AN UPDATE ON THE KFIB EXPERIMENT – G. E. Youngblood and R. H. Jones (Pacific Northwest National Laboratory)*, W. Kowbel (MER Corporation), Paul de Heij (NRG Petten) and Akira Kohyama (Kyoto University).

OBJECTIVE

The primary objective of the experiment called "KFIB" is to assess the thermal conduction **(K)** properties of several advanced SiC **FIB**ers before and after irradiation. The thermal conductivity of SiC/SiC composites made from these fibers (with various SiC-type matrices and architectures) will also be measured before and after irradiation. Models used to predict the transverse and in-plane thermal conductivity of these composites as a function of temperature and dose also will be assessed.

SUMMARY

An updated sample test matrix for the KFIB experiment is presented. The pre-irradiation test results for all fiber and SiC/SiC composite materials included in the test matrix, presented at the 4th IEA Workshop on SiC/SiC for Fusion Structural Applications held in Frascati, Italy, October 12-13, 2000, are reviewed. The KFIB samples have been delivered for capsule loading. They are scheduled to be irradiated from October-November, 2001 through April-May, 2002 to similar doses (2-4 dpa-SiC) in the ATR (Idaho Falls) at 300°C and the HFR (Petten) at 625 and 975°C. To assess irradiation enhanced creep (IEC) in SiC fibers, a bend stress relaxation module was added to the HFR sample matrix at each irradiation temperature.

PROGRESS AND STATUS

The KFIB experiment is coordinated at the Pacific Northwest National Laboratory (PNNL), but involves several other organizations who have furnished materials, materials analysis or irradiation facilities for the experiment [1].

Irradiation Tests and Schedule

Two irradiation tests are planned. A low temperature irradiation test will take place in the ATR reactor at 300°C to a dose of 3.6 dpa-SiC. A moderate and a high temperature irradiation test will take place in the HFR Petten reactor at 625°C and 975°C to a dose of about 2.5 dpa-SiC. Both irradiations will commence in October-November, 2001 and will be completed by April-May, 2002. Post-irradiation examinations (PIE) will be carried out in late 2002.

Sample Test Matrix

Preliminary KFIB sample test matrices have been reported [1-2]. However, two types of SiC/SiC composite bend bars and two fiber bend stress relaxation modules recently were added to the HFR Petten sample test matrix. An updated KFIB sample test matrix is given in Table 1.

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Table 1. Updated KFIB sample test matrix (7/31/01)

Uniaxial Fiber + Ceraset™ Discs (6.2 mm dia x 2.0 mm thk)	**HFR Petten (625°C and 975°C)**	**ATR (300°C)**
Hi-Nicalon[1] (parallel)	4	2
Hi-Nicalon[1] (perpendicular)	2	2
Tyranno SA-3[2] (parallel)	4	2
Hi-Nicalon Type S[1] (parallel)	4	2
Amoco K1100 graphite[3] (parallel)	0	2
SiC/SiC Composite Discs (6.2 mm dia x 2.0 mm)		
2D-Nicalon S/4xC-SiC multilayers/CVI-SiC[4]	6	4
3D-Nicalon S/PIP - Ceraset™ 1400C[5]	4	4
2D-8HS SA-Tyrannohex HP (100% transverse)[2]	4	3
2D-8HS SA-Tyrannohex HP (50% transverse)[2]	0	2
2D-5HS Nicalon S/150 nm PyC/CVR-PIP SiC[6]	6	0
SiC/SiC Bend Bars (30 mm x 6 mm x 2 mm)		
2D-PW Dupont Hi Nic/150 nm PyC/CVI-SiC[7]	8	0
2D-5HS Nicalon S/150 nm PyC/CVR-PIP SiC[6]	3	0
High-Purity CVD-SiC - Reference		
CVD-SiC (6.0 mm dia x 2.5 mm discs)[8]	5	3
CVD-SiC (25.5 mm long x 1.85 mm sq. bars)[8]	4	0
Fiber Bend Stress Relaxation Modules		
Tyranno SA-3 (10 μm dia)[2]	3	0
Hi-Nicalon S (13 μm dia)[1]	3	0
Textron 2-mill (50 μm dia)[9]	2	0

[1] Nippon Carbon Co., Yokohama, Japan
[2] Ube Industries Ltd., Ube City, Japan
[3] Amoco Corp., USA
[4] Hypertherm High Temperature Composites, Inc., Huntington Beach, CA
[5] JAERI, Tokai-mura, Japan
[6] MER Corp., Tucson, AZ
[7] Dupont Lanxide Composites, Newark, DE
[8] Rohm and Haas (formerly Morton Advanced Materials), Woburn, MA
[9] Textron Specialty Materials, Lowell, MA

The Hi-Nicalon™ and Hi-Nicalon™ Type S SiC fibers were fabricated by the Nippon Carbon Co. using an electron-beam radiation curing process [3]. The Hi-Nicalon™ fiber has excess C (C/Si = 1.39), while the Type S fiber is nearly stoichiometric SiC (C/Si = 1.05). The Tyranno™ SA-3 fiber, also nearly stoichiometric SiC, was fabricated by Ube Industries Ltd. using an oxidative curing process and high temperature (1800°C) sintering. It is noted that the tested Tyranno™ SA-3 fiber, a new smaller diameter version of Tyranno™ SA, has improved thermal creep and high temperature strength compared to the Tyanno™ SA fiber [4]. The Amoco K1100™ graphite fiber is pitch-derived and given a high temperature treatment so that highly graphitic platelets preferentially align parallel to the fiber axis to optimize the axial thermal conductivity [5]. MER currently is developing a composite design that uses stitched bundles of K1100™ graphite fiber in the z-direction to improve the transverse thermal conductivity of a 2D-SiC/SiC composite [6].

To determine the thermal conductivity of advanced SiC or graphite fibers, a composite rod (7-mm dia. x 20 mm) was constructed with uniaxially aligned fibers in an amorphous Ceraset™ matrix. As discussed previously, several discs were sliced from each rod for thermal diffusivity measurements [1-2]. Irradiation enhanced creep (IEC) will be assessed for two small diameter polymer-derived SiC fibers (Hi-Nicalon™ type S and Tyranno™ SA-3) and a SiC fiber made by chemical vapor deposition (Textron™ 2-mill) by a bend stress relaxation (BSR) test [7].

The unirradiated properties and the rationale for examining the thermal conductivity of the various SiC/SiC composite types listed in Table 1 were presented previously [1-2]. Several bars and thermal diffusivity discs made from monolithic CVD-SiC were included in the KFIB sample matrix as reference materials [8]. Two types of SiC/SiC composite bend bars were added to the HFR sample matrix: (1) 2D-5HS Nicalon S/150 nm PyC/CVR-PIP SiC, a material made by MER using a satin weave (5HS) Hi-Nicalon type S fabric with a hybrid matrix that combines a chemical vapor reaction (CVR) and a polymer infiltration and pyrolysis (PIP) process, and (2) Hi-Nicalon PW/150 nm PyC/CVI-SiC, a material made by Dupont Lanxide with a plain weave (PW) Hi-Nicalon fabric, a thin 150 nm PyC fiber coating and a chemical vapor infiltration (CVI) matrix (the so-called SiC/SiC composite reference material). The latter material has already been examined extensively in helium swelling experiments [9], and is included in the ongoing JUPITER 14J irradiation experiment [10].

Thermal Conductivity Analysis

The effective thermal conductivity of a composite fiber disc, K_{eff}, is determined from:

$$K_{eff} = \alpha_{eff} \bullet \rho_{bulk} \bullet C_p \qquad \text{(W/mK)} \qquad [1]$$

where C_p is the calculated heat capacity and α_{eff} and ρ_{bulk} are the measured thermal diffusivity and bulk density values, respectively. In cases where the sample consisted of two phases, the heat capacity was calculated by a rule of mixtures. For those cases, the following analytic expressions were derived to fit heat capacity data from the literature for SiC and graphite [11] and the Hi-Nicalon™ fiber composition [12]:

$$C_p(\text{SiC}) = 1.0337 + 0.0001949(T) - 36{,}582/T^2 \qquad \text{(J/gK)} \quad [2a]$$

$$C_p(\text{graphite}) = 1.8256 + 0.0002943(T) - 356.3/T \qquad \text{(J/gK)} \quad [2b]$$

$$C_p(\text{Hi-Nic}) = 1.0549 + 0.0002367(T) - 39{,}361/T^2 \qquad \text{(J/gK)} \quad [2c]$$

A parallel conduction model was assumed for the fiber disc with uniaxially aligned fibers in an amorphous Ceraset matrix:

$$K_{eff} = f_f K_f + f_m K_m \qquad [3]$$

where f_i and K_i are the volume fraction and the thermal conductivity values for the fiber (f) and matrix (m) constituents, respectively. The uniaxial composite discs were purposely made with a high fiber packing fraction so that $f_m < f_f$; and the Ceraset matrix was cured at only 1100°C to preserve its amorphous microstructure so that $K_m < K_f$. Therefore, the $f_m K_m$ term in Eq. [3] is only a small correction term.

From Eq. [3], K_f-values were estimated up to 1000°C, and the fiber thermal diffusivity (α_f) was calculated from:

$$K_f = \alpha_f \bullet \rho_f \bullet C_f \qquad [4]$$

where ρ_f and C_f are the fiber density and heat capacity values as a function of temperature, respectively.

The laser flash technique for measuring the thermal diffusivity and calculating the thermal conductivity via Eq. [1] assumes that the disc sample is homogeneous among other things [13]. The technique has also been demonstrated to be appropriate for randomly dispersed and continuous fiber-reinforced composites under certain conditions [14]. The relative dimensional and thermal conductivity values of the composite constituents set these conditions. They require that local perturbations from a uniform one-dimensional temperature gradient through the disc sample are not too severe. This will occur if the time constant for lateral heat conduction between the constituents is much smaller than the time constant for axial conduction through the sample. As demonstrated by Lee [14], the largest error expected when using the composite technique for determining the thermal conductivity of fine diameter fibers is due to the difficulty in accurately determining the fiber volume fraction term in Eq. [3].

Results

Example base-line thermal diffusivity and conductivity values for unirradiated fiber disc and composite samples are presented in Figures 1(a-f)[1].

In Figure 1a, the calculated K_f-values for all the fibers included in the KFIB test matrix are compared to the K-values for CVD-SiC. For completeness, the K_f-values for the Nicalon™ CG fiber taken from the literature were added to the figure. The K_f-values cover three orders of magnitude, with the K_f-values for the Tyranno™ SA-3 and Hi-Nicalon™ type S being the highest for the SiC fibers; and the K_f-values for the K1100™ graphite fiber being considerably higher than CVD-SiC K-values. It is expected that K_{eff} for SiC/SiC composites made with the Tyranno™ SA-3 or Hi-Nicalon™ type S fibers would exhibit higher thermal conductivity values compared to often tested SiC/SiC made with Nicalon™ CG or even Hi-Nicalon™ fibers. The data also suggest that the anisotropic, but highly conductive K1100™ graphite fiber might be used to boost the thermal conductivity of SiC/SiC composites in special applications, as MER is attempting to do.

In Figure 1b, the thermal diffusivity of the 13 different CVD-SiC samples are shown for the RT-400°C range. The different curves exhibit characteristic 1/T temperature dependence, but also a scatter (±20%) between each other. Even though many of the discs were cut from the same plate, the ±20% spread in α-values illustrates the importance of monitoring changes in diffusivity for the same sample before and after irradiation to properly assess degradation due to radiation effects.

In Figure 1c, the transverse thermal conductivity values for ten different samples of the 2D-Nicalon™ S/C-SiC multilayer/CVI-SiC are presented. The K-values are distributed over a wide range between 5-14W/mK. The samples with higher bulk density values, ranging from

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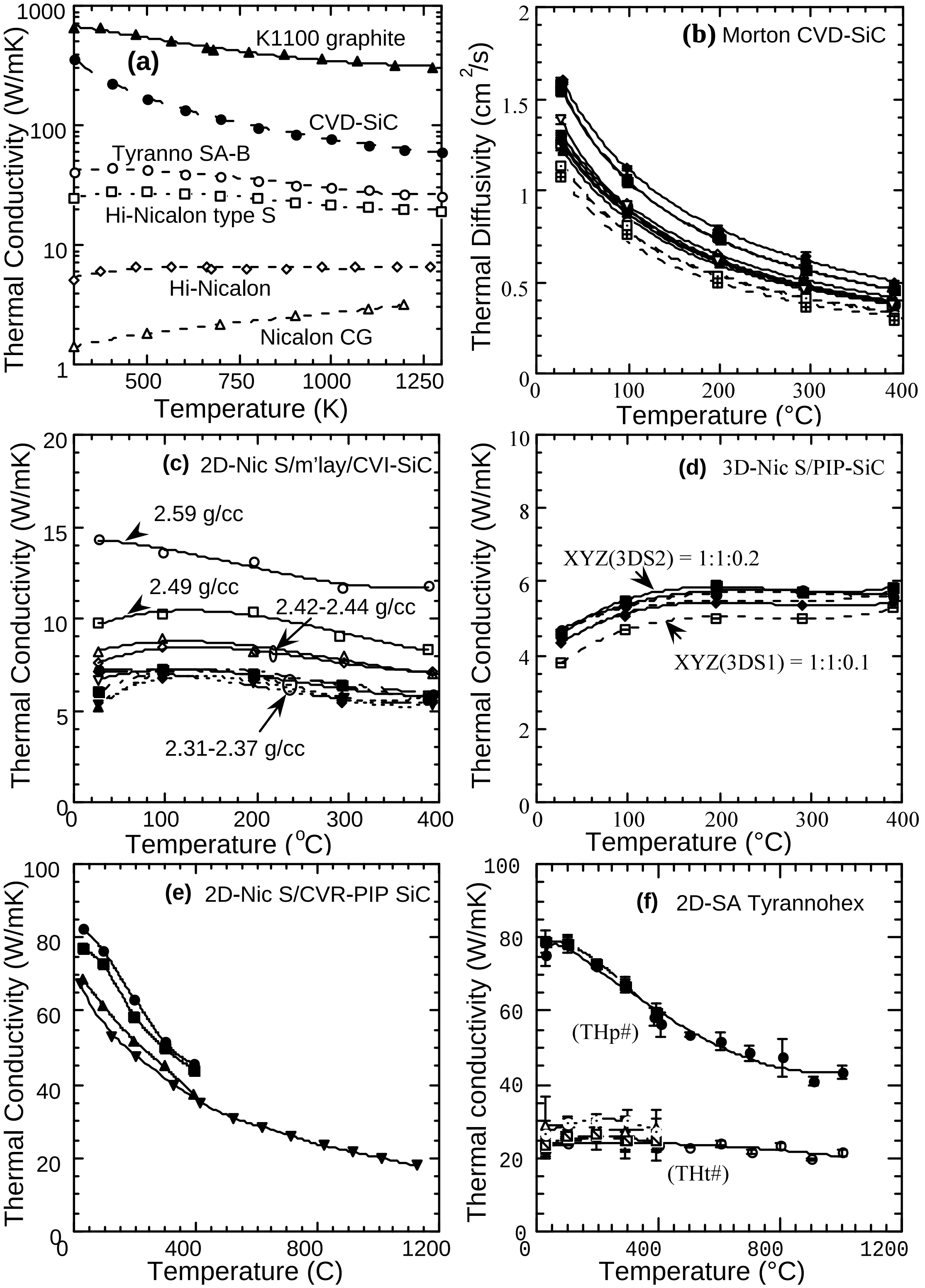
[1] Figure 1(a-f) – next page: (a) Comparison of the axial thermal conductivity for several fiber types with the thermal conductivity of CVD-SiC, (b) The measured thermal diffusivity of 13 unirradiated CVD-SiC samples, (c) The thermal conductivity of unirradiated 2D-Nicalon S/multilayer/CVI-SiC KFIB samples, (d) The thermal conductivity of unirradiated 3D-Nicalon S/PIP-SiC KFIB samples, (e) The thermal conductivity of 2D-Nicalon S/CVR-PIP SiC KFIB samples, and (f)The thermal conductivity of 2D-SA Tyrannohex KFIB samples for two orientations – in plane (upper curves) and transverse (lower curves).


(a)
Thermal Conductivity (W/mK)
1000
100
10
1
K1100 graphite
CVD-SiC
Tyranno SA-B
Hi-Nicalon type S
Hi-Nicalon
Nicalon CG
500
750
1000
1250
Temperature (K)
(b) Morton CVD-SiC
Thermal Diffusivity (cm^2/s)
2
1.5
1
0.5
0
0
100
200
300
400
Temperature (°C)
(c) 2D-Nic S/m'lay/CVI-SiC
Thermal Conductivity (W/mK)
20
15
10
5
0
2.59 g/cc
2.49 g/cc
2.42-2.44 g/cc
2.31-2.37 g/cc
0
100
200
300
400
Temperature (^{o}C)
(d) 3D-Nic S/PIP-SiC
Thermal Conductivity (W/mK)
10
8
6
4
2
0
XYZ(3DS2) = 1:1:0.2
XYZ(3DS1) = 1:1:0.1
0
100
200
300
400
Temperature (°C)
(e) 2D-Nic S/CVR-PIP SiC
Thermal Conductivity (W/mK)
100
80
60
40
20
0
0
400
800
1200
Temperature (C)
(f) 2D-SA Tyrannohex
Thermal conductivity (W/mK)
100
80
60
40
20
0
(THp#)
(THt#)
0
400
800
1200
Temperature (°C)

2.31 up to 2.59 g/cc as labeled on the curves, exhibited higher K-values. All samples with densities less densities less than 2.5 g/cc exhibited K-values less than 10 W/mK at ambient. These limited results suggest that achieving high quality composite by the CVI-method might be more difficult when multilayer coatings are used.

In Figure 1d, the thermal conductivity values for six samples of the 3D-Nicalon™ S/PIP-SiC composite are presented. The K-values all fall within a narrow range of about 5-6 W/mK and are similar for the composites made with either 0.1 or 0.2 relative fiber volume fraction in the z-direction. The PIP-SiC matrix for these composites was cured at 1400°C, a temperature too low to enhance the composite thermal conductivity by crystallizing the matrix.

In Figure 1e, the thermal conductivity values for four samples of the 2D-Nicalon™S/CVR-PIP composite are presented. The K-values averaged about 75 W/mK at ambient, and slightly exceeded 20 W/mK at 1000°C. The high K-values were obtained after giving the composite a HTT of 1800°C, which was sufficiently high to crystallize the matrix. However, to retain composite mechanical properties the SiC fiber must be thermally stable at such a high HTT.

In Figure 1f, the measured K-values are presented for samples of hot-pressed 2D-SA Tyrannohex™ composite cut so that the fibers were aligned with two different orientations. The two THp# samples had 50% of the fibers parallel and the other 50% transverse to the heat flow direction; and the K-values ranged from about 80 W/mK at ambient down to 42 W/mK at 1000°C. However, the seven THt# samples had 100% of the fibers transverse to the heat flow direction; and the K-values ranged from only 25 W/mK at ambient down to 21 W/mK at 1000°C for this group. Two reasons account for the large differences in K-values for these two groups with different fiber orientations. The thermal conductivity of the Tyranno™ SA fibers is quite high (64 W/mK at ambient [4]), so contributes significantly to K_{eff} for the THp# samples with conduction along 50% of the fibers. On the other hand, the THt# samples contain no fibers parallel to the heat flow direction and approximately twice as many transverse interfacial layers as the THp# samples. Apparently, the numerous 50-100 nm thick carbon interlayers that formed between the SA Tyranno fibers during the hot-pressing effectively act as thermal barriers and significantly contribute to lowering the transverse K-values for the THt# samples [15].

FUTURE WORK

The thermal conductivity for several types of advanced SiC fibers and for a highly oriented graphite fiber will be analyzed before and after irradiation. Similar analyses will be performed for composites made with these fibers by using appropriate thermal conductivity models. The results should be available by late-2002.

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[14] Hung Joo Lee, “Thermal Diffusivity in Layered and Dispersed Composites,” PhD Thesis, Purdue University, May 1975.

[15] G. E. Youngblood, D. J. Senor, R. H. Jones and Samuel Graham, “The Transverse Thermal Conductivity of 2D-SiC/SiC Composites,” p. 109 in FMSPR for the period June 30, 2000 (DOE/ER-0313/28).