SLURRY MOLDING OF CARBON-CARBON COMPOSITES AND THEIR APPLICATIONS

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Introduction

The increasing use of advanced solid state devices which have extremely high heat loads is requiring the development of passive thermal management substrates. In avionics applications, since the payload weight must be minimized, designers are constantly seeking low density materials to serve as both structural members and thermal management components. However, traditional materials such as copper and aluminum have a low specific thermal conductivity (κ/ρ). Carbon-carbon composites, on the other hand, offer several advantages, including high specific thermal conductivity, low density, tailorability, and excellent thermal shock resistance. However, traditional carbon-carbon processing (CVI or HIPIC) can take several months to complete, yielding expensive components [1, 2].

Recently an alternative processing method for carbon-carbon composites which employs slurry molding to reduce time and costs has been explored. The development of the material and process are reported here.

Experimental

Carbon-carbon composite preforms were made using the slurry molding technique illustrated in Fig. 1. Chopped or milled carbon fibers are dispersed in water with powdered Mitsubishi ARA mesophase pitch resin at a ratio of 1:1.5 by weight. Both 6.35-mm Amoco P-55 mesophase pitch-based carbon fibers (with AP-200 sizing) and 200µm Amoco ThermalGraph DKD-x high thermal conductivity fibers (no sizing) were evaluated to examine effects of fiber properties and length on processability and thermophysical properties of the finished composites. In order to improve strength, 25mm Fortafil 3(c) PAN-based carbon fibers were added in small quantities to each mixture. The specific recipes for the samples are listed in Table I. The slurry was then vacuum molded over a sieve screen into the form of a cylinder 114-mm dia. x 127-mm thick and dried in air at 50°C. The “greenform” was compression molded at 650°C, 11 MPa, with a heating rate of 0.1°C/min. The molded part was carbonized under flowing nitrogen at 5°C/min to 1050°C.

A pitch vacuum impregnation/carbonization (PVIC) cycle was performed on selected samples. These parts were immersed in molten mesophase pitch at 390°C and 0.1 millibar pressure. Vacuum was applied and released four times over a two hour period. The impregnated parts were then stabilized in air at 220°C for 18 hours and carbonized at 1050°C. All samples were graphitized at 2800°C for 1 hour. Total fabrication time was 8 days.

Results and Discussion

The as-received ARA-24 mesophase pitch bloated significantly upon carbonization at 650°C. To solve this problem, the pitch was heat-treated under vacuum at 400°C for two hours to remove low molecular weight volatiles. The carbon yield increased from 80 to 86% from heat treatment. However, some off gassing occurred, resulting in minor bloating and lower than expected composite densities. When the heat treated pitch was partially stabilized in air at 220°C for 5 hours (~4% weight gain), the parts did not bloat during carbonization, yielding densities of $1.45 \pm .05$ g/cm^3. The stabilization increased the carbon yield from 86 to 89%.

The slurrying technique results in dispersed fibers rather than tows, yielding a transversely isotropic composite (in-plane properties are isotropic but different from the out-of-plane properties). This fine dispersion of individual fibers leads to a very fine microstructure with no intact tows present in the composite. Higher transverse thermal conductivities, due to a more continuous network of the pitch-based matrix, were thus anticipated.

Figure 3 is a graph of the bulk density change during the PVIC cycles. The archemedian density increased to 1.68 g/cm^3 for the samples with P-55 fiber. The samples containing DKD-x fiber did not increase in density during the PVIC cycles. Since these fibers were not sized, poor wetability inhibited the infiltration with mesophase pitch.

Table I shows the thermal conductivity (κ) after graphitization. Densification produced significant improvements in thermal conductivities for the samples with P55 fiber. This is attributable to the increase in volume fraction of pitch derived carbon matrix.

The mechanical properties of the samples after graphitization are given in Table I. Strength and modulus of the composites with P55 fiber improved with density. The composites with DKD-x fibers exhibited low strengths. Again, since these fibers had no sizing, a poor fiber/matrix bond would result, yielding lower strengths.

Conclusion

It has been shown that slurry molding of carbon/carbon offers a fast method for producing transversely isotropic composites with high thermal conductivities. Composites with sized P55 fibers had excellent densities, strengths, and thermal conductivities. In fact, the in-plane thermal conductivity was higher than the as-received fibers, due to both the high volume fraction pitch matrix, and the high heat treatment temperature. Furthermore, the transverse thermal conductivity is comparable to composites made with traditional processes [1, 2].

However, composites fabricated with unsized DKD-x fibers had lower than expected thermophysical properties. Several factors may have contributed to the lower than anticipated properties. The lack of sizing on the DKD-x fibers resulted in a poor fiber matrix bond, causing the composite to exhibit low strength and modulus. The lack of fiber sizing inhibits wetting of the fiber by the pitch, translating into poor impregnation and densification. Last, the aspect ratio (L/D) of the DKD-x fibers was about 20:1; too short to contribute significantly to the mechanical properties of the composites. On the other hand, the P55 fibers have an aspect ratio of about 635:1, which is large enough to enhance the mechanical properties.

It is expected that this new fabrication process can produce composites at a lower cost than traditional CVI or HIPIC process because of the reduced (8 days total) processing time.

References

1. Savage, G. <u>Carbon-Carbon Composites</u>, Chapman & Hall, New York, NY (1993).

2. Murdie, N., C. P. Ju, J. Don, and M. A. Wright, "Carbon-Carbon Matrix Materials," in <u>Carbon-Carbon Materials and Composites</u>, J.D. Buckley (editor), Noyes Publications, New York (1989).

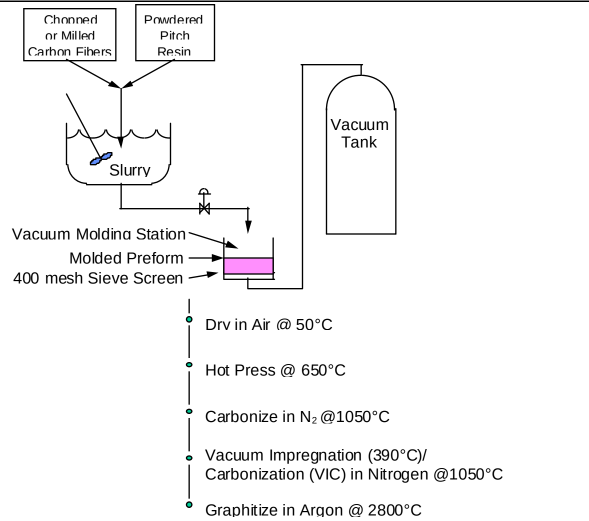

Figure 1. Slurry Molding Process.

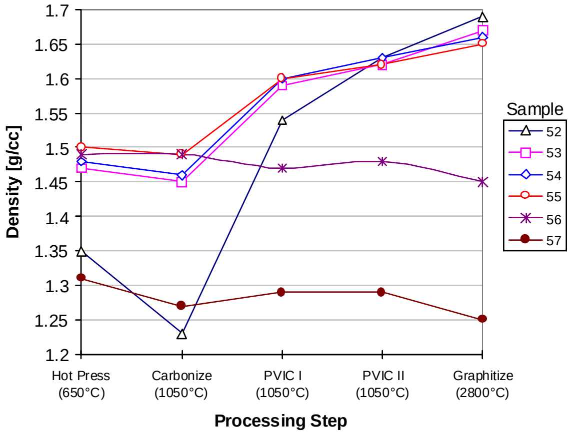

Figure 2. Bulk density changes during processing.

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Table I. Thermophysical properties of slurry molded composites. Each sample was hot-pressed to a total fiber fraction of approximately 38% by volume.

Sample #	Mass PAN Fiber	Pitch Fiber Type	Fiber Thermal Conductivity	Mass Pitch Fiber	Mass Pitch Matrix	Densification	Average Density	κ In-plane //	κ Out-of-plane ⊥	Flexural Strength	Flexural Modulus
	[g]		[W/m·K]	[g]	[g]		[g/cm^3]	[W/m·K]	[W/m·K]	[MPa]	[GPa]
50 - 51	30	P-55	120	70	150	As-Fab.	1.45	55.4	19.3	47.9 ± 14.8	25.7 ± 4.1
52 - 55	30	P-55	120	70	150	PVIC	1.68	131.8	43.2	76.3 ± 8.7	40.4 ± 5.3
56 - 57	30	DKD-x	600	70	150	PVIC	1.35	76.2	13.3	36.2 ± 3.6	19.0 ± 2.1