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Study of helium effects in SiC/SiC composites under fusion reactor environment

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Abstract

Helium release behavior from He-ion implanted SiC/SiC, monolithic β-SiC, and SiC fiber (Hi-Nicalon) was studied using the thermal desorption method with annealing temperatures between 500°C and 1600°C in 100°C increments. Helium release from SiC/SiC composites began below 500°C, while from monolithic β-SiC, helium release was observed above 1000°C. The magnitude of the helium released from β-SiC was 1/10 to 1/50 of the helium released from the composites. Helium release from the fiber became apparent above 1300°C. Helium release from composites at low temperature might be attributed to helium release from the carbon interphase. Behavior of transmuted helium in the SiC/SiC composites under fusion reactor conditions is discussed.

1. Introduction

Silicon carbide (SiC) has been proposed for use as a fusion-reactor structural material due to its low-induced activity after 14 MeV neutron irradiation and its high strength at high temperature. Since monolithic SiC is brittle, SiC-fiber-reinforced SiC-matrix composites (SiCf/SiC) have been developed to increase its toughness as a structural material. The recent status of the SiCf/SiC composites for fusion has been reviewed in [1–3].

In a fusion reactor environment, helium (He) will be produced in SiC in the first wall region at a rate of about 1500–2000 at. ppm He/(MWy/m^2) by transmutation reactions, with a concurrent displacement damage of 10–15 dpa/(MWy/m^2), depending on the details of the blanket structure and neutron spectrum [1,2]. In the end of its life (about 100 dpa), the He concentration will be 15 000–20 000 at. ppm. As the solubility of He is almost zero in all materials, and it stabilizes vacancy clusters, microstructural changes such as swelling and strength degradation may be accelerated in SiC/SiC composites under fusion reactor conditions.

Our previous work showed that after 1400°C annealing of helium implanted SiC/SiC composites, helium bubble formation was observed only in the SiC matrix of the SiC/SiC composite [4]. No bubbles were observed in the C interphase or in the SiC fiber. In order to predict material degradation by the generation of a large amount of He and displacement damage, an understanding of the behavior of He atoms in SiC/SiC composite is important. The behavior of He in monolithic SiC and graphite has been studied by several researchers [5–8] using He-release measurements and TEM observation, but the behavior of He atoms in SiCf/SiC composites has not been clarified. The purpose of this work is to study the He behavior in SiCf/SiC composites to predict transmuted He effects on microstructural development of SiC/SiC.

2. Experimental procedure

In this work, two types of composites (composites 1 and 2) were examined. The constituent materials, monolithic β-SiC and SiC fibers, were also examined. A two-dimensional SiCf/SiC composite reinforced by SiC weaves made of Hi-Nicalon fibers were used as

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specimens. Hi-Nicalon fiber weaves in the composites were coated by graphite using a chemical vapor deposition (CVD) process and β-SiC matrices were deposited on SiC weaves by chemical vapor infiltration (CVI). The resulting carbon coating thickness was approximately 1.2 μm for composite 1 and 0.15 μm for composite 2. Both composites have high fracture strength (500–600 MPa) and relatively high density (about 2.5 g/cm^3) [9]. These composites were fabricated by Dupont Lanxide in the USA. The composites were manufactured in the form of plates. Bend bars of width 4 mm, length 25 mm, and thickness 2 mm were cut from the plates. Implanted specimens were cut from the bars as thin plates. The implanted surface was a cross-sectional surface of the bend bar, and the implanted surface was polished using 1 μm diamond paste.

A monolithic, CVD processed, high purity β-SiC was supplied by Mitsui Heavy Industry as a 5-inch dummy disk for semiconductor process. The specimens were cut to 2 mm in width, 3 mm in length, and 0.3 mm in thickness. Hi-Nicalon fibers were used as the SiC fiber specimen. The fiber diameter was about 14 μm, and fibers of 60 mm length were used as specimens. The sum of the implanted volume of these fiber specimens was about 1/10 of the monolithic SiC specimen.

Helium implantation was carried out using a dynamitron accelerator at Tohoku University. 3.00 and 2.95 MeV He ion implantations with a degrader system were utilized to obtain a smooth depth distribution of He in the specimen. The degrader wheel consists of 11-step thickness aluminum foils. The He implantation was conducted two times for each specimen, firstly irradiation by 3.00 MeV He ions, and then by 2.95 MeV He ions. The calculated He distribution in the SiC is shown in Fig. 1.

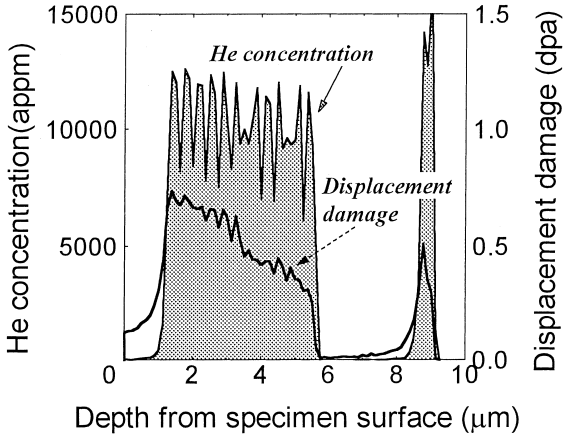

Fig. 1. Calculated depth distribution of implanted He in SiC by TRIM [10].

The total implanted helium concentration in SiC calculated from the irradiation fluence was about 10 000 at. ppm in the range from 1 to 5.5 μm depth. Displacement damage in the He-implanted volume was approximately 0.5 dpa using $E_d = 45$ eV. The implantation temperature was about 100°C, as measured by an infrared pyrometer. Fig. 2 shows a schematic view of the irradiation conditions and specimen geometry with material data.

Helium release from each specimen was determined by static-mode isotope dilution gas mass spectrometry during heating in a resistance-heated graphite crucible that was placed in a high-temperature vacuum furnace [11]. The absolute amount of ^{4}He released was measured relative to a known quantity of added ^{3}He 'spike'. The ^{3}He spikes were obtained by expanding and partitioning

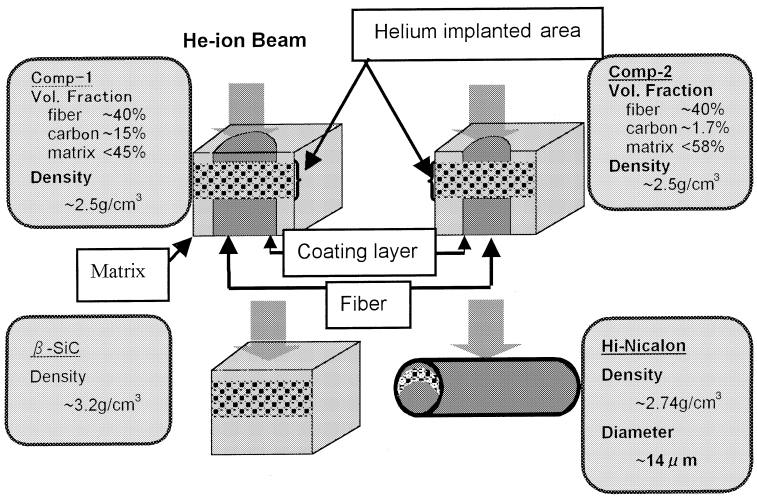

Fig. 2. Schematic view of helium implantation and specimen data.

a known quantity of gas through a succession of calibrated volumes [12]. The mass spectrometer was calibrated for mass sensitivity during each series of runs by analyzing the known mixture of ^{3}He and ^{4}He.

The stepped-anneal analysis was conducted while the temperature of the sample crucible was increased in 100°C increments, starting at 500°C and increasing to a maximum of between 1400°C and 1600°C. Each target temperature was held for a period of 30 min. The time period between each analysis was varied from a minimum of about 5 min to a maximum of about 10 min depending on the observed ^{4}He release rate at each temperature. To compare helium release between an unimplanted sample and an implanted one, He release from unimplanted composite 1 was measured as 'unimplanted 1'.

Following the temperature dependence of He release measurement, the specimens were relocated, in their original crucible, to different locations in the same furnace for subsequent high-temperature analyses to estimate the total amount of implanted He in the specimen. For the high-temperature analyses, the graphite crucibles were heated to their maximum sustainable temperature for a period of 10 to 20 s. Crucible temperature is estimated to approach 3000°C during this process. The melting point of SiC is approximately 2800°C, and therefore, He remaining after the step-annealing measurement was released from the specimen during this heat treatment.

3. Results and discussion

The results of the helium release measurements are given in Fig. 3. On the abscissa is the cumulative annealing time from the start of an analysis. The annealing temperature history is shown as a stepped solid line in this figure. Measured helium levels have been corrected for background helium build-up in the analysis furnace as a function of time. The background ^{4}He level, which is shown as a dashed line in Fig. 3, generally rises linearly with time and is largely due to the diffusion of helium from the atmosphere through the pyrex furnace top. The background helium build up rate of this system was 7.5×10^{-8} atoms/s.

The observed helium releases during the stepped-anneal measurements were non-linear with time at each temperature level. Fig. 4 shows the desorption rate of He during each sampling period. Helium release showed step like behavior at each temperature, with each step being characterized by a fairly rapid initial release rate, followed by a leveling off of the rate with time. The time dependence of the He release rate at each annealing temperature shows that the He diffuses out from the implanted region. The decay curve of the release rate shows that the holding time of 30 min at each annealing temperature can be considered long enough to accumulate released helium from the implanted area by diffusional processes.

The total amount of released He after the 3000°C anneal is about 40% of implanted number of He atoms estimated by beam current. The lower than expected amount might be attributable to scattering of beam by the degrader system. To compensate for both the uncertainty in the actual number of implanted He atoms and the difference of implanted volume between the fibers and the other specimens, the ratio of increments of accumulated He at each temperature (ΔN_T^S) to the sum of the total released He atoms by step annealing ($\sum N_T^S$) and that by high-temperature measurement (N^H) was used. The results are shown in Fig. 5. Except for the implanted composite 1 specimens, the He release from implanted specimens generally increased with increasing temperature, with the majority of the He being released

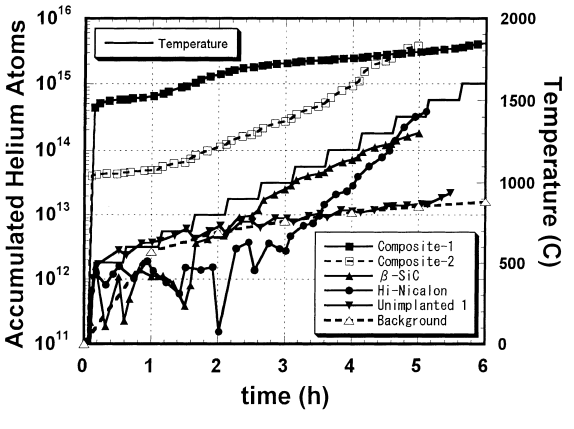

Fig. 3. Helium accumulation curve during step annealing.

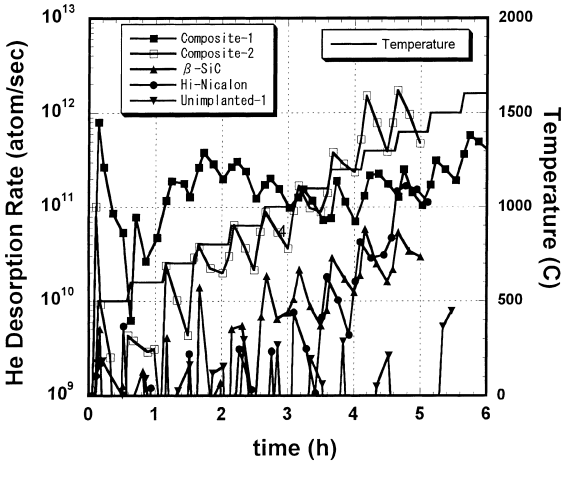

Fig. 4. Helium desorption rate at sampling period.

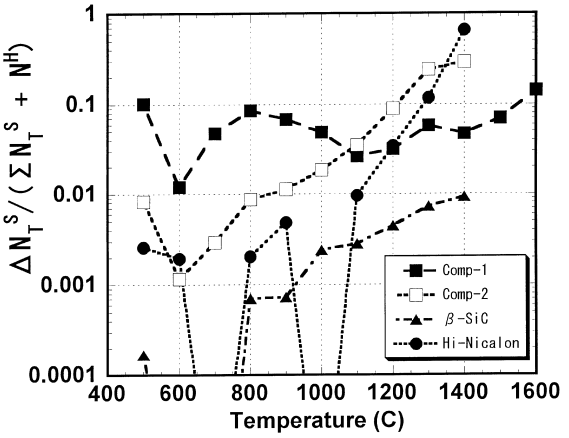

Fig. 5. Temperature dependance of accumulation rate to total dissolved helium.

above about 1000°C. The implanted composite 1 specimen, however, showed the highest incremental helium release at the lowest temperature step of 500°C, with additional significant releases at 800°C and 1600°C.

The He release peaks from the implanted composites at 500°C and 800°C are considered to be He release from the C interphase of the composites. The magnitude of the He release rate of the composite 2 specimen at 500°C and 800°C was about 1/10 of composite 1. Taking into account the difference in C volume fraction of these composites, the He release in these temperature regions is due to the release from the C phase. Atsumi et al. [8] showed that He released from graphite below 400°C.

Helium release from monolithic SiC was observed above 1000°C. Compared to the implanted composite, the magnitude of the He release rate of monolithic SiC was about 1/10 to 1/50. However, the temperature dependence of He release from monolithic SiC was similar to that of composite 2 and Hi-Nicalon above 1000°C. Therefore, He release of composites above 1000°C mainly comes from the SiC part in the composites such as the SiC matrix and the SiC fibers. The difference in magnitude of the release rate between the composites and the monolithic-SiC may be attributed to microstructural differences, such as C coating layer, small cracks and the growth boundary of β-SiC existing in composites. These features may enhance the He release by acting as diffusion channels.

A large He release from SiC fiber was observed above 1100°C, and it became larger than that of the monolithic SiC above 1300°C. This difference between the SiC matrix and the SiC fiber also comes from microstructural difference between these materials. Hi-Nicalon fiber consists of SiC micro-grains with a diameter of about 5 nm, and a small amount of C exists between each SiC micro-grain [13]. The C phase in the fiber might act as a diffusion channel for He release at higher temperatures (<1400°C). The remaining He in the SiC matrix and the SiC fiber was completely released at still higher temperature (>1400°C). This result agrees with He release from neutron irradiated boron-doped sintered SiC [7].

Previous work [3] showed that He bubbles did not precipitate on fiber/C/matrix interface. The results of this work show that this might be attributed to the faster diffusion rate of He in C phase compared to that of the fiber and the matrix. Under fusion irradiation conditions, the expected operating temperature will be 800–1100°C. In actual fusion irradiation conditions, displacement damage clusters will be produced much greater quantity than in this work in spite of defect recombination. Therefore, diffusion of He may be suppressed by trapping at vacancy type defects. Void swelling in SiC has been observed at temperatures above 1200°C [14], and therefore small vacancy type clusters formed in this temperature region may promote the formation of fine and dense He clusters in the SiC part of SiC/SiC composites such as the SiC matrix or the micro-grains in SiC fibers. Interface de-bonding by He bubble formation in composites may not occur under fusion reactor conditions because the interface and the region around it may not act as an effective He trapping site. Helium in the C phase and in the SiC phase in the vicinity of the C phase may diffuse out through the C phase. We also reported He effects on microstructural development using a dual ion irradiation experiment at 800°C and 950°C up to 10 dpa/1000 at. ppm He [15], and in this experiment, large He-defect clusters were not observed in this temperature region. Further investigations are required to determine the effects of small He defect clusters on physical properties such as strength and thermal diffusivity.

4. Summary

Helium release behavior of He-ion implanted SiCf/SiC composites, monolithic SiC, and SiC fibers(Hi-Nicalon) was measured. The He release peak regions from C, and SiC matrix, and SiC fibers were observed. The majority of the He was released from SiC matrix and SiC fiber above 1300°C, and therefore, He-defect clusters in SiC/SiC composites operating at fusion reactor temperatures (800–1100°C) may exist as dense, fine clusters. The C coating layer on SiC fiber and residual C in SiC fiber may act as a diffusion channel in SiC/SiC composites operating under fusion reactor conditions and suppress He bubble formation at C/SiC interface.

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