High Thermal Conductivity SiC/SiC Composites for Fusion Applications

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ABSTRACT

SiC-SiC composites offer excellent potential for fusion energy as well as numerous commercial applications. High manufacturing cost and very low through-the-thickness (transverse) thermal conductivity (below 15 W/mK) hinders wider applications of SiC/SiC composites. To overcome these obstacles, a low-cost, Chemical Vapor Reaction (CVR) process was developed to fabricate crystalline, high purity SiC fibers. Several variations of the CVR process were used to fabricate SiC/SiC composites. An unirradiated, 4 mm thick SiC/SiC composite was completely CVR-converted from a converted SiC fiber preform with a pitch graphite matrix, and exhibited a bulk density of 2.65 g/cc, 10% open porosity, RT and 1000^{o}C thermal conductivity values of 70 and 35 W/mK, respectively, and a RT bend strength of 200 MPa.

1. INTRODUCTION

Silicon carbide (SiC) has been considered as a structural material because of its outstanding mechanical properties [1-3]. To provide improved strength and toughness, continuous fiber reinforced SiC composites (SiC_f-SiC), are being developed and are being considered for fusion reactor applications [4].

Unfortunately, while the thermomechanical properties of SiC/SiC can be improved over that of monolithic SiC, the through-the-thickness thermal conductivity is significantly reduced. A high purity SiC of theoretically full density made by chemical vapor deposition (CVD) generally will have a room temperature thermal conductivity value >300 W/mK [5], while through-the-thickness conductivities of SiC/SiC CVI-composites have been observed to be <12 W/mK at RT [1].

To achieve high-purity, crystalline β-SiC fiber, an economical process was developed to directly convert graphite fiber to SiC by a chemical vapor reaction (CVR) [3]. The crystalline β-SiC microstructures which result from CVR-processing exhibit exceedingly clean grain boundaries which contribute to maintaining overall high phonon thermal transport in a CVR-converted SiC fiber or matrix. Previously, thin high thermal conductivity were fabricated by a hybrid PIP/CVI and CVR-SiC processes [6]. This paper examines thicker SiC-SiC composites made with CVR-SiC fibers and CVR-SiC matrix. Effect of processing conditions on thermomechanical properties is reported.

2. EXPERIMENTAL PROCEDURE

A T-300 1K 8HS fabric was used as the graphite precursor. The CVR process was carried out using a SiO gas generator. The conversion of the carbon precursor to SiC was controlled by the generator's chemistry and the operating temperature. The conversion level was measured by direct oxidation of the CVR converted fibers and by SEM examination of cross section of the oxidized SiC converted fibers. Density of the densified SiC-SiC composites was measured using the Archimedes principle. Mechanical testing was performed using a four point bending fixture with a 15:1 span to depth ratio. SEM was used to examine fracture surfaces, as well as the topography of SiC converted carbon fibers. Thermal diffusivity was measured using the laser flash method. Thermal conductivity was calculated as a product of density, thermal diffusivity and heat capacity. XRD was used to perform the phase identification of the SiC matrix.

3. RESULTS AND DISCUSSION

3.1 Effect of fabric architecture on composite properties.

Previously, T-300 1K plain weave carbon precursor was used [6]. That fabric yielded very good thermomechanical properties of thin SiC-SiC composites. However, as the composite thickness increases, the intrinsic closed porosity increases, Figure 1.

Figure 2 shows the effect of composite density on the thermal conductivity. The high porosity had a dual effect on the thermal conductivity: It reduced the thermal diffusivity and it reduced the density.

Increased composite thickness resulted in reduced composite density. In the case of 10 mm thick composites, it was difficult to achieve at least 2.0 g/cm^3 composite density. In order to alleviate this problem, a T-300 1K 8HS carbon fabric was used as the precursor. The rationale for this approach was based on the very extensive data base with C-C composites [7].

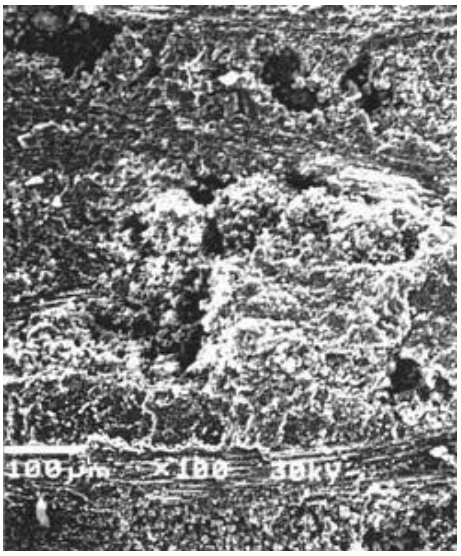

Figure 1. Cross-section of SiC-SiC composites made with T-300 1K plain weave fabric: 4 mm thick.

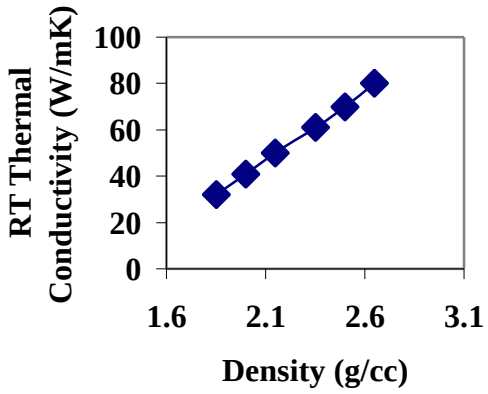

Figure 2. Effect of composite density on thermal conductivity.

3.2 Fiber properties.

Figure 3 shows the single fiber tensile strength values for single fibers extracted from T-300 1K 8HS fabric. A strength of about 1.5 GPa was attained for about 90% CVR-SiC converted fibers. A 100% CVR-SiC converted fiber was obtained, but the tensile strength was difficult to assess (increased stiffness). The issue of residual carbon-carbon with respect to irradiation resistance is addressed in a Jupiter experiment.

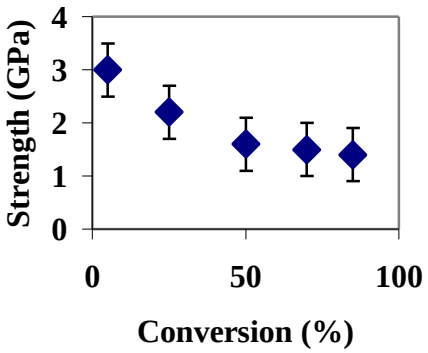

Figure 3. Single fiber strength versus SiC conversion.

The tensile strength of these fibers needs to be assessed on the basis of their heat treatment history. The 1.5 GPa tensile strength corresponds to a heat treatment in excess of 1700°C. Figure 4 shows the corresponding SEM micrographs (90% SiC conversion). Minimum cracking is observed at this conversion level. It is worth mentioning that for this heat treatment temperature, Nicalon S type fiber would exhibit a similar strength [8].

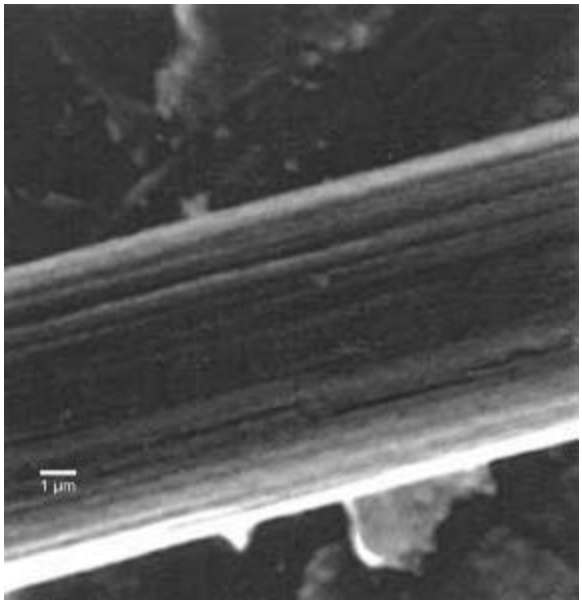

Figure 4. SEM micrograph of 90% CVR-SiC fibers.

Figure 5 shows a TEM micrograph of CVR-SiC fiber (SiC/carbon) interface. About 0.25 – 1.0 µm crystallites are observed, combined with a minimum amount of porosity. The relatively large grain size of the CVR-SiC fiber possibly accounts for its good creep resistance [3].

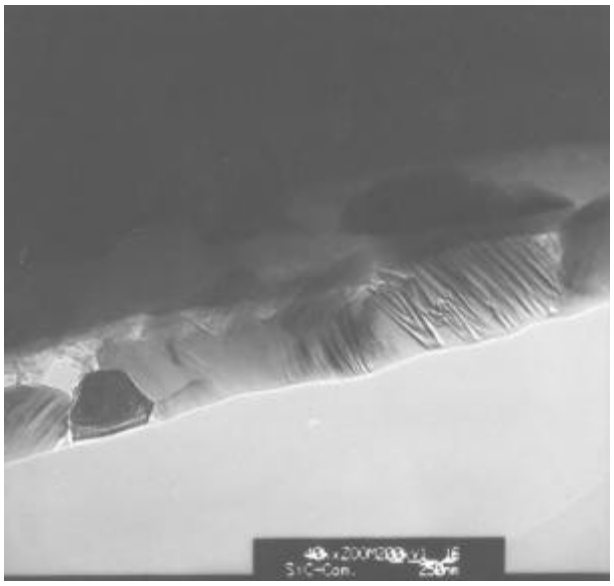

Figure 5. TEM of CVR-SiC fiber.

3.3 Composite properties with T-300 1K 8HS precursor.

The CVR-SiC/CVR-SiC composite, 4 mm thick had a RT through-the-thickness thermal conductivity of 75 W/mK, Figure 6. This represents almost an 7-fold increase over the state-of-the-art SiC-SiC composite thermal conductivity. Figure 6 also shows the thermal conductivity of the CVR-SiC/CVR-SiC composite as a function of temperature. The thermal conductivity experiences a parabolic behavior reaching 35 W/mK at 1000^{o}C. The importance of this result can be further emphasized by comparing it to the 6 W/mK, 1000^{o}C thermal conductivity exhibited by the state-of-the-art CVI-matrix SiC-SiC composite. The parabolic decay of the CVR-SiC/CVR-SiC coupon's thermal conductivity is typical of the higher thermal conductivity matrix.

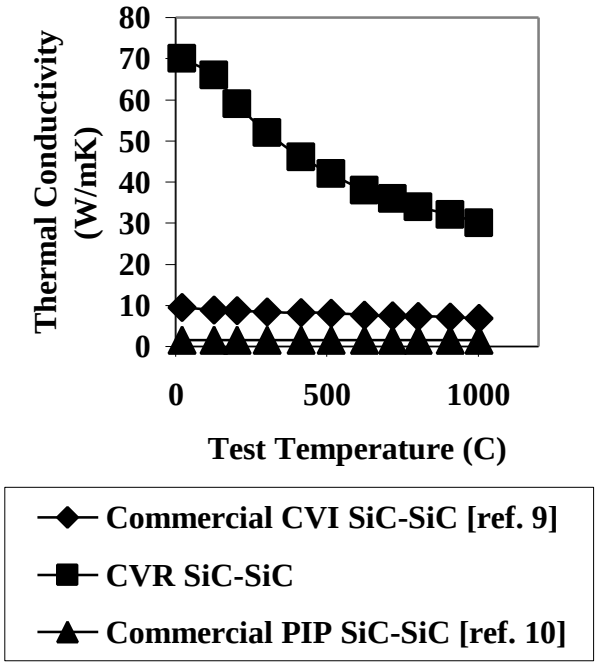

Figure 6. Thermal conductivity of CVR-SiC/CVR-SiC composite.

Figure 7 shows a cross section of the CVR-SiC/CVR-SiC composite (4 mm thick). No transverse cracking or delamination is observed. Very good fiber bundle penetration is achieved. Thus, the use of T-300 1K 8HS precursor alleviated the problems associated with the plain weave T-300 1K precursor.

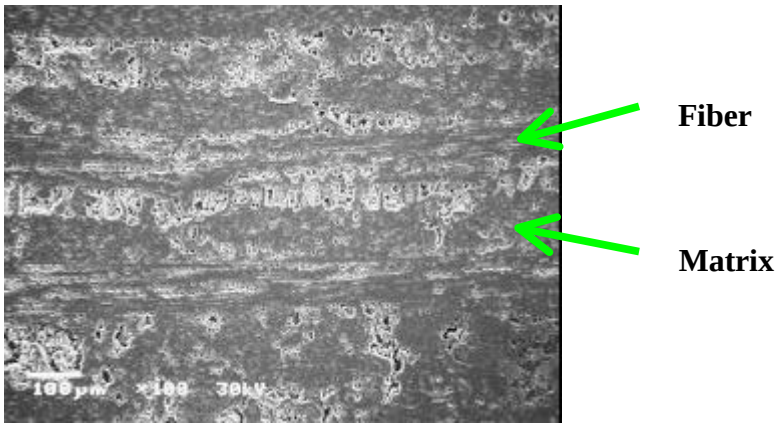

Figure 7. Cross-section of CVR-SiC/CVR-SiC composite.

Figure 8 shows a stress-strain curve of CVR-SiC/CVR-SiC composite. The proportional limit occurs at about 150 MPa (similar to the state-of-the-art SiC-SiC composites made with CVI-SiC

matrix and CG Nicalon and Hi-Nicalon fibers, respectively). The flexural strength of 200 MPa is observed combined with a non-brittle fracture. The flexural strength of the CVR-SiC/CVR-SiC composite is lower, however it is not yet fully optimized, than that of the CVI-matrix composites (300 MPa and 350 MPa for CG Nicalon and Hi-Nicalon, respectively). However, the CVR-SiC/CVR-SiC composite was made with no interfacial coating.

The non-brittle behavior of CVR-SiC/CVR-SiC composites is controlled by fiber-matrix interactions during the composite processing. At the carbon matrix stage, the SiC-C composite can be made very ductile via the control of the processing conditions.

At the CVR-SiC/CVR-SiC stage, the fiber matrix interactions are also controlled by mechanical bonding and interfacial porosity created during the CVR conversion. Thus, a porous SiC interfacial layer can be created in-situ. In addition, there should be no strong bonding between the SiC porous interfacial layer and the CVR fiber (only mechanical interaction is envisioned). Thus, the CVR-SiC process is envisioned to generate a porous SiC interfacial layer poorly bonded to the CVR-SiC fibers. The combination of these two factors can possibly explain the non-brittle behavior of the CVR-SiC/CVR-SiC composite.

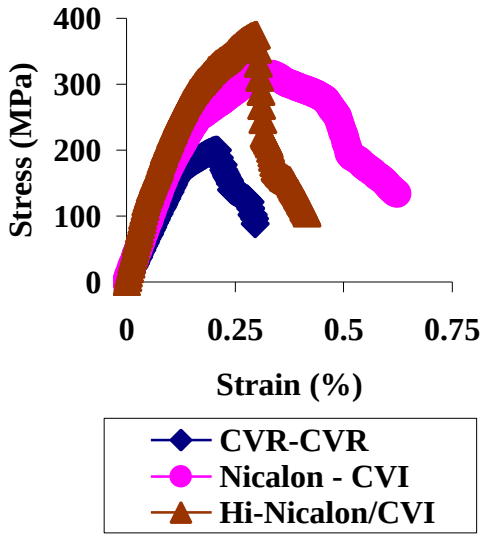

Figure 8. Stress-strain curve for CVR-SiC/CVR-SiC composite.

Table I shows the thermomechanical properties of different SiC-SiC composites. It illustrates a clear improvement in through-the-thickness thermal conductivity of CVR-SiC fiber based composites over Hi-Nicalon based composites. In addition, CVR-SiC derived fiber offers greatly reduced cost over Hi-Nicalon SiC fiber, and CVR-based composite processing greatly reduces the composite processing cost (it is a one-step gas-based processing).

TABLE I. Properties of SiC-SiC Composites

Type	Density (g/cc)	Ther. Cond. RT (W/mK)	Ther. Cond. 1000 ℃ (W/mK)	Flexural Strength RT (MPa)	Open Porosity (%)
Hi Nicalon/CVI	2.55	13	10	350	15
(PIP/CVI hybrid)	2.55	86	28	350	8
(CVR-CVR)	2.65	75	35	250	15

4. CONCLUSIONS

1. Thick CVR-SiC/CVR-SiC composites exhibit excellent thermal properties (70 W/mK at RT and 35 W/mK at 1000^{o}C).
2. CVR-SiC/CVR-SiC composites exhibit good mechanical properties (200 MPa flexural strength, non-brittle behavior).
3. CVR-SiC/CVR-SiC composites offer excellent cost reduction potential.

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