CREEP BEHAVIOR FOR ADVANCED POLYCRYSTALLINE SIC FIBERS - G. E. Youngblood and R. H. Jones (Pacific Northwest National Laboratory),[1] G. N. Morscher[2] (Case Westerm Reserve University) and Akira Kohyama (Institute of Advanced Energy, Kyoto University, Kyoto 611, Japan)

OBJECTIVE

The objective of this work is to examine irradiation enhanced creep behavior in advanced polycrystalline SiC fibers.

SUMMARY

A bend stress relaxation (BSR) test has been utilized to examine irradiation enhanced creep in polycrystalline SiC fibers which are under development for use as fiber reinforcement in SiC/SiC composite. Qualitative, S-shaped 1hr BSR curves were compared for three selected advanced SiC fiber types and standard Nicalon CG fiber. The temperature corresponding to the middle of the S-curve (where the BSR parameter m = 0.5) is a measure of a fiber's thermal stability as well as its creep resistance. In order of decreasing thermal creep resistance, the measured transition temperatures were Nicalon S (1450 °C), Sylramic (1420 °C), Hi-Nicalon (1230 °C) and Nicalon CG (1110 °C).

PROGRESS AND STATUS

Introduction

As part of the Joint DOE/Monbusho Program to support materials development for fusion energy, PNNL has initiated a systematic study of the potential effects of irradiation creep in SiC/SiC composites and SiC fibers [1,2]. A previous report described a simple bend stress relaxation (BSR) test designed to examine the creep behavior of irradiated and unirradiated SiC fibers [3]. This report presents some new thermal creep (BSR) results for several unirradiated advanced SiC fiber types. The selected advanced fibers, namely Nicalon S™ and Hi-Nicalon™ manufactured by Nippon Carbon Co. and Sylramic™ manufactured by Dow Corning Corp, were all textile grade types suitable for incorporation into SiC_f/SiC composite. A planned test matrix designed to examine the creep behavior of these advanced fibers during irradiation also is presented.

Theoretical and Experimental Review

Theoretical and experimental details of the BSR test, developed initially by researchers at NASA Lewis [4], were presented previously [3]. Briefly, an initial elastic bend strain is applied to a single fiber by wrapping several coiled loops onto a SiC mandrel of radius R_o. The loops are captured by a SiC sleeve that slips over the mandrel. For the test designed to examine the potential effects of irradiation enhanced creep, the fiber loops will be subjected to a specific time (t), temperature (T), and irradiation dose (ϕ) at the fixture imposed strain $e_o = r_f/R_o$, where e_o represents the maximum strain at the outer edge of the coiled fibers and r_f is the fiber radius. To separate thermal from irradiation creep effects, thermal control BSR tests are carried out without irradiation for equivalent times and temperatures.

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In both these tests, a BSR parameter "m" quantifies the stress relaxation that occurs during either treatment:

$$m = 1 - R_o/R_{(T \text{ or } \phi)} \approx 1/[1+(e_c/e_o)] \quad (1).$$

In Eq. (1), R_T or R_ϕ is the arc radius of a relaxed fiber segment measured at room temperature after the thermal or the irradiation treatment, respectively. In the approximation, e_c is representative of the tensile creep strain that would result in a constant stress creep test for exposure temperatures and times equivalent to the BSR test temperatures and times and where the stress is taken equal to e_oE and E is the elastic modulus [5]. Thus, a constant strain BSR fiber creep experiment can yield basic creep data, i.e., stress and time dependence exponents and thermal activation energies. Unlike a conventional constant stress creep test, a fiber BSR test can be conveniently designed to take place in a reactor, thus its utility for examining the potential effects of irradiation enhanced creep. In BSR tests, values of m will range from 0 to 1 with m = 1 or 0 indicating the occurrence of no relaxation or complete relaxation by creep, respectively, while intermediate values of m indicate partial relaxation. The temperature dependence of m itself gives a convenient qualitative ranking of the creep resistance of various fiber types. Furthermore, Morscher and DiCarlo [4] have shown that the drop off in fiber strength with increasing temperature correlates with the transition temperature range of the BSR parameter m. Thus, a quantitative measure of the fiber thermal stability is the temperature at which m = 0.5 for a particular exposure time, i.e., 1 hr, 100 hr, 1000 hr, etc.

If actual creep strains are desired, Eq. (1) can be inverted to estimate e_c from measured m-values for either thermal creep (TC)- or irradiation creep (IC)- BSR tests. Then the irradiation dependent strain increment $\Delta e = e_c(\phi,T,t,e_o) - e_c(T,t,e_o)$ for an irradiation fluence (or dose) ϕ and is given by

$$\Delta e = e_o\{1/m(\phi,T,t,e_o) - 1/m(T,t,e_o)\} \quad (2).$$

The term in brackets is the difference in reciprocal m-values for the IC- and the TC-BSR tests for equivalent treatment temperature T, time t and maximum applied fiber strain e_o.

In this reporting period, several TC-BSR tests were carried out after heat treating the fiber coils for one and 100 hour periods in argon in a muffle tube furnace. Some of the TC-BSR tests were carried out in the same fixtures as will be used later in the IC-BSR tests; other tests were carried out by NASA Lewis in a similar BSR test rig. For all cases, e_o was about 0.2%.

The fibers tested at PNNL or NASA Lewis were selected from the same fabrication lots. Further characterization of representative fibers selected from these lots is being carried out at PNNL. Average fiber diameters are being determined by optical or SEM images of typical fiber cross-sections, densities by a liquid gradient column, and room temperature tensile strengths by using a Micropull™ device designed for this purpose.

Results and Discussion

The measured, lot specific fiber properties are given in Table 1 and are marked with an asterisk. The listed property values not marked with an asterisk were provided by the manufacturer and pertain to production fiber properties.

Table 1. Properties of SiC Fibers Selected for the IC-BSR Tests

Property		Nicalon CG	Hi-Nicalon	Sylramic	Nicalon S
Fiber diameter	(um)	14*	12*	10*	11*
Density	(g/cm3)	2.55*	2.74*	3.0	3.10
Tensile strength	(GPa)	3.0*	3.4*	3.1	2.6
Tensile modulus	(GPa)	190	270	400	420
Elongation	(%)	1.4	1.0	0.8	0.6
Chemical composition	Si (wt %)	56.6	62.4	69	68.9
	C	31.7	37.1	29	30.9
	O	11.7	0.5	-	0.2
	C/Si (atomic)	1.31	1.39	1.0	1.05
Crystallite grain size	(nm)	<2*	5-10*	500	11
Thermal Stability	(°C)	1110*	1230*	1420*	1450*

* measured values for these fiber lots. Other values were provided by the manufacturer.

In Figure 1, the 1 hr BSR parameter m is plotted as a function of reciprocal temperature for the three advanced SiC fibers as well as for commercially available ceramic grade Nicalon CG fiber.

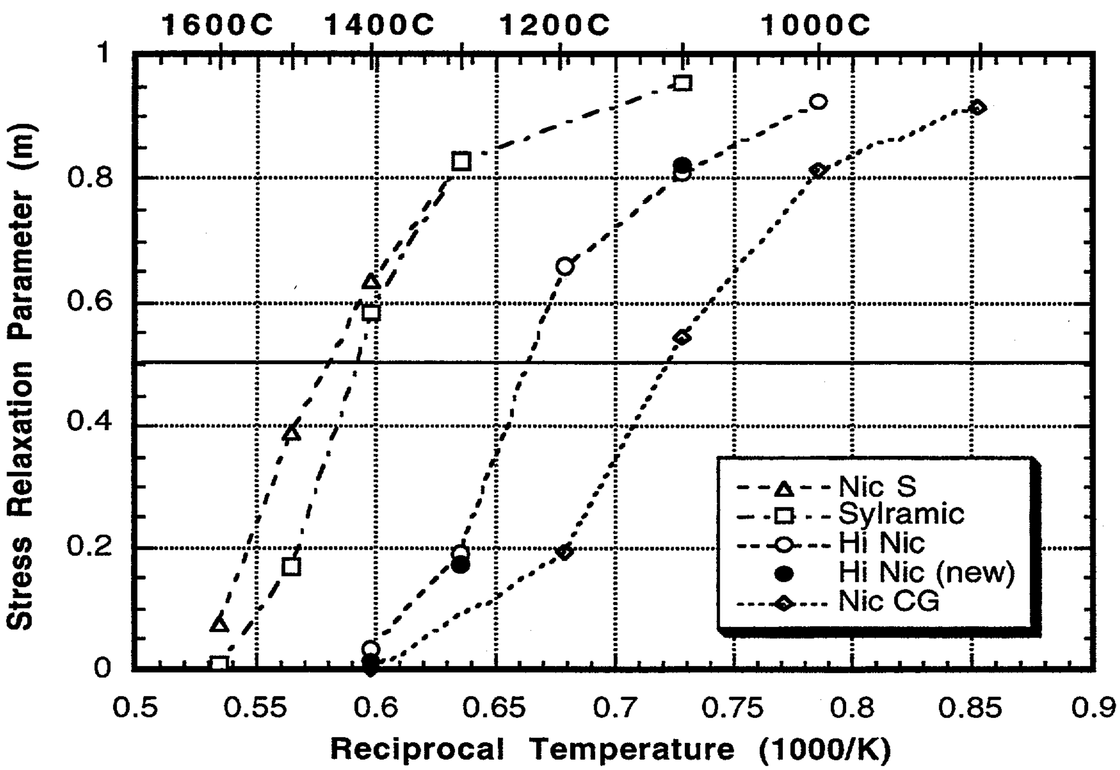

Figure 1. Comparison of one hour BSR thermal creep results for advanced SiC and Nicalon CG fibers ($e_o = 0.2\%$).

Qualitatively, the S-shaped curves have similar shapes. The transition from a slightly crept fiber (m > 0.9) to a near totally crept fiber (m < 0.1) takes place over about 350°C for all these fibers. The S-shaped curve for the fibers exhibiting the highest creep resistance are shifted to the left. Thus, in order of decreasing creep resistance, Nicalon S has the highest creep resistance, slightly higher than Sylramic, both of which are higher than Hi-Nicalon which is higher than Nicalon CG. The trends exhibited by these data generally agree with the trends delineated by Morscher and DiCarlo [6]; that is, lower oxygen content is the primary determinant of high creep resistance, while lower excess carbon content and near stoichiometric composition also contribute to a higher creep resistance in a SiC type fiber. Additional m-values for 20 and 100 hr exposures at 1000 and 1100°C were reported previously [3]. When compared to the corresponding 1000 and 1100°C 1 hr. m-values, the 20 and 100 hr values would be displaced to lower temperatures, i.e., to the right of these 1 hr data, as expected.

For each S-curve, the temperature where m = 0.5 is estimated and listed in Table 1 as the fiber "thermal stability." The data for the Hi-Nicalon fiber were obtained for two different lots. Although the common 1 hr m-values are in good agreement for these two different lots, the measured tensile strength (3.4 GPa) for the "new" lot of Hi-Nicalon fiber is somewhat greater than the listed production tensile strength value of 2.8 GPa. It is possible that the thermomechanical properties of Hi-Nicalon have a sensitive dependence upon fabrication details, which were still in a state of development when these fibers were obtained. Therefore, the higher strengths measured for the new lot of Hi-Nicalon may indicate some optimizing in its fabrication has taken place. For instance, Yun et al [7] have shown that annealing Hi-Nicalon in argon for one hour at 1500°C (well above its fabrication curing temperature of about 1300°C) significantly increases its average grain size and creep resistance while the tensile strength decreases.

CONCLUSIONS

The 1 hr TC-BSR test results indicated that the three advanced SiC fibers selected for later irradiation creep testing (Nicalon S, Sylramic and Hi-Nicalon), exhibited greater thermal creep resistance than standard Nicalon CG fiber, as expected. Using the temperatures at which m = 0.5 as a quantitative measure of thermal stability and creep resistance, the selected fibers ranked in order of highest thermal creep resistance first: Nicalon S (1450°C), Sylramic (1420°C), Hi-Nicalon (1230°C) and Nicalon CG (1110°C).

FUTURE WORK

To optimize SiC_f/SiC composite properties for fusion energy applications, it might be desirable to include a final high temperature anneal after fabrication. In particular, Hi-Nicalon strength and creep resistance have been shown to be sensitive to such a treatment, as noted above. Therefore, annealed Hi-Nicalon will be added to future fiber BSR testing. Also in the next reporting period, the 100 hr TC-BSR testing will be completed and reported for the selected SiC fibers.

The planned IC-BSR test matrix is presented in Table 2. The first IC-BSR irradiation cycle is scheduled to start in January, 1997 in the ATR reactor in Idaho Falls. Its irradiation test temperature will be 370°C, well below thermal creep regime temperatures. However, if irradiation creep is temperature independent down to 370°C, as preliminary data suggest [1], any observed creep will be totally irradiation induced. The following three tests will examine fiber creep in the more fusion relevant temperature range (800 - 1000°C), a range where both thermal and irradiation induced creep could be important. These latter tests will examine the dose dependence to 25 dpa-SiC as well as the applied strain dependence from 0.1 to 0.25%.

Table 2. SiC Fiber IC-BSR Test Matrix

Irrad Temp (°C)	Dose (dpa-SiC)	Time (hrs)	Applied Strain (%)	Initial Stress* (MPa)	Facility-Start Date
370	2	1500	0.2	400-800	ATR-Jan 97
1000	2	1500	0.1, 0.2, 0.25	200-1000	ATR-Jul 97
800	10	6400	0.1, 0.2, 0.25	200-1000	HFIR-Jan 98
1000	25	6400	0.1, 0.2, 0.25	200-1000	HFIR-Jan 98

* Depends on the fiber elastic modulus.

REFERENCES

1. F. A. Garner, G. E. Youngblood and M. L. Hamilton, "Review of Data on Irradiation Creep of Monolithic SiC," Fusion Materials Semiannual Progress Report for Period Ending December 31, 1995. DOE/ER--0313/19.

2. M. L. Hamilton, C. A. Lewinsohn, R. H. Jones, G. E. Youngblood, F. A. Garner and S. L. Hecht, "Progress in the Development of a SiC_f/SiC Creep Test," Fusion Materials Semiannual Progress Report for Period Ending June 30, 1996. DOE/ER--0313/20.

3. G. E. Youngblood, M. L. Hamilton and R. H. Jones, "Technique for Measuring Irradiation Creep in Polycrystalline SiC Fibers," Fusion Materials Semiannual Progress Report for Period Ending June 30, 1996. DOE/ER--0313/20.

4. Gregory N. Morscher and James A. DiCarlo, "A Simple Test for Thermomechanical Evaluation of Ceramic Fibers," *J. Amer. Ceram. Soc.,75(1),136-40 (1992).*

5. James A. DiCarlo, "Creep Limitations of Current Polycrystalline Ceramic Fibers," *Composites Science and Technology 51, 213-222 (1994).*

6. Gregory N. Morscher and James A. DiCarlo, "Creep and Stress Relaxation Properties in Relation to Microstructure for SiC Fibers," *HITEMP Review Vol. III, Oct. 25-27, 1993. NASA CP19117.*

7. H. M. Yun, J. C. Goldsby and J. A. DiCarlo, "Effects of Thermal Treatment on Tensile Creep and Stress-Rupture Behavior of Hi-Nicalon SiC Fibers," *Ceram. Eng. Sci. Proc. 16(5), 987-996 (1995).*