FY95 End of Fiscal Year Letter
(01 Oct 1994 - 30 Sep 1995)

ONR CONTRACT INFORMATION

Contract Title: High Thermal Conductivity Carbon/Carbon Composites
Performing Organization: Clemson University
Principal Investigator: Dr. Dan D. Edie; (803) 656-3056;
fax: (803) 656-0784
Contract Number: N00014-92-J-4104 (CU REF:05-5002)
R & T Project Number: C/C 9200---01
ONR Scientific Officer: Steven G. Fishman
**Title and Subtitle of Report:**
High Thermal Conductivity Carbon/Carbon Composites

**Author(s):**
Dr. Dan D. Edie

**Performing Organization Name(s) and Address(es):**
Clemson University
Box 340909, 123 Earle Hall
Chemical Engineering
Clemson, SC 29634-0909

**Sponsoring/Monitoring Agency Name(s) and Address(es):**
Office of Naval Research
Code 1513:ETF
Ballston Tower One
800 North Quincy Street
Arlington, VA 22217-5660

**Supplementary Notes:**

**Distribution Availability Statement:**
Unlimited

**Abstract (Maximum 200 words):**
The objective of this project was to develop a low-cost, high thermal conductivity carbon/carbon composite with a mesophase pitch-based matrix. A low-cost, continuous powder coating process was developed which can produce a flexible pre-impregnated pitch-based towpreg. A combination of a pressure-carbonization technique and heat treatment of the mesophase pitch was utilized to enhance composite properties by increasing the composite density. Three different fibers, T300 PAN-based, P55 pitch-based, and an experimental high thermal conductivity mesophase pitch-based, were incorporated as the filler phase in the composites. The thermal conductivity of the graphitized T300/AR-120 and the P55/AR-120 composites was 80.5 and 135.5 W/m-K, respectively. The composites reinforced with the experimental ribbon fibers exhibited 3-D anisotropy, with a thermal conductivity, transverse to the fibers, of 213.5 W/m-K, higher than that parallel to the fibers, 145 W/m-K. These results indicate that fiber shape can affect matrix properties in carbon/carbon composites. Using finite element methods, a model was developed to predict the effect of specific material variables, such as fiber fraction, fiber structure, matrix structure, fiber/matrix interface, and void fraction, on the thermal conductivity of the composite.
A. Research Goals

The objective of the project was to develop a low-cost, high thermal conductivity carbon/carbon composite with a mesophase pitch-based matrix. In addition, it was proposed that the project employ a pressure-carbonization technique in order to increase the composite density to approximately 1.5 g/cc, resulting in an increase in both the mechanical and thermal properties of the composites.

B. Significant Results

A low-cost, continuous powder coating process has been developed which can produce a flexible pre-impregnated pitch-based towpreg. The process was used to produce towpreg from Mitsubishi AR mesophase pitch powder and three different carbon fibers: T300 PAN-based fiber, P55 pitch-based fiber, and an experimental high thermal conductivity pitch-based ribbon fiber. The pitch powder was deposited on individual fibers, rather than on bundles of fibers. As a result, the pitch-based towpreg was very flexible and easy to handle. This flexibility will allow the forming of multidimensional pre-impregnated preforms which can be simply hot pressed into composites.

The thermal conductivity (parallel to the fibers) of the graphitized T300/AR-120 and P55/AR-120 composites was 80.5 and 135.5 W/m-K, respectively. These results, along with x-ray analysis, indicated a significant development of preferred crystalline order (parallel to fibers) upon graphitization at 2400°C. The composites reinforced with ribbon fibers exhibited 3-D anisotropy, with a thermal conductivity (transverse to the fibers) of 213.5 W/m-K, higher than that parallel to the fibers (145 W/m-K). These results indicated that fiber shape can affect matrix properties in carbon/carbon composites.

A finite element model was developed to predict the thermal conductivity of carbon/carbon composites, both parallel and transverse to the fibers. This model accounts not only for the anisotropic nature of the fibers and matrix, but also for random porosity and different types of fiber/matrix bonding (Figure 1). The model was able to accurately predict the average thermal conductivity of the composites produced in this study. Figure 2 shows graphically the agreement between predicted and measured thermal conductivity for one set of composites. Other composites showed similar agreement. The model successfully predicted the thermal conductivity parallel to the fiber axis for P55 and ribbon fiber composites. The model also predicted the thermal conductivity perpendicular to the fibers for P55 pitch-based fiber composites, but significantly underestimated the conductivity of ribbon fiber composites.

Figure 1. Exaggerated illustration of intramatrix cracking and fiber matrix debonding both parallel and perpendicular to the fiber axis in a ribbon fiber composite.
Figure 2. Plot of predicted versus measured thermal conductivity, parallel to the fibers, of individual T300/AR-120 composites heat treated to 2400°C.

C. Future Research

1. Ribbon fiber and heat-treated mesophase pitch should be used to produce high thermal conductivity carbon/carbon composites.

2. The towpreg fabrication method can be used to widen the processing window to include alternative pitches, which may form better composites in less time.

3. The finite element model produced for this study can be used to study the effect of specific material characteristics, such as fiber texture and fiber/matrix bonding, on the composite thermal conductivity.
D. List of Publications/Reports/Presentations

1. Papers Published in Refereed Journals


2. Non-Refereed Publications and Published Technical Reports


3. Presentations
   a. Invited

   b. Contributed
      (See item 2 above.)

4. Books (and sections thereof)

Enclosure (2)
E. LIST OF HONORS/AWARDS

<table>
<thead>
<tr>
<th>Name of Person Receiving Award</th>
<th>Recipient’s Institution</th>
<th>Name, Sponsor and Purpose of Award</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dan D. Edie</td>
<td>Clemson University</td>
<td>Elected to the Executive Council of the American Carbon Society</td>
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</tbody>
</table>
F. Participants

James W. Klett, completed Ph.D. in Chemical Engineering and graduated from Clemson in December, 1994.

Richard M. Dayrit, minority student currently enrolled as an M.S. student in Chemical Engineering.

Both of the above are U.S. citizens.

G. Other Sponsored Research During FY 95

This Grant
"High Thermal Conductivity Carbon/Carbon Composites," Office of Naval Research, $95,100, 0% of time, (1992-1995) $95,100

Other Grants
"High Thermal Conductivity Fibers," Sponsored by the Great Lakes Composite Consortium, $220,000/yr, 30% of time, 1/1/92 to 12/31/95.

"High Thermal Conductivity Fibers from PBO," Sponsored by ONR, 0% of time, $31,000/yr, 7/31/94 to 8/1/97.

"Production of Carbon Monofilament- Phase II," Sponsored by MSNW, $150,000/yr, 5% of time, 3/3/95 to 5/3/96.

"Supercritical Extraction for High Thermal Conductivity Fibers," Sponsored by DEPSCoR, $100,000/yr, 15% of time, 9/1/94 to 8/31/97.

"Engineering Fibers and the Micromechanics of Their Composites," Sponsored by NSF, $95,000/yr, 17% of time, 7/1/91 to 6/31/94.
H. SUMMARY OF FY95
PUBLICATIONS/PATENTS/PRESENTATIONS/HONORS/ PARTICIPANTS
(Number Only)

a. Number of Papers Submitted to Referred Journal but not yet published: ___ 1

b. Number of Papers Published in Refereed Journals: ___ ___

c. Number of Books or Chapters Submitted but not yet Published: ___ ___

d. Number of Books or Chapters Published: ___ ___

e. Number of Printed Technical Reports & Non-Refered Papers: ___ 2

f. Number of Patents Filed: ___ ___

g. Number of Patents Granted: ___ ___

h. Number of Invited Presentations at Workshops or Prof. Society Meetings: ___ 1

i. Number of Contributed Presentations at Workshops or Prof. Society Meetings: ___ 2

j. Honors/Awards/Prizes for Contract/Grant Employees: ___ 1
   (selected list attached)

k. Number of Graduate Students and Post-Docs Supported at least 25% this year on contract grant:
   Grad Students: TOTAL ___ 2
   Female ___ ___
   Minority ___ ___
   Post Doc: TOTAL ___ 1
   Female ___ ___
   Minority ___ ___

Enclosure (4)
1. Number of Female or Minority PIs or CO-PIs

   New Female
   Continuing Female
   New Minority
   Continuing Minority  

Enclosure (4) contd.