THE STRUCTURE AND PROPERTIES OF CARBON FIBER BASED ADSORBENT MONOLITHS


Oak Ridge National Laboratory,
P.O. Box 2008, Oak Ridge, TN 37831-6088, USA.

1. Introduction
Carbon fiber monoliths manufactured by a novel slurry molding process from isotropic pitch-derived fibers are being developed at ORNL for gas separation and storage applications [1]. Low density ($\rho = 0.2 - 0.3$ g/cm$^3$) monoliths have been successfully demonstrated to have an acceptable pressure drop for gas separation applications and are currently being developed for CO$_2$/CH$_4$ separations, whereas monoliths with densities in the range $\rho = 0.4 - 0.6$ g/cm$^3$ have been shown to have natural gas storage capacities of $>$100 V/V at 500 psi pressure and room temperature.

Here we report our recent studies of the structure, porosity, and properties of these novel monoliths. Data to be presented and discussed includes a SEM assessment of the structure of standard and hybrid monoliths, porosity assessments by N$_2$ adsorption at 77K, methane storage capacity measurements, and thermal conductivity determinations.

2. Experimental
The composites were made via a slurry molding process in which the carbon fibers and powdered phenolic resin binder were first mixed with water, followed by molding over a porous screen. The water is drawn through the screen under the influence of a vacuum, depositing the fibers and resin onto the screen. The resultant green-form is dried, cured and carbonized to yield a finished carbon composite product with a bulk density in the range 0.15 – 0.4 g/cm$^3$. If greater densities are required the green-form may be hot pressed after drying, followed by carbonization, yielding densities in the range 0.5-0.9 g/cm$^3$ (Fig. 1).

Thermal conductivity, as a function of temperature, was measured using the LASER flash, thermal-pulse method. The thermal diffusivity was calculated from the thermal transient, and the materials thermal conductivity then calculated from:

$$K = \alpha \cdot \rho \cdot C_p$$

Where $\alpha$ is the thermal diffusivity (m$^2$/s), $\rho$ is the bulk density (kg/m$^3$), and $C_p$ is the specific heat at the measurement temperature (J/kg K).

3. Results and Discussion
Porous carbon monoliths based on carbon fibers represent a unique class of carbon materials which have tailorable pores structures and, therefore, physical and mechanical properties. By controlling fiber length distribution, fiber fraction, fiber type, processing route, and activation conditions, one may markedly alter the resultant properties of the monoliths.

Fig. 1. The manufacturing process for carbon fiber based adsorbent monoliths.
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The natural gas storage capacities of several of our monoliths made from isotropic pitch fiber are given in the Table below.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Weight Activity @ 15 psi (g/100g)</th>
<th>Retentivity @ 15 psi (%)</th>
<th>Storage Capacity @ 500 psi (V/V)</th>
</tr>
</thead>
<tbody>
<tr>
<td>SMS-2B</td>
<td>6.8</td>
<td>29</td>
<td>76</td>
</tr>
<tr>
<td>SMS-3B</td>
<td>8.9</td>
<td>23</td>
<td>84</td>
</tr>
<tr>
<td>SMS-5A</td>
<td>9.8</td>
<td>20</td>
<td>104</td>
</tr>
</tbody>
</table>

The maximum storage volume attained to date in our monoliths is 104 V/V, which is less than our target of 150 V/V. The thermal conductivity of carbon adsorbents is of significance because large temperature gradients may develop when a storage bed is charged or discharged. We have demonstrated the ability of our monoliths to heat when an electrical current is passed through them (Fig. 2), offering the potential to overcome the bed cooling that occurs with rapid discharge of natural gas.

![Fig. 2 Current-voltage plot for a carbon fiber monolith.](image)

Another approach to minimizing the temperature gradients that develop in a storage bed is to increase the thermal conductivity of the adsorbent carbon. To this end, we have developed hybrid monoliths that contain small fractions of mesophase pitch-derived carbon fibers. Our hybrid monoliths exhibit thermal conductivities in the range 0.2–0.9 W/m.K depending on the blend and density of the monolith. In comparison, a packed bed of granular carbon at comparable density would have a thermal conductivity of approximately 0.1 W/m.K [1]. The thermal conductivities of several of the hybrid monoliths are shown in Fig. 3.

![Fig. 3. The temperature dependence of the thermal conductivity of carbon fiber based hybrid adsorbent monoliths.](image)

The improved thermal conductivity of our monoliths is attributed to the bonding between the fibers and the incorporation of high thermal conductivity, mesophase pitch-derived carbon fibers. These features are visible in the SEM micrograph in Fig. 4.

![Fig. 4. SEM micrograph of a hybrid monolith.](image)

4. Conclusions
A novel, carbon fiber based, adsorbent gas storage monolith with improved thermal conductivity has been developed.

5. References