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The effect of neutron-irradiation on the shear properties of SiC/SiC composites with varied interface

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Abstract

The materials used in this study were unidirectional Hi-Nicalon^{TM} SiC fibers reinforced SiC matrix composites. SiC/SiC composites were fabricated by chemical vapor infiltration (CVI) method. Fibers were coated with either pyrolytic carbon, multiple SiC layers or 'porous' SiC by CVI method prior to matrix deposition. They were irradiated in the High Flux Isotope Reactor at damage level of 0.5 dpa. Irradiation temperatures were either 300°C or 500°C. Interfacial shear properties and mechanical properties were evaluated. The correlation between interfacial shear properties and mechanical properties was examined. The 'porous' SiC interface showed a strong bond when compared with the pyrolytic C and multiple SiC interface in the non-irradiated state. The interfacial shear properties were substantially degraded by irradiation with the 'porous' SiC showing the most degradation. © 2000 Elsevier Science B.V. All rights reserved.

1. Introduction

The high temperature mechanical properties and low activation make SiC/SiC composites very attractive as fission and fusion reactor materials [1]. At present, interfacial properties between the fiber and matrix of neutron-irradiated SiC/SiC composites limit mechanical performance [2]. This limitation has been attributed primarily to shrinkage in the SiC-based fibers due to irradiation-assisted oxidation [3], irradiation-induced recrystallization of microcrystalline fibers [4–6], and to large dimensional changes of the carbon [7] interface applied to the fiber. To avoid or at least minimize these radiation effects, the recent trend in SiC fiber development is toward lower oxygen content, reduced free carbon and enhanced crystallinity. Previous work regarding ion-irradiation effect on carbon interface microstructure indicated that the basal planes of the irradiated graphite-like carbon appear to be chopped into small fragments and consequently amorphous-like structures were observed [8]. The development of more radiation-resistant SiC composites is based on the use of stoichiometric SiC fibers with lower oxygen and SiC-based interfaces. Recently, stoichiometric SiC fibers have been developed including Hi-Nicalon type-S^{TM} [9], Sylramic^{TM} and Tyranno SA^{TM}, however, development of SiC-based interface is still desired.

The objective of this work is to develop improved interfacial properties under neutron-irradiation. For this purpose, a multiple SiC interface and a 'porous' SiC interface were applied to low oxygen Hi-Nicalon^{TM} fibers. The effect of irradiation on interfacial shear properties was evaluated directly by single fiber indentation tests. The interfacial shear properties were correlated with mechanical properties.

2. Experimental procedure

2.1. Materials

The materials used in this study were unidirectional Hi-Nicalon^{TM} SiC fiber reinforced SiC composites. The

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atomic oxygen content of Hi-Nicalon™ fiber is reported to be less than 0.5% [10]. Hi-Nicalon™ is not a stoichiometric SiC fiber, but its properties under neutron-irradiation are relatively understood [5,6]. SiC/SiC composites were fabricated by chemical vapor infiltration (CVI) at Hypertherm high-temperature composites. Fibers were coated with either carbon, multiple SiC or 'porous' SiC by CVI prior to matrix deposition, as shown in Fig. 1. Each coating thickness was about 600 nm. Mixtures of methyltrichlorosilane, argon, methane and hydrogen gases were used to deposit the 'porous' SiC interface on fiber. In the multiple SiC interface, the first 100 nm thick SiC layer was deposited followed by a thin, interrupted layer of pyrolytic carbon. Four SiC layers were fabricated with interrupted pyrolytic carbon in the multiple SiC interface [11].

Irradiation was carried out in the hydraulic tube facility of High Flux Isotope Reactor (HFIR) at Oak Ridge National Laboratory. A dose equivalent of 0.5×10^{25} nm^{-2} ($E > 0.1$ MeV) = 0.5 dpa was used. The sample temperature was achieved by gas-gap conduction of the nuclear heat. The irradiation capsule environment was static helium. Nominal temperatures were 300°C and 500°C.

2.2. Mechanical tests

A load-controlled Nanoindenter™ microindentation hardness tester was used for single fiber indentation tests. Specimens for these tests were sliced and fixed on a holder with the fiber direction perpendicular to holder surface. After mechanical polishing, isolated fibers were selected with a video microscope. Fibers were pushed in by Berkovich type pyramidal diamond indenter tip with maximum load 0.4 N. An illustration of single fiber indentation test, and a SEM image of pushed-in fiber are shown in Fig. 2.

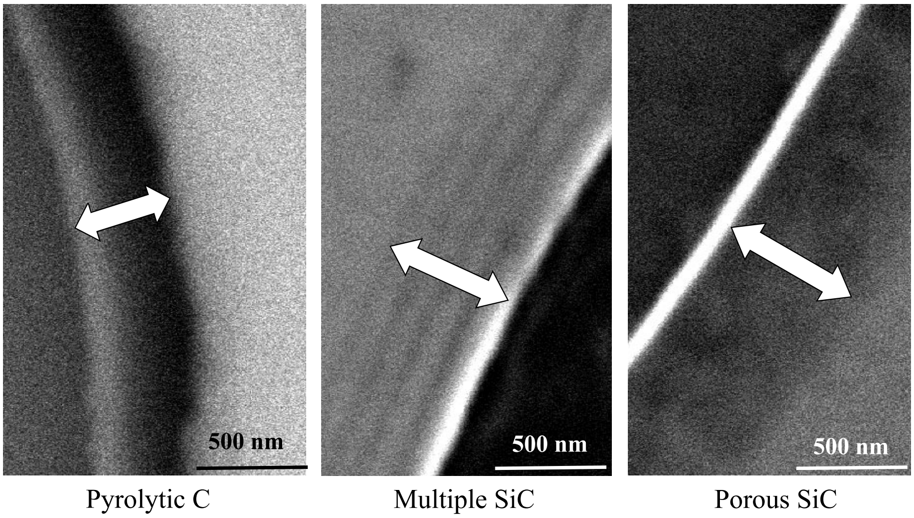
Four-point bend tests were carried out at ambient temperature. The sample geometry was $25^{l} \times 2.5^{w} \times 2^{t}$ mm^3. The load and support spans were 5 and 20 mm. The crosshead speed was 8.5×10^{-3} mm/s.

3. Results and discussions

3.1. Effect of neutron-irradiation on interfacial shear properties

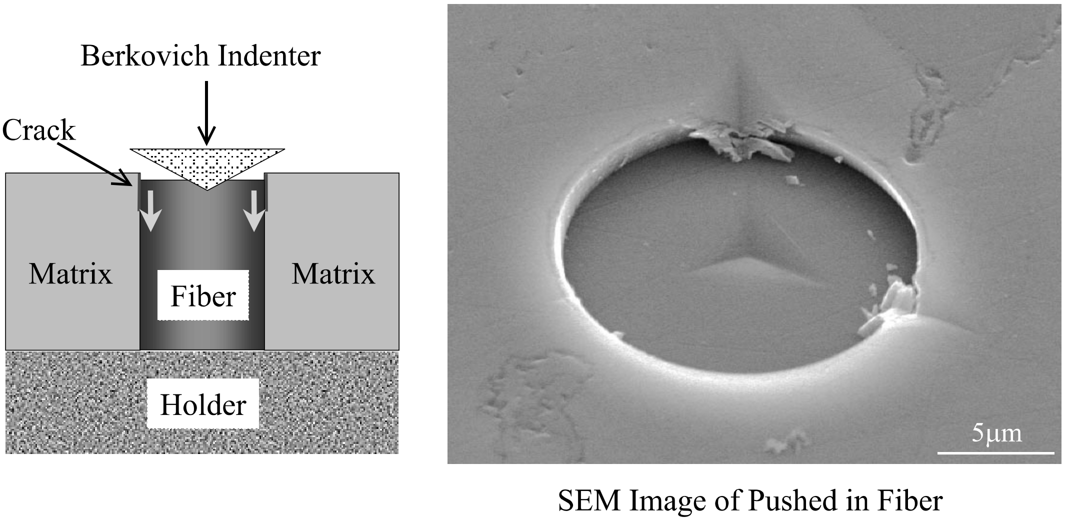
Single fiber indentation tests were carried out on both unirradiated materials, and materials irradiated at 0.5 dpa, 300°C with pyrolytic C, multiple SiC or 'porous' SiC interfaces. Typical loading curves of the 'porous' SiC interface materials are shown in Fig. 3. Following irradiation, interfacial shear properties, which include clamping stress and frictional stress, are substantially degraded, and the loading curve is shifted to the right. Critical loads for crack initiation, which is labeled 'fiber push-in', were not found in the irradiated samples. Samples with a pyrolytic C or multiple SiC interfaces showed weak interfacial shear properties similar to those of the irradiated 'porous' SiC interface.

Generally 'fiber push-in' load is used to evaluate interfacial shear properties in single fiber push-in test [12], however, it was very difficult to identify the 'fiber push-in' loads in every condition, and therefore, impossible to evaluate interfacial shear properties quantitatively from 'fiber push-in' load. To evaluate interfacial shear properties quantitatively from load vs displacement curves, the loads at 0.9 μm indent were measured. A displacement of 0.9 μm was chosen as this corresponds to



Pyrolytic C Multiple SiC Porous SiC

Fig. 1. SEM images of the pyrolytic carbon, multiple SiC and 'porous' SiC interfaces.



SEM Image of Pushed in Fiber

Fig. 2. Schematic illustration of the single fiber indentation test, and an SEM image of a pushed-in fiber.

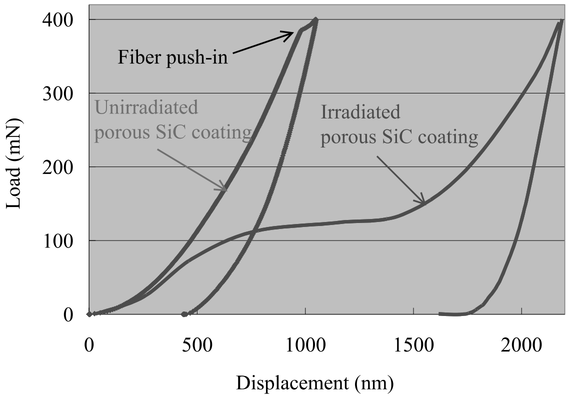


Fig. 3. Effect of neutron-irradiation on single fiber indentation loading curve of 'porous' SiC samples.

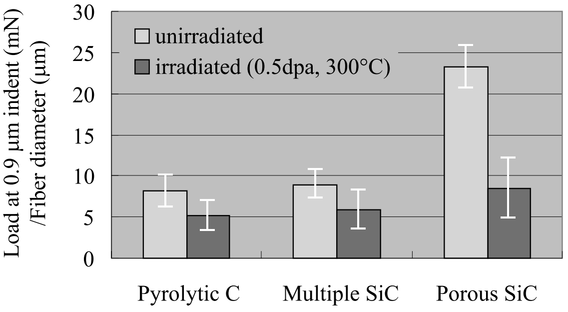


Fig. 4. Effect of neutron-irradiation on interfacial shear properties.

load/depth for which interfacial debonding has taken place, but is not to the point at which the indenter contacts the matrix. The measured load included clamping strength, frictional strength in the debonded area, and strength to compress a fiber. To reduce the effect of fiber size, measured loads were divided by the fiber diameter. The effect of neutron-irradiation on interfacial shear properties is shown in Fig. 4. The average load divided by diameter is compared. The error bar shows +/−1 standard deviation.

The 'porous' SiC interfaces showed a stronger bonding as compared to the pyrolytic C and multiple SiC interfaces, especially in the non-irradiated state. The pyrolytic C and multiple SiC interfaces showed almost the same interfacial shear properties, although the multiple SiC interfaces were a little stronger than pyrolytic C interface. In the case of the 'porous' SiC, it is believed that chemical interaction at the fiber interface led to stronger bonding. Following irradiation, interfacial shear properties for all interfaces were degraded, with the 'porous' SiC interface showing the largest degradation. Additionally, the scatter in the data increases following irradiation. This degradation is likely due to shrinkage of the fiber causing partial or total interfacial debonding. This has led to a wider distribution in interfacial properties as seen by the slightly larger statistical scatter (Fig. 4). Irradiation did not cause only degradation of interface but also shrinkage of fibers and therefore, part of interfacial debonding had occurred. This is considered as one of the reasons for degradation of interfacial shear properties and increasing of data scatter.

3.2. Effect of neutron-irradiation on mechanical properties

Four-point bend tests were carried out for unirradiated and neutron-irradiated SiC/SiC composites. The

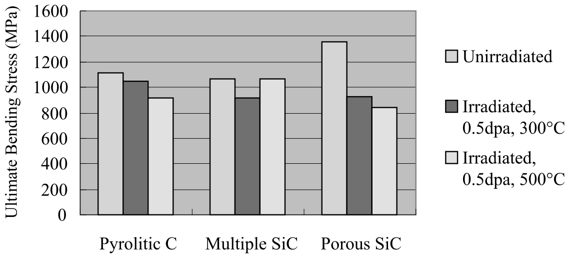


Fig. 5. Effect of neutron-irradiation on ultimate flexural stress.

damage level of irradiated samples was 0.5 dpa, and they were irradiated at 300°C or 500°C. The ultimate flexural stresses (UFS) of unirradiated and irradiated samples are compared in Fig. 5. Following irradiation, the UFS degraded, with 'porous' SiC interface showing the largest degradation, although the pyrolytic C and multiple SiC samples also degraded with irradiation except for the multiple SiC sample irradiated at 0.5 dpa and 500°C. It is reported that swelling of stoichiometric SiC depends on temperature [13]. In the case of the multiple SiC sample, the difference of UFS might be due to the differential swelling.

The UFS degradation appeared to correspond to the relationship between the unirradiated and the irradiated interfacial shear properties shown in Fig. 4; however, pyrolytic C UFS was the highest among samples irradiated at 0.5 dpa, 300°C, although 'porous' SiC interfacial shear strength was the highest at the same irradiation condition. In the case of 'porous' SiC samples, about 60% of interfacial shear strength was lost during irradiation at 0.5 dpa, 300°C, although about 30% of UFS was lost by the same irradiation. This degradation indicates that interfacial shear properties are more sensitive to neutron-irradiation than are the properties of the Hi-Nicalon™ SiC fiber and the CVI SiC matrix.

These mechanical results are consistent with previous results on materials processed with similar interface structures but with a 2-D plain weave fiber architecture [11,14].

4. Conclusions

To improve interfacial properties under neutron-irradiation, SiC-based interface structures were studied. Single fiber indentation tests and four-point bend tests were carried out.

The conclusions are:

1. The 'porous' SiC interface showed the strongest bond as compared with the pyrolytic C and multiple SiC interface in the unirradiated state. In the case of 'porous' SiC, it is believed that chemical bonding was responsible for the stronger bonding.
2. Irradiation at 300°C and damage levels of 0.5 dpa, degraded interfacial shear properties with the 'porous' SiC interface composites showing the most degradation. Irradiation resulted in increasing the scatter in the data of interfacial shear properties.
3. The fracture properties as measured by four-point bend tests were degraded by irradiaton, except for the multiple SiC sample irradiated at 0.5 dpa and 500°C. Again the 'porous' SiC showed the largest degradation. These results appear to correspond to the interfacial shear properties.
4. Interfacial shear properties are more sensitive to neutron-irradiation than Hi-Nicalon™ SiC fiber and CVI SiC matrix properties.

Acknowledgements

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