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Neutron irradiation effects on high-crystallinity and near-stoichiometry SiC fibers and their composites

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Abstract

Key characteristics required for the development of fusion-grade SiC/SiC composites are high-crystallinity and near-stoichiometry. To identify the primary mechanisms of degradation caused by neutron irradiation, the radiation behavior of the constituent SiC fibers needs to be examined. In this study, single filament tensile tests were conducted after neutron irradiation on a recently developed highly-crystalline and near-stoichiometric SiC fiber; Hi-Nicalon™ Type-S. Hi-Nicalon™ Type-S fiber exhibited excellent strength retention up to 7.7 dpa independent of irradiation temperature up to 800 °C. The radiation stability of the Hi-Nicalon™ Type-S fiber directly contributed to the excellent radiation performance in composites made with this fiber. The observed 14–20% decrease of elastic modulus due to neutron irradiation of Hi-Nicalon™ Type-S fiber had a minor effect on composite strength.

1. Introduction

Silicon carbide based composites (SiC/SiC), considered to be a candidate as a structural fusion-grade material, require excellent radiation stability of all constituents: reinforcing fiber, matrix and interphase. Silicon carbide, in nature, exhibits excellent radiological properties such as low induced radio-activation and low after-heat [1]. Moreover, it was revealed that high-crystallinity and near-stoichiometry β-SiC matrix, formed by chemical vapor infiltration (CVI), exhibited superior dimensional and mechanical stabilities under neutron irradiation up to 10 dpa [2,3].

In contrast, SiC-based fibers containing glassy silicon-oxycarbide phases (SiC_xO_y) such as Nicalon™ and Hi-Nicalon™ (Nippon Carbon Co. Ltd.) exhibited severe dimensional and mechanical property changes due to extensive shrinkage caused by crystallization of the amorphous SiC phase and the presence of radiation-instable impurities such as excess carbon [4]. As a result, fiber shrinkage and swelling of high-crystallinity SiC matrix caused stress-induced debonding of the fiber/matrix (F/M) interphase [5].

Recently developed Hi-Nicalon™ Type-S (Nippon Carbon Co. Ltd.) and Tyranno™-SA (Ube Industries, Ltd.) fibers have offered the possibility of improved radiation stability due to the similar microstructures with radiation-stable β-SiC; high-crystallinity and near-stoichiometry. In fact, SiC/SiC composites with these fibers did exhibit excellent tolerances in shape and mechanical properties under neutron irradiation [6,7]. To further understand why these composites exhibited improved radiation performance, the effect of neutron irradiation on fiber constituent alone needs to be assessed.

The objective in this study is to identify the effect of neutron irradiation on mechanical properties of highly-crystalline and near-stoichiometric SiC fiber by single filament tensile testing. Specifically, the effect of

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high-temperature irradiation on composite strength up to 800 °C is evaluated. The effects of neutron irradiation on chemical vapor deposited (CVD) SiC and advanced SiC/SiC composites are also assessed to explore the confirmed development of fusion-grade SiC/SiC composites.

2. Experimental

Highly-crystalline and near-stoichiometric SiC fiber; Hi-Nicalon™ Type-S, was used. Key characteristics of Hi-Nicalon™ Type-S fiber are listed in Table 1.

Neutron irradiation was carried out in the High Flux Isotope Reactor (HFIR) at Oak Ridge National Laboratory as a part of the HFIR-14J experiment. Irradiation temperatures were 300, 500 and 800 °C, and the irradiation fluence was 6.0, 6.0 and 7.7 dpa, respectively. It was assumed that 1 dpa corresponds to 1.0×10^{25} n/m^2 ($E > 0.1$ MeV). Experimental details are described elsewhere [8].

Fig. 1 shows a design of the cardboard specimen holder. Gauge length of tested fibers was 25.4 mm. Both ends of a single filament were adhered on the cardboard by epoxy. Viscous grease was spread on the fiber surface to capture broken fiber segments. Tensile strain was calculated from the distance between two stripes on the flags measured by a laser extensometer. The stiffness of test fixture is high compared with fiber and its effect on fiber strain was considered quite a small. For stress calculation, the fiber diameter was estimated by taking the average of three measurements on each individual fiber by video microscopy.

Prepared cardboard specimen was mounted by the connecting pins to main fixture and the center of the cardboard was cut by scissors taking care not to damage the tested fiber just before the testing. Crosshead displacement rate was 0.5 mm/min (strain rate: 2.0×10^{-4} s^{-1}). Fibers broken within the gauge length were recognized as valid. All of the valid data were analyzed by Weibull statistics. A detailed description of the test methodology was referred in the modified standard by Lara-Curzio [9].

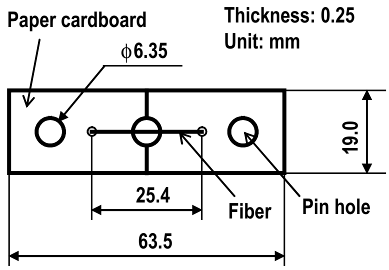

Fig. 1. Schematic illustration of cardboard specimen holder for single filament tensile test.

3. Results and discussion

3.1. Effect of neutron irradiation on Hi-Nicalon™ Type-S SiC fiber

Table 2 and Fig. 2 show tensile strength test results for Hi-Nicalon™ Type-S fiber. It is noted that 5–10% of measurement errors of the fiber diameter affected the large scatters of tensile strength and tensile modulus. A slight decrease in Weibull modulus is observed but there was no overall degradation in tensile strength after neutron irradiation. Little effect of the irradiation temperature on Weibull mean strength was observed (approximately +5%, after irradiation).

Fig. 3 shows normalized Weibull mean strength represented as the retention ratio after neutron irradiation. According to Osborne et al. [4], strength increase by neutron irradiation was typical for amorphous based SiC fibers; Nicalon™ and Hi-Nicalon™, due to crystallization of the amorphous SiC_xO_y phase or excess carbon. In contrast, Hi-Nicalon™ Type-S fiber maintained its tensile strength at the original level under neutron irradiation.

The dose dependency of tensile strength for Hi-Nicalon™ Type-S fiber was very similar to that of flexural strength for crystalline and pure SiC, i.e. CVD-SiC [2,3,10]. Both CVD-SiC and Hi-Nicalon™ Type-S fiber, in principle, kept its strength under heavy neutron irradiation under the same surface finish in materials

Table 1
Key characteristics of silicon carbide fibers

	Hi-Nicalon™ Type-S (HNL-S)	Hi-Nicalon™ (HNL)	Nicalon™ (CG-NL)
C/Si atomic ratio	1.05	1.39	1.34
Oxygen content (wt%)	0.2	0.5	11.7
Tensile strength (GPa)	2.6	2.8	3.0
Tensile modulus (GPa)	420	270	220
Elongation (%)	0.6	1.0	1.4
Density (Mg/m^3)	3.10	2.74	2.55
Diameter (μm)	11	14	14

Table 2
Tensile properties of Hi-Nicalon Type-S fibers irradiated at 300, 500 and 800 °C

Irradiation (T_{irr}, dose)	Weibull modulus	Weibull characteristic strength, σ_0 (GPa)	Weibull mean strength, $\bar{\sigma}$ (GPa)	Weibull standard deviation, s (GPa)	Tensile modulus (GPa)	Number of specimens
None	6.0	3.4	3.2	0.6	400 ± 51	22
300 °C, 6.0 dpa	4.6	3.6	3.3	0.8	316 ± 76	38
500 °C, 6.0 dpa	4.3	3.5	3.2	0.8	338 ± 66	36
800 °C, 7.7 dpa	4.8	3.5	3.2	0.7	344 ± 51	27

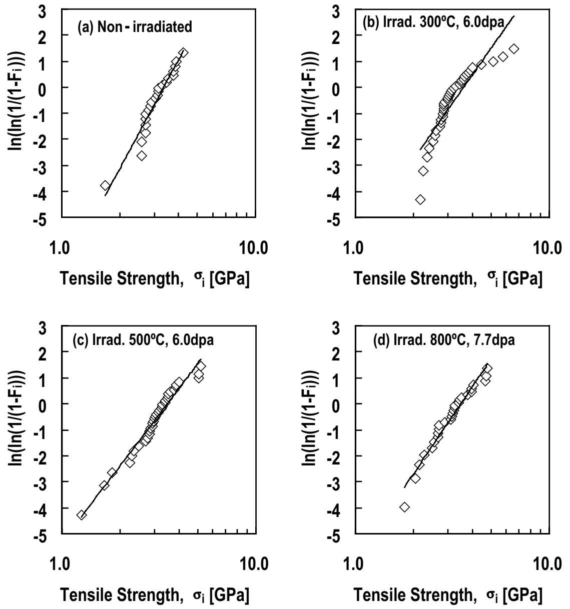

Fig. 2. Weibull statistical plots for tensile strength of Hi-Nicalon^{TM} Type-S fibers; (a) non-irradiated and irradiated at (b) 300 °C, (c) 500 °C and (d) 800 °C.

preparation for CVD-SiC. A slight increase of flexural strength by Snead et al. might be explained by the increase of fracture toughness [10]. However, the primary mechanism is still uncertain.

Fig. 4 shows the effect of neutron irradiation on the tensile modulus of Hi-Nicalon^{TM} Type-S fiber. Specifically, the tensile modulus tended to decrease attaining a maximum change of 20% for the 300 °C irradiation, 16% at 500 °C and 14% at 800 °C. Conversely, crystallization of Nicalon^{TM} with its amorphous SiC_xO_y phase and Hi-Nicalon^{TM} with excess carbon had significant effects on the change of their lattice parameters resulting in the increase of the elastic modulus. However, it is noted that Hi-Nicalon^{TM} fiber swells slightly in the low fluence neutron irradiation range due to its well-organized crystal structure [4]. In this range, no significant change of the tensile modulus was identified.

The degradation of the elastic modulus for Hi-Nicalon^{TM} Type-S fiber was also very similar to that for CVD-SiC [2,11,12]. The tendency toward greater reduction in elastic modulus for lower temperature irradiation is attributed to the higher amount of surviving defects consistent with the higher level of saturation swelling at low temperature. Moreover, well-crystallized, higher initial modulus structures such as

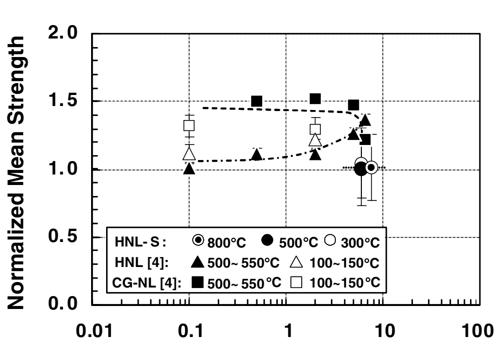

Fig. 3. Effect of neutron irradiation on Weibull mean strength of Nicalon^{TM} (CG-NL), Hi-Nicalon^{TM} (HNL) and Hi-Nicalon^{TM} Type-S (HNLS).

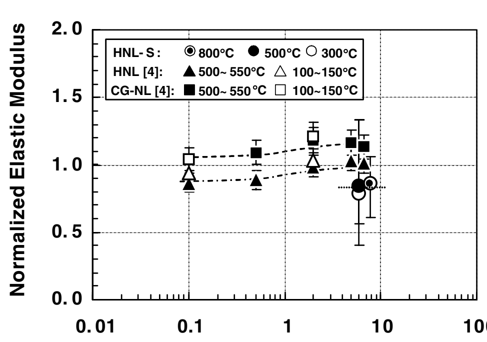

Fig. 4. Effect of neutron irradiation on elastic modulus of Nicalon^{TM} (CG-NL), Hi-Nicalon^{TM} (HNL) and Hi-Nicalon^{TM} Type-S (HNLS).

CVD-SiC are more sensitive. However, at higher temperatures, surviving defects following neutron cascades are reduced, leading to smaller elastic modulus change.

3.2. Effect of neutron irradiation on highly-crystalline and near-stoichiometric SiC/SiC composites

Several mechanical test results after neutron irradiation have been reported for advanced SiC/SiC composites [6,7,13]. SiC/SiC composites with high purity SiC fiber and matrix exhibited stable flexural strength under high dose (7.7 dpa) and high-temperature (800 °C) irradiation. The radiation stability in tensile strength for Hi-Nicalon™ Type-S fiber contributed the excellent radiation performance in composite strength. However, the 10–20% of degradation in flexural modulus of composites was identified, even at low-fluence neutron irradiation up to 1 dpa [13].

Conversely, recent our work offers no degradation of the tensile modulus of composites after neutron irradiation. Fig. 5 shows the preliminary test result about the effect of neutron irradiation on tensile properties of Hi-Nicalon™ Type-S fiber reinforced CVI-SiC matrix composites with single PyC, multilayered (ML) SiC/PyC and pseudo porous SiC interphase after neutron irradiation at 800 °C up to 1 dpa performed in the Japan Materials Testing Reactor (JMTR) at Oarai. Materials tested are summarized in elsewhere [14]. This indicates that the radiation-induced change in tensile modulus of fiber is not the primary mechanism for the degradation of the composite modulus. Moreover, Fig. 5 shows that the significant reductions of the tensile strength and proportional limit stress were identified in only a porous SiC interlayer composite. This also implies the importance of the F/M interphase to maintain good radiation performance in composite strength. Further investigations are addressed to confirm this issue.

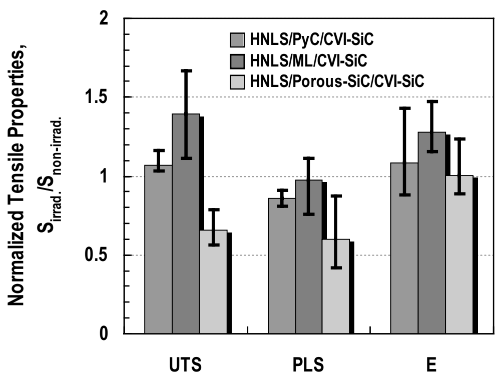

Fig. 5. Effect of neutron irradiation on ultimate tensile strength (UTS), proportional limit stress (PLS) and elastic modulus (E) of Hi-Nicalon™ Type-S fiber reinforced CVI-SiC matrix composites with single PyC, multilayered SiC/PyC and porous SiC interphase.

4. Conclusions

In order to identify the effect of neutron irradiation on recently developed highly-crystalline and near-stoichiometric SiC fiber; Hi-Nicalon™ Type-S, single filament tensile tests were carried out after neutron irradiation. Hi-Nicalon™ Type-S fiber exhibited no statistically significant degradation in tensile strength up to 7.7 dpa at 300–800 °C, leading to the excellent radiation performance in composite strength. The observed 14–20% decrease of elastic modulus due to neutron irradiation of Hi-Nicalon™ Type-S fiber had a minor effect on composite strength. However, this needs to be verified in the following microstructural observation.

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