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Effect of neutron irradiation on tensile properties of unidirectional silicon carbide composites

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Abstract

Tensile properties of unidirectionally reinforced Hi-Nicalon™ Type S SiC fiber, chemically vapor-infiltrated (CVI) SiC-matrix composites, with either pyrolytic carbon (PyC) or multilayered PyC/SiC interphase, were characterized following neutron irradiations to the maximum fluence of 7.7×10^{25} n/m^2 at 380 and 800 °C. The stress–strain behavior of the multilayered interphase composites remained unmodified after irradiation. The PyC interphase composite increased in ultimate tensile stress and strain to failure following neutron irradiation, whereas the proportional limit stress exhibited a slight decrease. Potential mechanisms for these changes include accommodation of misfit stress through irradiation creep, reduced interfacial friction, and differential swelling among individual composite constituents.

1. Introduction

Silicon carbide (SiC) continuous fiber-reinforced SiC-matrix composites (SiC/SiC composites) are promising candidate materials for advanced blanket concepts for fusion reactors [1,2]. Those blanket concepts propose using a SiC/SiC wall, and/or channel, as the pressure boundary for helium and/or lead–lithium cooled structures, or as the insulating insert for the lead–lithium flow channels [2–4]. One of the most common requirements for such applications is that the SiC/SiC components retain their mechanical integrity under the combined loading of a high neutron flux and stress. For example, many of the conceptual advanced blanket designs assume a maximum design stress of ~200 MPa for SiC/SiC [3]. Even for the flow channel insert applications, where the expected external stress level is small, the magnitude of thermal stress can reach ~100 MPa or higher depending on design parameters [5]. Recent studies conclude that the non-irradiated strength of advanced SiC/SiC composites satisfies these requirements, and that neutron irradiation may not significantly deteriorate the strength [6].

Historically, the effect of neutron irradiation on the strength of SiC/SiC composites has been evaluated primarily by three- or four-point flexural tests. It was well understood that such a flexural test is suitable only for the purpose of screening experiments, as the flexural strength values can not be deconvoluted to obtain the intrinsic strength parameters of fibrous composites [7]. However, a testing

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standard for tensile properties of ceramic matrix composites was not available until ASTM standard C1275 was published in 2000. Moreover, the test standard and generally accepted testing guidelines required multiple specimens, each of which is substantially larger than can be accommodated in typical irradiation capsules. Recently, effort focusing on the establishment of a miniature test specimen technology for the tensile properties of ceramic matrix composites successfully led to recommendations of small specimen geometries and test procedures for irradiation effect studies [8].

It is now well established that the flexural strength of near-stoichiometric SiC fiber-reinforced, chemically vapor-infiltrated SiC-matrix composites, or advanced radiation-resistant CVI SiC/SiC composites, undergoes little or no degradation under neutron irradiation to ~8 dpa at 300–800 °C [9,10]. In this work, the effects of neutron irradiation in similar SiC/SiC composites are evaluated utilizing the miniature tensile test procedures developed for the irradiation effect studies. A unidirectional reinforcement architecture was employed in order to better investigate the effects of irradiation on constitutive mechanical properties of the continuous fiber-reinforced composites.

2. Experimental

The materials used were CVI SiC-matrix composites unidirectionally reinforced with Hi-Nicalon™ Type S near-stoichiometric SiC fibers. To facilitate the pseudo-ductile fracture, pyrolytic carbon (PyC) and multilayered (ML) PyC/SiC coatings were applied onto the fiber serving as the fiber-matrix interphases. Typical fiber volume fraction (V_f) and the total composite porosity are ~30% and ~16%, respectively. Attributes of the materials studied are summarized in Table 1. These materials are identical to those used in the previous studies and the specimens were taken from the same plates [10].

The miniature straight tensile bar specimens of 50 mm × 4 mm × ~1.5 mm were neutron-irradiated in two facilities in the High Flux Isotope Reactor (HFIR) at Oak Ridge National Laboratory; to 1.8×10^{25} n/m^2 ($E > 0.1$ MeV, the same shall apply throughout) at 380 °C in the peripheral target position rabbit facility, and to 7.7×10^{25} n/m^2 at 800 °C in RB-14J removable beryllium reflector facility.

Tensile tests were performed at room temperature, incorporating incremental unloading/reloading sequences but otherwise following the general guidelines of ASTM Standard C1275-00. The crosshead displacement rate was 0.5 mm/min, corresponding to a strain rate of $\sim 4 \times 10^{-4}$ s^{-1}. A pair of strain gauges attached at the gauge section center on both faces was used to determine both the tensile and the bending strains. The maximum fractional bending strain in successful tests appeared to be less than ~10%. The average strain was used for analysis of tensile properties. The proportional limit stress (PLS) was defined as the stress of 5% stress deviation from the extrapolated tangent modulus fit, as discussed elsewhere [11].

3. Results

In Fig. 1(a) and (b), the tensile stress–strain relationship is compared for non-irradiated and irradiated samples of the PyC and ML interphase composites, respectively. It is immediately noticed that no significant deterioration of strength took place for either composite. The non-irradiated PyC interphase composite failed at a stress slightly greater than the proportional limit, whereas some of the irradiated PyC interphase samples exhibited ultimate tensile stresses which greatly exceed the proportional limit stresses. It should also be noted that the strain to failure for the irradiated PyC interphase composite is substantially larger than the non-irradiated composite. As to the ML interphase composite, no significant difference in apparent stress–strain relationship is observed between the non-irradiated and the irradiated conditions.

The ultimate tensile stress of the unidirectional composites is plotted in Fig. 2(a) as a function of neutron dose. Data from an irradiation experiment in the Japan Materials Test Reactor (JMTR, Oarai, Japan) for the identical materials are included [12].

Table 1
List of materials studied

Material	Fiber	Interphase	Matrix	V_f (%)	Density (g/cm^3)	Porosity (%)
UD-HNLS/PyC	Hi-Nicalon™ Type-S	PyC$^{520-720\text{ nm}}$	SiC^{CVI}	~30	~2.6	~17
UD-HNLS/ML	Hi-Nicalon™ Type-S	$5 \times$ (PyC$^{20\text{ nm}}$/SiC$^{100\text{ nm}}$)	SiC^{CVI}	~30	~2.7	~15

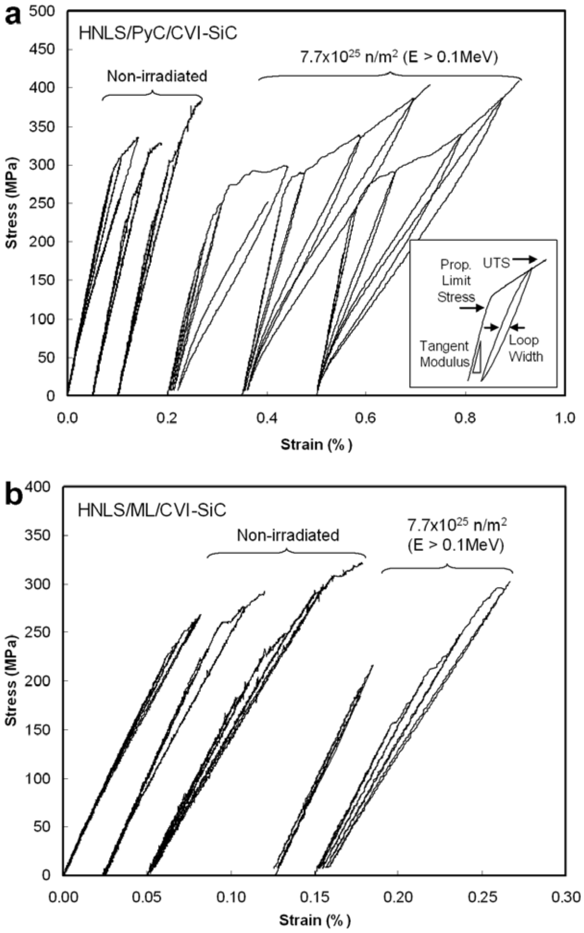

Fig. 1. Tensile stress–strain curves for non-irradiated and irradiated unidirectional Hi-Nicalon™ Type S composite with ~720 nm-thick PyC interphase (a) and multilayered ($PyC^{20\ nm}$/$SiC^{100\ nm}$)$_5$ interphase (b).

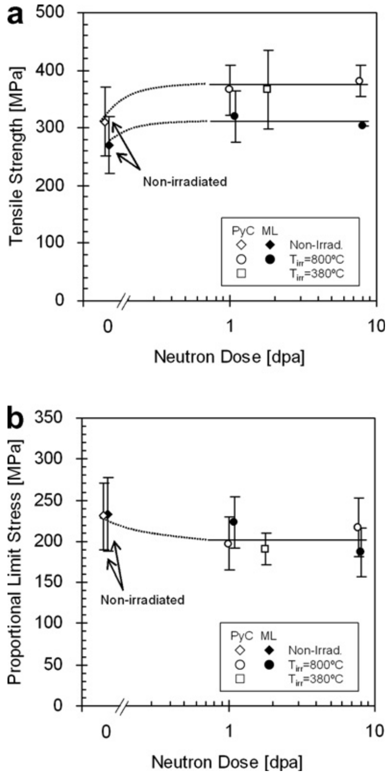

Fig. 2. Effect of neutron irradiation dose on ultimate tensile stress (a) and proportional limit tensile stress (b) of unidirectional Hi-Nicalon™ Type S composites. Error bars represent standard deviations. Data points at ~1 dpa and 800 °C are for identical materials irradiated in JMTR [12].

The error bars represent the standard deviations. Although there is no change in strength when the statistical uncertainty for individual data points is considered, the data suggest a slight irradiation-induced strength enhancement, as all the irradiated average strength values are higher than the non-irradiated strength value for the identical material for both the PyC and ML interphases.

In Fig. 2(b), the influence of neutron dose on the proportional limit tensile stress is presented. Although the proportional limit stress did not largely change following neutron irradiation, it appeared to slightly decrease after irradiation for both the PyC and ML interphases.

The measured tensile properties are summarized in Table 2. An effect of irradiation on increasing the strain to failure (ε_f) for the PyC-interphase composite, which is consistent with the observation in Fig. 1(a), is noticeable. The modification of tangent moduli (E) was not statistically significant. For

Table 2
Summary of non-irradiated and irradiated tensile properties

Material	Condition	E (GPa)	UTS (MPa)	PLS (MPa)	ε_f (%)
UD-HNLS/PyC	Non-irradiated	357 (30)	311 (60)	231 (42)	0.16 (0.06)
	1.8 dpa at 380 °C	354 (23)	366 (67)	197 (33)	0.40 (0.11)
	7.7 dpa at 800 °C	320 (11)	381 (26)	217 (36)	0.38 (0.02)
UD-HNLS/ML	Non-irradiated	375 (18)	271 (49)	232 (45)	0.09 (0.03)
	7.7 dpa at 800 °C	356 (34)	302	186 (30)	0.12

Numbers in parentheses represent standard deviations.

unidirectional composites, the longitudinal tangent modulus is determined only by the elastic moduli of the fiber and the matrix, both consist of beta-phase SiC. The expected change in elastic modulus of beta-phase SiC by irradiation in this temperature range is up to several percent [13]. Therefore, it is reasonable to expect that only very minor change in tangent modulus would be observed with this data scatter.

4. Discussion

The most important result obtained in this work is the demonstrated insensitivity of tensile strength of the advanced SiC/SiC composites to neutron irradiation. However, a significant effect of irradiation on the tensile behavior of the PyC-interphase composite beyond the proportional limits was observed. Features of the apparent irradiation effect on tensile behavior of the PyC-interphase composite are: (1) extended non-linear deformation beyond the proportional limit, (2) slightly reduced proportional limit stress, and (3) enlarged width of the unloading–reloading hysteresis loop. The increased ultimate stress and strain to failure are primarily the consequences of extended non-linear deformation.

The width of the unloading–reloading hysteresis loop at the half peak stress ($\delta\varepsilon_{1/2}$) can be related with the interfacial sliding stress (τ) and the mean matrix crack spacing ($\bar{d}$) by the following equation [14]:

$$\delta\varepsilon_{1/2} = \frac{b_2(1-a_1V_\mathrm{f})^2\sigma_\mathrm{p}^2}{8f^2\tau E_\mathrm{m}}\cdot\frac{r}{\bar{d}} = K\frac{\sigma_\mathrm{p}^2}{\tau E_\mathrm{m}\bar{d}}, \qquad (1)$$

where σ_p is the peak stress, E_m is the matrix modulus, r is fiber radius, a_1 and b_2 are the Hutchinson–Jensen parameters, and K is a constant that is not affected by irradiation [15]. As mentioned earlier, irradiation has not significantly affected the elastic modulus. As a result, any change in the hysteresis loop width is related to changes in the interfacial sliding stress and crack spacing. From this equation, the enlarged hysteresis loop width implies a lower sliding stress and/or higher crack density. The interfacial sliding stress is anticipated to degrade, since the near-isotropic graphite first contracts and then starts to swell accompanying a major strength degradation, either of which should cause a decrease in sliding stress. Hence, it is reasonable to assume that reduced interfacial friction has potentially contributed to the observed hysteresis loop widening. However, a detailed interfacial shear properties analysis in the companion work by Nozawa et al., did not indicate a significant irradiation-induced change in the interfacial friction for identical material [16]. Therefore, the hysteresis loop widening may have been caused primarily by the higher matrix crack density in the irradiated samples.

The matrix cracking stress (σ_mc) can be given by the equations below [17,18]:

$$\sigma_\mathrm{mc} = \sigma_\mathrm{mc}^0 - \sigma_\mathrm{T}, \qquad (2)$$

$$\sigma_\mathrm{mc}^0 = \left[\frac{6\tau\gamma_\mathrm{m}}{r}\cdot\frac{V_\mathrm{f}^2E_\mathrm{f}E_\mathrm{c}^2}{(1-V_\mathrm{f})E_\mathrm{m}^2}\right]^{1/3}, \qquad (3)$$

where σ_mc^0 is the matrix cracking stress in an internal stress-free condition, σ_T is the misfit stress defined in Ref. [17], γ_m is matrix fracture energy, and E_f and E_c are Young's moduli of fiber and composite, respectively. The matrix fracture energy stays unchanged or possibly increases with irradiation [13,19]. However, any increase is likely small, since the proposed primary toughening mechanism for the chemically vapor deposited SiC is associated with cleavage fracture of large grains and therefore may not effectively operate for the predominantly fine-grained microstructures in the CVI SiC matrix. Assuming that the interfacial sliding stress and all other parameters in Eq. (3) would not be significantly influenced, σ_mc^0 should be insensitive to neutron irradiation.

The misfit stress in the as-infiltrated condition originate from the mismatch in the coefficient of thermal expansion (CTE) between the fiber and the matrix. The manufacturer-claimed CTE of $\sim 5.1\times10^{-6}\ \mathrm{K}^{-1}$ (20–500 °C) for Hi-Nicalon™ Type S fiber is slightly greater than that for vapor-deposited SiC. This should result in a compressive axial component of misfit stress for the matrix, thus increasing the apparent matrix cracking stress and the proportional limit stresses. Since the irradiation temperatures in this study are lower than the matrix infiltration temperature of 1100–1200 °C, the misfit stress will be relaxed by the irradiation creep deformation of both the fibers and the matrix, leaving a reduced misfit stress when the specimens are cooled down to room temperature. Thus, irradiation creep can mitigate the internal stress and consequently lower the matrix cracking stress [20].

In an ideal condition, the matrix crack density is proportional to the matrix damage parameter (D) as defined in the following equation [21]:

$$D = \frac{E_\mathrm{c} - E^*}{E^*}, \qquad (4)$$

where E^* is the tangent modulus of the unloading curve. In Fig. 3, the matrix damage parameters for non-irradiated and irradiated PyC-interphase composites are compared as a function of the peak tensile stress. The lower onset stress and the greater slope for the irradiated samples clearly indicate their increased matrix cracking susceptibility. It is also noted from Fig. 3 that the matrix crack saturation is almost reached for the irradiated samples, while the non-irradiated samples fail before crack saturation. The tensile failure of composites before matrix crack saturation often implies excess interfacial friction. Although the measured interfacial friction change was not significant, it is possible that a minor change in the friction contributed to the observed enhanced non-linear deformation of the irradiated samples. It is important to note that this does not necessarily imply that the interphase system is incorrect for a true multiaxial composite system. The external tensile stress required for introduction of matrix cracks is much lower in the woven architectures which are used for practical applications.

From Fig. 3, it is obvious that a similar matrix crack density occurs at substantially different stress levels in non-irradiated and irradiated samples. For example, a matrix damage parameter of 0.5 occurs at ~280 MPa in irradiated specimens, whereas it occurs at ~330 MPa in the non-irradiated specimens. When the hysteresis behavior is compared at these peak stress levels for their respective conditions, the nearly closed non-irradiated loops contrast with the widely open irradiated loops. This implies that the radiation-induced mitigation of compressive internal stress in the matrix is the cause of the observed behavior, rather than an unlikely substantial reduction in interfacial friction discussed earlier. In fact, the magnitude of compressive matrix stresses roughly estimated from the regression analysis of the tensile data decrease from 78 ± 21 MPa for non-irradiated to 34 ± 24 MPa for irradiated specimens. Irradiation can also mitigate the axial tensile residual stress in the fibers through irradiation creep, in the same way as reducing the compressive residual stress in the matrix, thus increasing the apparent failure strain of the fibers. Therefore, the observed irradiation effect on the extended non-linear deformation of the PyC-interphase composites may be attributed to: (1) a reduced residual stresses in matrix and fibers by irradiation creep, and (2) possibly reduced interfacial frictional stress. Potential differential swelling among the matrix, interphase, and fibers, which was not investigated in this work, may also contribute to the macroscopic irradiation effects in the composites.

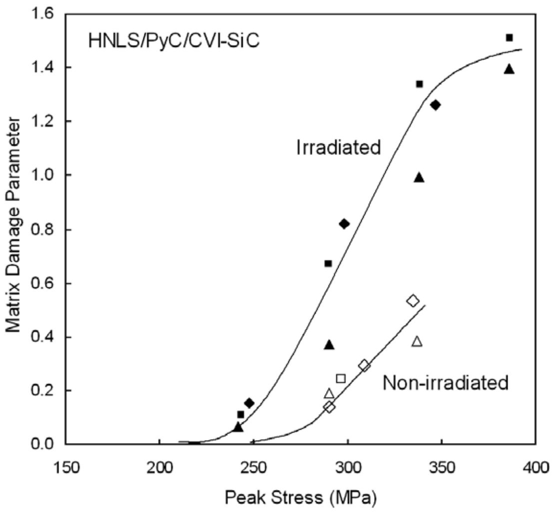

Fig. 3. Effect of neutron irradiation on evolution of matrix damage parameter with increasing peak tensile stress.

The most likely reason for the unmodified tensile behavior for the ML interphase composite is that the higher interfacial friction combined with the high matrix cracking stress, which is particular to the unidirectional reinforcement architecture, promoted premature composite failure even after irradiation. The single fiber push-out measurement and analysis indicate that both the interfacial debond and frictional stresses for the ML interphase are substantially higher than those for the PyC interphase [16].

5. Conclusions

Neutron irradiation upto a fluence of 7.7×10^{25} n/m^2 at 380 and 800 °C did not induce a deterioration of tensile strength of unidirectional Hi-Nicalon™ Type S SiC fiber-reinforced, CVI SiC-matrix composites with either PyC or ML PyC/SiC interphase. The tensile stress–strain behavior of the ML interphase composites remained unmodified after irradiation. In the PyC interphase composite, the ultimate tensile stress and the strain to failure improved at the slight expense of the proportional limit stress. Potential mechanisms for these changes include the irradiation creep-induced misfit stress accommodation, reduced interfacial friction, and the differential swelling among individual composite constituents.

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