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Study of hydrogen effects on microstructural development of SiC base materials under simultaneous irradiation with He- and Si-ion irradiation conditions

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Abstract

The microstructural development of SiC/SiC composite under the multi-ion beam irradiation up to 10 dpa at 800 and 1000 °C was investigated by transmission electron microscopy. Microstructural evolution in each composites component was dependent on the component grain structures, the helium or hydrogen implanting mode and the irradiation temperature. In this study, the synergistic effects of H and He gas elements on cavity formation in the composite component are discussed.

1. Introduction

Silicon carbide (SiC) has been proposed as a candidate structural blanket material for fusion reactor applications due to its low activation and its high strength at high temperature. SiC/SiC composites have been developed using advanced SiC fiber to improve mechanical and thermal conductivity properties [1,2].

Previous works examined the neutron irradiation resistance of SiC/SiC composite that were made with low oxygen content, highly crystalline and stoichiometric composition SiC fiber such as Hi-Nicalon Type S and Tyranno SA [3]. However, in addition to the displacement damage, helium (He) and hydrogen (H) are produced in SiC by nuclear transmutation reactions by 14 MeV neutrons. Gas productions were estimated to be approximately 20–2000 at.ppm He/(MW/m^2) and 10–800 at.ppm He/(MW/m^2) in the ARIES IV blanket design [1].

The effects of these gas elements in SiC are summarized as follows: helium is insoluble in SiC and is considered to stabilize vacancy-type clusters produced by displacement damage, and to enhance cavity formation [4–6]. The solubility of hydrogen is also very low. Hydrogen is trapped at both Si- and C-sites produced by displacement damage, where it forms molecules of C–H or Si–H [7]. Thermal desorption spectrometry has shown that the mobility of hydrogen in SiC increased above 800 °C [8]. Therefore, hydrogen potentially could enhance cavity formation in the temperature range 800 °C and above, which corresponds to the operating temperature range of a fusion reactor using SiC/SiC composite. The effects of the simultaneous production of displacement damage, helium and hydrogen in SiC are necessary to predict the material lifetime under fusion reactor environment. The purpose of this study is to clarify the synergistic effect of hydrogen on the cavity formation in SiC/SiC matrix and fiber components.

2. Experimental

The two-dimensional SiC/SiC composite with Hi-Nicalon Type-S fibers examined in this work was

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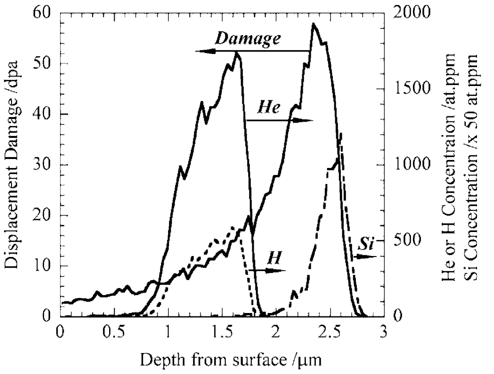

Fig. 1. The depth distribution of the displacement damage, He, H and Si in Sic calculated with TRIM-code [10].

fabricated at Oak Ridge National Laboratory (ORNL) [9]. The SiC fiber weaves in the composite were coated with pyrolitic carbon (PyC) using a chemical vapor deposition (CVD) process. The resulting thickness of the PyC coating was approximately 150 nm. The β-SiC matrix was deposited onto a SiC fabric lay-up by the forced-flow chemical vapor infiltration (FCVI) process.

Simultaneous multi-ion beam irradiation was performed in the TIARA (Takasaki Ion Accelerators for Advanced Radiation Application) facility of JAERI (Japan Atomic Energy Research Institute). Energy degraders were utilized to obtain broad depth distributions of He and H along the displacement damage distribution produced by the Si-ion irradiation. Fig. 1 shows the calculated depth distribution of the implanted Si, He and H, and the displacement damage in SiC calculated using the TRIM code [10] with a displacement energy of 35 eV (Si) and 20 eV (C). The irradiation dose in the examined area was approximately 10 dpa. The He and H concentration to dpa ratios were 130 at.ppm He/dpa and 40 at.ppm H/dpa, respectively. The irradiation temperatures were 800 and 1000 °C. Combinations of simultaneous ion irradiation were Si (Single), Si + H ($\mathrm{Dual_H}$), Si + He ($\mathrm{Dual_{He}}$) and Si + He + H (Triple).

Microstructural observations were carried out using a transmission electron microscope (TEM, Hitachi HF-2000) at 200 keV. The TEM samples were prepared using a Focused Ion Beam machine (FIB, Hitachi FB-2000A) at the JAERI Tokai Laboratory. Cross-sectional observations were performed on the SiC fiber, the SiC matrix and the PyC coating layer.

3. Results and discussion

Figs. 2 and 3 present TEM images of typical regions within the SiC matrix irradiated at 800 and 1000 °C, respectively. The observed cavities were located mainly on grain boundaries for the present irradiation conditions. A summary of microstructural observations and average cavity sizes and number density are given in Table 1.

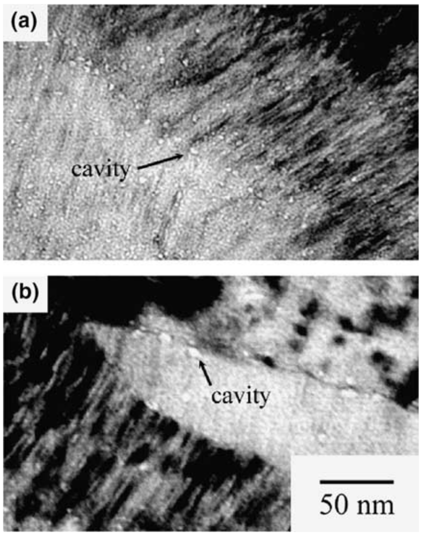

Fig. 2. TEM images of gas co-implanted region of the SiC matrix irradiated by (a) $\mathrm{Dual_{He}}$-ion and (b) Triple-ion beam at 800 °C.

Cavities were observed after $\mathrm{Dual_{He}}$- and Triple-ion beam irradiation at 800 °C only in the co-implanted regions of the SiC matrix, as shown in Fig. 2. The average cavity size in the SiC matrix after Triple-ion beam irradiation was larger than that after $\mathrm{Dual_{He}}$-ion beam irradiation. The number density of cavities in SiC matrix, estimated to be 0.9×10^{22} $\mathrm{m^{-3}}$ was almost the same for either the Triple- or $\mathrm{Dual_{He}}$-ion beam irradiation at 800 °C.

For the SiC matrix irradiated at 1000 °C, cavities were observed after $\mathrm{Dual_{He}}$- and Triple-ion beam irradiation in the co-implanted regions, shown in Fig. 3(b) and (c), respectively. In contrast, cavities were not observed after Single-ion beam (Fig. 3(a)) or after $\mathrm{Dual_H}$-ion beam irradiation. At 1000 °C, the cavity size in the SiC matrix after Triple-ion beam irradiation showed almost the same value as Dual-ion beam irradiation. These results suggest that cavity nucleation in the SiC matrix was enhanced above at least 800 °C by the synergistic effects of displacement damage and He or He + H, but synergistic effects of displacement damage and H on cavity nucleation were not observed for the present conditions.

However, near the PyC coating layer interface cavities were observed in the SiC matrix immediately from the irradiation surface to the gas-implanted depth after the all case of irradiation at 1000 °C, as shown in Fig. 3(d) and (e). The range of cavity nucleation was approximately 300 nm from the PyC/matrix interface for

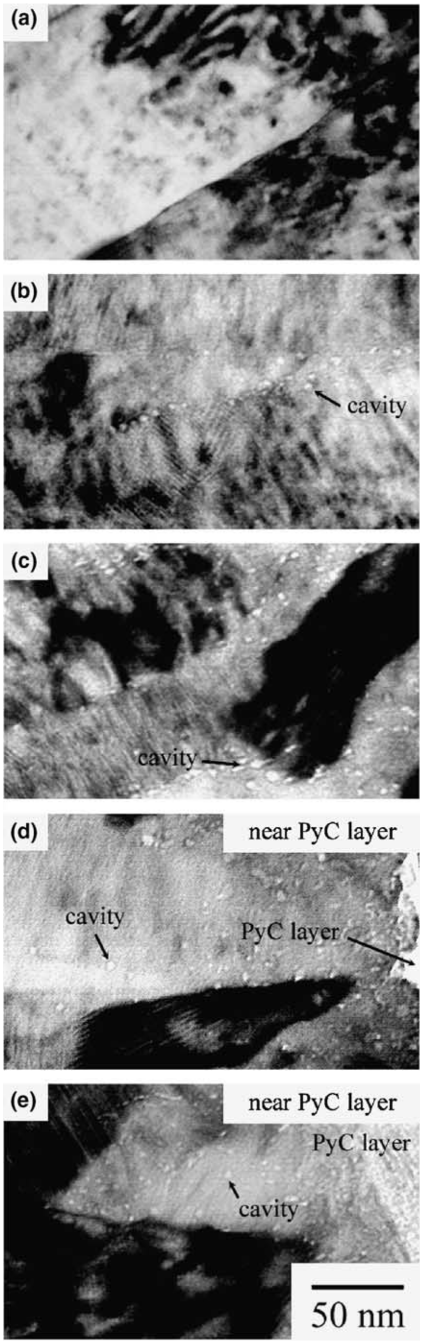

Fig. 3. TEM images of gas co-implanted region of the SiC matrix irradiated by (a) Single-ion, (b) Dual$_{He}$-ion and (c) Triple-ion beam at 1000 °C, and for the region of the SiC matrix near the PyC coating layer irradiated by (d) Single-ion and (e) Triple-ion beam.

Single- and Dual$_{H}$-ion beam irradiation, and approximately 400 nm for Dual$_{He}$- and Triple-ion irradiation. These results indicate that cavity nucleation was enhanced in the SiC matrix near the PyC interface region. Furthermore, the cavity number density in the SiC matrix near the PyC coating interface was larger for the Triple-ion beam compared to Dual$_{He}$-ion beam. The spatial distribution of cavities by irradiation mode near the interface region is summarized in Fig. 4(a) and (b). The cavity density near the carbon coating layer was approximately three times higher than away from the carbon coating layer after Triple-ion beam irradiation. The enhancement of cavity nucleation after Triple-ion beam irradiation near the PyC coating was likely due to the synergistic effect of displacement damage and He and H co-implanting.

Microstructural observations of the SiC fiber irradiated at 1000 °C are shown in Fig. 5. Small cavities were observed only in He + H co-implanted region of the SiC fiber after Triple-ion beam irradiation at 1000 °C. Cavities were not observed in SiC fiber for other irradiation conditions. This result indicates that cavity nucleation is possible in the SiC fiber if H coexists with displacement damage and He.

These results indicate that the existence of He in SiC is needed for cavity formation. He interstitials likely are trapped at the vacancies. Further He interstitials are absorbed and He-vacancy clusters grow. The H atoms also are trapped at both Si- and C-vacancy sites produced by displacement damage, and form H_2 molecules, or C–H and Si–H molecules. The C–H and Si–H bonds dissociate at approximately 800 and 600 °C, respectively [8]. For these experimental conditions, almost all the Si–H and C–H molecules should dissociate, and hydrogen should easily diffuse and form molecule H_2 in a cavity. Because the number density of cavities was limited in the SiC matrix after the 800 °C irradiation, enough H_2 molecules existed in the cavities stabilize them and the cavity size increased after Triple-ion beam irradiation. At 1000 °C, cavity nucleation in the SiC matrix was enhanced in SiC matrix by He ion beam and the number density of cavities increased compared to that at 800 °C. Apparently, the contribution of the H_2 for cavity growth became relatively smaller after 1000 °C irradiation. At 1000 °C, hydrogen molecules likely play the same role as He in the growth of He-vacancy type clusters. Possibly, H_2 more easily diffuses in SiC at 1000 °C compared to helium, and cavity nucleation might then be enhanced by the effect of the overall increasing gas-implantation rate after Triple-ion beam irradiation, compared to Dual$_{He}$-ion beam irradiation.

Non-uniform spatial distribution of the cavities was observed in the SiC matrix after 1000 °C irradiation, as shown in Table 1 and in Fig. 4. These observations might be affected by PyC coating interface, the difference of grain size obtained by the FCVI process in the vicinity of the PyC layer, impurities and so on, but the details need further clarification.

Table 1
Summary of microstructural observations and average cavity sizes for materials irradiated at 800 and 1000 °C

Temperature (°C)	Single			$\mathrm{Dual_H}$			$\mathrm{Dual_{He}}$			Triple		
	Matrix		Fiber	Matrix		Fiber	Matrix		Fiber	Matrix		Fiber
	(a)*	(b)**		(a)*	(b)**		(a)*	(b)**		(a)*	(b)**	
800	–		–	–		–	3.7 0.9		–	4.4 0.9		–
1000	4.2	–	–	4.0	–	–	4.7 1.6	5.0 1.5	–	4.6 3.3	4.7 2.2	2.7

–: cavities were not observed; value: average cavity size (nm) (the upper section) and average cavity number density ($\times 10^{22}\mathrm{m}^{-3}$) (the lower section).
(a)*: near the carbon coating layer for SiC matrix irradiated at 1000 °C.
(b)**: away from carbon coating layer for SiC matrix irradiated at 1000 °C.

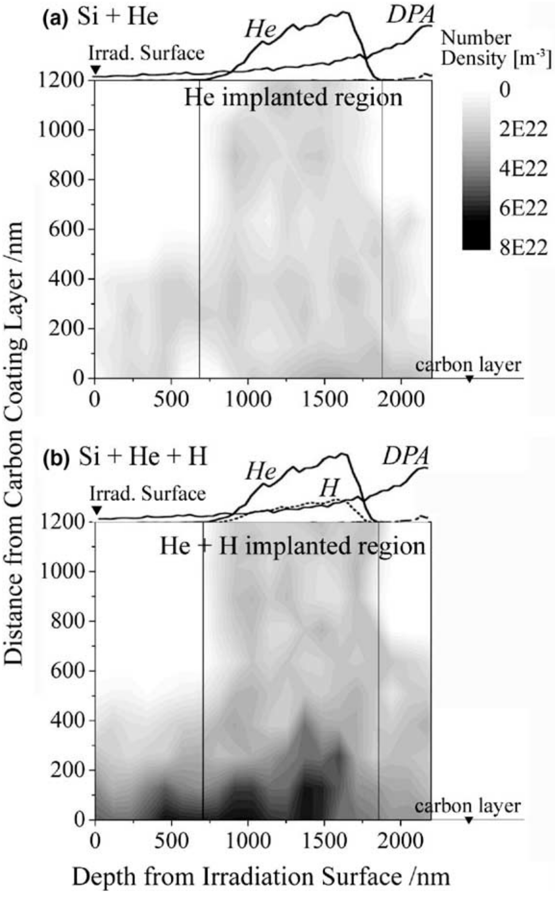

Fig. 4. The spatial distribution of cavity number density (m^{-3}) in the SiC matrix irradiated by (a) $\mathrm{Dual_{He}}$-ion and (b) Triple-ion beam at 1000 °C.

Cavities were not observed in the PyC coating layer for these irradiation conditions. Previously, thermal desorption of He from a graphite phase was reported to occur below 400 °C [11]. Possibly, He in the PyC coating layer readily diffuses to the interface during irradiation, and cavities do not form in the PyC coating layer itself.

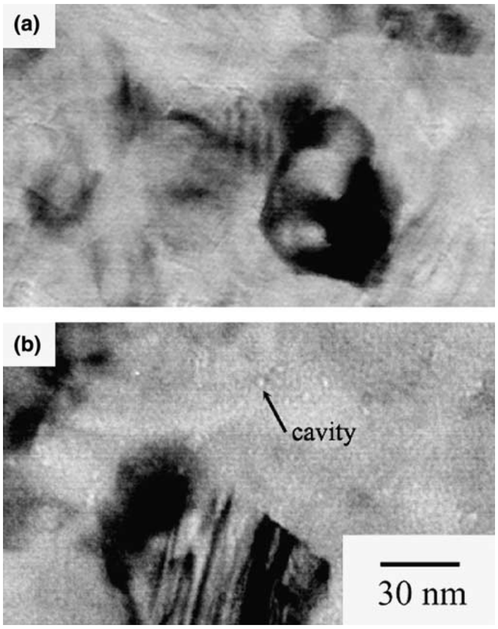

Fig. 5. TEM images of gas co-implanted region of the SiC fiber irradiated by (a) $\mathrm{Dual_{He}}$-ion and (b) Triple-ion beam at 1000 °C.

In the SiC/SiC composite, the SiC fiber is the primary load-bearing element and determines the strength of SiC/SiC composite. At approximately 1000 °C, due to the co-existence of displacement damage, He and H, mechanical property degradation of SiC/SiC composite might be caused by the cavities on the grain boundaries in the SiC fiber and the high density of cavities in the SiC matrix near the PyC coating layer. But the compression stress by the cavity formation has possibility of improvement of mechanical properties of the composites.

Nevertheless, a synergistic degradation of mechanical property due to displacement damage and co-existence of He and H in SiC has not been observed using nanoindentation method [4]. Further investigation will be required to actually predict the lifetime performance of SiC/SiC composites under true fusion conditions.

4. Summary

The microstructural development of cavity formation in SiC/SiC composite after multi-ion beam irradiation up to 10 dpa at 800 and 1000 °C were investigated by TEM. The following results were obtained:

1. (1) Cavities were observed mainly on SiC grain boundaries. The grain boundary is preferred nucleation site for cavity formation for these experimental conditions.
2. (2) Because the SiC fiber consists of fine grains (≈20 nm diameter), the cavity formation was only observed after Triple-ion beam irradiation at 1000 °C. Apparently, cavity nucleation in the SiC fiber can be instigated by the coexistence of H with displacement damage and He at 1000 °C.
3. (3) In the SiC matrix, the cavity size was larger at 800 °C and the cavity density was larger at 1000 °C after Triple-ion compared with $\mathrm{Dual}_{\mathrm{He}}$-ion beam irradiation. This result suggests that the coexistence of H with displacement damage and He strongly affected cavity nucleation by enhancing thin cavity growth at 800 °C and dense cavity nucleation at 1000 °C.

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