3.1 Strength and Microstructures of the Fiber/Matrix Interface of Neutron-Irradiated SiC/SiC Composites — T. Nozawa and H. Tanigawa (Japan Atomic Energy Agency), P. Dou, H. Bei, Y. Katoh, L. L. Snead, and R. E. Stoller (Oak Ridge National Laboratory)

OBJECTIVE

This work was carried out under the collaboration on fusion materials between the U.S. DOE and the Japan Atomic Energy Agency (JAEA). This study primarily aims to identify the dose-dependent strength of the fiber/matrix (F/M) interface of SiC/SiC composites coupled with summarizing the irradiation-induced microstructural changes of SiC fiber and matrix, and pyrolytic carbon (PyC) as an F/M interphase. High-dose irradiation up to 70 dpa was specifically focused on. Identifying the micro- and macro-strength interaction is another objective of this study.

SUMMARY

To investigate the irradiation effects on interfacial shear properties such as an interfacial debond shear strength (IDSS) and an interfacial friction stress (IFS), the single fiber push-out tests were applied to Hi-Nicalon Type-S fiber reinforced chemical-vapor-infiltration (CVI) SiC matrix composites neutron irradiated up to 70 dpa at 300~800 ºC. Test results demonstrated no significant irradiation-enhanced degradation of interfacial shear properties expected at any doses (i.e., a slight decrease at lower doses <10 dpa and apparent improvement at higher doses >10 dpa). The irradiation-induced stress is a primary source to affect the F/M interfacial stresses. At higher doses ~70dpa, volume expansion by disordering of PyC possibly induced a residual clamping stress, eventually improving interfacial shear properties. A good correlation between the F/M interfacial strength and the macro strength for certain irradiation conditions was obtained. Interfacial functions sustained but fiber (Hi-Nicalon Type-S) deterioration was implied by irradiation at 70dpa. In the future we must clearly define the irradiation-induced change in thermal expansivity. The differential swelling between the fiber and matrix and irradiation creep are also critically important.

PROGRESS AND STATUS

Introduction

A silicon carbide composite (SiC/SiC composite) is a candidate for the advanced blanket system of a fusion DEMO reactor due to the excellent thermo-mechanical and physical/chemical properties. Since composite's functions are readily affected by the fiber/matrix (F/M) interfacial properties, the irradiation stability of the interface needs to be proven for the practical design of the DEMO components.

The authors have investigated the effects of neutron irradiation on the interfacial shear properties and test results by lower dose irradiation (<7.7 dpa) were well-summarized in [1]. Through the entire work, the F/M interfacial strength was evaluated by the single fiber push-out test technique with the modified shear-lag model, which gives the IDSS and the IDS with consideration of thermal stress, irradiation-induced stress, toughness-induced stress and Poisson's expansion. From the key findings of this work, both the IDSS and the IFS first decreased and slightly recovered with increasing irradiation up to 10 dpa. The probable explanation of such changes is a radiation-induced dimensional change of PyC, which shows a turn-around behavior of irradiation-induced swelling. Also it was found that the decrease of the interfacial friction stress at 7.7 dpa at 800 ºC

was significant. It is speculated that a decreasing surface roughness by the irradiation-enhanced change of the cracking path at the F/M interface from the inside PyC interphase to the fiber surface affects the result, although this is still uncertain because of the scarce of data of the irradiation-induced morphology change of the PyC interface. In contrast, at higher doses up to 40 dpa, there was no significant degradation of the bend strength, implying no potential degradation of the F/M interfacial strength. This is somehow surprising since it has been believed that carbon materials are generally instable by higher dose irradiation and needs to be confirmed by the individual interface tests. Of particular important is to identify a correlation between the nano-strength and the irradiation-induced microstructures.

EXPERIMENTAL PROCEDURE

Material

A satin-weave (S/W) CVI-SiC/SiC composite was tested. The reinforcement was highly-crystalline and near-stoichiometric Hi-Nicalon Type-S SiC fibers. The multilayered PyC/SiC interface (20 nm PyC x 5 with 100 nm SiC inserts) was formed (Fig. 1). The fiber volume fraction was approximately 35 %. The details of S/W composites are described in [2, 3], respectively.

Neutron Irradiation

Rectangular flexural specimens with a size of 50.2 x 6.35 x 2.63 mm were irradiated by the HFIR flux trap fixed rabbit tube facility at Oak Ridge National Laboratory. The irradiation doses and temperatures were ~70 dpa and 325~800 ºC, respectively.

Post Irradiation Experiments

The original rectangular flexural specimen was cut into the miniature bars with as size of 25 x 3 x 1.2 mm. The bulk strength was then obtained by the 4-point bend test with loading/support spans of 10/20 mm. After the bend tests, the interfacial shear strengths were evaluated by the single fiber push-out test using a nano-indenter. By applying the modified double shear-lag (Hsueh) model, both the IDSS and the IFS are finally obtained. Note that the thermal and irradiation creep were not presently considered in the model due to the lack of data. The detailed analytical procedure was described in [1]. The microstructures of neutron irradiated composites were observed by the scanning electron microscopy (SEM) and the transmission electron microscopy (TEM).

RESULTS

Irradiation Effects on Interfacial Shear Properties

Figure 1 shows the effects of neutron irradiation on the interfacial shear properties. Both the IDSS and the IFS first decreased and increased with increasing neutron dose. Of particular emphasis is no serious degradation even in carbon instability region. Instead, high-dose irradiation further improved interfacial shear properties. The temperature dependence is unclear but seemingly high-temperature irradiation tends to impact more on the interfacial shear properties.

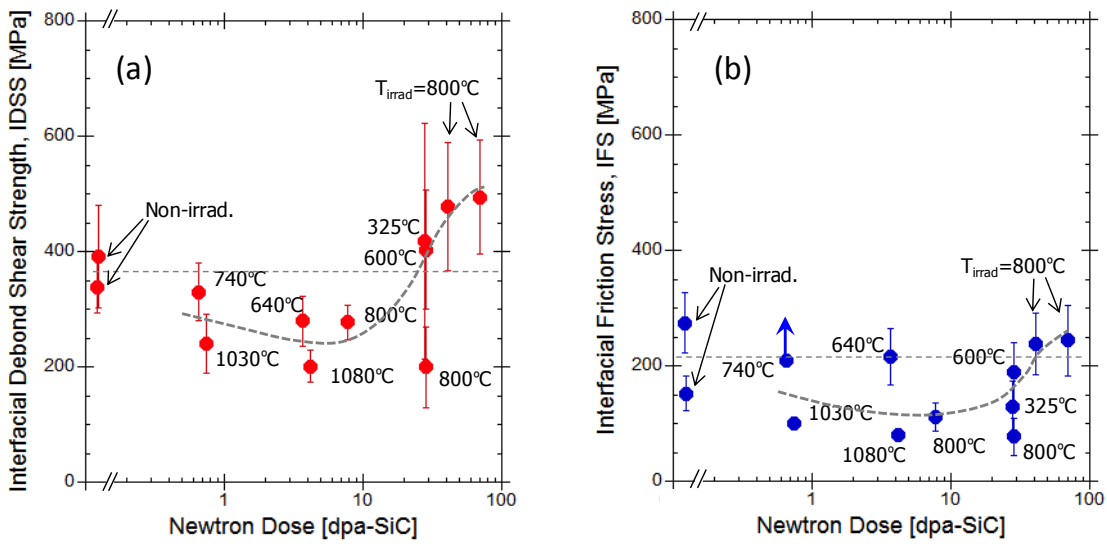

Fig. 1. Neutron irradiation effects on (a) the interfacial debond shear strength (IDSS) and (b) the interfacial friction stress (IFS) of Hi-Nicalon Type-S/multilayer/CVI-SiC composites.

The interfacial shear properties generally depend on (1) the residual strain and (2) the fiber surface roughness. Accordingly to the model calculation [1], comparably minor effect of the coefficient of thermal expansion (CTE) change of PyC itself is expected. There is in contrast a significant impact of the CTE changes of the fiber or the matrix even if there is a slight gap of CTEs between the fiber and the matrix. Radiation-induced CTE modification of the Hi-Nicalon Type-S SiC fiber and the CVI-SiC matrix is uncertain and needs to be identified in further experiments.

Irradiation-induced residual strain also influences the radial clamping stress at the F/M interface. Because of the significant swelling of PyC, i.e., “turn-around” behavior, and this undoubtedly affects the irradiation behavior of the interfacial shear properties. But this is unclear at higher doses because of instability of PyC itself. In contrast, a mild change of axial stresses between the fiber and the matrix is expected assuming the same swelling quantity of them. However, differential swelling between the fiber and the matrix may induce further stresses. If the fiber swells more than the matrix, the residual clamping stress tends to increase more, potentially improving the interfacial shear properties. This issue also needs to be clarified in future.

Figure 2 shows the typical microstructures of the F/M interface of Hi-Nicalon Type-S/multilayer/CVI-SiC composites by irradiation at 70 dpa at 800 ºC. It is clearly shown that parts of PyC were disordered by irradiation. From this fact, the most probable explanation for the significant improvement of the interfacial shear properties by irradiation up to ~70 dpa is that disordered PyC due to instability by the high-dose irradiation gives a significant residual compressive stress at the F/M interface by its volumetric expansion. Because of instability of PyC itself, there was a fact that a crack can easily propagated within PyC. In contrast, it is very hard to identify the admissible degradation of the Hi-Nicalon Type-S SiC fiber and the SiC matrix. This needs to be further evaluated carefully.

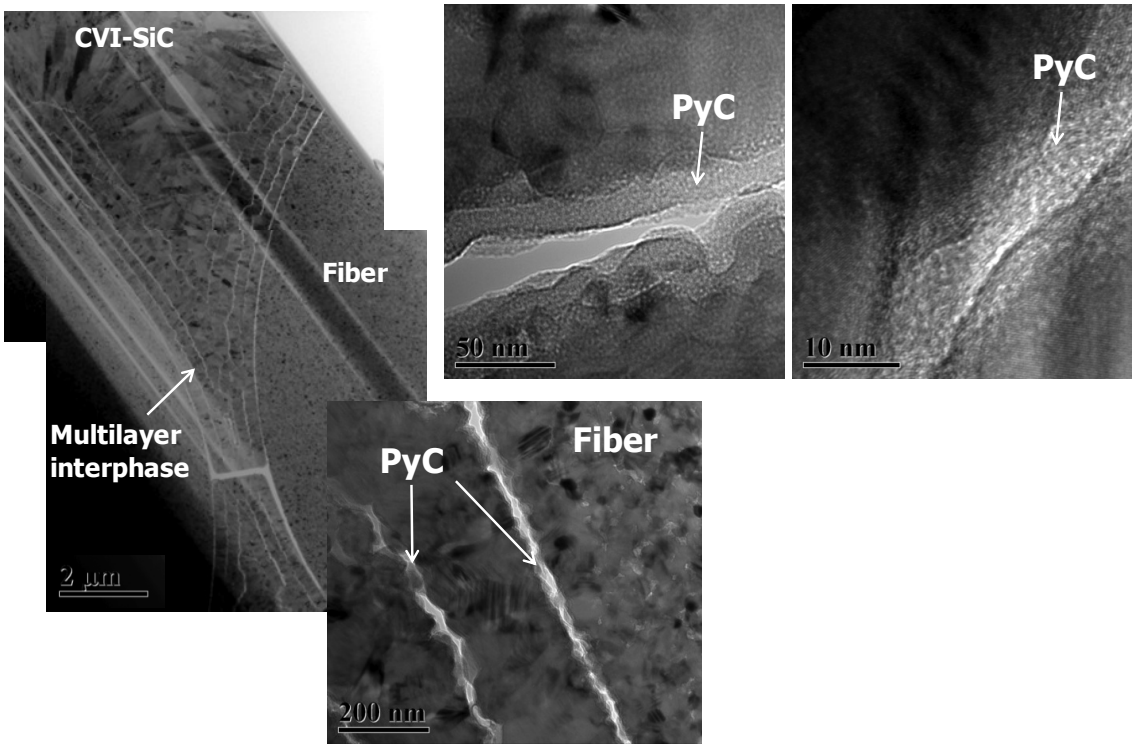

Fig. 2. Typical microstructures of the F/M interface of Hi-Nicalon Type-S/multilayer/CVI-SiC composites.

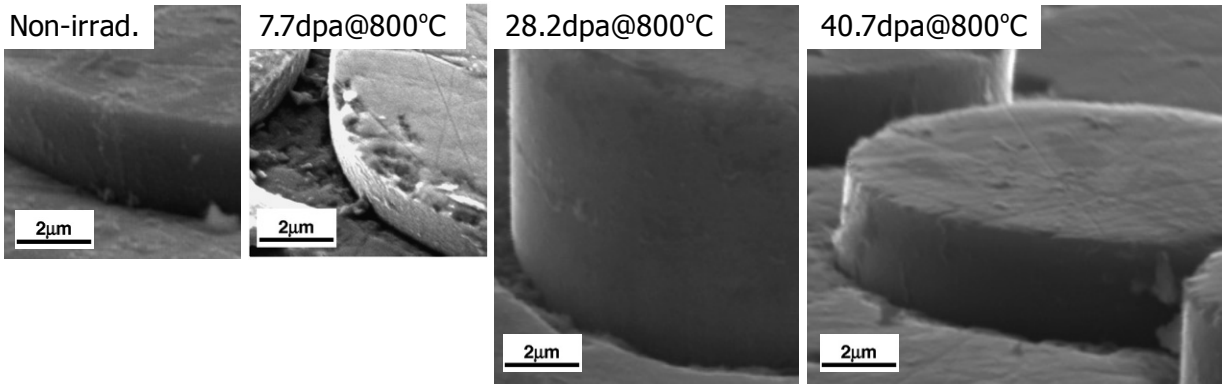

Fig. 3. Typical surface appearances of pushed-out Hi-Nicalon Type-S fibers.

Figure 3 shows the typical surface appearances of pushed-out fibers. There seems no clear change in the surface roughness. It was therefore concluded that contribution of the roughness-induced clamping stress was very minor.

Correlation between Bulk Strength and Interfacial Shear Properties

Figure 4(a) shows neutron dose dependencies of the proportional limit tensile/flexural stress (PLS) and the IDSS, both of which are normalized by the non-irradiated test result. The PLS slightly decreased by irradiation but did not depend on the fluence so much, while, as aforementioned, the IDSS first decreased a little bit and remarkably improved with increasing neutron fluence. Katoh et al [4] claimed that the PLS depends on the true matrix cracking stress and the misfit stress (i.e., irradiation-induced stress). By separately discussing these two parameters, the test results supported no significant change of the true matrix cracking stress by irradiation reported. The post irradiation strength therefore depends primarily on the misfit stress. Since the IDSS is also dependent on the residual stress, the post irradiation PLS therefore depends on IDSS too. This is true at least by irradiation up to 10 dpa. But, from this point of discussion, the PLS at higher doses was not recovered more than expected from the result of the IDSS; the IDSS shows remarkable improvement by irradiation up to >40 dpa (Fig. 4(b)).

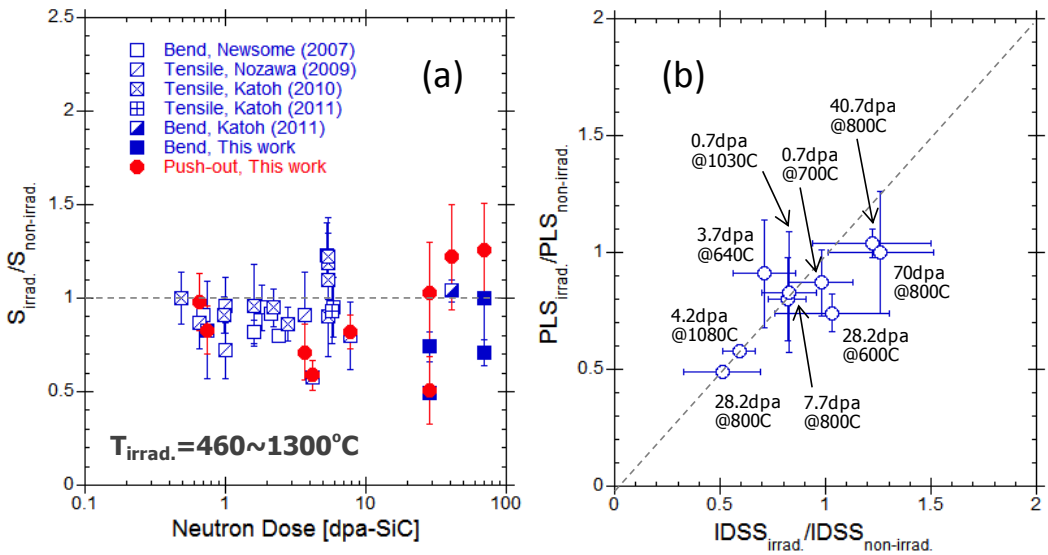

Fig. 4 – (a) Neutron dose dependencies of the proportional limit tensile/flexural stress (PLS) and the interfacial debond shear strength (IDSS) and (b) correlation between the PLS and the IDSS.

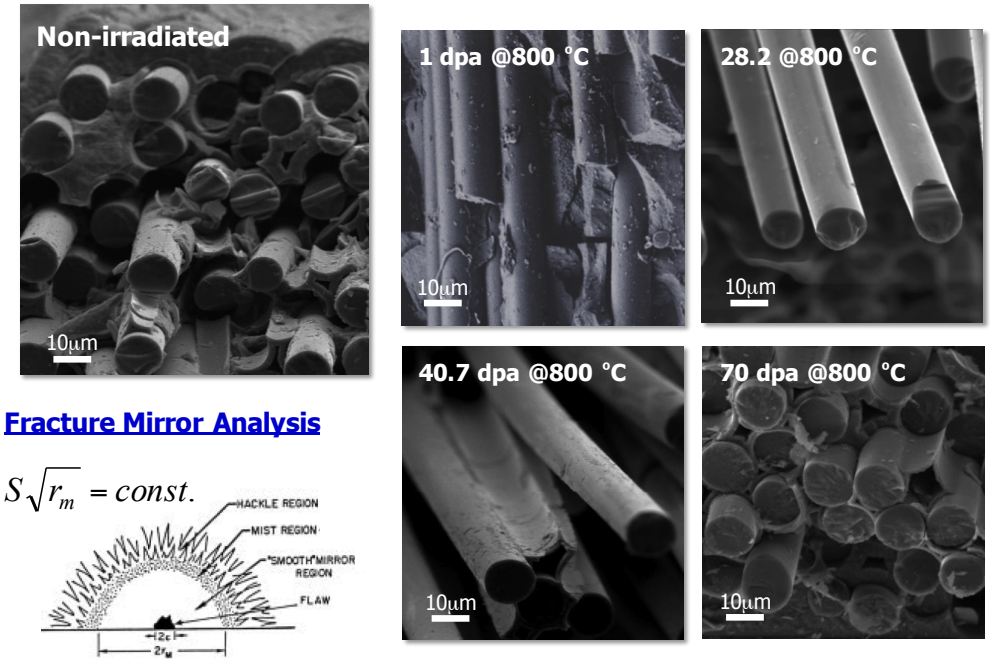

Fig. 5. Typical flexural fracture surface images of Hi-Nicalon Type-S/multilayer/CVI-SiC composites before and after irradiation.

Figure 5 shows typical fracture surface images of Hi-Nicalon Type-S composites before and after irradiation. In each test condition, composites show moderate fiber pullouts, although the fiber pullout length is short due to the strong interfacial friction stress. For the non-irradiated case and some lower dose irradiation cases, the cross section of the fractured fiber is like a broken surface of amorphous based fibers, while, as clearly shown for the 70 dpa irradiation case, the fracture mirror was appeared by higher-dose irradiation. Theoretically, the individual fiber strength depends on the mirror radius as explained in embedded drawing in Fig. 5. In general, the fiber strength decreased with increasing the mirror radius. With the fact that the fracture mirror first appeared by higher dose irradiation (~70 dpa), the possible fiber degradation would be a key reason for the reduction of the bulk composite strength. Detailed microstructural observation will be planned to confirm this.

REFERENCES

[1] T. Nozawa, Y. Katoh, and L. L. Snead, *J. Nucl. Mater*. **384** (2009) 195.
[2] Y. Katoh, T. Nozawa, L. L. Snead, K. Ozawa, and H. Tanigawa, *J. Nucl. Mater.* **417** (2011) 400.
[3] G. Newsome, L. L. Snead, T. Hinoki, Y. Katoh, and D. Peters, J. *Nucl. Mater.* **371** (2007) 76.
[4] Y. Katoh, K. Ozawa, T. Hinoki, Y. Choi, L. L. Snead, and A. Hasegawa, *J. Nucl. Mater*. **417** (2011) 416.