



Analysis of recovery process of low-dose neutron irradiation-induced defects in silicon nitride-based ceramics by thermal annealing



Areerak Rueanngoen^{a,*}, Koumei Kanazawa^a, Masamitsu Imai^b, Katsumi Yoshida^b, Toyohiko Yano^b

^a Department of Nuclear Engineering, Graduate School of Science and Engineering, Tokyo Institute of Technology, O-okayama, Meguro-ku, Tokyo 152-8550, Japan

^b Research Laboratory for Nuclear Reactors, Tokyo Institute of Technology, O-okayama, Meguro-ku, Tokyo 152-8550, Japan

ARTICLE INFO

Article history:

Available online 12 August 2014

ABSTRACT

Two kinds of silicon nitride ceramics consisting of different polymorphs were neutron-irradiated up to 8.5×10^{24} n/m² ($E > 0.1$ MeV) at 563 K, and their annealing behaviors were compared to those of previously reported SiAlON polymorphs subjected to the same irradiation condition. The macroscopic length change of α - and β -Si₃N₄ and α - and β -SiAlON were 0.11%, 0.06%, 0.12% and 0.14%, respectively. Based on swelling data and microstructural observations, the low dose neutron irradiation-induced defects in silicon nitride-based ceramics were considered to be primarily point defects. In order to investigate the kinetics of defect recovery, these irradiated specimens were isothermally and isochronally annealed continuously up to 1473 K. Macroscopic length change decreased gradually with increasing annealing temperature. Recovery curves of isochronal annealing of α -Si₃N₄ and α -SiAlON were similar, and those of β -Si₃N₄ and β -SiAlON were also similar. The recombination rate constant as a first-order reaction increased with the increasing of the isothermal annealing temperature. A two-stage recovery process was considered between the irradiation temperature and 1473 K. The activation energies at higher temperatures were almost double those at lower temperatures in both Si₃N₄ and SiAlON. At lower temperatures range the recovery should occur by annihilation of close-spaced Frenkel pairs. On the other hand, at higher temperatures, the recovery process may be governed by the annihilation of separated Frenkel pairs. In addition, the activation energies for defect recovery in Si₃N₄ were larger than defects in SiAlON. Recovery characteristics of α - and β -phases were different in both crystals that are suggested to be due to differences in crystal structures.

© 2014 Elsevier B.V. All rights reserved.

1. Introduction

Dimensional stability, and mechanical and thermal property stability under neutron irradiation are key factors for selecting materials for nuclear applications. At the beginning of the material survey, silicon nitride-based ceramics are one of the candidates for fusion applications, such as magnetic insulators, radio frequency transmission windows, and near first-wall components [1–6]. At present, these ceramics are not candidates for use as primary structural materials in fusion reactors, which may be due to the shortage of experimental studies. However, they are still of interest for fusion devices due to their excellent mechanical and thermal properties. Abe et al. [6] and Ioki et al. [7] reported that the rotor blades of turbo molecular pumps (TMP) made from silicon nitride have been developed for a fusion device. The ceramic TMP system will be subjected to a neutron flux as high as $\sim 10^{17}$ n/m²s, when installed in a diverter module near the core plasma. In recent

studies, it was reported that silicon nitride possessed greater dimensional stability than other ceramics such as SiC, Al₂O₃ and AlN after neutron irradiation [8,9], as will be mentioned later.

High energy neutron-irradiation induces several property changes in materials. The population and distribution of interstitials and vacancies and the population of extended defects and defect clusters depend on irradiation temperature, neutron flux, neutron energy, fluence and materials. The short-range and long-range recombination of point defects are important processes affecting the recovery of properties by thermal annealing for low dose irradiated materials. Annealing can remove some irradiation-induced defects, and thereby restore the physical, thermal, and mechanical properties. Annealing behaviors of neutron irradiated ceramics were reported in a relatively large number of papers [8,10–15], but studies on annealing behavior of silicon nitride ceramics are still limited.

Swelling of four kinds of ceramics after neutron irradiation up to 2.8 – 7.3×10^{26} n/m² at 775–1039 K was reported by Akiyoshi and Yano [9,11]. They reported that α -Al₂O₃ and AlN showed larger swelling than β -SiC and β -Si₃N₄. This difference was largely

* Corresponding author.

E-mail address: areerak_k@yahoo.com (A. Rueanngoen).

contributed to differences in their crystal structures. In the case of Al-based materials, the closest-packed plane is the (0001) basal plane and interstitial loops were observed to accumulate on the basal planes without restriction of number density. The resulting anisotropic swelling causes inter-granular micro-cracks, and causes large volumetric swelling. In contrast, for Si-based materials, closest-packed planes are induced cubic {111} and hexagonal {10 $\bar{1}$ 0} planes of β -SiC and β -Si₃N₄, respectively. Interstitial loops form under the restriction of growth or nucleation of new loops, due to the topological interaction of extra planes. This situation restricts the accumulation of individual interstitials, and then forced recombination with vacancies may occur. Therefore, volumetric swelling of these crystals is limited. Moreover, the nature of atomic bonding of these four kinds of compounds is different; SiC and Si₃N₄ are mainly covalently bonded while Al₂O₃ and AlN are between ionic and covalent bonding, which may affect the overall irradiation response.

Macroscopic length changes of β -Si₃N₄ irradiated up to fluences of 2.8×10^{26} n/m² at 753 K and 4.2×10^{26} n/m² at 1003 K were not completely recovered by thermal annealing up to 1773 K [13]. It was assumed that, during thermal annealing, formation of voids or difficulty of recovery of point defects/loops would be the reason for incomplete recovery. This assumption was supported by HREM observation [16], i.e., a dense population of dislocation loops were observed in heavily irradiated specimens. However, up to now, the neutron irradiation response and recovery behavior of silicon nitride-based ceramics are still unclear. In order to study their recovery behavior, the radiation response of low-dose neutron-irradiated materials is explored here is easier to clarify the stability of point defects under these conditions.

In our previous work [17], macroscopic length recovery of α - and β -SiAlON by isochronal and isothermal annealing was measured to clarify defect-recovery behavior during thermal annealing. We reported then that recovery processes were governed by the recombination of different types of Frenkel pairs dependent on materials polymorph. In this study, two polymorphs of silicon nitride ceramics were selected to study the recovery behavior of defects in different crystal structures, and to compare to that of α - and β -SiAlON. They were neutron-irradiated up to a fluence of 8.5×10^{24} n/m² ($E > 0.1$ MeV) at 563 K, at the same level as in the previous studies [17,18]. From transmission electron microscope observations of these four specimens after irradiation and after post-irradiation annealing, defect-clusters such as interstitial dislocation loops or voids were not observed [19], so that irradiation-induced defects were considered to be mostly point defects. The macroscopic length recovery was measured in-situ by continuous thermal annealing up to 1473 K in order to clarify the recovery process. Finally, the differences in length recovery tendencies were discussed based on crystal structure differences and similarity between the various polymorphs.

2. Experimental procedures

Two kinds of hot-pressed silicon nitride ceramics were used in this study, α - and β -Si₃N₄ were commonly prepared from α -Si₃N₄ (89.0 wt%), MgO (3.0 wt%), SiO₂ (3.0 wt%) and Y₂O₃ (5.0 wt%) powder mixtures, and sintered at different temperatures [18]. For comparison, data for two polymorphs of SiAlON were referenced from the previous study [17]. The z-value of β -SiAlON was 1.78 (Si_{4.22}Al_{1.78}O_{1.78}N_{6.22}). α -SiAlON contained strontium and yttrium as charge compensating ions, and the chemical formula was (Sr_{0.7}, Y_{0.3})_{0.56}Si_{9.75}Al_{2.25}O_{0.75}N_{15.25} [17]. Hot-pressed specimens were cut into rectangular bars with sizes of ~ 2 mm $\times$ ~ 4 mm $\times$ ~ 25 mm. These specimens were irradiated in the Japan Materials Testing Reactor (JMTR). All specimens were concurrently irradiated

up to a fluence of 8.5×10^{24} n/m² ($E > 0.1$ MeV) at 563 K. The temperature of these specimens during irradiation was measured with a thermocouple, which was placed in the capsule near the specimens. The macroscopic length changes in term of $\Delta L/L$, of α - and β -Si₃N₄ and α - and β -SiAlON were 0.11%, 0.06%, 0.12% and 0.14%, respectively [18].

In order to investigate the kinetics of the defect-recovery processes, specimen length changes were continuously isothermally and isochronally annealed up to 1473 K using a precision dilatometer (DIL 402C, NETZSCH). The specimens for the dilatometer measurement were rectangular bars with sizes of ~ 2 mm $\times$ ~ 2 mm $\times$ ~ 25 mm. The irradiated specimens were annealed up to 1473 K with a step-heating temperature interval of 50 K in helium. The specimen temperature was kept constant for 3 h of each temperature step. The heating rate between each step was 5 K/min. The length change measurement was corrected by measuring a single crystal Al₂O₃ as a suitable reference material. Unirradiated specimens of each lot were measured with the same temperature profile, to establish magnitude of thermal expansion. The net recovery was obtained by subtracting this data from the data of the neutron-irradiated specimens. The minimum detection sensitivity of present dilatometer was 10 nm, corresponding to 0.00004% of the specimen with a length of 25 mm.

3. Results and discussion

3.1. Recovery after isochronal annealing

In principle, recovery processes of irradiated materials are governed by the recombination of interstitial atoms and vacancies, which is related to length change during thermal annealing for low-dose irradiated materials. Recovery tendencies of the irradiated α - and β -Si₃N₄ after isochronal thermal annealing for 3 h up to 1473 K are shown in Fig. 1. Those of the irradiated α - and β -SiAlON are referenced for comparison [17]. Macroscopic irradiated lengths of α - and β -Si₃N₄ and α - and β -SiAlON were mostly recovered to their original unirradiated lengths, which suggested that the defects were largely removed. The measured recovery rates of α -Si₃N₄ and α -SiAlON were similar in shape. Both lengths began to decrease starting at the annealing temperature of slightly above the irradiation temperature, and gradually decreased with increasing annealing temperature. However, length changes of α -phases of both compounds from 1123 K to 1273 K were very small compared to those of lower and higher temperatures, as indicated with an arrow in Fig. 1. Beyond this temperature range, the length decreased again with increasing annealing temperature up to 1473 K, the highest temperature of this measurement. In the case

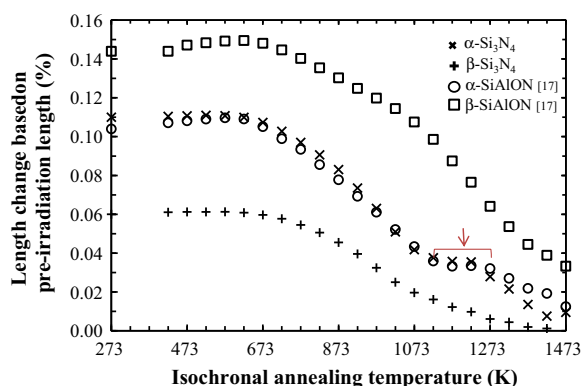


Fig. 1. Length change of irradiated Si₃N₄ and SiAlON [17] by isochronal annealing for 3 h up to 1473 K.

Download English Version:

<https://daneshyari.com/en/article/1565009>

Download Persian Version:

<https://daneshyari.com/article/1565009>

[Daneshyari.com](https://daneshyari.com)