PROPERTIES OF HIGH THERMAL CONDUCTIVITY CARBON-CARBON COMPOSITES FOR THERMAL MANAGEMENT APPLICATIONS

Ilan Golecki[1*], *L. Xue*[1], *D.M. Dewar*[2], *A.F. Anderson*[2], *C.K. Duncan*[2], *J. van Horik*[2], *K. Eubank*[2], *T. Walker*[3]
[1-3]*AlliedSignal, Inc., USA:* [1]*Research and Technology, Morristown, NJ 07962;* [2]*Aerospace Equipment Systems, Torrance, CA 90504;* [3]*Aircraft Landing Systems, South Bend, IN 46628.*
[*]*Phone (973) 455-4938; Fax (973) 455-4339 or -3008; E-mail: Ilan. Golecki@alliedsignal.com*

Introduction and Motivation

Carbon-Carbon (C-C) fiber-matrix composite materials (C-C's) possess unique characteristics which make them attractive for a wide spectrum of current and potential applications, e.g., brake pads for commercial and military airplanes and for race cars, heat exchangers [1-3], re-entry cones for missiles, aerospace structural components, heat sinks and spreaders for power electronics, and bipolar plates for proton-exchange-membrane fuel cells. C-C's have very low densities in the range 1.6-2.2 g/cm^3, lower than that of aluminum (2.7 g/cm^3) and four times lower than that of stainless steel. C-C's have higher thermal conductivities than those of copper and silver and the highest thermal conductivity per unit density among materials suitable for thermal management applications, see Fig. 1. They have high mechanical strength, which *increases* with temperature, by contrast to metals and ceramics, the strength of which *decreases* with temperature. C-C's also evidence graceful failure under load, as do ceramic fiber-matrix composites, while graphite and monolithic ceramics fail abruptly when their ultimate strength is exceeded. C-C's also possess high toughness and can be used in a wide range of temperatures and in severe and chemically aggressive environments; they require protection from oxidation for continuous use above ~350°C in an oxidizing environment [see companion paper, I. Golecki et al., this conference]. Many properties of C-C composites, such as the coefficient of thermal expansion (CTE), can be tailored by choosing the type of fiber, fabric, matrix and processing conditions. Here we describe several properties of C-C materials which we have developed for heat management applications at elevated temperatures [1-3].

Materials, Methods and Results

We used several types of continuous, pitch-based carbon fibers from different manufacturers, such as Amoco or Nippon Graphite. Pitch-based fibers are graphitizable and amenable to the fabrication of complex-shaped articles. We fabricated two-dimensional, thin-gauge, single- and multiple-ply, woven structures, having e.g., plain weave or different satin weaves. Both flat and intricately shaped configurations, exemplified by Fig. 2, were made. The measurements reported here were obtained on flat pieces. These preforms were densified using both resin and/or chemical vapor infiltration (CVI). Several resin systems were employed, as well as different CVI precursors and processing conditions [4]. Annealing was performed in the temperature range 1800-3000°C. For proprietary reasons, specific processing details are omitted. The microstructures of the samples were characterized using polarized-light microscopy, scanning electron microscopy and X-ray diffraction. In-plane thermal conductivities were measured using the Fourier and Kohlrausch methods [2]. The CTE was measured to 600°C. The tensile strength was determined using standard techniques.

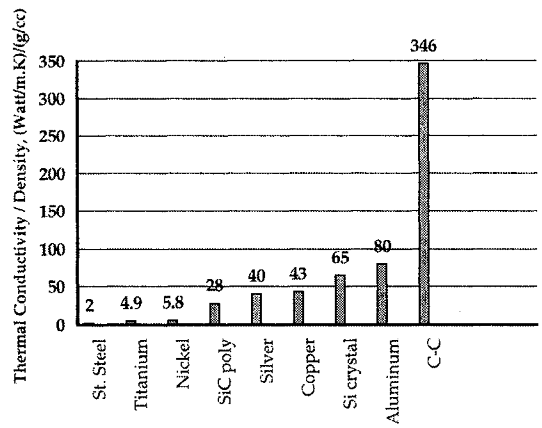

Figure 1. Specific thermal conductivity near room temperature of materials of interest in thermal management.

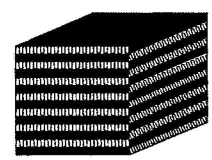

Figure 2. Schematic of a counterflow heat exchanger core.

The in-plane thermal conductivities of two single-ply, ca. 0.18 mm (7 mil) thick, 2-D woven C-C preforms are shown in Fig. 3. These samples had a balanced weave and had been heat treated. The two samples "A" and "B" were part of a set made from the same fibers, which underwent nominally the same processing sequences. The data for a high-purity, type AXF-5Q Poco graphite sample is also shown. The solid triangle and the open circle were measured by means of the Fourier technique in air in one laboratory, while the series of open triangles and solid circles were measured using the Kohlrausch technique in vacuum in another laboratory. The Fourier measurements were performed on pieces several inches on a side, while the Kohlrausch measurements were made using four narrow and long strips laid up on one another. In the C-C samples, the thermal conductivity is highest close to room temperature and decreases monotonically with temperature. The graphite sample behaves similarly, except there seems to be a broad maximum at ~ 100°C. Most materials exhibit a broad maximum in the temperature dependence of the thermal conductivity [5]. The decrease in thermal conductivity with increasing temperature is due to increased scattering of the phonons. The differences seen in the Kohlrausch measurements of samples "A" and "B" could be due either to a scatter in the properties and/or in the measurement technique. For sample "A", the agreement between the Kohlrausch and the Fourier values close to room temperature is excellent, but

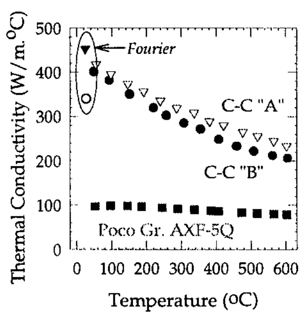

Figure 3. In-plane thermal conductivities of two-dimensional, 7 mil thick, single-ply, heat-treated Carbon-Carbon composite panels and of Poco graphite (grade AXF-5Q).

the agreement is not as good for sample "B". The thicknesses were normalized for the purposes of this comparison and standard metal samples (e.g. aluminum or copper) were used to check the experimental setup. Since these are absolute measurements carried out at different laboratories by different operators, using different approaches and experimental setups, it is not known with high certainty to which particular cause these differences could be attributed. Using a larger statistical database, better trends become apparent.

These C-C samples have room temperature conductivities of 340-460 W/m.K, more than four times higher than that of the graphite sample. However, the thermal conductivities of the C-C samples decrease with temperature faster than that of the graphite sample; at 600°C, the ratio is 2.5-3 to 1. The through-plane thermal conductivity of such 2-D C-C panels is much lower than the in-plane values, due to the fact that in 2-D C-C's, many of the physical properties are dominated by the fiber properties, even though graphitized, rough-laminar CVI carbon has excellent physical properties as well. The transverse properties of such pitch fibers are much worse than the longitudinal properties. This is due to the fact that the a-b carbon planes in the fibers are aligned longitudinally, in the fiber direction, and the c vector is in the transverse, radial direction. In graphite, the mechanical, thermal and electrical properties are much superior in the a-b plane, compared to the values between planes, in the c direction.

The in-plane thermal expansion behavior of two C-C samples is shown in Fig. 4. Sample "I" is different from sample "II" in both fiber type and weave type, but the heat treatment temperatures are the same and higher than in Fig. 3. Both types of composites have a *negative* in-plane TCE between room temperature and 250-300°C, as reported previously for C fibers and C-C composites [6]. After an initial decrease, the TCE increases with increasing temperature for both materials. Most materials show the latter type of behavior [5]. The "wiggles" in the TCE could be due to residual experimental noise. It is noteworthy that the two samples, even though heat treated at the same temperature, exhibit substantial differences in their TCE's. This has important implications for design of practical articles and for oxidation protection coatings. In C-C composites, one of the issues is to develop an oxidation protective coating which is reasonably well matched thermally to the substrate. The reason is that in addition to the somewhat unusual negative TCE in the fiber direction, such C-C composites exhibit positive and larger TCE's in the through-plane direction [6].

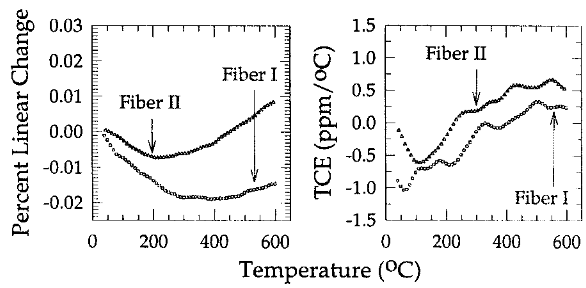

Figure 4. In-plane thermal expansion of two-dimensional, two-ply, heat-treated Carbon-Carbon composite panels.

Summary

We have achieved very high in-plane thermal conductivities of 400-700 W/m.K and through-plane thermal conductivities of 20-70 W/m.K in thin-gauge, 2-D C-C composite panels, using relatively inexpensive pitch-based carbon fibers and a relatively moderate thermal annealing schedule. Such materials are highly suitable for a number of thermal management applications, including heat exchangers and heat spreaders for electronic components. Densification of such components has been completed in less than one day, much faster than required using conventional, published approaches. Densities of 2.0-2.2 g/cm^3 have been achieved routinely. The microstructure of the carbon matrix produced by chemical vapor infiltration is rough-laminar, with extinction angles in the range 19-21°. Tensile strengths are in the range 200-400 MPa (30-60 ksi). Complex-shaped structures were fabricated from 0.08-0.25 mm (3-10 mil) thick plies and a density of 28 fins per inch has been demonstrated in C-C heat exchanger structures.

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