DESIGN OF PARTICLE REINFORCED HEAT CONDUCTING
COMPOSITES WITH INTERFACIAL THERMAL BARRIERS

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Design of particle reinforced heat conducting composites with interfacial thermal barriers

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Abstract

Two phase particle reinforced heat conducting composites are considered. We treat the case when there is an interfacial thermal barrier between phases. We provide rules of thumb for selecting the particle size distribution and minimum particle size when designing particle reinforced composites for optimal heat dissipation. The rules are based on new energy dissipation inequalities obtained in the work of Lipton (1996, Journal of Applied Physics, 80:5583-5586.)

Key Words. Heat conduction, interfacial thermal barriers, size effects.

1 Introduction

The effect of particle size on the thermal energy dissipated inside a particle reinforced composite conductor is addressed. We consider the technologically important case when there is an interfacial thermal barrier resistance between phases. In the context of electronic packaging, it is necessary for the packaging material to efficiently transport heat away from the device. Packaging made from an electrically insulating matrix material with particles or fibers of high thermal conductivity are attractive for this purpose, [1].

Experiments show that for small particles, the presence of an interfacial barrier can diminish or even negate the effect of a highly conducting reinforcement, see, [2] and [3]. This phenomena is in striking contrast to what occurs for perfectly bonded composites where there is no interfacial thermal barrier. Indeed, for perfectly bonded composites it is known, that the addition of highly conducting particles will always reduce the total heat dissipation independently of particle size. In this note we provide rules of thumb for selecting the particle size distribution and minimum particle size when designing a particle reinforced composite.

The thermal conductivity associated with the reinforcement is denoted by $c_r$ and that of the matrix by $c_m$. Here both conductors are assumed isotropic, and $c_r$, $c_m$ are scalar quantities. The reinforcement is assumed to have a better heat conductivity than the matrix, i.e., $c_r > c_m$. The interfacial thermal barrier is characterized by a scalar $\beta$ with dimensions of conductivity per unit length.

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The composite domain is denoted by Ω and its volume is given by |Ω|. The resistivity inside the composite is described by, \( c^{-1}(x) \) taking the values \( c_r^{-1} \) in the particles and \( c_m^{-1} \) in the matrix. For any vector \( \vec{j} \) in \( \mathbb{R}^3 \) we prescribe a heat flux \( \vec{j} \cdot n \) on the boundary of Ω and the thermal energy dissipated inside the composite is \( c_e^{-1} \vec{j} \cdot \vec{j} \) where,

\[
\frac{c_e^{-1} \vec{j} \cdot \vec{j}}{c_r^{-1}} = \min_{j \in V} \{ C(j) \}
\]  

with

\[
C(j) = |Ω|^{-1} \left\{ \int_Ω c^{-1}(x) |j|^2 dx + \beta^{-1} \int_Γ (j \cdot n)^2 ds \right\},
\]

and

\[
V = \{ j : \int_Ω |j|^2 dx < \infty, \text{div} j = 0, \, j \cdot n = \vec{j} \cdot n \text{ on } \partial Ω \}.
\]

Here \( ds \) is the element of surface area, and the vector \( n \) is the unit normal pointing into the matrix phase. The minimizer \( j_{A_r} \) is precisely the heat flux in the composite and is related to the temperature \( u_{A_r} \) by the constitutive law: \( j_{A_r} = c(x) \nabla u_{A_r} \). The constant tensor \( c_e \) represents the effective conductivity of the composite.

We write down the geometric criterion that determines when the effects of the interfacial thermal barrier over the benefit of a highly conducting reinforcement. This criterion is general and applies to any reinforcement. In order to give the criterion, we introduce the scalar \( R_{cr} \) given by:

\[
R_{cr} = \beta^{-1}(c_m^{-1} - c_r^{-1})^{-1}
\]

Here \( R_{cr} \) has dimensions of length. This quantity provides a measure of the relative magnitude of the interfacial barrier resistance with respect to the mismatch between the resistivity tensors of the matrix and reinforcement. For a given particle or fiber reinforcement denoted by "\( \Sigma \)" the geometric parameter of interest is its second Stekloff eigenvalue \( \rho_2 \).

The second Stekloff eigenvalue has dimensions of conductivity per unit length and is given by:

\[
\rho_2 = \min_{\text{div}(c_r \nabla \varphi) = 0} \frac{\int_{\partial \Sigma} \frac{c_r \nabla \varphi \cdot n}{c_r}^2 ds}{\int_{\Sigma} c_r \nabla \varphi \cdot \nabla \varphi dx},
\]

cf., Kuttler and Sigillito [4]. The Stekloff eigenvalue is a ratio measuring the relative importance between the particle’s ability to dissipate heat and the total heat flux leaving through the particle boundary. For spheres filled with an isotropic conductor this ratio is proportional to the reciprocal of the sphere radius and is given by \( \rho_2 = \frac{c_r}{a} \).

We consider the replacement of matrix material with a particle \( \Sigma \) of conductivity \( c_r \) and denote the associated effective conductivity tensor by \( \hat{c}_e \). The criterion on the particle geometry is given in the following theorem recently established in [5]:

**1.1 Energy Dissipation Inequality**

Given a reinforcement particle "\( \Sigma \)" if \( \rho_2 \) satisfies,

\[
R_{cr}^{-1} \leq c_r^{-1} \rho_2,
\]

then

\[
c_e \geq \hat{c}_e.
\]
2 Rules of thumb on minimum particle dimensions for suspension design

It follows from the energy dissipation inequality that if both conducting phases are isotropic and if $\Sigma$ is a sphere of radius $a$ that:

2.1 Size effect for spheres

$$c_e \geq \tilde{c}_e$$

if

$$a \leq R_{cr} = \beta^{-1}(c_m^{-1} - c_r^{-1})^{-1}. \quad (2.2)$$

This inequality motivates the following:

2.1 For polydisperse suspensions of spheres, the best conductivity properties are obtained from suspensions consisting only of spheres with radii greater than or equal to $R_{cr}$

More generally, we consider starlike inclusions $\Sigma$ filled with isotropic conductor $c_r$ embedded in an isotropic matrix with conductivity $c_m$. We suppose $\Sigma$ is starlike for the point "x" inside $\Sigma$ and denote the minimum distance from the point "x" to a tangent plane on the particle boundary by $h_m(x)$. The maximum and minimum distance from "x" to the particle boundary are denoted by $r_M(x)$ and $r_m(x)$ respectively. We apply the isoperimetric inequalities of Bramble and Payne [6] to estimate $\rho_2$ from below:

$$c_r^{-1}\rho_2 \geq \frac{1}{r_M^2} \left[ \left( \frac{r_m}{r_M} \right)^2 \frac{h_m}{r_M} \right]$$

2.2 Size effect for ellipsoidal reinforcement

Given an ellipsoidal reinforcement $\Sigma$ with major and minor axes specified by $a$ and $c$ respectively, then:

$$c_e \geq \tilde{c}_e$$

if

$$a \left( \frac{a}{c} \right)^3 \leq R_{cr} \quad (2.5)$$

This inequality motivates the following:

2.2 When constructing suspensions of particles made from ellipsoids one does best using only those with major and minor axes for which $a \left( \frac{a}{c} \right)^3 \geq R_{cr}$.

Next we consider cylindrical inclusions of length $\ell$ and radius $R$. If $\ell/2 \geq R$ then $r_M = ((\ell/2)^2 + R^2)^{1/2}$ and $r_m = h_m = R$. On the other hand if $\ell/2 \leq R$, then: $r_m = h_m = \ell/2$. (For both cases we have chosen the reference point to be the center of mass for the cylinder.) Such inclusions can be used to model chopped fiber suspensions. We have:
2.3 Size effect for cylindrical inclusions with $\ell/2 \geq R$:

$$c_e \geq c_e, \quad (2.6)$$

if

$$\frac{((\ell/2)^2 + R^2)^2}{R^3} \leq R_{cr}. \quad (2.7)$$

2.4 Size effect for cylindrical inclusions with $\ell/2 \leq R$:

$$c_e \geq c_e, \quad (2.8)$$

if

$$\frac{8((\ell/2)^2 + R^2)^2}{\ell^3} \leq R_{cr}, \quad (2.9)$$

Rules of thumb for the design of chopped fiber reinforced composites follow immediately from these inequalities.

We remark that the physical dimensions of the composite domain $\Omega$ enter into the design problem. Indeed, it follows from the inequalities (2.6), (2.7) and (2.8), (2.9) that:

2.3 If the dimensions of the domain are such that only fibers with $\ell/2 \geq R$ satisfying (2.7) or fibers with $\ell/2 \leq R$ satisfying (2.9) can be placed inside $\Omega$, then one obtains the best results by not reinforcing at all.

3 Rules of thumb based on the size distribution of particles

We introduce design criteria based upon the size distribution of particles. The region occupied by the reinforcement particles is denoted by $A$ and the union of all particle matrix interfaces is denoted by $\Gamma$. We introduce the surface energy tensor $M$ defined by:

$$M_{ij} = |A|^{-1} \int_{\Gamma} n_i n_j ds. \quad (3.1)$$

Here, $n_i$ is the $i^{th}$ component of the outward pointing unit normal on the particle matrix interface. For a heat flux of the form $\vec{j} \cdot n$ prescribed on the boundary of the composite domain we have the following criterion the particle reinforced configuration.

3.1 Reinforcement Criterion

If

$$\frac{(M_j \cdot \vec{j})}{|\vec{j}|^2} \leq R_{cr}^{-1} \quad (3.2)$$

then the energy dissipated inside the reinforced composite is less than the energy dissipated when there is no reinforcement, (ie., $c_e^{-1} \vec{j} \cdot \vec{j} \leq c_m^{-1} \vec{j} \cdot \vec{j}$).

We set $\lambda_M$ to be the largest eigenvalue of $M$. Since,

$$\lambda_M = \max_{\vec{j} \in R^3} \frac{(M_j \cdot \vec{j})}{|\vec{j}|^2} \quad (3.3)$$
it follows immediately that if a reinforcement configuration satisfies:

\[ \lambda_M \leq R_{cr}^{-1} \]  \hspace{1cm} (3.4)

then

\[ c_e \geq c_m I, \]  \hspace{1cm} (3.5)

where \( I \) is the 3 \times 3 identity matrix. We now apply these observations and consider a suspension made from isotropically conducting spheres of different radii embedded in a matrix of isotropic conductivity. We suppose that we know the volume distribution of sphere radii within the suspension. For a polydisperse suspension of spheres with radii \( a_1, a_2, \ldots, a_N \) we suppose that the volume occupied by spheres of radius \( a_i \) is given by the function \( V(a_i) \) where \( \sum_{i=1}^{N} V(a_i) = |A| \). For a prescribed volume distribution function \( V(a) \) we write the mean of the reciprocal radii as:

\[ < a^{-1} > = |A|^{-1} \sum_{i=1}^{N} a_i^{-1} V(a_i), \]  \hspace{1cm} (3.6)

For this case calculation gives

\[ M_{ij} = < a^{-1} > I_{ij}. \]  \hspace{1cm} (3.7)

For polydisperse suspensions of spheres (3.4), (3.5), and (3.7) imply:

if \( < a^{-1} >^{-1} \geq R_{cr} \),

then \( c_e \geq c_m I, \)  \hspace{1cm} (3.8)

where \( c_m \) is the matrix conductivity.

This motivates the following:

\textbf{3.1 Reinforced polydisperse suspensions of spheres with size distributions satisfying:}

\[ < a^{-1} >^{-1} \geq R_{cr} \]  \hspace{1cm} (3.10)

\textit{have better overall conductivity properties than the unreinforced conductor.}

Last we show how to establish the reinforcement criteria. We start by writing the energy dissipation inside the reinforced composite as:

\[ c_e^{-1} \tilde{j} \cdot \tilde{j} = \min_{j \in V} \{ C(j) \} \]  \hspace{1cm} (3.11)

with

\[ C(j) = |\Omega|^{-1} \left\{ \int_{\Omega} c_m^{-1} \tilde{j} \cdot \tilde{j} dx - \int_A (c_m^{-1} - c_r^{-1}) \tilde{j} \cdot \tilde{j} dx + \beta^{-1} \int_{\Gamma} (\tilde{j} \cdot \tilde{n}) ds \right\}. \]  \hspace{1cm} (3.12)

Next the energy dissipated inside the unreinforced domain is given by:

\[ c_m^{-1} \tilde{j} \cdot \tilde{j} = \min_{j \in V} \tilde{C}(j) \]  \hspace{1cm} (3.13)
where
\[
\tilde{\mathcal{C}}(j) = |\Omega|^{-1} \int_\Omega c_m^{-1} j \cdot j dx.
\] (3.14)

It is easily seen that the constant current \( \tilde{j} \) is the minimizer for (3.13). Moreover, it is also an admissible trial field for the variational principle (3.11). Substitution of \( \tilde{j} \) into (3.11) gives the estimate:
\[
c_e^{-1} \tilde{j} \cdot \tilde{j} \leq c_m^{-1} \tilde{j} \cdot \tilde{j} + |\Omega|^{-1} L(\tilde{j})(c_m^{-1} - c_r^{-1}) \int_\Gamma (\tilde{j} \cdot n)^2 ds,
\] (3.15)

where
\[
L(\tilde{j}) = R_{cr} - \frac{\int_A |\tilde{j}|^2 dx}{\int_\Gamma (\tilde{j} \cdot n)^2 ds} = R_{cr} - \frac{||\tilde{j}||^2}{M \tilde{j} \cdot \tilde{j}}.
\] (3.16)

Clearly \( c_e^{-1} \tilde{j} \cdot \tilde{j} \leq c_m^{-1} \tilde{j} \cdot \tilde{j} \) when \( L(\tilde{j}) \leq 0 \) and the reinforcement criteria follows.

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