$Note$

Thermal Conductivity of Composites

P. G. Klemens 1

Received September 12,]989

A general expression for the effective thermal conductivity of inhomogeneous media in terms of the Fourier' components of the spatial variation of the conductivity is applied to composites consisting of inclusions in a continuous matrix. It is reformulated in terms of the mean square fluctuations of the conductivity. Specific cases treated are spherical inclusions and long cylinders, both random and with preferred directions. The results hold provided the difference in thermal conductivities is small or provided the concentration of inclusions is not too large. The theory fails if the thermal conductivity of the matrix is much smaller than that of the inclusions. The same considerations also apply to electrical conductivity.

KEY WORDS: composites; concrete; conductivity; fibers; inclusions; oriented fibers; porosity; spheres; thermal conductivity.

1. INTRODUCTION

In a previous paper [1] an expresion was obtained for the thermal conductivity of an inhomogeneous medium, provided the local value of the thermal conductivity $\kappa(r)$ can be expressed in terms of Fourier components

$$
\kappa(\mathbf{r}) = \kappa_0 + \sum_{\mathbf{q}} \kappa(\mathbf{q}) e^{i\mathbf{q} \cdot \mathbf{r}} \tag{1}
$$

where κ_0 is the volume-average of $\kappa(\mathbf{r})$. Defining the effective thermal

¹ Department of Physics and Institute of Materials Science, University of Connecticut, Storrs, Connecticut 06269, U.S.A.

conductivity κ_e in terms of the rate of entropy production and the average temperature gradient, the following expression was found:

$$
\kappa_{\rm e} = \kappa_0 - \frac{1}{\kappa_0} \sum_{\mathbf{q}} |\kappa(\mathbf{q})|^2 \cos^2 \theta \tag{2}
$$

where θ is the angle between the wave-vector q and the direction of the average temperature gradient.

The sum of $|\kappa(q)|^2$ over all wave-vectors can be identified with the mean square fluctuation of $\kappa(\mathbf{q})$, i.e.,

$$
\sum_{\mathbf{q}} |\kappa(\mathbf{q})|^2 = V_0^{-1} \int d\mathbf{r} [\kappa(\mathbf{r}) - \kappa_0]^2
$$
 (3)

where V_0 is the volume of the domain of integration.

Equation (2) places an upper limit on the thermal conductivity, namely, the volume-average κ_0 . In many cases one may expect the second term to be small. Equation (2) is useful only if the Fourier components $\kappa(q)$ can be obtained and the summation over all wave-vectors carried out. However, in isotropic cases, where in the average $\cos^2 \theta = 1/3$, one can use Eq. (3).

The present paper considers simple models of two-phase composites: a continuous matrix containing inclusions which are either spheres or long cylinders. Equation (3) is used to evaluate the second term in Eq. (2), which reduces κ_e below κ_0 . The results are expressed in terms of the fractional volume occupied by the inclusions.

The procedure fails when the fractional volume of inclusions becomes too large, since correlations in their position must then become important. It also fails in the limit when then conductivity of the continuous matrix becomes very small, since there is then no entropy production in the matrix. Finally, Eq. (2) is no longer useful when the second term approaches κ_0 in magnitude, since any error in the evaluation would then be magnified.

2. SPHERICAL INCLUSIONS

Consider a random assembly of spherical inclusions in a matrix of volume V_0 . Let κ_1 be the conductivity of the matrix, κ_2 that of the inclusions, and f the volume fraction of inclusions. Then

$$
\kappa_0 = (1 - f)\kappa_1 + f\kappa_2 \tag{4}
$$

and

$$
V_0^{-1} \int d\mathbf{r} \left[\kappa(\mathbf{r}) - \kappa_0 \right]^2 = f \kappa_2^2 + (1 - f) \kappa_1^2 - \kappa_0^2
$$

= $(1 - f) f (\kappa_1 - \kappa_2)^2$ (5)

Using Eqs. (2) and (3), since $\cos^2 \theta = 1/3$ in the case of isotropy,

$$
\kappa_e = \kappa_0 - f(1 - f)(\kappa_1 - \kappa_2)^2 / 3 \kappa_0
$$

= $\kappa_1(1 - f) + \kappa_2 f - \frac{1}{3}(1 - f)f \frac{(\kappa_1 - \kappa_2)^2}{\kappa_1(1 - f) + \kappa_2 f}$ (6)

This equation is symmetrical in the two components, and the departure from κ_0 is independent of the sign of $\kappa_1 - \kappa_2$. When $\kappa_1 - \kappa_2$ is small compared to κ_0 , it should hold over the entire composition range.

In the special case when $\kappa_2 \ll \kappa_1$, so that the inclusions have negligible conductivity,

$$
\kappa_e = \kappa_1 (1 - 4f/3) = \kappa_0 - f \kappa_1 / 3 \tag{7}
$$

For example, there is a 10% departure from the volume-averaged conductivity if $f = 0.3$. Equation (7) breaks down when f becomes too large, and also there are conceptional difficulties in all cases when $f > 0.5$.

Equation (7) applies in all cases when there are voids in a continuous matrix, ranging from ceramics with some porosity to bubbles blow into concrete to reduce its thermal conductivity.

The case $\kappa_1 \ll \kappa_2$ presents difficulties. A straightforward application of Eq. (6) would yield

$$
\kappa_e = \kappa_2 (4f - 1)/3 \tag{8}
$$

so that κ_e would appear to be negative for $f < 0.25$. This result is clearly unphysical; it is a consequence of the assumptions of the theory breaking down. If the matrix has zero conductivity, there is no entropy production in the matrix. Entropy production can then occur only if the inclusions are numerous enough to touch frequently and to form a continuous percolation path [2]. The present method is therefore unsuited to all cases when the conductivity of the majority phase is very small. In particular, this includes foams, for which the minority phase has a relatively high conductivity but is constrained to be continuous.

3. LONG FIBERS

Consider a long fiber of cross-sectional area A in a matrix of crosssectional area A_0 , so that $f = A/A_0$. Again, let κ_1 be the conductivity of the matrix and κ , that of the fiber material. As before, the sum of the squares of the Fourier components is given by Eq. (5). Now, however, the wavevectors of the Fourier components all lie in a plane perpendicular to the fiber axis. If the fiber has a circular cross section, they are distributed isotropically in that plane.

Let the fiber axis make an angle ϕ with the direction of the temperature gradient. A wave-vector q, lying in the normal plane, has a component q_1 in the plane formed by the fiber axis and the temperature gradient and a component q_2 normal to that plane. Thus

$$
\cos \theta = \sin \phi \qquad \text{for} \quad q_1
$$

\n
$$
\cos \theta = 0 \qquad \text{for} \quad q_2
$$
 (9)

Averaging over all directions of q in the plane normal to the fiber axis, so that in the average $q_1^2 = \frac{1}{2}q^2$,

$$
\langle \cos^2 \theta \rangle = \sin^2 \phi / 2 \tag{10}
$$

and from Eqs. (3) and (5) ,

$$
\sum_{\mathbf{q}} |\kappa(\mathbf{q})|^2 \cos^2 \theta = \frac{1}{2} \sin^2 \phi f (1 - f) (\kappa_1 - \kappa_2)^2
$$
 (11)

For an assembly of long parallel fibers, with the temperature gradient making an angle ϕ with the fiber axes, κ_e becomes

$$
\kappa_e = \kappa_0 - \frac{1}{2\kappa_0} \sin^2 \phi f (1 - f)(\kappa_1 - \kappa_2)^2
$$
 (12)

In the special case when all fibers are parallel to the temperature gradient, $\sin \phi = 0$, and $\kappa_e = \kappa_0$, as expected.

If the fibers are oriented completely at random, so that in the average $\sin^2 \phi = 2/3$, one reverts to Eq. (6). This equation applies to inclusions of all shapes, provided they are randomly oriented, so that the overall medium is isotropic, that is without preferred directions. Thus, for randomly oriented fibers

$$
\kappa_e = \kappa_0 - f(1 - f)(\kappa_1 - \kappa_2)^2 / 3\kappa_0 \tag{13}
$$

Another case of interest is when all fibers lie normal to a given direction and are randomly oriented in the normal plane. Let the temperature

Thermal Conductivity of Composites 975

gradient make an angle α with that plane. A particular fiber has its axis at an angle β with the projection of grad T in that plane. The angle ϕ between the fiber axis and grad T is then given by

$$
\cos^2 \phi = 1 - \sin^2 \phi = \cos^2 \alpha \cos^2 \beta \tag{14}
$$

and averaging over all orientations of β , so that $\cos^2\beta = \frac{1}{2}$,

$$
\left\langle \sin^2 \phi \right\rangle = 1 - \frac{1}{2} \cos^2 \alpha \tag{15}
$$

Substituting this into Eq. (12),

$$
\kappa_e = \kappa_0 - \left[(1 + \sin^2 \alpha) / 4 \kappa_0 \right] f (1 - f) (\kappa_1 - \kappa_2)^2 \tag{16}
$$

The thermal conductivity is thus anisotropic, is a minimum in the direction normal to the plane of the fibers, when $\sin \alpha = 1$, and a maximum in directions lying in that plane. The conductivity is below the volume-averaged conductivity κ_0 in all cases. It is interesting that κ_e is a minimum in the direction normal to the fiber plane irrespective of whether the conductivity of the fibers is larger or smaller than that of the matrix.

Again, these expressions hold provided

$$
f(1-f)(\kappa_1 - \kappa_2)^2 \ll \kappa_0^2 \tag{17}
$$

In the case of fibers of very low conductivity in a conducting matrix, where $\kappa_0 = (1 - f)\kappa_1$ and $\kappa_2 = 0$, Eq. (13) becomes

$$
\kappa_{\rm e} = \kappa_1 (1 - 4f/3) \tag{18}
$$

This would apply, for example, to the case of glass fibers in a metal matrix provided the fractional fiber volume is not too large.

In the opposite case of conducting fibers in an insulating matrix, the theory breaks down, for the same reason as discussed in Section 2, and the problem becomes a percolation problem.

4. SUMMARY

The general equation (2) for the effective thermal conductivity has been applied to the cases of spherical inclusions and fiber inclusions. Estimates are given for the departure from the volume-averaged conductivity. For those cases where this departure is only a small fraction of the conductivity, the results appear to be reliable. The theory fails for the case of a nonconducting matrix.

ACKNOWLEDGMENTS

The author wishes to thank City University, London, for their hospitality during his sabbatical leave, where this work was done. Interest in the problem was stimulated by discussions with Dr. D.C. Spooner, of the British Cement Association. Dr. J. M. Javanainen, of the University of Connecticut, suggested the connection expressed by Eq. (3).

REFERENCES

1. P. G. Klemens, *Int. J. Thermophys.* 10:1213 (1989).

2. S. Kirkpatrick, *Rev. Mod. Phys.* 45:574 (1973).