Effective Thermal Conductivity of a Thin, Randomly Oriented Composite Material

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ABSTRACT

The thermal conductivity of a randomly oriented composite material is modeled using a probabilistic approach in order to determine if a size effect exists for the thermal conductivity at small composite thicknesses. The numerical scheme employs a random number generator to position the filler elements, which have a relatively high thermal conductivity, within a matrix having a relatively low thermal conductivity. The results indicate that, below some threshold thickness, the composite thermal conductivity increases with decreasing thickness, while above the threshold the thermal conductivity is independent of thickness. The threshold thickness increases for increasing filler fraction and increasing $k_f/k_m$, the ratio between the filler and matrix thermal conductivities.
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NOMENCLATURE

$h_0, h_1$ \quad \text{heat transfer coefficients to the cold and hot thermal reservoirs, respectively [W m}^{-2} \text{K}^{-1}]$

$h_c$ \quad \text{thermal contact conductance at the filler surface [W m}^{-2} \text{K}^{-1}]$

$H$ \quad \text{composite thickness [m]}

$k_{eff}$ \quad \text{effective composite thermal conductivity [W m}^{-1} \text{K}^{-1}]$

$k_f$ \quad \text{filler thermal conductivity [W m}^{-1} \text{K}^{-1}]$

$k_m$ \quad \text{matrix thermal conductivity [W m}^{-1} \text{K}^{-1}]$

$k_{mean}$ \quad \text{mean composite thermal conductivity [W m}^{-1} \text{K}^{-1}]$

$l$ \quad \text{element dimension [μm]}

$L$ \quad \text{composite width [m]}

$m$ \quad \text{number of cells (elements) in the thickness direction}

$n$ \quad \text{number of cells (elements) in the width direction}

$n_f$ \quad \text{total number of filler elements}

$q$ \quad \text{total heat flow through the composite [W]}

$R_{i,j}$ \quad \text{thermal resistance of the $i,j$ node [m K W}^{-1}]$

$R_{tot}$ \quad \text{total thermal resistance of the composite [m K W}^{-1}]$

$T_0, T_1$ \quad \text{temperatures of cold and hot thermal reservoirs, respectively [K]}
Predicting the thermal, electrical, and mechanical properties of composite materials is very important to their application, since it is both time consuming and expensive to measure these properties, given the wide possible ranges of filler and matrix materials, the filler orientation and dimensions, and the fraction of the total composite that is occupied by the filler. Numerous studies have been reported which describe methods for calculating the effective thermal (or electrical) conductivities for fiber-reinforced composites [see, e.g., Maewal et al., 1976; Han and Cosner, 1981; Peterson and Fletcher, 1987; and Mottram, 1992]. However, apparently most, if not all, of these investigations focused on geometries in which the composite material was infinite in extent. Since the structure of fiber-reinforced composites is generally periodic, this allowed the use of a "unit cell" approach, in which the effective transport properties of an elemental cell containing representative properties of fiber and matrix material, in an appropriate configuration, are the same as those of the composite medium as a whole. This approach works well, provided that the geometry of the actual composite material at hand is relatively large, so that it contains a sufficient number of the unit cells. This unit-cell approach, however, will not be applicable if the composite geometry is limited in at least one dimension, such that the unit cell cannot be truly representative of the entire composite structure.

Another class of composite materials is those in which the filler, such as the fibers in a fiber-reinforced composite, is randomly oriented throughout the matrix. Examples of this type of composite include in-plane randomly oriented fiber composites [Peterson and Fletcher, 1987], and thermally conductive compounds consisting of a grease or epoxy which contains a large...
number of small particles for enhancing the compound thermal conductivity, such as copper or silver particles. These thermally conductive compounds are generally applied as a thin layer between two solid materials in order to increase the thermal conduction across the interface. Hence, their geometry can be severely restricted in the thickness direction. It is anticipated that the effective compound thermal conductivity, $k_{\text{eff}}$, will be independent of thickness for sufficiently large thicknesses, but that $k_{\text{eff}}$ will vary at smaller thicknesses.

This report is concerned with predicting $k_{\text{eff}}$ for composites containing a randomly oriented filler material using a probabilistic approach. A range of filler and matrix thermal conductivities is examined. Specifically, our analysis determines the minimum thickness necessary to achieve the "bulk" value of $k_{\text{eff}}$, i.e., the minimum thickness needed to produce a $k_{\text{eff}}$ that is independent of sample thickness.

THEORETICAL APPROACH

Due to the typical geometry of a thermal compound application, in which a thin layer of the compound is sandwiched between two solids, the two-dimensional model shown in Fig. 1 is employed. All elements are taken to be square-shaped for simplicity. The matrix elements, having a thermal conductivity $k_m$, are white, while the filler elements, having a thermal conductivity $k_f$, are shaded. The positions of the matrix elements are determined in a random procedure which is discussed below. The dimensions of each square element are $l \times l$. The thickness, $H$, is equal to the number of elements in the thickness direction, $m$, multiplied by $l$. The width, $L$, is equal to the number of elements in the width direction, $n$, multiplied by $l$. In all cases, $L \geq 2H$ in order to minimize any effects due to the left- and right-hand boundaries. The
composite is positioned between two solid materials having temperatures $T_1$ (upper) and $T_0$ (lower). Heat transfer between the solid materials and the composite is characterized by the heat transfer coefficients $h_1$ (upper) and $h_0$ (lower). For generality, a thermal contact conductance, $h_c$ (not shown in Fig. 1), is assumed to exist between the filler elements and any of their neighboring elements.

**Thermal Resistance Network**

Since we are considering only steady-state heat transfer, it is expedient to model the thermal conduction through the composite material using a thermal resistance network. Figure 2 shows a small portion of this network near the lower left-hand corner of the composite. Each node represents the temperature of one of the elements in Fig. 1. The boundary conditions at the left and right-hand sides of the composite, at positions $j = 1$ and $j = n$, are taken to be insulated, so that no heat flows in those directions.

The thermal resistances surrounding the $i,j$ node are shown in Fig. 2, where $R_{ij,U}$ is the "upper" resistance, $R_{ij,R}$ is the "right" resistance, $R_{ij,B}$ is the "bottom" resistance, and $R_{ij,L}$ is the "left" resistance. These resistances are determined by the two nodal elements which they straddle. For example, if the $i,j$ node is a filler element, and the $i,j+1$ node is a matrix element, $R_{ij,R}$ is given by

$$R_{ij,R} = R_{i,j} + R_{i,j+1} \quad (1)$$

where
\[ R_{i,j} = \frac{1}{h_c} + \frac{1}{2k_{i,j}} \] (2)

and

\[ R_{i,j+1} = \frac{1}{2k_{i,j+1}} \] (3)

where in this case \( k_{i,j} = k_f \) and \( k_{i,j+1} = k_m \). Note that the thermal contact conductance, \( h_c \), only appears where there is a filler element. If a filler element occupied the node \( i,j+1 \) rather than a matrix element, then \( h_c \) would also appear in the expression for \( R_{i,j+1} \). The other resistances surrounding the node \( i,j \) are determined in a completely analogous manner, except for the top \( (i = 1) \) and bottom \( (i = m) \) rows of the matrix, where the heat transfer coefficients \( h_1 \) and \( h_0 \) come into play, as shown for the bottom row in Fig. 2.

The system of linear equations is generated in a straightforward manner by summing up all the heat flows for each node and equating to zero. For example, for the \( i,j \) node located in the interior of the matrix, as shown in Fig. 2, we have

\[ \frac{T_{i-1,j} - T_{i,j}}{R_{ij,U}} + \frac{T_{i,j+1} - T_{i,j}}{R_{ij,R}} + \frac{T_{i+1,j} - T_{i,j}}{R_{ij,B}} + \frac{T_{i,j-1} - T_{i,j}}{R_{ij,L}} = 0 \] (4)

One such equation can be written for each node, producing a system of equations of the form \( Cx = B \), where \( C \) is an \( (m \times n) \times (m \times n) \) coefficient matrix, \( B \) is a vector of length \( m \times n \), and \( x \) is the output vector of length \( m \times n \) which contains the nodal temperatures. This matrix equation is solved using LU decomposition [Press et al., 1992], where \( L \) represents "lower triangular" and \( U \) "upper triangular".
Determining the Positions of the Filler Elements

The total number of filler elements, $n_f$, is a function of the specified filler fraction, $\varphi$, where $\varphi$ ranges between 0 and 1:

$$n_f = \varphi \times (m \times n)$$

where $p = m \times n$ is the total number of nodes. We employ a random number generator [Press et al., 1992] to determine which nodes are occupied by the filler. Since the output of the random number generator is between 0 and 1, we divide this range into $p$ intervals:

$$0 < z < \frac{1}{p}, \quad \frac{1}{p} \leq z < \frac{2}{p}, \quad \frac{2}{p} \leq z < \frac{3}{p}, \quad \ldots, \quad \frac{p-1}{p} \leq z < 1$$

where each interval represents one node. The random number generator is then utilized to produce a random number between 0 and 1. The interval in which that number falls is assigned to be a filler node. This process is repeated until all $n_f$ filler nodes are assigned. If, during the course of this procedure, two or more random numbers fall within the same interval, only the first such number is kept, since that node is already assigned to be a filler element. Any subsequent numbers falling within the same interval are discarded, and additional random numbers are generated in order to ensure that exactly $n_f$ nodes are occupied by filler elements.

Determining the Composite Thermal Conductivity

Determining $k_{\text{eff}}$ for the composite material is actually not trivial, even once all the nodal temperatures have been determined. The reason lies in the highly anisotropic nature of the heat flow within the composite. Referring to Fig. 2, recall that the heat transfer coefficients to isothermal reservoirs, rather than the composite temperature itself, are specified at the upper and
lower boundaries. If we assume the thermal resistances due to $h_1$, $h_0$, and the conduction within the composite are in series, we arrive at the following expression for the total thermal resistance, $R_{tot}$, between $T_1$ and $T_0$:

$$R_{tot} = \frac{1}{h_1L} + \frac{H}{k_{eff}L} + \frac{1}{h_0L}$$

Intuitively, Eq. (7) seems reasonable, because we can imagine that we should be able to assign well-defined values for each of the three terms on the right-hand side of Eq. (7). However, in reality, for finite values of $h_1$ and $h_0$, there is no well-defined single temperature on the upper and lower boundaries of the composite, and hence the assumption implicit in Eq. (7) that these three resistances lie in series is not rigorously correct. This problem, however, is averted for the results reported here, since we take both $h_1$ and $h_0$ as tending towards infinity, which yields uniform upper and lower boundary temperatures for the composite. The resolution of this issue when $h_1$ and $h_0$ are assigned finite values is left for the future.

The composite thermal conductivity, $k_{eff}$, is determined here by first calculating $R_{tot}$ using

$$R_{tot} = \frac{T_1 - T_0}{q}$$

where $q$ is the total heat flow through the composite. In practice, $q$ is determined by summing all the individual nodal heat flows at either the upper or lower composite boundary, which must yield equal values of $q$. In fact, the consistency of the numerical scheme is checked by comparing $q$ calculated at the upper boundary with that calculated at the lower boundary, and ensuring that the values are equivalent.
Finally, $k_{\text{eff}}$ is determined by equating the value for $R_{\text{tot}}$ calculated from Eq. (8) with Eq. (7), yielding

$$k_{\text{eff}} = \frac{H}{LR_{\text{tot}} \frac{1}{h_1} - \frac{1}{h_0}}$$

(9)

Once $k_{\text{eff}}$ is calculated in this manner for a single set of random numbers, and hence filler positions, this procedure is repeated a number of times, with a new set of random numbers assigned for each repetition. This yields a distribution of $k_{\text{eff}}$ values, from which a mean value of $k_{\text{eff}}$ is calculated ($k_{\text{mean}}$). The value of most interest is $k_{\text{mean}}$, since that is what is most likely to occur in a real application. A total of 200 iterations are performed to determine $k_{\text{mean}}$. An example of the $k_{\text{eff}}$ distribution for $\varphi = 0.20$, $m = 20$, $n = 40$, $k_m = 0.2$ W m$^{-1}$ K$^{-1}$, and $k_f = 400$ W m$^{-1}$ K$^{-1}$ is shown in Fig. 3. These particular values of $k_m$ and $k_f$ correspond to a thermal grease matrix with copper particles serving as the filler. For this particular case, $k_{\text{mean}}$ is 0.2975 W m$^{-1}$ K$^{-1}$. It should be noted that the distribution in Fig. 3 is by no means typical. For larger values of $\varphi$, and/or small values of $m$, especially where $k_f \gg k_m$, occasional large values of $k_{\text{eff}}$ result which skew the distributions towards the higher values. Such outlying points would presumably become less and less relevant as the total number of iterations is increased. However, in the interests of maintaining a reasonable computation time while achieving satisfactory accuracy, the number of iterations for the present results is fixed at 200. The error caused by this limitation, as well as from other sources, is discussed in the next section.
Numerical Uncertainty

There are essentially four contributions to error in our numerical scheme: (i) insufficient number of iterations to achieve a satisfactory distribution; (ii) insufficient $L/H$ ratio to eliminate edge effects; (iii) errors originating from the random number generator; and (iv) roundoff and truncation errors. For (i), this error seems most severe where $k_f$ differs greatly from $k_m$. A comparison between a test run of 1000 iterations with our standard run of 200 iterations indicates an error as high as ±10%. For (ii), sample calculations indicate that maintaining $L/H \geq 2$ results in an error of less than ±1%. For (iii), the random number generator employed is satisfactory, according to Press et al. (1992), except for an extraordinarily high number of calls, on the order of $10^8$. In the present case, the maximum number of calls is on the order of $10^5$, so that we neglect any error due to the random number generator. Finally, for (iv), these errors are difficult to estimate, but are thought to be negligible compared with error (i). Therefore, the error caused by an insufficient number of iterations is the dominant contributor, and we thus estimate the total numerical uncertainty at ±10%.

RESULTS AND DISCUSSION

The results shown below indicate the effects of filler fraction ($\varphi$), matrix and filler thermal conductivities ($k_m$ and $k_f$), filler thermal contact conductance ($h_c$), and filler size $l$ on the mean composite thermal conductivity ($k_{\text{mean}}$), as determined from an average of 200 iterations. In each case, $k_{\text{mean}}$ is plotted versus the composite thickness, as represented by the number of elements, or cells, in the thickness direction. All calculations are performed for $h_0 = h_1 = 10^{30}$ W m$^{-2}$ K$^{-1}$, i.e., effectively infinite, $T_1 = 1$ K, and $T_0 = 0$ K. In each case the ratio $L/H$ is at least
2, and for smaller values of \( m \), \( L/H \) is set at 5 so that edge effects due to the left and right boundaries can be neglected.

**Effect of Filler Fraction (\( \varphi \))**

Figure 4 presents \( k_{\text{mean}} \) as a function of \( \varphi \). The matrix and filler thermal conductivities are \( k_m = 0.2 \text{ W m}^{-1} \text{ K}^{-1} \) and \( k_f = 400 \text{ W m}^{-1} \text{ K}^{-1} \), respectively. These values correspond approximately to a matrix consisting of thermal grease, such as one of the Apiezon greases, and the filler consisting of copper particles, all at room temperature. Any effect of filler contact resistance is neglected here by taking \( h_c = 10^{30} \text{ W m}^{-2} \text{ K}^{-1} \).

Figure 4 shows the anticipated results, in that at large thicknesses, the curves generally approach a horizontal asymptote which signifies that \( k_{\text{mean}} \) becomes independent of thickness at that point. At small thicknesses, \( k_{\text{mean}} \) tends to increase with decreasing thickness. Thus, each curve can be divided into two regions: the "small" region at lower thicknesses, where \( k_{\text{mean}} \) varies with thickness, and the "bulk" region at higher thicknesses, where \( k_{\text{mean}} \) is independent of thickness. The thickness at which the "bulk", or asymptotic, behavior is reached increases with increasing \( \varphi \). For \( \varphi = 0.20 \), the boundary between the "small" and "bulk" regions occurs near \( m = 7 \), that is, for a composite thickness of only 7 elements, or cells. The size of each element, \( l \times l \), is determined by the average filler size, so that if the filler consists of solid copper particles, the "bulk" region is obtained for a thickness of 7 copper particles. For \( \varphi = 0.40 \), the boundary between the two regions occurs near \( m = 13 \), and for \( \varphi = 0.60 \), the boundary apparently lies beyond \( m = 25 \). Therefore, the region in which the "small" size effect strongly impacts \( k_{\text{mean}} \) becomes much larger with increasing \( \varphi \).
What causes $k_{\text{mean}}$ to increase at small thicknesses? First of all, note that this trend is in agreement with a study on percolation in a unidirectional fiber composite consisting of perfectly conducting, circular fibers enmeshed within a nonconducting matrix [Joy and Strieder, 1979]. Our model differs from the percolation model in that both the matrix and the filler have a finite conductivity, which allows conduction even without the percolation mechanism. However, we believe that a percolation type of mechanism is responsible for the enhanced thermal conduction at small thicknesses. It seems that as the thickness is decreased, there is a greater propensity for the filler particles to "line up", thus creating a relatively high thermal conductivity path, or thermal "short", which increases the overall composite thermal conductivity. The probability for this kind of filler alignment apparently decreases with increasing thickness, such that beyond some given thickness—the boundary between the "small" and "bulk" regions—none of these high thermal conductivity paths can occur, yielding a $k_{\text{mean}}$ which is thereafter independent of composite thickness.

For consistency, the "bulk" values of $k_{\text{mean}}$ calculated for large thicknesses should agree with standard estimates of the thermal conductivity of composite materials, given known values of $\varphi$, $k_m$, and $k_f$. Unfortunately, however, there is no single generally accepted formula that can be used to provide a reliable estimate of $k_{\text{mean}}$. Two approaches that provide upper and lower bounds to $k_{\text{mean}}$ are to (i) assume the matrix and filler lie in series (lower bound), and (ii) assume the matrix and filler lie in parallel (upper bound) (Peterson and Fletcher, 1987). The corresponding relation for the series approximation is
\[ k_{\text{mean}} = \frac{1}{\varphi \left( \frac{1}{k_f} + \frac{1}{k_m} \right)} \]  \hspace{1cm} (10)

and that for the parallel approximation is

\[ k_{\text{mean}} = \varphi k_f + (1 - \varphi) k_m \] \hspace{1cm} (11)

A comparison of the values calculated from each of these relations, together with the bulk, or large-thickness, values taken from Fig. 4, is provided in Table 1. It is seen that the bulk values taken from Fig. 4 always lie between the bounds calculated from Eqs. (10) and (11), thus demonstrating, as much as possible, the consistency of the present results for large thicknesses.

So far, nothing has been said concerning any effect of element size, \( l \), on \( k_{\text{mean}} \). With reference to Eq. (2), \( l \) impacts the thermal resistance \( R_{i,j} \) only in the term involving the filler contact conductance \( h_c \). Therefore, where \( h_c \to \infty \), as in the results in Fig. 4, changing \( l \) has no effect on \( k_{\text{mean}} \). Later in this report, the effect of varying \( l \) for finite \( h_c \) will be evaluated.

Finally, note the small fluctuations in the curves in Fig. 4, which are especially apparent for the \( \varphi = 0.20 \) and \( \varphi = 0.40 \) curves. These fluctuations are the result of the probabilistic nature of our theoretical approach. As discussed in the previous paragraph, there is some possibility, at small thicknesses, for the filler particles to align themselves to create a high thermal conductivity path. This can result in a value for \( k_{\text{eff}} \) that is a factor of 15 greater than \( k_{\text{mean}} \), thus skewing the distribution and yielding a relatively high value for \( k_{\text{mean}} \). Upon further investigation, it is revealed that this effect is somewhat dissipated by taking a greater number of iterations to determine \( k_{\text{mean}} \). However, in an actual composite material, there is also some probability for
the highly conducting particles to align themselves in this way, so that the fluctuations predicted
by our theoretical model could also occur. Predicting exactly when and where such fluctuations
will occur, however, is impossible because of the random nature of the composite.

Effect of $k_m$ and $k_f$

The composite thermal conductivity $k_{mean}$ is plotted versus thickness for three different
sets of $k_m$ and $k_f$ in Fig. 5. For each curve, $\phi = 0.60$, $l = 5 \mu m$, and $h_0 = h_1 = h_c = 10^{30}$ W m$^{-2}$
K$^{-1}$. The upper curve corresponds to a copper filler within a grease matrix, like the results in
Fig. 4. The middle curve, where $k_m = 0.19$ W m$^{-1}$ K$^{-1}$ and $k_f = 46$ W m$^{-1}$ K$^{-1}$, corresponds to a
polymide matrix with an alumina filler. The lowest curve, where $k_m = 0.3$ W m$^{-1}$ K$^{-1}$ and $k_f = 1.0$ W m$^{-1}$ K$^{-1}$, corresponds to the G-10 composite, which consists of woven glass fibers within
an epoxy matrix. Note, however, that since G-10 is a woven, not a random, composite, these
results will not apply specifically to G-10. These three combinations of $k_m$ and $k_f$ are chosen to
provide a broad range of thermal conductivities, while still being relevant for some practical
applications.

Although all three curves indicate that $k_{mean}$ increases with decreasing thickness, the
effects on the two upper curves are most pronounced. For the lowest curve, where $k_m$ and $k_f$
differ by only a factor of three, there is only a very slight increase in $k_{mean}$ at lower thicknesses.
Apparently, $k_m$ and $k_f$ must be sufficiently different to achieve a significant size effect on $k_{mean}$
at small thicknesses. Furthermore, the change in $k_{mean}$ between its maximum and minimum
values increases with increasing difference between $k_f$ and $k_m$, or $k_f/k_m$. For the upper curve
where \( k_f/k_m = 2000 \), \( k_{mean} \) changes by a factor of 7.5, while for the middle curve \( (k_f/k_m = 242) \), \( k_{mean} \) changes by only a factor of 4.5.

For the upper curve, the horizontal asymptote is still not reached for \( m = 25 \), indicating that the boundary between the "small" and "bulk" regions lies at still greater thicknesses. For the middle curve, the boundary occurs near \( m = 12 \), and for the lowest curve, at \( m < 5 \). Thus, it is clear that the greater the difference between \( k_m \) and \( k_f \) as measured by the ratio \( k_f/k_m \), the greater the thickness over which the size effect on \( k_{mean} \) is important.

**Effect of Filler Contact Resistance**

Figure 6 shows the effect of varying \( h_c \) on \( k_{mean} \). The material properties assumed are the same as those in Fig. 4: \( k_m = 0.2 \) W m\(^{-1}\) K\(^{-1}\) and \( k_f = 400 \) W m\(^{-1}\) K\(^{-1}\), while \( \phi = 0.40 \). As anticipated, as \( h_c \) decreases (i.e., the thermal contact resistance between the filler particles and the matrix or other filler particles increases), \( k_{mean} \) decreases. For the thermal grease/copper particle combination considered here, the actual value of \( h_c \) at room temperature would probably be relatively high—perhaps close to \( 10^7 \) W m\(^{-2}\) K\(^{-1}\), or even higher.

Three of the curves in Fig. 6 display the size effect on \( k_{mean} \) at small thicknesses. However, one curve, that for \( h_c = 10^5 \) W m\(^{-2}\) K\(^{-1}\), is essentially flat. The reason for this can be understood by examining Eq. (2), the relation for \( R_{i,j} \). For \( h_c = 10^5 \) W m\(^{-2}\) K\(^{-1}\), the first term on the right-hand side, \( 1/h_c l \), is equal to 2.0 m K W\(^{-1}\), while the second term, \( 1/2k_{i,j} \), is equal to 0.00125 m K W\(^{-1}\). The sum of these terms is almost equal to the thermal resistance of the matrix, or \( 1/2k_m = 2.5 \) m K W\(^{-1}\). Since the matrix nodes and filler nodes have approximately
the same resistance, the distribution of filler nodes is insignificant, and hence there is no observable size effect on $k_{\text{mean}}$.

It was briefly mentioned earlier that, for $h_c \rightarrow \infty$, there is no impact of varying $l$ on $k_{\text{mean}}$. For the finite values of $h_c$ in Fig. 6, however, the choice of $l$ is important, as it would be in practical applications where $h_c$ will always be some finite value. The next and final result section explores how varying $l$ at finite values of $h_c$ impacts $k_{\text{mean}}$.

**Effect of Filler Size ($l$)**

Figure 7 demonstrates that increasing the size of the filler particles, as measured by the length $l$, tends to both increase the bulk value of $k_{\text{mean}}$, as well as to increase the size effect on $k_{\text{mean}}$ at smaller thicknesses. The filler contact conductance $h_c$ is fixed at $10^7$ W m$^{-2}$ K$^{-1}$, and $\varphi$ is set at 0.40. Note that since the thickness is measured in terms of the number of cells, increasing $l$ increases the actual thickness of the composite.

As described earlier, $l$ impacts the thermal resistance only through the first term in Eq. (2), $1/h_c l$. Therefore, increasing $l$ tends to lower $R_{i,j}$ for the filler particles, by making $h_c$ less and less important relative to $k_f$. Since $R_{i,j}$ decreases, $k_{\text{mean}}$ increases, as demonstrated in Fig. 7. Because the same term in Eq. (2) is affected, the trends in Fig. 7 are qualitatively similar to those in Fig. 6.

**DISCUSSION**

One simplifying feature of the present analysis is the assumption of square-shaped filler particles. In reality, the filler particles, especially in thermal compound applications, tend to be
spherical, thus reducing the area of contact between touching particles and possibly reducing the size effect demonstrated here. For the future we can identify three unresolved matters concerning the thermal conductivity of thin, random composites: (i) the effect of spherical, or other non-square, particles on $k_{\text{mean}}$; (ii) the effect of finite $h_0$ and $h_1$ on $k_{\text{mean}}$; and (iii) experimental verification of the size effect on $k_{\text{mean}}$ predicted by our model. Our forthcoming work will focus on these three areas, including an experiment on a commonly used thermal compound, in which the compound thickness will be carefully controlled and measured to produce thermal conductivity data that can be directly compared with our model.

CONCLUSIONS

A probabilistic model of the thermal conductivity of a two-component, randomly-oriented composite material indicates that a significant size effect exists for the thermal conductivity at small thicknesses. Specifically, below some threshold thickness beyond which the thermal conductivity is independent of thickness, the thermal conductivity increases with decreasing thickness. This effect appears to be due to the increased probability that for small thicknesses the filler particles, which have a relatively high thermal conductivity relative to the matrix, are able to align themselves in the thickness direction, thus providing a high-thermal-conductivity path which increases the overall composite conductivity. The threshold thickness below which the size effect is important increases with increasing filler fraction and increasing $k_f/k_m$, the ratio between the filler and matrix thermal conductivities.

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REFERENCES


Table and Figure Captions

Table 1.  Limiting values of $k_{mean}$ for the conditions shown in Fig. 4, in W m$^{-1}$ K$^{-1}$.

Figure 1.  Schematic thermal model of the two-dimensional, randomly oriented composite material.

Figure 2.  Lower left-hand portion of the thermal resistance network.

Figure 3.  Histogram of $k_{eff}$ values for $k_m = 0.2$ W m$^{-1}$ K$^{-1}$, $k_f = 400$ W m$^{-1}$ K$^{-1}$, $\varphi = 0.20$, $m = 20$, $n = 40$, $h_0 = h_1 = h_c = 10^{30}$ W m$^{-2}$ K$^{-1}$, and $l = 5$ $\mu$m.

Figure 4.  Effect of varying fiber fraction $\varphi$ on $k_{mean}$ ($k_m = 0.2$ W m$^{-1}$ K$^{-1}$, $k_f = 400$ W m$^{-1}$ K$^{-1}$, $h_0 = h_1 = h_c = 10^{30}$ W m$^{-2}$ K$^{-1}$, and $l = 5$ $\mu$m).

Figure 5.  Effect of varying fiber and matrix thermal conductivities on $k_{mean}$ ($\varphi = 0.60$, $h_0 = h_1 = h_c = 10^{30}$ W m$^{-2}$ K$^{-1}$, and $l = 5$ $\mu$m).

Figure 6.  Effect of varying the filler contact conductance on $k_{mean}$ ($k_m = 0.2$ W m$^{-1}$ K$^{-1}$, $k_f = 400$ W m$^{-1}$ K$^{-1}$, $\varphi = 0.40$, $h_0 = h_1 = 10^{30}$ W m$^{-2}$ K$^{-1}$, and $l = 5$ $\mu$m).

Figure 7.  Effect of varying the filler size on $k_{mean}$ ($k_m = 0.2$ W m$^{-1}$ K$^{-1}$, $k_f = 400$ W m$^{-1}$ K$^{-1}$, $\varphi = 0.40$, $h_0 = h_1 = 10^{30}$ W m$^{-2}$ K$^{-1}$, and $h_c = 10^{7}$ W m$^{-2}$ K$^{-1}$).
Table 1. Limiting values of $k_{mean}$ for the conditions shown in Fig. 4, in W m$^{-1}$ K$^{-1}$.

<table>
<thead>
<tr>
<th>$\varphi$ (from Fig. 4)</th>
<th>Bulk</th>
<th>Series Approximation</th>
<th>Parallel Approximation</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.20</td>
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<td>0.250</td>
<td>80.2</td>
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<tr>
<td>0.40</td>
<td>0.658</td>
<td>0.333</td>
<td>160.1</td>
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<tr>
<td>0.60</td>
<td>19.8</td>
<td>0.500</td>
<td>240.1</td>
</tr>
</tbody>
</table>
Figure 1. Schematic thermal model of the two-dimensional, randomly oriented composite material.
Figure 2. Lower left-hand portion of the thermal resistance network.
Figure 3. Histogram of $k_{\text{eff}}$ values for $k_m = 0.2 \text{ W m}^{-1} \text{ K}^{-1}$, $k_f = 400 \text{ W m}^{-1} \text{ K}^{-1}$, $\varphi = 0.20$, $m = 20$, $n = 40$, $h_0 = h_1 = h_c = 10^{30} \text{ W m}^{-2} \text{ K}^{-1}$, and $l = 5 \mu\text{m}$.
Figure 4. Effect of varying fiber fraction $\phi$ on $k_{\text{mean}}$ ($k_m = 0.2$ W m$^{-1}$ K$^{-1}$, $k_f = 400$ W m$^{-1}$ K$^{-1}$, $h_0 = h_1 = h_c = 10^{30}$ W m$^{-2}$ K$^{-1}$, and $l = 5$ $\mu$m).
Figure 5. Effect of varying fiber and matrix thermal conductivities on $k_{\text{mean}}$ ($\varphi = 0.60$, $h_0 = h_1 = h_c = 10^{30}$ W m$^{-2}$ K$^{-1}$, and $l = 5 \mu$m).
Figure 6. Effect of varying the filler contact conductance on $k_{\text{mean}}$ ($k_m = 0.2 \, \text{W} \, \text{m}^{-1} \, \text{K}^{-1}$, $k_f = 400 \, \text{W} \, \text{m}^{-1} \, \text{K}^{-1}$, $\varphi = 0.40$, $h_0 = h_1 = 10^{30} \, \text{W} \, \text{m}^{-2} \, \text{K}^{-1}$, and $l = 5 \, \mu\text{m}$).
Figure 7. Effect of varying the filler size on $k_{\text{mean}}$ ($k_m = 0.2 \text{ W m}^{-1} \text{ K}^{-1}$, $k_f = 400 \text{ W m}^{-1}$ K$^{-1}$, $\phi = 0.40$, $h_0 = h_1 = 10^{30} \text{ W m}^{-2} \text{ K}^{-1}$, and $h_C = 10^7 \text{ W m}^{-2} \text{ K}^{-1}$).