about 3 times I_N and that τ_3 and Γ is well represented by the "intrinsic" void radius, the τ_2 component mainly contributes to the broad component but not the narrow component in the ACAR curve. Further it should be noted that p-Ps self-annihilation gives the narrow component in the ACAR curve and the τ_1 lifetime very close to the vacuum lifetime (0.125ps) with a intensity of $I_1 = 1 - I_2 - I_3 = 20\%$, very nearly equal to I_N, as seen in Fig.3. At this stage, however, we could not completely exclude a possibility that Ps compound formation take place at some defects and that this compound gives the τ_2 component and the broad component but no narrow component. This problem must wait a future study.

By neutron irradiation τ_2 decreases to the nearly constant lifetime of 0.47 ns, irrespective of the glass samples and shows no change with neutron dose (Fig.2 and Fig.3). However, I_2 increases with neutron dose, accompanying decrease in I_3. These results clearly show that τ_2 component is due to annihilation of positrons trapped at irradiation-induced defects. We should to point out that these defects are sensitively introduced by irradiation as seen after electron irradiation (Tab.2). A displacement-atom ratio (dpa) by 3MeV electrons with a dose of 1×10^{18} e/cm^2 is estimated to be 1×10^{-4} for a threshold energy of 10 eV for oxygen [10]. For metals and semiconductors 3 MeV-electron irradiation with dose of such a low dpa can induce single vacancies but practically no vacancy-clusters giving lifetime of about 0.5 ns. Then it should be emphasized that after the low-dpa irradiation τ_2 has been shortened to about 0.47 ns. It is unlikely that this lifetime component is due to positron trapping in clusters of irradiation-induced vacancies.

Isochronal annealing behavior of the lifetime parameters is shown in Fig.5:(a) HRP and (b) ES. In the HRP glass τ_1 gradually decreases with annealing temperature and exhibits a small stage around 450°C, while τ_2 shows sluggish increase with temperature. In contrast to these small changes in the lifetimes, the intensities I_2 and I_3 recover successively with temperature; I_2 decreases but I_3 increases in a compensatory way with annealing temperature, and they attain their intensities of unirradiated state after annealing around 800°C. It should be emphasized that I_2 and I_3 exhibit enhanced recovery around 450°C which corresponds to the stage for τ_1. In the ES glass overall recovery of the lifetime parameters is close to that in the HRP. However, τ_2 for ES increases much faster than that for HRP with annealing temperature above 400°C and shows complete recovery around 750°C.

Fundamental irradiation-induced point defects in silica glass are oxygen vacancies, peroxy bridges (globally equivalent to Frenkel defects), peroxy radicals, silicon vacancies and interstitials [10]. Because of its small displacement energy we can assume major defect species are associated with oxygen. These defects usually trap holes or electrons and sometimes relax with small atomic displacement. The resulting defects are the E' center, peroxy radicals (POR), non-bridging oxygen hole center (NBOHC) and so on.

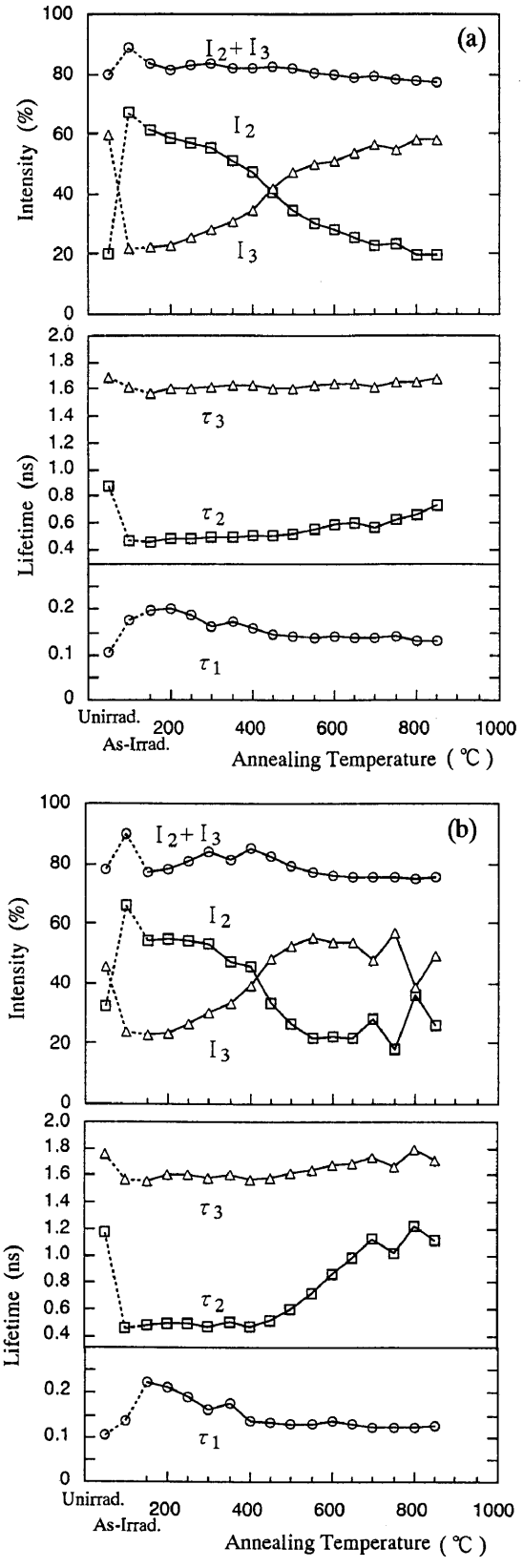

Fig. 5 Annealing behavior of the positron lifetime parameters for (a) HRP, and (b) ES.

These have been extensively studied by ESR and optical absorption measurements [9,10,16]. It has been reported that formation and natures of defects in silica glass are strongly influenced by impurities such as OH groups[9,16]. Griscom showed that after gamma-ray irradiation E' centers recovers completely at about 300°C in low-OH silica glass while at about 400°C in high-OH silica glass, and that POR in low-OH silica glass are stable and their complete recovery occurs 700°C 700°C. Nagasawa et al. made isochronal annealing experiments after gamma-ray irradiation on optical silica glass fiber and showed recovery of E' centers (below 300°C), NBOHC (below 400°C) and POR (below 600°) [24]. Considering these previous studies, we can explain the present results of isochronal annealing as follows. Firstly the small increase by annealing at 150°C and sucsessive recovery up to 400°C in τ_1 are due to E' centers which will trap positrons and gives positron lifetime of 0.30 ns as seen for vacancies in silicon crystals [15]. Successive recovery of I_2 up to about 400°C is also possibly associated with E' center. Secondly further recovery in I_2 can be attributed to POR which is known to anneal out around 700°C. As stated above the lifetime τ_2 increases rapidly with annealing temperature above 400°C in ES, while very slowly in HRP. This difference may be due to OH impurities because OH content is very high in ES but quite low in HRP.

Defects in silica glass have been studied mostly by ESR method which is very sensitive tools for paramagnetic defects but gives no signals for diamagnetic defects. On the contrary positron annihilation method has no such restriction. Positrons are sensitively trapped at vacancy-type defects and voids. Further because of its positive charge, positrons are likely to be attracted to negatively-charged defects and give a large trapping rate. Then positron annihilation is expected to be sensitive to charge states of defects [25]. Then ESR and optical absorption experiments on the same samples for positron annihilation will be very useful to studies of defects in silica glass. Such experiments are now in progress.

4. Summary

Positron lifetime and angular correlation of annihilation radiation (ACAR) measurements were used to detect structural subnano-size voids and defects in fused and synthetic silica glass before and after irradiation with fast neutrons up to 8.3×10^{18} n/cm^2 at about 150°C or 3 MeV electrons up to 1.0×10^{18} e/cm^2 below 50°C. Positronium formation in structural voids with radius of about 0.3 nm is proved by the longest lifetime of about 1.7 ns and the narrow component at the top portion of ACAR curves. The other long lifetime (τ_2) component ranging from 0.7 to 1.2 ns is found to be very sensitively dependent on the u nirradiated glass samples. After neutron-irradiation, however, this lifetime was shortened to about 0.47 ns irrespective to the samples and its intensity was increased with neutron dose. Fast recovery in τ_2 was observed for a synthetic silica containing high hydroxyl (OH) impurities (1200 ppm) above 400°C, but only slow recovery up to 900°C for electrically-fused silica with low OH content (10ppm). These results were compared with previous ESR and optical absorption work, which suggests that the τ_2 component are due to the irradiation-induced peroxy- radicals. Small increase in the short lifetime (τ_1) was observed and interpreted to positron trapping at E' centers and possibly nonbridging oxygen hole centers induced by irradiation.

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