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Original article

Mechanical property degradation of high crystalline SiC fiber–reinforced SiC matrix composite neutron irradiated to ~100 displacements per atom☆

Takaaki Koyanagi[a,*], Takashi Nozawa[b], Yutai Katoh[a], Lance L. Snead[c]

[a] Oak Ridge National Laboratory, 1 Bethel Valley Road, Oak Ridge, TN 37831, USA
[b] National Institutes for Quantum and Radiological Science and Technology, 2-166 Omotedate, Obuchi, Rokkasho, Aomori 039-3212, Japan
[c] Stony Brook University, 100 Nicolls Rd, Stony Brook, NY 11794, USA

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ABSTRACT

For the development of silicon carbide (SiC) materials for next-generation nuclear structural applications, degradation of material properties under intense neutron irradiation is a critical feasibility issue. This study evaluated the mechanical properties and microstructure of a chemical vapor infiltrated SiC matrix composite, reinforced with a multi-layer SiC/pyrolytic carbon–coated Hi-Nicalon^{TM} Type S SiC fiber, following neutron irradiation at 319 and 629 °C to ~100 displacements per atom. Both the proportional limit stress and ultimate flexural strength were significantly degraded as a result of irradiation at both temperatures. After irradiation at 319 °C, the quasi-ductile fracture behavior of the nonirradiated composite became brittle, a result that was explained by a loss of functionality of the fiber/matrix interface associated with the disappearance of the interphase due to irradiation. The specimens irradiated at 629 °C showed increased apparent failure strain because the fiber/matrix interphase was weakened by irradiation-induced partial debonding.

1. Introduction

Continuous silicon carbide (SiC) fiber–reinforced SiC matrix (SiC/SiC) composites are leading candidates for use as structural materials for advanced nuclear reactor components such as fusion blanket components [1–3], fuel cladding and channel boxes for light water reactors [4–6], and fuel cladding and control rods for gas-cooled fast reactors [7,8]. They are of interest because of their inherent high dissociation temperatures, chemical stability [9], relatively low neutron absorption [10], high toughness [11], relatively mature fabrication technology [11,12], and so on. To enable the successful development of SiC/SiC composites for advanced reactors, the effects of high-dose fusion or fission neutron irradiation on their mechanical and physical properties are critical phenomena that must be considered. The neutron doses for SiC/SiC components are expected to reach beyond 100 displacements per atoms (dpa), depending on the reactor design [13]. For example, SiC/SiC composite fuel cladding and core structures for gas-cooled fast reactors and SiC/SiC composite fusion reactor components are expected to receive intense neutron irradiation to ~100 dpa during their service lives [7]. Although the lifetime neutron dose SiC/SiC composites will receive is not specified for each application, it is possible that the material degradation associated with high-dose irradiation will be a limiting factor for the use of SiC/SiC composite materials in radiation environments.

Very high-dose neutron irradiation of a β SiC monolith up to 1.9×10^{27} n/m^2 (E > 0.1 MeV) at 370–650 °C was achieved in fast breeder reactors by Yano et al. [14–16]. They investigated irradiation-induced dislocation loops by transmission electron microscopy (TEM) and x-ray diffraction. They found growth of the dislocation loops with increasing neutron dose, although the growth was moderate; the average loop diameter was ~15 nm for the specimen irradiated to 1.0×10^{27} n/m^2 (E > 0.1 MeV). Moreover, a series of high-dose neutron irradiations was conducted at 300–800 °C on a specific type of SiC/SiC composite: a chemical vapor infiltrated (CVI) SiC/SiC composite with Hi-Nicalon^{TM} Type S (HNS) SiC fibers and a multi-layer pyrolytic carbon (PyC)/SiC interphase [17–20]. The important findings from these studies are (1) saturation swelling of the SiC/SiC composite up to ~70 dpa, (2) a saturation of degradation in the thermal diffusivity from a few dpa to ~70 dpa, and (3) irradiation-temperature–dependent degradation of flexural properties (resistant at higher radiation

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* Corresponding author at: Oak Ridge National Laboratory, 1 Bethel Valley Road, Oak Ridge, TN 37831-6140, USA.
E-mail address: koyanagit@ornl.gov (T. Koyanagi).

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temperatures) beyond ~40 dpa. These results encouraged the use of SiC/SiC composites at higher irradiation temperatures and, at the same time, motivate further development of irradiation-resistant SiC/SiC composite materials for applications at relatively low irradiation temperatures. In this study, the series used in the high-dose irradiation study was extended to higher neutron doses of up to 105 dpa. The main objective of this study is to use microscopy analysis to understand the mechanism of degradation of the mechanical properties of irradiated SiC/SiC composites, which was not clearly identified in previous work [19,20].

2. Experimental

The SiC/SiC material investigated was HNS fiber (lot number 2D2685-02-I5-01, year 2000) –reinforced CVI SiC/SiC composites fabricated by Hypertherm THC Inc. (now a subsidiary of Rolls Royce Inc.) The fiber architecture was a two-dimensional five-harness satin weave with a 0/90° stacking configuration. The fiber/matrix interphase was a five-layer ring-stack of ~10 nm thick PyC and ~100 nm thick SiC. This is a fiber/matrix interface design with a minimized thickness of the PyC phase [21]. The fiber volume fraction was approximately 35%. The mean density of the composite was 2.52 g/cm^3. The HNS fiber contained a carbon impurity phase, and the C/Si atomic ratio was 1.05 based on the material data sheet from the manufacturer. Also investigated was a monolithic chemical vapor deposited (CVD) SiC (CVD Silicon Carbide^{TM} high-resistivity grade, Rohm and Haas Co.). The material purity was 99.9995%, as guaranteed by the vendor. The composite and monolithic materials were machined into flexural bars with dimensions of 50.8 (long) × 6.3 (wide) × 2.8 (thick) mm. For the composite, one of the fiber reinforcement directions was aligned to the specimen length direction. The direction of the fiber fabric stacks corresponded to the specimen thickness direction. More details of the specimen fabrication and preparation can be found elsewhere [17].

Neutron irradiation was conducted in the flux trap facility of the High Flux Isotope Reactor at Oak Ridge National Laboratory (ORNL). The nominal irradiation temperatures were ~300 and ~500 °C. The neutron dose was up to 105 dpa with an equibalance of 1 dpa = 1×10^{25} n/m^2 (E > 0.1 MeV) [17]. The total duration of the irradiation was 1000–1150 days. The irradiation conditions for the specimens are summarized in Table 1. Part of the specimen surfaces was contaminated during irradiation because of contact between the specimens and the metallic components of the irradiation vehicle. However, no significant reaction layer was found by observation, and the effect was minimal in a previous study using same irradiation capsule design [20]. The actual irradiation temperatures were determined by the recovery behavior of the instantaneous coefficient of thermal expansion of a SiC passive thermometer made of CVD SiC, during annealing using a NETZSCH DIL 402CD horizontal dual pushrod dilatometer. The annealing was conducted at a heating rate of 1 °C/min. and a cooling rate of 2.5 °C/min. Details of the method of determining irradiation temperature were reported in [22]. The error in temperature determination by data analysis was typically 10–15 °C [22]. Note that the specimens were designed to be irradiated in contact with the thermometer. The SiC passive temperature monitor provided the temperature at the end of irradiation. Because this study used the same irradiation vehicle design used in previous work, and the actual irradiation temperature of the previous specimens was close to the nominal temperature for doses of ~40 and ~70 dpa [20], the desired irradiation temperature was expected to be achieved if the temperature of the thermometer was reasonably close to the nominal temperature.

The dimensions of each specimen were obtained using a micrometer, which measured the length swelling at an accuracy of 0.05%. Swelling along the specimen width and thickness directions was not reported because of a significant deviation in the data. Mechanical properties of both the irradiated and the nonirradiated specimens were evaluated by four-point flexural testing at room temperature, according to ASTM C1341. The support span and the loading span were 40 and 20 mm, respectively. The proportional limit stress (PLS) was determined as a 5% deviation in stress from the initial linearity. The dynamic Young's moduli of the SiC/SiC composites were determined using the impulse excitation of vibration method in accordance with ASTM standard C1259, using an Integrated Material Control Engineering resonant frequency and damping analyzer. The fracture surfaces of the composites were characterized using a Hitachi 4800S scanning electron microscope (SEM) for nonirradiated specimens and an FEI Quanta Dual Beam SEM/focused ion beam (FIB) for irradiated specimens.

As-irradiated specimens were also observed using a FEI Versa Dual Beam SEM/FIB. The TEM specimens were prepared using the FEI Versa Dual Beam SEM/FIB operated at 30 kV for rough milling and 2 and 5 kV for final thinning, followed by low-energy ion milling using a Fischione Model 1040 NanoMill operated at 600 and 900 eV. TEM observation was also conducted using a JEOL JEM2100F with TEM and scanning TEM (STEM) modes operated at 200 kV. STEM–electron energy loss spectroscopy (EELS) evaluation was conducted using a Gatan GIF Quantum post-column energy filter and Gatan Digital Micrograph software. The spectrometer correction angle was 4.2 mrad. Background removal of the EELS spectra was conducted using a Power Law fit model for carbon K-edge spectra. The TEM specimen thickness was determined by an EELS log-ratio method.

3. Results

3.1. Mechanical properties and swelling

Flexural tests of nonirradiated specimens were conducted in the previous study [17]. This study used the same grade of material for irradiation and the same support/load span length for the flexural test. Fig. 1 shows the flexural behavior of nonirradiated and irradiated CVI SiC/SiC composites. The PLS and ultimate flexural strength (UFS) determined are listed in Table 1. The nonirradiated material shows typical fracture behavior for a tough composite, i.e., nonlinear fracture behavior beyond PLS. On the other hand, the specimen irradiated at 319 °C to 92 dpa failed in a brittle manner; i.e., PLS was equal to UFS, and both PLS and UFS were significantly degraded. The specimen irradiated at 629 °C to 99 dpa showed nonlinear fracture behavior with apparent increased failure strain. Complete failure could not be achieved because of contact between the fixture and the specimen under flexural strain

Table 1
Summary of irradiation conditions, dynamic Young's modulus, flexural properties, and swelling. Actual irradiation temperatures were determined using passive SiC thermometry. Parentheses indicate one standard deviation. Thirty nonirradiated specimens were tested in previous work [17].

Material	Design irradiation temperature [°C]	Actual irradiation temperature [°C]	Dose [dpa]	Specimen ID	Dynamic Young's modulus [GPa]	Transient modulus of elasticity [GPa]	PLS [MPa]	UFS [MPa]	Length swelling [%]
CVI SiC/SiC	Not applicable		0	Not available	228 (21)	179 (5)	395 (55)	453 (46)	0
CVI SiC/SiC	300	319	92	S06 S94	186 208	135 164	143 124	256 273	0.48 0.56
CVI SiC/SiC	500	629	99	S104 S110	187 219	142 168	225 206	256 273	0.38 0.35
CVD SiC	300	257	73	B42 B80	388 379	Not available			0.82 0.79
CVD SiC	500	534	105	B29 B38	Not available				0.64 0.63