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MODELING THE THERMAL CONDUCTIVITY OF FIBER-REINFORCED CERAMIC COMPOSITES

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ABSTRACT

A review of models for the prediction of the thermal conductivity of uni-directional fiber-reinforced composites will be presented. The ability of these models to give an accurate prediction of the composite thermal conductivity depends on the amount of information known about the constituent phase properties under the assumption that these properties do not change as a result of processing. Also presented are models that take into account the effects of fiber coatings.

INTRODUCTION

Fiber reinforced composites are an important class of technological materials that have a wide range of applications, from the sporting goods industry to the automotive and aerospace industries. These materials offer improved thermal and mechanical properties over many conventional materials for applications where high strength, high fracture toughness to weight ratios and resistance to mechanical and thermal shock are of prime importance. Depending on the application of the composite, the thermal conductivity may be one factor in the choice of the materials selected to form the final composite.

It is the purpose of this paper to present a review of certain models found in the literature that will enable the determination of the composite thermal conductivity based upon the properties, amounts and other microstructural information of the constituent phases. The degree of complexity of the models presented is determined by this information both prior to and after processing and includes the constant thermal conductivities, volume fractions, fiber and matrix shapes, fiber packing arrangements and geometries, to name a few.

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In the derivation of thermal conductivity models for uni-directional fiber reinforced composite materials certain assumptions are made to make the problem more tractable. These assumptions include but are not limited to a) the composite is macroscopically homogeneous, b) the fiber and matrix are each locally isotropic and homogeneous, c) the constituent properties of each phase are known and have not changed as a result of processing, d) the composite is fully dense and e) no interactions have occurred between the fiber and the matrix, which would lead to the formation of an interfacial boundary layer. Two models that include the effects of an interfacial boundary layer and will be discussed subsequently. Assumptions specific to a particular model will be discussed at the appropriate time.

The thermal conductivity, k , parallel and perpendicular to the fiber direction are of primary importance. For heat flow parallel or along the fiber direction, the effective thermal conductivity, k_a , is written as [1]

$$k_a = v_m k_m + v_f k_f \quad (1)$$

where the v 's are the volume fractions of each phase and the subscripts m and f refer to the matrix and fibers respectively. This equation can be considered as the upper bound to the composite thermal conductivity and is known as the parallel model or the "simple rule of mixtures". A plot of Equation 1 is shown in Figure 1 (curve 1) as a function of fiber volume fraction. The values used for k_m and k_f are 10 and 100 W/(m·K), respectively.

Figure 1 Comparison of various fiber reinforced composite thermal conductivity models.
 (1) Parallel model. (2) Series model. (3) Rayleigh's model or Hashin upper bound.
 (4) Hashin lower bound. (5) Davies' model.

When the heat flow direction is transverse or perpendicular to the fibers the determination of the effective thermal conductivity is much more difficult. As a first approximation the fibers and matrix can be considered to be in series and the thermal conductivity, k_t , is then given by [1]

$$k_t = \frac{1}{\frac{v_m}{k_m} + \frac{v_f}{k_f}} \quad (2)$$

This equation is known as the series model or the "inverse rule of mixtures" and can be considered to be the lower bound of the composite thermal conductivity. A plot of k_t using this model is shown in Figure 1 (curve 2). The combination of the parallel and series models are the maximum and minimum values that the effective thermal conductivity may have and are called the primitive bounds.

Rayleigh [2] proposed a model for k_t that considered a dilute suspension of cylindrical objects arranged in a rectangular array. This expression for the transverse thermal conductivity is written as

$$\frac{k_t - k_f}{k_t + k_f} = v_m \frac{k_m - k_f}{k_m + k_f} \quad (3)$$

As a comparison to the primitive bounds, a plot of Equation 3 is also shown in Figure 1 (curve 3). The result shows that Rayleigh's model falls between the upper and lower primitive bounds and may be considered to give a tighter upper bound than the parallel model.

Hashin [3] has derived bounds which are narrower than the primitive bounds. This model is known as the composite cylindrical assemblage model and his expression for the upper bound in terms of the thermal conductivity is given by

$$k_t = k_f + \frac{v_m}{\frac{1}{k_m - k_f} + \frac{v_f}{2k_f}} \quad (4)$$

It can be shown that Equation 4 is equivalent to Equation 3 by making use of the fact that $v_m + v_f = 1$. The lower bound of Hashin, which is written as

$$k_t = k_m + \frac{v_f}{\frac{1}{k_f - k_m} + \frac{v_m}{2k_m}} \quad (5)$$

may also be determined from Equation 4 by interchanging the subscripts m and f. The lower bound is also plotted in Figure 1 (curve 4). The Hashin bounds give tighter bounds than the series

and parallel models when the only information available is the thermal conductivity and amount of each phase.

Davies [4] has derived an expression for the dielectric constant of fibers embedded in an isotropic matrix. The derivation is in part based on effective medium theory or a self consistent scheme in that each fiber is divided into a series of short segments and the presence of other fibers is taken into account by assuming that each segment is embedded in a matrix having the bulk properties of the composite, such as the dielectric constant or in this case the thermal conductivity. His expression, written in terms of the thermal conductivity, is

$$k_t = k_f + \frac{2 v_m k_t (k_m - k_f)}{k_t + k_m} \quad (6)$$

A plot of Equation 6 is shown in Figure 1 (curve 5). The result is that Davies' model falls between the Hashin bounds. McCullough [5] points out that there is a problem with effective medium theories in that they tend to over estimate the composite thermal conductivity for moderate filler concentrations. The amount of filler depends on the constituent properties and geometrical factors.

Springer and Tsai [6] have derived an expression that requires numerical techniques to determine k_t . They have also derived an approximate expression that depends upon the fiber shape and packing arrangement. For a cylindrical fiber in a square packing array their expression is

$$k_t = k_m(1 - 2\sqrt{v_f/\pi}) + \frac{k_m}{B} \left(\pi - \frac{4}{\sqrt{1 - (B^2 v_f/\pi)}} \tan^{-1} \frac{\sqrt{1 - (B^2 v_f/\pi)}}{1 + \sqrt{B^2 v_f/\pi}} \right) \quad (7)$$

where

$$B = 2 \left(\frac{k_m}{k_f} - 1 \right).$$

By inspection it can be seen that this equation is valid for only a certain range of matrix to fiber thermal conductivity ratios which limits the fiber volume fraction.

Miller [7] has shown that for a general class of two phase composite materials, the bounds on the dielectric constant originally derived by Beran [8] for a three dimensional isotropic homogeneous medium, may be expressed in terms of the volume fractions and two geometrical factors that characterize the geometry of the matrix and filler phases. A cell model was used to characterize the geometry and it was assumed that the medium could be subdivided into cells whose properties are independent of adjacent cells. Beran and Silnutzer [9] have extended this analysis to include k_t for a fiber reinforced composite assuming that the fiber arrangement is statistically homogeneous and isotropic in the plane perpendicular to the fiber direction. The expression for the upper (k_{tu}) and lower (k_{tl}) bounds are

$$k_{tu} = k_m \left\{ 1 + v_f (\alpha - 1) - \frac{0.5 v_f v_m (\alpha - 1)^2}{1 + (\alpha - 1) [v_f + 2 v_m^2 G_f - 2 v_f^2 G_m]} \right\} \quad (8a)$$

$$k_{tl} = \frac{k_f}{v_f + v_m \alpha - \frac{0.25 (1 - \alpha)^2 v_f v_m}{[(\alpha - 1) [v_f^2 G_m - v_m^2 G_f] + 0.5 \alpha + 0.5 (1 - \alpha) v_f]}} \quad (8b)$$

where $\alpha = k_f / k_m$. The geometrical factors, G_m and G_f , are in the range 0.25 to 0.5 inclusively. A value of 0.25 corresponds to circles and 0.5 for two dimensional needles. A comparison of these bounds to those of Hashin are shown in Figure 2 with $G_f = 0.25$ and $G_m = 0.5$.

Figure 2 Comparison of the Beran and Silnutzer bounds, curves 3 and 4, with the Hashin bounds, curves 1 and 2.

As this figure shows, these bounds fall between the Hashin bounds and the lower bound agrees within about 10% with the Hashin bound throughout the entire range of the fiber volume fraction. Beran and Silnutzer point out that the fiber packing arrangement is of significant importance in that a change in the packing geometry may significantly alter k_t .

Elsayed and McCoy [10] extended the analysis of fiber positioning and derived bounds that contain an additional shape factor and two packing parameters.

Han and Cosner [11] presented a numerical technique to determine k_t by analyzing the heat conduction through a representative unit cell composed of rectangular and staggered arrays of fibers with various fiber to matrix thermal conductivities. A diagram of the packing arrangement and unit cells for these two geometries is shown in Figure 3. The results of the analysis for various packing angles, γ , show that for fibers with larger thermal conductivities than the matrix, there is a much higher degree of anisotropy than if the matrix is the higher conductivity material. They also point out that the effective conductivity is highly sensitive to fiber orientation and that k_t / k_m is

Figure 3 Unit cells for the rectangular and staggered fiber packing arrangements of Han and Cosner [11]. γ is the fiber packing angle.

principally governed by the amount of separation of the fibers perpendicular to the direction of heat flow.

Using fractal techniques and a generalized unit cell, Pitchumani and Yao [12] derived an expression for k_t in terms of two parameters that describe the local fractal dimensions transverse and parallel to the direction of heat flow, d_t and d_p , respectively. The generalized unit cell is constructed to include the analysis of both the rectangular and staggered arrays and is based upon the construction of an average Wigner - Seitz cell. This construction was extended to form a hybrid unit cell composed of the rectangular and staggered unit cells in order to derive an expression for the thermal conductivity of random media. The two fractal dimensions provide a method for quantifying the relative fiber arrangements and describe the degree of "connectedness" of the fibers. When $d_p = d_t$ and the packing angle is 45° their expression reduces to that of Springer and Tsai and for other parameters their results compare well with those of other investigators such as Han and Cosner.

During processing, reactions may take place between the fibers and matrix that lead to the formation of an interfacial boundary layer. Another condition may occur if there exists a large thermal expansion mismatch between the fiber and matrix. Gaps may form between the fiber and matrix when the composite is cooled from processing temperatures. These conditions would violate two of the assumptions upon which the previous models were derived. These assumptions were that the constituent properties do not change as a result of processing and that no interactions have occurred between the matrix and fibers. To prevent chemical interactions at the interface of the fiber and matrix, the fiber may be coated with another material. Coatings may also be applied to enhance a property. This new material adds another factor that will affect the properties of the composite and must be included in any theory that describes the composite thermal conductivity. The parallel model for the axial thermal conductivity given by Equation 1 may be modified with the addition of a term that includes the volume fraction and thermal conductivity of the coating. As a first approximation for the transverse thermal conductivity, the series model given by Equation 2

may be modified by including a term for the coating in the denominator of the same form as the matrix and fiber.

Hasselman and Johnson [13] have studied the effects of a thermal boundary resistance by modifying Rayleigh's model for a dilute suspension of cylinders. Their expression for the transverse composite thermal conductivity is

$$k_t = k_m \frac{\left\{ \left(\frac{k_f}{k_m} - 1 - \frac{k_f}{ah_c} \right) v_f + \left(1 + \frac{k_f}{ah_c} + \frac{k_f}{k_m} \right) \right\}}{\left\{ \left(1 + \frac{k_f}{ah_c} - \frac{k_f}{k_m} \right) v_f + \left(1 + \frac{k_f}{ah_c} + \frac{k_f}{k_m} \right) \right\}} \quad (9)$$

where a is the fiber diameter and h_c is the interfacial thermal conductance. When $h_c \rightarrow \infty$ this expression reduces to the Rayleigh model, Equation 3. A plot of Equation 9 for various thermal conductances is shown in Figure 4.

Figure 4 The transverse composite thermal conductivity given by Equation 9 for an interfacial barrier resistance between a dilute suspension of fibers and matrix with a range of values of k_f/ah_c .

This figure shows the effect of the increase of k_f/ah_c from 0, which corresponds to perfect contact between the fiber and matrix, to ∞ , which corresponds to no contact between the fiber and matrix. In this case the interfacial barrier acts like porosity.

Hatta and Taya [14] have developed a theory for the effective thermal conductivity of coated fiber composites that includes the effect of changing the aspect ratio of the fiber. Their results show that if the thermal conductivity of the coating is larger than that of the matrix, the composite thermal

conductivity is higher than without the coating and lower if the coating has a lower thermal conductivity. When the coating thermal conductivity is greater than both the fiber and matrix and the aspect ratio of the fiber increases, the coating is more effective in enhancing the overall composite thermal conductivity. However when the aspect ratio reaches a value that depends upon the thermal conductivity of each phase, the effect of this enhancement decreases.

SUMMARY

In this paper we have presented a review of several composite thermal conductivity models found in the literature for uni-directional fiber reinforced materials. The review includes models that place bounds on the composite thermal conductivity and models that take into account fiber coatings and thermal barrier resistances. These models incorporate the constituent phase properties such as conductivities, volume fractions, packing geometries and fiber and matrix shapes that would help in the selection process of materials for particular applications prior to processing.

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