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Effect of annealing on thermal diffusivity in ceramics irradiated by electrons and neutrons

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Abstract

α-Al_2O_3, AlN, β-Si_3N_4, and β-SiC ceramic specimens were irradiated by a 30 MeV electron beam up to 0.01 dpa at 80 ˚C, and they started to recover thermal diffusivity with annealing from 300–500 ˚C. In previous studies, similar ceramics were heavily neutron-irradiated in a fast reactor, JOYO, and the neutron-irradiated β-Si_3N_4 and β-SiC also showed recovery above 400 ˚C, though they were irradiated at 502 ˚C or 738 ˚C. This result is explained by the hypothesis that a 'primary' irradiation occurred at the set temperature, and after that, a 'secondary' irradiation occurred at a lower temperature during the shutdown process of the reactor. In contrast, the neutron-irradiated α-Al_2O_3 and AlN did not show such unexpected recovery, but showed recovery above 800-900 ˚C. This difference is explained on the basis of structural models of dislocation loops namely, the 'pileup' model and the 'nano-partition' model.

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1. Introduction

Thermal diffusivity is one of the most important characteristics of ceramic materials used in fusion reactors. In ceramics, unlike metals, heat is mainly carried by phonons. Phonon transportation is obstructed by lattice defects, especially vacancies. It has been reported that neutron-irradiated specimens show severe degradation in thermal diffusivity because of irradiation induced defects [1, 2].

In previous studies [1, 3, 4], thermal diffusivity or conductivity during irradiation was estimated from post-irradiation measurements using several assumptions. One assumption is that the amount of 'transient defect' during irradiation is small enough . Here, 'transient defect' is considered that it exists for a very short time during irradiation. Dynamic effects due to 'transient defects' during irradiation are an important issue to understand the processes that produce defects [5, 6]. However, only a few in-situ experimental studies have been conducted [1, 7, 8] except investigations of electron excitation like radiation induced conductivity (RIC) [9–11] or luminescence measurements [12, 13]. Hence, an in-situ measurement of positron annihilation lifetime (PAL) during ion-beam irradiation is now on early practice [1, 7], and the results validate the assumption to a large extent.

However, another assumption is required to enable the estimation mentioned above, that is, the irradiation temperature is constant. That means that in a test irradiation environment, the temperature is kept constant during irradiation, while a material used in a real-life fusion reactor encounters a constant heat load. Specimens that are neutron-irradiated heavily contain many types of defect, such as interstitial atoms, vacancies, dislocation loops, voids, and so on. These defects interact with each other and their amount and size are changed

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during irradiation at elevated temperature. Irradiation temperature changes the reaction velocity, accordingly it changes the distribution of defects. Then the modified distribution of defects changes thermal diffusivity of the specimen. Under a constant heat flux environment, the temperature of a specimen is determined by its thermal diffusivity. Therefore, these correlations make it difficult to fix the temperature of the specimen during irradiation.

To resolve this problem, an analysis based on reaction kinetics is required. In this analysis, the reaction between interstitial atoms, vacancies, dislocation loops, and voids is explained by their density and reaction velocity. In the simplest model, the correlation is described as below.

- $I + I_n \rightarrow I_{n+1}$
- $I + V \rightarrow 0$
- $V + V_n \rightarrow V_{n+1}$

where I: interstitial atom, I_n: cluster of interstitial atoms that contains n atoms, V: vacancy, V_n: cluster of vacancies that contains n vacancies, and 0 represent no defect. In ceramics, a cluster of interstitial atoms is usually an interstitial dislocation loop, and a cluster of vacancies is usually a void. During neutron irradiation, these reactions conflict each other, so it is difficult to determine the velocity of each reaction. In previous studies, specimens that were neutron-irradiated heavily were annealed to study their recovery behavior [1, 14–18]. The recovery of thermal diffusivity was compared to that of macroscopic swelling and it partly represented the distribution of these defects. However, the discussions were qualitative and not quantitative.

On the other hand, electron irradiation induces a simple Frenkel pair. In this study, 30 MeV electron accelerator KURRI(Kyoto University Research Reactor Institute)-Linac was used to induce point-defects in bulk ceramic specimens and recovery behavior with annealing was compared to that of the heavily neutron-irradiated specimens.

The maximum energy of a primary knock-on atom (PKA) during the 30 MeV electron irradiation is obtained as $E_{\mathrm{p.max}} = 2E(E + 2m_0c^2)/Mc^2$ where $E_{\mathrm{p.max}}$: maximum PKA energy, E : incident energy of electron, m_0 : rest mass of an electron, c : velocity of light and M : mass of a target. When the mass target is 16 amu, $E_{\mathrm{p.max}}$ is calculated as 125 keV. However, the average PKA energy was only 225 eV even by 30 MeV incident electrons, and the Kinchin-Pease model gives a displacement damage function as 3.75. This result shows that even using the 30 MeV electron irradiation, induced defects are mostly point defects like the Frenkel pair. Furthermore, the electron-irradiation in this study was performed at 80 ℃ in cooling water.

The neutron irradiation was performed up to several tens dpa above 500 ℃, and many interstitial dislocation loops were observed by transmission electron microscopy (TEM) [15, 17, 17–19]. This different defect distribution leads to different behavior with annealing.

2. Experimental

In this study, α-Al_2O_3, AlN, β-Si_3N_4 and β-SiC were selected as typical ceramic materials. It is well known that β-SiC is the most important material for nuclear applications, and α-Al_2O_3 is used as an insulator. In addition, AlN was selected because of its high thermal diffusivity before irradiation, which makes it easy to estimate the effect of induced defects. β-Si_3N_4 showed higher thermal diffusivity and lower swelling than β-SiC after high neutron irradiations [20], and this makes β-Si_3N_4 a candidate material for nuclear applications. Properties of the non-irradiated specimens used for electron-irradiation are listed in Table 1. The specimens used for neutron-irradiation were different from specimens used for electron-irradiation, and they were reported in detail in a previous work [1].

Electron irradiation on ceramic specimens was performed using the 30 MeV electron Linac at the Kyoto University Research Reactor Institute. The irradiation was operated in pulse mode with a peak current of 400 mA for 4 μs, and the frequency was 70-80 Hz, so beam current averaged about 110-140 μA in 2 cm^2. The range of the 30 MeV electron beam in ceramics is deeper than 5 cm, so the defects were induced equally allover the ϕ 10 mm×0.5 mm specimen with a dose of 1.0×10^{-2} dpa. Specimens were irradiated in flowing cold water in an aluminum chamber and irradiation temperature was kept at about 80 ℃.

The neutron irradiations were performed in the Japanese experimental fast reactor JOYO, and capsules of the specimens were enclosed in the Core Material Irradiation Rigs, CMIR-4. In the rigs, irradiation temperature of a specimen was determined by gamma heating of the specimen, sodium flow in the compartment, and heat conduction of the double-walled tubes. After the irradiation, the irradiation temperature of each capsule was confirmed using a Thermal Expansion Difference temperature (TED) monitor. Each capsule contained α-Al_2O_3, AlN, β-Si_3N_4 and β-SiC specimens, and they were irradiated under the same condition. The capsule