HIGH THERMAL CONDUCTIVITY, MESOPHASE PITCH-DERIVED CARBON FOAM

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INTRODUCTION

The extraordinary physical properties of carbon fiber result from the unique graphitic morphology of the spun (extruded) filaments [1]. Contemporary advanced structural composites exploit these properties by creating a disconnected network of graphitic filaments held together by a matrix suitable for the application. Carbon foam derived from a pitch precursor, on the other hand, can be considered as an interconnected network of graphitic ligaments and, thus, should exhibit isotropic material properties [2]. The foam represents a potential reinforcing phase for structural composite materials. Because of the continuous graphitic network, the foam-reinforced composites will display higher isotropic thermal conductivities than carbon fiber reinforced composites. Furthermore, the lack of interlaminar regions, which develop in traditional prepregged carbon fiber reinforced composites, should result in enhanced mechanical properties such as shear strength and fracture toughness.

A new, less time consuming process for fabricating pitch-based graphitic foams without the traditional blowing and stabilization steps has been developed. It is believed that this new foam will be less expensive and easier to fabricate than traditional foams. Therefore, it should lead to a significant reduction in the cost of carbon-based thermal management and structural materials (i.e. foam-reinforced plastics and foam core composites).

EXPERIMENTAL

The foam is produced using a proprietary method developed at Oak Ridge National Laboratory in the Carbon and Insulation Materials Technology Group. The process does not utilize a thermodynamic flash (blowing) to produce the foam and, most importantly, the unique method eliminates the requirement to stabilize the foamed pitch prior to carbonization (typically an oxidative stabilization step). The method is fairly versatile and can be easily adjusted to control pore/cell size and density. Mitsubishi ARA24 naphthalene-based synthetic pitch was used at the precursor for the graphitic foam. All foam samples were graphitized at 5°C/min in Argon to 2800°C and soaked for 1 hour.

Samples of the graphitized foam were either vacuum impregnated with an epoxy resin or densified with carbon by chemical vapor infiltration with methane to a final density of 1.3 g/cm³.

RESULTS AND DISCUSSION

Figure 1 is a SEM micrograph of the graphitized foam illustrating that the foam exhibits a predominately open cell structure at a density of 0.54 g/cm³. The mean pore diameter measured with mercury porosimetry was indicated to be 91 microns. However, the actual cell diameter is approximately 250 microns, as seen in Figure 2, resulting in a structure equivalent to 100 pores per inch (ppi). This, along with large ligament thickness contributes to an accessible surface area of 4 m²/g, which is very useful in porous media heat transfer.

The ligaments of the foam appear completely different from vitreous reticulated carbon foams produced commercially; e.g. significantly thicker and exhibiting a spherical morphology. As the foam is produced, the shear stresses from the expansion of the bubbles cause the liquid mesophase crystals to align parallel to the surface of the bubbles. During carbonization and graphitization, the resultant aligned mesophase forms highly ordered graphitic structures parallel to the surface of the bubble and along the axis of the ligaments. This was confirmed with optical microscopy.
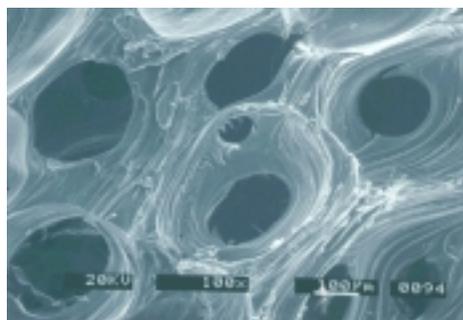
Figure 1. Scanning electron micrograph of foam graphitized at 2800°C.

The 002 x-ray difraction peak was very narrow and asymmetric; indicative of the existence of highly ordered graphitic crystals. Using the Bragg/Scherrer method, the interlayer (d_{002}) spacing was calculated to be 0.3362 nm. This is significantly closer to pure graphite (0.3354 nm) than most high performance carbon fibers, such as Vapor Grown Carbon Fibers (VGCF) and K1100 [3,4]. The crystallite size in the c-direction ($L_{c,002}$) was calculated to be 442 nm, and the crystallite size in the a-direction ($L_{a,100}$) was calculated to be 203 nm. These crystallite sizes are larger than typical high thermal conductivity carbon fibers [3,4,5]. Table 1 is a comparison of X-ray diffraction results and estimated thermal conductivities of various carbon fibers to the graphitic foam. The foam has the lowest d-spacing and the highest $L_{a,100}$.

The compressive strengths of the foam and densified foam are presented in Table 2. As can be seen, the foam exhibits strengths superior to aluminum foams at similar densities. Furthermore, an order of magnitude improvement in strength is achieved through densification of the foam with either an epoxy resin or a graphtic carbon (CVI).

Table 1. X-ray diffraction parameters

Fiber	HTT [°C]	$L_{c(002)}$ [nm]	$L_{a(100)}$ [nm]	d_{002} [nm]	Estimated κ [W/m·K]
P-120[3]	As-received			.3392	640
Clemson Ribbon[5]	2400	323	124	.3380	950
K1100[4]	As-received	85	51	.3366	950
VGCF[3]	2800			.3366	1950
Foam Ligament	**2800**	**203**	**442**	**.3362**	**>1000**

Table 3 presents the thermal properties of the foam versus other thermal management materials. While the thermal conductivity of the foam is not as high as copper, the specific thermal conductivity is over four times greater than copper and three times greater than aluminum. Also, the thermal conductivity and specific thermal conductivity of the graphite foam is significantly higher than aluminum foams with similar densities. Although the thermal conductivity of the graphite foam is not as high as a typical 2-D cloth, mesophase pitch densified carbon-carbon composite (C/C), the specific conductivity of the foam is higher. Most importantly, the out-of-plane conductivity of the foam is significantly higher than the carbon-carbon composite.

Table 2. Foam compressive strengths

Material	Specific Gravity	Compressive Strength MPa	Modulus GPa
Foam	**0.54**	**3.4**	**0.180**
Foam/Epoxy	**1.26**	**34.3**	**0.560**
Foam/Carbon CVI	**1.3**	**31.6**	**0.850**
Aluminum Foam[6]	0.5	~1.0	~1.0

Table 3. Thermal properties compared to other materials

	S.G	Thermal Conductivity // W/m·K	Thermal Conductivity ⊥ W/m·K	Specific T. C.* // W/m·K	Specific T. C.* ⊥ W/m·K
Graphite Foam	**0.54**	**106**	**106**	**198**	**198**
Copper	8.9	400	400	45	45
Aluminum	2.7	150	150	56	56
Aluminum Foam[6]	~0.5	12	12	24	24
Typical C/C	1.8	250	50	138	28

*defined as thermal conductivity divided by specific gravity

CONCLUSIONS

A novel technique to produce high thermal conductivity, open celled graphitic foam has been developed. The existence of very sharp 002 and 100 X-ray diffraction peaks confirms the presence of highly oriented graphitic planes. In fact, the interlayer spacing and crystallite sizes were better than VGCF filaments, which have been estimated to exhibit a thermal conductivity as high as 1950 W/m·K, suggesting that the ligaments of the foam will exhibit similar properties. These properties, combined with the continuous graphite network throughout the foam, result in an isotropic thermal conductivity greater than 100 W/m·K and a specific thermal conductivity over 4 times that of copper, an industry standard for thermal management. The high isotropic thermal conductivity of a foam-core composite will provide superior thermal management characteristics than 2-dimensional carbon-carbon composites used in aerospace applications by both reducing weight and improving through thickness thermal conductivity.

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