MODELING THE THERMAL CONDUCTIVITY OF CARBON FIBERS

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Introduction

The excellent thermal properties of mesophase pitch-based carbon fibers makes them an ideal material for applications in which high rates of heat dissipation and low mass are required. They are used to enhance the thermal conductivity of polymers and metals for high performance applications such as heat sinks for military avionics. However, the high cost of mesophase pitch-based fibers has limited their use in high volume applications. Continuing research at Clemson University has been focused on developing a low-cost high thermal conductivity carbon fiber. The measurement of the thermal conductivity of carbon fibers is difficult, time consuming, and requires specialized equipment. There are only two apparatuses available that are able to directly measure the thermal conductivity of carbon fibers with reasonable accuracy (the thermal potentiometer and the Angstrom's apparatus). Other researchers have also used laser flash analysis of unidirectional composites. However, this method is considered less reliable than the other two techniques, and it only indirectly measures thermal conductivity.

The correlation between electrical resistivity and thermal conductivity derived by Lavin and coworkers [1] has allowed many researchers to estimate the thermal conductivity for carbon fibers based on their electrical resistivity (an easier measurement to perform). However, this correlation must be used with caution, since it is only valid for fibers produced from the same precursor and with electrical resistivity values within the range used to derive the correlation.

A more reliable approach would be to develop a predictive model for thermal conductivity, which is based on the structure of the fiber, since it has been proven that the structure of the carbon fibers directly affects their thermal conductivity. Structural information can be easily obtained utilizing readily available x-ray techniques.

In this article, models for fiber structure (structural parameters obtained from x-ray analysis) will be presented and used to predict the thermal conductivity of carbon fibers. Two models will be discussed. The first model is based on a theory for short-fiber composite materials, while the second model utilizes the concept of homogenization to create a periodic composite model.

Short-Fiber Composite Model

From a structural point of view, carbon fibers could be considered a composite material comprised of two components: a) a matrix or mesophase fraction which did not crystallize during the heat treatment process, and b) a reinforcing material which consists of the mesophase material that crystallized into graphite-like structure (this component is responsible for most of the thermal conductivity of the fiber).

In fact, the structure of the carbon fiber resembles that of an aligned short-fiber composite (or discontinuous fiber-reinforced composite) with the crystallites serving as the short fibers. See Figure 1.

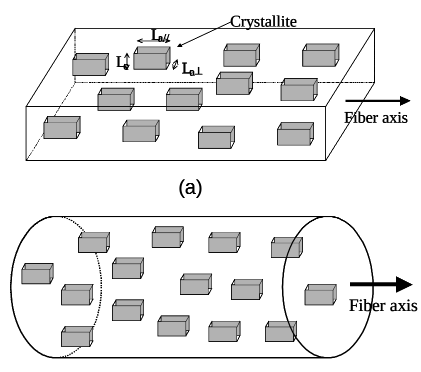

Figure 1. Schematic of a (a) ribbon fiber and (b) round fibers visualized as an aligned short-fiber composite.

* Currently employed by Oak Ridge National Laboratory.

The properties of short-fiber composites depend on the dimensions of the reinforcing fiber. The Halpin-Tsai equations [2] are used to predict the longitudinal and transverse properties of aligned short-fiber composites. These simple equations approximate the results of more exact micromechanics analyses. In carbon fibers the principal direction of thermal transport is parallel to the fiber axis (longitudinal). The Halpin-Tsai equations for longitudinal properties are [2]:

$$\frac{\kappa_L}{\kappa_m} = \frac{1 + (2 \cdot l / d) \cdot \eta_L \cdot V_f}{1 - \eta_L \cdot V_f}, \quad (1)$$

where

$$\eta_L = \frac{(\kappa_f / \kappa_m) - 1}{(\kappa_f / \kappa_m) + 2 \cdot (l / d)}, \quad (2)$$

where

κ_L= thermal conductivity of the composite in the longitudinal direction,

κ_m = thermal conductivity of the matrix; this is estimated from carbon/carbon composite data, and is equal to 64.3 W/m·K [3].

κ_f = thermal conductivity of the reinforcing fiber; for a perfect graphite crystal, the thermal conductivity would be 2000 W/m·K. However, the crystallites within the fiber are not perfect graphite. Therefore, the conductivity of the crystallite is assumed to be 2000*cos(Z).

V_f = volume fraction of the reinforcing fiber; the degree of graphitization, g_p, will be utilized as a measurement of the volume fraction of the reinforcing fiber or crystallites.

l = length of the reinforcing fiber; the crystallite dimension along the graphene planes in the direction parallel to the fiber axis, $L_{a//}$, is length of the reinforcing fiber or crystallite.

d = diameter of the reinforcing fiber; $2*((L_{a\perp}*L_c)/\pi)^{1/2}$, where d is the diameter of a given circle of area equal to the cross-sectional area of the crystallite.

Periodic Composite Model

Another approach that may be used to estimate the conductivity of a carbon fiber is to apply homogenization theory. Generally speaking, homogenization theory predicts "an averaged" value of the thermal properties for a composite with a periodic structure, based on the properties of the constitutive components [3, 4]. This theory assumes that the composite is periodic in all directions; the unit, which is periodically repeated within the composite, is called the 'fundamental component'.

The first step in applying homogenization theory is to identify, or make an approximation to, the underlying periodic structure of the composite or 'fundamental component'. For the case of the carbon fibers, two 2-D periodic structures were assumed, and these are shown schematically in Figure 2.

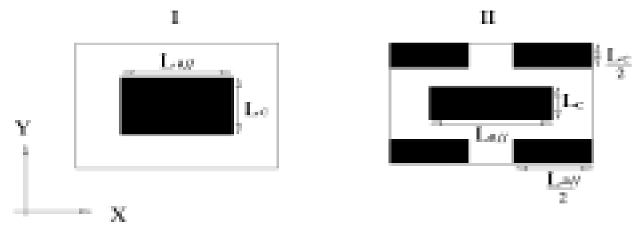

Figure 2. Periodic structures used for the periodic composite model.

Once the periodic structure, $\wp$, has been identified, then matrix K(x,y) denotes the thermal conductivity of $\wp$ at any point (x,y). The homogenized value of the thermal conductivity for the composite, K^o (constant matrix), satisfies the following relation,

$$\lambda \cdot K^o \cdot \lambda = \min_{v \in H^1(\wp)} \frac{1}{Area\ of\ \wp} \int_{\wp} (\lambda + \nabla v) \cdot K \cdot (\lambda + \nabla v) dA, \quad (3)$$

where λ is any 2x1 vector and H($\wp$) denotes a suitable collection of periodic functions on $\wp$ (see [3,4] for details).

Given the structural data for a carbon fiber, an approximation of K(x,y) is constructed. The homogenized matrix K^o can then be estimated from the above equation by using a finite element approximation.

Discussion and Conclusions

The structural data obtained from the ribbon fibers, and commercial round P-series fibers (produced by BP Amoco) are listed in Table 1. These parameters were substituted into the two models. The results obtained are presented in Table 2.

The agreement between the measured and the estimated values of thermal conductivity is fairly good for both ribbon and round fibers. However, the short-fiber composite model seems to give better agreement for the round fibers.

significantly affect the thermal conductivity along the fiber axis.

Figure 3 plots these results versus the experimental values of thermal conductivity for ribbon and round carbon fibers. Figure 3 also shows linear least-squares fits of the measured thermal conductivities compared to those estimated by each of the models. As the figure shows, the periodic composite model using structure II yielded the closest fit to experimental data. The figure also shows that the short-fiber composite model appears to slightly underestimate the thermal conductivity of the carbon fibers.

Table 1. Parameters utilized in the periodic composite model for ribbon and round fibers.

Set	$g_p=V_f$	L_c (Å)	$L_{a//}$ (Å)	$L_{a\perp}$ (Å)	Z (°)
Ribbon					
903-S1	0.7264	132	125	458	3.4
212-S1	0.7964	194	114	421	3.3
212-S4	0.7419	217	111	341	2.8
212-S9	0.8612	152	129	397	2.1
903-S4	0.7968	222	115	369	2.2
Round					
P-25	0.02	26	23	14	32
P-55	0.20	124	136	58	14
P-75	0.34	146	134	84	11
P-100	0.68	227	360	235	5.6
P-120	0.74	251	467	309	5.6

Table 2. Estimated and measured values of thermal conductivity for ribbon and round fibers.

Fiber Set	Thermal Conductivity (W/m·K)			
	Short-fiber composite	Periodic Composite (I)	Periodic Composite (II)	Measured values
Ribbon				
903-S1	328	434	470	321
212-S1	400	555	571	567
212-S4	325	452	461	565
212-S9	591	737	781	696
903-S4	402	554	567	421
Round				
P-25	68	62	31	22
P-55	117	108	110	120
P-75	154	153	159	185
P-100	434	377	463	520
P-120	546	457	590	640

As Table 2 shows, the difference between the results obtained from the periodic composite model using structure I and structure II is not very significant. This indicates that the relative position of the crystallites in the direction perpendicular to the fiber axis does not significantly affect the thermal conductivity along the fiber axis.

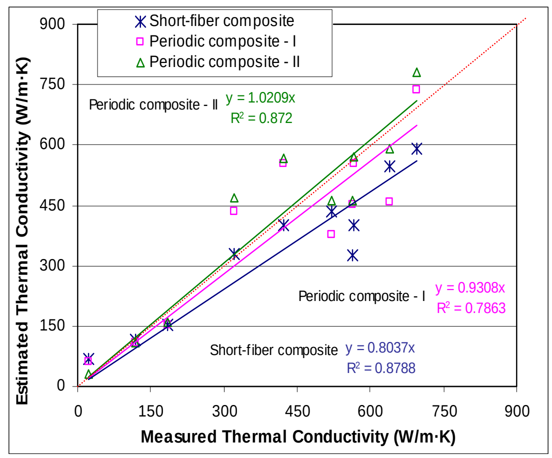

Figure 3. Measured versus estimated values of thermal conductivity for ribbon and round fibers.

Conclusions

Two models that directly relate the structure of carbon fibers to their thermal conductivity were developed. These models demonstrate that it is possible to estimate fiber properties directly from structural measurements. Other relationships, such as the Lavin's and APPI's, are merely property correlations. Thus, they only indirectly reflect the influence of the fiber structure on its properties.

The short-fiber composite model provides a quick and easy way of estimating the thermal conductivity of carbon fibers. However, it is not as accurate as the periodic composite model.

References

[1] Lavin J. G., Boyington D. R., Lahijani J., Nysten B. and Issi J-P., "The correlation of thermal conductivity with electrical resistivity in mesophase pitch-based carbon fiber," Carbon 31 (6) pp. 1001-1002, 1993.

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[4] Ervin, V.J., "Estimating the Thermal Conductivity of Composites using Homogenization," in press.