HIGH THERMAL CONDUCTIVITY MESOPHASE PITCH-DERIVED CARBON FOAMS: EFFECT OF PRECURSOR ON STRUCTURE AND PROPERTIES

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Introduction

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. Potentially, the process will lead to a significant reduction in the cost of carbon-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 (melting point of 237°C), 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.

Results and Discussion

Figures 1 (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 (0.6 g/cm^3) than foams produced with the ARA24 mesophase pitch (0.55 g/cm^3). 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. 1) between most of the cells. It is evident from the images in Figure 1 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. 1).

It can be seen in the ARA24 derived foams that the graphitic structure is less aligned in the junctions between ligaments, (J in Fig. 1), and possesses more folded, mosaic texture. It is postulated that this arises from the lack of shear stresses at this location during forming.

Figures 2 (a) and (b) are optical micrographs of the Mitsubishi ARA24 pitch-derived foams under cross-polarized light. These micrographs confirm that the graphene layer planes are highly oriented parallel to the surface of the bubbles (along the axis of the ligaments), indicated by the monochromatic regions (MC in Fig. 2). The mesophase alignment is predominantly parallel to the surface of the bubbles, with a disruption of the alignment in the junctions (J in Fig. 2). The folded texture regions (F) in Figure 2 (b), illustrate there is more fold-sharpening and crystal misalignment in the junctions of the ARA24 derived foams.

An examination of the Conoco pitch-derived foams (Figures 2 (c) and (d)) also reveals the presence of highly aligned ligaments. However, images of the junctions (J) suggest that the Conoco-derived foams contain better orientation of the mesophase in these regions. This may be a result in the differences in the precursor mesophase and their respective rheological properties.

Figure 3 is the x-ray diffraction spectra for both foams. The d_{002} spacing 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 K1100 and vapor grown carbon fibers (VGCF) and better than most synthetic carbons. The crystallite sizes (L_a and L_c)were similar to typical high thermal conductivity carbon fibers.

The thermal conductivity of the graphitized ARA24 foam 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/cm^3 (density was varied by varying processing conditions).

Conclusions

The manufacture and properties of high thermal conductivity carbon 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 (36). These properties, combined with the continuous graphitic network result in a specific thermal conductivity up to 6 times greater than that of copper.

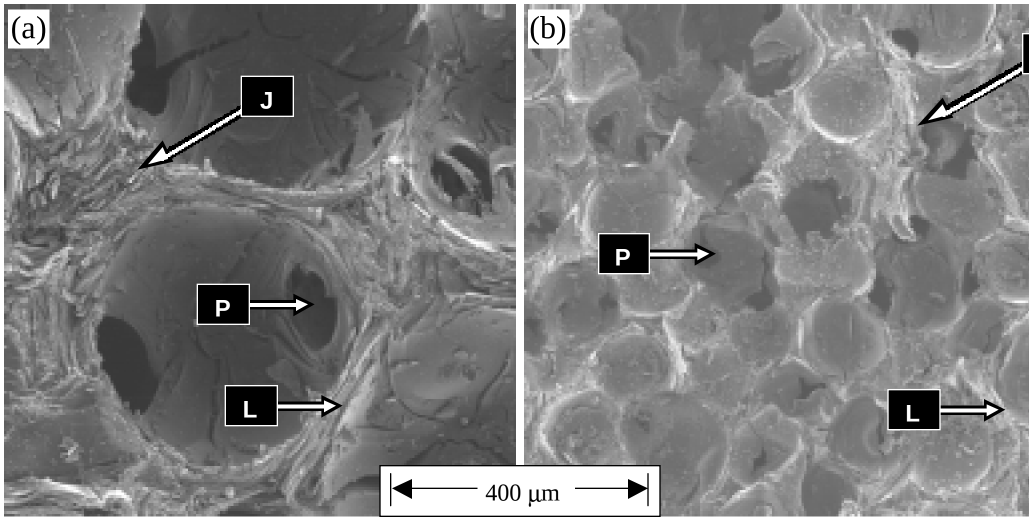
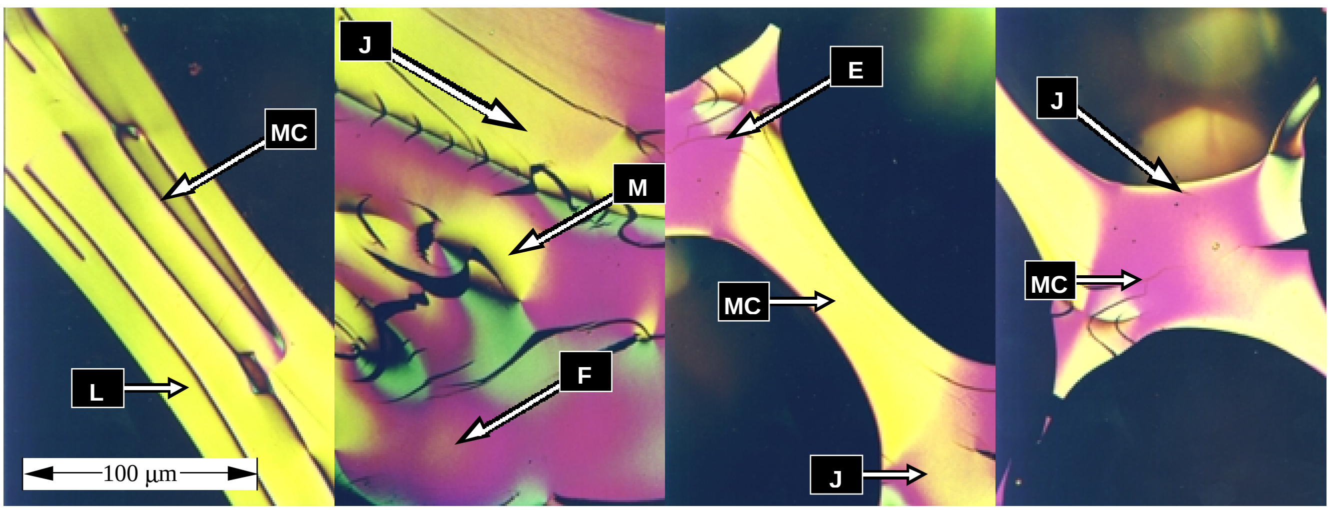


Figure 1. Structure of (a) Mitsubishi ARA and (b) Conoco Mesophase pitch-derived carbon foams carbonized at 1000°C.



(a) Ligament in ARA24 Foam (b) Junction in ARA24 Foam (c) Ligament in Conoco Foam (d) Junction in Conoco Foam

Figure 2. Comparison between ARA24 and Conoco pitch derived foams carbonized at 1000°C.

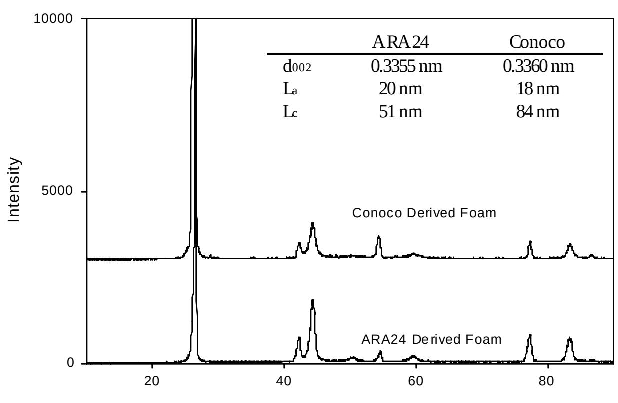


Figure 3. X-ray diffraction patterns of ARA24 and Conoco derived foams graphtized at 2800°C.

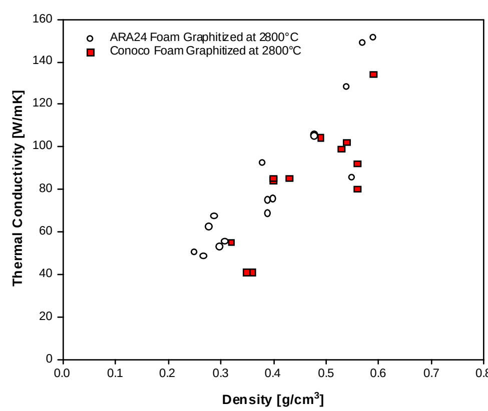


Figure 3. Thermal conductivity of ARA24 and Conoco derived foams as a function of density.

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