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Journal of Nuclear Materials 289 (2001) 10–15

journal of nuclear materials

www.elsevier.nl/locate/jnucmat

Failure mechanisms in continuous-fiber ceramic composites in fusion energy environments

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Abstract

Silicon carbide composites are attractive for structural applications in fusion energy systems because of their low activation and afterheat properties, excellent high-temperature properties, corrosion resistance, and low density. These composites are relatively new materials with a limited database; however, there is sufficient understanding of their performance to identify key issues in their application. To date, dimensional changes of the constituents, microstructural evolution, radiation-enhanced creep, and slow crack growth have been identified as potential lifetime limiting mechanisms. Experimental evidence of these mechanisms, the factors that control them, and their implications on component lifetime will be discussed. © 2001 Published by Elsevier Science B.V.

1. Introduction

Composites consisting of silicon carbide reinforced by small diameter (≈10–20 μm) silicon carbide fibers have many advantageous properties for use in fusion energy systems [1–4]. Although it is difficult to obtain experimental data for the effects of irradiation on the mechanical properties of these composites, several potential effects can be identified by analogy to the results from studies performed on materials outside of radiation environments and from the limited studies performed in radiation environments. These studies suggest that there are a number of important processes that may also determine the lifetime and behavior of these composites under anticipated fusion energy system conditions [5–18]. One such process is the occurrence of radiation-enhanced creep of the constituents. Another important process is the accumulation of stresses due to radiation-induced dimensional change of the composite constituents. Features and implications of these processes will be described.

Several mechanisms leading to time-dependent failure of silicon carbide fiber-reinforced silicon carbide matrix composites (SiC_f/SiC_m) have been observed in conditions outside of radiation environments [5–11]. Although tensile fracture and creep are well-known failure processes, a number of mechanisms lead to subcritical crack growth in SiC_f/SiC_m [5–7,11]. These mechanisms lead to a reduction in the crack-bridging tractions provided by the reinforcing fibers and a subsequent increase in stress intensity at the tip of matrix cracks. Since matrix cracks can exist due to overstresses, machining of notches or holes, impact, thermal shock etc., subcritical crack growth is expected to be a lifetime limiting mechanism at stresses and temperatures below those leading to creep or tensile failure.

In conditions outside of radiation environments, three primary mechanisms of subcritical crack growth have been observed in SiC_f/SiC_m composites [5–7]. The mechanism that controls the rate of subcritical crack growth is dependent on the environmental conditions. In particular, the oxygen concentration and temperature have been found to define the range of conditions under which these mechanisms operate in laboratory experiments.

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0022-3115/01/$ - see front matter © 2001 Published by Elsevier Science B.V.
PII: S0022-3115(00)00676-0

In inert environments, the temperature dependency measured for subcritical crack growth [6] was similar to that reported for fiber creep [19–22]. In addition, the temperature increase required to obtain the same rate of subcritical crack growth in composites containing creep-resistant fibers as in composites containing less creep-resistant fibers was the same as that required to obtain the same amount of stress relaxation, presumed to be due to creep, in fiber 'bend stress relaxation' experiments [19]. Henager et al. [23] have made a micromechanical model of a crack bridged by discrete, time-dependent forces (representing creeping fibers) that agrees well with experimental observations of crack growth. These observations indicated that fiber creep controls the rate of subcritical crack growth in environments lacking oxygen. This mechanism of subcritical crack growth is referred to as the fiber relaxation mechanism (FRM).

Although an interphase is required to allow fibers to debond during crack propagation and to avoid fiber fracture, removal of the interphase by oxidation also reduces the tractions supplied by the fibers and leads to crack growth. Oxidation of the interphase produces volatile species that diffuse throughout the composite. Studies have shown that fibers surrounded by partially oxidized interphases are more compliant than those with unoxidized interphases [24]. In addition, the temperature dependency measured for crack growth in oxygen [6] was found to be the same as that measured for carbon oxidation [25]. These observations suggest that interphase removal controls the rate of subcritical crack growth in certain regimes of temperature and oxygen concentration (≈500–1000°C, <70 000 ppm O_2). This mechanism of subcritical crack growth is referred to as the interphase removal mechanism (IRM).

A third mechanism of subcritical crack growth causes rapid failure of SiC_f/SiC_m composites [5–10]. In environments containing high concentrations of oxygen, at temperatures above ≈500°C, the formation of a brittle oxidation reaction product on the fiber surface, or bonding of the fiber to the matrix, leads to rapid fiber failure. The failure of fibers that bridge matrix cracks removes the tractions supplied by those fibers and increases the stress intensity at the crack tip. This mechanism of crack growth is referred to as the oxidation embrittlement mechanism (OEM).

A fourth mechanism of subcritical crack growth has been proposed [26], but rarely observed. This mechanism is proposed to occur at temperatures where the oxidation reaction product flows viscously. The result of this viscous flow is to allow stress relaxation to occur, similar to fiber creep. This mechanism is referred to as the viscous interphase mechanism (VIM).

2. Methods

A variety of techniques were used to obtain the experimental and analytical results presented here. Subcritical crack growth experiments were performed on single edge notched beam specimens, loaded in flexure under controlled conditions of temperature and oxygen concentration, to characterize subcritical crack growth behavior [7,11]. Flexural strength measurements were also performed to investigate the fracture behavior of irradiated composite specimens [15]. Fiber push-in testing was performed to characterize the compliance of fibers with partially oxidized interphases [24]. The dimensional variation of fibers, prior and subsequent to irradiation exposure, was measured using precise length measurements and liquid immersion techniques [14].

Torsional creep experiments were performed on individual fibers during irradiation by a light ion beam. The fibers were coated with nickel, outside of the 8-mm gauge length, to allow gripping. Torque was applied to the grip by a galvanometer-like electromagnetic system. The magnitude of the torque was directly proportional to an electric current. The angular displacement was measured by an optically coupled system. The fiber was heated directly by the application of an electric current. The fiber temperature was measured by a two-color pyrometer. Prior to turning on the ion particle beam, the specimens were equilibrated, under stress and at temperature, for 10–48 h.

A model of the composite as three concentric cylinders was used to calculate stresses induced by dimensional changes caused by irradiation [18]. Equilibrium of forces in a long cylindrical element results in

$$\sigma_r = \frac{\mathrm{d}}{\mathrm{d}r}(r\sigma_\theta) \tag{1}$$

and

$$\sigma_\theta = \frac{\mathrm{d}}{\mathrm{d}r}(r\sigma_r), \tag{2}$$

where σ represents stress and the subscripts r and θ represent radial and tangential (circumferential) directions, respectively. These requirements can be satisfied by stresses of the form

$$\sigma_r = A - \frac{B}{r^2} \tag{3}$$

and

$$\sigma_\theta = A + \frac{B}{r^2}. \tag{4}$$

Using the equilibrium conditions, stress–strain relationships, continuity and compatibility requirements and appropriate boundary conditions, a system of equations describing the axial stresses in the fiber and the interphase and the radial pressure at the interphase

boundaries can be derived [18]. Given appropriate materials properties, applied stresses and a desired temperature, the equations can be solved for the desired stresses and pressures.

In this work, strain components for the fiber, interphase and matrix were described by functions dependent on the neutron fluence. Mathematical expressions were found, by trial and error, that described available experimental data [12–14] for the effect of neutron irradiation on the dimensional stability of silicon carbide fibers, carbon fibers (used to simulate the interphase material) and chemically vapor deposited silicon carbide (CVD SiC). Substitution of these expressions into the equilibrium conditions, stress–strain relationships, continuity and compatibility requirements and appropriate boundary conditions yielded a solution for the axial stresses in the fiber and the interphase and the radial pressure at the interphase boundaries that were dependent on the neutron fluence. The solutions were evaluated for two types of silicon carbide fibers, Hi-Nicalon® (Nippon Carbon Company, Tokyo, Japan) and Dow-X (Dow Corning, Midland, MN, USA), at 293 and 1273 K. The Dow-X fiber was an experimental material that was further developed by Dow Corning (Midland, MI) and is now available as Sylramic™ fiber.

3. Results and discussion

Earlier reports have indicated that the creep of silicon carbide and silicon carbide fibers is enhanced by irradiation. Price [12,13] estimated the creep rate of monolithic CVD SiC from measurements of stress relaxation after irradiation. Price claimed that the creep rate was linearly proportional to the applied stress and the flux

$$\dot{\varepsilon}_c = K\sigma_{app}\varphi, \tag{5}$$

where K is a constant, σ_{app} represents the applied stress, and φ represents the flux. For an applied stress of 100 MPa at 900°C after a fluence of 5 dpa ($E > 0.18$ MeV), at the end of approximately six months, the creep rate predicted by Eq. (5) is 2.7×10^{-11} s^{-1}. In comparison, for an applied stress of 100 MPa, the thermally induced creep rate at 900°C is 1.05×10^{-13} s^{-1} [27]. Therefore, irradiation enhances the creep rate by more than an order of magnitude.

Torsional creep during light ion irradiation of fibers consisting of a 33 μm diameter carbon fiber coated with 67.5 μm of silicon carbide, deposited by CVD (SCS-6 monofilaments, Textron Speciality Materials, Lowell, MA, USA) was measured (Fig. 1). The results clearly show that radiation increased the creep rate of these fibers. Similar results have been obtained for Hi-Nicalon fibers.

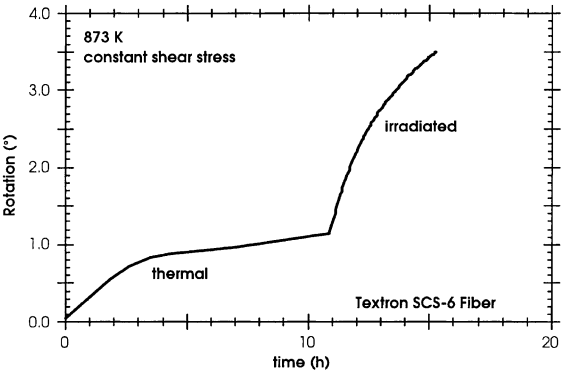

Fig. 1. Torsional creep of SCS-6 silicon carbide fibers before and during light ion irradiation.

In a previous work, it was observed that the ratio of the rate of subcritical crack growth for composites containing two different polymer-derived silicon carbide fibers (ceramic grade Nicalon® and Hi-Nicalon® fibers) was directly proportional to the ratio of the fiber creep rates [7]. By analogy, the rate of subcritical crack growth in radiation environments, controlled by FRM, may be proportional to the ratio of irradiation-enhanced fiber creep to thermal creep. Using this assumption, the rate of predicted irradiation-enhanced subcritical crack growth is compared to that observed outside radiation environments in Fig. 2. Radiation is predicted to increase the rate of subcritical crack growth by an order of magnitude.

Previous studies have also demonstrated how IRM enhances the rate of subcritical crack growth in SiC_f/SiC_m by removal of the interphase material [6,7]. Measurements of the push-in behavior of fibers that had been exposed for progressively longer times, in air at 800°C, showed that the fibers appeared more compliant, due to a longer gauge length, as the interphase recession length increased (Fig. 3) [24]. Fibers in composites with

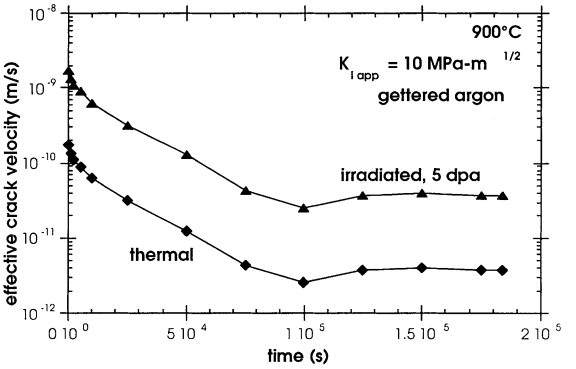

Fig. 2. Predicted rate of subcritical crack growth for an SiC_f/SiC_m composite containing 40 volume fraction Hi-Nicalon fibers.

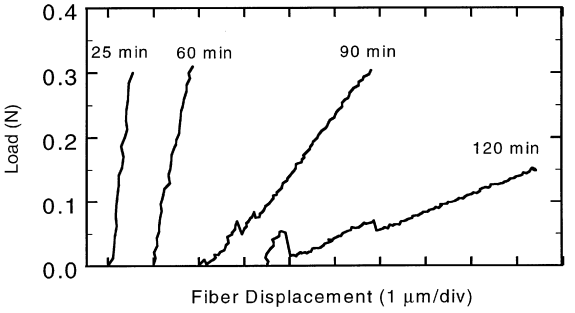

Fig. 3. The push-in behavior of fibers exposed for progressively longer periods of time at 800°C, in air, with progressively greater lengths of interphase recession.

intact but debonded interphases exhibit non-linear compliance behavior such that the fiber displacement is quadratically dependent on the force on the fiber [28–35]. Subsequent to interphase recession, however, frictional forces between the fiber and the matrix are removed and the fiber displacement is linearly dependent on the force on the fiber (see Fig. 3). Henager et al. have developed a micromechanical model to describe the effect of time-dependent, non-linear bridging forces on crack growth [23]. Henager et al. have modeled time-dependent bridging forces with either quadratic or linear compliance functions. The model results show the same general behavior as experimentally observed crack growth (Fig. 4). Therefore, loss of bridging tractions due to IRM is expected to increase the effective crack velocity and results in a crack velocity that is linearly dependent on time.

To analyze the effects of irradiation on the mechanical behavior of SiC_f/SiC_m in radiation environments, it is essential to know the stress state in the composite constituents and the relevant micromechanical relationships. It is assumed that the micromechanical relationships are the same as those observed in experiments outside of radiation environments. Thus, creeping fibers with intact interphases will obey a quadratic compliance relationship; those with interphases removed by oxidation, or some other process, will obey a linear compliance relationship. Various studies have found that irradiation causes dimensional change in the constituents of SiC_f/SiC_m composites [12–14]. Silicon carbide fibers have been found to shrink [14]. CVD silicon carbide exhibits swelling that saturates after a dose of only a few dpa to a value that is dependent on the temperature [12,13]. Although the density of graphitic carbon remains constant, the material swells in a direction normal to the basal planes and shrinks in the directions of the planes [14].

Based on the available literature, equations describing the dimensional change of each of the constituents were developed. These equations were used to describe the irradiation-induced strain in the constituents of the concentric cylinder model described earlier. The model was used to predict the resulting stresses for two different fibers of interest to fusion materials designers. One fiber, Hi-Nicalon®, exhibited shrinkage due to irradiation [14]. The other fiber, Dow-X, had been subjected to a high-temperature heat treatment to convert it to polycrystalline silicon carbide and it exhibited a limited amount of swelling [14].

At 1000°C, the stresses predicted in the constituents of the concentric cylinders model of a composite reinforced by Hi-Nicalon® fibers increase with increasing dose (Fig. 5). The axial stress in the fiber exceeds 3 GPa at a dose below 5 dpa. The axial stress in the carbon interphase remains below 1 GPa for a dose up to 100 dpa. The radial pressure at the boundary between the fiber and the interphase increases monotonically and reaches 1.5 GPa at a dose between 10 and 15 dpa. The radial pressure at the boundary between the interphase and the matrix is initially highly compressive (≈−9 GPa), but becomes tensile after a dose of about 5 dpa and then has almost the same value as the radial pressure at the boundary between the fiber and the interphase. These results are only valid if the constituents remain intact

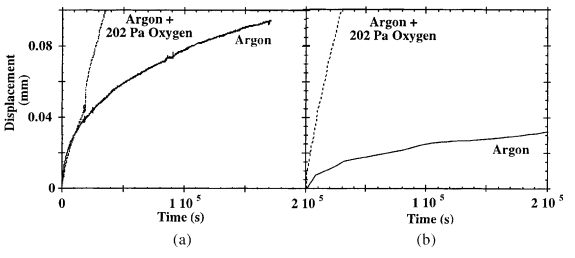

Fig. 4. A comparison of (a) measured, and (b) calculated SENB specimen midpoint displacement as a function of time in inert and oxygen containing environments.

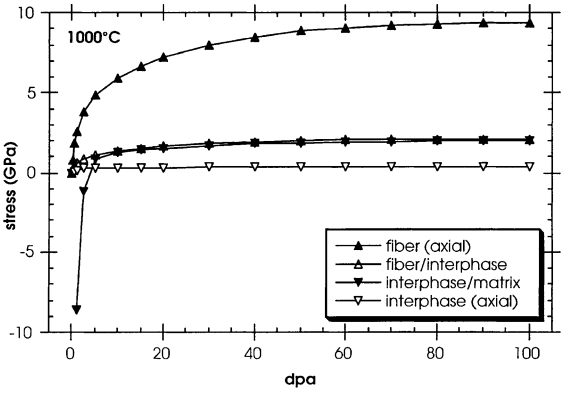

Fig. 5. Irradiation-induced stresses in a Hi-Nicalon reinforced composite.

and bonded, which is highly unlikely. The high-temperature strength of Hi-Nicalon® fibers has been reported as 1.9 GPa when tested in air [36] and between 2 and 3 GPa when tested after exposure in argon [37]. Therefore it is likely that fiber failure will occur between 0.5 and 2.5 dpa. Fiber failure will relieve some of the irradiation-induced stress, but the stress will continue to accumulate under exposure to radiation resulting in multiple fracture of fibers. This process should result in composites with low values of fracture toughness and fracture surfaces having limited fiber pullout. Since fibers fail, this process is similar to the OEM mechanism described earlier. This mechanism severely and rapidly degrades the mechanical properties of SiC_f/SiC_m and is extremely undesirable.

In comparison, the model predicts lower stresses in the Dow-X fiber than the Hi-Nicalon® fiber at either 1000°C or 100°C (Fig. 6). At 100°C, the axial stress in the Dow-X fiber reaches a local maximum between 2.5 and 10 dpa. The stress at this maximum is approximately 2.6 GPa. Although the Dow-X fiber was an experimental material, the strength of a later version, Sylramic™, is reported to be 3.2 GPa [36]. Debonding of the carbon interphase was observed during fiber push-in testing at applied axial stresses of approximately 1.5 GPa. At 1000°C, the predicted axial stress in the Dow-X fiber is significantly lower than at 100°C whereas the radial pressure at the boundary between the fiber and the interphase is relatively unchanged. In composites reinforced with Dow-X fibers, therefore, debonding may occur prior to fiber failure during high-temperature operation. By analogy with results from experiments outside of radiation environments, this process could lead to IRM-like crack growth. There is a difference between IRM and irradiation-induced debonding, however. Whereas oxidation progressively removes interphase material along the axis of fibers from their intersection with matrix cracks, irradiation-induced debonding is a bulk phenomenon. Therefore, although IRM-like crack growth may occur, it is more likely that the fibers will separate from the matrix producing a material that behaves like a bundle of fibers. The fracture surface of such materials will contain fibers with large pullout lengths, but the overall load carrying capacity of the material will be low.

The model used to predict the stresses due to irradiation-induced dimensional changes in composites with either type of fiber does not account for relaxation processes. As discussed earlier, irradiation is also anticipated to enhance the creep rate of silicon carbide. Therefore the stresses in actual components will be time-dependent and the axial stresses in the fibers will probably be lower than those shown in Figs. 5 and 6. Although fiber fracture will accommodate some of the irradiation-induced stress, the final length of broken fibers will be determined by a combination of stress transfer, creep relaxation, and interphase debonding with the outcome being that there will be longer lengths of fiber than if stress relaxation or debonding did not occur. Likewise, irradiation-enhanced creep will also occur in composites with axial stresses in the fibers lower than those required to cause fracture of a fiber. In these composites irradiation-induced creep may reduce the axial stress in fibers, but at the expense of increasing the radial tension at the boundary between the fiber and the interphase, since creep is a constant volume process. Therefore irradiation-induced creep may accelerate the IRM-like processes in composites containing fibers with lower irradiation-induced dimensional mismatches. Axial stresses in the fibers could be minimized by use of a fiber with similar irradiation-induced dimensional change as the matrix, and in this case irradiation-enhanced FRM may become a primary failure mechanism.

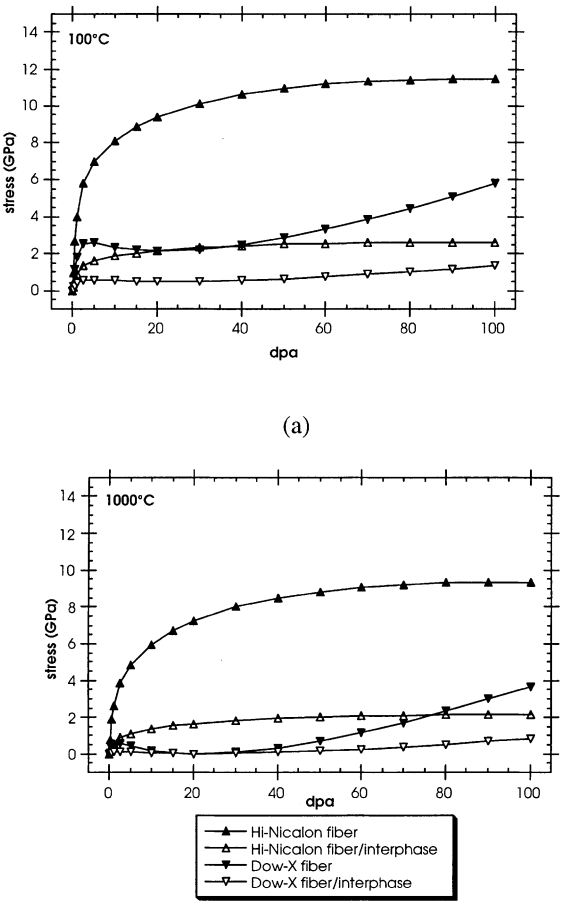

Fig. 6. Predicted axial stresses in Hi-Nicalon and Dow-X fibers, and the predicted radial pressure at the boundary between the fiber and the interphase at (a) 100°C, and (b) 1000°C.

4. Summary

Irradiation enhances creep and dimensional changes in silicon carbide composite constituents. Irradiation-enhanced creep is expected to be one order of magnitude higher than thermal creep. Irradiation-induced mismatch strains give rise to significant stresses in the constituents of SiC_f/SiC_m composites under irradiation. Fibers with behavior closer to that of the matrix material have less axial tensile stress than fibers that shrink during irradiation but the radial pressure at the boundary between the fiber and the interphase may eventually cause debonding. Fiber failure and debonding will degrade the mechanical behavior of SiC_f/SiC_m composites during irradiation.

Acknowledgements

This work was supported by The Office of Fusion Energy Science and The Office of Basic Energy Science under US Department of Energy (DOE) contract DE-AC06-76RLO 1830 with Pacific Northwest National Laboratory, which is operated for DOE by Battelle. The authors would like to thank Dr J.I. Eldridge, NASA Glenn Research Center, for performing the fiber push-in testing.

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