Voids in α-Al_2O_3 Irradiated by 85 MeV ^{19}F Ions

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Abstract The voids in α-Al_2O_3 irradiated by 85 MeV ^{19}F ions of 5.28×10^{16} cm^{-2} have been observed during the post-annealing by a positron annihilation lifetime technique for the first time. The voids start to appear at 450℃. The void radius keeps nearly constant at ~0.29nm and the number of voids increases with increasing the annealing temperature from 550℃ to 750℃. Afterwards, the radius of voids increases rapidly with the annealing temperature and reaches 1.10nm at 1050℃.

Key words heavy ion irradiation, thermal annealing, void, positron annihilation

1 Introduction

α-Al_2O_3 is an important material used for the first wall of fusion reactors and the windows of laser and solar cells and for monitoring neutron radiation damage. The radiation effects in α-Al_2O_3 irradiated by high-fluence neutrons, especially void nucleation phenomenon, is a currently interesting topic. Iwata, one of the authors, predicts the void formation in α-Al_2O_3 and some other materials when the irradiating neutron fluence and post irradiation annealing temperature both exceed the certain values or thresholds and the voids grow with the increasing of the annealing temperature above the threshold. The α-Al_2O_3 irradiated by $E_n \geqslant 1$ MeV neutrons of 1.5×10^{20} cm^{-2} and 3×10^{20} cm^{-2} has been studied. No voids were detected up to the post irradiation annealing temperature of 1200℃ on the α-Al_2O_3 irradiated by neutrons of 1.5×10^{20} cm^{-2}. Nevertheless, the voids with a radius of 0.7 nm were observed on the α-Al_2O_3 irradiated by neutrons of 3×10^{20} cm^{-2} after the post irradiation annealing at 850℃[1]. In order to determine the temperature threshold and to understand the detailed evolution of void nucleation with the annealing temperature, the present work was motivated to study the dependence of void nucleation on the post irradiation annealing temperature for the α-Al_2O_3 irradiated by 85 MeV ^{19}F ions of 5.28×10^{16} cm^{-2} that is equivalent to the 3×10^{20} cm^{-2} neutron irradiation. The created voids were examined by a positron annihilation lifetime technique that is a powerful tool for investigating the voids in metals, insulators and other materials, especially for studying the early stage of void nucleation[2-5].

2 Experiment

The 11mm × 11mm × 1mm α-Al_2O_3 R-cut single crystal samples were used in the experiment. The samples were irradiated by 85 MeV ^{19}F ions from the HI-13 tandem accelerator. The irradiation fluence was 5.28×10^{16} cm^{-2} that is equivalent to the neutron fluence of 3×10^{20} cm^{-2}. The irradiation was performed at room temperature.

The post-irradiation annealing was conducted under nitrogen atmosphere for 40 minutes from 100℃ to 1050℃ in steps of 50℃.

The detection of vacancies and voids were carried out

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by a positron lifetime technique. The positron lifetime spectra were measured at room temperature after irradiation and annealing at different temperatures by means of a conventional BaF_2 fast-fast coincidence positron lifetime spectrometer with a time resolution of 210ps. Two identical samples were arranged as a sandwich with a 0.8 MBq positron source in the center. The lifetime spectra each contained 1.5×10^6 counts and were analyzed with an LT program[6]. Besides the source components, the measured lifetime spectra were fitted with two or three lifetime components. The fitting variance was all less than 1.3.

3 Results and discussion

The lifetime spectra were well fitted with two lifetime components below 450℃ and, the third long-lifetime component had to be added above 450℃, otherwise, the fitting variance was unacceptable. The obtained lifetimes τ_1, τ_2 and τ_3 and their intensities I_2 and I_3 ($I_1 + I_2 + I_3 = 1$) are shown in Fig.1 as a function of the post irradiation annealing temperature.

The bulk lifetime τ_b of positrons in α-Al_2O_3 is 150 ps[1]. It can be seen from Fig.1 that the measured lifetime τ_1 is due to positron annihilation in the bulk and is almost independent of the annealing temperature. No changes were observed in τ_2 up to the annealing temperature of 450℃, afterward, τ_2 increases with the increasing of annealing temperature. τ_2 is ascribed to the lifetime of positrons trapped at the Al vacancy clusters (V_{Al}-V_{Al}) and Al-O vacancy clusters (V_{Al}-V_O) below 450℃. The lifetime of positrons trapped at the inner surface of voids is about 500 ps[7,8]. Therefore, above 450℃ τ_2 is assumed to be a weighted average lifetime of the positrons trapped at the above-mentioned clusters and those trapped at the inner surface of voids described below. The increase of τ_2 demonstrates the growing of the clusters. A long lifetime τ_3 starts to appear at 450℃. τ_3 increases to 870 ps at 550℃, then keeps constant at a value of ~ 880 ps up to 750℃, and at higher temperatures, τ_3 increases rapidly with the increasing of annealing temperature and reaches 2430 ps at 1050℃. The intensity I_3 increases first with the annealing temperature increase, arrives at a maximum of ~ 3.5 % at 750℃ and then starts decreasing at 950℃. From its value τ_3 is ascribed to the annihilation of ortho-positronium (Ps) formed in voids[1].

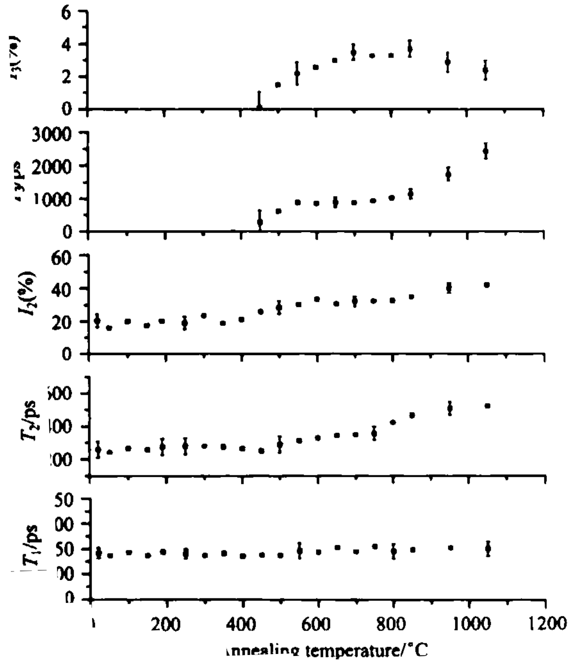

Fig.1. Positron annihilation lifetime and its intensity as a function of annealing temperature in α-Al_2O_3 irradiated by 85 MeV ^{19}F ions of 5.28×10^{16} cm^{-2}.

The Ps self-annihilation lifetime is related to the void size, and the radius of voids can be calculated from the measured lifetime[9]:

$$R_v = R_o - R_w,$$

where R_v is the void radius, R_w is the overlapping of P_s wave function with molecules on the void wall, and R_o is related to the lifetime of P_s in the ground state in the infinity deep spherical (IDS) square-well potential model by

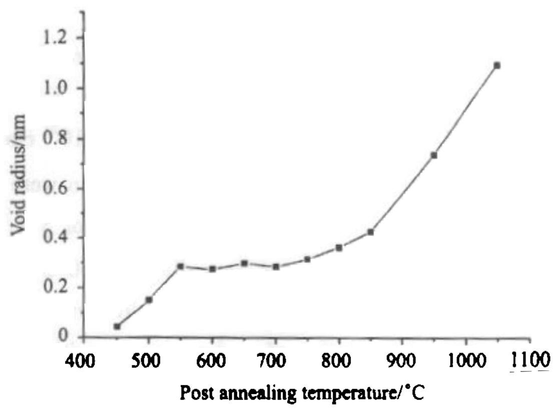

Fig.2. Dependence of void radius observed in α-Al_2O_3 irradiated by 85 MeV ^{19}F ions of 5.28×10^{16} cm^{-2} on annealing temperature.

$\tau = 1.92R_0$. Fig.2 shows the variation of the void radius with the post-annealing temperature in the α-Al_2O_3 irradiated by 85 MeV ^{19}F ions of 5.28×10^{16} cm^{-2}. It can be seen that the void radius first increases with the annealing temperature, keeps nearly constant at ~ 0.29 nm in the temperature region between 550℃ and 750℃, and increases with temperature rapidly and reaches 0.74 nm and 1.10 nm at 950℃ and 1050℃, respectively.

The Al atoms are displaced by the irradiating ^{19}F ions in α-Al_2O_3. The produced holes are captured by O ions adjacent to Al vacancies to form V-center. This V-center has 3 holes shared by O ions surrounding the Al vacancy, which is the void nucleation center[10]. No long lifetime was observed in the as-irradiated α-Al_2O_3. During thermal annealing Al vacancies freely migrate, leading to the formation of voids with the V-center as a nucleation center. The present results show that voids could not be formed until the annealing at 450℃ that is the threshold of annealing temperature. On the other hand the void formation needs a certain number of Al vacancies. We did the positron lifetime measurements on the α-Al_2O_3 irradiated with $E_n > 1$ MeV neutrons to a fluence of 1.5×10^{20} cm^{-2}. No voids were observed in it up to 1200℃. In the present experiment the α-Al_2O_3 was irradiated by 85 MeV ^{19}F ions of 5.28×10^{16} cm^{-2}, which is equivalent to the $E_n > 1$ MeV neutron irradiation to a fluence of 3.0×10^{20} cm^{-2}, and the voids were detected above 450℃. As shown in Fig.1, τ_3 is almost independent of annealing temperature between 550℃ and 750℃, while its intensity I_3 increases. Assuming the intensity I_3 is proportional to the number of voids, we can conclude that from 550℃ to 750℃ thermal energy is mainly used to create voids and the void number increases with the increasing of annealing temperature, while the value of τ_3 or void size is steady. After that τ_3 increases rapidly with increasing annealing temperature, indicating the growing of voids.

In summary, the detailed evolution of void nucleation with the post-irradiation annealing temperature has been determined on the α-Al_2O_3 irradiated by 85MeV ^{19}F ions of 5.28×10^{16} cm^{-2} by a positron annihilation lifetime technique for the first time. The void nucleation starts at the post irradiation annealing temperature of 450℃. From 550℃ to 750℃ the radius of created voids does not change with temperature and takes a value of 0.29 nm, while the number of voids increases with the increasing of annealing temperature. Afterwards the void radius increases rapidly with increasing the annealing temperature and reaches 1.10nm at 1050℃. The present experiment also demonstrates the applicability of positron lifetime technique in investigating irradiation-induced voids, especially the early stage of void nucleation in metals, insulators etc.

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85MeV ^{19}F 重离子辐照 α-Al_2O_3 的空洞研究*

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摘要　$5.28\times10^{16}cm^{-2}$ 85MeV ^{19}F 辐照的 α-Al_2O_3 中,在其热退火过程中采用正电子湮没方法首次观察到了空洞. 450℃退火开始产生空洞,550℃到750℃空洞半径约为0.29nm不随温度变化,但浓度随温度增加而增加;高于750℃,空洞半径随温度升高迅速增大,1050℃时空洞的半径达1.10nm.

关键词　重离子辐照　热退火　空洞　正电子湮没

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