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Microstructural stability of SiC and SiC/SiC composites under high temperature irradiation environment

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Abstract

Silicon carbide continuous fiber-reinforced silicon carbide matrix composites (SiC/SiC composite) are attractive as the structural material of advanced energy systems, including nuclear fusion. The irradiation may affect the fiber/matrix interphases which are responsible for the pseudo-ductile fracture behavior of SiC/SiC composites. In this work, the investigation of the microstructural evolution of SiC/SiC composites in a fusion environment is performed by the dual-ion irradiation method. Reinforcements were Tyranno™-SA and Hi-Nicalon™ Type-S. The displacement damage rate was up to 100 dpa. The irradiation temperature and He/dpa ratio were up to 1673 K and 60 appm, respectively. The microstructural modification induced by the dual-ion irradiation especially occurred in the interphase. The advanced SiC fiber did not shrink and the C/SiC multilayer interphase showed a superior microstructural stability against the dual-ion irradiation at high temperatures.

1. Introduction

Silicon carbide continuous fiber-reinforced silicon carbide matrix composites (SiC/SiC composites) have a lot of attractive properties, such as high strength, high chemical stability and low activation in thermo-nuclear applications. The low activation property makes SiC/SiC composites to be the most desirable material for the components of the advanced blanket system in fusion reactors [1].

Under an irradiation environment, the swelling and degradation of strength are the most essential mechanical and structural issues. The irradiation affects each constituent of the SiC/SiC composite and these effects are different for fiber, matrix and interphase. The recent development of SiC fiber reduces the oxide concentration and its composition and is close to stoichiometric SiC. These advanced SiC reinforcement has a crystallized microstructure. Due to the enhanced crystallinity the reinforcement behaves similar to the matrix in an irradiation environment. This is expected to keep the interphase stability and mechanical property of SiC/SiC composites [2].

The irradiation affects the carbon layer in the interphase. A most pronounced irradiation effect is the disorder of the graphitic structure. The irradiation increases the basal plane interspacing in the graphitic lattice and results in an anisotropic volume change. The improvement of the interphase structure is necessary to reduce this influence, so a C/SiC multilayer interphase structure is suggested for SiC/SiC composites [3].

The evaluation method for fusion materials is one of the important studies for materials development. A fusion reactor and a 14 MeV fast neutron source do not yet exist, so fission reactors are used for neutron irradiation experiments. The primary knock-on atom (PKA) energy and irradiation temperature in a fission reactor are different from those in a fusion reactor [4]. Especially in a fusion reactor, 14 MeV neutrons cause helium production from (n, α) nuclear reactions. The

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insoluble helium gas is considered to affect the irradiation induced displacement damage behavior [5]. For structural materials of the blanket system in a fusion reactor, SiC/SiC composites are exposed to high temperatures and heavy irradiation environments. The temperature and the neutron dose are estimated to be about 1273 K and up to 100 dpa, respectively [6].

For the evaluation of materials under this severe condition, the dual-ion irradiation method is suitable [7,8]. The ion irradiation method has been studied for the simulation of 14 MeV fast neutron irradiation. An incident ion particle has enough PKA energy to induce a cascade damage similar to 14 MeV fast neutrons. In addition, the ion irradiation allows to modify the irradiation condition. Two different species are simultaneously irradiated to a material in a dual-ion irradiation. Heavy ions are used for inducing displacement damage and helium ions are implanted to play the part of produced helium by nuclear reactions under fusion conditions. The evaluation of helium production in SiC is almost impossible in a fission reactor, the dual-ion irradiation is more important to investigate its property in a fusion environment.

The objective of this work is to evaluate irradiation effects under near-fusion environment by the dual-ion irradiation and TEM observation on the microstructural stability of advanced SiC reinforced SiC composites [9,10].

2. Experimental

The material used in this study are advanced SiC fiber-reinforced SiC composites. Reinforcements are Hi-Nicalon™ Type-S (Nippon Carbon Co.) and Tyranno-SA™ (Ube Industries Co.), which are near-stoichiometric SiC fibers with low oxygen and high crystallinity. The composites were produced by the chemical vapor infiltration (CVI) method. The interphases consists of a pyrolytic carbon (PyC) layer and a C/SiC multilayer, which is developed for fusion applications. The thickness of the PyC interphase was about 600 nm. The C/SiC multilayer interphase has five thin carbon layers. The thickness of each layer in the multilayer is about 20 nm. The CVI processing of materials was performed at Oak Ridge National Laboratory (ORNL). For an ion-beam irradiation, the composites were cut to square shape and the irradiated surface was polished by diamond powders. The dimensions of the specimens were 4.0 mm × 2.0 mm × 2.0 mm. The irradiation surfaces were chosen to be normal to the fiber direction. The ion-beam irradiation was carried out at the Dual-Beam for Energy Technology (DuET) Facility, Kyoto University. The specimens were irradiated with 5.1 MeV silicon ions and simultaneously implanted with 650 keV helium ions. The energy of the helium ions was degraded and slowed down by a thin aluminum foil degrader placed in the irradiation chamber. The depth profiles of damage and ions were calculated by the TRIM-98 code. The sublattice-averaged displacement energy and the density of SiC are assumed to be 35 eV and 3.21 g/cm^3, respectively. The damage level was defined as the average of the dual-ion irradiation region. The irradiation was performed up to 100 dpa. Irradiation temperature and flux were up to 1473 K and 1×10^{-3} dpa/s, respectively. The He/dpa ratio of simultaneously irradiated helium was 60 appm. The SiC composites were sliced about 0.1–0.3 mm thick by a dicing saw. A focused ion beam (FIB) processing [3] was performed to prepare thin foils for microstructural investigations by transmission electron microscopy (TEM). The foil orientation was normal to the irradiation surface. The microstructural investigation was performed with a conventional JEOL JEM-2010 TEM.

3. Results

The cross-sectional TEM micrographs of the carbon interphase in a dual-ion irradiated Tyranno-SA/PyC/CVD-SiC composite are shown in Fig. 1. Fig. 1(a) was taken near the surface and Fig. 1(b) is a dual-ion irradiated interphase. Fig. 1(c) shows the unirradiated interphase of the PyC layer. The irradiation direction is from the upper side of the micrograph. In Fig. 1(a), the fiber surface is flat to that of matrix; shrinkage of the fiber by irradiation induced crystallization does not occur and the expansion rates of fiber and matrix are almost the same. In the interphase, the dual-ion irradiated carbon layer in Fig. 1(b) significantly expands compared with the unirradiated layer in Fig 1(c). Anisotropic volume change occurred by irradiation induced disorder of carbon atoms and the increasing rate of carbon layer thickness is 16.7%. In Fig. 1(a), part of the carbon layer is lost near the surface.

The TEM micrograph of a dual-ion irradiated Tyranno-SA/Multilayer/CVD-SiC composite is shown in Fig. 2. This specimen consists of five carbon layers in the interphase. The dual-ion irradiated region where helium is implanted is limited to a range of 400–1400 nm depth from the surface. Each layer of the interphase does not lose its surface and significant expansion is not observed. The thickness of each carbon layer decreases with increasing displacement damage. The decreasing rate of the thickness is about −0.1 nm/dpa. In the dual-ion irradiated region of the matrix, the production of helium cavities was observed which are shown in Fig. 3. The helium cavities were mainly produced on the grain boundaries. There were no helium cavities outside the dual-ion irradiation region.

Helium microcavities are observed in SiC layers between carbon layers of the interphase, and also on the

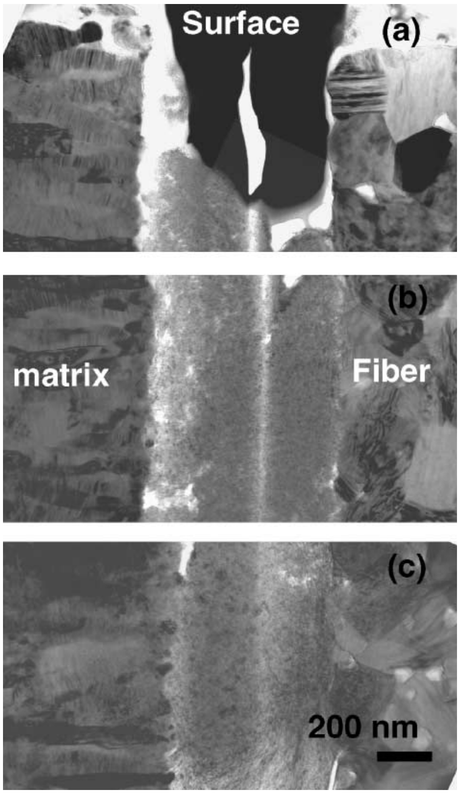

Fig. 1. Cross-sectional TEM image of PyC interphase in Tyranno-SA/PyC/CVI-SiC composite irradiated at 1273 K and 100 dpa. (a) Near surface, (b) dual-ion irradiated range and (c) unirradiated range.

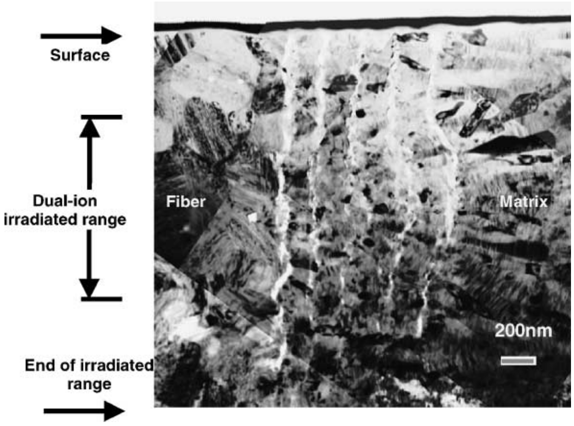

Fig. 2. TEM micrograph of Tyranno-SA/multilayer/SiC-CVI composite after dual-ion irradiation up to 100 dpa at 1273 K.

grain boundaries of Tyranno-SA fiber. TEM images of microcavities on the grain boundaries in Tyranno-SA fiber are shown in Fig. 4. The Hi-Nicalon Type-S/multilayer/CVI-SiC composite irradiated at 1273 K up to 100 dpa shows the same modification as the Tyranno-SA/multilayer/CVI-SiC composite except for the microcavity production in the fiber. Helium cavities are not observed in the dual-ion irradiated Hi-Nicalon Type-S fiber. The evaluation results of cavities are shown in Fig. 5. The swelling by cavities increases with increasing displacement damage rate, and the amount of swelling is about 0.3% at 100 dpa. The cavity density almost decreases with increasing displacement damage. The mean cavity radius increases proportionally with the displacement damage from about 3 nm at 50 dpa to 5 nm at 100 dpa. The cavities are not recognized in higher displacement damage conditions outside the helium implanted region. The observed cavities are not the voids by displacement damage but the helium cavities.

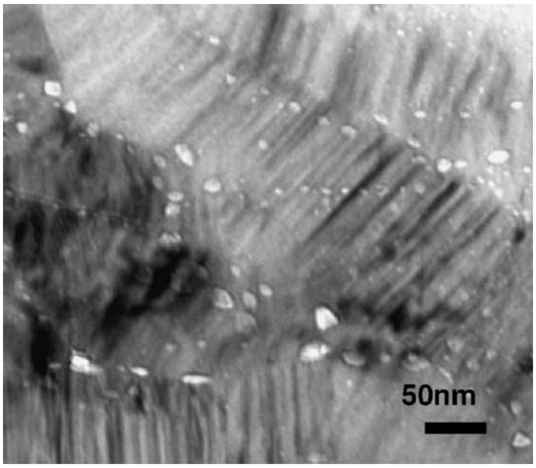

Fig. 3. Helium cavities production on grain boundary in dual-ion irradiated region of matrix SiC.

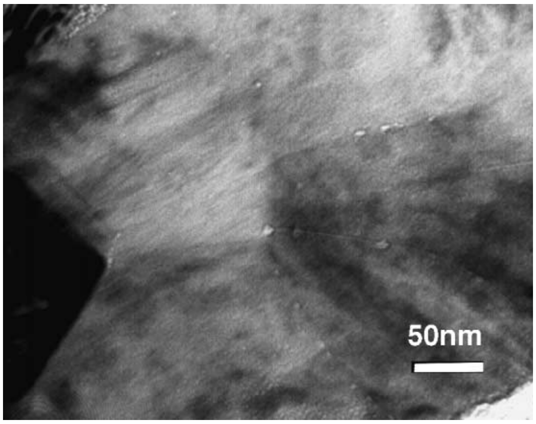

Fig. 4. TEM image of microcavities on grain boundary in dual-ion irradiated region of Tyranno-SA fiber.

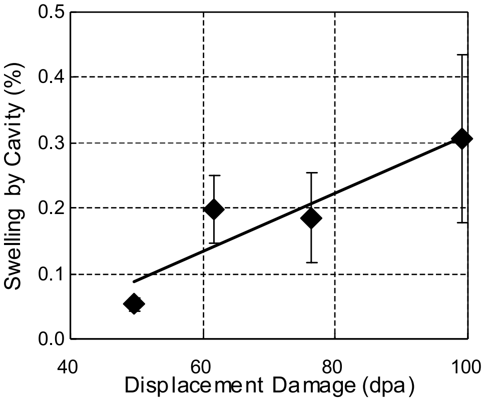

Fig. 5. Swelling by helium cavity in matrix SiC irradiated at 1273 K.

4. Discussion

Tyranno-SA fiber is developed for fusion applications to guarantee microstructural stability under a heavy irradiation environment. This fiber almost behaves similar as the SiC matrix. The irradiation conditions of the present work have been established close to a fusion environment, in order to evaluate the effects after heavy irradiation in a fusion reactor. Advanced SiC fibers are promising candidates for the reinforcement for structural materials in a fusion reactor.

The interphase structure is one of the key issues in SiC/SiC composites. A PyC is normally used as the interphase of composites. As shown in Fig. 1, advanced fibers show enough stability, but the interphase expansion is too large and results in the deformation of matrix and fiber. At the near surface of the interphase, the carbon layer is significantly lost in Fig. 1(a). The sputtering by irradiation mainly seems to cause this loss. The effect of carbon disorder on the mechanical property still remains unclear, but the loss of the carbon layer shows the possibility for an embrittlement by the accumulation of disorder. From these results, large volumes of carbon layers are apparently unsuitable for fusion materials [11].

For the improvement of the microstructural stability in the composite, it is necessary to improve not only the fiber but also the interphase structure. The C/SiC multilayer interphase is developed to improve the property against irradiation and oxidation [12]. In the present work, the multilayer interphase improves the effects of volume expansion and sputtering. On the other hand, an increase of the displacement damage results in a decrease of the thickness of each carbon layer in interphase. The irradiation induced carbon mixing and slight swelling of SiC are estimated to be caused by this phenomenon. The depth of 5.1 MeV Si ion deposition is calculated to be $\simeq$2500 nm, chemical reactions do not reduce the thickness of the carbon layer in a dual-ion irradiated region. The effects of interphase thickness reduction are not sure, one possibility is the degradation of the mechanical properties caused by the loss of the carbon layer in the interphase. Another possibility is an increase of the interface toughness of matrix and fiber caused by the slight swelling of the SiC in matrix, interphase and fiber. For the development of the interphase structure, it is necessary to consider both interphase expansion and loss of carbon layer.

At 1273 K, the effect of helium becomes important because of swelling by the helium cavity. The void swelling regime of SiC is above 1473 K [13]. Due to helium implantation followed by annealing, helium precipitates on the grain boundary of the matrix by annealing at 1673 K for 1 h [14]. The production of cavities occurs at about 1273 K in SiC under a fusion environment. The temperature of 1273 K is in the range of the operation temperature of a fusion blanket using SiC/SiC composite.

The cavities in a matrix grow with increasing displacement damage rate. The cavity density seems to have a peak against the displacement damage because the size of most cavities are too small to be detected in the TEM micrograph at low dose rates. Actually, at relatively low doses, a large quantity of microcavities are estimated to exist in the materials.

The swelling by cavities is about 0.3% at 100 dpa in the present work [15]. The evaluation of swelling by cavities was analyzed by TEM micrographs; the influence of swelling by point-defects is not ignorable. A previous study of neutron irradiation reported that the volume swelling at 1273 K was about 0.5% [3]. The behavior of cavities is not independent of point-defects, but the total swelling by cavities and point-defects in a SiC matrix is estimated to be lower than 1% at 100 dpa and 1273 K.

The crystallized SiC fibers have smaller grains and much more grain boundaries than the SiC matrix. Due to the high grain boundary density, the precipitation of helium cavities is hard to detect. In Hi-Nicalon Type-S fibers, which have smaller grains, grain boundaries have enough capacity to keep helium without precipitation. Tyranno-SA fiber has a larger grain size which is close to that of the SiC matrix, therefore, microcavities precipitate on the grain boundaries. From these results, the effect of cavities on advanced SiC fibers is different from that on the SiC matrix. The large amount of helium cavity production makes the interphase unstable because of the difference in the rate of swelling. In the present condition, the dimensional behavior of the fiber is not different from that of the SiC matrix.

In the present work, at the early stage of helium cavity production, the swelling by cavities in SiC is small enough. But it is not sufficient to determine the critical swelling rate at 1273 K. The production of helium cavities means helium migration at 1273 K, therefore, the flux rate of irradiation becomes important. At 873 K, the flux rate does not significantly affect the point-defect swelling because of small migration [16]. But at 1273 K, the point-defect migration becomes larger and helium migration occurs, the evaluation of the microstructural behavior depending on the flux rate is important.

5. Conclusion

Dual-ion irradiation studies have been performed for the advanced silicon carbide continuous fiber-reinforced silicon carbide matrix composite (SiC/SiC composite) for fusion application. The purpose of this work is to study the microstructural evolution of the interphase in a high-temperature, high-fluence irradiation and helium existence environment. The reinforcements were Tyranno-SA and Hi-Nicalon Type-S. The interphase structures were C/SiC multilayers and PyC layers. The irradiation temperature, averaged displacement damage rate and helium ratio were 1273 K, 100 dpa and 60 appm/dpa, respectively.

The C/SiC multilayer interphase is suggested to improve the microstructural stability and showed superior dimension stability in an irradiation environment. But a reduction in the thickness of each carbon layer occurred. The irradiation induced carbon mixing or slight swelling of SiC are estimated to cause this reduction.

The helium cavity production occurred at 1273 K and the amount of swelling by cavities was about 0.3% at 100 dpa. In the present work, the swelling by cavities is small enough but the flux dependence and higher temperature investigations are necessary to determine the swelling effect in a fusion environment.

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