Surface Morphology Changes in a SiC/SiC Composite as Caused by Simultaneous Triple-Ion-Beam Irradiation*1

Shuhei Nogami1, *2, Akira Hasegawa1, Tomitsugu Taguchi2, Katsunori Abe1 and Reiji Yamada2

1Department of Quantum Science and Energy Engineering, Tohoku University, Sendai 980-8579, Japan
2Department of Material Science, Japan Atomic Energy Research Institute, Tokai 319-1195, Japan

Surface morphology changes of silicon carbide (SiC) fiber reinforced SiC matrix (SiC/SiC) composite materials occurring after simultaneous triple-ion-beam irradiation were studied. Irradiation tests were performed with helium (He) ions, hydrogen (H) ions, and self-ions (carbon (C) ions or silicon (Si) ions). The peak displacement damage was 10 dpa (displacements per atom), and the irradiation temperatures were 600, 800 and 950°C. The concentrations of He and H at the damage peak region were 1000 atomic ppm and 385 atomic ppm, respectively. Observations of the irradiated surface and the measurement of morphology changes were performed. The shrinkage of SiC fibers and the apparent shrinkage of the interfacial material, carbon, between the matrix and the fibers at the irradiated surface were observed. These phenomena were mainly attributed to displacement damage caused by irradiation.

(Received April 25, 2000; Accepted November 22, 2000)

Keywords: silicon carbide, composite, fusion reactor material, surface change, ion irradiation, helium effects, hydrogen effects, scanning electron microscope, scanning probe microscope

1. Introduction

Radiation resistance and low induced-radioactivity after high energy neutron irradiation are important issues for material selection for fusion reactors. Based on these criteria, silicon carbide (SiC) materials have been considered as a candidate material for the blanket-shield and first-wall structures of a fusion reactor because of their inherently lower induced-radioactivity by 14 MeV-neutron irradiation and their high temperature strength. Monolithic SiC materials are too brittle to serve as a structural material, so continuous SiC-fiber reinforced SiC-matrix (SiC/SiC) composites have been developed for fusion reactor applications because they have higher fracture toughness than monolithic SiC. In fusion reactor conditions, displacement damage and transmutant gas atoms such as helium (He) and hydrogen (H) will be simultaneously formed in materials during neutron irradiation. The amount of He and H will be much higher than that of other candidate materials such as ferritic steels and vanadium alloys. The solubility of He in SiC is negligibly small and that of H is also small. Therefore, these gases may combine with defects and form cavities. The effect of these gas atoms on microstructural evolution needs to be investigated.

It has been reported that crystallized SiC materials swell1) and SiC fiber, which includes small β-SiC grains, shrinks2) after neutron irradiation because of differing microstructural changes during irradiation. SiC/SiC composite materials are composed of crystalline β-SiC-matrix, interphase-materials, such as carbon, and SiC-fiber. The morphological changes are considered to be occurred after irradiation because of their individual dimensional changes.

The purpose of this study is to investigate the morphology changes of the surface of SiC/SiC composites under fusion conditions by using simultaneous triple-ion-beam (self-ion, He-ion and H-ion) irradiation using accelerators.

2. Experimental

The SiC/SiC composite materials used in this study were fabricated with two-dimensional SiC fiber weaves infiltrated with β-SiC using a chemical vapor infiltration (CVI) process. The SiC fibers were Hi-Nicalon™, and these fibers are composed of small β-SiC grains (about 0.5 nm), 0.5 mass% oxygen and free carbon.3) The atomic ratio of carbon to silicon in Hi-Nicalon™ is about 1.39.4) The pyrolitic carbon was deposited on Hi-Nicalon™ fibers using a CVI process. The pyrolitic carbon serves as a fiber/matrix interphase material, and the thickness of the C-layer was 1.2 μm. These composites were fabricated by DuPont Laxnide in the USA. The specimens were cut perpendicularly to the fiber weave and mechanically polished into a mirror-state with a final thickness of 300 μm. Then the specimens were cut into disks with a diameter of 3 mm.

A simultaneous dual-ion-beam irradiation and two kinds of simultaneous triple-ion-beam irradiations were carried out at the Takasaki Ion Accelerator for Advanced Radiation Application (TIARA) facility of the Japan Atomic Energy Research Institute (JAERI). The irradiation conditions are shown in Fig. 1. The combinations of ions and their accelerated energy were 6.50 MeV-C + 1.70 MeV-He (upper figure), 4.30 MeV-C + 1.20 MeV-He + 380 keV-H (middle figure), and 7.84 MeV-Si + 1.20 MeV-He + 380 keV-H (lower figure), respectively. The projected ranges and depth distributions of displacement damage and atomic concentration were calculated with the TRIM code5) using a 45 eV displacement threshold energy5) and a material density of 2.74 × 103 kg/m3,3) which corresponds to that of Hi-Nicalon™. The peak displacement damage by self-ions was about 10 dpa (displacements per atom) and the displacement damage rate was

---

1) This Paper was Presented at the Spring Meeting of the Japan Institute of Metals, held in Yokohama, on March 29, 2000.
2) Graduate Student, Tohoku University.
were in the range of the gas production ratio in the first-wall region of a fusion reactor. Irradiation temperatures, which were measured using an infrared pyrometer during irradiation, were 600, 800 and 950°C. The surface morphology changes after irradiation were observed using an optical microscope and a scanning electron microscope (SEM) at 10 kV, and step-height measurements were performed using a scanning probe microscope (SPM).

3. Results and Discussion

3.1 Irradiated surface observation

Figure 2 shows a typical observation of the irradiated surface of the SiC/SiC composites using an optical microscope and an SEM. This specimen was irradiated up to 10 dpa using Si-ions at 600°C, and the gas concentrations were 1000 atomic ppm He and 385 atomic ppm H. In this experiment, a 2 mm diameter region on a specimen surface was irradiated with ion beams. The change in the specimen surface morphology after irradiation can be seen by comparing Fig. 2(a) to Fig. 2(b). This morphology change was not only due to the heat treatment during irradiation but also due to the ion beam irradiation because the CVI process used to fabricate the composite specimens was performed at a temperature of about 1100°C, which is higher than the irradiation temperatures. In addition, the region with the morphology changes was restricted only to the irradiated area. Figures 2(c) and (d) show the boundary between the irradiated region and the unirradiated region of this specimen. In Fig. 2(d), the specimen was tilted to 45°. From this image, the sinkage of the C inter-

---

**Fig. 1** The calculated depth distributions of He concentration, H concentration and displacement damage, where the value of density corresponds to that of SiC fiber (Hi-Nicalon™).

**Fig. 2** A typical observation of the irradiated surface of the SiC/SiC composites. (a): An optical microscope observation of the surface before irradiation. (b): An optical microscope observation of the surface after irradiation. (c), (d): An optical microscope observation (c) and an SEM observation (d) of the boundary between the irradiated region and the unirradiated region.
phase layer is easily observed. Similar morphology changes were observed in all the irradiated specimens.

Using a transmission electron microscope (TEM), Nogami et al. reported on the cross-sectional observation of SiC/SiC composites after dual-ion-beam irradiation using He-ions and C-ions. Figure 3 shows a typical TEM observation of the irradiated surface of a SiC/SiC composite. This specimen was irradiated at 950°C up to 10 dpa using C-ions and He-ions (1000 atomic ppm He). The thin-foil for TEM observation was prepared using a focused ion beam (FIB) device using 30 keV Ga-ions. Tungsten (W) was deposited on the surface before the thinning process to protect the initial surface from Ga-ion beam irradiation. Before irradiation, the specimen surface was flat and each surface of matrix, C-layer and fiber were considered to be in the initial one shown in Fig. 3. The micrograph shows not only the sinkage of the C-layer from the surface but also the morphology change of SiC fibers during irradiation. Nogami et al. reported that this morphology change of SiC fibers was due to the volumetric shrinkage after dual ion-beam irradiation.

3.2 Shrinkage of SiC fibers after irradiation

Figure 4 shows a typical step-height measurement result of an irradiated surface as measured using an SPM. The SPM was scanned along the dotted line in Fig. 4(a), and Fig. 4(b) shows the measurement along the dotted line. In this result,
the morphology changes between the irradiated region and the unirradiated region were not detected. A device more sensitive than an SPM is needed to observe a height change between an irradiated and unirradiated region of SiC matrix. In this study, the height between the surface of SiC matrix and that of SiC fiber \( (L_{\text{m}}) \) in the irradiated region and that between the surface of SiC matrix and that of C-layer \( (L_{\text{cm}}) \) were measured.

The bump-height of the SiC matrix \( (L_{\text{m}}) \) was assumed from the results of volumetric swelling of CVD (Chemical Vapor Deposition) processed \( \beta \)-SiC materials after neutron irradiation as reported by Snead et al.\(^3\) Only displacement damage (a few dpa) was produced in the materials in this neutron irradiation tests studied by Snead et al. By considering this report, the volumetric swelling \( (\Delta V_m) \) of CVD-SiC caused by displacement damage was estimated to be about +0.7%, +0.4% and +0.3%, at 600°C, 800°C and 950°C, respectively. The volumetric swelling \( (\Delta V_m) \) is usually estimated from the following equation for the linear swelling:

\[
\Delta V_m (\%) \sim 100 \times \frac{L_{\text{m}}}{R_p}
\]

where \( R_p \) is the projected range of ion beams calculated using the TRIM code. From these results and this equation, the bump-height \( (L_{\text{m}}) \) caused by displacement damage was calculated to be about 21 nm at 600°C \( (R_p = 3 \mu m) \), about 12 nm at 800°C \( (R_p = 3 \mu m) \), about 18 nm at 800°C \( (R_p = 4.5 \mu m) \), and about 14 nm at 950°C \( (R_p = 4.5 \mu m) \), respectively.

The volume ratio of shrunk SiC fiber was estimated by the shrinkage parameter \( (L_{\text{cm}} - L_{\text{m}})/R_p \). Figure 5 shows the dependence of the shrinkage parameter on irradiation conditions. It appears that self-ions irradiation causes the majority of the shrinkage. This can be seen by considering the specimens irradiated at 800°C with only C-ions because only displacement damage was produced by irradiation with self-ions. The volumetric shrinkage due to displacement damage was considered to be mainly due to irradiation induced crystallization of the amorphous phase and growth of \( \beta \)-SiC grains in Hi-Nicalon because the shrinkage of Hi-Nicalon was also observed after neutron irradiation tests, in which He and H were not produced, and grain sizes of \( \beta \)-SiC in Hi-Nicalon grew larger than those before irradiation by TEM observation.\(^3\)

The difference of the shrinkage parameter between specimens irradiated with C-ions and with Si-ions was observed. The different shrinkage parameters may be due to the difference in the damage distribution shown in Fig. 1. Although the displacement damage at the peak was almost the same in both specimens, the total damage quantities integrated across the irradiated region by Si-ions were larger than those by C-ions.

At 800°C, H irradiation enhanced volumetric shrinkage while helium had almost no effect, while the effect of He and H on morphology changes was not clearly observed at 600°C. Nagata et al.\(^5\) reported that deuteron (D) atoms were retained in SiC through the formation of Si-D or C-D bonds up to about 700°C and were released above about 700°C. By considering this result and free energy changes of some compounds such as CH₄ and C₂H₆,\(^1\) hydrogen atoms were proposed to combine with dangling bonds of C and Si atoms below 800°C and were probably released as gases such as CH₄ and SiH₄ from SiC-fiber at 800°C. Hi-Nicalon™ fiber contains 17 atomic% excess carbons. Therefore, the release of H in the form of CH₄ at 800°C from SiC fiber might also enhance the shrinkage of SiC fibers. Hasegawa et al.\(^1\) reported that the He in CVD-SiC and Hi-Nicalon™ fibers was immobile below 900 to 1000°C, and therefore, the He effect may not be obvious below 800°C. The effect of He on microstructure development is discussed elsewhere.\(^8,\)\(^1\) For example, helium bubbles were observed in dual-ion-beam irradiation (C and He) at 950°C.

### 3.3 Morphological change of C-layer after irradiation

Figure 6 shows the dependence of the shrinkage of the C-layer below the irradiated surface as a function of irradiation conditions. The amount of shrinkage was estimated by \( (L_{\text{cm}} - L_{\text{m}}) \). The data from He-ion, H-ion and C-ion irradiation at the temperature of 800°C was not measured by an SPM because
the thickness of the C-layer was thin (0.15 μm).

Engle et al.\textsuperscript{13} proposed a mechanism to describe the dimensional change of graphite materials due to displacement damage. Swelling perpendicular to c-plane and shrinkage parallel to a-axis is responsible for the dimensional change caused by neutron irradiation. The microstructure of the C-layer in SiC/SiC composite specimens is almost the same as graphite materials and the a-axis is parallel to the fiber direction. Therefore, the shrinkage of the C-layer can be attributed to shrinkage parallel to a-axis, which was mainly induced by displacement damage due to the irradiation of self-ions (C or Si-ions).

The difference in the shrinkage quantities between specimens irradiated with C-ions and with Si-ions was observed. This may be due to the difference of the damage distribution and the total amount of displacement damage. The shrinkage volume parallel to a-axis of the specimens irradiated by Si-ions is larger than by C-ions.

In this study, the effect of He and H irradiation on the morphology change of C-layer was not clearly observed in the temperature between 600 and 950°C. The shrinkage of the C-layer in this work is thought to be mainly due to displacement damage. However, to better understand the mechanism of the volume change by triple-ion-beam irradiation, microstructure development should be observed.

4. Summary

Morphology changes of the surface of SiC/SiC composites after triple-ion-beam irradiation using He-ions, H-ions and C-ions or Si-ions at temperature between 600 and 950°C were studied, and the following results were obtained:

1. Surface morphology changes of irradiated SiC fibers were observed, and the changes were mainly attributed to volumetric shrinkage caused by displacement damage.

2. At 800°C, H enhanced the shrinkage of SiC fibers while helium had almost no effect on the volumetric shrinkage.

3. Sinkage of the C-layer below the irradiated surface of the SiC matrix was observed. This sinkage is mainly due to shrinkage caused by displacement damage.

4. The effect of He and H on the sinkage of C-layers was not clearly observed for the irradiation conditions in this experiment.

Acknowledgement

The authors are grateful to Dr. R. H. Jones (Pacific Northwest National Laboratory) for providing SiC/SiC composites. This work was partly supported by the JUPITER (Japan-USA Program of Irradiation Testing for Fusion Research) program. The authors are also grateful to Dr. K. Toji and Mr. K. Motomiya (Tohoku University) for their help with thin-foil processing using an FIB device. This work was undertaken as part of the Universities-JAERI collaboration research project. We wish to thank all the staff of the electrostatic accelerator group of TIARA for their operation of the accelerator.

REFERENCES