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Investigations on variation of defects in fused silica with different annealing atmospheres using positron annihilation spectroscopy

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ABSTRACT

The laser damage resistance properties of the fused silica can be influenced by the microstructure variation of the atom-size intrinsic defects and voids in bulk silica. Two positron annihilation spectroscopy techniques have been used to investigate the microstructure variation of the vacancy clusters and the structure voids in the polishing redeposition layer and the defect layer of fused silica after annealing in different atmospheres. The fused silica samples were isothermally annealed at 1000 K for 3 h in a furnace under an air atmosphere, a vacuum atmosphere and a hydrogen atmosphere, respectively. The positron annihilation results show that ambient oxygen atmosphere only affects the surface of the fused silica (about 300 nm depth) due to the large volume and low diffusion coefficient of the oxygen atom. However, hydrogen atoms can penetrate into the defect layer inside the fused silica and then have an influence on vacancy defects and vacancy clusters, while having no effect on the large voids. Besides, research results indicate that an annealing process can reduce the size and concentration of vacancy clusters. The obtained data can provide important information for understanding the laser damage mechanism and improving laser damage resistance properties of the fused silica optics.

1. Introduction

Because of its excellent optical properties, fused silica has been widely used for transmissive optics in inertial confinement fusion (ICF) facilities such as the National Ignition Facility (NIF), the Laser MegaJoule (LMJ) and the SG-III laser facility [1–3]. High power laser-induced damage (LID) in fused silica has attracted extensive scientific research interests since it extremely restricts the output of the large laser system. A number of studies have showed that subsurface damage (SSD) includes micro-flaws and mechanical scratches resulting from grinding and polishing treatments are responsible for laser damage initiation at 351/355 nm of fused silica. The removal of these precursors induces an increase of the laser-induced damage threshold [4]. Due to the fabrication process and the impurities in raw materials and the other well-known factors, the actually formed structure will deviated from the perfect random network structure of $Si(O_{1/2})_4$ tetrahedron. Breakage of Si–O covalent bond, loss of oxygen ions and oxygen/silicon interstitials are observed in pure fused silica. The intrinsic microstructural point defects associated with non-bridging oxygen hole centers (NBOHC) and oxygen deficient centers (ODC) have also been found to have direct relations to LID in fused silica [5–7]. The formation and the spatial distribution of the NBOHC and ODC defects about the 3ω laser damage initiation crater in fused silica has been determined by Wong et al. [8]. Kucheyev and Demos [9] have investigated an increase in the concentration of NBOHC and ODC defects after high fluence laser irradiation. Exposure of fused silica to high fluence UV (355 nm) laser leads to Si/O stoichiometry variation as well as to the formation of ODC defects in fused silica [10].

A conventional method of removing point defects is to thermally anneal the material in a furnace. Shen et al. [11] have pointed out that only relatively modest temperatures (approximately 400 °C)

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and times are required to anneal out isolated point defects. The damage threshold on indentation improves from 8 to 35 J/cm^2 after annealing at approximately 750 °C. Thus, investigation of defects variation under different annealing processes is very important in improving laser damage resistance properties of the fused silica optics.

The laser damage resistance properties of the fused silica can be influenced by the microstructure variation of the atom-size intrinsic defects and voids in bulk silica. It is very difficult to non-destructively characterize the tiny atomic size modifications of intrinsic defects and voids in thermally treatment of high quality fused silica. Positron annihilation spectroscopy (PAS) technique is a powerful method and sensitive to study such nanometer-scaled open spaces such as vacancy-type defects in metals, free volumes in polymers, and voids in oxides [12–15]. Positrons tend to localize at open spaces in these materials and annihilate with the surrounding electrons [16–18]. Positron in the silica glass can form positronium (Ps), a bound state of a positron and an electron. Ps exists in two spin states, the short-lived para- positronium (p-Ps) and the long-lived ortho- positronium (o-Ps). Ps is formed for 25% in the antiparallel spin singlet state, p-Ps, and for 75% in the spin triplet state, o-Ps [19]. These states have a vacuum lifetimes of 125 ps and 140 ns, respectively. The p-Ps lifetime in a-SiO_2 was found to be 156 ± 4 ps. This was because that the screening of the Coulomb interaction between the constituent particles by electrons of the medium and that the average distance between the electron and positron in positronium becomes larger than its vacuum value [20]. The o-Ps lifetime is shortened to the order of a few nanoseconds in a void of the matter by the so called pick-off process. In particular, about 80% of positrons form Ps in silica glass (SiO_2); the Ps is localized in the structural subnanovoid in the glass [21]. The pick-off annihilation rate of the *o*-Ps is related to the average size of the subnanovoids in fused silica [22,23]. In recent years, many significant results have been achieved by applying positron annihilation techniques into fused silica defect studies [24–26]. In this work, we attempt to characterize the microstructure defects variation in fused silica under different atmospheres annealing processes. For this purposes, we employed two different PAS techniques: Doppler broadening annihilation spectroscopy (DBS) coupled to a slow positron beam and Positron annihilation lifetime spectroscopy (PALS). The investigation of DBS coupled to a variable-energy slow positron beam is possible to provide a defect distribution from a few nanometers to several microns of the samples by tuning positron implantation energy from few eV to about 20 keV. The PALS is possible to detect defect information about one hundred microns in solid materials. In addition, we present the results of systematically studies on characteristics of microstructure defects. The obtained results can provide important information for understanding the laser damage mechanism of fused silica.

2. Experimental materials and methods

2.1. Sample preparation

In this section, the samples preparation is firstly presented and then, the details of PAS experiments are given. Fused silica samples (JGS1) with a dimension of 15 mm × 15 mm × 1 mm and a surface roughness of less than 1 nm RMS (root mean square) were used as substrates.

Samples of C1 were untreated fused silica samples. The other samples were isothermally annealed at 1000 K for 3 h in a furnace under an air atmosphere (C2), a vacuum atmosphere (C3) and a hydrogen atmosphere (C4), respectively. 5 K/min heating rate was used to the heating process. 5 K/min cooling rate with a temperature range from 1000 K to 500 K and natural cooling below 500 K were applied. The details on the sample preparation are presented in Table 1.

2.2. Positron annihilation spectroscopy (PAS)

PAS experiments were done at room temperature. Two PAS techniques were employed to research defects in fused silica, using radioactive ^{22}Na isotopes as a positron source. The positron–electron annihilation in matter is completely dominated by the emission of two 511 keV photons in the opposite direction in the center-of-mass system. Since the momentum of the positron in the delocalized state is much lower than the electron moment in the solid material, the annihilation parameters such as Doppler-shift provide information on the electronic structure. Firstly, we discuss briefly the Doppler broadening positron annihilation spectra by measuring the 511 keV positron-electron pair annihilation line at room temperature coupled to a slow variable mono-energetic positron beam. The doppler broadening of the line positron annihilation is characterized by so called *S* and *W* parameters, which are defined as the ratio of the central region (511 ± 0.8 keV) and wing region ((511 ± 1.7 to 511 ± 3.1 keV) to the total area of the 511 keV annihilation peak, respectively. The *S*-parameter describes mainly annihilation with low momentum valence electrons. Correspondingly, *W*-parameter describes mainly annihilation with high momentum core electrons. Consequently, an increase (decrease) in *S (W)* parameter indicates the presence of vacancy type defects. The *S* and *W* values were recorded as a function of the positron implantation energy E_p between 0.25 and 20.0 keV using a slow positron beam. For the keV energy of positron, the probability of its implantation depth is given by Refs. [27,28]:

$$P(z,E) = \frac{m\left[\rho\Gamma\left(1+\frac{1}{m}\right)\right]^m z^{m-1}}{(AE^n)^m}\exp\left\{-\frac{\left[\rho\Gamma\left(1+\frac{1}{m}\right)\right]^m z^m}{(AE^n)^m}\right\} \quad (1)$$

where *P* is the probability of positron implantation depth, *A* is the empirical parameters, *m* is the shape parameter, *z* is the depth from the surface in nm, *E* is the positron beam energy in keV and ρ is the material density in g/cm^3. In addition, the empirical parameters and the density of fused silica are equal as follows: $A = 40$, $m = 2$, $n = 1.6$ and $\rho = 2.2$ g/cm^3. Therefore, the positron mean implantation depth in fused silica varied from 0 to 2.2 μm by tuning positron implantation energy from few eV to 20 keV. The positron implantation depth profiles are represented in Fig. 1 for several positron incident energies in the range 1.5–20 keV. Therefore, Doppler broadening annihilation coupled to a slow positron beam is used to detect the surface state of the sample and the surface defect information of the polishing redeposition layer of fused silica.

As a second technique, we carried out the Positron annihilation lifetime experiments. The positron lifetime is measured as the time-interval between the detection of a 1.27 MeV γ-ray, which is emitted almost simultaneously with a positron in the decay of ^{22}Na, and the 511 keV γ-ray emitted as a by-product of the positron annihilation. The positron annihilation rate is proportional to the local electron density. Therefore, the measurement of the positron lifetime allows the distinction between annihilation of “free” positrons and positrons trapped in lattice defects such as vacancies. After subtracting the source and background components, the lifetime spectra were fitted to the following expression:

$$L(t) = \mathrm{Re}\otimes\sum_i \frac{I_i}{\tau_i}\exp(-t/\tau_i) \quad (2)$$

where τ_i is one of the lifetime components of the spectra, and I_i is