EFFECT OF NEUTRON IRRADIATION ON TENSILE PROPERTIES OF UNIDIRECTIONAL SILICON CARBIDE COMPOSITES–Y. Katoh, T. Nozawa, L. L. Snead, and T. Hinoki (Oak Ridge National Laboratory)

OBJECTIVE

The objective of this work was to determine the effects of neutron irradiation in chemically vapor-infiltrated Hi-Nicalon™ Type-S SiC/SiC composites. 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. A miniature tensile test procedure developed for the irradiation effect studies by the authors was utilized.

SUMMARY

Tensile strength properties of unidirectionally reinforced Hi-Nicalon™ Type S SiC fiber, CVI SiC-matrix composites with either PyC or multilayered $(PyC/SiC)_n$ interphase was characterized after neutron irradiation to the maximum fluence of 7.7 x 10^{25} n/m^2 at 380 and 800°C in High Flux Isotope Reactor at Oak Ridge National Laboratory. The stress/strain behavior of the multilayered interphase composites remained unmodified after irradiation. The PyC interphase composite improved the ultimate tensile stress and the strain to failure by neutron irradiation, in slight expense of the proportional limit stress. Potential mechanisms for these changes include the irradiation creep-induced misfit stress mitigation, reduced interfacial friction, and the differential swelling among individual composite constituents. Substantial difference in irradiation effect on non-linear deformation of SiC/SiC composites is expected between unidirectional and multi-dimensional architectures.

PROGRESS AND STATUS

Introduction

Silicon carbide (SiC) continuous fiber-reinforced SiC-matrix composites (SiC/SiC composites) are the promising candidate materials for the advanced blanket design concepts for fusion reactors [1,2]. Those blanket concepts assume the 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 primary common requirements for such applications is that the SiC/SiC components retain their mechanical integrity under a combined loading of high flux neutrons and the substantial internal / external stresses. For example, many of the conceptual advanced blanket design works assume the maximum design stresses of ~ 200 MPa for SiC/SiC blanket structures [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 depending on design parameters [5]. Recent studies support that the non-irradiated strength of the advanced SiC/SiC composites satisfies these requirements, and 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-point or four-point flexural tests. It was well understood that a flexural test is suitable only for the purpose of screening-type experiments, as the flexural strength values can not be deconvoluted to intrinsic mechanical properties of fibrous composites [9]. However, a testing standard for tensile properties of ceramic matrix composites had not been available until ASTM standard C1275 was published in year 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 lead to provide recommendation of small specimen geometries and test procedures for the irradiation effect studies [8].

It is well perceived 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 by neutron irradiation to ~ 8 dpa at 300–800°C [9,10]. In this work, the effects of neutron irradiation in similar SiC/SiC composites were evaluated utilizing the miniature tensile test procedures developed for the irradiation effect studies. Also, 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.

Experimental Procedure

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 $(PyC/SiC)_n$ coatings were applied onto the fiber as the fiber-matrix interphases. Typical fiber volume fraction and the 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 50mm x 4mm x ~ 1.5mm were neutron-irradiated in two facilities at High Flux Isotope Reactor (HFIR) at Oak Ridge National Laboratory (Oak Ridge, TN, USA); to 1.8×10^{25} n/m^2 (E > 0.1 MeV, the same shall apply hereinafter) 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.

The tensile tests were performed at room temperature, incorporating incremental unloading / reloading sequences but in otherwise following the general guidelines in ASTM Standard C1275-00. The crosshead displacement rate was 0.5mm/min. A pair of strain gauges attached at the gauge section centers on both faces was used to determine both the average 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 at 5% stress deviation from the extrapolated tangent modulus fit [11].

Results

In Figs. 1 (A) and (B), tensile stress/strain relationship is compared for non-irradiated and irradiated samples of the PyC and multilayered interphase composites, respectively. It is immediately noticed that no significant deteriorating irradiation effect on strength took place in either composite. The apparent feature of non-irradiated PyC interphase composite is the general failure at the stress slightly beyond the proportional limit, whereas some of the irradiated PyC interphase samples exhibit the ultimate tensile stresses which largely 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 that for the non-irradiated composite. As to the multilayered interphase composite, no significant difference in apparent stress/strain relationship is observed between the non-irradiated and the irradiated conditions.

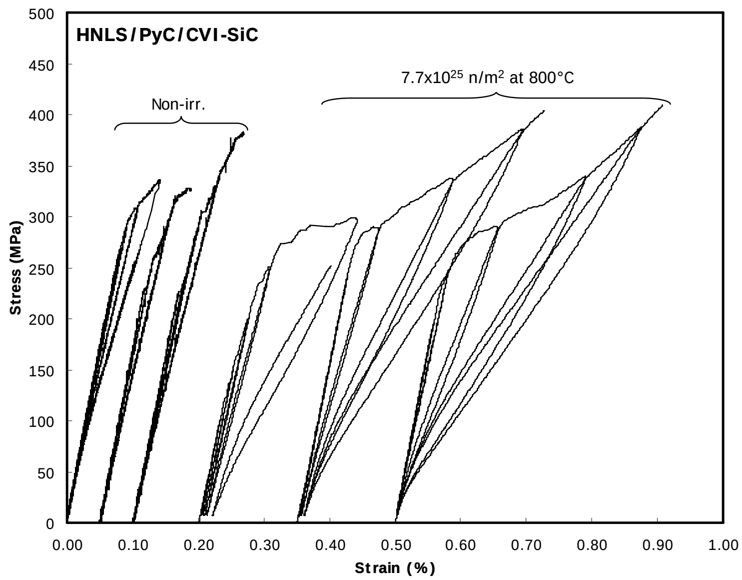
The ultimate tensile stress of the unidirectional composites is plotted in Fig. 2 (A) as a function of neutron

Table 1. List of materials studied

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

dose. Data from an irradiation experiment in Japan Materials Test Reactor (JMTR, Oarai, Japan) for the identical materials were included [12]. The error bars represent the standard deviations in Fig. 2 (A) and the following figures. Although the statistical uncertainty may not be eliminated when comparing between particular data points, it is reasonable to admit the slight irradiation-induced strength enhancement, as all the irradiated average strength values are higher than the non-irradiated strength value for the identical material.

(A)



(B)

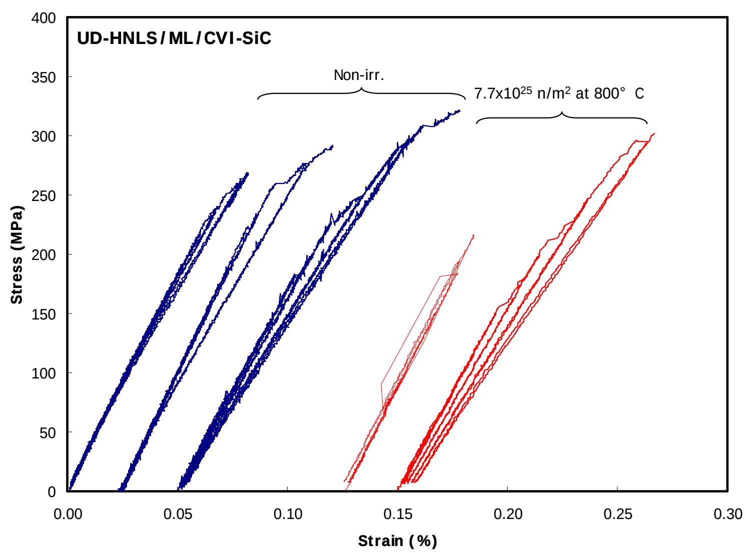
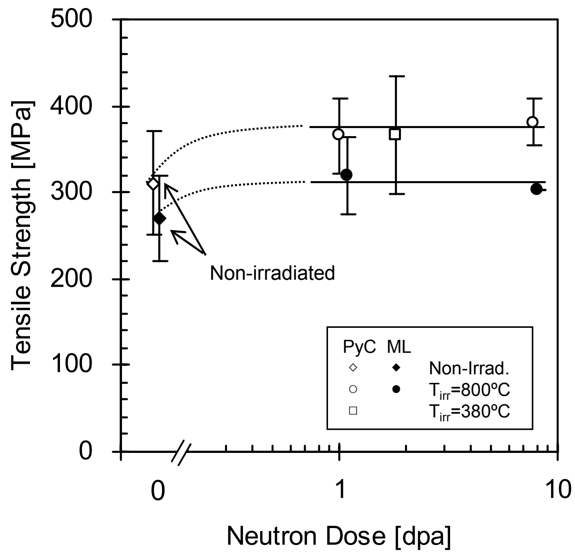


Fig. 1. Tensile stress - strain curves for non-irradiated and irradiated unidirectional Hi-Nicalon™ Type S composite with ~ 720 nm - thick PyC interphase (A) and multilayered (PyC^{20nm} / SiC^{100nm})$_5$ interphase (B).



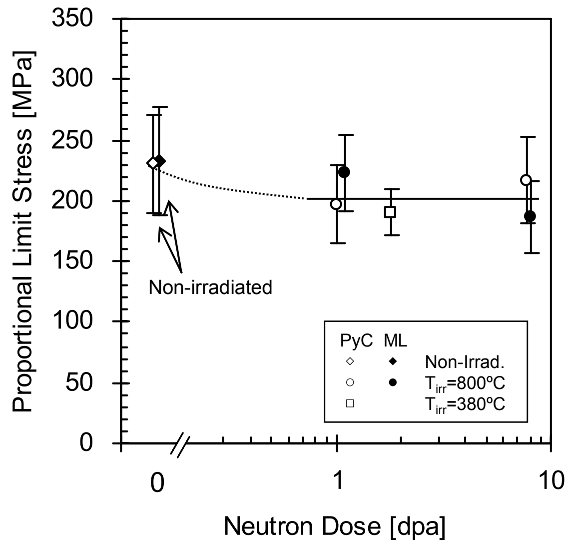


Fig. 2. Effect of neutron irradiation dose on ultimate tensile stress (A) and proportional limit tensile stress (B) of unidirectional Hi-Nicalon™ Type S composites. Error bars represent standard deviations.

In Fig. 2 (B), the influence of neutron dose on the proportional limit tensile stress is presented. The proportional limit stress did not appear to be very sensitive to neutron irradiation, but is likely to slightly decrease after irradiation.

The tensile properties measured are summarized in Table 2. The irradiation effect on the strain to failure for the PyC-interphase composite, which is consistent with the observation in Fig. 1 (A), is noticeable. The modification of tangent moduli was not statistically significant. For unidirectional composites, the longitudinal tangent modulus is determined only by the elastic moduli of the fiber and the matrix, both comprise of beta-phase SiC in the materials studied. The expected change in elastic modulus of beta-phase SiC by irradiation in this temperature range is a few to several percent [13]. Therefore, it is reasonable with this data scatter not to admit the tangent modulus change.

Table 2. Summary of non-irradiated and irradiated tensile properties.
Numbers in parentheses represent standard deviations.

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)
	1.8×10^{25} n/m^2 @ 380°C	354 (23)	366 (67)	197 (33)	0.40 (0.11)
	7.7×10^{25} n/m^2 @ 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)
	7.7×10^{25} n/m^2 @ 800°C	356 (34)	302	186 (30)	0.12

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, significant effect of irradiation on the tensile behavior of the PyC-interphase composite beyond the proportional limits was observed. The apparent irradiation effect features on tensile behavior of the PyC-interphase composite are 1) the extended non-linear deformation beyond the proportional limit, 2) the slightly reduced proportional limit stress, and 3) the enlarged width of the unloading - reloading hysteresis. The increased ultimate stress and the strain to failure are primarily the consequences of the 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].

$$\delta\varepsilon_{1/2} = \frac{b_2(1-a_1 f)^2 \sigma_p{}^2}{8 f^2 \tau E_m} \cdot \frac{r}{\bar{d}} \tag{1}$$

where f is fiber volume fraction, σ_p is the peak stress, E_m is the matrix modulus, r is fiber radius, and a_1 and b_2 are the Hutchinson-Jensen parameters [15]. According to this equation, the enlarged hysteresis loop width implies the lower sliding stress and/or higher crack density, since other parameters do not significantly change by irradiation. Since the strength degradation of PyC interphase is anticipated, it is reasonable to assume that reduced interfacial friction 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 the identical material [16]. Therefore, the hysteresis loop widening may have been caused 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].

$$\sigma_{mc} = \sigma_{mc}^0 - \sigma_T \tag{2}$$

$$\sigma_{mc}^0 = \left[\frac{6\tau\gamma_m}{r} \cdot \frac{V_f{}^2 E_f E_c{}^2}{(1-V_f)E_m{}^2}\right]^{1/3} \tag{3}$$

where σ_{mc}^0 is the matrix cracking stress in an internal stress-free condition, σ_T is 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 increased by irradiation [13,19], but it is likely that the extent of potential increase is 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 highly fine-grained microstructures in the SiC matrix deposited by CVI. Assuming all other parameters, including the interfacial sliding stress, in Eq. 3 would not be significantly influenced, σ_{mc}^0 can be insensitive to neutron irradiation.

The misfit stress in an as-infiltrated condition is originated from the mismatch in coefficient of thermal expansion (CTE) between the fiber and the matrix. The manufacturer-claimed CTE of ~ 5.1 x 10^{-6} K^{-1} (20–500°C) for Hi-Nicalon™ Type S fiber is slightly larger than that for vapor-deposited SiC. This should result in compressive axial component of misfit stress for the matrix, thus increases apparent matrix cracking stress and the proportional limit stress. 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 reduced misfit stress when the specimens are cooled down to room temperature. Thus irradiation creep can mitigates the internal stress and consequently lower the matrix cracking stress.

The matrix crack density is ideally proportional to matrix damage parameter (D) in the following definition [20].

$$D = \frac{E_c - E^*}{E^*} \quad (4)$$

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 larger slope for the irradiated samples clearly indicate the increased matrix cracking susceptibility. It is also noted in Fig. 3 that the matrix crack saturation is almost reached for the irradiated samples, while the non-irradiated samples fail before the crack saturation. The tensile failure of composites before the 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. However, this does not mean that the applied interphase was too strong for this composite system, because the external tensile stress required for introduction of matrix cracks is much lower in woven architectures, which are used for practical applications.

From Fig. 3, it is obvious that the similar matrix crack density occurs at substantially different stress levels in non-irradiated and irradiated samples. For example, matrix damage parameter of 0.5 occurs at ~ 280 MPa in irradiated conditions, whereas it occurs at ~ 330 MPa in a non-irradiated condition. When the hysteresis behavior is compared at these peak stress levels for respective conditions, the nearly closed irradiated loops contrast the widely open non-irradiated loops. This implies the radiation-induced mitigation of compressive internal stress in the matrix, rather than the unlikely substantial reduction in interfacial friction discussed earlier. In fact, the magnitude of compressive matrix stresses roughly estimated from the tensile regression lines decrease from 78 ±21 MPa non-irradiated to 34 ±24 MPa irradiated. 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 cause prolonging

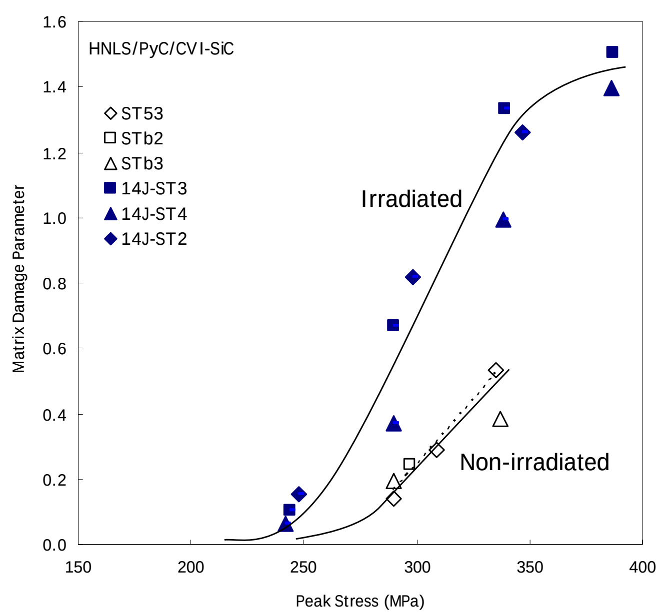


Fig. 3. Effect of neutron irradiation on evolution of matrix damage parameter with increasing peak tensile stress.

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) 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 have also contributed to the macroscopic irradiation effects in the composites.

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

Conclusions

Neutron irradiation to the maximum fluence of 7.7 x 10^{25} n/m^2 at 380 and 800°C did not impose deteriorating effect on tensile strength of unidirectional Hi-Nicalon™ Type S SiC fiber-reinforced, CVI SiC-matrix composites with either PyC or multilayered $(PyC/SiC)_n$ interphase. The tensile stress/strain behavior of the multilayered interphase composites remained unmodified after irradiation. The PyC interphase composite improved the ultimate tensile stress and the strain to failure in slight expense of the proportional limit stress. Potential mechanisms for these changes include the irradiation creep-induced misfit stress mitigation, reduced interfacial friction, and the differential swelling among individual composite constituents. Substantial difference in irradiation effect on non-linear deformation of SiC/SiC composites is expected for between unidirectional and multi-dimensional architectures.

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