Evaluation of Fiber/Matrix Interfacial Strength of Neutron Irradiated SiC/SiC Composites Using Hysteresis Loop Analysis of Tensile Test

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Advanced SiC/SiC composites composed of highly-crystalline and near-stoichiometric SiC fibers, SiC matrix by CVI method, and PyC interphase were irradiated up to \(1.0 \times 10^{23}\) n/m² (\(E > 0.1\) MeV, \(\sim 1\) dpa) at 1273 K.Unload/reload cyclic tensile test and hysteresis loop analysis were carried out in order to identify neutron irradiation effects on interfacial properties of advanced SiC/SiC. The composites exhibited good irradiation resistance in ultimate tensile strength (\(\sim 10\%\) increase), but there were slight reduction of elastic modulus and proportional limit stress (PLS) (\(\sim 10\%\)). Wider hysteresis loops and lower gradient of the curves beyond PLS from the strain–stress curves, and longer pullouts of fiber from the SEM observation were observed, which imply weaker F/M interactions after neutron irradiation. From the hysteresis loop analysis, degradation of interfacial sliding stress and negative increment of misfit stress after neutron irradiation were obtained.

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1. Introduction

Silicon carbide (SiC) and its composites (SiC/SiC) are attractive structural materials for fusion reactors and gas fast reactors because of their superior mechanical properties at high temperature and good irradiation resistance.¹ The mechanical properties of SiC/SiC composites after irradiation depend on the properties of their constituents, especially fiber/matrix (F/M) interface.²–⁶ SiC/SiC composites reinforced with less crystalline and non-stoichiometric SiC fibers have shown the interfacial debonding due to mismatch of swelling behavior of the fiber and β-SiC matrix after neutron irradiation. In contrast, due to highly crystalline and near stoichiometric SiC fibers, advanced SiC/SiC composites have exhibited excellent irradiation stability in ultimate bend/tensile strength.⁶,⁷

In our previous work,⁸ tensile, flexural and inter-laminar shear properties of advanced SiC/SiC composites with various interfaces (pyrolytic carbon (PyC), multilayered SiC/PyC (ML) and pseudo- porous SiC) after neutron irradiation up to \(1 \times 10^{23}\) n/m² at mainly 1025 K was examined. Advanced SiC/SiC composites exhibited good irradiation resistance in ultimate bend/tensile strength, but the result of a slight loss of proportional limit stress (PLS) in the monotonic tensile test and \(\sim 40\%\) degradation of interlaminar shear strength in the double notched specimen (DNS) test suggested degradation of F/M interphase. However, irradiation effects on F/M interphase remain uncertain because it is well known that PLS depends on various constituent properties of composites and inter- laminar shear strength also might be dependent on size, geometry and distribution of pores. Therefore, irradiation effects on the interfacial role became one of the most important issues to be discussed.

The main purpose of this study is to identify neutron irradiation effects on interfacial properties of advanced SiC/SiC composites. For this purpose, we applied unloading/reloading tensile test and hysteresis loop analysis.

2. Experimental Procedure

2.1 Materials

Unidirectional (UD) SiC/SiC composites were prepared by isothermal chemical vapor infiltration (I-CVI) process at Hyper-Therm High-Temperature Composites, Inc. They were composed of Hi-Nicalon™ Type-S fibers as reinforcements with single pyrolytic carbon (PyC). The thickness of PyC coating was 520, 720 nm (PyC520, PyC720), the density of SiC/SiC was 2.58 Mg/m³, and the fiber volume fraction was 29%. Materials fabrication is detailed elsewhere.⁹,¹⁰

2.2 Neutron irradiation

Neutron irradiation was performed in the Japan Materials Testing Reactor (JMTR) at Oarai, Japan. Neutron fluence and temperature were \(1.0 \times 10^{23}\) n/m² (\(E > 0.1\) MeV, correspond to 1 dpa) and 1273 K, respectively.

2.3 Tensile test

Tensile tests were carried out in accordance with ASTM C 1275 at room temperature by using a face-loaded miniature specimen developed in previous works.¹¹,¹² The size of tensile test specimens was \(4 \times 1.5 \times 50\) mm except unirradiated PyC720. First, 1.0 mm-thick aluminum tabs were attached on both ends of rectangular specimens. Second, a couple of 6.0 mm-length gauges were adhered in the center of gauge sections on both sides. Prepared specimens were mounted in the test flame by connection of wedge-type grips gripping system was kept in the plastic bags during the test. Tensile tests were conducted under the displacement control of 0.5 mm/min. The detailed tensile setup is in,⁸ and more details are discussed elsewhere.¹¹,¹² In addition, the size of unirradiated PyC720 was \(2 \times 1.5 \times 50\) mm, and detailed procedures of the tensile test of unirradiated PyC720 were given elsewhere.²,⁹
2.4 Microstructural observations

After the tests, the fractured surfaces of tensile specimens were observed by scanning electron microscopy (SEM).

3. Results

Typical tensile stress/strain curves of PyC520 after neutron irradiation were shown in Fig. 1. Initial linear region and non-linear behavior beyond it during tensile loading were observed in all composites; ~435 MPa of ultimate tensile stress (UTS) with ~235 MPa of proportional limit stress (PLS) and ~319 GPa of elastic modulus, i.e. unirradiated composites, fractured at lower stress ~365 MPa of UTS with ~268 MPa of PLS and ~365 GPa of elastic modulus. As shown in Fig. 1, wider maximum hysteresis loop width and larger permanent strain after neutron irradiation were observed.

Table 1 exhibits the summary of unirradiated and irradiated tensile properties after neutron irradiation up to 1 dpa at 1273 K; (a), (b) unirradiated, (c), (d) irradiated.

Figure 2 shows the relative tensile properties after neutron irradiation up to 1 dpa at 1273 K. PyC composites exhibited good irradiation resistance in UTS (% increase), but a slight loss of both elastic modulus (~10%) and PLS (~10%) were observed after neutron irradiation up to 1 dpa at 1273 K.

Figure 3 exhibits the typical fractured surfaces by SEM observation of the unirradiated and irradiated composites. Longer fiber pullouts were observed in the irradiated composites (0–287 μm, 94 μm in average), comparing with the unirradiated composites (0–121 μm, 46 μm in average). In addition, some regions of the fractured surfaces in the unirradiated composites were flat with no pullouts.

4. Discussions

4.1 Neutron irradiation effects on tensile elastic modulus

The slight loss of tensile elastic modulus after neutron irradiation at 1273 K was similar to the decrease irradiated at 1073 K. According to previous reports, moduli of Hi-Nicalon™ Type-S single fiber by tensile test and CVD-SiC composite by nano-indentation techniques decreased ~10% at 1 dpa, and kept this level up to at least 10 dpa in the temperature range from 423 to 823 K. At 1023 to 1273 K, the modulus was observed to decrease more, reaching ~20% at 1 dpa and ~30% at 10 dpa.

Table 1 Summary of unirradiated and irradiated tensile properties. Averages and standard deviations were shown. PyC means PyC520 and PyC720. The parentheses in Number of test indicate the number of tests fractured successfully.

<table>
<thead>
<tr>
<th>Specimen</th>
<th>Irradiation condition</th>
<th>$E$/GPa</th>
<th>PLS/MPa</th>
<th>UTS/MPa</th>
<th>Number of test</th>
</tr>
</thead>
<tbody>
<tr>
<td>PyC520</td>
<td>Unirrad.</td>
<td>314.9 ± 31.3</td>
<td>150.1 ± 3.7</td>
<td>349.1 ± 16.1</td>
<td>2 (2)</td>
</tr>
<tr>
<td>PyC520</td>
<td>1273 K, 1 dpa</td>
<td>296.8 ± 16.3</td>
<td>175.5 ± 45.6</td>
<td>409.2 ± 19.6</td>
<td>3 (2)</td>
</tr>
<tr>
<td>PyC720</td>
<td>Unirrad.</td>
<td>329.1 ± 33.2</td>
<td>236.9 ± 16.3</td>
<td>287.4 ± 37.3</td>
<td>5 (5)</td>
</tr>
<tr>
<td>PyC720</td>
<td>1273 K, 1 dpa</td>
<td>266.1 ± 5.8</td>
<td>189.4 ± 33.9</td>
<td>435.5 ± 0.0</td>
<td>3 (1)</td>
</tr>
<tr>
<td>PyC</td>
<td>Unirrad.</td>
<td>325.0 ± 33.3</td>
<td>212.1 ± 41.6</td>
<td>325.0 ± 42.9</td>
<td>7 (7)</td>
</tr>
<tr>
<td>PyC</td>
<td>1273 K, 1 dpa</td>
<td>281.4 ± 19.6</td>
<td>182.4 ± 40.8</td>
<td>418.0 ± 20.2</td>
<td>7 (3)</td>
</tr>
</tbody>
</table>
elastic modulus exhibited nearly no reduction at least up to 3 dpa, following 10% decrease at ≈ 8 dpa. Therefore, the small amount of degradation in elastic modulus of advanced SiC/SiC after neutron irradiation might be occurred.

### 4.2 Neutron Irradiation Effects on PLS

Generally, proportional limit stress, i.e., matrix cracking stress \( \sigma_{mc} \), is closely dependent on interfacial properties and moduli of fiber and matrix and misfit stress.\(^{17,18} \)

\[
\sigma_{mc} = E_f \left[ \frac{6\sigma_T^2 E_m f E_i}{(1-f) E_m^2} \right]^{1/3} - \sigma^T
\]

where \( \sigma^T \) is the misfit stress, \( \sigma_m \) is the matrix fracture energy, \( E_i \) is the Young’s modulus of fiber, \( E_m \) is that of matrix, \( E \) is that of composites, \( r \) is the interfacial sliding stress, \( f \) is the fiber volume fraction, and \( R \) is the fiber radius, respectively. As discussed in Section 4.1, the irradiation-induced change of elastic moduli of highly crystalline SiC matrix and fiber should be minor at 1273 K up to 1 dpa.

Wider hysteresis loop width and lower gradient of the curves beyond PLS (Fig. 1), and longer fiber pullouts (Fig. 3) imply the degradation of interfacial properties. In order for further investigation of interfacial properties, the method of unloading/reloading hysteresis loop analysis methodology proposed by Vagaggini, Domergue, and Evans was applied.\(^{18-20} \)

In this analysis, inelastic strain index, which is important for estimating the interfacial property, and Young’s modulus of material with matrix cracks were obtained from the reciprocal moduli during reloading tensile-strain curves from different peak stress (\( \sigma_p \); the transition stress from reloading to unloading). The detailed method for unidirectional composites is given in elsewhere.\(^{18} \)

Figure 4 exhibits the inelastic strain index (\( L \)) of advanced SiC/SiC composites after neutron irradiation, which is given by,

\[
L = \frac{b_2(1-a_1 f)^2}{4f^2 E_m} \frac{R}{d} d
\]

with \( \delta \) being the matrix crack spacing, and \( a_1, b_2 \) the Hutchinson and Jensen parameters.\(^{21} \) As defined in eq. (2), \( L \) is dependent on interfacial stress and mean matrix crack spacing. Unfortunately, as the mean matrix crack spacing was not measured in individual tested samples, the saturated matrix crack spacing (\( \delta_s \)) was determined by the crack spacing at fiber bundles of the fractured surfaces by SEM observation, in accordance with the matrix damage parameter (\( D \)) which is defined as,

\[
\frac{E/E^* - 1}{B} = \frac{\delta}{\delta_s} \equiv D
\]

where \( B \) is a constant for a particular composite (fixed \( f \) and \( E_i/E_m \)) and \( E^* \) is the Young’s modulus of the composite with matrix cracks, respectively. As shown in eq. (3), \( D \) is in inversely proportion to matrix crack spacing, and is almost saturated around 300 MPa of peak stress (Fig. 5), so we applied the saturated matrix crack spacing from the SEM observation (\( \delta_{s, \text{unirrad}} = 58 \mu m, \delta_{s, \text{irrad}} = 23 \mu m \) ) to eq. (2) at 306 MPa for the unirradiated specimens and 289 MPa for the irradiated. As a result, each interfacial sliding stresses was obtained (\( \tau_{\text{unirrad}} = 35 \) MPa, \( \tau_{\text{irrad}} = 21 \) MPa). Actually, about 40% degradation of the interfacial sliding stress after neutron irradiation up to 1 dpa at 1273 K was confirmed.

As shown in eq. (1), PLS also depends on misfit stress, which correlates with residual stress in composites. Figure 6 exhibits the misfit stress for large debond energy as a function of peak stress using the following method in Ref. 20). The conversion of misfit stress to residual stress components is provided elsewhere.\(^{18} \) Negative increment of misfit stress was detected after neutron irradiation; \( \sigma_{\text{irrad}}^\tau \) was in range of +80 ~ −10 MPa, −4 MPa in average at the highest peak stress, and \( \sigma_{\text{irrad}}^\tau \) was −60 ~ −230 MPa, −150 MPa, respectively.
This result indicates that slight difference of swelling between each constituent (fiber and PyC interphase, matrix and PyC) might affect the residual stress in PyC interphase and cause the degradation of the interfacial property, ignoring the influence of interphase thickness and roughness of fiber. Or it is also considered that the degradation might be caused by volumetric change of PyC interphase itself. According to Kaae,\textsuperscript{22} low density isotropic PyC shrinks and has anisotropy in volumetric change after neutron irradiation up to 1 dpa. Therefore, further researches about swelling behaviors of SiC constituents (fiber, matrix) and PyC interphase after irradiation are required respectively in order to clarify the detailed mechanism of the degradation.

4.3 Neutron irradiation effects on UTS

It is well known that degradation of interphase affects composite maximum strength in unidirectional composites,\textsuperscript{23}

\[
\sigma_u = f \sigma_c \left( \frac{2}{m+2} \right)^{\frac{m+1}{m+2}}
\]

\[
\sigma_c = \left( \frac{\alpha_0^{m+1} L_0}{R} \right)
\]

where \(\sigma_u\) is the ultimate tensile strength, \(\sigma_c\) is the characteristic strength, \(\alpha_0\) is the Weibull mean strength, \(m\) is the shape parameter, and \(L_0\) is the gauge length, respectively. In this study, there was up to 30% increment in UTS after neutron irradiation regardless of the decrease of interfacial sliding stress as discussed in Section 4.2. However, low fluence neutron irradiation probably caused moderate degradation in interfacial properties of PyC layer, resulting in the fullest potential of fiber strength. Most cracks emerged in matrix penetrated into fibers through the strong-bonded F/M interface for the unirradiated samples. (The result of SEM observation showed that some regions of the fractured surfaces in unirradiated composites were flat with no pull-outs.) This indicates that the maximum strength of composites significantly depends on the performance of intact fibers. There have no degradation in \(\beta\)-SiC\textsuperscript{24,25} and Hi-Nicalon\textsuperscript{TM} Type-S fiber\textsuperscript{14} and primary role of interphase is to control the fracture behaviors (brittle or quasi-ductile). From these aspects, it can be concluded that the major achievements to provide good irradiation tolerances of UTS.

5. Conclusions

In order to identify neutron irradiation effects on interfacial properties of advanced SiC/SiC composites, unload/reload cyclic tensile test and hysteresis loop analysis were carried out. The advanced SiC/SiC composites exhibited good irradiation resistance in UTS (~30% increase), but a slight reduction of PLS was obtained. Wider hysteresis loops and lower gradient of the curves beyond PLS from the strain-stress curves, and longer fiber pullouts from the SEM observation were observed, which imply weaker F/M interactions. Additionally, from the hysteresis loop analysis, degradation of interfacial sliding stress was detected. Therefore it is considered that a slight reduction of PLS might be caused by the degradation of interfacial sliding stress.

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REFERENCES