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Neutron irradiation and post annealing effect on sapphire by positron annihilation

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ABSTRACT

Sapphire single crystals grown by an improved Kyropoulos-like method are irradiated by fast neutron flux. The irradiated doses of neutron are 10^{18} and 10^{19} n/cm^2. The infrared transmission spectra of sapphire were studied before and after irradiation. The irradiated samples were annealed at 200, 400, 600, 800 and 1000 °C for 10 min in ambient atmosphere. Positron annihilation studies have been carried out before and after neutron irradiation. The experimentally measured positron lifetime in the pristine specimen is 143 ps. There were aluminum vacancies produced in sapphire crystals after neutron irradiation. The positron lifetime increased with the dose of neutron flux. A longer value τ_2 was found after annealing at 600 °C, which indicated vacancies were aggregated with each other. The second long-time component τ_2 has been found to increase with the annealing temperature. There was almost no change in peak position of the CDB spectra after neutron irradiation and isothermal annealing. The chemical environment of core in sapphire did not change greatly after neutron irradiation.

1. Introduction

Sapphire is rhombohedral in cell structure and belongs to the D_{3d} space group with ionic bond. Pure sapphire is colorless and shows little absorption in the ultraviolet–visible range. The physical properties of sapphire, such as its high melting point, hardness, strength and resistance to radiation damage, have led to serious consideration of this material as an insulator for the first wall of a nuclear fusion reactor. A variety of methods have been employed to study the effects of reactor irradiation on sapphire, such as electron spin resonance (ESR), optical absorption (OA) and thermoluminescence (TL). The positron annihilation gamma-rays depend strongly on the local electronic environment of the annihilation sites (Ganguly et al., 2006). Positron annihilation spectroscopy (PAS) can yield detailed information regarding both the electron density and the electron momenta in the region from which the positron annihilates (Siegel, 1980). PAS can be used to identify the nature of Al vacancies where positrons are trapped and defects in polyaniline (Muthe et al., 2009; Nam et al., 2009). The coincidence Doppler broadening (CDB) enables us to identify the chemical element whose electron annihilates with the positron, by measuring the electron momentum distribution in the high-momentum region, given by the positron annihilation with the inner orbital electrons (Nagai et al., 2003).

Studies of the defects produced in sapphire date from the in-reactor irradiations of Levy and Dienes reported in 1954 (McHargue et al., 2009). Sapphire:Mn irradiated by neutron flux has been studied by EPR spectra (Libin et al., 1996). The change of positron lifetime with the temperature in sapphire was studied. It indicated the change from $\tau = 150$ ps at 296 K to $\tau = 142$ ps at 900 K (Forster et al., 1992). There has been considerable interest in effects of high-dose irradiation on sapphire because of their potential use in future thermonuclear reactors (Tanabe et al., 1994). However, the defects evolution after neutron irradiation and annealing were still unclear. Radiation-resistant sapphire and composites based on it are promising structural and electro-insulating materials for atomic power station and reactor and thermonuclear installations (Abdukadyrova, 2003). Furthermore, radiation effects of sapphire crystal depend on not only irradiation conditions but also on its chemical composition and preparation process (Wang et al., 2009a, 2009b). Sapphire crystal grown by an improved Kyropoulos-like method was irradiated by fast neutron flux. This paper presents on the influence of neutron irradiation to the dose level of 10^{18} and 10^{19} n/cm^2 by positron annihilation. The subsequent effects of annealing sapphire after irradiation were studied.

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2. Materials and methods

2.1. Materials

Sapphire single crystals were grown by an improved Kyropoulos-like method. A detailed growth process of sapphire crystal was described elsewhere (Xu et al., 2007). Impurity analyses were performed using proton-induced X-ray emission (PIXE). The results indicated Ti, Cr, Mn, Fe, Co and Ni were not found in the specimen (Wang et al., 2009a, 2009b). Samples were prepared by cutting the as-grown sapphire crystal perpendicularly to optical axis into small pieces with the size of 10 mm × 10 mm × 2 mm and final both-sides polishing.

2.2. Fast neutron irradiation

Fast neutron irradiation was carried out in light water reactor, where a neutron flux in the reactor core was about 6.6×10^{12} $\text{cm}^{-2}\,\text{s}^{-1}$ with an average energy of about 10 MeV and the fluence was up to 10^{18} and 10^{19} n/cm^2. Specimens were individually wrapped in Al foil, which removes low energy thermal neutrons when irradiated and placed in a dry air-filled radiation basket adjacent to the submerged reactor core. The samples were water-cooled during irradiation. The water temperature was about 40–45 °C and the temperature of samples were about 70–80 °C. Following irradiation, samples were washed with 8 M HNO_3, 48%HF and distilled water to remove surface contamination derived from the Al foil. The heat treatment temperature was from 200 to 1000 °C. Specimens were heated at 10 °C/min and equilibrated at the final temperature for 10 min.

2.3. IR spectra measurement

The infrared transmission spectra of sapphire were studied before and after irradiation. IR spectra were recorded on a Perkin–Elmer spectrum one FT-IR spectrometer in the 1600–4000 cm^{-1} region at room temperature.

2.4. Positron lifetime and CDB spectra measurement

Positron lifetime and the CDB spectra were recorded at room temperature using a fast-low coincidence spectrometer consisting of two BaF_2 detectors for a resolution of 186 ps. A positron source 7.4×10^{5} Bq of ^{22}Na deposited on a thin Kapton foil of 7 μm in thickness was sandwiched between the two identical specimens for positron lifetime measurements. The CDB spectra were measured by using two HPGe detectors. The energies of annihilating gamma-ray pairs were simultaneously recorded by the two detectors located at an angle of 180° relative to each other. The overall energy resolution was 1.33 keV, which corresponds to the momentum resolution of $5.2\times10^{-3}m_0c$. The sample–detector distance is 36 cm and the strength of ^{22}Na positron source is 1.78 MBq. Total counts of more than 2×10^{7} for each of the measurements were accumulated. To highlight the element-specific difference between the measured CDB spectra, we obtained ratio curves of the spectra, which are given by the ratio of the spectra to that for a reference sample. The details are described elsewhere (Wang et al., 2009a, 2009b).

3. Results and discussion

3.1. IR spectra

Fig. 1 contains the IR transmission spectra for sapphire before and after neutron irradiation, the analysis revealed a trend towards the irradiation producing slight decrease in transmission. It is likely that much of the decrease in transmission is induced by scattering (Regan et al., 2001). However, the transmission was still above 80% in the region 2140–4000 cm^{-1}, which had no impact on its performance as infrared window.

3.2. Positron lifetime spectrum

The results of positron annihilation spectroscopy are illustrated in Table 1 for both the as-irradiated and post-annealed sapphire samples. The experimentally measured positron lifetime in the pristine specimen prepared by an improved Kyropoulos-like method is 143 ps, which was less than that reported (150 ps) in Hasegawa et al. (1994) and Xu et al. (1997). After neutron irradiation, the positron lifetimes τ_1 were 165 and 177 ps, under the fluence of 10^{18} and 10^{19} n/cm^2, respectively.

The positron lifetime measurements reflect the change in local electron density around vacancy. In pristine sapphire, the positron lifetime is related to the positron trapped in the as-grown state. The as-irradiated spectrum was well fitted with one lifetime component. The τ_1 component is ascribed to saturation trapping of positrons in single vacancies and small vacancy complexes, and τ_2 component results from those annihilating in large vacancy agglomerates. The intensity I_2 denoted the relative number of

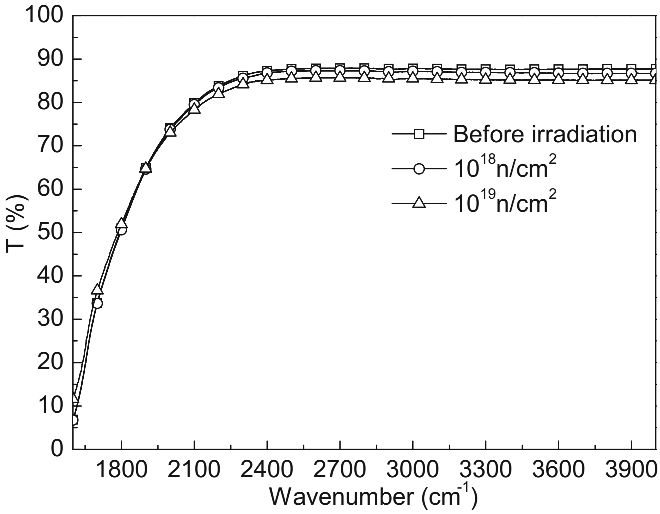

Fig. 1. FT-IR spectra for neutron irradiated sapphire.

Table 1
Positron annihilation lifetimes and their relative intensities in sapphire crystals.

Specimens	τ_1 (ps)	τ_2 (ps)	I_1 (%)	I_2 (%)
pristine	143		100	
10^{18} n/cm^2 irradiation	165		100	
10^{19} n/cm^2 irradiation	177		100	
10^{19} n/cm^2 and 200 °C annealing	171		100	
10^{19} n/cm^2 and 400 °C annealing	174		100	
10^{19} n/cm^2 and 600 °C annealing	150	255	59.5	40.5
10^{19} n/cm^2 and 800 °C annealing	163	356	75.4	24.6
10^{19} n/cm^2 and 1000 °C annealing	160	381	62.3	37.7