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Effect of dual-beam-irradiation by helium and carbon ions on microstructure development of SiC/SiC composites

S. Nogami [a], A. Hasegawa [a,*], K. Abe [a], T. Taguchi [b], R. Yamada [b]

[a] *Department of Quantum Science and Energy Engineering, Graduate School of Engineering, Tohoku University, Aramaki-aza Aoba 01, Aoba-ku, Sendai, 980-8579, Japan*

[b] *Department of Material Science, JAERI, Tokai, 319-1195, Japan*

Abstract

Microstructural changes of SiC-fiber reinforced SiC-matrix (SiCf/SiC) composites after simultaneous helium-ion and carbon-ion irradiations were studied. SiCf/SiC composite specimens were irradiated up to 10 dpa and 1000 at. ppm He at 1073 and 1223 K. Cross-sectional microstructural observation and irradiated surface analyses were performed. Dimensional changes in SiC-fibers and pyrolytic graphite layers were observed but a dimensional change was not observed in the SiC-matrix. Microstructural features such as voids and dislocation loops were not observed in these experimental conditions.

1. Introduction

Silicon carbide (SiC) is considered to be an attractive material for the first-wall and blanket-shield because of its inherently lower induced radioactivity under high energy neutron irradiation and because of its good high temperature mechanical properties. Monolithic SiC is too brittle to serve as a structural material, so continuous SiC-fiber reinforced SiC-matrix composites (SiCf/SiC) have been developed as a candidate structural material for fusion reactor applications. The recent status of SiCf/SiC composites for fusion applications has been reviewed in other works [1,2].

Transmutant gas atoms such as helium (He) and hydrogen (H) are produced in materials during 14 MeV-neutron irradiation. Helium production in SiC will be approximately 2000 at. ppm/MWa/m^2 at the first-wall. The ratio of He concentration to the displacement damage under neutron irradiation will reach about 130 at. ppm/dpa in SiC, a much larger value than for other candidate materials such as ferritic steels and vanadium alloys [3,4]. Helium is insoluble in virtually all materials and is captured easily with vacancy-type clusters. For this reason, the swelling may be somewhat enhanced due to irradiation. Consequently, helium may promote void nucleation during irradiation if the temperature is high enough. So it is important to investigate the effects of He on microstructural development in SiCf/SiC composites to qualify these materials for fusion reactor service.

Since an intense 14 MeV-neutron irradiation facility does not exist, He effects on mechanical properties have been studied using accelerator-based He implantation techniques. Hasegawa et al. [5,6] investigated the He effects on mechanical properties and microstructural change of SiCf/SiC composites using relatively high energy single He-ion beam irradiation. In these studies, the He/dpa ratio was somewhat higher ($\sim$25 000) than fusion reactor conditions. Recently, dual and triple-beam-facilities utilizing He, H and self-ions have become available to simulate fusion irradiation conditions by varying the beam intensities to provide He/dpa and H/dpa ratios that are expected to occur in fusion reactors.

In the present paper, the effect of He on the microstructural development concurrently with displacement damage of SiCf/SiC composites was studied using dual-beam-irradiation with He and C ions.

* Corresponding author. Tel.: +81-22 217 7923; fax.: +81-22 217 7925.

E-mail addresses: nogami@jupiter.qse.tohoku.ac.jp (S. Nogami), akira.hasegawa@jupiter.qse.tohoku.ac.jp (A. Hasegawa), katsunori.abe@jupiter.qse.tohoku.ac.jp (K. Abe), taguchi@popsvr.tokai.jaeri.go.jp (T. Taguchi), reiji@popsvr.tokai.jaeri.go.jp (R. Yamada).

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2. Experimental

In this study, a two-dimensional SiCf/SiC composite and a monolithic SiC (β-SiC) were irradiated. SiCf/SiC composite was fabricated by DuPont Lanxide with 0/90 SiC-fiber weaves of Hi-Nicalon™ infiltrated with β-SiC using the chemical vapor infiltration (CVI) process. The fiber/matrix interface was composed of a pyrolytic graphite layer deposited by the chemical vapor deposition (CVD) process. The thickness of graphite layer was 1.2 μm. Hi-Nicalon™ is composed of β-SiC grains whose size is about 5–10 nm and substantial residual oxygen (<0.5 wt%) and carbon (C/Si atomic ratio ~1.39) [1]. In preparation for ion-beam irradiation, the composite samples were sliced perpendicular to the lamination layers of textile into 500 μm thick slices using a low speed diamond saw. The slices were mechanically thinned to 300 μm thickness with diamond paste, and then cut into 3 mm diameter disks with a diamond slurry drill.

Monolithic β-SiC samples were also prepared in the same shape as the composites ones. The monolithic SiC were fabricated using the CVD process by Mitsui Engineering and Shipbuilding Co., Ltd. and were composed of cylindrical β-SiC grains with about 500 nm diameter, which are polycrystal SiC highly oriented to (1 1 1).

Dual-beam-irradiation was carried out at the Takasaki ion accelerators for advanced radiation application (TIARA) facility of JAERI/Takasaki. Specimens were simultaneously irradiated at temperatures of 1073 and 1223 K with 6.50 MeV C^{2+} ions by a tandem accelerator and with 1.70 MeV He^{+} ions by a single-ended accelerator. Specimen temperature during irradiation was monitored using an infrared pyrometer. The projected range of the two beams in Hi-Nicalon™ was 4.5 μm, which was calculated with the TRIM code [7] using a 45 eV displacement threshold energy [8] and a 2.74 g/cm^3 density. Fig. 1 shows the calculated depth distribution of He and C concentrations, and it also shows the displacement damage in Hi-Nicalon™. The peak displacement damage and the damage rate were about 10 dpa and 1.1×10^{-3} dpa/s, respectively. Helium concentration was 0 or 1000 at. ppm at the damage peak of C ions.

After irradiation, irradiated surfaces were examined using a scanning electron microscope (SEM). A step-height measurement using a scanning probe microscope (SPM) was also made. For cross-sectional microstructural observation, specimens were fixed to β-SiC plates with resin onto a portion of the unirradiated surface and mechanically sliced by a dicing saw into 50 μm thickness. Then, a part of irradiated area was thinned to about 50 nm thickness from the irradiated surface to about 10 μm depth by a focused ion-beam (FIB) device (Hitachi FB-2000A) using 30 kV-Ga ion-beam at the

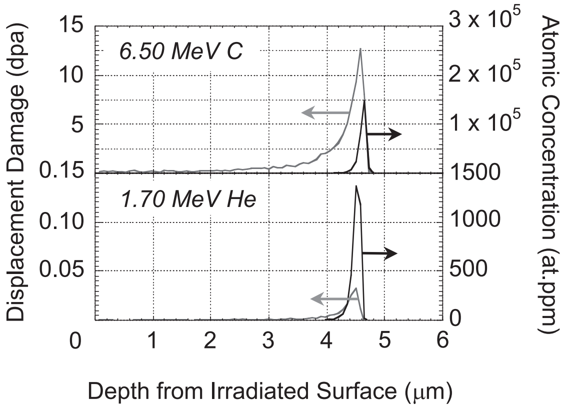

Fig. 1. Calculated depth distribution of C and He concentrations and displacement damage in Hi-Nicalon by the TRIM code. Density: 2.74 g/cm^3, displacement threshold energy: 45 eV.

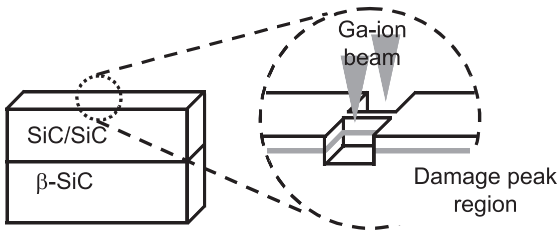

Fig. 2. Schematic illustration of the thin foil processing by an FIB device.

Tohoku University. The schematic illustration of the thin foil processing is shown in Fig. 2. The cross-sectional microstructural observation was performed using a field emission transmission electron microscope (FE-TEM, Hitachi HF-2000) at 200 kV.

3. Results and discussion

Cross-sectional microstructural observations by TEM for SiCf/SiC composite specimens irradiated up to 10 dpa and 0 at. ppm He at 1073 K, 1000 at. ppm He at 1073 K and 1000 at. ppm He at 1223 K are shown in Fig. 3. Neither amorphization nor He bubbles were observed in the SiC matrix. SiC matrix showed to have excellent stability, with respect to amorphization and He retention under this dual-beam-irradiation conditions. These results are consistent with previous works. Snead et al. [9] reported the threshold irradiation dose for the amorphization of SiC and also that amorphization was not observed for doses of 10 dpa at irradiation temperatures between 1073 and 1223 K. Sasaki et al. [10] studied He release behavior of neutron irradiated

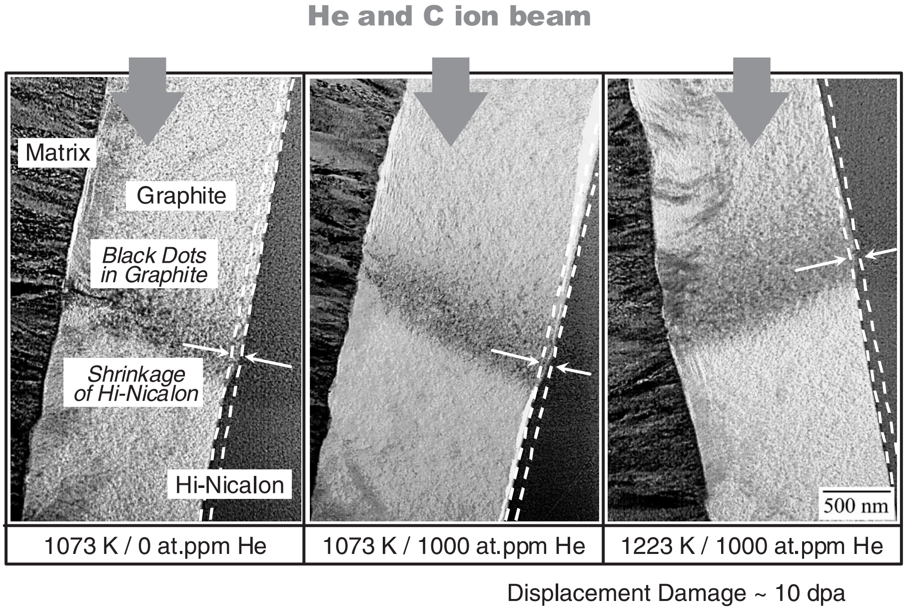

Fig. 3. Cross-sectional microstructural observations of SiCf/SiC composites around the peak damage region.

pressureless-sintered-SiC. In this material He release became significant at temperatures higher than about 1273 K [10]. Helium is considered immobile in this temperature region.

Shrinkage of Hi-Nicalon™ from the irradiated surface to the region of the projected range was also observed, which was indicated by dashed lines in Fig. 4. From results of neutron irradiation study, shrinkage of SiC fibers by irradiation is thought to be caused by crystallization of Si–C–O and graphite phase in SiC fibers [11]. A change in β-SiC grain size was not observed for the three specimens, as determined by selected dark-field imaging of TEM. This may be attributed to a relatively small crystallized volume in the fiber.

Shrinkage of the graphite layer parallel to the fiber axis direction at the irradiated surface and a high density of black-dots in the peak damage region were observed for the three specimens. These black-dots are thought to be clusters of point defects. Dimensional changes are considered to be mainly attributed to the formation of these defects by dual-beam ion irradiation. Kelly et al. [12] reported a mechanism of radiation damage in graphite in which swelling occurred perpendicular to and shrinkage parallel to the basal planes, which are

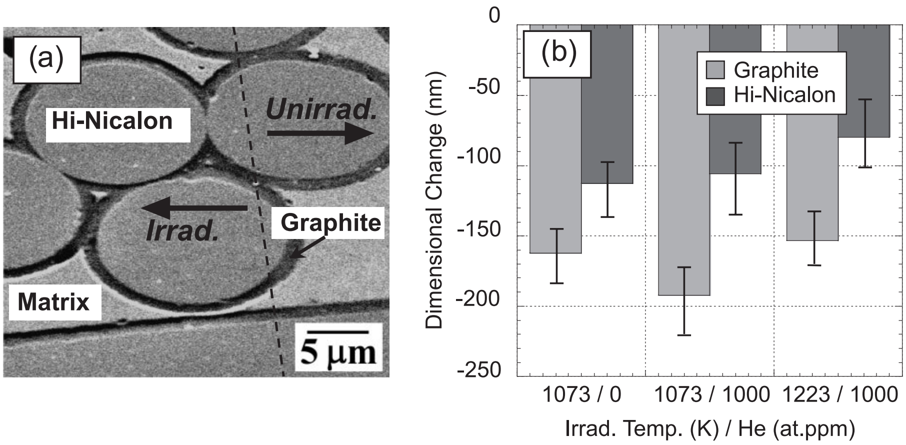

Fig. 4. Results of surface observation of SiCf/SiC composites after dual-beam-irradiation: (a) an SEM observation. Irradiation condition; displacement damage: 10 dpa, He concentration: 1000 at. ppm, irradiation temperature: 1073 K, (b) SPM measurements.

caused by the migration and preferred realignment of lattice defects.

Results of irradiated surface observation by SEM are shown in Fig. 4(a). Step-height differences among graphite layers, SiC-matrix and Hi-Nicalon™ were observed for all irradiated composites. In contrast, this type of morphology change was not observed in unirradiated regions. Step-height changes of monolithic SiC specimens, irradiated under the same conditions as composite specimens, were not observed by SPM measurements. Swelling of SiC is considered to be mainly due to void formation at higher temperature (>1523 K) [13] and amorphization at lower temperature ($\sim$77 K to RT) [9]. Neither void formation nor amorphization were observed in the SiC-matrix of composite specimens, so the SiC-matrix was considered not to change in volume under the irradiation conditions. Based on this assumption, the observed step height difference from the SiC-matrix to the Hi-Nicalon™ and that from the Hi-Nicalon™ to the graphite layer was due to volume shrinkages of both the Hi-Nicalon™ and the graphite layer. As the CVI process to fabricate the composite specimens was performed at the temperature of about 1373 K, the shrinkage of Hi-Nicalon™ did not occur by heat treatments at 1223 K. Fig. 4(b) shows the dependence of dimensional changes on irradiation temperature and He content. Under these irradiation conditions, no differences of volume shrinkage for Hi-Nicalon™ and graphite layer were clearly observed. The relative shrinkage of the graphite layer compared to SiC matrix at the irradiated surface was large, and it might be attributed to microstructural change at the region of the projected range. Radiation-induced sublimation at the irradiated surface of the graphite layer may be considered for the shrinkage, too.

Fig. 5 shows the microcracks and debondings within the graphite layer. These were mainly observed in front and behind the peak damage region. On the other hand,

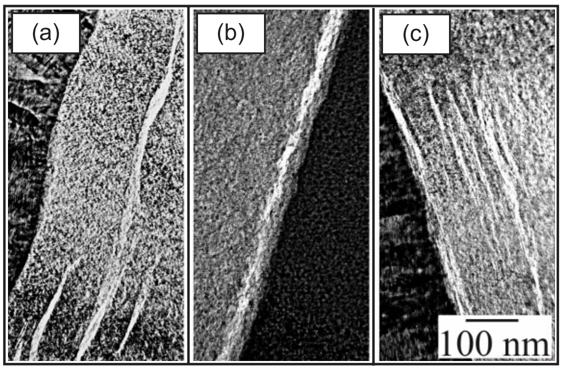

Fig. 5. Microcracks and debondings in graphite layers of irradiated SiCf/SiC composites. Irradiation temperature and He concentration: (a) 1073 K, 0 at. ppm, (b) 1073 K, 1000 at. ppm, and (c) 1223 K, 1000 at. ppm.

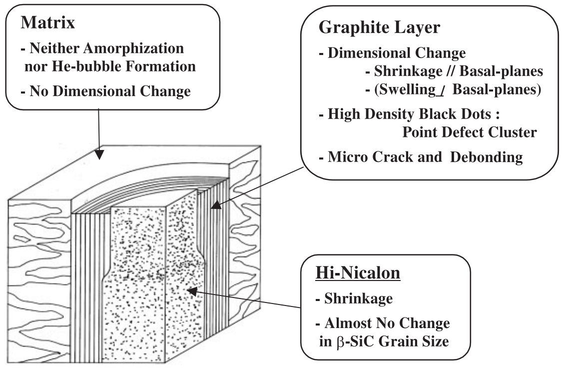

Fig. 6. Schematic illustration of summary.

these cracks were not observed in the peak damage region. This indicates swelling perpendicular to the basal-planes of the graphite layer in the peak damage region. The cracks and debondings may be attributed to large differences in dimensional change among the matrix, pyrolytic graphite and Hi-Nicalon™ in front and behind the peak damage region.

These results showed dimensional changes peculiar to each phase, however, these changes were mainly attributed to displacement damage (10 dpa) by the C-ions. A schematic summary of the observations is illustrated in Fig. 6.

The effect of He on the microstructure of graphite layer and Hi-Nicalon™ was not clearly observed in this work, therefore He was not considered to have a significant influence on the microstructural development of SiCf/SiC composites under these irradiation conditions. It is probably because the mobility of He atoms and vacancies are too low to effect a microstructural development in this temperature region [14]. Further higher temperature irradiation study should be required to clarify He effects in fusion reactor conditions.

Recently, highly crystalline and nearly stoichiometric SiC-fibers such as Tyranno SA and Nicalon-S [15] have been developed. These SiC-fibers are expected to have better dimensional stability under irradiation because of their crystalline and stoichiometric compositions. Additionally, several new fiber-coating materials, such as SiC/C-multilayer, have been developed. New composites containing Nicalon-S and these new coating materials are expected to have much better dimensional stability and better mechanical properties under irradiation conditions.

4. Summary

To study the effect of He on microstructural development of irradiated SiCf/SiC composites, dual-beam-irradiation using He-ions and C-ions was carried out at 1073 and 1223 K. The following microstructural observations and irradiated surface analyses were obtained:

1. Neither amorphization nor He bubble formation in the SiC matrix were observed.
2. Axial shrinkage of Hi-Nicalon™ was observed; however, changes in β-SiC grain size in Hi-Nicalon™ were not observed.
3. Axial shrinkage and concurrent swelling of the graphite layer perpendicular to the fiber were observed. These dimensional changes may be attributable to preferred orientation of the point defect clusters formed in the peak damage region of the graphite.
4. Microstructural changes observed in this study were considered to be mainly attributed to displacement damage (10 dpa) caused by the C-ions. Helium effects were not clearly observed in SiCf/SiC composites under these experimental conditions.

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