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Processing and Properties of SiC/$MoSi_2$-SiC Composites Fabricated by Melt Infiltration

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PROCESSING AND PROPERTIES OF SiC/$MoSi_2$-SiC COMPOSITES FABRICATED BY MELT INFILTRATION

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ABSTRACT

Hi-Nicalon SiC fiber reinforced $MoSi_2$-SiC matrix composites (SiC/$MoSi_2$-SiC) have been fabricated by the melt infiltration approach. The composite consists of ~60 vol%, 2-D woven BN/SiC coated Hi-Nicalon SiC fibers and ~40 vol% $MoSi_2$-SiC matrix. The room temperature tensile properties and thermal conductivity of the SiC/$MoSi_2$-SiC composites were measured and compared with those of the melt infiltrated SiC/SiC composites. The influence of fiber architecture on tensile properties was also evaluated. Results indicate that the primary modulus, stress corresponding to deviation from linearity, and transverse thermal conductivity values for the SiC/$MoSi_2$-SiC composites are significantly lower than those for the SiC/SiC composites. Microcracking of the matrix due to the large difference in thermal expansion between $MoSi_2$ and SiC appears to be the reason for the lower matrix dominated properties of SiC/$MoSi_2$-SiC composites.

INTRODUCTION

Advanced high temperature materials are needed for successfully developing future generations of aerospace propulsion and power systems. Because of its high melting point, elevated temperature oxidation resistance, brittle to ductile transition, and high electrical and thermal conductivity, molybdenum silicide ($MoSi_2$) is considered to be a promising high temperature structural material for next generation turbine components [1]. However, the use of $MoSi_2$ has been hindered due to the brittle nature of the material at low temperatures, inadequate

creep resistance at high temperatures and poor oxidation resistance from 450 to 550 ^{0}C [1]. In recent years significant progress has been made in improving the properties of $MoSi_2$. For example, creep resistance has been improved by alloying $MoSi_2$ with W or Re [2,3]; the low temperature toughness problem has been improved by particulate or fiber reinforcements [4-6]; and the intermediate temperature pest oxidation problem has been mitigated by dispersions of Si_3N_4 or Ge [7,8].

Currently, unreinforced and reinforced $MoSi_2$ materials are fabricated by hot pressing or hot isostatically pressing. With these methods only simple shapes can be fabricated. Even though $MoSi_2$ is electrically conductive and can be elecro-discharge machined, fabrication of complex shape components from a hot pressed block is very expensive and time consuming. Therefore, cost effective, near net shape processing approaches are needed for utilization of $MoSi_2$ components in turbines. Among many fabrication approaches, the non-reactive melt infiltration approach appears to be best suited for $MoSi_2$. This approach has been successfully adopted for fabrication of SiC/SiC composites. One draw back of this process is that it leaves excess silicon metal, which may limit the upper temperature capability of the material. However, excess silicon can be converted to $MoSi_2$ by displacement or combination reaction by selecting Mo_2C or Mo_5Si_3 as a starting material [9].

This study had three objectives: First, to fabricate SiC/$MoSi_2$-SiC composites by the MI approach; second, to measure their properties and compare them with those of the SiC/SiC composites fabricated by a similar approach; third, to determine potential performance advantages of 2-D woven SiC/$MoSi_2$-SiC composites for high temperature applications.

EXPERIMENTAL

The starting materials for composite fabrication were BN/SiC coated Hi-Nicalon^{TM} SiC fiber preforms, $MoSi_2$ and SiC powders. The preforms of BN/SiC coated Hi-Nicalon^{TM} SiC fibers were purchased from Honeywell Advanced Composites Inc. The Nippon Carbon Company, Japan fabricated the Hi-Nicalon fibers and Albany International Techniweave, Inc. Rochester, prepared the woven fiber mats. Three different 2-D fiber architectures were examined: PW (plain weave), 5HS (5 harness satin), and 8HS. The $MoSi_2$ and SiC powders with average particle diameters of ~75 and ~3 μm, respectively, were purchased from H.C. Stark. For composite fabrication, the preforms were fabricated by stacking 8 layers 2-D woven SiC fiber mat in a graphite fixture and depositing an ~0.5 μm thick BN layer and a 3-5 μm thick SiC layer (both by chemical vapor infiltration (CVI)). The nominal dimensions of the preforms after CVI coating were 229-mm (L) x 152-mm (W) x 2-mm (T). The preforms contained

~ 40 vol. % SiC fibers, ~18-30 vol% BN/SiC interface coating, and 20-40% interconnected open porosity.

The preforms were cut into smaller specimens of dimensions with 152 mm (L) x 12.5 mm (W) x 2 mm (T)) using a diamond impregnated metal bonded cutoff wheel and water based grinding fluid. The cut specimens were soaked in a degreasing solution for 24 hrs, and then dried in an oven maintained at 100 ^{0}C.

The $MoSi_2$ and SiC powders were dry attrition milled in a stainless steel vessel using 6-mm WC balls as grinding medium. The outer surface of the vessel was cooled with liquid nitrogen. A typical batch contained a 50:50 mixture of $MoSi_2$ and SiC powders. A grinding medium to powder ratio of 20:1 was used. Attrition milling was performed for 16 hrs in flowing argon gas. The milled powder was stored in a glove box filled with nitrogen. The average particle diameter after attrition milling was ~ 5 μm.

For slurry preparation, the attrition milled $MoSi_2$ and SiC powder mixtures were blended with a commercially available binder, dispersant, and wetting agent. NH_4OH was added to the slurry to adjust p_H to ~9.

Composite fabrication

Slurry infiltration into the preform specimen was conducted in a stainless steel (SS) chamber equipped with three ports. One port was connected to a forepump for evacuating the chamber; one port was used for poring slurry into the chamber; and one was connected to a compressed air source for pressurizing the chamber. A plaster of Paris mold with a cavity was fabricated. The mold, with the specimen, was placed inside the SS chamber. The chamber was evacuated to 10^{-4} Torr and then the port connected to the vacuum pump was closed off. Through the second port, the slurry was poured over the preform placed on the plaster of Paris mold and the port was closed off. Through the third port, compressed air was bled into the SS chamber until chamber pressure reached 3 MPa. The air pressure was maintained for several hours until the slurry infiltrated into the preform, and the water present in the slurry had completely seeped into the plaster of Paris mold. Subsequently, any excess powder present on the surface of the preform specimen was dusted off. The $MoSi_2$-SiC particle-filled specimen was transferred to a vacuum furnace lined with graphite heating elements and fixtures. An appropriate amount of electronic grade silicon was placed over the specimen. The furnace was evacuated to 10^{-4} Torr, heated to 1420 ^{0}C for 30 minutes allowing the silicon melt to infiltrate the particle filled specimen, and then cooled slowly to room temperature.

Specimen preparation

Some of the specimens were sectioned, mounted in a metallographic mold, ground successively on 40 μm down to 1-μm diamond particle impregnated metal disks, and polished in a vibratory polisher on a micro-cloth using 0.3 μm diamond powder paste. The mounted specimens were coated with a thin layer of carbon or palladium in a vacuum evaporator to avoid charging during observation in a scanning electron microscope (SEM).

For tensile testing, dog-bone shaped specimens were machined from the composite plate using an ultrasonic, SiC-slurry impact machine. At each specimen end, two glass fiber-reinforced epoxy tabs of dimension 37 mm x 12 mm x 1 mm were bonded, leaving ~60 mm for the gage section. A spring-loaded clip-on gage was attached to the gage section of the specimen to monitor the strain during the tensile test. The specimens were tested at room temperature until failure in a servo-controlled tensile testing machine equipped with self-aligning grips at a crosshead speed of 1.3 mm/min.

For comparison purposes, a panel of Hi-Nicalon SiC/SiC composite fabricated by the MI process was purchased from Honeywell Advanced Composites Inc. This composite was tested using the same approach that was used with the Hi-Nicalon SiC/$MoSi_2$-SiC composites.

For thermal diffusivity measurement, ~9-mm × 9-mm specimens were cut from a larger composite specimen. Thermophysics Laboratory, Blacksburg, VA, measured the transverse thermal diffusivity of the Hi-Nicalon SiC/$MoSi_2$-SiC and SiC/SiC composite specimens in argon at temperatures from 25 to 1400 ^{0}C.

RESULTS

Microstructure

SEM photographs of the cross-section of the Hi-Nicalon SiC/$MoSi_2$-SiC composites are shown in Fig. 1. This figure indicates the matrix is well infiltrated in the pore spaces of the SiC preform. The light regions in figure 1(a) are the $MoSi_2$-SiC matrix and the darker regions are the BN/SiC coated Hi-Nicalon woven mats. Figures 1(b) and 1(c) are higher magnification photographs of the matrix and interface regions. The brighter particles in Fig. 1(b) are $MoSi_2$ and the dark particles are SiC. The phase surrounding the $MoSi_2$ and SiC particles is silicon. Figure 1(c) shows no evidence of reaction between the CVI SiC coating and the matrix.

Tensile properties

The room-temperature tensile stress-strain behavior for the 5HS Hi-Nicalon SiC/SiC and SiC/$MoSi_2$-SiC composites is shown in figure 2. Also included in the

figure for comparison is the stress-strain behavior of 5HS Hi-Nicalon SiC/Epoxy composite. The stress-strain behaviors generally show two regions: an initial elastic region and a non-linear region. Comparison of the stress-strain behaviors indicates major differences in the stress corresponding to deviation

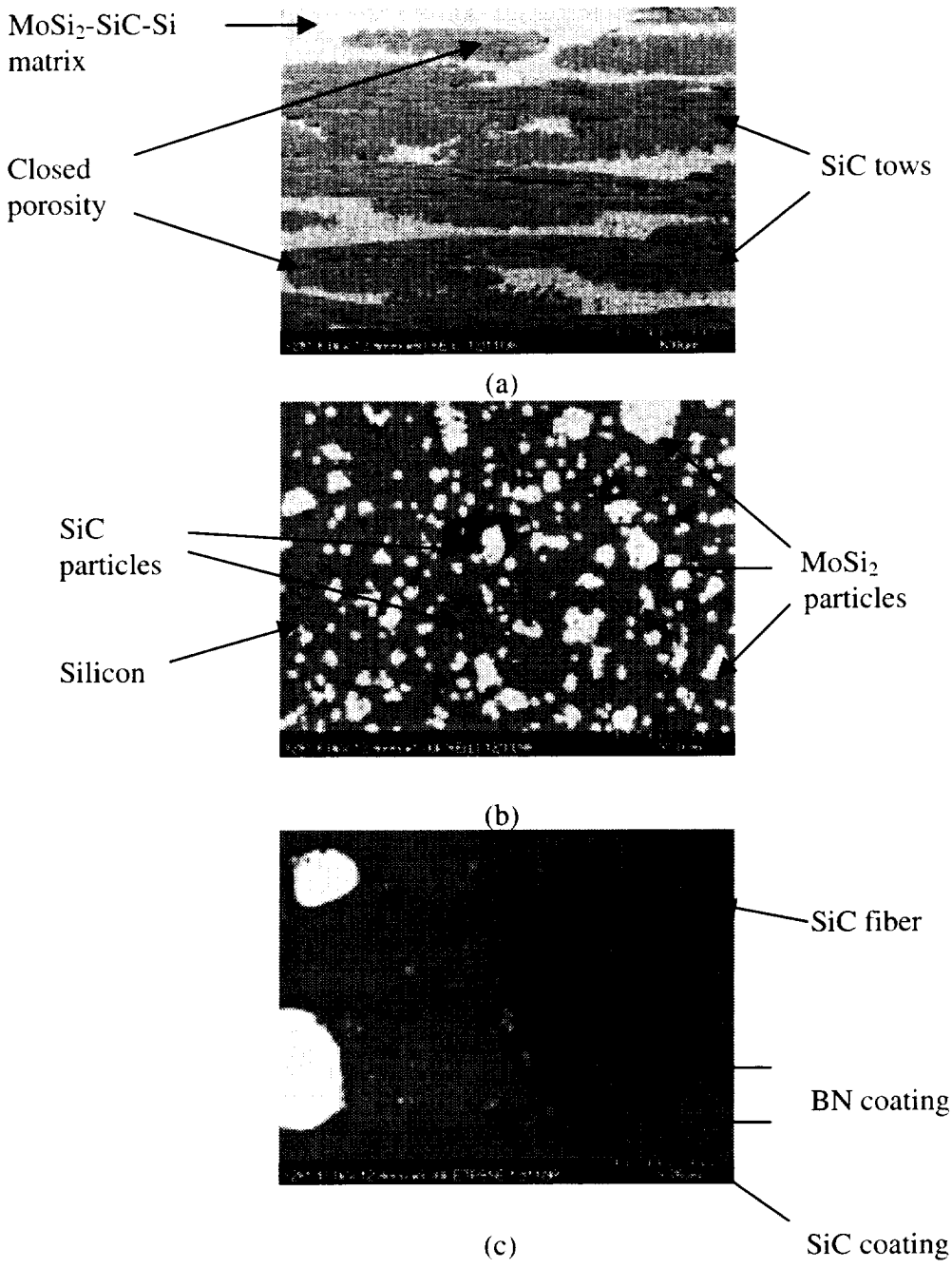


Fig.1 SEM micrographs of the cross section of a 5HS Hi-Nicalon SiC/$MoSi_2$-SiC composite showing various microstructural features: (a) distribution of the matrix within the preform, (b) distribution of particles in the matrix, and (c) the CVI SiC coating/matrix interface region.

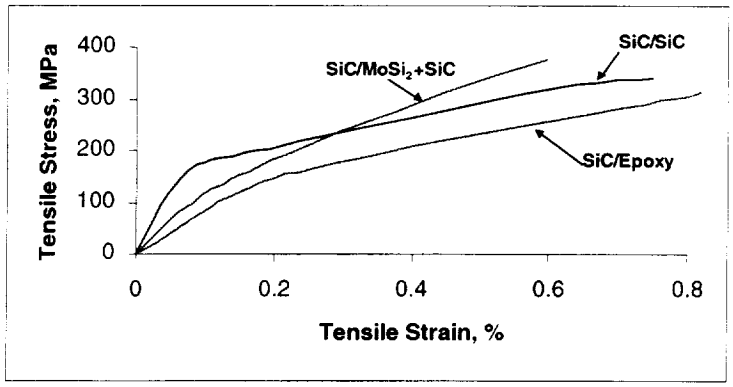


Fig. 2 Room temperature tensile stress-strain behavior for the 5HS Hi-Nicalon SiC/$MoSi_2$-SiC, SiC/SiC, and SiC/epoxy composites

from linearity (DFL) and the primary elastic modulus, but the ultimate tensile strengths for all three composites are nearly the same suggesting minimal degradation of SiC fibers, or the BN/SiC interface coatings during processing. The room-temperature tensile property data for the PW, 5HS, and 8HS Hi-Nicalon SiC/$MoSi_2$-SiC, 5HS SiC/SiC and 5HS SiC/epoxy composites are summarized in Table I. Data represent an average of 3 specimens for each composite type. Noticeable features in Table I are that the fiber architecture shows no major effect on the tensile properties of SiC/$MoSi_2$-SiC composites. Also, the primary modulus and DFL stress for SiC/$MoSi_2$-SiC composites are moderately higher than that for the Nicalon SiC/epoxy composites, but lower than that for the SiC/SiC composites. Theoretically, the initial elastic modulus of SiC/$MoSi_2$-SiC composites and SiC/SiC composites should be the same because $MoSi_2$ and SiC materials have similar elastic moduli. It is suspected that the lower primary elastic

Table I Room temperature tensile data for Hi-Nicalon SiC/Epoxy, SiC/$MoSi_2$-SiC, and SiC/SiC composites.

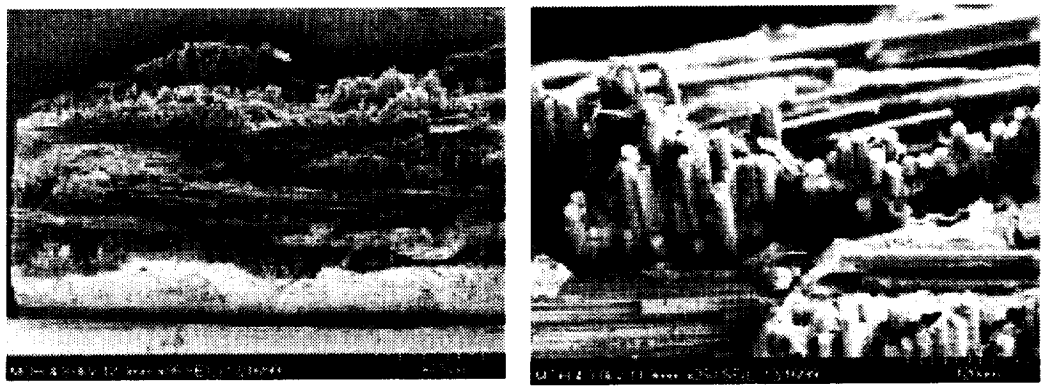
Composites	Lay up	DFL, MPa	DFL, %	E, GPa	UTS, MPa	UTS, %
SiC/Epoxy	5HS	66±4	0.06±0.015	116±21	328±30	0.7±0.07
SiC/$MoSi_2$-SiC	PW	37±4	0.03±0.004	131±7	264±20	0.7±0.07
SiC/$MoSi_2$-SiC	5HS	62±26	0.05±0.021	125±24	329±52	0.5±0.09
SiC/$MoSi_2$-SiC	8HS	49±3	0.03±0.003	160±3	304±8	0.8±0.04
SiC/SiC	5HS	105±12	0.05±0.01	215±18	340±26	0.8±0.05

modulus of SiC/$MoSi_2$-SiC composites may be due to microcracking of the matrix due to large difference in thermal expansion between $MoSi_2$ and SiC particles.

The SEM photographs of the fracture surfaces of SiC/$MoSi_2$-SiC composites are shown in Fig. 3. The fracture surface is jagged with limited fiber pull out.

Thermal conductivity

The variation of transverse thermal conductivity with temperature in argon is plotted in Fig. 4 for Hi-Nicalon SiC/$MoSi_2$-SiC and SiC/SiC composites. At room temperature, the transverse thermal conductivity for SiC/SiC composites is



(a) (b)

Fig. 3 SEM photographs of the tensile fracture surface of the 5HS Hi-Nicalon SiC/$MoSi_2$-SiC composites.

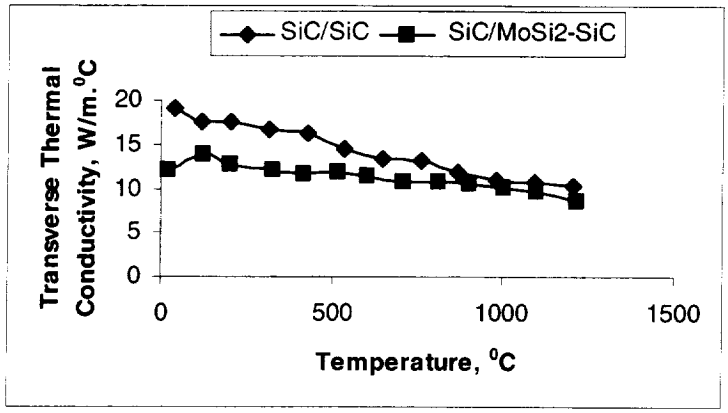


Fig. 4 Variation of transverse thermal conductivity with temperature for the Hi-Nicalon SiC/$MoSi_2$-SiC composites and SiC/SiC composites in argon.

higher than that for the SiC/$MoSi_2$-SiC composites. As test temperature increases, the difference between the thermal conductivity of these two materials decreases, and beyond 1100^0C, the thermal conductivity of both materials is nearly the same.

SUMMARY OF RESULTS

Hi-Nicalon SiC fiber reinforced $MoSi_2$-SiC composites have been fabricated by the MI process. The thermal conductivity and room temperature tensile strengths of the SiC/$MoSi_2$-SiC composites have been measured and compared with state-of-the-art SiC/SiC composites fabricated by a similar processing method. Major findings are the following.

(1) Microstructurally, the Hi-Nicalon SiC fiber and the CVI SiC coating are stable during MI processing of the SiC/$MoSi_2$-SiC composites.

(2) Matrix dominated properties such as the primary elastic modulus and DFL stress for the SiC/$MoSi_2$-SiC composites are lower than those for the SiC/SiC composites, possibly due to microcracking of the matrix.

(3) High temperature thermal conductivity of the SiC/$MoSi_2$-SiC composites is similar to that of the SiC/SiC composites.

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13. ABSTRACT ***(Maximum 200 words)***

Hi-Nicalon SiC fiber reinforced $MoSi_2$-SiC matrix composites (SiC/$MoSi_2$-SiC) have been fabricated by the melt infiltration approach. The composite consists of ~60 vol%, 2-D woven BN/SiC coated Hi-Nicalon SiC fibers and ~40 vol% $MoSi_2$-SiC matrix. The room temperature tensile properties and thermal conductivity of the SiC/$MoSi_2$-SiC composites were measured and compared with those of the melt infiltrated SiC/SiC composites. The influence of fiber architecture on tensile properties was also evaluated. Results indicate that the primary modulus, stress corresponding to deviation from linearity, and transverse thermal conductivity values for the SiC/$MoSi_2$-SiC composites are significantly lower than those for the SiC/SiC composites. Microcracking of the matrix due to the large difference in thermal expansion between $MoSi_2$ and SiC appears to be the reason for the lower matrix dominated properties of SiC/$MoSi_2$-SiC composites.

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