Neutron Tolerance of Advanced SiC-Fiber / CVI-SiC Composites

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Abstract: Fusion blankets employing a silicon carbide (SiC) fiber-reinforced SiC matrix composite (SiC/SiC composite) as the structural material provide attractive features represented by high cycle efficiency and extremely low induced radioactivity. Recent advancement in processing and utilization techniques and application studies in ceramic gas turbine and advanced transportation systems, SiC/SiC composites are steadily getting matured as industrial materials. Reference SiC/SiC composites for fusion structural applications have been produced by a forced-flow chemical vapor infiltration (FCVI) method using conventional and advanced near-stoichiometric SiC fibers and extensively evaluated primarily in Japan-US collaborative JUPITER program. In this work, effect of neutron irradiation at elevated temperatures on mechanical property of these composites is characterized. Unlike in conventional SiC/SiC composites, practically no property degradation was identified in advanced composites with a thin carbon interphase by a neutron fluence level of approximately 8dpa at 800C.

1. Introduction

Fusion power reactors with silicon carbide (SiC) fiber-reinforced SiC matrix composite (SiC/SiC composite) branket structures provide attractive features represented by very high heat cycle efficiency and extremely low induced radioactivity. Utilization of SiC/SiC composites has so far been regarded as a high risk option in blanket design studies, because of the inexperience as industrial materials, unconfirmed neutron irradiation response and lack of proven joining and hermetic sealing techniques [1]. However, recent advancement in processing and utilization techniques and application studies in ceramic gas turbines, ceramic heat exchangers and advanced transportation systems, SiC/SiC composites are steadily getting matured as industrial materials [2]. Therefore, the effects of neutron irradiation on mechanical and physical integrity are among the most important technical issues for SiC/SiC composites for nuclear applications.

SiC/SiC composites comprize three distinctive constituents, namely, fibers, matrices and interphases. Although both the fibers and the matrices are SiC or SiC-based materials by definition, their microstructures and chemical and phase compositions very mach vary depending on the raw materials and the processing technique. The interphase is a thin and compliant layer which provides a relatively weak bond between the fiber and the matrix for enabling pseudo-ductile fracture in the inherently brittle ceramic. Recent trend for improved radiation stability is to make the most use of stoichiometry and crystalline SiC, or minimize the amount of amorphous(-like) SiC, carbon, silica and boron nitride, all of which are less stable against neutrons. Among several techniques for the SiC/SiC composite processing, chemical vapor infiltration (CVI) is the most appropriate method for production of reference materials for irradiation studies, since it deposits stoichiometry crystalline cubic (beta) SiC as the matrices. Therefore, in the Japan-US collaborative JUPITER program for fusion materials research [3], SiC/SiC composites with different advanced SiC fibers and interphases had been produced by a CVI method and neutron-irradiated. In this paper, we report the influence of neutron irradiation at elevated temperatures on mechanical property of the these reference advanced SiC fiber composites with pyrolytic carbon (PyC) interphases.

TABLE 1: REPRESNTATIVE PROPERTIES AND CHEMICAL COMPOSITIONS OF SiC (-BASED) FIBERS USED FOR COMPOSITE PRODUCTION.

	Hi-Nicalon™	Hi-Nicalon™ Type-S	Tyranno™-SA
C/SiC atomic ratio	1.39	1.05	1.07
Oxygen content (wt%)	0.5	0.2	<0.5
Microstrcuture	Nano-crystalline SiC and amorphous C	Polycrystalline	Polycrystalline
Tensile strength (GPa)	2.8	2.6	2.6
Tensile modulus (GPa)	270	420	400
Elongation (%)	1.0	0.6	0.7
Mass density (g/cm^3)	2.74	3.1	3.0
Average diameter (um)	14	11	7

2. Experimental Procedure

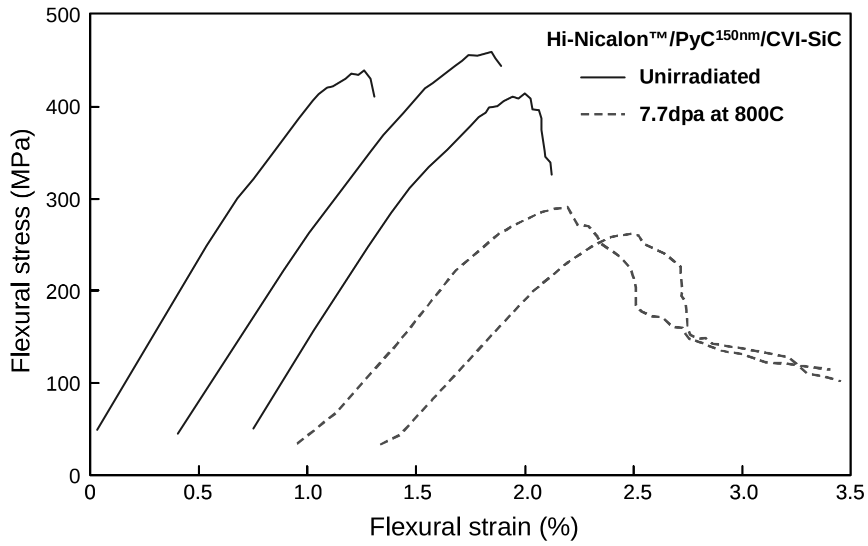
The materials were produced at Oak Ridge National Laboratory (ORNL) through forced-flow temperature gradient CVI method using Hi-Nicalon™ Type-S (Nippon Carbon Co., Tokyo, Japan) and Tyranno™-SA (Ube Industries Ltd., Ube, Japan) near-stoichiometry SiC fibers as well as conventional Hi-Nicalon™ SiC fibers for comparative study [4]. Properties and chemical compositions reported by the manufacturers are compiled in Table 1. The conventional Hi-Nicalon™ fiber contains significant amount of excess carbon and does not have highly crystalline structure. PyC coating, typically 150nm-thick, had been applied to the fiber fabric, which are plain-woven or satin-woven, prior to the matrix densification in order to produce the interphase. Typical ultimate tensile strength, proportional limit tensile stress and tensile modulus of the unirradiated composites are 250MPa, 50-100MPa and 250GPa, respectively.

Neutron irradiation was performed in a RB-13J and 14J capsules of High Flux Isotope Reactor (HFIR) at ORNL to a maximum fluence level of about 9 dpa ($9x10^{25}$ n/m^2, E>0.1MeV) at 300-800C. The composites were square-cut into miniature bend-bars for flexural tests. Three-point and four-point flexural tests were carried out at ambient temperature prior to and after the irradiations. The crosshead displacement rate was 0.5 mm/min. Fracture surfaces were observed by scanning electron microscope (SEM) following the flexural tests.

3. Results and Discussion

Stress-strain relationships during the flexural tests on the unirradiated and irradiated SiC/SiC composites are presented in Fig.1. For the case of composites reinforced with conventional Hi-Nicalon™ fibers, severe degradation in ultimate stress is obvious. It is reasonable to attribute this observation to an irradiation-induced shrinkage of non-stoichiometry SiC-based fibers, since the flexural modulus is also largely reduced and long tails in stress-strain curves present, suggesting an initial debond and a very small friction at the fiber-to-matrix interface.

On the other hand, the composite with advanced Hi-Nicalon™ Type-S fibers did not apparently show any sign of mechanical property degradation. This also supports the above implication of conventional composite's degradation mechanism, because a potential



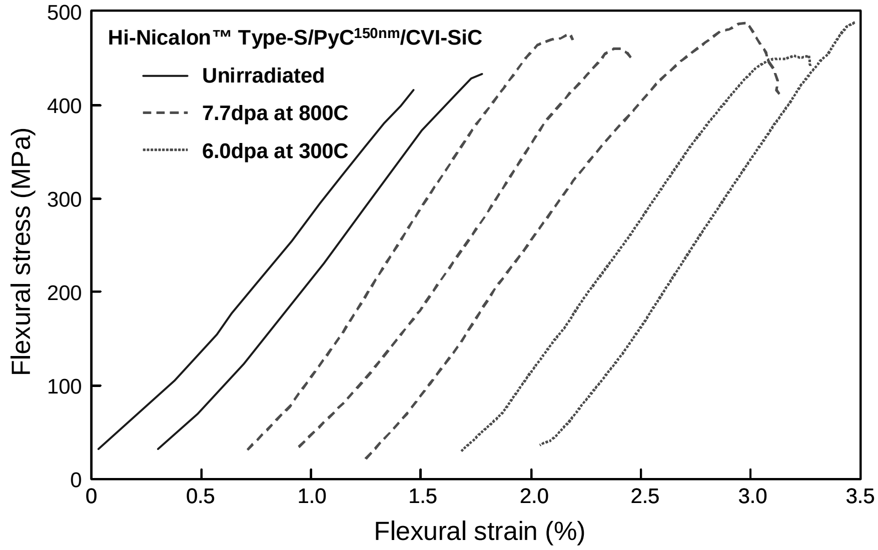


Fig.1. Stress-strain relationship during tensile tests on unirradiated and irradiated Hi-Nicalon™ / CVI-SiC (upper) and Hi-Nicalon™ Type-S / CVI-SiC (lower) composites with PyC interphases.

irradiation-induced degradation in the PyC interphase appears not to cause macroscopic flexural property degradation for the case of advanced composite. The observation in Fig.1 suggests minor increase in tensile modulus and ultimate strength, instead of decrease, in the irradiated Hi-Nicalon™ Type-S composite, although it may not be statistically significant [5]. Potential microstructural modifications at around the interphase and its consequence on other properties and at higher neutron fluence levels remain unrevealed.

The composites reinforced with Tyranno™-SA advanced near-stoichiometry SiC fibers did not suffer from irradiation-induced strength degradation, either. Therefore, we can conclude that CVI-SiC-matrix composites with crystalline and near-stoichiometry SiC fiber reinforcement and 150nm-thick PyC interphases do not degrade macroscopic mechanical properties by neutron irradiation up to about 10 dpa in a temperature range of 300-800C.

Figure 2 compiles the results of neutron irradiation effect on ultimate strength of the SiC/SiC composites. Data points labeled 2, 3 and 4 are from the present work. Irradiated-to-unirradiated ratios of ultimate flexural strength are plotted in order to enable comparison with

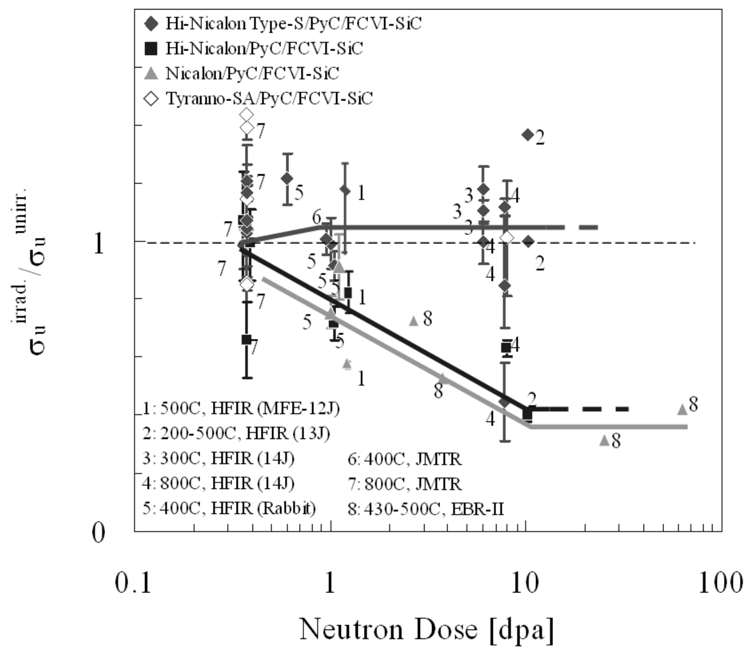


Fig.2. Effect of neutron irradiation at elevated temperatures on normalized ultimate flexural strength (ratio of irradiated strength to unirradiated one) of advanced and conventional reference F-CVI SiC/SiC composites. (The data compilation includes data from literatures that are not listed in this paper.)

data on conventional materials. As clearly depicted in Fig.2, unlike the conventional SiC/SiC composites which drastically degrade mechanical property by 10dpa, advanced SiC-fiber composites exhibit a superior stability in ultimate strength after potential minor increase by approximately 1dpa. The lack of further strength degradation beyond ~10dpa in conventional composites indicates irradiation stability of silicon carbide once saturation in chemical and grain structure evolution is achieved.

Pseudo-ductility of the composites was also maintained after irradiation, as an example of fracture surfaces with an appropriate fiber pull-out is presented in Fig.3. However, among the advanced SiC fiber composites, only Hi-Nicalon™ Type-S composite with a thick (500nm) PyC interphase, underwent detectable strength degradation by about 8dpa at 800C. Therefore, irradiation damage and chemical instability within PyC interphases in a fusion environment will be an important issue for future development of SiC/SiC composites for blanket applications. Effects of irradiation to higher fluence levels on microstructures advanced SiC fiber composites with PyC interphase as well as other advanced interphases need to be explored by both neutron and ion irradiations.

4. Conclusions

Reference SiC/SiC composites for neutron irradiation study were produced by CVI method employing conventional and advanced SiC(-based) fibers and PyC interphases.

The composites with crystalline and near-stoichiometry SiC fiber reinforcement and 150nm-thick PyC interphases did not show any sign of macroscopic mechanical property degradation by neutron irradiation up to about 10 dpa in a temperature range of 300-800C.

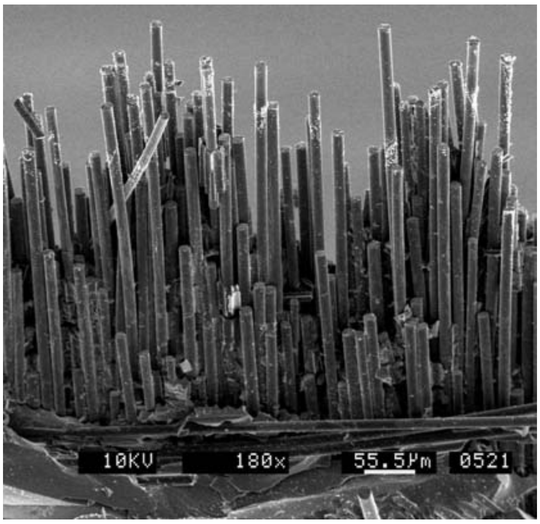


Fig.3. Fracture surface of Hi-Nicalon™ Type-S/PyC/FCVI-SiC composites following neutron irradiation to approximately 8dpa at 800C.

The effect of neutron irradiation to higher fluence levels, especially as the consequences of microstructural modification at interphases, remains as a very important technical issue.

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