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Mechanical property change and swelling behavior of SiC fiber after light-ion irradiation

A. Hasegawa *, S. Nogami, T. Aizawa, K. Katou, K. Abe

Department of Quantum Science and Energy Engineering, Tohoku University, 01 Aramaki-aza-Aoba, Aoba-ku, Sendai 980-8579, Japan

Abstract

Studies on the effects of helium or hydrogen gases on the mechanical properties and size stability of SiC fibers were examined using accelerator implantation techniques. Three types of Nicalon fibers (Nicalon-CG, Hi-Nicalon, and Hi-Nicalon type-S) were examined. Three levels of implanted gas concentrations from 50 to 5000 appm were conducted. Swelling of the fibers was observed after helium implantation. The amount of swelling depended on the helium concentration and the type of fiber. The swelling had vanished after the post-implantation annealing at 1000 °C. The slight decrease of tensile strength was observed after the He implantation. It remained after the post-implantation annealing up to 1400 °C. The significant effects of hydrogen implantation on the strength and swelling were not observed in this work.

1. Introduction

SiC fiber reinforced SiC composites are under consideration as candidate structural materials for future fusion reactors because of their high temperature strength and their low induced radioactivity after 14 MeV neutron irradiation [1,2]. On the other hand, the amount of transmuted elements especially Helium (He) and Hydrogen (H) in SiC after 14 MeV neutron irradiation reach higher levels compared to other candidate materials such as vanadium alloys or low-activation ferritic steels. The He/dpa ratio is predicted to be about 100 appm/dpa in the first wall region, it means the total amount of He will be about 1 at.% after 100 dpa irradiation [3]. The amount of hydrogen will reach also about several thousands appm after 100 dpa [3]. The solubilities of He and H in SiC are almost zero, and thus, they may strongly interact with open interfaces such as grain boundaries or with point defects leading to degradation of the material properties. The fiber is the primary load-bearing element in continuous fiber reinforced composites, and therefore the degradation of the mechanical properties of SiC fibers induced by displacement damage and transmuted elements caused by high energy neutron irradiation is a critical issue for fusion applications.

Based on previous studies, degradation of SiC fiber after neutron irradiation is caused by oxidation from residual oxygen and by the growth of SiC grains from amorphous phase in fibers [4–6]. These microstructural changes of fibers induced degradation of SiC/SiC composites [7,8]. To improve irradiation resistance and high temperature property of SiC/SiC composites, high purity and highly crystallized SiC fibers, such as Hi-Nicalon type-S [9] or Tyranno-SA [10], have been developed [11].

On the other hand, grain boundaries in crystalline SiC fiber may act as bubble formation sites and cause the degradation of mechanical properties because He bubbles were observed in the monolithic β-SiC after He implantation and annealing at 1200 °C [12,13]. Hydrogen is also insoluble in SiC, therefore a large amount of H-ion implantation caused surface cracking of monolithic-SiC [14].

The purpose of this work is to study the effects of He or H gases on the mechanical properties and the size stability of crystalline SiC fibers.

* Corresponding author. Tel.: +81-22 217 7923; fax: +81-22 217 7925.

E-mail address: akira.hasegawa@qse.tohoku.ac.jp (A. Hasegawa).

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2. Experimental procedure

Polymer derived SiC fibers were examined in this work. These are Nicalon-CG, Hi-Nicalon and Hi-Nicalon type-S which were supplied by Nippon Carbon Ltd. The physical properties of these fibers have been published previously [9]. The main characteristics of the fibers are shown in Table 1. The Hi-Nicalon type-S, which is mainly examine in this work, is a near stoichiometric and crystalline SiC fiber.

Three MeV He^+-ions and 1.6 MeV H_2^+-ions were irradiated to these specimens using accelerator implantation technique. The irradiation experiments were carried out using a Dynamitron accelerator of the Faculty of Engineering at Tohoku University. The gage length of the tensile tested fiber was 25 mm. These gas-ion beams irradiated at a central part of the gage area. The length of irradiated area of the gage length of fiber was about 3 mm. The implantation range of 3 MeV He^+- and 1.6 MeV H_2^+-ions, that was equivalent to 850 keV H^+-ions, in the monolithic SiC is about 8 μm. The beam current was less than 0.1 μA, and irradiation time was about 6 h for 5000 appm implantation. Irradiation temperature was monitored by an infrared pyrometer, and it was below 100 °C during the irradiation. A rotating-energy degrader system was used to obtain a uniform gas atom distribution throughout a depth of 1–5 μm below the specimen surface. The SiC fibers were irradiated on one side first, and then the fibers were inverted and irradiated again.

Three He level concentrations were examined; these were about 50, 500 and 5000 appm respectively. In the case of H-ions, 500 and 5000 appm were conducted. The displacement damage (dpa) calculated by TRIM code were 0.2 dpa for 5000 appm He irradiation and 0.015 dpa for 5000 appm H irradiation respectively. The implanted concentrations were estimated by the irradiation fluence. Post-implantation annealing was carried out at 750, 1000, 1200 and 1400 °C in vacuum. The annealing time was 1 h. After the annealing, single fiber tensile test was conducted at room temperature in air. The crosshead speed was 0.5 mm/min. Microstructural observations were carried out using a transmission electron microscope (TEM).

Table 1
Typical properties of PCS-derived SiC fibers and monolithic SiC [9]

Materials	Oxygen (wt%)	C/Si	Grain size (nm)	Density (g/cm^3)
Nicalon-CG	11.7	1.31	<1	2.55
Hi-Nicalon	0.5	1.39	5	2.74
Hi-Nicalon type-S	0.2	1.05	>10	3.10
β-SiC[a] (Morton)	<0.1	1.0		3.20

[a] Monolithic SiC (CVD processed).

3. Results and discussions

Fig. 1 shows example of irradiated Hi-Nicalon type-S fibers, which were bent as a result of one side He implantation to 5000 appm. The bending of the fibers for tensile test was not observed if a fiber was irradiated in both sides. The average fiber diameters are about 14 μm, therefore, the bending is due to the fact that only about 1/2 of the fiber cross-sectional area were swelled by the ion irradiation.

The bent fiber was not observed in 50 appm He-implanted Nicalon type-S fiber, and significant bending fiber appeared above 500 appm He implantation. The bending angle of He-ion implanted Hi-Nicalon type-S fibers increased with increasing He concentration. The calculated swelling of the implanted area is estimated using a 2-dimensional assumption proposed by Jung et al. [16]. Linear swelling of Hi-Nicalon type-S based on its bent angle was about 0.41% (5000 appm He), 0.23% (500 appm) and about 0% (50 appm) respectively. On the other hand the swelling was not observed in the 5000 appm H-ion implanted Hi-Nicalon type-S fiber. These results are summarized in Table 2.

Many researchers reported swelling of SiC. Jung et al. [16,17] reported swelling of monolithic 6H–SiC after neutron, He- and H-ion implantation. The magnitude of

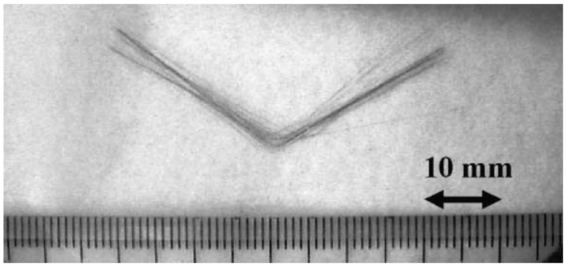

Fig. 1. Example of bent fiber after He implantation at RT. Implanted concentration: 5000 appm, fiber: Hi-Nicalon type-S.

Table 2
Summary of strength change of fibers ($\sigma_{implanted}/\sigma_{unirrad}$) with and without implantation

Fibers	Implanted element	Implanted concentration		
		50 appm	500 appm	5000 appm
Hi-Nicalon type-S	H	–	1.03 (0)	0.96 (0)
	He	– (0)	0.89 (0.23)	0.84 (0.41)
Hi-Nicalon	He	–	0.94 (0.13)	–
Nicalon-CG	He	–	0.89 (0)	–

Values in parentheses mean the swelling (%) of fiber by one side implantation of He or H. Implanted temperature: RT.

liner swelling of Hi-Nicalon type-S after He implantation was almost the same level of Jung's data. SiC was easily amorphized at room temperature for damage levels of approximately 0.1–0.5 dpa by ion beam irradiation, and produced a volumetric expansion [19]. The retention and clustering of defects such as carbon interstitials produced by cascade also contribute the swelling [20].

The volume change of SiC fibers by this work depended on fiber types. The swelling was 0.13% in 500 appm He-implanted Hi-Nicalon and the fiber bend was not observed in the 500 appm He-implanted Nicalon-CG. The Nicalon-CG contains large amounts of uncrystallized phases and residual carbon phases. Hi-Nicalon also contains these phases but with a smaller amount compared to Nicalon-CG. Hi-Nicalon type-S is an almost crystalline fibers. These results show that the swelling of as-irradiated fiber related to the amount of crystalline phases, therefore swelling of the fibers may be attributed to the disordering of crystalline phase of SiC in the fiber.

In the case of H-implanted Hi-Nicalon type-S fibers, fiber bend was not observed. The calculated dpa of 5000 appm H-implantation was about 0.015 dpa and it was about 0.2 dpa for 5000 appm He-implantation, therefore, the amount of displacement damage (dpa) induced by H-ion irradiation is not sufficient to generate enough disordering in SiC crystals in the fiber.

When the bent fibers were annealed at temperatures greater than 1000 °C, the fibers became unkinked. In the monolithic SiC, swelling induced by displacement damage at room temperature anneals out when heat treated at a temperature about 1000 °C [17]. This result also supports the bending is caused by the displacement damage related microstructural changes such as disordering of SiC phase.

The fiber bend and another additional dimensional change did not appear after 1400 °C annealing. Previous works showed that small and fine He bubbles formed in monolithic β-SiC after 1200 °C annealing [12,13,17], but our result means swelling caused by the He bubble formation was not so significant that He-implanted fibers bent again.

The effect of He on fiber strength shows in Fig. 2. It is the Weibull plots of annealed fiber strength with and without 500 appm He implantation. The Weibull moduli are in the range of 4.1–6.6. The Weibull modulus tends to decrease with increasing annealing temperature both with and without He implantation.

Fig. 3 summarizes the tensile strength change due to annealing up to 1400 °C in Hi-Nicalon type-S both with and without He implantation. The tensile strength decreased with increasing He concentration below 1000 °C. Strength degradation of He-implanted fiber did not recover in spite of the disappearance of the swelling, which caused fiber bent, and this means that the microstructural changes which caused the swelling in fiber did not affect the strength change directly. The dependence on He concentration of the strength degradation became small in specimens annealed at 1400 °C.

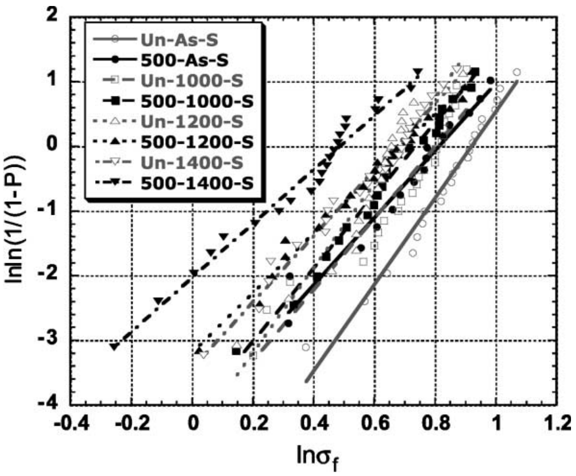

Fig. 2. Weibull plots of tensile strength of He-implanted and annealed Hi-Nicalon type-S. Unimplanted and annealed data are also plotted. He concentration: 500 appm.

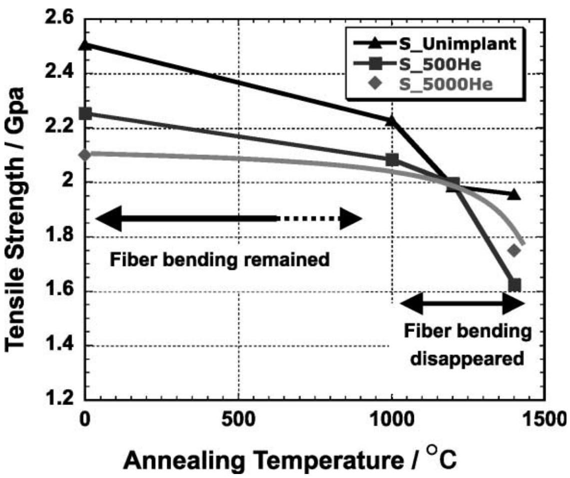

Fig. 3. Average strength change of He-implanted and annealed Hi-Nicalon type-S fibers.

A decrease in strength after He implantation was also observed in the Nicalon-CG and Hi-Nicalon fibers. Table 2 shows the strength of various SiC fibers both with and without He implantation. Again, fiber strength degrades by He implantation but the strength change behavior does not depend on the magnitude of the swelling.

In the case of H-ion implanted fibers, strength degradation was not observed as the He-implanted fibers in as-implanted and annealed at 750 °C fibers. Fig. 4

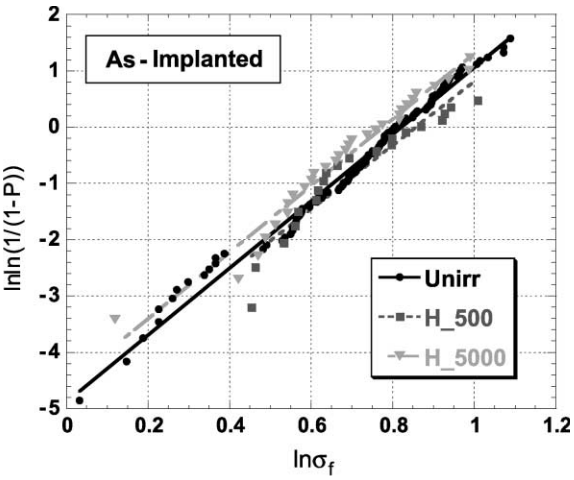

Fig. 4. Weibull plots of H-implanted Hi-Nicalon type-S fibers.

shows the Weibull plots of tensile tested fiber with and without H implantation. Hydrogen in SiC could be mobile at around 750 °C annealing [14,15], therefore bubble formation in SiC was expected after the annealing. This result shows that clustering of implanted H does not cause strength degradation of Hi-Nicalon type-S fiber under the testing condition.

Microstructural observation using TEM showed that significant amorphous regions were not observed in He-implanted Hi-Nicalon type-S fiber, and He bubbles were not observed in the fibers implanted with 500 appm He and annealed at 1400 °C. In the previous work which was conducted to He-implanted SiC/SiC composites, He bubbles were not observed in SiC fibers (Hi-Nicalon) implanted with 5000 appm He and annealed at 1400 °C, but bubbles were observed at grain boundaries in the CVI processed SiC matrix [13]. Serious degradation of fiber strength, which was expected by He bubble formation on the grain boundaries of crystalline SiC fibers, was not observed in this work. In the case of stainless steels, He bubble formation caused serious grain-boundary embrittlement even in the several appm level of He containing samples when He bubble formed on grain boundaries. The resistance to He embrittlement of the SiC fiber might be attributed to the smaller grain size (20–50 nm) and microstructure of grain-boundary area which might trap He and suppress He bubble nucleation and growth.

Our previous work using He desorption technique showed that more than 90% of the He remained in Hi-Nicalon type-S after annealed at temperatures up to 1400 °C [18]. Thus, it can be considered that the implanted He remains in the Hi-Nicalon type-S fiber even after 1400 °C annealing, but the He could not aggregate to form visible He clusters in the fiber. In the case of Nicalon-CG and Hi-Nicalon fibers, uncrystallized phases located between the SiC nano-crystals in the fiber could be considered as trapping site or as diffusion path of the He, but these phases have not been clearly observed in Hi-Nicalon type-S fiber in this work. Further investigation using high resolution TEM will be necessary to clarify the He trapping mechanism in the SiC fibers. This issue will be important to clarify the soundness of crystalline fibers to use in fusion environment where displacement damage, He and H generation will coexist.

4. Summary

The tensile strength and swelling behavior of Nicalon type fibers were studied with and without He and H implantation, and post-implantation annealing up to 1400 °C were conducted.

(1) Swelling of fibers was observed after He implantation at around room temperature. The amount of swelling depended on the helium concentration and the type of fiber. Hi-Nicalon type-S fiber showed the biggest swelling. Any swelling that had occurred during implantation had vanished after annealing at 1000 °C.
(2) The tensile strength of fibers decreased with increasing the He concentration. The degradation of tensile strength remained after post-implantation annealing up to 1400 °C, but He bubble formation in Hi-Nicalon type-S was not observed by TEM. A drastic degradation of fracture strength, which was caused by He bubble formation at grain boundaries, was not observed.
(3) Swelling and strength degradation of Hi-Nicalon type-S fiber were not observed after 5000 appm Hydrogen implantation.

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