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Effect of simultaneous ion irradiation on microstructural change of SiC/SiC composites at high temperature

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Abstract

The effect of simultaneous triple ion irradiation of He, H and Si on microstructural evolution of two kinds of SiC/SiC composites (HNS composite (using Hi-Nicalon type S SiC fiber) and TSA composite (using Tyranno SA SiC fiber)) at 1000 °C has been investigated. The microstructure observations of SiC/SiC composites irradiated to 10 dpa were examined by transmission electron microscopy. He bubbles were hardly formed in matrix of TSA composite, but many helium bubbles and some cracks were observed at grain boundaries of matrix of HNS composite. He bubbles and cracks were not, on the other hand, observed in the both fiber fabrics of HNS and TSA composites. Debonding between fiber and carbon layer following irradiation region was not observed in the both composites. Under these irradiation conditions, TSA composite showed the better microstructural stability against ion beams irradiation than one of HNS composite.

1. Introduction

Ceramic matrix composites show excellent mechanical properties at high temperature and also non-catastrophic failure behavior. These materials, therefore, are attractive candidates for structural applications at high temperature. For these reasons, SiC/SiC composites are being actively investigated for the first wall and blanket components in power fusion reactors [1,2], and additionally because of their low residual radioactivity after neutron irradiation [3].

Under the fusion condition, He and H atoms are produced by 14.1 MeV fusion neutrons transmutation reactions of (n, α) and (n, p). He and H production rates in SiC in first wall region are approximately 1500–2000 and 800 appm/MWa/m^2 corresponding to a gas/dpa ratio of 130 appm He/dpa and 50 appm H/dpa, respectively [4,5]. The first wall transmutation rates in SiC are larger than the other candidate materials such as ferritic steels and vanadium alloys [3,5]. Because He is insoluble in virtually all materials and is captured easily with vacancy clusters, microstructural changes and strength degradation may be accelerated in SiC/SiC composites.

Previous studies reported the effect of He on the microstructural change of SiC [6,7]. The He/dpa ratios were, however, more than 20 000 appm/dpa in these studies and too high as compared with the ratio under the fusion condition. Recently the synergistic effect of displacement damage and He atoms on the microstructural changes in SiC/SiC composites was studied at almost the same He/dpa ratio under fusion conditions [8]. These composites were, however, irradiated at lower temperature than 800 °C. These materials will be used at

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temperatures ranging from 800 to 1200 °C in a fusion power reactor [1,2]. In this study we, therefore, investigated the synergetic effect of simultaneous triple irradiation of He, H and Si ions on microstructural change of SiC/SiC composites at higher temperature than 800 °C and at almost the same He/dpa and H/dpa ratio under fusion conditions.

We adopted two kinds of the advanced SiC fiber fabrics that were near-stoichiometric SiC fibers: Hi-Nicalon type S [9] and Tyranno SA [10] in this study, because these fiber fabrics possess superior mechanical and thermal properties as compared with their predecessors such as ceramic grade Nicalon and Hi-Nicalon. Snead et al. reported the effect of neutron irradiation on mechanical property of SiC/SiC composites using ceramic grade Nicalon or Hi-Nicalon fiber fabric, and the strength were reduced by irradiation because of the degradation in interfacial strength [11,12]. This degradation in interfacial strength was directly measured following irradiation and attributed to densification of the ceramic grade Nicalon or Hi-Nicalon fiber [11,12]. Therefore the dimensional stability of the composites using these advanced fiber fabrics following triple ion beam irradiation was also investigated.

2. Experimental procedure

2.1. Materials and preparation

The 2D plane weave of Hi-Nicalon type S and Tyranno SA SiC fiber fabrics were chosen in this study. The two types of SiC/SiC composites examined in this study were fabricated using these fibers by forced thermal gradient chemical vapor infiltration (F-CVI) method at Oak Ridge National Laboratory. Carbon layers as the interphase layers between fiber and matrix were applied prior to F-CVI method to fabricate SiC/SiC composites. The fabrication conditions of both composites by F-CVI were almost the same. Igawa et al. reported details of the fabrication procedure [13]. The characterization of both composites was shown in Table 1. The surface of the specimen was polished by grinding wheel with diamond abrasive (average grain size: 1 μm). The shape of specimen was 1 mm width, 5 mm length and 0.4 mm thickness.

2.2. Irradiation

The triple ion irradiation was carried out at TIARA (Takasaki Ion Accelerators for Advanced Radiation Application) facility of JAERI. The specimens were simultaneously irradiated at 1000 °C by 6.00 MeV Si^{2+} ions, 1.20 MeV He^{+} and 250 keV H^{+} ions. He^{+} ion implantation was performed using an aluminum foil energy degrader in order to control He distribution in the depth range of about 1.5–2.4 μm from the specimen surface. Fig. 1 shows the distributions of He and H concentration and displacement damage as a function of depth from the surface in SiC calculated by TRIM code [14]. In this study, the displacement threshold energies of Si and C were assumed to be 35 and 20 eV, respectively [15]. The irradiation was performed to 10 dpa at the depth of 2 μm as shown in Fig. 1. The resultant He/dpa and H/dpa ratio were 60 and 50 appm/dpa, respectively, which would correspond to a region behind the first wall of a fusion power reactor.

2.3. Microstructure observation

The focused ion beam processing was used to prepare foil specimens for transmission electron microscopy (TEM) observation. The foil specimens contained the fiber, matrix and interphase layer in the irradiation and non-irradiation regions as shown in Fig. 2. Microstruc-

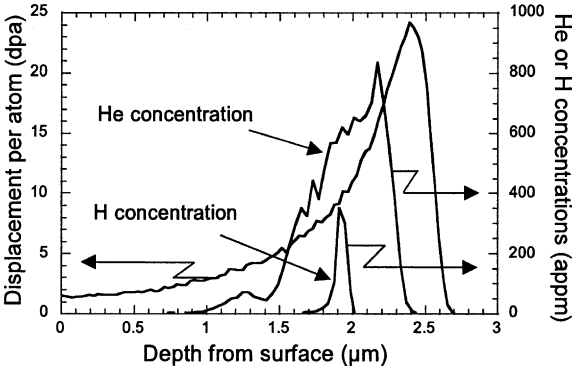

Fig. 1. Distribution of He, H concentrations and displacement damage as a function of depth from surface in SiC calculated by TRIM code.

Table 1
Characterization of the SiC/SiC composites fabricated by F-CVI method

Composite ID	Kind of fiber fabric	Fiber volume fraction (%)	Density (g/cm^3)	Porosity (%)	Average thickness of carbon layer (nm)
HNS	Hi-Nicalon type S	33	2.43	23.5	159
TSA	Tyranno SA	37	2.67	15.1	107

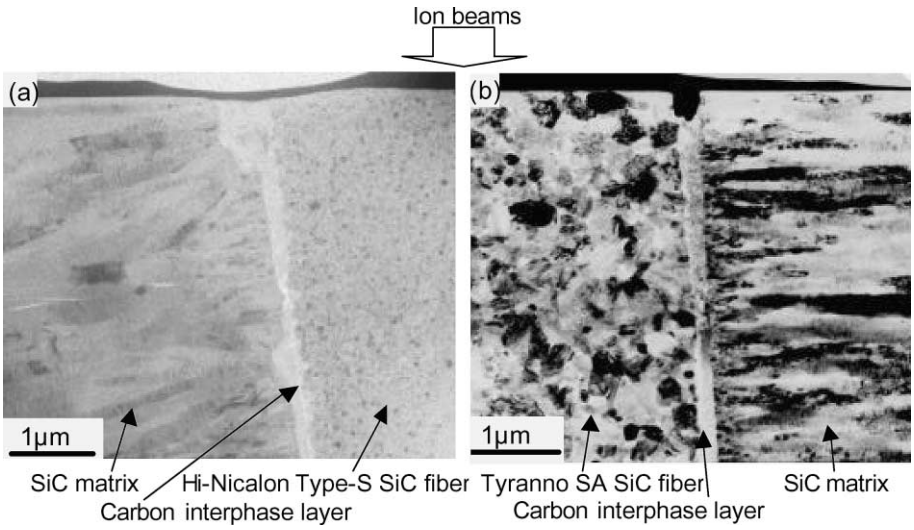

Fig. 2. Cross-section TEM microphotographs of (a) HNS and (b) TSA composites irradiated at 1000 °C by simultaneous triple ion beams.

tural observation was performed with Hitachi HF-2000 field-emission TEM operated at 200 kV.

3. Results and discussion

The cross-section TEM microphotographs of both SiC/SiC composites (HNS and TSA composites) irradiated at 1000 °C under simultaneous triple ion beams are shown in Fig. 2. Both composites have a 150 nm carbon layer as an interphase layer between fiber and matrix. However, the thicker carbon regions at the vicinity of the surface and 3.5 μm depth from the surface were observed in the HNS composite only. These thicker carbon regions were produced by fabrication process, not the effect of ion beams irradiation. The reason is that the region at 3.5 μm depth from the surface is non-irradiated area as shown in Fig. 1. In the matrices of both composites, β-SiC grains grew in the radial direction of the SiC fibers and formed elongated grains. The Hi-Nicalon type S fiber consists of randomly oriented fine β-SiC grains (~20 nm). And the Tyranno SA consists of larger β-SiC grains (~100 nm) than those in Hi-Nicalon type S.

The cross-section TEM microphotographs of the irradiated SiC matrices at 1.9 μm depth from the surface (displacement damage with implanted gases region) in either composites were shown in Fig. 3. Amorphization was not observed in both matrices. Snead and Zinkle reported that amorphization of SiC by irradiation did not occur at above 200 °C [16]. He bubbles and some cracks were observed in the matrix of the HNS composite. The average size of He bubbles and these cracks were 5.9 and 512 nm, respectively. These He bubbles and cracks were formed in the range of 1.7–2.5 and 2–2.5 μm depth from the surface, respectively. Almost all of He bubbles and cracks were formed at the grain boundaries in the matrix. It is considered that the cracks

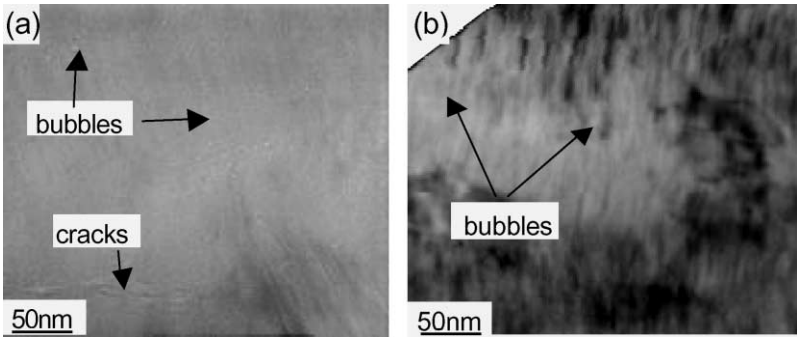

Fig. 3. Cross-section TEM microphotographs of SiC matrix in (a) HNS and (b) TSA composites.

would be produced by He bubble concentration at grain boundary. Few He bubbles were also observed in the SiC matrix of the TSA composite, but cracks were not. The average size of He bubbles was 7.4 nm. The size of He bubbles in the matrices of both composites was almost the same. The number of He bubbles was, however, much smaller than that of SiC matrix in HNS composite. It is, therefore, considered that the cracks were not formed in the matrix of TSA composite.

Neither cracks nor He bubbles were observed in both Hi-Nicalon type S and Tyranno SA. He bubbles are formed by combining He ion with vacancies. Because the vacancies induced by the irradiation reached the grain boundaries soon owing to the small grain size (~100 nm), the concentration of vacancies was low. Therefore the frequency of combining the implanted He ions with the vacancy reduced. It was considered that this reduction prevented He bubbles from growing.

The lack of He bubbles in both the SiC matrix and Tyranno SA of TSA composite may be considered to be the difference of impurities in the composites. Tyranno SA fiber included 2 wt% aluminum as the impurity [10,17]. During the fabrication process of the composite in TSA composite, the impurity of Tyranno SA fiber might be diffused into the matrix. The amount of impurity in TSA composite would be, therefore, much higher than that of HNS composite. The cause of no formation of He bubbles in Tyranno SA fiber and the suppression of cavity formation in the matrix may be caused by the degradation of mobility of vacancy or/and helium atoms, owing to the interaction of impurity atoms with it. And another reason is also considered that the irradiation temperature between HNS and TSA composite might be slightly different.

Sasaki et al. reported He bubbles formation in B-doped sintered β-SiC after neutron irradiation above 1400 °C (total He produced by neutron irradiation was 2000 appm and the displacement damage dose of the materials was less than 1 dpa) [18]. Hojou and Izui reported that He bubbles were formed in a sintered SiC implanted by more than 40 000 appm He ions at 1000 °C [6]. The concentration of the implanted He of this study, which was about 600 appm as shown in Fig. 1, was much smaller than those of the previous studies. He bubbles in the SiC matrices were nevertheless formed at around 1000 °C in this study. The displacement damage of this study was much larger than those of the previous studies. This result, therefore, indicated the synergetic effect of the high displacement damage (~10 dpa) and implanted gases on the microstructural change of the composites. It is also found that He bubbles formation at around 1000 °C depends strongly on the amount of implanted He, displacement damage and impurities of the composites.

From the above results, it was found that TSA composite showed better microstructural stability against ion beams irradiation compared to the HNS composites.

The cross-section TEM microphotographs of carbon interphase layers and the corresponding selected area electron diffraction patterns are shown in Fig. 4. A microstructural change of the carbon interphase layer was found in the irradiation region. The carbon interphase layer in the non-irradiation region consists of a non-graphitic carbon structure which contained very small turbostratic units in random array that were cross-linked by disorganized carbon, while the turbostratic carbon structure was formed in the irradiated region

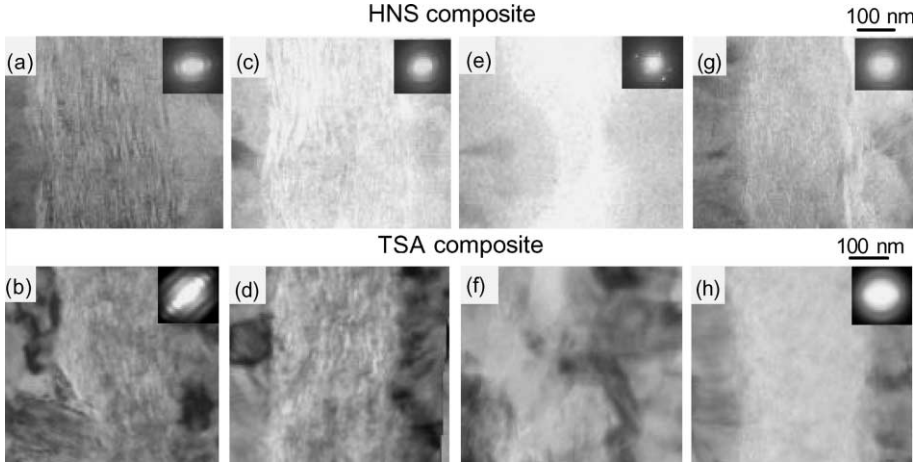

Fig. 4. Cross-section TEM microphotographs of carbon interphase layers and the corresponding selected area electron diffraction patterns for only damage region (a and b), damage with gas implanted region (c and d), projected range Si ion (e and f) and non-irradiation region (g and h).

(region of displacement damage only and region with displacement damage with implanted gases). Though the microstructures in the displacement damage region and in the displacement damage with implanted gases region of carbon interphase layers of both composites were contracted, the effect of implanted gases on microstructural changes was not clearly observed. Also He bubbles were not observed in the carbon interphase layer of both composites. The diffusion coefficient of He in carbon was more than about 30 times larger than in SiC at 1000 °C [19], and almost all implanted He ions in the carbon interphase layer were released during the irradiation at 1000 °C. To form turbostratic carbon structures, a threshold displacement damage and the presence of He atoms may be necessary. A new crystalline phase was formed in the carbon interphase layer of the projected range of Si ions. Roe et al. reported that a SiC layer was formed from a diamond layer implanted with up to 2.0 at.% Si and annealing at 800 °C [20]. The concentration of Si atoms at the projected range of Si ions in both composites was about 2.5 at.% calculated by TRIM code [14]. Therefore the new crystalline phase was thought to be SiC.

According to previous reports [8], the microstructure observation of the ion irradiated SiC/SiC composites (using Hi-Nicalon fiber with a carbon interphase layer) at 800 °C exhibited the debonding at the boundary between the carbon interphase layer and the fibers. This debonding was due to the shrinkage of Hi-Nicalon fiber induced by ion beams irradiation. While the microstructure observation of both composites irradiated in this study did not exhibit the debonding in the irradiation region of the interphases between carbon layer and fiber. The reason is that the shrinkage of the advanced SiC fibers did not occur by the irradiation because of their near-stoichiometry and highly crystalline SiC structure. It was found that the composites using Hi-Nicalon type S or Tyranno SA fibers had an excellent dimensional stability of the interphase between fiber and carbon interphase layer against ion beam irradiation at around 1000 °C.

4. Conclusions

The synergetic effect of simultaneous triple irradiation of He, H and Si ions on the microstructural change of SiC/SiC composites (Hi-Nicalon type S or Tyranno SA fiber with carbon interphase layer) at 1000 °C has been investigated. The He/dpa and H/dpa ratios were about 60 and 50 appm/dpa, which are relevant to the fusion condition. The microstructures of SiC/SiC composites irradiated to 10 dpa were examined by TEM. The following results were obtained:

(1) He bubbles and cracks were observed in the SiC matrix of HNS composite, and few He bubbles were observed in the SiC matrix of TSA composite. On the other hand, He bubbles and cracks were not observed in Hi-Nicalon type S fibers and Tyranno SA fibers. The average size of He bubbles was almost the same in SiC matrices of both composites, but the amount of He bubbles in TSA was much smaller compared to HNS composite. It is speculated that the impurity in TSA composite prevented He ions from moving easily. It was, therefore, found that TSA composite showed a better microstructural stability against ion beam irradiation in comparison to HNS composite.

(2) The carbon interphase layer showed a non-graphitic carbon structure in the non-irradiation region, while the turbostratic carbon structure was formed in the irradiated region. A new crystalline SiC-like phase was formed in the carbon interphase layer of the projected range of Si ions.

(3) Debonding between fibers and carbon layers in the irradiation region was not observed in HNS and TSA composites. Microstructural analyses showed that SiC/SiC composites using advanced SiC fibers had excellent dimensional stability of the interphase between fibers and carbon interphase layers against ion beams irradiation at around 1000 °C.

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