

Graphitic Foam Thermal Management Materials for Electronic Packaging

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ABSTRACT

The goal of this program is to utilize the recently developed high conductivity carbon foam for thermal management in electronics (heat exchangers and heat sinks). The technique used to fabricate the foam produces mesophase pitch-based graphitic foam with extremely high thermal conductivity and an open-celled structure. The thermal properties of the foam have been increased by 79% from 106 to 187 W/m-K at a density of 0.56 g/cm³ through process optimization. It has been demonstrated that when the high-thermal-conductivity graphitic foam is utilized as the core material for the heat exchanger, the effective heat transfer can be increased by at least an order of magnitude compared to traditional designs. A once-through-foam core/aluminum-plated heat exchanger has been fabricated for testing in electronic modules for power inverters.

INTRODUCTION

Contemporary thermal management has centered around aluminum and copper heat sinks and substrates. This is due to the very high thermal conductivity (180 W/m-K for aluminum 6061 and 400 W/m-K for copper). However, when weight is taken into account, the specific thermal conductivity (thermal conductivity divided by specific gravity) is only ~54 and 45 W/m-K respectively. Therefore, in automotive applications, where weight is a significant concern, it is imperative that a lighter weight thermal management material be found.

Mesophase pitch-derived graphitic foam, on the other hand, can be considered as an interconnected network of graphitic ligaments and, thus, should exhibit isotropic material properties. More importantly, such a foam will exhibit extremely high thermal conductivities along the ligaments of the foam (up to 5 times better than copper) and, therefore, will exhibit high bulk thermal conductivities. Metallic foams, on the other hand, are also being explored as a potential thermal management material. However, the thermal conductivities are still low, 5 - 50 W/m-K (1). Existing carbon foams are typically reticulated glassy carbon foams with a pentagonal

dodecahedron structure (2, 3, 4), illustrated in Figure 1, and exhibit thermal conductivities less than 1 W/m-K (1, 5, 6, 7). The pitch-derived graphitic foams reported here exhibit a spherical morphology, and present a unique solution to this problem by offering high thermal conductivity with a low weight.

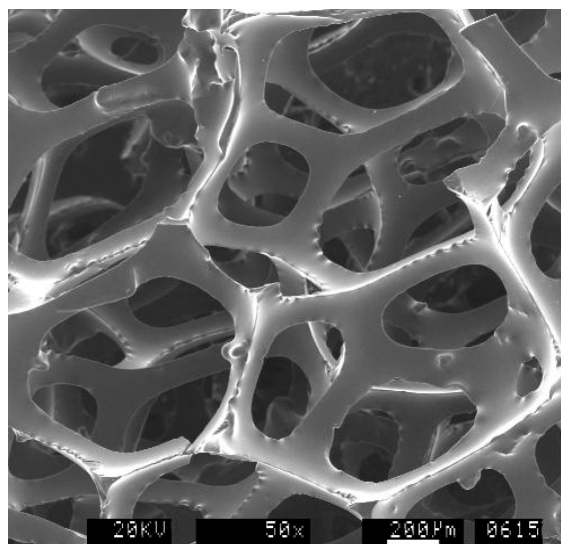


Figure 1. Typical reticulated glassy carbon foam produced by ERG Corporation.

Two devices are currently used for thermal management: heat exchangers, which transfer heat energy from one area of a device to another, and heat sinks, which absorb heat. Currently, most cooling heat exchangers for high-power electronics use layers of water-cooled aluminum or copper plate mounted below the electrical circuitry to transfer heat from hotter areas to cooler areas. This presents a unique problem in that if a crack or leak forms, then the water would short the circuitry and destroy the units. By using high thermal conductivity graphitic foam as the core material for these heat exchangers, the effective transfer of heat can be significantly increased while reducing the size and weight of the heat exchanger. But more importantly, it is

potentially possible to utilize air to cool the device, thereby removing the water from the system.

A new, less time consuming process for fabricating mesophase pitch-based graphitic foams without the traditional blowing and stabilization steps has been developed at Oak Ridge National Laboratory (ORNL) and is the focus of this research. Initially these foams possess a thermal conductivity of 106 W/m-K at a relatively low density of 0.54 g/cm³. Potentially, the process will lead to a significant reduction in the cost of graphitic-based thermal management materials (i.e. foam-reinforced composites and foam core sandwich structures).

EXPERIMENTAL

In this research, two 100% mesophase pitches were used to produce graphitic foam: Mitsubishi ARA24 naphthalene-based synthetic pitch with a melting point of 237°C (henceforth called ARA24 Mesophase), and a proprietary mesophase pitch from Conoco Corporation labeled Conoco Dry Mesophase (melting point of 355°C). All foam samples were carbonized at 0.2 °C/min to 1000°C and then graphitized at 10°C/min in Argon to 2800°C with a 2 hour soak at temperature.

In order to develop a fundamental understanding of the foam structure and graphitic morphology, and therefore develop an ability to tailor the thermal properties, samples were examined using a scanning electron microscope. Also, the thermal conductivity, κ , of the foam was determined with a xenon flash diffusivity technique. The thermal diffusivity, α , was first measured on samples 12-mm diameter by 12-mm thick on a custom built machine in the High Temperature Materials Laboratory at Oak Ridge National Laboratory. The sample density, ρ , and specific heat capacity, C_p , (measured with a laser flash technique to be 713 J/Kg·°C) were then used to calculate the thermal conductivity with the following relation:

$$\kappa = \alpha \cdot \rho \cdot C_p. \quad (1)$$

Finally, the same foam samples used for thermal diffusivity were machined to cylinders 12-mm diameter by 6-mm thick for x-ray diffraction studies, giving an understanding of the relationship between processing conditions and the graphitic structure of the foam.

Several 38.1 mm thick foam blocks were made from ARA24 Mesophase. Sandwich panels were constructed from a 12.7 mm thick, 152.4 mm diameter foam core sections machined from the thick blocks. Both aluminum 3003-H14 and copper 110, 0.635 mm thick, were used as facesheets. A thermally conductive film adhesive manufactured by Thermagon Inc., T-gon 1/KA-08-128 (0.203 mm, 8 W/m-K), was used to bond the facesheets to the foam core with a cure at 0.241 MPa, 150°C for 30

minutes. Although a slightly higher pressure was recommended for curing the film adhesive, 0.241 MPa was found to be sufficient for bonding to the foam.

Flexural tests were conducted on 107 mm by 27.9 mm samples according to ASTM C393-94 for four-point bending with two-point loading and one-quarter span. This specimen geometry was chosen to produce core shear or bond failure. Compression testing was conducted at 5.08 mm/min for 19 mm square samples.

The thermal conductivity of the laminated samples was determined by the same method as that for the basic foam.

RESULTS AND DISCUSSION

Figures 2 (a) and (b) are scanning electron micrographs of the pore structure of the Mitsubishi ARA24 and Conoco derived foams, respectively, heat treated at 1000°C. The Conoco pitch yielded foams with marginally higher densities than foams produced with the ARA24 mesophase pitch. The ARA24 pitch-derived foams exhibited a larger mean pore size than the Conoco pitch-derived foams (275 μm vs. 60 μm). The higher melting temperature of the Conoco pitch yields higher viscosities during processing, and therefore smaller bubble sizes.

Both foams exhibit a spherical cell structure with open, interconnected pores (P in Fig. 2) between most of the cells. It is evident from the images in Figure 2 that the graphitic structure in both foams is oriented parallel to the cell walls and highly aligned along the axis of the ligaments (L in Fig. 2).

It can be seen in the ARA24 derived foams that the graphitic structure is less aligned in the junctions between ligaments, (J in Fig. 2), and possesses more folded, mosaic texture. It is postulated that this arises from the lack of stresses at this location during forming.

Figure 3 is the x-ray diffraction patterns from both foams. The d_{002} spacing as determined using the x-ray diffraction patterns and the Bragg equation was calculated to be 0.3355 nm for the ARA24 foam and 0.3360 nm for the Conoco derived foam. This is significantly better than existing high performance carbon fibers such as Amoco K1100 fibers (0.3366) and vapor grown carbon fibers (VGCF) (0.3366) and better than most synthetic graphites. The crystallite sizes (L_a and L_c) as determined from the x-ray diffraction data and the Scherrer equation were similar to typical high thermal conductivity carbon fibers.

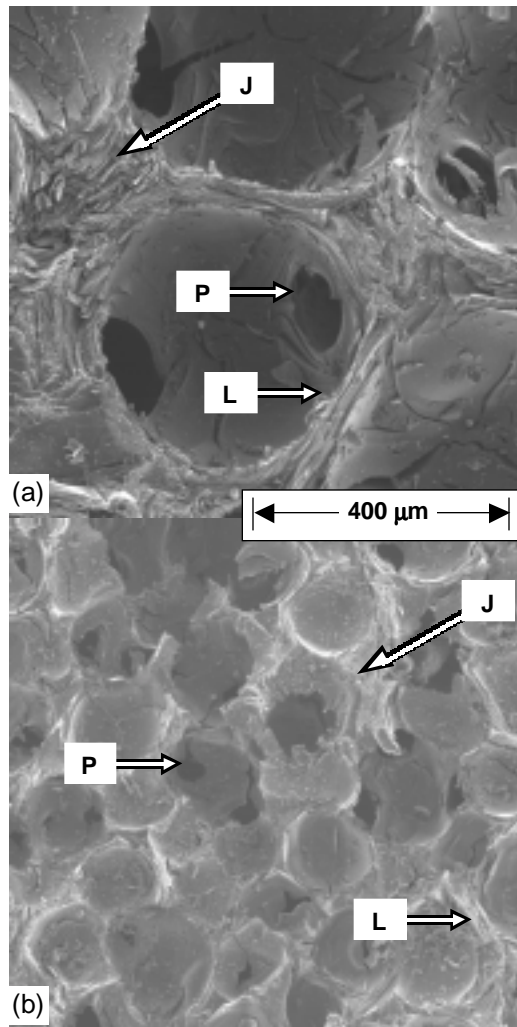


Figure 2. Structure of (a) Mitsubishi ARA Mesophase and (b) Conoco Mesophase pitch-derived graphitic foams carbonized at 1000°C.

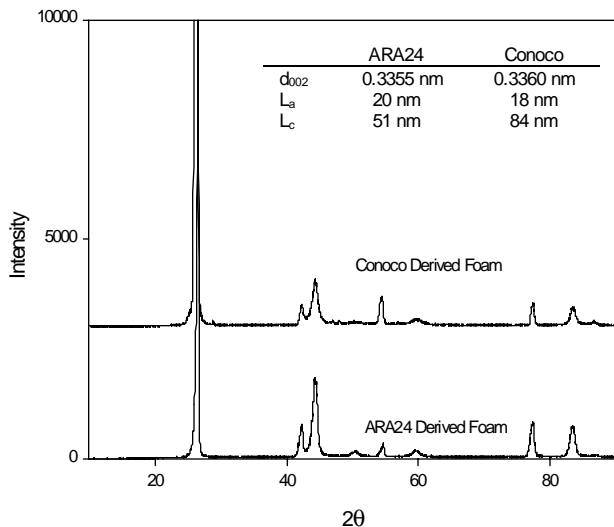


Figure 3. X-ray diffraction patterns of ARA24 and Conoco derived foams graphitized at 2800°C.

The thermal conductivity of the ARA24 mesophase foam graphitized at 10°C/min ranged from 50 to 150 W/m·K and the Conoco derived foams exhibited thermal conductivities ranging from 40 to 135 W/m·K (Figure 4). This is remarkable for a material with such a low density, 0.27 to 0.57 g/cm³ (density was varied by varying processing conditions).

Under close examination of the SEM images in Figure 2, microcracks and delaminations of the graphite planes can be observed. This is most likely due to the thermal stresses induced during carbonization and graphitization due to the low thermal conductivity of the foam prior to graphitization. It was postulated that reducing the heating rates during this process would minimize thermal gradients, and thus minimize thermal stresses, resulting in fewer microcracks and delaminations. In fact, when the graphitization rate was slowed to 4°C/min, the thermal conductivity of the ARA24 derived foams increased significantly to nearly 190 W/m·K (Figure 4). This is significantly better than the targeted 50% increase in thermal conductivity.

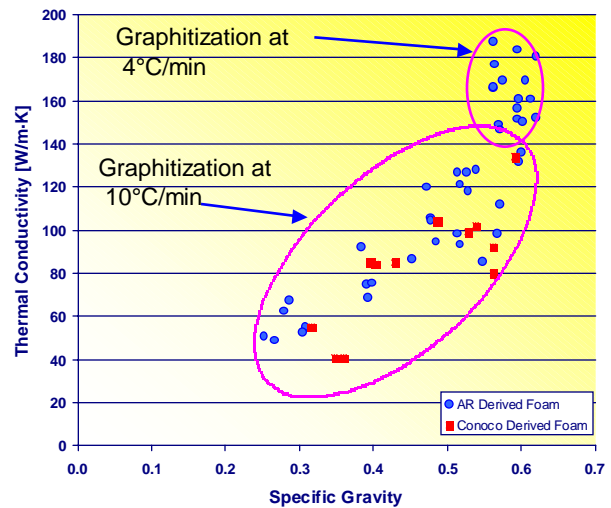


Figure 4. Thermal conductivity of ARA24 and Conoco derived foams as a function of density.

Four sandwich panels were fabricated for testing: two with aluminum facesheets and two with copper facesheets. The flexure specimens exhibited classic shear failure with only a slight delamination of the foam from the facesheet. The core shear stress ranged from 1.49 to 2.35 MPa, while the shear modulus ranges from 47.9 to 111 MPa. The values for panels with copper facesheets had a significantly higher core shear stress and core shear modulus that has not been explained.

A typical load-displacement curve for the compression tests shows that the foam core crushes with a fairly uniform load over a large displacement. The compression strength and modulus ranged from 1.2 to 2.5 MPa and 44 to 176 MPa, respectively.

The results of the thermal conductivity testing of the sandwich panels indicated that the sandwich specimens had a through the thickness thermal conductivity of between 50 and 65 W/m-K with little difference between the aluminum and the copper sandwich panels. Although the thermal conductivity was lower than the basic foam due to the relatively low conductivity adhesive, the specific conductivity of the sandwich panels is comparable to aluminum.

The average adhesive thickness in the sandwich panels was between 0.127 and 0.203 mm. With a thermal conductivity of only 8 W/m-K the interface was the limiting factor for the through thickness conductivity. Methods to improve the through thickness thermal conductivity include using a higher conductivity adhesive and decreasing the adhesive thickness. Several additional sandwich panels have been successfully bonded with thinner bondlines of filled epoxies (approximately 0.0254 mm). Also, a brazing technique has been developed for bonding aluminum facesheets (thermal conductivity of the brazing material is approximately 45 W/m-K).

APPLICATIONS

Since the foam is open cellular, it is a prime candidate for use as a porous media heat exchanger for a power electronic substrate. Currently, most substrates for high power electronics include a water-cooled aluminum or copper plate mounted below the circuitry. It can be shown that the effective heat transfer coefficient can be raised from $\sim 250 \text{ W/m}^2\text{-K}$ for current designs to over $10,000 \text{ W/m}^2\text{-K}$ for flow through a porous graphite foam. It was proposed that a once through foam core/aluminum plated substrate be fabricated to replace the current substrates, see Figure 5. The foam core thickness, geometry, channel patterns, foam cell size, and heat treatment temperature will be evaluated to give optimum heat removal at the lowest flow rate of cooling fluid.

In a separate test, heat transfer coefficients for a shell and tube heat exchanger with graphites foam as the core were measured as high as $11,000 \text{ W/m}^2\text{-K}$. This test validated the ability of the foams for removing large amounts of heat with cooling air instead of water.

Furthermore, independent tests of the foam material at Florida International University have confirmed the dramatic improvement in heat transfer coefficients when the foam is used as a porous heat transfer media. The major conclusion of this research is that natural convection is not enough to improve the heat transfer: the air must be forced through the foam to realize the full potential of the material.

A closed-form mathematical model was developed at the University of Tennessee to predict the thermal conductivity of the foam based on the structure and

density. This model is being expanded to study the heat transfer characteristics of the foam under forced flow.

Last, extensive meetings with Chrysler, Lockheed Martin, Boeing, Modine, Peterbilt and a NASCAR racing team are being conducted to develop this material for radiator systems and other electronic cooling.

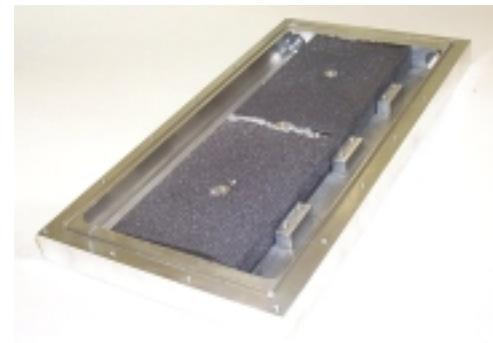
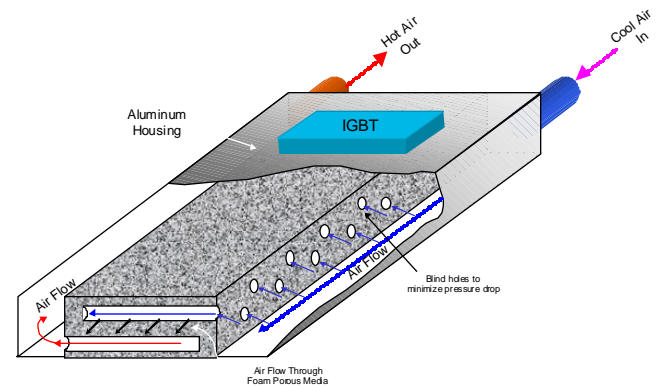


Figure 5. Schematic and photograph of design for foam-based heat exchanger substrate for power electronics cooling.

CONCLUSIONS

The manufacture and properties of high thermal conductivity graphites foams have been reported. It was shown that pitch precursor characteristics will affect foam structure and properties such as bubble size, ligament structure, and thermal conductivity. The highly aligned ligaments have similar structures to high thermal conductivity carbon fibers, such as K1100 and VGCF. In fact, the d-spacing were less than VGCF, which have exhibited thermal conductivities as high as 1950 W/m-K . These properties, combined with the continuous graphitic network result in a specific thermal conductivity up to 6 times greater than that of copper. Through this essential materials characterization, it was determined that slower heating rates during carbonization and graphitization would result in a dramatic improvement in thermal conductivity, nearly 75% better than the initial values.

Standard laminating techniques were shown to be viable for producing foam core sandwich panels. However, either thin bondlines or brazed interfaces were found necessary to preserve the high thermal conductivity. Through further development, graphites foam can replace

honeycomb in applications that require high thermal conductivity and low weight.

High heat transfer coefficients have been measured and heat exchangers have been designed and built with the knowledge learned in this program. Ongoing research will conduct extensive tests and redesigns to build a proper heat exchanger substrate for power electronics cooling and other cooling applications for PNGV.

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REFERENCES

1. Gibson LJ, Ashby, MF. Cellular Solids: Structures & Properties, Pergamon Press, New York, 1988.
2. Glicksman LR, Torpey M. Proceedings of the Polyurethane World Congress, Aachen, Germany, 1987.
3. Glicksman LR, Marge AL, Moreno JD. Developments in Radiative Heat Transfer, ASME HTD – 1992;203.
4. Kuhn J, Int. J. Heat Mass Transfer 1992;35(7):1795-1801.
5. Glicksman, LR, Schuetz M, Sinofsky M. A Study of Radiative Foam Heat Transfer through Foam Insulation. Report prepared by Massachusetts Institute of Technology under subcontract No. 19X-09099C, 1988.
6. Ultramet Product Literature, 1998.
7. Doermann D, Sacadura JF. J. of Heat Transfer 1996;118:88-93.