FABRICATION OF SIC/SIC CERAMIC COMPOSITES USING ADVANCED FIBERS – N.L. Vaughn, L.L Snead, R.A. Lowden (Oak Ridge National Laboratory); A. Kohyama and Y. Katoh (Kyoto University, Japan); J.L. Bailey, J.J. Henry, and A.M. Williams (Oak Ridge National Laboratory)

OBJECTIVE

This paper presents a summary of the development of fiber-reinforced ceramic composites using a SiC matrix derived by forced-flow chemical vapor infiltration SiC and near stoichiometric SiC-based fibers.

SUMMARY

FCVI SiC/SiC composites are being fabricated and will be incorporated in the upcoming 14J experiment in conjunction with the Jupiter and Crest Projects. Three different fiber compositions, three different fiber architectures, and two different interface thicknesses are being evaluated simultaneously. All of the fibrous preforms received a pyrolytic carbon surface coating between the fiber and matrix interface. Many of the non-irradiated samples have been characterized and trends examined.

PROGRESS AND STATUS

Experimental Details

Evaluation of three different fiber types and architectures has been investigated (see Table 1), and an initial comparison of their properties has been completed. Initial comparisons of the Hi-Nicalon™, Hi-Nicalon™ Type-S, and Tyranno™ SA fiber composites will be used to determine the next phase of study. Composites fabricated from satin weave fabric were layed up in a 0-90° orientation. Composites made from plain weave fabric were layed up in a 0±30° orientation. The bottom third of each composite was buffered using ceramic grade Nicalon to minimize waste during the initial densification attempts. Due to the satin weave architecture a volumetric fiber loading of approximately 35% was the maximum. Once the preforms were made, each received a pyrolytic carbon coating derived from decomposition of propylene at 1100°C at a pressure of 40 torr. The resulting average calculated interphase coating thickness was between 150 to 200nm. Each fibrous preform was then infiltrated/densified with a SiC matrix using forced flow chemical vapor infiltration (FCVI) from decomposition of methyltrichlorosilane. The infiltration run times varied from 11 to 17 hours at 1200°C with ambient pressure.

Table 1: Fiber Types and Respective Architectures Examined to Date

Fiber Type	Chopped	Plain Weave	Satin Weave
CG-Nicalon	●	●	
Hi-Nicalon		●	●
Hi-Nicalon Type S		●	●
Tyranno SA		●	●

Once densified the composites were removed from their graphite holders and cut into flexure bars (30 x 6 x 2.2mm) according to the 14J irradiation capsule bend bar geometry. The samples were then weighed and measured and broken using four-point bending with an upper span of 10mm and a lower span of 20mm. Using the failure loads and dimensions the ultimate stress and matrix microcrack stress of each sample was calculated and an average determined for each composite.

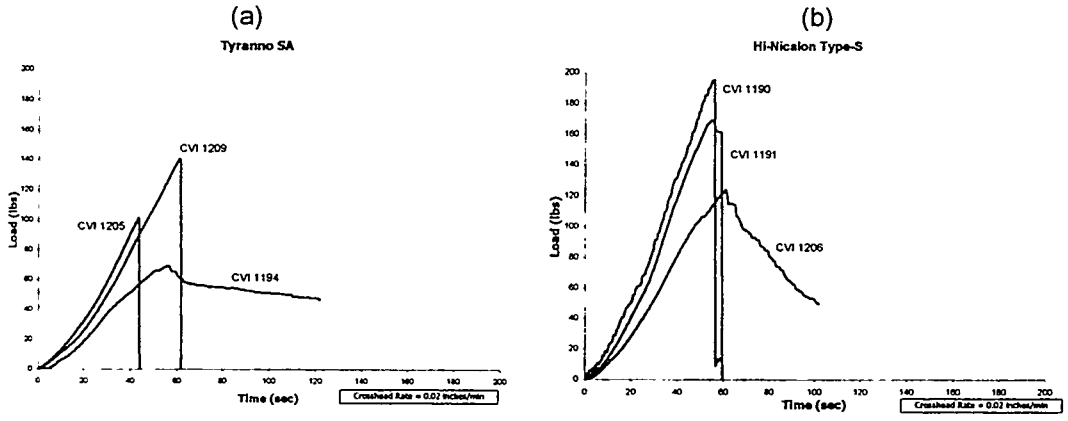
Some difficulty was experienced in fully densifying the Tyranno SA fabric, especially in the satin weave. Aforementioned, the buffer layer was removed to improve infiltration, but showed minimal improvement in the Tyranno SA fabric. Thus the run conditions were altered from the standard conditions.

All of the samples described in this report have yet to be irradiated. Composite samples fabricated from all three fiber types will be incorporated into the upcoming 14J experiment in the HIFR at the Oak Ridge National Lab and planned lower dose rabbit irradiations. Further information will be available after such time. This report covers only non-irradiated samples.

Results

Twelve flexure bars were cut from each sample and six were characterized to determine as-fabricated properties. The remaining six bars from each sample will be incorporated into radiation testing or other characterization procedures. The results from flexure testing can be seen in Figure 2 and Table 2. The dimensions of the flexure bars were 30 x 6 x 2.2mm. The samples were broken using four point bending with an upper span of 10mm and a lower span of 20mm. All testing was done at room temperature.

Figure 1: Representative Load Displacement curves for the three composites of differing fiber type a) Tyranno SA fiber reinforced; b) Hi-Nicalon Type-S fiber reinforced; c) Regular Hi-Nicalon fiber reinforced



(c)

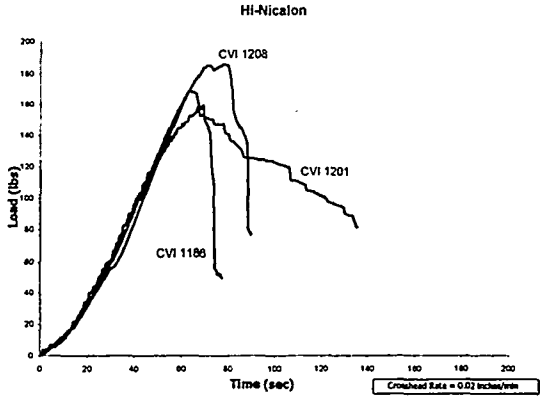


Table 2: Comparison of Average Strengths and Theoretical Densities (8 tests each); error represents one standard deviation

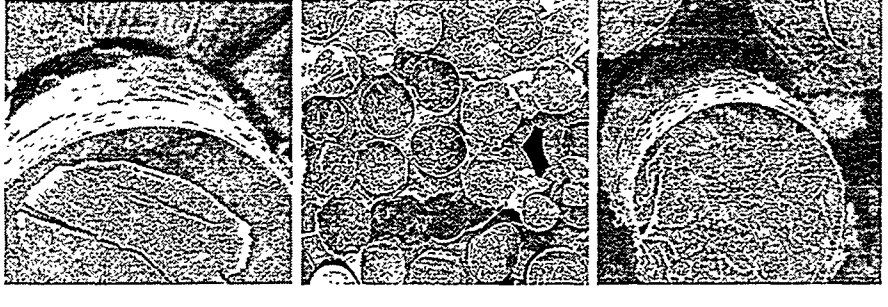
Fiber Type	*UTS (MPa)*	*MM σ (MPa)*	*Theor. Density (%)*
Tyranno SA	202±62	211±51	78±2%
Hi-Nicalon	367±56	342±67	85±4%
Hi-Nic Type S	352±50	327±44	84±4%

Discussion of Results

Comparing ultimate bend strength and matrix microcracking strength of each composite, it was determined that the composites made using both plain and satin weave Hi-Nicalon were the best performers of the initial composites compared. Though the Type-S and Tyranno composites did not return as high strengths as the regular Hi-Nicalon, their strengths were encouraging and they demonstrated great potential for the applications being considered. The lesser performance of the Type-S composite may be ascribed to the thinner than planned interphase coating observed through SEM of the fracture surface (see Figure 2). This most likely contributed to the brittle behavior exhibited by the Type-S composites as seen in Figure 1.

Figure 2: SEM of fracture surface from CVI 1206 a Type-S fiber composite a) fiber-matrix interface; b) brittle fracture surface; c) different fiber-matrix interface

(a) (b) (c)



Initially the Tyranno SA fiber composites seemed inferior to the Hi-Nicalon family of composites. But after closer examination the poor performance of the Tyranno composites could be connected to the inability to properly densify the composites. Multiple factors led to the overall low density of the Tyranno composites. The fiber architecture was the first suspect. The bundles of fibers in all three composites densified well, however none of the composites had good inter-layer densification. This was most evident in the Tyranno fiber composites. Large voids between layers were created due to very rapid densification. Such voids between the layers of fabric could have the effect of reduced interlaminar shear and strength properties. Secondly, the choice to buffer the Tyranno composites (to save material) intensified the poorly infiltrated areas within the fabric. This was due to the higher growth rate within the Nicalon fabric ultimately causing sealing off of the gas flow to the Tyranno region. By removing the buffer layer and using the desired fiber types through the entire part, overall properties of the composites were enhanced. Further investigation is recommended on the effects of fiber architecture and layup on the final strength for Tyranno composites.

Figure 3: Cross-section of CVI1201 (Hi-Nicalon Composite), line indicates buffer zone

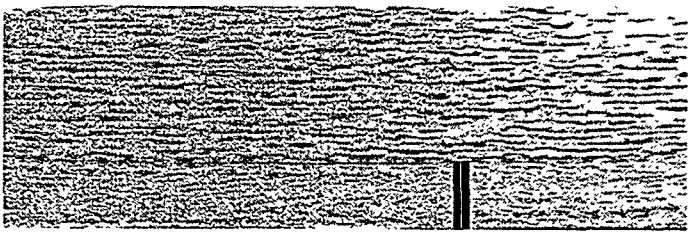


Figure 4: Schematic of Forced Flow Chemical Vapor Infiltration Process

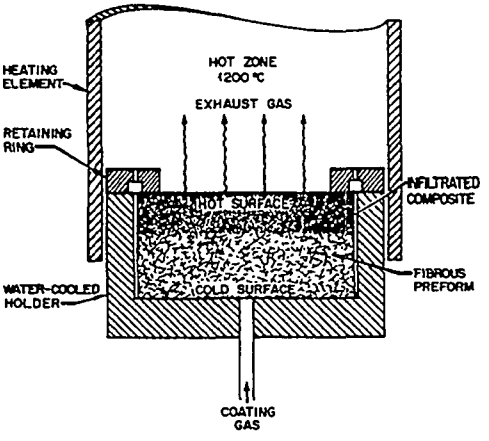
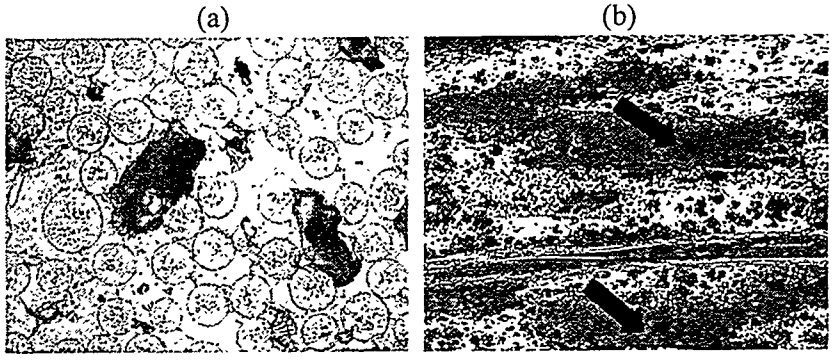


Figure 5: a) 100X picture of Type-S composite fiber bundle; b) 5X picture of Type-S composite showing porosity between fabric layers (arrows indicate pores)

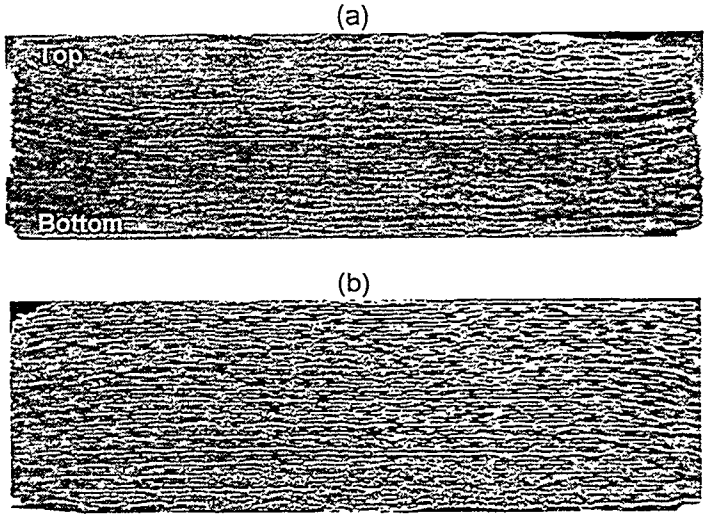


Once the buffer layer was removed, infiltration improved greatly. However, simple layup adjustment did not remedy the inter-layer pores. Therefore, the process conditions were altered. By adjusting the infiltration parameters, slightly better infiltration was achieved both in the fiber bundles and between layers. The parameters were adjusted as seen in Table 2 and improvements were noted immediately (Figure 6). Further development of the FCVI approach to infiltrating these advanced fibers is recommended.

Table 3: *Changes in Run conditions for Tyranno plain weave composite.*

Infiltration Parameter	Old Conditions	New Conditions
Surface Temperature (°C)	1200°C	1150°C
MTS Flow Rate (g/min)	0.300	0.250
Hydrogen Flow Rate (sccm)	500	500

Figure 6: Cross-section of CVI1205 and CVI1209 respectively – Tyranno Composite with No Buffer fabricated using a) old run conditions and b) new run conditions



CONCLUSIONS

The advanced fibers performed well when used to replace current lower-grade ceramic fibers in regards to matrix microcracking and ultimate bend strength. Initially, no processing conditions were altered to accommodate the advanced fibers though parameters such as thermal conductivity, fiber diameter and fabric weave need to be considered for a more thorough evaluation of these materials. Ordinarily these factors and others are considered to predict the optimum deposit kinetics during infiltration. By incorporating factors such as fabric architecture and fiber surface roughness, further property improvements will be achieved. Ultimate bend strengths near the 400 MPa range indicate great promise for the use of these new fibers. Though low theoretical density of the composites contributed to inferior properties, further development of the infiltration process should further improve properties. It is recommended that alternate forms of infiltration be investigated alongside FCVI, such as liquid-phase infiltration. Additionally, development of new and current interface coatings is an essential part of the development of these fibers for ceramic composites. Each fiber type reacted differently to the pyrolytic carbon coating, therefore coatings may need to be derived specifically for a given fiber type and architecture. Though the Tyranno fibers did not perform exceptionally during initial studies, past research indicates good results from both Nicalon and Tyranno fibers as preforms for FCVI. Further development of SiC/SiC composites using Type-S, Tyranno, and other advanced fibers is therefore recommended.