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Physics Letters A 313 (2003) 218–222
PHYSICS LETTERS A
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Critical resistance for multi-phase composite materials

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Received 24 January 2002; received in revised form 22 April 2003; accepted 23 April 2003

Communicated by R. Wu

Abstract

The effect of interfacial resistance on the effective conductivity of a multi-phase composite material was studied. The composite under study is composed of a matrix surrounding different types of circular cylinders arranged in rectangular order. It was assumed that the interfacial resistance is concentrated on the surface of the cylinders. For any direction of calculating the effective conductivity of the system, a condition was found in which the effect of cylinders of one type can be neglected. This condition may be estimated by $R \leqslant k - 1$, where R and k are the non-dimensional interfacial resistance and the relative conductivity of the neglected cylinders, respectively. The case $R = k - 1$ applies when the same relation exists between the interfacial resistance and conductivity of all types of cylinders.

PACS: 44.10.+i; 72.80.Tm; 41.20.Cv

Keywords: Interfacial resistance; Conductivity; Multi-phase composites

Multi-phase systems, which consist of inclusions of different shapes and properties embedded in a matrix, can be found in a wide range of practical processes and are of considerable technological importance. Starting with the work of Maxwell [1] and Rayleigh [2], who considered the problem of calculating the effective conductivity of two-phase systems composed of spheres and cylinders, McPhedran later extended the discussion to three-phase composites having the CsCl structures [3]. More recently, Whites et al. [4,5], in the context of the dielectric constant, have developed a formulation for the efficient numerical calculation of the effective property of multi-phase composites. Since the problem of calculating dielectric constant is mathematically identical to that of calculating thermal or electrical conductivity [6,7], similar results can be expected.

Most studies have assumed that the interface is ideal, but the interfacial resistance may occur due to a variety of phenomena [8], such as the presence of a thin gap with a third material between the inclusions and the matrix [9] and disparity in the physical properties [10] (Kapitza resistance). Taking this effect into account is very important, since the effective conductivity may change significantly, and a system with conducting inclusions may behave like one that has non-conducting inclusions.

Chiew [8] showed that for composite materials with a random array of uniform spherical inclusions, there may exist a critical situation in which the system does

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0375-9601/03/$ – see front matter
doi:10.1016/S0375-9601(03)00749-7

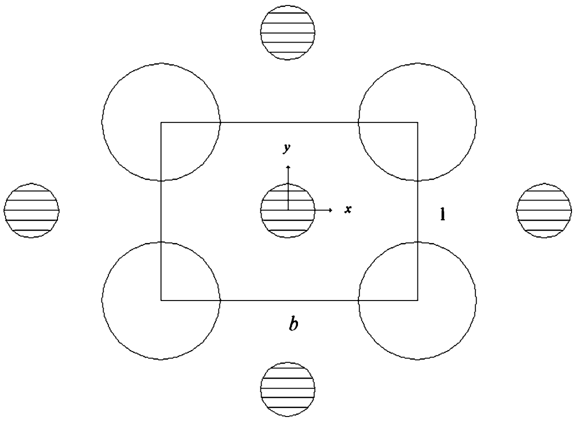

Fig. 1. The structure of the three-phase composite under study.

not sense the presence of the inclusions. Studies into the behavior of composites that consist of periodic arrays of uniform spheres [11,12] and random and periodic arrays of uniform cylinders [13,14] revealed that for all these cases a critical situation arises if the non-dimensional interfacial resistance between the inclusions and the matrix is equal to the relative conductivity of the inclusions minus one.

Here, we extend the discussion to multi-phase composite materials. In order to simplify the presentation, let us consider a three-phase system that consists of a matrix and two types of circular cylindrical inclusions that are arranged in rectangular order with periodicities equal to a unity in the y-direction and b in the x-direction, as depicted in Fig. 1. Let us assume that a uniform field of magnitude E_{ext} has been applied along the x-axis in the negative direction. At the surface of any cylinder of type i $(i = 1, 2)$, we may consider a dimensionless interfacial resistance [15], R_i, and express the boundary conditions as follows:

$$\frac{k_i}{R_i a_i}(T_i - T_m) = -k_i \frac{\partial T_i}{\partial r} = -\frac{\partial T_m}{\partial r}, \quad r = a_i, \tag{1}$$

where a and k represent the radius and the relative conductivity of cylinders, respectively. T shows the temperature function and m refers to the matrix. Using the Rayleigh method for the purpose of solving the Laplace equation through the system provides a system of algebraic equations in which the nth equation $(n = 1, \ldots, \infty)$ of the set reads

$$\frac{B^i_{2n-1}}{\gamma^i_{2n-1} a_i^{4n-2}} + \sum_{m=1}^{\infty} \binom{2n+2m-3}{2n-1} \times \left(S^1_{2n+2m-2} B^i_{2m-1} + S^2_{2n+2m-2} B^{2-\delta_{i2}}_{2m-1}\right) = E_{\text{ext}} \delta_{n1}, \tag{2}$$

where B^i_{2n-1} are unknowns, S^i_{2n} are the lattice sums [1] over cylinders of type i, δ_{ij} represents the Kronecker delta (1 for $i = j$, otherwise 0) and γ^i_{2n-1}, which can be referred to as multipolar polarizabilities, are of the form

$$\gamma^i_{2n-1} = \frac{1 - k_i + R_i(2n-1)}{1 + k_i + R_i(2n-1)}. \tag{3}$$

By applying the Fourier law, the effective conductivity of the system can be derived as follows:

$$k_e = 1 - \frac{2\pi(B^1_1 + B^2_1)}{b E_{\text{ext}}} \tag{4}$$

or more generally, for the case of N types of cylinders in the unit cell as $k_e = 1 - 2\pi \sum_{i=1}^{N} B^i_1/(bE_{\text{ext}})$. The values of B^1_1 and B^2_1 can be obtained numerically by solving the algebraic system of equations given in (2), however, coarsely truncating (2), we may explicitly derive these values. If we perform a triangular truncation of the second order of (2) and use the resultants B^1_1 and B^2_1 in (4), we can obtain an analytical relation for the effective conductivity, which can be applied to low-volume fractions, i.e.,

$$k_e = 1 - \sum_{i=1}^{2} \frac{2 f_i}{(\lambda_i \lambda_{2-\delta_{i2}} - \xi_i \xi_{2-\delta_{i2}})/(\lambda_{2-\delta_{i2}} - \xi_{2-\delta_{i2}})} \tag{5}$$

with

$$\lambda_i = \frac{1}{\gamma^i_1} + c_1 f_i - c_2 \gamma^i_3 f_i^4 - c_3 \gamma_3^{2-\delta_{i2}} f_i f^3_{2-\delta_{i2}}, \tag{6}$$

$$\xi_i = c_4 f_i - c_5\left(\gamma^i_3 f_i^4 + \gamma_3^{2-\delta_{i2}} f_i f^3_{2-\delta_{i2}}\right), \tag{7}$$

where f_i denotes the volume fraction of cylinders of type i. The constants for the case $b = \sqrt{3}$ for deriving the effective conductivity in the x-(parallel) and y-(perpendicular) directions are listed in Table 1, calculating highly accurate values for the lattice sums using integral representation technique [16].

Fig. 2 shows the results for the effective conductivity in the presence of the interfacial resistance. The

Table 1
The calculated values for $c_1, \ldots, c_5$ used in the analytical formula (5) for determining the effective conductivity in the parallel and perpendicular directions for the case $b = \sqrt{3}$

	Parallel	Perpendicular
c_1	0.187018134	1.812981866
c_2	1.310523128	1.310523128
c_3	1.310523128	1.310523128
c_4	1.812981866	0.187018134
c_5	−1.310523128	−1.310523128

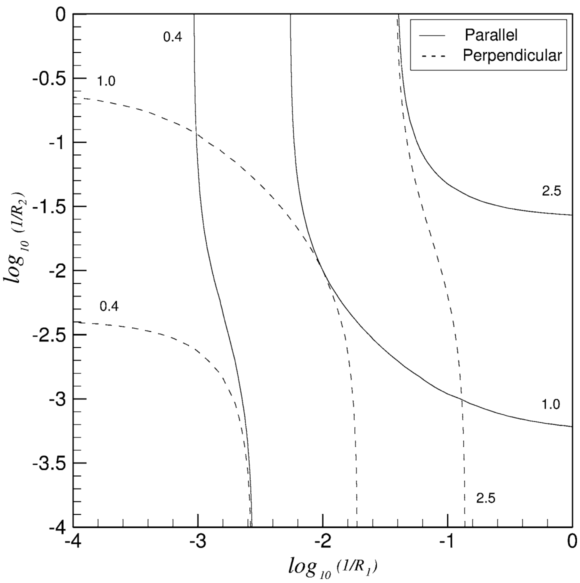

Fig. 2. The contours of the effective conductivity with respect to the interfacial resistance of the cylinders. $f_1 = 0.4$, $f_2 = 0.2$, $k_1 = 101$, $k_2 = 101$ and $b = \sqrt{3}$. As can be seen for the cases in which the interfacial resistances are very large, the effective conductivity of the system is less than the conductivity of the phases (see $k_e = 0.4$). This is because under these conditions the cylinders act as cylinders the conductivity of which is less than that of the matrix.

volume fractions are $f_1 = 0.4$ and $f_2 = 0.2$, the conductivities of the cylinders were assumed to be $k_1 = 101$, $k_2 = 101$, and the periodicity in the parallel direction is $b = \sqrt{3}$. The effective conductivity was derived numerically by solving Eq. (2) and then applying Eq. (4). 100 unknowns of B^i_{2n-1} $(i = 1, 2)$ were considered in the process of the solution. As can be seen, the effective conductivity of the system can be highly affected, and based on the values of R_1 and R_2, the system may yield a conductivity outside the limit of the conductivity of the phases. Providing that $B^1_1 + B^2_1 = 0$, the system is subject to situations in which the effect of the inclusions can be neglected ($k_e = 1$ in Fig. 2). Of these states, the case $B^1_1 = B^2_1 = 0$ is of particular interest. This case occurs when $R_i = k_i - 1$ $(i = 1, 2)$, and as a result based on Eq. (3) the dipole polarizabilities (γ^1_1, γ^2_1) are zero. Obtaining a value of zero for the dipole polarizabilities means that the state of the system resembles that of a system of perfect interfaces which is made up of cylinders the conductivity of which is equal to a unity. Therefore, we may expect that the effective conductivity would be independent of the volume fractions and the direction of the calculation of the effective conductivity.

While the effect of both types of cylinders can be exactly neglected when $R_i = k_i - 1$ $(i = 1, 2)$, in general the effect of the inclusions of type i $(i = 1$ or 2) cannot be neglected when only $R_i = k_i - 1$. In other words, systems with $k_i = 1$, $R_i = 0$ (two-phase system) and k_i, $R_i = k_i - 1$ (three-phase system) are not equivalent when $R_{2-\delta_{i2}} \neq k_{2-\delta_{i2}} - 1$. Although for both cases, B^i_1 is zero and the effective conductivity can be calculated simply by using $k_e = 1 - 2\pi B_1^{2-\delta_{i2}}/(bE_{\text{ext}})$ but for the imperfect interface case, the terms B^i_{2n-1} $(n > 1)$ are not zero and are present in the procedure of the calculation of $B_1^{2-\delta_{i2}}$, as is evident in Eq. (2). For the perfect interface case, all the terms of B^i_{2n-1} $(n > 1)$ are zero and do not affect the value of $B_1^{2-\delta_{i2}}$. This means that the field distributions inside the matrix for the two- and three-phase systems may be dissimilar. The degree of discrepancy can only be numerically determined and depends on the geometrical considerations, resistance and conductivity of the inclusions. Fig. 3 shows a comparison between the effective conductivity of the two systems for a series of given data. As can been seen, the results for the three-phase system underestimate the conductivity of the two-phase system. The reason can be understood when considering that for the three-phase case $\gamma^i_{2n-1} > 0$ $(n > 1)$, while for the two-phase one they are zero. Increasing f_i increases the error, since the higher-order terms play an important role in the response of the system. In the dilute limit, the systems can be used equivalently. This is also evident from Eq. (5), as ignoring the higher orders for the case $\gamma^i_1 = 0$ gives $k_e = 1 - 2f_{2-\delta_{i2}}/(1 + c_1\gamma_1^{2-\delta_{i2}})$, which is the conductivity of the system, which neglects the effect of the type-i inclusions.

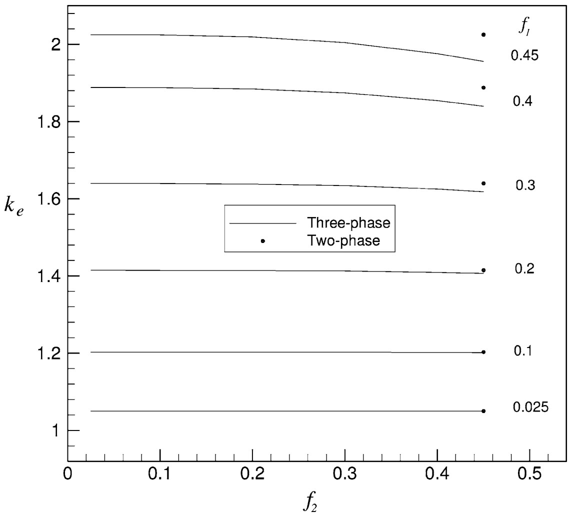

Fig. 3. The effective conductivity of the two- and three-phase systems. For the three-phase system, $k_1 = 101$, $k_2 = 101$, $R_1 = 10$, $R_2 = k_2 - 1 = 100$ and $b = \sqrt{3}$. The two-phase system consists of the matrix and type-one cylinders with the same properties as those given in the three-phase case.

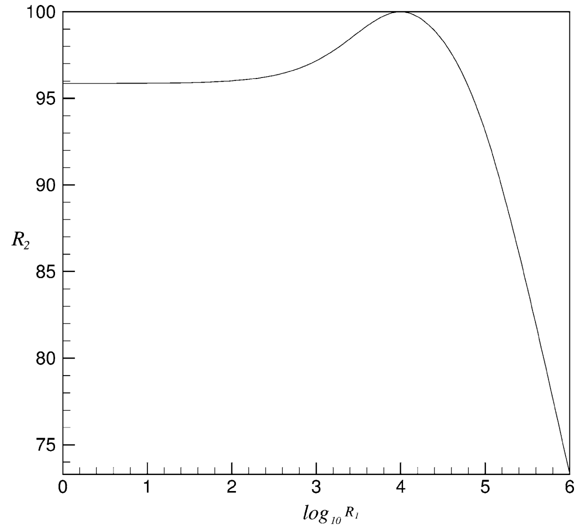

Fig. 4. The calculated value for R_2, in which the effect of the type-two cylinders can be neglected. $f_1 = 0.45$, $f_2 = 0.45$, $k_1 = 101$, $k_2 = 101$ and $b = \sqrt{3}$.

By reducing R_i from the value $k_i - 1$, we may obtain a situation in which the three-phase system exactly gives the effective conductivity of the two-phase one. The expectation of finding such a situation stems from the fact that cylinders the conductivity of which is greater than that of the matrix, boost the conductivity of the system. Fig. 4 reports such an interfacial resistance for type-two cylinders as a function of the interfacial resistance of type-one cylinders for another series of given data. As can be seen, only when $R_1 = k_1 - 1$, we get $R_2 = k_2 - 1$.

For the case of N types of cylinders in the unit cell, by extending Eqs. (2) and (4), it can be shown that when $R_i = k_i - 1$ $(i = 1, \ldots, N)$, the effect of all types of cylinders can be neglected and the system simply behaves like a homogeneous system with the conductivity of the matrix. In other cases, the resistance, in which the effect of type-i cylinders in the direction of the calculation the effective conductivity can be neglected, may be estimated by $R_i < k_i - 1$.

In summary, the effective conductivity of a multi-phase composite material that is made up of a periodic structure in the presence of the interfacial resistance was studied. The situations in which the effect of one or all types of cylinders can be neglected due to the interfacial resistance were explained. The structure considered in this study was composed of circular cylinders in a periodic arrangement, but when the results given in Refs. [11–13] are considered, a similar outcome can be expected for the case of random arrangements as well as for that of spherical inclusions (random and periodic).

Acknowledgements

The authors wish to thank Professor J. Huang for helpful discussions on the calculation of the lattice sums and for communicating his results prior to publication.

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