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INTERPHASE INTEGRITY OF NEUTRON IRRADIATED SIC COMPOSITES

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ABSTRACT

SiC/SiC composites were fabricated from Hi-Nicalon™ fibers with carbon, porous SiC and multilayer SiC interphases. These materials were then irradiated in the High Flux Beam Reactor with fast neutrons at 260 and 900-1060°C to a dose of 1.1×10^{25} n/m^2 corresponding to 1.1 displacements per atom (dpa). Results are presented for bend strength of both non-irradiated and irradiated materials. Within the interphases studied the multilayer SiC interphase material showed the least degradation (8-20%) in ultimate bend stress. while porous SiC underwent the greatest degradation (~35%). The fiber matrix interphases are studied with TEM for both non-irradiated and irradiated materials. While no irradiation induced microstructural evolution of the interphase was observed, debonding of the interphase from the fiber was observed for all cases. This debonding is attributed to tensile stresses developed at the interface due to densification of the Hi-Nicalon™ fiber. Residual stress analysis of the fiber matrix interface indicates that the irradiation-induced densification of Hi-Nicalon™ and the volumetric expansion of the CVD SiC matrix cause tensile stresses well in excess of those which can be withstood by these. or any other viable SiC composite interphase.

INTRODUCTION

A key factor affecting ceramic composite performance is the fiber/matrix interphase deposited onto the fiber preform prior to the infiltration of the matrix material. The most widely utilized interphase in the CVI SiC/Nicalon™ system is carbon. This layer. typically less than 1 µm in thickness. has the effect of reducing the fiber/matrix interfacial strength. A weak interphase will readily debond and arrest a crack propagating through the matrix. In monolithic ceramics. or ceramic composites with very high fiber/matrix interfacial strength. a single crack can freely propagate through the material causing catastrophic failure. Since toughening from impeding propagation of cracks is the primary advantage of SiC/SiC composites compared to monolithic SiC, properly engineered interphases are critical to the performance of these materials.

One concern when considering composite materials for fusion energy applications is the material stability under the expected high-energy neutron exposure. It is known that irradiation causes significant thermophysical changes in monolithic SiC. For example. at temperatures less than 1000 °C. isotropic swelling occurs in silicon carbide in response to the production of simple point defects reaching saturation by ~0.5dpa.[1] Swelling in crystalline SiC is largest for low irradiation temperatures with saturation room temperature linear expansion of about 1%.[2] As irradiation temperature increases. lattice expansion decreases to near zero at 1000°C.[1,2] While the irradiated strength of monolithic silicon carbide has had relatively limited study, recent work[3,4] has reinforced similar work by Price[5] suggesting no reduction should occur for exposure less than 10^{26} n/m^2. However. both the physical and dimensional properties of CG-Nicalon™ SiC-based fiber are unstable at relatively low neutron fluence[6-11] and opposite to what occurs in fully dense SiC. The densification of the ceramic grade (CG) Nicalon™ (about 5% at 1dpa) has been blamed for the substantial (~40%) reduction in bend strength occurring in composites fabricated from this first production grade SiC fiber.[12]

DISCLAIMER

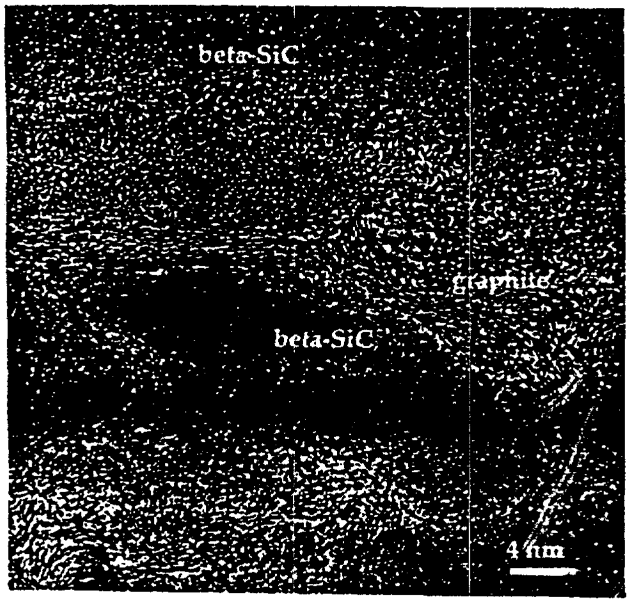

Figure 3- HRTEM image of porous SiC.

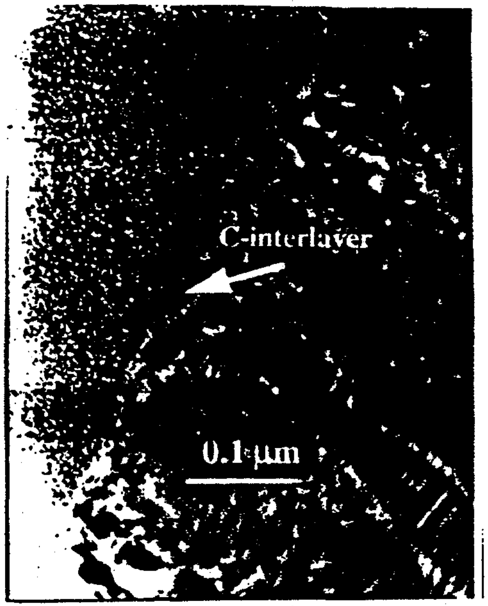

Figure 4- TEM image of multilayer SiC .

Composites containing the multilayer SiC fiber coating were fabricated using isothermal CVI of Hi-Nicalon™ fiber preforms by Hyper-Therm High-Temperature Composites, Inc. An initial layer of pyrolytic carbon 20nm thick was deposited on the fiber preform to prevent the previously discussed interaction with the SiC processing phases. The first SiC layer of the multilayer fiber coating is then deposited followed by a 20 nm interrupted layer of pyrolytic carbon. A TEM image of the interface between the fiber and the first interlayer is given in Figure 4. The composite architecture for all composites was 1800 denier (500 filament yarn) plain weave fabric and was laid-up inside a graphite holder with a fiber volume fraction of approximately 40%. The fabricated composite void fraction was 10-12%.

Composite samples were machined to dimensions of 2.5 x 3 x 25 mm for the irradiation study. The porous SiC material surfaces were machined such that the top and bottom surfaces had exposed fabric where the surface layer had been ground flat. The multilayer SiC material had a thin CVD SiC overcoat (~100µm) so that the top and bottom fabric layer were not exposed. The fabric orientation was such that the fabric was in the plane of the width and length axes of the bend bar. All edges of the bend bars were ground flat, thus some of the fibers were machined away for the porous SiC and pyrolitic carbon interfacial materials. All materials were cleaned in acetone and isopropyl alcohol prior to the irradiation capsule assembly.

An irradiation capsule was inserted into the V-16 in-core thimble of the High Flux Beam Reactor at the Brookhaven National Laboratory. The duration of the irradiation was one reactor cycle corresponding to an approximate fast neutron fluence of 1.1x 10^{25} n/m^2 (E>0.1 MeV). This corresponds to ~1.1 dpa in SiC, assuming a sublattice-averaged displacement energy of 40 eV. The irradiation temperature for the multilayer-SiC interfacial composite was 385°C and was constant within 5°C throughout the irradiation. Two subcapsules were used for the porous-SiC interphase composite. The lower temperature subcapsule, which was made of type-6061 aluminum alloy, had a constant irradiation temperature of 260°C, constant within 5°C throughout the irradiation. The higher temperature subcapsule, which was fabricated from vanadium, initially achieved a temperature of 1060°C and then decreased in temperature linearly with time to a final temperature of 910°C. It is believed that this decrease in temperature is due to swelling of the vanadium subcapsule. Post-irradiation examination of the SiC samples did not show any evidence of sample reaction with the subcapsules.

All TEM was carried out on mechanically thinned and polished samples. For the case of the neutron irradiated specimens, foil thickness prior to Ar ion milling was left at greater >200µm to minimize grinding damage to the interface. Bend testing was conducted at room temperature using a 4-point bend fixture with load and support spans of 0.5 and 2.0cm, respectively. The cross-head displacement speed was 8.5x10^{-4} cm/s. When spare samples were available, furnace annealing of controls at representative reactor temperature and atmosphere were performed to ensure oxidation or other factors were not responsible for composite strength degradation.

RESULTS AND DISCUSSION

Results from the room temperature bend testing for both non-irradiated and 1.1dpa irradiated composites are given in Table 1. It is noted that data of irradiated bend bars are from two samples at each condition, thus are statistically limited. However, several samples of each type were tested in the non-irradiated condition yielding statistically meaningful data. The matrix micro cracking stress was taken from the load-displacement curve as the departure from linearity and is therefore a macroscopic matrix stress. Undoubtedly, limited matrix micro cracking occurred at stresses below this value. The results from the furnace anneal of control samples was within statistical scatter of the non-annealed control samples, although the ultimate fracture stress for the annealed multilayer SiC interphase appears to be low. From Table 1, the macroscopic matrix microcracking stress is unchanged within statistical limitations. The ultimate fracture stress appears to be reduced in each case, with the multilayer SiC interphase showing the least reduction (~8-20%) and the porous SiC interphase showing the most (~35%). SEM of fracture surfaces shown elsewhere[14] exhibit exaggerated fiber pull-out.

Interphase Type	Matrix Stress (MPa)	Ultimate Stress (MPa)
Multilayer SiC		
As processed	250±32 (6tests)	507±75
Annealed, 385°C	264±17 (5tests)	438±21
Irradiated, 385°C	290.244	462.371
Pyrolytic Carbon		
As processed	214±38 (4tests)	375±51
Irradiated, 385°C	172, 285	235.347
Porous SiC		
As processed	298±22 (8tests)	515±19
Annealed, 260°C	335, 332	507.510
Irradiated, 260°C	268, 251	347.344
Annealed, 1000°C	287, 415	457.507
Irradiated, 1042-910°C	210.201	332.304

Table 1- Bend test results

Transmission electron microscopy reveals debonding at the fiber-interphase boundary for all three interphase materials. For the case of the porous SiC interphase, debonding within the fiber also occurred, though debonding at the boundary was still more common. Non-irradiated (fig 5A,C) and irradiated (fig 5B,D) interphases are shown in Figure 5. A debond width of ~50 nm is clearly visable at the boundary between the fiber and first multilayer SiC interphase (fig 5B). Debonding was observed for regions opaque to the 300 keV electrons indicating that debonding is not due to preferential milling by the argon ions. The porous SiC micorgraph (fig 5D) gives an examples of an failure which occurred within the fiber. Debond width in this case is on the order of 80 nm. Debonding for the pyrolitic carbon interphase (not shown) occurs at the interface between the fiber and the carbon interphase.

Assuming non-irradiated physical properties for fiber and matrix and the irradiation-induced volumetric changes for Hi-Nicalon™ and CVD SiC given in fig. 1, a residual stress analysis for the fiber/matrix interface has been performed. A complete explanation of this analysis is to be published elsewhere.[17] Micromechanical analysis of an infiltrated bundle was simplified by considering a system of three concentric cylinders fiber/interphase/matrix along with the following assumptions. 1) all the phases in the composite are linear elastic and isotropic.

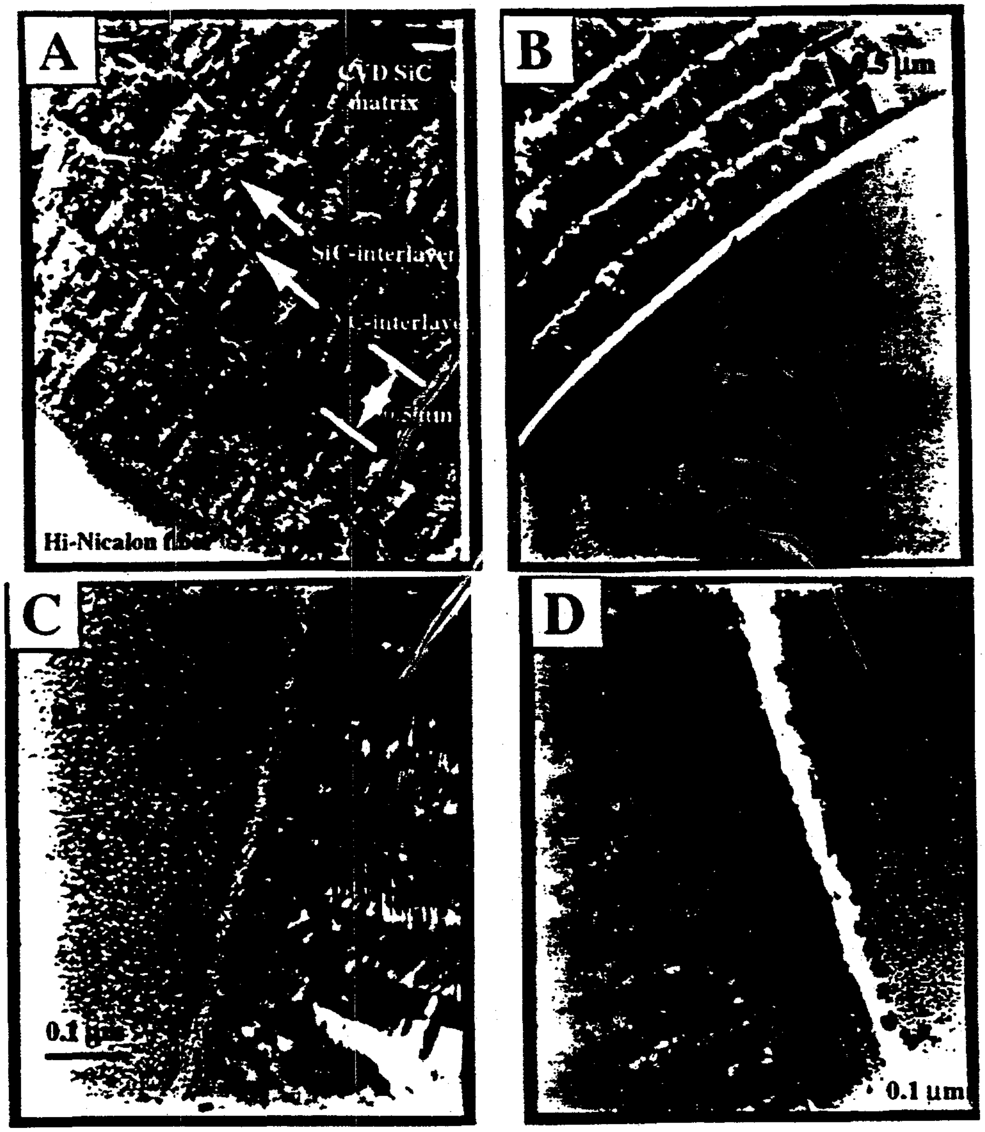

Figure 5- TEM of Composite Interphases :
A,B) non-irradiated and irradiated multilayer.
C,D) non-irradiated and irradiated porous SiC.

2) there is no relaxation of stresses during irradiation or during cooling from the fabrication temperature. 3) fibers and matrix exhibit volumetric changes as a result of neutron irradiation. Figure 6 shows the predicted radial stress at the fiber-fiber coating interface with neutron irradiation. Note that those stresses would exist only if continuity of radial displacement existed at the interface. At the 1.1 dpa dose level of this study fig. 6 yields a residual stress above 1 GPa, which is clearly higher than could be withstood by the interphases in this study, thus leading to predicted debonding.

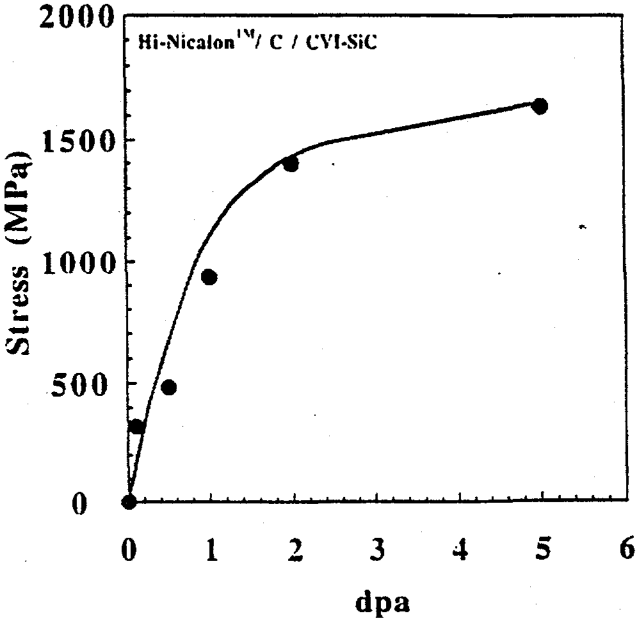

Figure 6- Irradiation-induced residual stress.

CONCLUSIONS

1) Bend strength results indicate that at 1.1dpa the ultimate fracture strength is decreased for Hi-Nicalon™ composites. The relative degree of decrease appears to depend on the interphase type with multilayer SiC interphase suffering the least reduction (~8-20%), followed by the pyrolitc carbon interface (~22%) and the porous SiC interphase (~35%).
2) Transmission electron microscopy reveals debonding at or near the interface between the fiber and interphase.
3) Residual stress analysis indicates that radial tensile stresses develop at the interphase during irradiation due to induced densification of the Hi-Nicalon™ fiber. These stresses are considerably higher than can be withstood by these or any viable SiC/SiC interphase, indicating that Hi-Nicalon™ is of limited application in neutron irradiation environments.

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