Lappeenrannan teknillinen yliopisto Lappeenranta University of Technology

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TRANSPORT PROPERTIES OF MULTI-PHASE COMPOSITE MATERIALS

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ABSTRACT

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This research concerns the calculation of the effective transport properties of multi-phase composite materials. The composite materials under investigation are composed of a periodic array of inclusions embedded in a matrix. The geometry of the inclusions taken into account are circular cylindrical, elliptical cylindrical and spherical. The inclusions can be solid or multiply coated. The method used for these studies is Rayleigh's method. Highly accurate values for the lattice sums have been obtained using recently developed algorithms. A series of explicit formulations used to facilitate the calculation of the effective transport property of the systems under study are reported here.

The behavior of the systems as a function of the transport properties of the phases is inspected. For multi-phase composites with cylindrical inclusions highly an-isotropic behavior is observed.

The procedure for finding the equivalent systems of multi-coated systems with circular cylindrical inclusions, when the sum of the transport property of the layers is equal to zero, is studied. It is found that to construct the equivalent system in multi-coated systems, many layers should be modified. Furthermore, multi-coated systems with elliptical cylindrical and spherical inclusions do not have similar equivalent systems. The results are verified using a numerical simulation method.

The effect of interfacial resistance on the effective conductivity of multi-phase systems with circular cylindrical and spherical inclusions is studied, assuming that the interfacial resistance is concentrated on the surface of the inclusions. It is found that there is a specific condition in which the effect of one phase on multi-phase systems in the direction of calculation of the effective conductivity can be neglected. This condition may be estimated by $R \le k-1$, 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.

Two resistor models are used for deriving the upper and lower bounds of the effective transport property. It is found that the physical behavior of these bounds can be different from the natural behavior of the systems.

Keywords: transport property, multi-phase, composite materials. UDC 536.2 : 620.18 : 517.927

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Lappeenranta, May 2003

Ali Moosavi

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Appendinx I

Moosavi, A., Sarkomaa, P. (2002a), 'The resonant behavior of multi-phase composite materials', *Int. J. Mod. Phys. C.*, Vol. 13, pp. 405-417.

Appendix II

Moosavi, A., Sarkomaa, P., Polashenski, W. (2002b), 'The effective conductivity of a composite materials with cubic arrays of multi-coated spheres', *Appl. Phys. A*, In press (Published online: 4 December 2002, DOI 10.1007/s00339-002-1456-y, pp.1-8).

Appendix III

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Appendix IV

Moosavi, A., Sarkomaa, P. (2003b), 'Critical resistance for multi-phase composite materials', *Phys. Lett. A*, In press.

Appendix V

Moosavi, A., Sarkomaa, P. (2003c), 'An investigation into the behavior of multi-phase composite materials: Coated systems', *J. Phys. A: Gen. Math.*, Submitted.

NOMENCLATURE

Arabic letters

radius of the cylindrical or spherical inclusions
distance between the inclusions in the <i>x</i> -direction
distance between the inclusions in the y-direction for 3-D structures
constants
electric flux density
volume fraction
magnitude of external field
total volume fraction
heat flux vector
thermal conductivity
electrostatic potential
unit vector in the direction <i>x</i>
unit vector in the direction y
Bessel function of kind <i>i</i> and order <i>n</i>
number of the inclusions in the unit cell
number of the layers in coated systems
normal outward unit vector
order
a parameter encountered in calculating the lattice sums
interfacial resistance
radial distance
lattice sum
extra heat flux resulting from the inclusions
extra heat flux resulting from the interface
temperature
volume
cartesian coordinates
spherical harmonics of order (l, m)

Greek letters

\boldsymbol{d}_{n1}	Kronicker delta (1 if $n = 1$ otherwise zero)
e	dielectric constant
f	azimuthal angle
\boldsymbol{g}_n	multipolar polarizability
k	wave number
m	elliptic cylindrical coordinate
q	polar angle
S	surface area of the inclusions or boundary
у	potential function

Superscripts

d	dynamic
+	upper bound
-	lower bound
*	complex conjugation
,	perpendicular direction

Subscripts

е	effective
ext	external
Ι	interface
Р	particle

Acronyms

SC	simple cubic
BCC	body-centred cubic
FCC	face-centred cubic

1. INTRODUCTION

1.1 Composite materials: definition, classification and importance

There is no unique definition for composite materials [Schwartz, 1984], and different definitions are available; however, for our purposes, the term composite material refers to a system that consists of two or more distinct phases that are separated from each other by an interface. The phases are insoluble in each other. Composites usually have three parts; namely, a matrix (background), structural constituents and an interface. The matrix is a homogeneous material which surrounds the other phases and the interface is the boundary or surface between the phases. One way of classifying composites is according to the specification of the structural constituents. The structural constituent can take many forms such as particles, fibers, laminates, fillers, flakes or combinations of the above.

The above definition brings to mind many systems that are of clear and essential technological importance. Examples are wood, foam, oil-filled porous rocks, colloidal suspensions, emulsions, solid rocket propellant, concrete, reinforced materials, to name just a few. Therefore the study of the behaviour and property of composites has a lot of potential and for years has attracted researchers in different fields including physics, biology, engineering, mathematics, materials science, geophysics and hydrology.

1.2 The subject of this study

In this research, the purpose is to determine the effective property of multiphase composite materials that are made up of periodic structures [Moosavi and Sarkomaa, 2002a; Moosavi et al., 2002b; Moosavi and Sarkomaa, 2003a; Moosavi and Sarkomaa, 2003c]. For the sake of greater familiarity, the formulations will be given mainly in the context of thermal conductivity, but the studied properties can be considered to be all those listed by Batchelor [1974] that are mathematically equivalent (recently Torquato [2002] has provided a similar table with more details). These properties include thermal and electrical conductivity, dielectric permittivity, magnetic permeability, mobility, the

effective mass in bubbly flow. The inclusions studied in this thesis are in the shape of circular cylinders or spheres; but also composites with periodic arrays of elliptical cylinders are examined. The basic formulations and results can be used for any purpose, although here the focus is applications in which the effect of one or more of the phases is neglected. For coated composites, the interesting behaviour reported for singly coated circular cylinders is considered. A further generalisation of this phenomenon is provided and extended to multicoated systems. For multiphase non-coated composites, cases in which the effects of phases with perfect and imperfect interfaces can be neglected, will be explained.

1.3 Motivation for selecting the cases for the study

1.3.1 Periodic composites with ideal inclusions

Only a few composites can be considered periodic and as having ideal inclusions. However, it should be noted that accurate calculation of the effective properties of real structures can be very difficult if not prohibitive. Ideal structures are under consideration here, since the mathematics involved in these systems is manageable and the effective property may be calculated to a high degree of accuracy. The results can be useful for understanding the interplay between microstructures and the effective property of real structures, and specifically those which can be approximated with the use of periodic structures. Also, the results provide a way of testing well-known classical numerical methods such as the boundary element method, finite difference method or finite element method [Baltz et al., 1997].

1.3.2 Coating composites

In many cases it is necessary to coat the inclusions; one such case is for the purpose of increasing the wettability and adhesion of the inclusions and the matrix. When the enhancing effect of the inclusions is not sufficient, a coating layer may modify the behaviour of the system. For protection from chemical reactions, a suitable coating layer can also be applied. In some cases, the coating layer is unwanted but occurs because of many problems in the production processes. For instance, due to a mismatch in thermal expansion, a coating layer with a third material may appear between the inclusions and the matrix. In some cases,

modelling a system as a coated one may provide reasonable results, even though the system is not exactly coated. This is the case for wet wood for which the moisture is considered to be a coating layer. Therefore, extending the formulations to coated systems may have many implications.

1.3.3 Composites with interfacial resistance

Most of the research has been performed assuming an ideal interface, but interfacial resistance may occur because of a variety of phenomena, such as physical irregularities at the boundaries, the presence of impurities, reaction on the surface of inclusions, acoustic mismatch at cryogenic temperature, or the presence of a gap between the inclusions and the matrix [Shai, 1982; Sanokawa, 1968; Eardley, 1973; Bhatt, 1992; Little, 1959; Torquato, 1995]. It is very important to take this effect into account, as the effective conductivity of the system may be significantly changed and a system with conducting inclusions may behave like a system that has non-conducting inclusions. In a more general case, the effect of interfacial resistance can be modelled assuming that there is a coating layer around the inclusions. However, in some cases, interfacial resistance can be characterised by introducing a non-dimensional parameter assuming that the interface has no thickness and that interfacial resistance [Little, 1959; Torquato, 1995] or when the Knudsen number in the gap is considerable.

1.3.4 Multiphase composites

The behavior demonstrated by multi-phase systems can be completely different from that understood on the basis of two-phase systems. Also, extending the discussion to multi-phase cases introduces further generalisations into the formulations and relations.

1.4 Earlier studies

Contrary to general belief, the knowledge required for producing composite materials is not new. For example, archaeological studies show that the inhabitants of Finland were able to produce fibre-reinforced ceramics 4000 years ago [Lukkassen, 2002]. The scientific investigation for deriving the effective property of composites is also old and begins from the work done by Maxwell [1873], Rayleigh [1892] and Einstein[1906].

In his treatise, Maxwell [1873] outlined the procedure for calculating the effective conductivity of composite materials that have a very small volume fraction of randomly arranged sphere-shaped inclusions, and derived a first-order relation of the volume fraction for the effective conductivity of these materials. A considerable improvement was made to Maxwell's relation by Jeffrey [1973] who considered the interactions between the spheres using a methodology put forward by Batchelor [1972] and was successfully able to obtain a second-order relation. Chiew and Glandt [1983] later presented a modified version of Jeffrey's formulation. Other research calculating the transport properties of composites with random arrangements can be found in reports by Sangani and Yao [1988], Bush and Soukoulis [1995], Kirchner et al. [1998] and others.

The research into periodic structures was initiated by Lord Rayleigh [1892]. Rayleigh took into account composites made up of periodic arrays of circular cylinder- and sphere-shaped inclusions. By describing the polarisation of each sphere in an external field using an infinite set of multipole moments, Rayleigh derived a relation for calculating the effective conductivity (electrical or thermal) of these materials of $O(f^{10/3})$, where *f* is the volume fraction. Rayleigh's methodology turned out to be problematic. Rayleigh decided to use an arbitrary method for calculating one parameter (lattice sum S_2) included in his methodology, without explaining the reason; therefore, his method has been questioned by several researchers [Jeffrey, 1973]. Mckenzie and McPhedran [1978] modified the Rayleigh method and explained the physics and mathematics in the problematic section of Rayleigh's procedure (see also Poulton et al. [1999]). Rayleigh's methodology has since been validated by many researchers. It is powerful and can be extended to electromagnetic [Nicorovici et al., 1994], elastostatic [McPhedran et al., 1994] and elastodynamic [Movchan et al., 1997] problems.

Runge [1925] studied the effective property of composites with periodic arrays of coated circular cylinders, but the core of the system considered by Runge had a property equal to that of a matrix. The discussion was generalised by Israelachvili et al. [1976] and Ninham and

Sammut [1976]. Mentuaful and Todreas [1994] derived a more developed explicit relation for the effective property of these composites. Nicrovici et al. [1993a] studied the case in more detail in the context of dielectric permittivity and reported strange behaviour for these composites. The same authors later extended the results to composites made up of randomly arranged coated circular cylinders and as well as to composites made up of periodic and random arrays of coated-spherical inclusions [Nicorovici et al., 1993b; Nicorovici et al., 1995]. Lu and Lin [1995; 1997] used the coating procedure for the purpose of modelling interfacial resistance in composite materials. Lu et al. [1996; 1997] have also presented the necessary changes needed to extend the Maxwell methodology to coated systems. For noncoated multi-phase composite materials, the work of McPhedran [1984] can be indicated who considered calculating the effective transport properties of three-phase composite materials having two different types of spherical inclusions arranged in the CsCl lattice (i.e., two intermeshed simple cubic lattices). More recently, Whites [2000] and Wu and Whites [2001] have developed a formulation for calculating efficiently numerically the effective property of multi-phase materials made up of periodic structures composed of spheres or cylinders, within the context of permittivity.

In addition to composites made up of inclusions in the shape of spheres or circular cylinders, composites made up of inclusions of other shapes have also been investigated, for example composites made up of inclusions in the shape of elliptical cylinders [Obdam et al., 1987; Nicorovici et al., 1996] or spheroids [Lu, 1998]. Schulgasser [1992] and Fel et al [2000] have provided some examples of structures whose the effective property can be easily found by using the Keller reciprocal relation [Keller, 1964]. A simple example could be a square checkerboard, which has been widely studied [Craster et al., 2001]. The case of a rectangular checkerboard, however, requires a considerable effort [Obnosov, 1999]. The effective conductivity of materials with square cylinders has been studied by Andrianov et al. [1999], who have applied the Padé approximation. More generally, Obnosov et al. [1999] have considered rectangular and triangular inclusions using complex variables.

Instead of considering a composite material and then calculating the effective property, it is possible to design composites with a specific property. Recently, Torquato et al. [2001] and Hyun et al. [2001, 2002] have adopted optimisation techniques for this purpose (the so-called

Target-Optimisation method). In this method, the unit cell is divided into many cells, each of which contains the material of the matrix or the other phases. Using numerical procedures, the cells are gradually moved until they yield a final structure that has the effective target property.

2. COMPOSITE MATERIALS WITH PERIODIC ARRAYS OF CIRCULAR CYLINDERS

In this chapter the problem of calculating the effective transport property of composite materials with circular cylindrical inclusions is discussed. First, the simplest case will be considered, i.e. one in which the cylinders are uniform and solid and the interface has no resistance, and the solution given by Rayleigh [1892] is explained. After that multiphase cases will be considered. Multiphase cases can occur as a result of the coating of the inclusions [Moosavi and Sarkomaa, 2002a] or the presence of different types of inclusions in the system [Moosavi and Sarkomaa, 2003a]. Both these cases will be taken into account separately and the behaviour of these systems explained. The effect of interfacial resistance will also be discussed [Moosavi and Sarkomaa, 2003b].

2.1 Two-phase composites with arrays of uniform and solid circular cylinders

Let us consider composite materials that are made up of uniform and solid inclusions of a circular cylindrical shape and arranged in a rectangular array with periodicities equal to a unity in the y-direction and b in the x-direction and immersed in a matrix with unit conductivity, as depicted in Fig. 2.1(a). The radius and the reduced conductivity of the inclusions will be titled by a and k, respectively. This way, the unit cell of the system would be a rectangular cylinder with sides equal to b and c, in the x- and y-directions, respectively, having a circular cylindrical inclusion with conductivity k and volume fraction $f = \mathbf{p} a^2/b$ in the middle, as shown in Fig. 2.1(b).

The problem under study is deriving the effective conductivity of the system in the x- and ydirections. The effective conductivity can be simply obtained along the axis of the cylinders by using the arithmetic average technique, i.e., 1 - f + kf.



Figure 2.1 A composite material composed of a matrix and dispersed phases shaped like circular cylinders arranged in a periodic arrangement (a), and the unit cell of the system (b).

Suppose that a uniform field of magnitude E_{ext} is applied externally along the *x*-axis of the system in the negative direction. Thereby, the temperature inside the cylinder and the matrix in the polar coordinates (r,q) where q is measured from the *x*-axis can be expressed as:

$$T_1 = C_0 + \sum_{l=1}^{\infty} r^l \left(C_l \cos l \boldsymbol{q} + C_l' \sin l \boldsymbol{q} \right) \qquad r \le a \qquad (2.1)$$

$$T_{2} = A_{0} + \sum_{l=1}^{\infty} \left[\left(A_{l} r^{l} + B_{l} r^{-l} \right) \cos l \boldsymbol{q} + \left(A_{l}' r^{l} + B_{l}' r^{-l} \right) \sin l \boldsymbol{q} \right] \qquad r > a$$
(2.2)

The periodicity of the system implies that C_0 and A_0 differ from one cell to another, but the other coefficients are essentially the same for all cells. Because of the symmetry of the temperature around q = 0, C'_l , A'_l and B'_l will be neglected. Also, the temperature is anti-symmetric around q = p/2, and therefore, *l* can only be an odd number. As a result, the temperature functions can be reduced to the following:

$$T_{1} = C_{0} + \sum_{n=1}^{\infty} C_{2n-1} r^{2n-1} \cos(2n-1) \boldsymbol{q} \qquad r \le a \qquad (2.3)$$

$$T_{2} = A_{0} + \sum_{n=1}^{\infty} \left[A_{2n-1} r^{2n-1} + B_{2n-1} r^{-2n+1} \right] \cos(2n-1) \boldsymbol{q} \qquad r > a \qquad (2.4)$$

Next, the unknown coefficients will be derived. In the interface, the temperature and heat flux are continuous. Thus, we have

$$T_1 = T_2, \qquad k \,\partial T_1 / \partial r = \partial T_2 / \partial r \qquad r = a$$
(2.5)

Applying the above-mentioned boundary conditions, the coefficients of the temperature functions can be related to each other, i.e.,

$$A_{2n-1} = \frac{B_{2n-1}}{g_{2n-1}a^{4n-2}}$$
(2.6)

$$C_{2n-1} = \frac{B_{2n-1}}{c_{2n-1}a^{4n-2}},$$
(2.7)

where

$$g_{2n-1} = \frac{1-k}{1+k}$$
 $c_{2n-1} = \frac{1-k}{2}$ (2.8)

As can be seen, g_{2n-1} , which we can be referred to as multipolar polarisability, and c_{2n-1} are not *n*-dependent, but since they are, in general (for the case of coated cylinders or in the presence of resistance), functions of *n*, they will be kept in this form.

The temperature functions cannot still be determined since the relations given in (2.6) and (2.7) are relations between the coefficients only and do not provide suitable equations in terms of the unknowns; a further series of relations between the coefficients is required. Many methods known nowadays, such as the Rayleigh method [1892], Zuzovski-Brenner method [1977], collocation scheme [Lu and Lin, 1994], inclusion model [Lu, 1998], and others have been considered for this purpose.

Here, the Rayleigh method is used, as it is the oldest and has received a lot of attention in the literature [Meredith and Tobias, 1960; Mcphedran and Mckenzie, 1978; Perrins et al., 1979; Cheng and Torquato, 1997]. It is based on the fact that the temperature around the origin, when origin not included, can be considered due to terms originating at infinity and at the other lattice sites. The reason is as follows: the temperature function given in Eq. (2.4)

consists of two types of terms. The terms that contain r^{-2n+1} are clearly due to the sources situated at the origin, since by increasing the distance from the origin these terms diminish. The terms that contain r^{2n-1} cannot be due to the sources at the origin because they increase when r increases; therefore they originate from other existing sources in the system. The external field for any point placed in (x, y) leads to a temperature increase equal to $E_{ext}x$. For this point, sources situated on the axis of the *j*th cylinder will also lead to a temperature

increase equal to
$$\sum_{n=1}^{\infty} B_{2n-1} \cos(2n-1) \boldsymbol{q}_j / r_j^{2n-1}$$
 where (r_j, \boldsymbol{q}_j) is polar coordinate of the point

when measured from the *j*th cylinder center. With this explanation the following relation can be written:

$$A_{0} + \sum_{n=1}^{\infty} A_{2n-1} r^{2n-1} \cos(2n-1) \boldsymbol{q} = E_{ext} x + \sum_{j \neq 0} \sum_{n=1}^{\infty} \frac{B_{2n-1}}{r_{j}^{2n-1}} \cos(2n-1) \boldsymbol{q}_{j}$$
(2.9)

The left-hand side of Eq. (2.9) can be considered as the real part of the relation

$$A_0 + \sum_{n=1}^{\infty} A_{2n-1} (x + iy)^{2n-1}$$
(2.10)

The right-hand side can also be written as the real part of the following:

$$E_{ext}(x+iy) + \sum_{j\neq 0} \sum_{n=1}^{\infty} B_{2n-1}[x-a_j+i(y-h_j)]^{-2n+1}$$
(2.11)

where a_j and h_j are coordinates of the center of the *j*th cylinder measured from the origin. Now successive differentiation with respect to *x* on both sides of the above is performed and the results at the origin evaluated (this can be done at any other point [McKenzie et al., 1978]). In this manner the following equations are obtained:

$$A_{1} + B_{1}S_{2} + \frac{3!}{2!}B_{3}S_{4} + \dots = E_{ext}$$

$$3! A_{3} + 3! B_{1}S_{4} + \frac{5!}{2!}B_{3}S_{6} + \dots = 0$$

$$5! A_{5} + 5! B_{1}S_{6} + \frac{7!}{2!}B_{3}S_{8} + \dots = 0$$

$$\dots$$

$$(2.12)$$

where $S_n = \sum_{j \neq 0} (\mathbf{a}_j + i\mathbf{h}_j)^{-n}$ are the lattice sums. For more details on calculating the lattice

sums, the reader can refer to chapter 6. The above set of equations can be rewritten as

$$A_{2n-1} + \sum_{m=1}^{\infty} \binom{2n+2m-3}{2n-1} S_{2n+2m-2} B_{2m-1} = E_{ext} \boldsymbol{d}_{n1}, \quad (n = 1, \dots, \infty)$$
(2.13)

where d_{n1} represents the Kronecker delta (1 for n=1, otherwise 0). Using relation (2.6), the following expression is finally obtained:

$$\frac{B_{2n-1}}{\boldsymbol{g}_{2n-1}a^{4n-2}} + \sum_{m=1}^{\infty} \binom{2n+2m-3}{2n-1} S_{2n+2m-2}B_{2m-1} = E_{ext}\boldsymbol{d}_{n1}$$
(2.14)

All the unknown coefficients can be derived by solving the above set of equations and using (2.6) and (2.7). By examination of this linear system, it is found that this is a system of linear algebraic equations with infinite unknowns. The aim here is not to try to solve the system of equations totally, but to concentrate, instead, on deriving only a few values of *B*. This is because considering B_{2n-1} for a sufficiently large *n* has no significant effect on the temperature values. The set of equations can be solved using different well-established methods such as lower-upper decomposition (LU) and singular value decomposition (SVD) methods. When the volume fraction or conductivity of the inclusions increases, more terms should be considered. To give the reader an estimation on the number of terms for consideration, some of the results of Perrins et al. [1979] for the square array (*b*=1) are reported in Table 2.1. As can be seen in this table the most difficult cases happen for perfectly conducting cylinders near to contact.

Table 2.1. The number of terms that should be considered to obtain an accuracy of 5 decimal digits (a maximum percent relative error of 0.01) for the effective conductivity of composites with periodic cylinders in a square array.

$f \setminus k$	2	10	50	~
0.1	1	1	1	1
0.3	2	2	2	2
0.5	2	4	4	4
0.7	4	10	12	8
0.75	6	18	24	36
0.78	8	36	48	100

This makes it possible to obtain the temperature functions, but there is still have no relation for calculating the effective conductivity of the system. This can be done using different methods. Rayleigh [1892] used Green's theorem which will be explained below for uniform solid cylinders. For the case of three-phase non-coated cylinders, however, a volume averaging method [Lu and Lin, 1995; Cheng and Torquato, 1997], which can be more confidently applied to a case involving an imperfect interface, will be explained.

2.1.1 Green's theorem and the effective conductivity

Based on Green's theorem for functions j and y with continuous second derivatives, the following relation, which is Green's second identity, can be written [Spiegel, 1968]:

$$\int_{V} (\mathbf{y} \nabla^{2} \mathbf{j} - \mathbf{j} \nabla^{2} \mathbf{y}) dv = \oint_{\mathbf{s}} \left(\mathbf{y} \frac{\partial \mathbf{j}}{\partial n} - \mathbf{j} \frac{\partial \mathbf{y}}{\partial n} \right) d\mathbf{s} , \qquad (2.15)$$

where V and s are the total volume and surface area, respectively. If both y and j satisfy the Laplace equation through the volume, the following can be written:

$$\oint_{s} \left(\mathbf{y} \, \frac{\partial \mathbf{j}}{\partial n} - \mathbf{j} \, \frac{\partial \mathbf{y}}{\partial n} \right) d\mathbf{s} = 0 \tag{2.16}$$

Now, let us suppose that y = x, j = T and that the medium being investigated is the unit cell of the composite material under study. As a result on can obtain

$$\oint_{\boldsymbol{s}_{cell}} \left(x \frac{\partial T}{\partial n} - T \frac{\partial x}{\partial n} \right) d\boldsymbol{s} = 0 \quad \boldsymbol{s}_{cell} = \boldsymbol{s}_{l} + \boldsymbol{s}_{r} + \boldsymbol{s}_{b} + \boldsymbol{s}_{t} + \boldsymbol{s}_{c}$$
(2.17)

Here, $\mathbf{s}_t, \mathbf{s}_r, \mathbf{s}_b, \mathbf{s}_t$ refer to the left, right, bottom and top surface boundaries of the unit cell, respectively. \mathbf{s}_c is the surface of the cylinder inside the unit cell. The integral on surfaces \mathbf{s}_b and \mathbf{s}_t is zero because $\partial x/\partial n = \partial T/\partial n = 0$. Also, we have

$$\int_{\boldsymbol{s}_l+\boldsymbol{s}_r} x \frac{\partial T}{\partial n} d\boldsymbol{s} = \int_0^1 (x_r - x_l) \frac{\partial T}{\partial x} dy = b \int_0^1 \frac{\partial T}{\partial x} dy = b G$$
(2.18)

$$\int_{\mathbf{s}_l+\mathbf{s}_r} T \frac{\partial x}{\partial n} d\mathbf{s} = (T_r - T_l) = -bE_{ext}$$
(2.19)

$$\int_{\boldsymbol{s}_c} \left(x \frac{\partial T}{\partial n} - T \frac{\partial x}{\partial n} \right) d\boldsymbol{s} = \int_0^{2\boldsymbol{p}} \left(A_1 a^2 + B_1 \right) \cos^2 \boldsymbol{q} \, d\boldsymbol{q} - \int_0^{2\boldsymbol{p}} \left(A_1 a^2 - B_1 \right) \cos^2 \boldsymbol{q} \, d\boldsymbol{q} = 2\boldsymbol{p}B_1 \quad (2.20)$$

In (2.18), G is the average gradient. From the above

$$b G - b E_{ext} + 2pB_1 = 0 (2.21)$$

can be obtained and, as a result, the effective conductivity of the system is

$$k_{e} = \frac{G}{E_{ext}} = 1 - \frac{2pB_{1}}{bE_{ext}} = 1 - \frac{2fg_{1}A_{1}}{E_{ext}}$$
(2.22)

As can be seen, knowing B_1 is enough for obtaining the effective conductivity of the system. The relative conductivity of the medium without inclusions is clearly equal to a unity; thereby, the term $2\mathbf{p}B_1/(bE_{ext})$ is produced as a result of the presence of inclusions in the matrix. This term can be positive (an impairing case), negative (an enhancing case) or equal to zero.

2.1.2 The effective conductivity for the perpendicular direction and Keller theorem

In general, the effective conductivity is a tensor of rank two (see Torquato [2002] for details). For the case $b \neq 1$, the effective conductivity in the y-direction is not equal to that in the x-direction and should be calculated. For this purpose, the same method as explained for the parallel direction may be applied. If the system is rotated by an angle of $\mathbf{a} = \mathbf{p}/2$, we get

$$\frac{B'_{2n-1}}{g_{2n-1}a^{4n-2}} + \sum_{m=1}^{\infty} {\binom{2n+2m-3}{2n-1}} S'_{2n+2m-2} B'_{2m-1} = E_{ext} d_{n1}, \qquad (2.23)$$

where S'_{l} are the lattice sums over the cylinders in this new position. It can be proven that $S'_{2n} = (-1)^n S_{2n}$ for n > 1 and also that $S'_2 = 2\mathbf{p}/b - S_2$ [Nicorovic and McPhedran, 1996]. The effective conductivity now is

$$k'_{e} = 1 - \frac{2\mathbf{p}B'_{1}}{bE_{ext}} = 1 - \frac{2f\mathbf{g}_{1}A'_{1}}{E_{ext}}$$
(2.24)

The effective conductivities in the parallel and perpendicular directions have been linked to each other through the well-known Keller reciprocal relation [Keller, 1964; Milton, 1988; Fel et al., 2000], i.e.,

$$k_{e}(k,1) \times k'_{e}(1/k,1) = 1$$
 (2.25)

This fact can be easily proven. By considering that reversing the conductivity of the phases only makes the sign of g negative and applying the above-mentioned property of the lattice sums (see also Perrins et al., [1979]) from relation (2.23), it is found that

$$\frac{(-1)^{n}B'_{2n-1}}{\boldsymbol{g}_{2n-1}a^{4n-2}} + \sum_{m=1}^{\infty} {\binom{2n+2m-3}{2n-1}} S_{2n+2m-2} (-1)^{m}B'_{2m-1} = E_{ext} \left(1 - \frac{2\boldsymbol{p}B'_{1}}{bE_{ext}}\right) \boldsymbol{d}_{n1}$$
(2.26)

By using Eq. (2.24) and comparing the above relation with (2.14), it is found

$$\frac{B'_{2n-1}}{k'_{e}(1/k_{1},1)} = (-1)^{n} B_{2n-1}$$
(2.27)

Writing the above relation for n = 1 and using Eq. (2.24) again gives

$$k'_{e}(1/k_{1},1) = 1 + \frac{2\mathbf{p}k'_{e}(1/k_{1},1)B_{1}}{bE_{ext}}$$
(2.28)

By applying Eq. (2.22), the following relation is finally obtained

$$k_e(k_1,1) \times k'_e(1/k_1,1) = 1$$
 (2.29)

2.1.3 Explicit forms for the effective conductivity

As can be seen in Table (2.1), for low volume fractions or when the conductivity of the cylinders is small, considering a few values of B_{2n-1} may yield reasonable results. It is more useful to derive an explicit relation for the effective conductivity of the system within these boundaries. On the basis of the method used for truncating (for example square or triangular manner) and the number of the unknowns taken into account, different expressions may be obtained [Manteufel and Todreas, 1994]. Using triangular truncation of the second order gives

$$\frac{B_1}{g_1 a^2} + S_2 B_1 + 3S_4 B_3 = E_{ext}$$
(2.30)

$$\frac{B_3}{g_3 a^6} + S_4 B_1 = 0, \qquad (2.31)$$

whence

$$k_{e} = 1 - \frac{2f}{\frac{1}{g_{1}} + c_{1}f - c_{2}g_{3}f^{4}},$$
(2.32)

where $c_1 = bS_2/p$ and $c_2 = 3S_4^2(b/p)^4$. In Table 2.2 the constants for some of the values of b

have been reported by calculating highly accurate values for the lattice sums [Movchan, 1997; Huang, 1999; Huang, 2001].

Table 2.2 The numerical constants of the explicit relation (2.32) for the parallel and perpendicular directions

	Parallel		Perpendicular	
b	c_1	<i>C</i> ₂	<i>C</i> ₁	c_2
1	1.00000000	0.305828278	1.00000000	0.305828278
$\sqrt{3}$	0.187018134	1.310523128	1.812981866	1.310523128
2	-0.09421981	2.312822989	2.094219807	2.312822989
3	-1.14159216	11.68912746	3.141592162	11.68912746
5	-3.23598776	90.19360281	5.235987756	90.19360281
10	-8.47197551	1443.097645	10.47197551	1443.097645

2.2 Multiphase composites with arrays of uniform and coated circular cylinders

The theory for coated cylinders (see Moosavi and Sarkomaa [2002a]) can be formulated in the same manner as that discussed in the case of solid cylinders. To start with, the solid cylinders given in Fig. 2.1 are replaced by the multi-coated ones shown in Fig. 2.2. As can be seen in Fig. 2.2, the multi-coated cylinders under study are made up of a core of radius a_1 and N-2 coating layers placed in the regions $a_{i-1} \le r \le a_i$ (i = 2, ..., N - 1), respectively. With these considerations, the volume fractions occupied by the core and coating layers can be obtained from $f_i = \mathbf{p}(a_i^2 - a_{i-1}^2)/b$, where $a_0 = 0$ and the total volume fraction can be expressed as $F = \sum_{i=1}^{N-1} f_i$. The conductivity of phase *i* is denoted by k_i . For the composite, the temperature profiles inside the core, coating layers and the matrix can be expressed as:

$$T_{1} = C_{0} + \sum_{n=1}^{\infty} C_{2n-1} r^{2n-1} \cos(2n-1)\boldsymbol{q}$$
(2.33)

$$T_{i} = A_{0}^{i} + \sum_{n=1}^{\infty} \left[A_{2n-1}^{i} r^{2n-1} + B_{2n-1}^{i} r^{-2n+1} \right] \cos(2n-1) \boldsymbol{q} \qquad (i = 2, \cdots, N)$$
(2.34)



Figure 2.2 The multicoated circular cylinder under study.

Following the same discussion as that given for solid cylinders, the following equation can be derived for the system:

$$A_0^N + \sum_{n=1}^{\infty} A_{2n-1}^N r^{2n-1} \cos(2n-1) \boldsymbol{q} = E_{ext} r \cos \boldsymbol{q} + \sum_{j \neq 0} \sum_{n=1}^{\infty} \frac{B_{2n-1}^N}{r_j^{2n-1}} \cos(2n-1) \boldsymbol{q}_j$$
(2.35)

After performing the required steps explained in section 2.1, the governing equations of the system can be derived as

$$\frac{B_{2n-1}^{N}}{\boldsymbol{g}_{2n-1}^{N}a_{N-1}^{4n-2}} + \sum_{m=1}^{\infty} \binom{2n+2m-3}{2n-1} S_{2n+2m-2} B_{2m-1}^{N} = E_{ext} \boldsymbol{d}_{n1}, \qquad (2.36)$$

where $\boldsymbol{g}_{2n-1}^{N}$ can be obtained by successively applying the following procedure

$$\boldsymbol{g}_{2n-1}^{i} = \frac{\left(k_{i} + k_{i-1}\right)a_{i-2}^{4n-2} + \left(k_{i} - k_{i-1}\right)\boldsymbol{g}_{2n-1}^{i-1}a_{i-1}^{4n-2}}{\left(k_{i} - k_{i-1}\right)a_{i-2}^{4n-2} + \left(k_{i} + k_{i-1}\right)\boldsymbol{g}_{2n-1}^{i-1}a_{i-1}^{4n-2}} \qquad i > 2$$

$$(2.37)$$

which is the result of implementing the boundary conditions between the phases, i.e.

$$T_i = T_{i+1}$$
 and $k_i \frac{\partial T_i}{\partial r} = k_{i+1} \frac{\partial T_{i+1}}{\partial r}$ $r = a_i$ $(i = 1, \dots, N-1)$ (2.38)

The effective conductivity of the system can be derived again using Green's theorem. In applying Green's theorem, only the surface of the boundaries of the unit cell and that of the outermost coated cylinder, which is in contact with the matrix, are considered. Therefore, it is not surprising that the same formula is derived as that for solid cylinders, i.e.,

$$k_{e} = 1 - \frac{2\mathbf{p}B_{1}^{N}}{bE_{ext}} = 1 - \frac{2F\mathbf{g}_{1}^{N}A_{1}^{N}}{E_{ext}}$$
(2.39)

2.2.1 Phase abandonment in multicoated composites

The behaviour of singly coated cylinders has been studied by Nicorovici et al. [1993a; 1993b; 1995]. The coating sequence effect has also been investigated by Lu [1997a]. Here the attention is mainly focused on the results reported by Nicorovici et al. The property selected by these authors was the dielectric constant, and therefore they were able to choose the property of the layers from $-\infty$ to $+\infty$. The dielectric constant expresses the ability of a dielectric to store electrostatic energy under the influence of an electric field. In electrostatic the equation $\nabla \cdot (\mathbf{e} \nabla H) = 0$ needs to be solved, where **e** is the dielectric constant and H the electric potential. There is also $D = -e \nabla H$ where D denotes electric flux density. These equations are mathematically equivalent to $\nabla (k\nabla T) = 0$ and $F = -k\nabla T$, respectively, in heat conduction. For some transport properties, such as dielectric constant, negative values are of physical significance and are realisable. Materials can be designed to exhibit negative values of certain transport properties, such as magnetic permeability (a constant of proportionality that exists between magnetic induction and magnetic field intensity), even though they do not normally exhibit negative values for these properties [Vessalago, 1968]. It is also possible to go further and design materials with two negative transport properties [Smith et al., 2000]. These materials may exhibit strange behaviour. For example, materials where both the

dielectric permittivity and magnetic permeability are negative (so-called left-handed materials) demonstrate a negative refractive index [Pendry et al., 2001].

Next, the discussion is extended to a multicoated case (see Moosavi and Sarkomaa [2002a]). The transport property is here denoted by \boldsymbol{e} . As can be seen in Eq. (2.36), the term $\boldsymbol{x}_{2n-1}^{N} = 1/(\boldsymbol{g}_{2n-1}^{N} a_{N-1}^{4n-2})$ plays an important role in the response of the system to the applied field, as this is the only part of the set of equations which contains information on the property and volume fraction of the layers. Therefore, if a completely different set of coating cylinders with N^* phases satisfies the condition $\boldsymbol{x}_{2n-1}^{N*} = \boldsymbol{x}_{2n-1}^{N}$, it can be applied instead of the original coated cylinders, and the effective transport property of the system in both states will be the same. For example, for the case of coating layer *i*, for which $\boldsymbol{e}_i = 0$, $\boldsymbol{g}_n^{i+1} = 1$ is derived. As can be seen, \boldsymbol{g}_n^{i+1} does not contain any information on the property and radius of the layers under layer *i*; therefore, they can be arbitrarily chosen. The same occurs when $\boldsymbol{e}_i = \pm\infty$, which yields $\boldsymbol{g}_n^{i+1} = -1$.

Now, a more interesting case is considered in which $\mathbf{e}_i + \mathbf{e}_{i-1} = 0$. Nicorovici et al. [1993a] showed that for a singly coated system, cases $\mathbf{e}_2 = -\mathbf{e}_1$ and $\mathbf{e}_2 = \mathbf{e}_1$ surprisingly demonstrate the same response to the applied field. In other words, coated cylinders can be replaced by solid ones that have a property equal to that of the core and a radius equal to that of the coating layer. The reason is clear. Let us derive \mathbf{x}_{2n-1}^3 :

$$\boldsymbol{x}_{2n-1}^{3} = \frac{(\boldsymbol{e}_{3} - \boldsymbol{e}_{2})(\boldsymbol{e}_{2} - \boldsymbol{e}_{1})a_{1}^{4n-2} + (\boldsymbol{e}_{3} + \boldsymbol{e}_{2})(\boldsymbol{e}_{2} + \boldsymbol{e}_{1})a_{2}^{4n-2}}{(\boldsymbol{e}_{3} + \boldsymbol{e}_{2})(\boldsymbol{e}_{2} - \boldsymbol{e}_{1})a_{1}^{4n-2} + (\boldsymbol{e}_{3} - \boldsymbol{e}_{2})(\boldsymbol{e}_{2} + \boldsymbol{e}_{1})a_{2}^{4n-2}} \times \frac{1}{a_{2}^{4n-2}}$$
(2.40)

When $\mathbf{e}_2 = \mathbf{e}_1$ or $\mathbf{e}_2 = -\mathbf{e}_1$, \mathbf{x}_{2n-1}^3 reduces to $(\mathbf{e}_3 + \mathbf{e}_1)/[(\mathbf{e}_3 - \mathbf{e}_1)a_1^{4n-2}]$. Since all the parameters remain unchanged, the responses of the systems are exactly the same. It can also be shown that the cases $\mathbf{e}_2 = -\mathbf{e}_3$ and $\mathbf{e}_2 = \mathbf{e}_1$ conditionally produce the same response. The condition is that for the two-phase case, the radius of the dispersed phase must be equal to a_2^2/a_1 , where a_1 and a_2 are the radii of the core and shell, respectively, for the three-phase system. This

result is important, since this way when a_1 and a_2 are both small but the ratio of a_2/a_1 is large, the three-phase system will give the response of a two-phase system with the radius of the inclusions equal to a_2^2/a_1 , which can be completely considerable. As a result, the response of a concentrated system can be produced with a system that has a very small total volume fraction.

In the following, a multicoated system when the sum of the dielectric constant of two dispersed layers, *i*-1 and *i*, is equal to zero, is considered. For this case

$$\mathbf{x}_{2n-1}^{i+1} = \frac{\mathbf{e}_{i+1} - \mathbf{e}_i + (\mathbf{e}_{i+1} + \mathbf{e}_i) \mathbf{x}_{2n-1}^i a_i^{4n-2}}{\mathbf{e}_{i+1} + \mathbf{e}_i + (\mathbf{e}_{i+1} - \mathbf{e}_i) \mathbf{x}_{2n-1}^i a_i^{4n-2}} \times \frac{1}{a_i^{4n-2}} \qquad i \ge 1$$
(2.41)

This relation would yield the same \mathbf{x}_{2n-1}^{i+1} and, as a result, the same \mathbf{x}_{2n-1}^{N} if layer *i* had the same property as layer *i*-1 and all the layers under layer *i*-1 were magnified by a factor of $(a_i/a_{i-1})^4$. Layer *i*-1 undergoes two changes; magnification by occupying the place of layer *i* and reduction due to the extension of layer *i*-2. Thus, this case can be materialised only if $a_{i-1} > \sqrt{a_i a_{i-2}}$. This means that for the case where the property of the layers is equal to $(\mathbf{e}_1, \pm \mathbf{e}_1, \dots, \pm \mathbf{e}_1, \mathbf{e}_2)$, the following can always be written:

$$\boldsymbol{e}_{e}(\boldsymbol{e}_{1},\boldsymbol{e}_{1},\cdots,\boldsymbol{e}_{1},\boldsymbol{e}_{2}) \times \frac{1}{\boldsymbol{e}_{e}(\boldsymbol{e}_{1},\pm\boldsymbol{e}_{1},\cdots,\pm\boldsymbol{e}_{1},\boldsymbol{e}_{2})} = 1$$
(2.42)

The system satisfies the Keller reciprocal relation (see chapter 4); and thus we have

$$\boldsymbol{e}_{e}(\boldsymbol{e}_{1},\ldots,\boldsymbol{e}_{N}) \times \boldsymbol{e}_{e}'(1/\boldsymbol{e}_{1},\ldots,1/\boldsymbol{e}_{N}) = 1$$
(2.43)

By applying (2.42) and (2.43), we obtain

$$\boldsymbol{e}_{e}(\boldsymbol{e}_{1}, \boldsymbol{e}_{1}, \cdots, \boldsymbol{e}_{1}, \boldsymbol{e}_{2}) \times \boldsymbol{e}_{e}'(1/\boldsymbol{e}_{1}, \pm 1/\boldsymbol{e}_{1}, \cdots, \pm 1/\boldsymbol{e}_{1}, 1/\boldsymbol{e}_{2}) = 1$$
(2.44)

In addition to inspecting \mathbf{x}_{2n-1}^{i+1} , let us now also study \mathbf{x}_{2n-1}^{i} when $\mathbf{e}_{i} + \mathbf{e}_{i-1} = 0$, i.e.,

$$\mathbf{x}_{2n-1}^{i} = \frac{(\mathbf{e}_{i} - \mathbf{e}_{i-1}) + (\mathbf{e}_{i} + \mathbf{e}_{i-1}) \mathbf{x}_{2n-1}^{i-1} a_{i-1}^{4n-2}}{(\mathbf{e}_{i} + \mathbf{e}_{i-1}) + (\mathbf{e}_{i} - \mathbf{e}_{i-1}) \mathbf{x}_{2n-1}^{i-1} a_{i-1}^{4n-2}} \times \frac{1}{a_{i-1}^{4n-2}} \qquad i \ge 1$$
(2.45)

An inspection of the above relation shows that when $\mathbf{e}_i = -\mathbf{e}_{i-1}$, another equivalent system can be obtained differently. In this system, the layers 1,...,*i*-3 have been extended by a factor of $(a_{i-1}/a_{i-2})^4$. The layer *i*-1 now has the property equal to that of layer *i*-2 and its outer radius is $a_{i-1} \times (a_{i-1}/a_{i-2})$. This system can be materialized if $a_{i-1} < \sqrt{a_i a_{i-2}}$.

The results of the present study have been verified using the finite element method. Consider, for instance, the following simple example. Based on the above formulations for a triply coated system, cases $\mathbf{e}_1 = -\mathbf{e}_2 = \mathbf{e}_3$ and $\mathbf{e}_1 = \mathbf{e}_2 = \mathbf{e}_3$ should give the same response to the applied field. The results for both systems are given in Figures 2.3(*a*, *b*). As can be seen in both systems, the fields through the matrix are exactly equal, which means that both systems have exactly the same effective property.



Figure 2.3 The equipotential contours inside a unit cell. The original system (a) and the equivalent system (b).

2.3 Three-phase composites with arrays of solid circular cylinders

In this section, the problem of calculating the effective conductivity of composites made up of two types of cylindrical inclusion arranged in a periodic array and embedded in a continuous phase is discussed [see Moosavi and Sarkomaa, 2003b], as depicted in Fig. 2.4. The configuration of the geometry selected for the study makes it possible to construct many periodic structures that can be extensively found in literature. First, the Rayleigh method will be developed for a three-phase composite system. Then, the algorithm is verified, comparing the results with some existing numerical results. Finally, the behaviour of the system is inspected. Only the mathematical algorithms for the *x*-direction will be explained. As outlined in section (2.1.2), the same mathematical formulation can be extended for the *y*-direction. Furthermore, the effective conductivities can be related to each other using the Keller theorem.



Figure 2.4 The structure of the three-phase materials under study

2.3.1 Governing equations

Suppose that the origin of cartesian coordinates has been placed at the centre of a cylinder of type one in a unit cell of the system in which the x- and y-axes are parallel to the sides of the unit cell (see Fig. 2.4). Furthermore, assume that the matrix of the composite under study has unit conductivity, and the periodicity of the system in the y-direction is equal to a unity for

greater generality. Also the periodicity of the system in the x-direction is denoted by b. Applying these conditions, the conductivity of the cylinders is denoted by k_1 and k_2 , the radiuses by a_1 and a_2 and the volume fractions by f_1 and f_2 for the cylinders of type one and two, respectively. Let us assume that a uniform field of magnitude E_{ext} has been applied along the x-axis of the system in the negative direction. By taking the center of one of the cylinders of type i (i = 1,2) as the origin of polar coordinates (r,q), the temperature within the cylinder can be given as

$$T_{i}(r,\boldsymbol{q}) = C_{0}^{i} + \sum_{n=1}^{\infty} C_{2n-1}^{i} r^{2n-1} \cos(2n-1)\boldsymbol{q}$$
(2.46)

and for the temperature outside the cylinder the following may be written

$$T_{m,i}(r,\boldsymbol{q}) = A_0^i + \sum_{n=1}^{\infty} \left(A_{2n-1,i} r^{2n-1} + B_{2n-1,i} r^{-2n+1} \right) \cos\left(2n-1\right) \boldsymbol{q}$$
(2.47)

where *m* stands for the matrix. Note that as in the previous cases in (2.48) and (2.49), the terms of even degree are not present because of the anti-symmetry of the temperature profiles around q = p/2. At the surface of each type of cylinder, we have

$$T_i = T_{3i}, \quad k_i \frac{\partial T_i}{\partial r} = \frac{\partial T_{3i}}{\partial r} \quad r = a_i$$
 (2.48)

By applying the above boundary conditions, the following relations can be obtained:

$$A_{2n-1}^{i} = \frac{B_{2n-1}^{i}}{\boldsymbol{g}_{2n-1}^{i} a_{i}^{4n-2}}$$
(2.49)

$$C_{2n-1}^{i} = \frac{B_{2n-1}^{i}}{\boldsymbol{c}_{2n-1}^{i} a_{i}^{4n-2}},$$
(2.50)

where

$$g_{2n-1}^{i} = \frac{1-k_{i}}{1+k_{i}}$$
 $c_{2n-1}^{i} = \frac{1-k_{i}}{2}$ (2.51)

In the same manner as explained for the two-phase case, the terms in (2.47) can be analyzed. By separating the effect of sources situated on the axes of the cylinders of type one and two the following can be written:

$$A_{0}^{i} + \sum_{n=1}^{\infty} A_{2n-1}^{i} r^{2n-1} \cos(2n-1) \boldsymbol{q} = E_{ext} x + \sum_{j\neq 0}^{\infty} \sum_{m=1}^{\infty} \frac{B_{2m-1}^{i}}{r_{j,i}^{2m-1}} \cos(2m-1) \boldsymbol{q}_{j,i} + \sum_{j}^{\infty} \sum_{m=1}^{\infty} \frac{B_{2m-1}^{2-\boldsymbol{d}_{i2}}}{r_{j,2-\boldsymbol{d}_{i2}}^{2m-1}} \cos(2m-1) \boldsymbol{q}_{j,2-\boldsymbol{d}_{i2}}, \qquad (2.52)$$

where $r_{j,i}$ and $q_{j,i}$ are measured from the center of the *j*th cylinder situated in the array of cylinders of type *i*. If the above equation is written in complex form [Moosavi and Sarkomaa, 2003a] and successive differentiation is performed with respect to *x* on both sides of the above equation, and the results evaluated at the origin of cylinder of type *i*, the final result after using (2.49) would appear as

$$\frac{B_{2n-1}^{i}}{\boldsymbol{g}_{2n-1}^{i}a_{i}^{4n-2}} + \sum_{m=1}^{\infty} \binom{2n+2m-3}{2n-1} \left(S_{2n+2m-2}^{1}B_{2m-1}^{i} + S_{2n+2m-2}^{2}B_{2m-1}^{2-\boldsymbol{d}_{1}} \right) = E_{ext}\boldsymbol{d}_{n1}, \qquad (2.53)$$

Here, $S_n^i = \sum_{j \neq 0} (\mathbf{a}_{j,i} + i\mathbf{h}_{j,i})^{-n}$ are the lattice sums over the cylinders of type *i*, where $\mathbf{a}_{j,i}$ and

 $\boldsymbol{h}_{j,i}$ are the coordinates of center of the cylinder *j* of type *i* in the current polar coordinates.

2.3.2 Volume averaging technique for determining the effective conductivity of the system

Based on Fourier's law, the effective conductivity of the system can be derived using the following formula:
$$\langle \boldsymbol{F} \rangle = -k_e \langle \nabla T \rangle$$
 (2.54)

where $\langle F \rangle = (1/V_{cell}) \int_{V_{cell}} F dV$ and $\langle \nabla T \rangle = (1/V_{cell}) \int_{V_{cell}} \nabla T dV$ are the average heat flux and temperature gradient over the unit cell respectively. The average heat flux can be considered

temperature gradient over the unit cell, respectively. The average heat flux can be considered because of the matrix and cylinders of type one and two, i.e.,

$$\left\langle \boldsymbol{F} \right\rangle = \frac{1}{V_{cell}} \left[\int_{V_1} \boldsymbol{F} \, dV + \int_{V_2} \boldsymbol{F} \, dV + \int_{V_3} \boldsymbol{F} \, dV \right]$$
(2.55)

or

$$\left\langle \boldsymbol{F} \right\rangle = -\frac{1}{V_{cell}} \left[\left(k_1 - 1 \right) \int_{V_1} \nabla T_1 dV + \left(k_2 - 1 \right) \int_{V_2} \nabla T_2 dV + \int_{V_{cell}} \nabla T dV \right], \quad (2.56)$$

where V_1 , V_2 and V_3 are the volumes of cylinders of type one and two and the matrix placed in the unit cell, respectively. The average temperature gradient can also be written as follows:

$$\left\langle \nabla T \right\rangle = \frac{1}{V_{cell}} \left[\int_{V_{cell}} \nabla T dV \right]$$
(2.57)

From Eqs. (2.56) and (2.57), we can obtain

$$\langle \boldsymbol{F} \rangle = \langle \boldsymbol{S}_{P} \rangle_{1} + \langle \boldsymbol{S}_{P} \rangle_{2} - \langle \nabla T \rangle,$$
 (2.58)

where

$$\left\langle \boldsymbol{S}_{P}\right\rangle_{i} = \frac{1-k_{i}}{V_{cell}} \int_{V_{i}} \nabla T_{i} dV$$
(2.59)

or equivalently, using Green's first identity [Spiegel, 1968], we have

$$\left\langle S_{P}\right\rangle_{i} = \frac{1-k_{i}}{V_{cell}} \int_{S_{i}} T_{i} \mathbf{n} dS$$
(2.60)

Here, s_i is the surface of inclusion of type *i* and *n* expresses the unit outward normal vector to the surface. After using the orthogonality properties of trigonometric functions, the following can be obtained:

$$\langle S_P \rangle_i = \frac{1 - k_i}{V_{cell}} \int_0^{2\mathbf{p}} C_1^i \cos^2 q \, dq \, \mathbf{i} = \frac{\mathbf{p} a_i^2 (1 - k_i)}{V_{cell}} C_1^i \, \mathbf{i} = \frac{2\mathbf{p} B_1^i}{V_{cell}} \mathbf{i}$$
 (2.61)

By substituting the resultants in Eq. (2.58) and taking into account $\langle \nabla T \rangle = E_{ext} \mathbf{i}$ and $V_{cell} = b$, the final result for the effective conductivity can be given as

$$k_{e} = 1 - \frac{2\boldsymbol{p}(B_{1}^{1} + B_{1}^{2})}{bE_{ext}} = 1 - \frac{2f_{1}\boldsymbol{g}_{1}^{1}A_{1}^{1} + 2f_{2}\boldsymbol{g}_{1}^{2}A_{1}^{2}}{E_{ext}}$$
(2.62)

or more generally, for a case in which M cylinders are placed in the unit cell, the effective conductivity of the system can be obtained using

$$k_{e} = 1 - 2\mathbf{p} \sum_{i=1}^{M} B_{1}^{i} / (V_{cell} E_{ext})$$
(2.63)

2.3.3 The effective conductivity in explicit forms

To assist calculating the conductivity of the system under study, some relations are here derived in explicit forms. Based upon the triangular truncation of second order, the following can be obtained:

$$k_{e} = 1 - \frac{2f_{1}}{(\boldsymbol{l}_{1}\boldsymbol{l}_{2} - \boldsymbol{x}_{1}\boldsymbol{x}_{2})/(\boldsymbol{l}_{2} - \boldsymbol{x}_{2})} - \frac{2f_{2}}{(\boldsymbol{l}_{1}\boldsymbol{l}_{2} - \boldsymbol{x}_{1}\boldsymbol{x}_{2})/(\boldsymbol{l}_{1} - \boldsymbol{x}_{1})},$$
(2.64)

with

$$\boldsymbol{I}_{i} = \frac{1}{\boldsymbol{g}_{1}^{i}} + c_{1}f_{i} - c_{2}\boldsymbol{g}_{3}^{i}f_{i}^{4} - c_{3}\boldsymbol{g}_{3}^{2-\boldsymbol{d}_{i2}}f_{i}f_{2-\boldsymbol{d}_{i2}}^{3}$$
(2.65)

$$\mathbf{x}_{i} = c_{4}f_{i} - c_{5}\left(\mathbf{g}_{3}^{i}f_{i}^{4} + \mathbf{g}_{3}^{2-\mathbf{d}_{i2}}f_{i}f_{2-\mathbf{d}_{i2}}^{3}\right),$$
(2.66)

where $c_1 = S_2^1 b/p$, $c_2 = 3(b/p)^4 S_4^{1^2}$, $c_3 = 3(b/p)^4 S_4^{2^2}$, $c_4 = S_2^2 b/p$ and $c_5 = 3S_4^1 S_4^2 (b/p)^4$. The higher orders may be ignored to obtain a simpler relation

$$k_e = 1 - \frac{2f_1}{w/u_2} - \frac{2f_2}{w/u_1}, \qquad (2.67)$$

where

$$\boldsymbol{w} = (1/\boldsymbol{g}_1^1 + c_1 f_1)(1/\boldsymbol{g}_1^2 + c_1 f_2) - c_4^2 f_1 f_2$$
(2.68)

$$\boldsymbol{u}_{i} = 1/\boldsymbol{g}_{1}^{i} + (c_{1} - c_{4})f_{i}$$
(2.69)

The constants for cases b = 1 and $b = \sqrt{3}$ (the parallel and perpendicular directions) are listed in Table 2.3.

$b \rightarrow$	1	$\sqrt{3}$ (Parallel)	$\sqrt{3}$ (Perpendicular)
c_1	1.00000000	0.187018134	1.812981866
<i>c</i> ₂	0.305827837	1.310523128	1.310523128
<i>C</i> ₃	7.645695831	1.310523128	1.310523128
<i>C</i> ₄	1.00000000	1.812981866	0.187018134
<i>c</i> ₅	-1.529139176	-1.310523128	-1.310523128

Table 2.3 The numerical constants of the analytical formula (2.64)

2.3.4 Behaviour of the system and phase abandonment

Before starting the discussion on the results of the three-phase system, it is helpful to verify the validity of the extension to the three-phase system. A series of calculations for two-phase composites with uniform cylinders arranged either in square (b = 1) or hexagonal orders $(b = \sqrt{3})$ will be performed and the results then compared with those reported by Perrins et al. [1979].



Figure 2.3 A comparison between the present results and those reported by Perrins et al. [1979].

The two-phase cases can be constructed from the three-phase one simply by applying $f_1 = f_2$ and $k_1 = k_2$. For this purpose highly accurate values for lattice sums over cylinders of type one and two were derived and Eq. (2.53) (*i*=1,2) was solved numerically using LU decomposition method [Press et al., 1986]. Taking into account 100 unknowns of $B_{2n-1,1}$ and $B_{2n-1,2}$ gives us a measure of obtaining accurate results for all the volume fractions and conductivities considered [Perrins et al., 1979]. In Fig. 2.3 the results are compared for both the square and hexagonal arrays for the most challenging case, i.e., the case of perfectly conducting cylinders. As can be seen for all the values of volume fractions, the results of the two studies are in excellent agreement.

Figure 2.4 shows a typical result for the effective conductivity of the system for both the parallel and perpendicular directions. The volume fractions are $f_1 = 0.4$ and $f_2 = 0.4$, and the periodicity in the x-direction was supposed to be $b = \sqrt{3}$.



Figure 2.4 The contours of the effective conductivity for the perfect interface case. $f_1 = 0.4$ and $f_2 = 0.4$

For deriving the conductivity of the system in the perpendicular direction either Eq. (2.53) can be solved and Eq. (2.62) applied or, alternatively, the Keller theorem can be used for this purpose. Through a careful examination of this figure, it appears that increasing or decreasing the conductivity of both types of cylinders may enhance or diminish the conductivity of the system, respectively, which is obvious and remains correct for both directions. Furthermore, the system demonstrates higher effective conductivity in the perpendicular direction. This behavior is a consequence of the rectangular shape of the unit cell which provides a more (less) important role for the cylinders with lower conductivity in the parallel (perpendicular) direction. Interestingly, for the case of mono-sized cylinders with $k_1 = \infty$ and $k_2 = 0$, increasing the volume fraction of the cylinders causes the conductivity of the system to approach zero in the parallel direction and approach infinity in the perpendicular direction (see Table 2.4). When perfectly insulating cylinders touch each other, they form a barrier which prevents heat flow in the parallel direction. This behavior can also be observed for all systems for which b > 1. For the case b = 1, however, the system is isotropic and the same results can be expected for both directions. For this case, the same type cylinders are not able to touch each other and a limited value for the effective conductivity of the system can be expected. Surprisingly, it was found that the effective conductivity of the system is simply a unity.

F	k _e	k'_{e}
0.1	0.983871	1.01639
0.2	0.937008	1.06723
0.3	0.863599	1.15794
0.4	0.769417	1.29969
0.5	0.660207	1.51468
0.6	0.539863	1.85232
0.7	0.408730	2.44660
0.8	0.261159	3.82908
0.9	3E-7	19.253
$p/(2\sqrt{3})$	0	-

Table 2.4 The effective conductivity of three-phase materials made up of mono-sized cylinders for which $k_1 = 0$ and $k_2 = \infty$.

This result can be confirmed using the Keller theorem as follows: since the system is isotropic and interchange between the material of the cylinders keeps the system unchanged, using Eq. (2.43) it is possible to get

$$k_{e}(k,1/k,1) \times k_{e}'(1/k,k,1) = k_{e}(k,1/k,1) \times k_{e}(k,1/k,1) = 1$$
(2.70)

Considering k = 0 proves our case. Sculgasser [1992] has shown that in a three-phase system with interchangeable phases (see Fig. 2.5), when one of the phases has conductivity equal to k and the two remaining phases are perfectly conducting and non-conducting, the effective conductivity of the system is k. From the above results, it is clear that it is not necessary for the first phase to be interchangeable, and it can simply be a matrix.

For the case b = 1 with non-equal sized cylinders, if $f_i \leq \frac{\mathbf{p}}{4} (\sqrt{2} - 1)^2$, $f_{2-\mathbf{d}_{i2}}$ can be increased freely to the touching value limit, and the effective conductivity of the system can approach infinity or zero, depending on the conductivity of the touching cylinders.



Figure 2.5 The structure investigated by Schulgasser et al. [1992]

Figure 2.6 shows the results of the effective conductivity for a system with a lower total volume fraction, i.e., $f_1 = 0.4$ and $f_2 = 0.2$. *b* as before is equal to $\sqrt{3}$. A comparison with Fig. 2.4 reveals that the case $k_1 = k_2 = 1$ is the only situation in which both systems for the given conductivities present the same effective conductivity. In this situation $\mathbf{g}_1 = \mathbf{g}_2 = 0$, which leads to $B_1^1 = B_1^2 = 0$, and as a result, $k_e = 1$.



Figure 2.6 The contours of the effective conductivity for the perfect interface case. $f_1 = 0.4$ and $f_2 = 0.2$.

Figures 2.4 and 2.6 also show that having cylinders with conductivities equal to the conductivity of the matrix is not the only condition for the effective conductivity to be equal to the conductivity of the matrix. In fact, this case is a special state of the following general situation:

$$B_1^1 + B_1^2 = f_1 \boldsymbol{g}_1 A_1^1 + f_2 \boldsymbol{g}_2 A_1^2 = 0$$
(2.71)

The importance of the situation $k_1 = k_2 = 1$ ($B_1^1 = B_1^2 = 0$) is that it is independent of the values of f_1 , f_2 and b, and for all theses situations, it would be found that $k_e = 1$, which is physically obvious. This behavior does not hold for the other values of the conductivities.

2.3.5 Heat transfer with interfacial resistance

If the dimensionless interfacial resistances [Torquato and Rintoul, 1995] are denoted by R_i (i = 1,2), the boundary conditions given in (2.48) will change to the following shape [Moosavi and Sarkomaa, 2003b]:

$$\frac{k_i}{R_i a_i} (T_i - T_{3i}) = -k_i \frac{\partial T_i}{\partial r} = -\frac{\partial T_{3i}}{\partial r} \quad \text{on } a_i \ (i = 1, 2)$$
(2.72)

Therefore, for the coefficients of the temperature profiles, we obtain

$$A_{2n-1}^{i} = \frac{B_{2n-1}^{i}}{\boldsymbol{g}_{2n-1}^{i} a_{i}^{4n-2}}$$
(2.73)

$$C_{2n-1}^{i} = \frac{B_{2n-1}^{i}}{c_{2n-1}^{i} a_{i}^{4n-2}},$$
(2.74)

where

$$\boldsymbol{g}_{2n-1}^{i} = \frac{1 - k_{i} + R_{i}(2n-1)}{1 + k_{i} + R_{i}(2n-1)} \qquad \boldsymbol{c}_{2n-1}^{i} = \frac{1 - k_{i} + R_{i}(2n-1)}{2}$$
(2.75)

When the interface is imperfect, the volume averaged heat flux may be formulated using the same formula as for a perfect case, but the volume-averaged temperature will contain extra terms because of the temperature jump in the interfaces, i.e.,

$$\langle \nabla T \rangle = \frac{1}{V_{cell}} \left[\int_{V_{cell}} \nabla T \, dV + \int_{s_1} (T_{31} - T_1) n \, dS + \int_{s_2} (T_{32} - T_2) n \, dS \right],$$
 (2.76)

Therefore, the volumes averaged of the temperature and flux can be related to each other by

$$\left\langle \boldsymbol{F} \right\rangle = \left\langle \boldsymbol{S}_{P} \right\rangle_{1} + \left\langle \boldsymbol{S}_{P} \right\rangle_{2} + \left\langle \boldsymbol{S}_{I} \right\rangle_{1} + \left\langle \boldsymbol{S}_{I} \right\rangle_{2} - \left\langle \nabla T \right\rangle, \qquad (2.77)$$

where

$$\left\langle \boldsymbol{S}_{P}\right\rangle_{i} = \frac{1-k_{i}}{V_{cell}} \int_{V_{i}} \nabla T_{i} dV$$
(2.78)

$$\left\langle \boldsymbol{S}_{I}\right\rangle_{i} = \frac{1}{V_{cell}} \int_{\boldsymbol{S}_{i}} (T_{3i} - T_{i}) \boldsymbol{n} \, dS \tag{2.79}$$

By performing the integrals, we can derive

$$\left\langle \boldsymbol{S}_{P}\right\rangle_{i} = \frac{2\boldsymbol{p}B_{1}^{i}}{V_{cell}} \cdot \frac{1-k_{i}}{R_{i}+1-k_{i}} \mathbf{i}$$
(2.80)

$$\left\langle \boldsymbol{S}_{I}\right\rangle_{i} = \frac{2\boldsymbol{p}\boldsymbol{B}_{1}^{i}}{V_{cell}} \cdot \frac{R_{i}}{R_{i}+1-k_{i}}\mathbf{i}$$
(2.81)

By substituting the resultants in Eq. (2.77), the same formula is interestingly obtained as for a perfect interface

$$k_{e} = 1 - 2\boldsymbol{p} \, \frac{B_{1}^{1} + B_{1}^{2}}{bE_{ext}} = 1 - \frac{2f_{1}\boldsymbol{g}_{1}^{1}A_{1}^{1} + 2f_{2}\boldsymbol{g}_{1}^{2}A_{1}^{2}}{E_{ext}}$$
(2.82)

Chiew [1987], in studying the effect of resistance on the conductivity of two-phase dispersions made up of a random array of spheres, showed that there may be a critical situation in which the system does not sense the presence of the inclusions and behaves like a uniform matrix. A study into the behaviour of composites consisting of periodic and random arrays of uniform cylinders [Lu and Lin, 1995 and Lu and Song, 1996] and periodic arrays of spheres [Lu, 1997b and Cheng and Torquato, 1997] revealed that for all these cases, a critical situation occurs if R = k - 1. Here, the discussion will be extended to three-phase systems. Again, the extension is validated by comparing the results with those of two-phase systems [Lu and Lin, 1995] as shown in Table 2.5.

Table 2.5 A comparison between the present results and those reported by Lu and Lin [1995] for the square array of the uniform cylinders.

	Lu and Lin	Present	Lu and Lin	Present	Lu and Lin	Present
	[1995]		[1995]		[1995]	
$f \setminus (1/R)$	8		0.1		0.0001	
0.2	1.49906	1.49906	1.48676	1.48676	0.718703	0.718703
0.5	3.07141	3.07141	2.98364	2.98364	0.413291	0.413291
0.6	4.32252	4.32252	4.12547	4.12547	0.327454	0.327454
0.7	7.36828	7.36828	6.68414	6.68414	0.241682	0.241682
0.74	10.8513	10.8513	9.18034	9.18033	0.204152	0.204152
0.76	15.1110	15.1110	11.6103	11.6103	0.182844	0.182844
0.78	33.79	33.79	16.8449	16.8449	0.15565	0.15565
0.785398	-	-	19.7137	19.7136	0.15	0.14

As can be seen, the results of the two studies agree very well except for the case f = 0.785398, 1/R = 0.0001. In the present study the number of terms included in the process of calculation was increased to more than 100 to see if the difference might be due to the number of terms. The obtained result was again about 0.14. Since the other values are greatly in agreement with those of Lu and Lin [1995], the suggested value for this case in this study is 0.14.

Figures 2.7 and 2.8 show the contours of the effective conductivity of the system for given conductivities. k_1 and k_2 were supposed to be 101 (this way, the logarithm of $R_1 = R_2 = 100$ would be an integer number) and volume fractions $f_1 = f_2 = 0.4$ in Fig. 2.7

and $f_1 = 0.4$, $f_2 = 0.2$ in Fig. 2.8. As can be seen, the contours of the effective conductivity are similar to those in Figs. 2.4 and 2.6, and the effective conductivity of the materials can be highly affected because of the presence of interfacial resistance, and based upon the values of R_1 and R_2 , the materials may exhibit a conductivity more, equal or even less than that of the matrix alone. Among the points rendering the presence of inclusions irrelevant, the case $R_i = k_i - 1(i=1,2)$ is of particular interest. In this situation, based on Eq. (2.75), the dipole polarisabilities are zero, which leads to $B_1^1 = B_1^2 = 0$, and the state of the system resembles that of a system of perfect interfaces made up of inclusions the conductivity of which is equal to a unity. Therefore, it can be expected that changing the volume fractions, b, does not alter the conductivity of the material from that of the matrix, which is correct.



Figure 2.7 Contours of the effective conductivity for the imperfect interface case. $f_1 = 0.4$, $f_2 = 0.4$, $k_1 = 101$ and $k_2 = 101$.

Examination of Figs. 2.7 and 2.8 also shows that for any given values of R_i , maximum there is a value of $R_{2-d_{12}}$ at which the system yields a conductivity equal to a unity in the parallel or perpendicular directions. If, for a given R_i and $R_{2-d_{12}}$, the system gives $k_e = 1$ in the parallel direction, it will not yield the same results for the perpendicular direction unless $R_i = k_i - 1$ (i = 1, 2). Furthermore, in some cases, the value of R_i in the parallel direction is so large that even when $R_{2-d_{i2}} = 0$, the enhancing effect is not enough to increase the effective conductivity to the point at which the effect of the inclusions can be neglected. Inversely, in some cases, even when $R_{2-d_{i2}}$ tends to infinity in the perpendicular direction, this does not reduce the conductivity of the system to that of the matrix.



Figure 2.8 Contours of the effective conductivity for the imperfect interface case. $f_1 = 0.4$, $f_2 = 0.2$, $k_1 = 101$ and $k_2 = 101$.

While the effect of both types of inclusion can be exactly neglected when $R_i = k_i - 1$ (*i*=1,2), in general, the effect of cylinders of type *i* cannot be neglected when only $R_i = k_i - 1$. In other words, systems with $k_i = 1$, $R_i = 0$ and k_i , $R_i = k_i - 1$ are not equivalent if $R_{2-d_{12}} \neq k_{2-d_{12}} - 1$. Nevertheless, in this condition we find that $g_1^i = 0$ and $B_1^i = 0$ but the terms B_{2n-1}^i (n > 1) are not zero and are present in the procedure of the calculation of $B_1^{2-d_{12}}$, as is evident from Eq. (2.53). For a perfect contact case, all the terms B_{2n-1}^i (n > 1) are zero and do not affect the value of $B_1^{2-d_{12}}$. The amount of discrepancy can only be numerically determined and depends on geometric considerations, resistance and the conductivity of the inclusions. A wide range of cases was tested in which $R_{2-d_{12}}$ increased from 0 to ∞ , conductivities from 1 to 10000 and volume fractions from 0 to 0.45. The two-phase system can be simulated causing the conductivity of one of the phases to approach a unity. Figure 2.9 shows a sample of the calculations.



Figure 2.9 The effective conductivity of the two- and three-phase systems. For the three-phase system, $k_1 = 101$, $k_2 = 101$, $R_1 = 10$ and $R_2 = k_2 - 1 = 100$. The two-phase system consists of a matrix and cylinders of type one with the same properties as those given in the three-phase case.

As can been seen, the results underestimate the conductivity of the two-phase system; however, for low volume fractions, the phase for which $R_i = k_i - 1$ can be easily replaced by the material of the matrix. This is also evident from relation (2.67), as it can be seen that when $g_1^i = 0$, $k_e = 1 - 2f_{2-d_{12}}/(1 + c_1g_1^{2-d_{12}})$ is obtained. When f_i is increased, the error increases since the terms of higher orders play an important role in the response of the system; however, for all the cases studied, the relative error remains less than five percent in the parallel and seven percent in the perpendicular direction. Based on the results for cases $b > \sqrt{3}$, smaller errors might be expected since the maximum values of f_1 and f_2 decrease.

Since the conductivity of the system is a continuous function of the parameters under study, for $R_i < k_i - 1$ a situation can be found in which a three-phase system gives exactly the same conductivity as a two-phase system. This is always possible, since inclusions with conductivity greater than that of the matrix, boost the conductivity of the system. Figure 2.10 shows that a discrepancy from $R_1 = 1 - k_1$ causes a certain amount of error if the cylinders of the second type, for which $R_2 = 1 - k_2$, are replaced with the material of the matrix; therefore the value of R_2 has been reduced to obtain the conductivity of a two-phase system.



Figure 2.10 Error encountered by replacing cylinders of type two, for which $R_2 = k_2 - 1$, with the material of the matrix and the calculated value for the resistance of the cylinders, in which the system gives exactly the conductivity of the two-phase system. $f_1 = 0.45$, $f_2 = 0.45$, $k_1 = 1001$, $k_2 = 101$.

3. COMPOSITE MATERIALS WITH PERIODIC ARRAYS OF ELLIPTICAL CYLINDERS

The studies pertaining to the determination of the effective transport properties of composites made up of elliptical cylinders are fewer in number and more recent than those for composites made up of circular cylinders. Zimmerman [1996] has studied the case of elliptical cylinders in the dilute limit using both the Maxwell and differential method and derived an expression for the effective conductivity of a material that has a very low volume fraction but a random orientation of elliptical cylinders. The case of periodic arrays has been considered by Lu [1994] who has applied the collocation scheme. The case of periodic arrangement has also been differently investigated by Nicorovici and McPhedran [1996] using Rayleigh's method and in the context of dielectric permittivity. These authors had first formulated the lattice sums in polar coordinates which showed to be problematic and, therefore, later revised the formulation [Yardley et al., 1999] by calculating the lattice sums in elliptical cylinder coordinates. In this section Rayleigh's method is developed to cover multi-coated elliptical cylinders (see also Moosavi and Sarkomaa [2003c]). It is tested whether the behaviour observed in circular cylinders can also be applied here. Clearly, the formulations can be used for any inspection related to the transport properties of composites with periodic arrays of coated elliptical cylinders.

The structure of the composites under study in this section is the same as that explained for the multiply coated circular cylinders in section 2.2. The only difference made here is to replace the circular cylinders with elliptical cylinders shown in Fig. 3.1. As can be seen, the distance between the two foci of the elliptical cylinders is equal to 2r.

The most suitable coordinates for the presentation are elliptic cylindrical coordinates, which implies that

$$x = \mathbf{r} \cosh \mathbf{m} \cos \mathbf{q} \tag{3.1}$$

$$y = \mathbf{r} \sinh \mathbf{m} \sin \mathbf{q} , \qquad (3.2)$$



Figure 3.1 The multicoated elliptical cylinder under study.

where q is measured from the x-axis. As is customary in the analysis, an external field of magnitude E_{ext} along the x-axis is applied in the negative direction. The temperature distribution [Nicorovici et al., 1996] inside the core, coating layers and matrix can be given in the form

$$T_{1} = C_{0} + \sum_{l=1}^{\infty} \left\{ \left[C_{l} \left(\mathbf{r}/2 \right)^{l} \cosh l \mathbf{m} \right] \cos l \mathbf{q} + \left[C_{l}^{\prime} \left(\mathbf{r}/2 \right)^{l} \sinh l \mathbf{m} \right] \sin l \mathbf{q} \right\}$$
(3.3)

$$T_{i} = A_{0}^{i} + \sum_{n=1} \left\{ \left| A_{l}^{i} (\mathbf{r}/2)^{l} \cosh l\mathbf{m} + B_{l}^{i} (2/\mathbf{r})^{l} e^{-l\mathbf{m}} \right| \cos l\mathbf{q} + \left| A_{l}^{\prime i} (\mathbf{r}/2)^{l} \sin l\mathbf{m} + B_{l}^{\prime i} (2/\mathbf{r})^{l} e^{-l\mathbf{m}} \right| \sin l\mathbf{q} \right\}$$

$$(i = 2, \dots, N) \quad (3.4)$$

In the above temperature functions, the terms C'_l, A'_l and B'_l cannot appear since the temperature is symmetric around q = 0. Also, the terms of even degree are excluded since the temperature is anti-symmetric around q = p/2; thus, l = 2n - 1 $(n = 1, \dots, \infty)$. Therefore, we have

$$T_{1} = C_{0} + \sum_{n=1}^{\infty} \left[C_{2n-1}(\mathbf{r}/2)^{2n-1} \cosh(2n-1)\mathbf{m} \right] \cos(2n-1)\mathbf{q}$$
(3.5)
$$T_{i} = A_{0}^{i} + \sum_{n=1}^{\infty} \left[A_{2n-1}^{i}(\mathbf{r}/2)^{2n-1} \cosh(2n-1)\mathbf{m} + B_{2n-1}^{i}(2/\mathbf{r})^{2n-1} e^{-(2n-1)\mathbf{m}} \right] \cos(2n-1)\mathbf{q}$$
$$(i = 2, \dots, N)$$
(3.6)

At the surface of the layers the following boundary conditions can be written:

$$T_i = T_{i+1}$$
 and $k_i \partial T_i / \partial \boldsymbol{m} = k_{i+1} \partial T_{i+1} / \partial \boldsymbol{m}$ $\boldsymbol{m} = \boldsymbol{m}_i \quad (i = 1, \dots, N-1)$ (3.7)

In this manner, A_{2n-1}^{i} , B_{2n-1}^{i} and C_{2n-1}^{i} can be related to each other. If Rayleigh's method is performed, as explained in the previous chapter (see also Nicorovici et al. [1996]), the governing equations for the system can be given by the following equation:

$$\boldsymbol{x}_{2n-1}^{N}B_{2n-1}^{N} + \sum_{m=1}^{\infty} S_{2n-1,m}B_{m}^{N} = E_{ext}\boldsymbol{d}_{n1}, \qquad (3.8)$$

where \mathbf{x}_{l}^{N} (l = 2n - 1) can be derived by imposing the boundary conditions (3.7) as follows:

$$\mathbf{x}_{l}^{i} = \frac{e^{-l\mathbf{m}_{i-1}}(k_{i} - k_{i-1}) + 2\mathbf{x}_{l}^{i-1}(\mathbf{r}/2)^{2n}[k_{i}\cosh(l\mathbf{m}_{i-1}) + k_{i-1}\sinh(l\mathbf{m}_{i-1})]}{2e^{-l\mathbf{m}_{i-1}}[k_{i}\sinh(l\mathbf{m}_{i-1}) + k_{i-1}\cosh(l\mathbf{m}_{i-1})] + 2\mathbf{x}_{l}^{i-1}(\mathbf{r}/2)^{2n}\sinh(2l\mathbf{m}_{i-1})(k_{i} - k_{i-1})} \times \frac{1}{(\mathbf{r}/2)^{2l}}e^{l\mathbf{m}_{i-1}}}$$

 S_{lm} are elliptic lattice sums [Yardley et al., 1999] defined as:

$$S_{lm} = \frac{1}{\hbar_m} (2/\mathbf{r})^{l+m} \sum_{j \neq 0} \mathbf{b}_{lm} [(\mathbf{a}_j + i\mathbf{h}_j)/\mathbf{r}]$$
(3.10)

(3.9)

where \mathbf{a}_{j} and \mathbf{h}_{j} are coordinates of the center of the *j*th cylinder measured from the origin. \hbar_{m} represents the Neumann symbol (1 for m = 0, 2 otherwise) and $\mathbf{b}_{lm}(\mathbf{z})$ is:

$$\boldsymbol{b}_{lm}(\boldsymbol{z}) = (-1)^{l} \hbar_{m} \sum_{h=0}^{\infty} \frac{(2h+l+m-1)}{j!(h+m)!(l-1)!} \times exp[-(m+2h+l)\cosh^{-1}\boldsymbol{z}] \times {}_{2}F_{1}[2h+m+l, 2h+m; l+1; exp(-2\cosh^{-1}\boldsymbol{z})]$$
(3.11)

Here, $_{2}F_{1}$ represents Gauss's hypergeometric function and is given by

$${}_{2}F_{1}(\ell, \boldsymbol{l}; \boldsymbol{t}; \boldsymbol{u}) = 1 + \frac{\ell \boldsymbol{l}}{1!\boldsymbol{t}}\boldsymbol{u} + \frac{\ell(\ell+1)\boldsymbol{l}(\boldsymbol{l}+1)}{2!\boldsymbol{t}(\boldsymbol{t}+1)}\boldsymbol{u}^{2} + \dots = \sum_{n=0}^{\infty} \frac{(\ell)_{n}(\boldsymbol{l})_{n}}{(\boldsymbol{t})_{n}} \frac{\boldsymbol{u}^{n}}{n!}$$
(3.12)

Applying Green's theorem will give the effective conductivity of the system as the following:

$$k_e = 1 - \frac{2\mathbf{p}B_1^N}{bE_{ext}} \tag{3.13}$$

Let us test the correctness of the extension to a multiply coated case. Since as compared to the non-coated case [Yardley et al., 1999], only \mathbf{x}_l^i has been changed, only this term is inspected here. One such inspection is to see whether (3.9) can be reduced to the corresponding relation of circular cylindrical inclusions in the limit, i.e. $\mathbf{r} \to 0$ and $\mathbf{m} \to \infty$. Using $\cosh x = (e^x + e^{-x})/2$ and $\sinh x = (e^x - e^{-x})/2$, we get

$$\mathbf{x}_{l}^{i} = \frac{(k_{i} - k_{i-1}) + \mathbf{x}_{l}^{i-1}(\mathbf{r}/2)^{2l} [k_{i}(e^{2l\mathbf{m}_{i-1}} + 1) + k_{i-1}(e^{2l\mathbf{m}_{i-1}} - 1)]}{k_{i}(1 - e^{-2l\mathbf{m}_{i-1}}) + k_{i-1}(1 + e^{-2l\mathbf{m}_{i-1}}) + \mathbf{x}_{l}^{i-1}(\mathbf{r}/2)^{2l}(e^{2l\mathbf{m}_{i-1}} - e^{-2l\mathbf{m}_{i-1}})(k_{i} - k_{i-1})} \times \frac{1}{(\mathbf{r}/2)^{2l}e^{2l\mathbf{m}_{i-1}}}$$
(3.14)

At the limit, we have $\lim_{\substack{r \to 0 \\ m_i \to \infty}} r e^{\frac{m_i}{r}} / 2 = a_i$ [Meixner and Schäfke, 1954]; therefore, the following relation can be obtained:

$$\mathbf{x}_{l}^{i} = \frac{k_{i} - k_{i-1} + (k_{i} + k_{i-1}) \mathbf{x}_{l}^{i-1} a_{i-1}^{2l}}{k_{i} + k_{i-1} + (k_{i} - k_{i-1}) \mathbf{x}_{l}^{i-1} a_{i-1}^{2l}} \times \frac{1}{a_{i-1}^{2l}},$$
(3.15)

which is exactly the relation derived for the case of multiply coated circular cylinders. Also, for the cases in which the coating layer i-1 is perfectly insulating or conducting, we derive

$$\mathbf{x}_{l}^{i} = \frac{1}{(\mathbf{r}/2)^{2l} \left(e^{2l\mathbf{m}_{l-1}} - 1\right)} \qquad (k_{i-1} = 0)$$
(3.16)

$$\mathbf{x}_{l}^{i} = -\frac{1}{(\mathbf{r}/2)^{2l} \left(e^{2l\mathbf{m}_{l-1}} + 1\right)} \qquad (k_{i-1} = \infty)$$
(3.17)

As can be seen, there is no information on the layers below the layer *i*-1, which means that the layer *i*-1 intersects the relation between the layer *i* and the layers below the layer *i*-1. Note that at the limit, (3.16) and (3.17) are reduced to $\mathbf{x}_{l}^{i} = 1/a_{l-1}^{2l}$ and $\mathbf{x}_{l}^{i} = -1/a_{l-1}^{2l}$, respectively. The above tests give the measures of the correctness of the extension in the *x*-direction. Similar to the discussion outlined in chapter 2, an extension in the perpendicular direction can also be readily performed.

Next, let us see whether the behaviour explained for multiply coated circular cylinders in the case $\mathbf{e}_i + \mathbf{e}_{i-1} = 0$ can also be applied here. This can be done again by inspecting \mathbf{x}_i^i . By comparing (3.14) and (3.15), it is found that the inverse of (3.15), $\mathbf{e}_i + \mathbf{e}_{i-1}$, is not explicit in (3.14) and, thereby, considering that $\mathbf{e}_i = -\mathbf{e}_{i-1}$ will not lead to any specific result. Therefore, the above-mentioned behaviour for circular cylinders cannot be applied to an arbitrary coated cylindrical system. Figure 3.1(*a*, *b*) shows that for this case, changing the dielectric constant of the coating layer from $\mathbf{e}_2 = \mathbf{e}_1$ to $\mathbf{e}_2 = -\mathbf{e}_1$ alters the field through the matrix.



Figure 3.1 The equipotential contours inside the unit cell of a system composed of singly coated elliptical cylinders. A potential gradient of unit magnitude was applied externally along the x-axis and the other external boundaries were insulated. (a) $\mathbf{e}_2 = \mathbf{e}_1$ (b) $\mathbf{e}_2 = -\mathbf{e}_1$.

4. KELLER'S THEOREM

The Keller reciprocal relation [Keller, 1964] has shown its usefulness in many studies and is worth being examined here. Keller showed that for a composite material consisting of a matrix and cylinder-shaped inclusions arranged in a regular array, the following relation is obtained:

$$k_{e}(k_{1},k_{2}) \times k_{e}'(k_{2},k_{1}) = k_{1}k_{2}, \qquad (4.1)$$

where k_e and k'_e represent the conductivity of the system in the symmetry axes x and y, respectively. Keller further claimed that the above-mentioned relation will work for composites made up of randomly arranged cylindrical inclusions. Mendelson [1974] later showed that this is indeed valid, considering that x and y are now the principal axes of the system. If the medium is statistically isotropic, relation (4.1) can be rewritten as

$$k_{e}(k_{1},k_{2}) \times k_{e}(k_{2},k_{1}) = k_{1}k_{2}$$
(4.2)

Furthermore, if the phases are statistically equivalent (interchangeable), we obtain

$$k_e(k_1, k_2) = k_e(k_2, k_1) = \sqrt{k_1 k_2}$$
(4.3)

The above relation is of interest since it is geometry-independent and gives in a very simple form the exact effective conductivity of systems for which deriving the effective transport properties requires a considerable mathematical effort (for example, the square checkerboard). More recently, Lu [1999] has considered composites with regular arrays of multicoated cylinders and derived the following:

$$k_{e}(k_{1},\ldots,k_{N}) \times k_{e}'(1/k_{1},\ldots,1/k_{N}) = 1$$
(4.4)

For multiphase non-coated composites, Fel et al. [2000] have also obtained the same relation. Here, extending Mendelson's methodology, the elegant strategy behind the relations will be shown. The technique is simple and uses the fact that any two-dimensional divergence-free field, when rotated locally at each point by 90°, produces a curl-free field and vice versa [Milton, 1988]. Composites made up of coated cylindrical inclusions are considered here, although with slight changes the discussion can be extended to multiphase composites made up of non-coated inclusions.

Consider a composite material consisting of a matrix and a series of cylinder-shaped inclusions that are placed in a random arrangement inside the matrix. The inclusions are uniform and multicoated with the property of the dispersed layers and the matrix being equal to k_1, \ldots, k_{N-1} and k_N (we can consider $k_N = 1$), respectively. Suppose that the system is statistically homogeneous, x and y are the principal axes and the potential functions, T_1^{j}, \ldots, T_N^{j} , are the solutions of the Laplace equation $(\nabla^2 T = 0)$ in the system. Here, j refers to *j*th cylinder. Therefore, on the surface between the layers, we have $\partial T_i^j / \partial \ell = \partial T_{i+1}^j / \partial \ell$ $(T_i^j = T_{i+1}^j)$, where ℓ is the distance measured along the interfaces and $k_i \partial T_i^j / \partial n = k_{i+1} \partial T_{i+1} / \partial n$, where *n* is the normal outward direction to the interfaces. If a series of functions $\mathbf{y}_i^j (i = 1, ..., N)$ is defined, in which $\nabla \times (\mathbf{y}_i^j \mathbf{k}) = -\mathbf{k} \times \nabla \mathbf{y}_i^j = k_i \nabla T_i^j$, we have $\partial \mathbf{y}_i^j / \partial y = k_i \partial T_i^j / \partial x$ and $\partial \mathbf{y}_i^j / \partial x = -k_i \partial T_i^j / \partial y$; therefore, \mathbf{y}_i^j satisfies the Laplace equation in the layers *i* of cylinder *j*. The first boundary condition for T_i^{j} will lead to finding $(1/k_i)\partial \mathbf{y}_i^j/\partial n = (1/k_{i+1})\partial \mathbf{y}_{i+1}^j/\partial n$ and the second one will give $\partial \mathbf{y}_i^j/\partial \ell = \partial \mathbf{y}_i^j/\partial \ell$ $(\mathbf{y}_i^j = \mathbf{y}_{i+1}^j)$. As a result we have a function, \mathbf{y} , which is the solution of the Laplace equation in the same composite but with the properties of the phases equal to $1/k_1, \ldots, 1/k_N$. Based on definition of the effective conductivity, we obtain

$$k_{e}(k_{1},...,k_{N}) \times k_{e}'(1/k_{1},...,1/k_{N}) = \frac{\int_{V}^{k} \nabla T dv}{\int_{V}^{V} \nabla T dv} \times \frac{\int_{V}^{k} \nabla \mathbf{y} dv}{\int_{V}^{V} \nabla T dv}$$
(4.5)

Here, *V* is the volume containing a sufficient number of inclusions. The analysis is started by deriving $\int_{V} \nabla T dv$ and $\int_{V} k \nabla T dv$. Getting help from Green's first identity [Spiegel, 1968] it is found that

$$\int_{V} \nabla T dv = \oint_{S} T ds = \oint_{S} T ds = \mathbf{i} \oint_{S} T n_{x} ds + \mathbf{j} \oint_{S} T n_{y} ds = \mathbf{i} \oint_{S} T n_{x} ds$$
(4.6)

where s is the boundary surface of the volume V. Here $\oint_s Tn_y ds$ has been ignored because of the selection of the principal axes. Furthermore, we have [Spiegel, 1968]:

$$\int_{V} \nabla \times (\mathbf{y}\mathbf{k}) dv = \oint_{s} ds \times (\mathbf{y}\mathbf{k})$$
(4.7)

Using this property, we obtain

$$\int_{V} k \nabla T dv = \int_{V} \nabla \times (\mathbf{y} \mathbf{k}) dv = \mathbf{i} \oint_{S} \mathbf{y} n_{y} ds - \mathbf{j} \oint_{S} \mathbf{y} n_{x} ds = \mathbf{i} \oint_{S} \mathbf{y} n_{y} ds$$
(4.8)

Here again, the selection of the principal axes has implied that $\oint_{s} yn_{x} ds = 0$.

Now, $\int_{V} \nabla \mathbf{y} dv$ and $\int_{V} k \nabla \mathbf{y} dv$ are studied. Using Green's first identity and applying $\oint_{S} \mathbf{y} n_{x} ds = 0$, we obtain $\int_{V} \nabla \mathbf{y} dV = \mathbf{i} \oint_{S} \mathbf{y} n_{x} ds + \mathbf{j} \oint_{S} \mathbf{y} n_{y} ds = \mathbf{j} \oint_{S} \mathbf{y} n_{y} ds$ (4.9)

Since $\nabla \mathbf{y}_i^j = -\mathbf{k} \times (k_i \nabla T_i^j)$ and $\oint_s Tn_y ds = 0$, also the following can be obtained:

$$\int_{V} k \nabla \mathbf{y} dv = \int_{V} \mathbf{k} \times \nabla T dv = -\int_{V} \nabla \times (T \mathbf{k}) dv = -\mathbf{i} \oint_{s} T n_{y} ds + \mathbf{j} \oint_{s} T n_{x} ds = \mathbf{j} \oint_{s} T n_{x} ds \qquad (4.10)$$

Here, the property given in (4.7) has been used. Substituting (4.6), (4.8), (4.9) and (4.10) in (4.5), we can derive

$$k_{e}(k_{1},\ldots,k_{N}) \times k_{e}'(1/k_{1},\ldots,1/k_{N}) = 1$$
(4.11)

5. COMPOSITE MATERIALS WITH PERIODIC ARRAYS OF SPHERES

The first composites studied by researchers to derive the effective transport properties were composites with spherical inclusions, as is Maxwell's case studies [1873] (see Moosavi et al. [2002b] for a review). In this chapter, these composites are studied in cases where the inclusions are placed in a periodic arrangement. The terminology used for the presentation is the same as that given by Cheng and Torquat [1997] who considered cases in which the unit cell of the system was a cube. In this chapter a more general state will be considered in which the unit cell of the system is a rectangular prism. In specific cases, the discussion can be reduced to those considered by the above authors. Also formulations for the case of multiphase systems (coated and non-coated) will be developed below. It should be noted that the case of composites with periodic spheres in a rectangular order was first investigated by Rayleigh [1892].

5.1 The case of a two-phase composite with uniform and solid spheres

Let us consider a composite material with a unit cell in the shape of a rectangular prism in which a sphere is placed at the centre and possibly at each corner or face of the unit cell. For greater generality, the sides of the unit cell will be considered *b*, *c* and a unity in the *x*-, *y*- and *z*-directions of the cartesian coordinates which has been placed at the centre of a sphere situated at the centre of the unit cell. Also, the continuous medium is supposed to have a unit conductivity. The radius of the spheres is denoted by *a* and the conductivity by *k*. Assuming that b=c=1, the unit cell of the system would be a cube, and simple cubic, body-centred and face-centred structures can be constructed.

As is clear, in general, the system is anisotropic; therefore, three different conductivities may be expected for the system in the x-, y- and z-directions. Since deriving the conductivity of the system in these directions is similar, the conductivity of the system is only derived in one direction, namely the x-direction. To perform this, a uniform field of magnitude E_{ext} is applied externally along the x-axis of the system in the negative direction. If a spherical coordinate (r,q,f) is set at the origin, in which q is measured from the x-axis and f from the xy plane, the temperature inside the sphere and the matrix in terms of spherical harmonics can be given by

$$T_{1} = \sum_{l=0}^{\infty} \sum_{m=-l}^{l} C_{lm} r^{l} Y_{lm}(\boldsymbol{q}, \boldsymbol{f})$$
(5.1)

$$T_{2} = \sum_{l=0}^{\infty} \sum_{m=-l}^{l} \left(A_{lm} r^{l} + B_{lm} r^{-l-1} \right) Y_{lm} (\boldsymbol{q}, \boldsymbol{f}), \qquad (5.2)$$

where C_{nm} , A_{nm} and B_{nm} are unknowns to be determined and $Y_{lm}(q, f)$ is a spherical harmonics of the order (l, m) defined as:

$$Y_{lm}(\boldsymbol{q},\boldsymbol{f}) = \sqrt{\frac{(l-|m|)!}{(l+|m|)!}} P_l^{|m|}(\cos \boldsymbol{q}) e^{im\boldsymbol{f}}$$
(5.3)

On the surface of the spheres, we can write down the following continuity relations can be written:

$$T_1 = T_2, \quad k \,\partial T_1 / \partial r = \partial T_2 / \partial r \qquad r = a$$
, (5.4)

which lead to

$$A_{lm} = \frac{B_{lm}}{\boldsymbol{g}_l a^{2l+1}} = \boldsymbol{x}_l B_{lm}$$
(5.5)

$$C_{lm} = \frac{B_{lm}}{c_l a^{2l+1}},$$
(5.6)

where

$$\boldsymbol{g}_{l} = \frac{1-k}{k+(l+1)/l}$$
(5.7)

$$c_{l} = \frac{1-k}{(2l+1)/l}$$
(5.8)

The temperature around the origin, when the origin is not included, can be considered due to terms originating at infinity and at the other lattice sites (see section 2.1); therefore we can write

$$\sum_{l=0}^{\infty} \sum_{m=-l}^{l} A_{lm} r^{l} Y_{lm}(\boldsymbol{q}, \boldsymbol{f}) = E_{ext} x + \sum_{i=0}^{\infty} \sum_{j=-i}^{i} \sum_{h\neq 0}^{\infty} B_{ij} \frac{Y_{ij}(\boldsymbol{q}_{h}, \boldsymbol{f}_{h})}{r_{h}^{i+1}}$$
(5.9)

where (r_h, q_h, f_h) is the spherical coordinate of a point measured from the lattice point *h*. Using Eq. (5.5) and considering that $x = r\cos q = Y_{10}(q, f)$, the above equation can be written in the following form:

$$\sum_{l=0}^{\infty} \sum_{m=-l}^{l} \frac{B_{lm}}{\boldsymbol{g}_{l} a^{2l+1}} r^{l} Y_{lm}(\boldsymbol{q}, \boldsymbol{f}) = \sum_{l=0}^{\infty} \sum_{m=-l}^{l} E_{ext} \boldsymbol{d}_{l1} \boldsymbol{d}_{m0} r^{l} Y_{lm}(\boldsymbol{q}, \boldsymbol{f}) + \sum_{i=0}^{\infty} \sum_{j=-i}^{i} \sum_{h\neq 0}^{\infty} B_{ij} \frac{Y_{ij}(\boldsymbol{q}_{h}, \boldsymbol{f}_{h})}{r_{h}^{i+1}}$$
(5.10)

As can be seen, the left-hand side and the first part of the right-hand side are given on the basis of the coordinate situated at the origin, but the second part on the right-hand side has been expressed on the basis of the coordinates placed at the centre of the other spheres. To proceed, all the terms have to presented on the basis of the coordinates situated at the origin. For this purpose, the general form of a theorem on spherical harmonics can be used, well known as the addition theorem [McKenzie et al, 1978; Greengard, 1988]. On the basis of this theorem, we have

$$\frac{Y_{ij}(\boldsymbol{q}_{h},\boldsymbol{f}_{h})}{r_{h}^{i+1}} = \sum_{l=0}^{\infty} \sum_{m=-l}^{l} (-1)^{i} J_{m}^{j} \boldsymbol{h}_{l+m}^{i-j} \boldsymbol{h}_{l-m}^{i+j} r^{l} \frac{Y_{l+i,j-m}(\boldsymbol{q}_{h0},\boldsymbol{f}_{h0})}{r_{h0}^{l+i+1}} Y_{lm}(\boldsymbol{q},\boldsymbol{f})$$
(5.11)

where

$$J_{m}^{j} = \begin{cases} \left(-1\right)^{\min\left(|m|,|j|\right)} & \text{if } m \cdot j > 0\\ 1 & \text{otherwise} \end{cases}, \qquad \boldsymbol{h}_{l}^{i} = \sqrt{\binom{l+i}{l}} \end{cases}$$
(5.12)

Here, (r_{h0}, q_{h0}, f_{h0}) expresses the centre of the *h*th sphere measured from the origin. By substituting the above relation in (5.10), we can obtain

$$\sum_{l=0}^{\infty} \sum_{m=-l}^{l} \frac{B_{lm}}{g_{l} a^{2l+1}} r^{l} Y_{lm}(\boldsymbol{q}, \boldsymbol{f}) = \sum_{l=0}^{\infty} \sum_{m=-l}^{l} E_{ext} \boldsymbol{d}_{l1} \boldsymbol{d}_{m0} r^{l} Y_{lm}(\boldsymbol{q}, \boldsymbol{f}) + \sum_{l=0}^{\infty} \sum_{m=-l}^{l} \sum_{h\neq 0} \sum_{i=0}^{\infty} \sum_{j=-i}^{i} (-1)^{i} J_{m}^{j} r^{l} \boldsymbol{h}_{l+m}^{i-j} \boldsymbol{h}_{l-m}^{i+j} \frac{Y_{l+i,j-m}(\boldsymbol{q}_{h0}, \boldsymbol{f}_{h0})}{r_{h0}^{l+i+1}} B_{ij} Y_{lm}(\boldsymbol{q}, \boldsymbol{f})$$
(5.13)

If the lattice sums for an array of spherical inclusions are defined as

$$S_{lm} = \sum_{h \neq 0} \frac{Y_{lm}(\boldsymbol{q}_{h0}, \boldsymbol{f}_{h0})}{r_{h0}^{l+1}}$$
(5.14)

Eq. (5.13) can be given as

$$\sum_{l=0}^{\infty} \sum_{m=-l}^{l} \frac{B_{lm}}{g_l a^{2l+1}} r^l Y_{lm}(\boldsymbol{q}, \boldsymbol{f}) = \sum_{l=0}^{\infty} \sum_{m=-l}^{l} E_{ext} \boldsymbol{d}_{l1} \boldsymbol{d}_{m0} r^l Y_{lm}(\boldsymbol{q}, \boldsymbol{f}) + \sum_{l=0}^{\infty} \sum_{m=-l}^{l} \sum_{i=0}^{\infty} \sum_{j=-i}^{l} (-1)^i J_m^{\,j} r^l \boldsymbol{h}_{l+m}^{i-j} \boldsymbol{h}_{l-m}^{i+j} S_{l+i,j+m} B_{ij} Y_{lm}(\boldsymbol{q}, \boldsymbol{f})$$
(5.15)

Since the above equation is valid for arbitrary values of r, q and f and spherical harmonics are linearly independent, the terms of the left- and right-hand sides of the above equation can be equated for a specific value of l and m, i.e.,

$$\frac{B_{lm}}{\boldsymbol{g}_l a^{2l+1}} = E_{ext} \boldsymbol{d}_{l1} \boldsymbol{d}_{m0} + \sum_{i=0}^{\infty} \sum_{j=-i}^{i} (-1)^i J_m^{j} \boldsymbol{h}_{l+m}^{i-j} \boldsymbol{h}_{l-m}^{i+j} S_{l+i,j+m} B_{ij}$$
(5.16)

Now, let us reconsider the structure under study and also the temperature functions given in (5.1) and (5.2). An examination reveals that l(i) has to be an odd number because of the skew symmetry of the temperature around q = p/2. Also, *m* must be considered as an even number because the temperature is symmetric around f = p/2. Furthermore, in the case c=1, m(j) should be considered a multiple of 4; therefore, $(-1)^i = -1$ and $J_m^j = 1$. After performing the necessary changes to bring the summation from negative *j* to positive values, Eq. (5.10) can be rewritten as follows:

$$\frac{B_{2n-1,m}}{g_{2n-1}a^{4n-1}} + \sum_{i=0}^{\infty} \sum_{j=0}^{2^{i-1}} \frac{\hbar_j}{2} \left(\mathbf{h}_{2n-1+m}^{2i-1-j} \mathbf{h}_{2n-1-m}^{2i-1+j} S_{2n+2i-2,|j-m|} + \mathbf{h}_{2n-1+m}^{2i-1+j} \mathbf{h}_{2n-1-m}^{2i-1-j} S_{2n+2i-2,j+m} \right) B_{2i-1,j} = E_{ext} \mathbf{d}_{n1} \mathbf{d}_{m0}$$
(5.17)

Here \hbar_j is the Neumann symbol (1 for j=0, 2 otherwise). This symbol was added because for j=0, the results of the summation is twice the one derived from Eq. (5.16). Since $Y_{l,-m}(\boldsymbol{q},\boldsymbol{f}) = Y_{lm}^*(\boldsymbol{q},\boldsymbol{f})$, where the asterisk stands for a complex conjugation, the sign of *m* in S_{lm} is limited to positive values.

As explained in chapter 2, considering a sufficient number of unknowns will lead to the required result. Based on reports given by McPhedran and Mckenzie [1978] and McKenzie et al. [1978] with 50 zonal unknowns together with azimuthal terms, the results are reasonable for volume fractions up to 0.523 (SC), 0.677 (BCC) and 0.733 (FCC) for any values of the conductivity of the inclusions.

5.1.1 Determining the effective conductivity of the system

In the same terminology used for the solid cylinder, for the average heat flux and temperature gradient we can write

$$\langle \boldsymbol{F} \rangle = M \langle \boldsymbol{S}_P \rangle - \langle \nabla T \rangle$$
 (5.18)

where M is the number of spheres in the unit cell and

$$\left\langle \boldsymbol{S}_{P}\right\rangle =\frac{1-k}{V_{cell}}\int_{V_{1}}\nabla TdV$$
(5.19)

$$\langle \nabla T \rangle = \frac{1}{V_{cell}} \int_{V_{cell}} \nabla T dV ,$$
 (5.20)

where V_1 is the volume of the sphere. Taking into consideration the temperature profiles given in Eqs. (5.1) and (5.2) and using orthogonality properties of spherical harmonics, we obtain

$$\langle S_{P} \rangle = \frac{1-k}{V_{cell}} \int_{s_{1}}^{T} n ds = \frac{1-k}{V_{cell}} \int_{0}^{2p} \int_{0}^{p} a^{2} C_{1,0} \cos^{2} q \sin q \, dq \, df \, \mathbf{i} = \frac{4pa^{2}(1-k)}{3V_{cell}} C_{1,0} \, \mathbf{i} = \frac{4pB_{1,0}}{V_{cell}} \, \mathbf{i}$$
(5.21)

Here, \mathbf{s}_1 represents the surface of the sphere. By substituting (5.21) in (5.18) and considering that $\langle \nabla T \rangle = E_{ext} \mathbf{i}$ and $V_{cell} = bc$, the final result for the effective conductivity can be given as

$$k_{e} = 1 - \frac{4MpB_{1,0}}{bcE_{ext}}$$
(5.22)

5.1.2 Explicit forms for the effective conductivity

Applying the truncation order 4 allows the solution of Eq. (5.17) to be expressed as follows:

$$k_{eff} = 1 - \frac{3f}{D}, \qquad (5.23)$$

where

$$D = 1/\boldsymbol{g}_1 + f - c_1 \boldsymbol{g}_2 f^{10/3} \frac{1 - c_4 \boldsymbol{g}_3 f^{11/3}}{1 + c_2 \boldsymbol{g}_2 f^{7/3}} - c_3 \boldsymbol{g}_3 f^{14/3} - c_5 \boldsymbol{g}_4 f^6 - c_6 \boldsymbol{g}_5 f^{22/3} + O(f^{25/3}) \quad (5.24)$$

The numerical constants, for the case in which the unit cell is a cube, are given in [Sangani and Acrivos, 1983] and are shown below in Table 5.1.

	SC	BCC	FCC
c_1	1.3047	1.29×10^{-1}	7.529×10^{-2}
c_2	4.054×10^{-1}	7.642×10^{-1}	-7.410×10^{-1}
<i>C</i> ₃	7.231×10^{-2}	2.569×10^{-1}	4.195×10^{-2}
C_4	2.305×10^{-1}	-4.129×10^{-1}	6.966×10^{-1}
<i>C</i> ₅	1.526×10^{-1}	1.13×10^{-2}	2.31×10^{-2}
C_6	1.05×10^{-2}	5.62×10^{-3}	9.14×10^{-7}

Table 5.1 The numerical constants in the explicit expression (5.24).

5.2 The case of a periodic array of coated spheres

The present author has recently reported a study performed on composites with cubic arrays of multi-coated spheres [Moosavi et al., 2002b]. For these composites, the changes required to develop the above procedures for two-phase systems in the case of multi-coated systems will be discussed below. The conductivity and radii of the core and coating layers are denoted by k_1, \ldots, k_{N-1} and a_1, \cdots, a_{N-1} , respectively. The temperature functions inside the layers can be given by

$$T_{1} = \sum_{n=0}^{\infty} \sum_{m=0}^{2n-1} C_{2n-1,m} r^{2n-1} Y_{2n-1,m} (\boldsymbol{q}, \boldsymbol{f})$$
(5.25)

$$T_{i} = \sum_{n=0}^{\infty} \sum_{m=0}^{2n-1} \left(A_{2n-1,m}^{i} r^{2n-1} + B_{2n-1,m}^{i} r^{-2n} \right) Y_{2n-1,m} (\boldsymbol{q}, \boldsymbol{f}) \qquad (i = 2, \dots, N)$$
(5.26)

At the surface of the core and coating layers, the boundary conditions

$$T_{i} = T_{i+1}, \quad k_{i} \partial T_{i} / \partial r = k_{i+1} \partial T_{i+1} / \partial r \quad r = a_{i} \quad (i = 1, \dots, N-1)$$
(5.27)

are applied, whence we can be related the coefficients, i.e.,

$$A_{2n-1,m}^{i} = \frac{B_{2n-1,m}^{i}}{\boldsymbol{g}_{2n-1}^{i}a_{i-1}^{4n-1}} = \boldsymbol{x}_{2n-1}^{i}B_{2n-1,m}^{i}, \qquad (5.28)$$

where

$$\mathbf{x}_{2n-1}^{i} = \frac{2n(\mathbf{e}_{i} - \mathbf{e}_{i-1}) + \mathbf{x}_{2n-1}^{i-1}[(2n-1)\mathbf{e}_{i-1} + 2n\mathbf{e}_{i}]a_{i-1}^{4n-1}}{[2n\mathbf{e}_{i-1} + (2n-1)\mathbf{e}_{i}] + \mathbf{x}_{2n-1}^{i-1}(2n-1)(\mathbf{e}_{i} - \mathbf{e}_{i-1})a_{i-1}^{4n-1}} \times \frac{1}{a_{i-1}^{4n-1}}$$
(5.29)

An equation of type (5.9) can be written for this system, i.e.,

$$\sum_{n=0}^{\infty} \sum_{m=0}^{2n-1} A_{2n-1,m}^{N} r^{2n-1} Y_{2n-1,m}(\boldsymbol{q},\boldsymbol{f}) = E_{ext} x + \sum_{i=0}^{\infty} \sum_{j=0}^{2i-1} \sum_{h\neq 0}^{\infty} B_{2i-1,j}^{N} \frac{Y_{2i-1,j}(\boldsymbol{q}_{h},\boldsymbol{f}_{h})}{r_{h}^{2i}}$$
(5.30)

After performing the other required steps, the final equation is

$$\frac{B_{2n-1,m}^{N}}{g_{2n-1}^{N}a_{N-1}^{4n-1}} + \sum_{i=0}^{\infty}\sum_{j=0}^{2^{i-1}}\frac{\hbar_{j}}{2} \left(\boldsymbol{h}_{2n-1+m}^{2i-1-j}\boldsymbol{h}_{2n-1-m}^{2i-1+j}\boldsymbol{S}_{2n+2i-2,|j-m|} + \boldsymbol{h}_{2n-1+m}^{2i-1+j}\boldsymbol{h}_{2n-1-m}^{2i-1-j}\boldsymbol{S}_{2n+2i-2,j+m} \right) B_{2i-1,j}^{N} = E_{ext}\boldsymbol{d}_{n1}\boldsymbol{d}_{m0}$$
(5.31)

By applying the same method used for the case of solid spheres, the effective conductivity of the system again can again be given by

$$k_e = 1 - \frac{4MB_1^N}{bcE_{ext}} \tag{5.32}$$

5.2.1 Comparison between the behaviour of coated spheres and circular cylinders

In this chapter the focus is shifted back to the general state, assuming that the transport property can be negative or positive. If one layer $(2 \le i \le N-1)$ is perfectly conducting $(\mathbf{e}_i = \infty)$ or non-conducting $(\mathbf{e}_i = 0)$, the layers under this layer will not contribute to the effective property of the system. These results can also be easily seen in Eq. (5.29), where the multipolar polarisability for the cases in question would be -1 and 1, respectively. Furthermore, two spheres with the same property can be unified and considered as being a unit layer, which is obvious and can be confirmed from Eq. (5.29).

In the section on cylindrical inclusions, the strange behaviour of coated cylinders when $\mathbf{e}_i + \mathbf{e}_{i-1} = 0$ was studied. Here, the aim is to find out whether the same behaviour can be observed for the case of multicoated spheres. Nicorovici et al. [1995] found that the case of singly coated spheres did not exhibit the same behaviour as that of coated cylinders. The reason is clear when it is kept in mind that all the behaviour was based on the mathematical expression for multipolar polarisability. Here, \mathbf{g}_{2n-1}^1 is a function of n and because of this, the results are not the same. A careful examination of this case, however, shows that by the first order, the cases $\mathbf{e}_2 = -0.5\mathbf{e}_1$ and $\mathbf{e}_2 = \mathbf{e}_1$ yield the same effective transport property; however, the inspection of (5.29) shows that even this result cannot be extended to the case of multicoated spheres. This makes sense physically, since the successive application of $\mathbf{e}_i = -0.5\mathbf{e}_{i-1}$ to infinitely coated layers causes the property of the outermost coating layer to approach zero, and this case exhibits the behaviour of non-conducting spheres of radius a_N $(N \to \infty)$ and not of spheres that have a property equal to \mathbf{e}_1 with a radius of a_N .

5.3 Three-phase non-coated spheres

In accordance with the three-phase case explained for circular cylinders, here a composite is considered in which the unit cell of the system is a rectangular prism with a sphere of type one at the center and a sphere of type two at each corner. The formulations can be readily developed for other structures. It should be noted that when b=c=1, the structure is CsCl that has been studied by McPhedran [1983]. For this case, the temperature of the phases and the matrix can be expressed as

$$T_{i} = \sum_{n=1}^{\infty} \sum_{m=0}^{2n-1} C_{2n-1,m}^{i} r^{2n-1} Y_{2n-1,m}(r, q)$$
(5.33)

$$T_{3,i} = \sum_{n=1}^{\infty} \sum_{m=0}^{2n-1} \left(A_{2n-1,m}^{i} r^{2n-1} + B_{2n-1,m}^{i} r^{-2n+1} \right) Y_{2n-1,m}(r, q),$$
(5.34)

where *i* can be either 1 or 2 showing the sphere type. As can be seen, the non-symmetry of the temperature around q = p/2 has been taken into account. Because of the symmetry of the

temperature around, also m will be considered to be an even number in general and for the case c=1, a multiple of 4. The boundary conditions are of the following shape:

$$T_i = T_{3,i}, \quad k_i \frac{\partial T_i}{\partial r} = \frac{\partial T_{3,i}}{\partial r} \quad \text{on} \quad r = a_i$$
 (5.35)

Therefore, for the coefficient of the temperature functions, we obtain

$$A_{2n-1}^{i} = \frac{B_{2n-1}^{i}}{\boldsymbol{g}_{2n-1}^{i} a_{i}^{4n-2}}$$
(5.36)

$$C_{2n-1}^{i} = \frac{B_{2n-1}^{i}}{\boldsymbol{c}_{2n-1}^{i} a_{i}^{4n-2}},$$
(5.37)

where

$$\boldsymbol{g}_{2n-1}^{i} = \frac{1 - k_{i}}{k_{i} + 2n/(2n-1)}$$
(5.38)

$$\boldsymbol{c}_{2n-1}^{i} = \frac{1 - k_{i}}{(4n - 1)/(2n - 1)}$$
(5.39)

By separating the effect of the inclusions of type one and two, we get

$$\sum_{n=0}^{\infty} \sum_{m=0}^{2n-1} A_{2n-1,m}^{i} r^{2n-1} Y_{2n-1,m}(\boldsymbol{q}, \boldsymbol{f}) = E_{0} r \cos \boldsymbol{q} + \sum_{n=0}^{\infty} \sum_{m=0}^{2n-1} \sum_{h\neq 0}^{\infty} B_{2n-1,m}^{i} r_{h}^{-2n} Y_{2n-1,m}(\boldsymbol{q}_{h}, \boldsymbol{f}_{h}) + \sum_{n=0}^{\infty} \sum_{m=0}^{2n-1} \sum_{h}^{\infty} B_{2n-1,m}^{i} r_{h}^{-2n} Y_{2n-1,m}(\boldsymbol{q}_{h}, \boldsymbol{f}_{h})$$
(5.40)

By following the same methodology as given for solid spheres we obtain the following linear system is obtained:

$$\frac{B_{2n-1,m}^{i}}{g_{2n-1}^{i}a_{i}^{4n-1}} + \sum_{l=0}^{\infty}\sum_{j=0}^{2l-1}\frac{\hbar_{j}}{2} \left[h_{2n-1+m}^{2l-1-j}h_{2n-1-m}^{2l-1+j} \left(S_{2n-2l-2,|j-m|}^{1}B_{2l-1,j}^{i} + S_{2n-2l-2,|j-m|}^{2}B_{2l-1,j}^{2} \right) + h_{2n-1+m}^{2l-1+j}h_{2n-1-m}^{2l-1-j} \left(S_{2n+2l-2,j+m}^{1}B_{2l-1,j}^{i} + S_{2n+2l-2,j+m}^{2}B_{2l-1,j}^{2-d_{i2}} \right) \right] = E_{ext}d_{n1}d_{m0}$$
(5.41)

where S_{nm}^{i} are the lattice sums over the spheres of type *i*. For a three-phase case, the volume averaged flux and temperature are related to each other by

$$\langle \boldsymbol{F} \rangle = \langle \boldsymbol{S}_{P} \rangle_{1} + \langle \boldsymbol{S}_{P} \rangle_{2} - \langle \nabla T \rangle$$
 (5.42)

$$\left\langle \boldsymbol{S}_{P}\right\rangle_{i} = \frac{1-k_{i}}{V_{cell}} \int_{V_{i}} \nabla T_{i} dV$$
(5.43)

After performing the integrals in the same manner as that explained for uniform spheres, we obtain

$$k_{e} = 1 - \frac{2p(B_{1}^{1} + B_{1}^{2})}{bcE_{ext}}$$
(5.44)

For the case of circular cylindrical inclusions, it was shown that the composite can be perfectly conducting in one direction and non-conducting in the other. However, for the case of spherical inclusions, this behavior does not hold true and these composites cannot be non-conducting since heat can pass through the gaps between the spheres. Furthermore, the Keller theorem is not valid here, and thus, for the case of mono-sized spherical inclusions, when $k_1 = 1/k_2$, the effective conductivity of the system is not equal to that of the matrix.

5.3.1 The case of an imperfect interface

The boundary conditions, including the effect of interfacial resistance, are of the following shape:

$$\frac{k_i}{R_i a_i} (T_i - T_{3,i}) = -k_i \frac{\partial T_i}{\partial r} = -\frac{\partial T_{3,i}}{\partial r} \quad \text{on } r = a_i$$
(5.45)

with

$$A_{2n-1}^{i} = \frac{B_{2n-1}^{i}}{g_{2n-1}^{i}a_{i}^{4n-2}}$$
(5.46)

$$C_{2n-1}^{i} = \frac{B_{2n-1}^{i}}{\boldsymbol{c}_{2n-1}^{i} a_{i}^{4n-2}},$$
(5.47)

where

$$\boldsymbol{g}_{2n-1}^{i} = \frac{1 - k_{i} + R_{i}(2n-1)}{2n/(2n-1) + k_{i} + 2nR_{i}} \quad \boldsymbol{c}_{2n-1}^{i} = \frac{1 - k_{i} + R_{i}(2n-1)}{(4n-1)/(2n-1)}$$
(5.48)

For this case, the relation between the volume averaged flux and temperature would be

$$\left\langle \boldsymbol{F} \right\rangle = \left\langle \boldsymbol{S}_{P} \right\rangle_{1} + \left\langle \boldsymbol{S}_{P} \right\rangle_{2} + \left\langle \boldsymbol{S}_{I} \right\rangle_{1} + \left\langle \boldsymbol{S}_{I} \right\rangle_{2} - \left\langle \nabla T \right\rangle, \tag{5.49}$$

where

$$\left\langle \boldsymbol{S}_{P}\right\rangle_{i} = \frac{1-k_{i}}{V_{cell}} \int_{V_{i}} \nabla T_{i} dV$$
(5.50)

$$\langle S_I \rangle_i = \frac{1}{V_{cell}} \int_{S_i} (T_{3i} - T_i) n \, dS \,,$$
 (5.51)

from which we obtain

$$\left\langle \boldsymbol{S}_{P}\right\rangle_{i} = \frac{2\boldsymbol{p}B_{1}^{i}}{V_{cell}} \cdot \frac{1-k_{i}}{R_{i}+1-k_{i}} \mathbf{i}$$
(5.52)

$$\left\langle \boldsymbol{S}_{I}\right\rangle_{i} = \frac{2\boldsymbol{p}\boldsymbol{B}_{1}^{i}}{V_{cell}} \cdot \frac{\boldsymbol{R}_{i}}{\boldsymbol{R}_{i}+1-\boldsymbol{k}_{i}}\mathbf{i}$$
(5.53)

By substituting the resultants in Eq. (5.48), we obtain a relation that resembles relation (5.44).

As is clear from (5.48), the critical resistance in the case of composites with mono-sized uniform spherical inclusions can be derived using the same formula as the cylindrical inclusions, i.e, R = k - 1. Furthermore, because of the mathematical similarity between (5.41) and (2.54), similar results can be expected as in the case of three-phase composites made up of cylindrical inclusions. Therefore, the effect of spherical inclusions of type *i* in any direction of the calculation of the effective conductivity can be neglected by adjusting the interfacial resistance in the range $R_i \le k_i - 1$.

6. LATTICE SUMS

The problem of calculating lattice sums arises when studying a variety of phenomena in various branches of science including materials science. These sums have the following form for fiber composites:

$$S_n = \sum_{\mathbf{w} \in \Lambda_2} \frac{1}{\mathbf{w}^n}, \qquad (6.1)$$

where $\Lambda_2 = \{ l_1 b + i l_2 | l_1, l_2 \in Z, l_1 b + i l_2 \neq 0 \}$. In three-dimensions, we also have

$$S_{nm} = \sum_{j \in \Lambda_3} \frac{Y_{nm} (\boldsymbol{q}_j, \boldsymbol{f}_j)}{r_j^{n+1}} \quad , \tag{6.2}$$

where $\Lambda_3 = \{(l_1b, l_2c, l_3) | l_i \in Z, (l_1, l_2, l_3) \neq (0, 0, 0)\}$. Here, (r_j, q_j, f_j) expresses the position of a point in spherical coordinates. In the thesis the main interest is in calculating S_n and S_{nm} for $n \ge 2$ and $m \ge 0$. For this purpose, the direct summation method can be used, although the results will converge very slowly. For this reason, many algorithms have been developed for the rapid and highly accurate calculation of lattice sums [Greengard and Rokhlin, 1997; Helsing, 1994; Movchan et al., 1997 and Huang, 1999] some of these algorithms will be explained in this section.

6.1 Integral representation of the lattice sums

Huang [1999] has shown that the problem of calculating lattice sums can be reduced to the one of calculating integrals. The mathematical basis for this method is the plane-wave representation of each pole and analytically summing up the resulting geometric series. Here, the method for a rectangular array of cylinders with periodicity equal to *b* and a unity in the *x*-and *y*-directions, respectively, is explained. For an arbitrary point in the complex plane (z = x + iy), depending on the region in which the point is situated, we can write
$$\frac{1}{z^n} = \frac{1}{(n-1)!} \int_0^\infty I^{n-1} e^{-Iz} dI \qquad (x>0)$$
(6.3)

$$\frac{1}{z^n} = \frac{1}{(n-1)!} \int_0^\infty (-1)^n \mathbf{I}^{n-1} e^{\mathbf{I}z} d\mathbf{I} \qquad (x < 0)$$
(6.4)

$$\frac{1}{z^{n}} = \frac{1}{(n-1)!} \int_{0}^{\infty} (-i)^{n} \mathbf{l}^{n-1} e^{i\mathbf{l}z} d\mathbf{l} \qquad (y > 0)$$
(6.5)

$$\frac{1}{z^{n}} = \frac{1}{(n-1)!} \int_{0}^{\infty} \mathbf{l}^{n-1} e^{-ilz} d\mathbf{l} \qquad (y < 0),$$
(6.6)

where $n = 1, ..., \infty$. The reason for the case n=1 is clear, as a simple analytical integration of the right-hand side yields the left-hand expression. For the other values of n, it is possible to differentiate both sides of the case n=1 with respect to z, n times, in order to obtain the above relations. To calculate the lattice sums given in Eq. (6.1), the lattice is divided into four regions, as depicted in Fig. (6.1).



Figure 6.1 The methodology used for calculating the lattice sums

As can be seen for regions 1 to 4, we have x>0, y>0, x<0 and y<0, respectively; therefore, the lattice sums in the regions can be derived in the following manner:

$$S_{n}(1) = \frac{1}{(n-1)!} \int_{0}^{\infty} I^{n-1} \sum_{k=1}^{\infty} \sum_{j=-k}^{k} e^{-I(bk+ij)}$$
(6.7)

$$S_{n}(2) = \frac{i^{n}}{(n-1)!} \int_{0}^{\infty} I^{n-1} \sum_{j=1}^{\infty} \sum_{k=-j+1}^{j-1} e^{I(ibk-j)}$$
(6.8)

$$S_{n}(3) = \frac{(-1)^{n}}{(n-1)!} \int_{0}^{\infty} \boldsymbol{I}^{n-1} \sum_{k=1}^{\infty} \sum_{j=-k}^{k} e^{-\boldsymbol{I}(bk+ij)}$$
(6.9)

$$S_{n}(4) = \frac{(-i)^{n}}{(n-1)!} \int_{0}^{\infty} I^{n-1} \sum_{j=1}^{\infty} \sum_{k=-j+1}^{j-1} e^{I(ibk-j)}$$
(6.10)

The expressions inside the integrals can be analytically summed up. In fact, we have [Dienstfrey and Huang, 2001]

$$\boldsymbol{t}_{1} = \sum_{k=1}^{\infty} \sum_{j=-k}^{k} e^{-I(bk+ij)} = \frac{e^{bI} - e^{iI} + e^{bI+iI} + e^{bI+2iI}}{\left(e^{bI} - e^{iI}\right)\left(e^{bI+iI} - 1\right)}$$
(6.11)

$$\boldsymbol{t}_{2} = \sum_{j=1}^{\infty} \sum_{k=-j+1}^{j-1} e^{I(ibk-j)} = \frac{e^{ibI} \left(1+e^{I}\right)}{\left(e^{I}-e^{ibI}\right) \left(e^{(ib+1)I}-1\right)}$$
(6.12)

If the expressions given in (6.7)-(6.10) are summed up, the required lattice sums are obtained. For odd values of *n*, expressions (6.7)-(6.10) cancel each other out and, as a result, the lattice sums would be zero. For even numbers, the result is

$$S_{2n} = \frac{2}{(2n-1)!} \int_{0}^{\infty} \mathbf{l}^{2n-1} [\mathbf{t}_{1} + i^{2n} \mathbf{t}_{2}] d\mathbf{l} , \qquad (6.13)$$

When calculating the above-mentioned integrals to a sufficient accuracy, the lattice sums can be obtained to a desired accuracy. Note that Eq. (6.13) cannot be used for calculating S_2 . In fact, this case is conditionally convergent, and completely different values can be obtained for this sum. Rayleigh calculated this sum arbitrarily in the direction of the applied field and his method was questioned because of this. Poulton et al. [1999] have summarized a vast selection of literature pertaining to this subject. Recently, Dienstfrey and Huang [2001] have performed an attempt to include the case S_2 in the integral representation. Similar formulations can be developed for particulate composites. For example, Huang has derived the following integral representation for a simple cubic arrangement of inclusions:

$$S_{nm} = C \boldsymbol{I} \int_{0}^{\infty} \boldsymbol{I}^{n-3} e^{-l} \int_{0}^{2\boldsymbol{p}} \left\{ \boldsymbol{t}_{ud} (\boldsymbol{l}, \boldsymbol{a}) \cos(m\boldsymbol{a}) + i^{n} (\cos \boldsymbol{a} + 1)^{m} (\sin \boldsymbol{a})^{n-m} \boldsymbol{t}_{nsew} (\boldsymbol{l}, \boldsymbol{a}) \right\} / \boldsymbol{h}(\boldsymbol{l}, \boldsymbol{a}) \right\} d\boldsymbol{a} d\boldsymbol{l}$$

$$(6.14)$$

where

$$C = 1 / \left(p \sqrt{(n-m)! (n+m)!} \right)$$
(6.15)

$$t_{ud} = I^{3} \{ 1 + 2\cos(I\cos a) + 2\cos(I\sin a) + 4\cos(I\cos a)\cos(I\sin a) + e^{-1} [1 - 2\cos(I\cos a) - 2\cos(I\sin a) - 4\cos^{2}(I\cos a) - 4\cos^{2}(I\sin a)] + e^{-21} [-1 + 4\cos(I\cos a)\cos(I\sin a)] - e^{-31} \}$$
(6.16)

$$\boldsymbol{t}_{nsew} = 2\boldsymbol{I}^{3} \left\{ 1 + \cos(\boldsymbol{l}\cos\boldsymbol{a}) + 2e^{-\boldsymbol{l}} \left[\cos(\boldsymbol{l}\cos\boldsymbol{a}) + \cos^{2}(\boldsymbol{l}\cos\boldsymbol{a}) \right] - e^{-2\boldsymbol{l}} \left[1 + \cos(\boldsymbol{l}\cos\boldsymbol{a}) + 2\cos(\boldsymbol{l}\sin\boldsymbol{a}) + 2\cos(\boldsymbol{l}\cos\boldsymbol{a})\cos(\boldsymbol{l}\sin\boldsymbol{a}) \right] \right\}$$
(6.17)

$$h(\mathbf{l}, \mathbf{a}) = \left\{ 1 - 2e^{-1} \cos[\mathbf{l}(\cos \mathbf{a} - \sin \mathbf{a})] + e^{-21} \right\} \left\{ 1 - 2e^{-1} \cos[\mathbf{l}(\cos \mathbf{a} + \sin \mathbf{a})] + e^{-21} \right\}$$
(6.18)

Note that for a simple cubic arrangement, non-zero cases happen when n is an even number and m a multiple of 4. Also, as explained, the above relation cannot be used for the case n=2.

6.2 Calculating lattice sums over reciprocal arrays

Consider the same rectangular array as studied in section (6.1). To calculate the propagation of electromagnetic waves through this medium, the following equation, which is well known as the Helmholtz equation, needs to be solved:

$$\left(\nabla^2 + \boldsymbol{k}^2\right) \boldsymbol{\hat{A}}(\boldsymbol{r}) = 0, \qquad (6.19)$$

where **k** is the wave number, **r** the position vector and \hat{A} can be an electric or magnetic field.

Such an investigation can be successfully performed using the Rayleigh method as outlined by many researchers [Poulton et al., 1999]. The final equation, similarly to the discussion in chapter 2, is, again, a linear algebraic system; however, it contains sums of the form $S_n^d(\mathbf{k}) = \sum_j J_n^2(\mathbf{k}r_j) e^{inq_j}$ over the cylinders, where J_n^2 is the Bessel function of the second

kind and order *n*. Since these lattice sums are frequency–dependent, they are here called them dynamic lattice sums. To obtain an absolutely convergent series and also accelerate the convergence, Chin et al. [1994] have proposed a method to calculate these lattice sums in terms of Bessel functions over the reciprocal array. The reciprocal array is a set of imaginary points constructed for the lattice under study by

$$\boldsymbol{r}_{h} = (r_{h}, \boldsymbol{q}_{h}) = h_{1} 2\boldsymbol{p}\mathbf{i}/b + h_{2} 2\boldsymbol{p}\mathbf{j} \qquad h = (h_{1}, h_{2}) \in Z^{2}$$

$$(6.20)$$

The methodology involves a considerable mathematical effort which relies on finding Green's function of Eq. (6.19) in the direct lattice and in the reciprocal array. Green's function is the solution of Eq. (6.19) when there is a Dirac function on the right–hand side. It can be shown that Green's function in the direct array involves dynamic lattice sums, and in the reciprocal array it consists of a convergent summation in terms of Bessel functions. Furthermore, Green's function in the direct lattice and the reciprocal array are related to each other [Chin et al., 1994; Nicorovici et al. 1995]; thus, relating these two functions, the dynamic lattice sums can be obtained in an absolutely convergent series over a reciprocal array. The final result for the rectangular array is as follows:

$$S_{2n}^{d}(\mathbf{k})J_{2n+q}^{1}(\mathbf{k}) = -\frac{4}{b}(-1)^{n}\sum_{h}\left(\frac{\mathbf{k}}{r_{h}}\right)^{q}\frac{J_{2n+q}^{1}(r_{h})}{r_{h}^{2}-\mathbf{k}^{2}}e^{-i2n\mathbf{q}_{h}}, \quad n \ge 1$$
(6.21)

where q > 0 is an arbitrary integer parameter. By increasing the value of q, the lattice sums converge more rapidly, but excessively large values of q may lead to instability in the numerical algorithm. At the long-wavelength limit ($\mathbf{k} \rightarrow 0$), Eq. (6.19) will be reduced to the Laplace equation. In this limit, the lattice sums would be static, i.e. those that are the interest of this thesis. A simplification of (6.21) for this limit [Movchan, 1997] gives

$$S_{2n} = (-1)^n \boldsymbol{p} \, \frac{(2n+q)!}{b(2n-1)!} \Biggl[\sum_{h\neq 0} \frac{J_{2n+q}^1(r_h)}{(r_h/2)^{2+q}} e^{-12n\boldsymbol{q}_h} + \frac{1}{(q+2)!} e^{-2i(\boldsymbol{q}_0+\boldsymbol{p}/2)} \boldsymbol{d}_{n1} \Biggr], \tag{6.22}$$

where q_0 is the direction of the incident field. All the required lattice sums for the rectangular array can be calculated in this way.

Equally well, the same methodology can be developed for particulate composites. The reciprocal array for a lattice with periodicity equal to b, c and a unity in the x-, y- and z-directions takes the following form:

$$\boldsymbol{r}_{h} = (r_{h}, \boldsymbol{q}_{h}, \boldsymbol{f}_{h}) = h_{1} 2p\mathbf{i}/b + h_{2} 2p\mathbf{j}/c + h_{3} 2p\mathbf{k}, \quad h = (h_{1}, h_{2}, h_{3}) \in \mathbb{Z}^{3}$$
 (6.23)

Nicorovici et al. [1995] have shown that the static lattice sums for this system can be derived from the following formula:

$$S_{nm} = \frac{(2n+2q+1)!}{(2n-1)!} \frac{4\mathbf{p}}{bc} \mathbf{i}^{n} \left[\sum_{h \neq 0} \frac{J_{n+q}^{1}(r_{h})}{r_{h}^{q+2}} Y_{nm}^{*}(\mathbf{q}_{h}, \mathbf{f}_{h}) + \frac{1}{(q+5)!} Y_{2,m}^{*}(\mathbf{q}_{0}, \mathbf{f}_{0}) \mathbf{d}_{n2} \right], \ n \geq 2, \ m \geq 0$$
(6.24)

where the asterisk stands for a complex conjugation and q_0 and f_0 are the directions of the applied field.

6.3 Results

Table (6.1) shows a sample of the results of the present study for the three-phase fiber composites, which has been obtained using the above methodologies. Although the integral representation requires more computer programming, it is faster than the second method; therefore, all the cases except n = 1 have been calculated using this method. For cylinders of type two, applying the integral representation technique yields the following:

$$S_{2n}^{2} = \frac{1}{\boldsymbol{u}^{2n}} + \frac{1}{(2n-1)!} \int_{0}^{\infty} \int_{0}^{2n-1} \left[\left(e^{l\boldsymbol{u}} + e^{-l\boldsymbol{u}} \right) \boldsymbol{t}_{1} + i^{2n} \left(e^{il\boldsymbol{u}} + e^{-il\boldsymbol{u}} \right) \boldsymbol{t}_{2} \right] d\boldsymbol{l} , \qquad (6.25)$$

where $\mathbf{u} = b/2 + i/2$. For calculating integrals (6.13) and (6.25), well-established methods such as Gauss-Laguerre integration, can be used for this purpose [Press et al., 1986].

N	S_n^1	S_n^2	$S_n^1 + S_n^2$
2	0.3392133718630	3.2883853566054	3.6275987284684
4	2.1744038488973	-2.1744038488973	0.0
6	-2.0154171446150	-3.8476145488104	-5.8630316934253
8	2.0262994706141	-2.0262994706141	0.0
10	-1.9919685438111	1.9919685438111	0.0
12	2.0041914554050	4.0054485162926	6.0096399716977
14	-1.9990818777902	1.9990818777902	0.0
16	2.0003051627592	-2.0003051627592	0.0
18	-1.9999213768630	-3.9997969795075	-5.9997183563705
20	2.0000338688335	-2.0000338688335	0.0
22	-1.9999887079801	1.9999887079801	0.0
24	2.0000041209456	4.0000075266341	6.0000116475798
26	-1.9999987455894	1.9999987455894	0.0
28	2.0000004181529	-2.0000004181529	0.0
30	-1.9999998662038	-3.9999997212317	-5.9999995874356
32	2.000000464611	-2.000000464611	0.0
34	-1.9999999845130	1.9999999845130	0.0
36	2.000000052497	4.000000103247	6.000000155744
38	-1.99999999982792	1.99999999982792	0.0
40	2.000000005736	-2.000000005736	0.0

Table 6.1 The calculated lattice sums for the case of a three-phase composite of a periodic array of cylinders when $b = \sqrt{3}$.

In this thesis, these integrals have been calculated using the Mathematica 3 software application, which offers the possibility to check the accuracy of the results. For the case S_2^1 , Eq. (6.22) has been used. q was selected to be 12, and the computation was performed over a 200×200 reciprocal array. Calculating S_2^1 , S_2^2 can be found simply by using $S_2^2 = 2\mathbf{p}/\sqrt{3} - S_2^1$. Note that for $n \gg 1$, the direct summation method can quickly yield accurate results.

7. RESISTOR MODELS FOR DERIVING THE BOUNDS FOR THE EFFECTIVE CONDUCTIVITY

From one perspective, two main approaches for calculating the effective transport properties of composite materials can be identified. According to the first strategy, the efforts are concentrated on directly solving the governing equations of the system [Rayleigh, 1892; Cheng and Torquato, 1997; Lu, 1994]. The second method differs from the first one in that there the aim is to develop bounds (upper-lower) for the effective transport properties of the system [Hashin and Shtrikman, 1962; Torquato et al., 1986; Helsing, 1993]. In general, fewer details of the system are required for the second method; however, using more specifications, tighter bounds may be derived. The best-known bounds are those proposed by Hashin and Shtrikman. Hashin and Shtrikman have shown that the bounds they obtained can be realised for some particular microstructures. The upper bounds can be attained using an assemblage of coated spheres in which the core is filled with a material of higher conductivity; the upper bound is attained using the same structure that changes the material of the core and shell. Since these structures introduce more disconnected-connected microstructures of the phases, Hashin-Shtrikman bounds can be considered to be optimal.

In this section, two simple resistor models are used (see Moosavi et al. [2002b]) for obtaining the bounds on the effective conductivity of systems consisting of simple cubic arrays of multicoated spheres (or square arrays of cylinders). Figures 7.1 and 7.2 outline the methods for finding two bounds for the effective conductivity of a system. For the lower bound, the methodology given in Fig. 7.1 is used. In this method, the unit cell is divided into infinite layers in the direction of the heat flow. The effective conductivity can be derived as

$$k_{e}^{+} = \sum_{i=1}^{N-1} \boldsymbol{p} \left[a_{i}^{2} - a_{i-1}^{2} \right] A_{i} + 1 - \boldsymbol{p} a_{N-1}^{2}, \qquad (7.1)$$

where

$$A_{i} = \frac{a_{i}^{2}}{a_{i}^{2} - a_{i-1}^{2}} \int_{q_{i}}^{p/2} \frac{\sin 2q dq}{\sum_{j=i}^{N-1} 2a_{j} \sqrt{1 - (a_{i} \sin q / a_{j})^{2}} (1/k_{j} - 1/k_{j+1}) + 1}$$
(7.2)



Figure 7.1 Quarter unit cell considered in the derivation of the effective conductivity by using the first resistor model

For deriving the upper bound, the strategy outlined in Fig. 7.2 is used. In this method, the unit cell is divided into infinite slices normal to the direction of the heat flow. The effective conductivity of the system can be given by



Figure 7.2 Quarter unit cell under inspection in the second resistor model

$$k_{e}^{-} = \frac{1}{\sum_{i=1}^{N-1} \frac{2(a_{i} - a_{i-1})}{B_{i}} + 1 - 2a_{N-1}},$$
(7.3)

where

$$B_{i} = \frac{1 - a_{i-1}/a_{i}}{\int_{0}^{q_{i}} \frac{\sin q dq}{p \left\{ k_{i} (a_{i} \sin q)^{2} + \sum_{j=i+1}^{N-1} k_{j} (a_{j}^{2} - a_{j-1}^{2}) - [a_{N-1}^{2} - (a_{i} \cos q)^{2}] \right\} + 1}$$
(7.4)

Following the same strategy as discussed above for spheres, it can be shown for the case of a square array of cylinders that

$$k_{e}^{+} = \sum_{i=1}^{N-1} 2[a_{i} - a_{i-1}]A_{i} + 1 - 2a_{N-1}, \qquad (7.5)$$

where

$$A_{i} = \frac{a_{i}}{a_{i} - a_{i-1}} \int_{q_{i}}^{p/2} \frac{\cos q \, dq}{\sum_{j=i}^{N-1} 2a_{j} \sqrt{1 - (a_{i} \sin q / a_{j})^{2}} (1/k_{j} - 1/k_{j+1}) + 1}$$
(7.6)

and for the lower bound

$$k_{e}^{-} = \frac{1}{\sum_{i=1}^{N-1} \frac{2(a_{i} - a_{i-1})}{B_{i}} + 1 - 2a_{N-1}},$$
(7.7)

where

$$B_{i} = \frac{1 - a_{i-1}/a_{i}}{\int_{0}^{q_{i}} \frac{\sin q \, dq}{2\left\{k_{i}a_{i}\sin q + \sum_{j=i+1}^{N-1} k_{j}\left[\sqrt{a_{j}^{2} - (a_{i}\cos q)^{2}} - \sqrt{a_{j-1}^{2} - (a_{i}\cos q)^{2}}\right] - \sqrt{a_{N-1}^{2} - (a_{i}\cos q)^{2}}\right\} + 1}$$

81

(7.8)

Figure 7.3 shows a typical comparison between the results of the bounds and exact results. The total volume fraction (*F*) was selected to be 0.2 ($f_1 = 0.1$ and $f_2 = 0.1$). It is perhaps interesting that in some cases the second resistor model yields better results. This is contrary to the natural thinking that since the case is coated and heat goes from one layer to another, considering the resistor in the series should yield better results. Also, it can been seen that the upper bound does not conform to the behaviour of the exact solution when the coating layer is perfectly conducting. The same can be observed when the coating layer is perfectly insulating.



Figure 7.3 Logarithmic plot of the effective conductivity for the simple cubic array of coating spheres as a function of conductivity of the core and shell. Despite of the accuracy, the first resistor model is unable to show the same behaviour as the exact solution when the shell is perfectly conducting. The same condition applies for the second resistor model when the shell is perfectly insulating.

8. Conclusion

This thesis studied the effective transport properties of multiphase composite materials made up of periodic arrays of circular cylinders, elliptical cylinders and spheres using a methodology indebted to Rayleigh [1892]. For each section, a series of explicit relations for easy calculating of the effective property were reported.

Composites containing inclusions in the shape of coated circular cylinders were considered, the behaviour of singly coated systems, when the sum of the transport property of the layers is zero, was extended to multicoated ones, and a generalisation of this phenomenon was provided. This thesis showed that the procedure for neglecting phases can occur in different methods and that these procedures can affect several layers and also explained the details of these procedures. Furthermore, a formulation for deriving the effective property of composites with a periodic array of confocally multicoated elliptical cylinders was presented and it was shown that, in general, the behaviour observed in composites made up of circular cylindrical inclusions does not hold for these coated composites. However, further study is required to determine whether the coating layers of other shapes (uniform coating, for example) can demonstrate this behaviour. This thesis also explained that contrary to the case of coated circular cylinders, the generalisation behaviour for composites containing singly coated spheres cannot be developed for composites with multicoated spheres. Whenever necessary, the results were verified using classical numerical techniques.

For the case of three-phase composites with inclusions in the shape of circular cylinders, this thesis showed that these materials can exhibit interesting behaviour. By having mono-sized inclusions in a position of contact when the unit cell is a rectangular cylinder and one phase is perfectly conducting and the other insulating, a composite material can be obtained that is super-conducting in one direction and non-conducting in another. If the rectangular array is changed to a square one in the mentioned case, surprisingly, the effect of all the dispersed phases can be neglected.

The effect of interfacial resistance on effective conductivity was studied by characterising interfacial resistance using a non-dimensional parameter. It was shown that this effect is very

important, and on the basis of the value of interfacial resistance, even the effective conductivity of the system can fall outside the range of the property of the phases. The number of situations in which the effect of both phases can be neglected is infinite; however, these situations are usually valid for one direction only. There is only one state that is common and if it occurs, the effect of the inclusions can be neglected despite the direction in which the effective property is calculated. This situation occurs when both phases have an interfacial resistance equal to their conductivity minus one. It was further demonstrated that there is an interfacial resistance, which can be estimated by $R_i \leq k_i - 1$, in which the effect of the inclusions of type *i* for one direction can be neglected. These results can be applied for the cases of inclusions in the shape of circular cylinders or spheres.

Two resistor models were used for deriving the bounds. The results of the bounds were compared for coated composites, and it was shown that having the resistor in a series does not guarantee that better results will be obtained in all cases. By combining these bounds in a specific manner, better results may be obtained than by using one of them alone. For instance, when an inclusion has a property close to that of the matrix, it is advisable to use a simple average value.

Although the formulation and results of this thesis on multi-phase composite materials have been verified in some cases by relevant theorems and numerical results, comparing them with experimental results can be a crucial task. Unfortunately, the experimental results found in subjects relevant to the study belong mostly to two-phase systems and, in some cases, to three-phase ones. These results have been used by many authors to validate the theoretical formulations and results [Meredith and Tobias, 1960; McPhedran and McKenzie, 1978; Perrins et al., 1979; Nicorovici et al., 1995; Yardley et al., 1999]. Since the formulations and results given in this thesis are in complete agreement with those of the mentioned authors, they have, therefore, already been compared to the experimental results in two- and three-phase systems. However, for the new formulations and results obtained on multi-phase systems in this thesis, the validation of the results with experimental results can be helpful and is suggested as a topic for further study in this field.

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THE RESONANT BEHAVIOR OF A MULTI-PHASE COMPOSITE MATERIAL

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We study the resonant behavior of a system consisting of a square array of multi-coated cylinders by calculating the effective dielectric constant of the system. The results were examined numerically using the finite element method.

Keywords: Dielectric constant; resonance; cylinder; multi-coated; finite element method.

1. Introduction

Finding the response of a system consisting of a periodic array of solid cylinders embedded in a homogenous matrix has a long history dating back to Lord Rayleigh.¹ Runge² extended Rayleigh's method to coated elements where the geometry was composed of an array of tubes and the core of the tubes was filled with the same material as the matrix. Israelachvili *et al.*³ reported the solution of the problem when the materials of core and matrix were different.

The inspection of the behavior of the solution as a function of the property of the core and shell divulged a new feature of the system. Nicorovici *et al.*⁴⁻⁶ discovered that the procedure of coating the cylinders with a material that has a dielectric constant, which is the negative of that of the cylinders ($\varepsilon_{\text{shell}} = -\varepsilon_{\text{core}}$) or matrix ($\varepsilon_{\text{shell}} = -\varepsilon_{\text{matrix}}$) can yield the response of a system with magnified cylinders. This means that a system with diminutive concentration may conditionally give the response of a concentrated system. These authors termed the system in these situations "partially-resonant" and the conditions that put the system in these particular states the "partial resonances" of the system. The term "resonance" may be misleading here. In fact, the above-mentioned features are a generalization of what happens in two-phase composites when $\varepsilon_{\text{core}} = -\varepsilon_{\text{matrix}}$, which is not a resonance.^{7,8} Therefore, one needs to consider that the term "partial resonance"

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only refers to a specific condition. The term "partial" was applied due to the fact that the response of the system in these situations is still limited.

In this report, we elucidate how the above-described behavior occurs in a system consisting of arrays of multi-coated cylinders. The motivation for this research stems from the further need to inspect the behavior of systems with components, which have a negative permittivity (ε) or negative permeability (μ) or systems having both ε and μ negative (the so-called left-handed medium⁹). These systems have shown interesting and unexpected results,^{4-6,9-11} which may also be of practical interest.¹²

Note that, although periodic arrays are idealized microstructures they may be realizable experimentally. One reason for studying these structures is that their properties can many times be easily computed. Also, the results of periodic systems can be useful for grasping the interplay between the microstructure and macroscopic properties of composites. We numerically show that a disordered structure can also be partially resonant.

The structure of this report is explained here. The following section describes the geometry under study. In Sec. 3, we briefly explain the procedure of deriving the response of the system. Section 4 predicts some results from the theoretical study of the system when it is in resonant states. Section 5 verifies the results of the previous section and finally, in Sec. 6, we summarize the key findings of this study.

2. Geometric Description

Consider a homogenous matrix with a unit dielectric constant surrounding an array of composite cylinders, which have a topology based upon the well-known square lattice. Each lattice point of the square array can be described by a lattice vector, r_n , defined as:

$$\boldsymbol{r}_n = h(n_1 \mathbf{e}_{x_1} + n_2 \mathbf{e}_{x_2}), \qquad (1)$$

where h is the characteristic length that expresses all the dimensionless distances n_1 and n_2 are arbitrary integers and the two basic vectors, \mathbf{e}_{x_1} and \mathbf{e}_{x_2} , form the orthonormal basis of the plane. Let θ be an angle measured from the x_1 -axis. Thus, one can write

$$x_1 = r\cos\theta, \quad x_2 = r\sin\theta. \tag{2}$$

The radiuses of the core and coating layers are determined by a_1, \ldots, a_{N-1} , respectively, as is shown Fig. 1. With these considerations, one can show that the volume fractions occupied by the core and coating layers can be obtained from

$$f_i = \pi (a_i^2 - a_{i-1}^2) \quad (i = 1, \dots, N - 1)$$
(3)

and the total volume fraction can be expressed as:

$$F = \sum_{i=1}^{N-1} f_i \,. \tag{4}$$



Fig. 1. The multi-coated cylinder under study.

 $a_0 = 0$ has been included here and in the following relations in order to reduce the number of mathematical notations. We also use the term "layer" for the core, coating layers and matrix, and the dielectric constant ratio between layers i-1 and i is represented by $\varepsilon_{i-1,i}$.

3. Mathematical Modeling

Considering the problem symmetrical allows it to be solved independent of the direction of the applied field, and without compromising generality, we assume a potential gradient of unit magnitude to be applied along the x_1 -axis. For the unit cell located at the origin, considering the general solution of the Laplace equation in polar coordinates (r, θ) and following Ref. 1, the potential (V) inside the layers may be given as:

$$V^{1} = \sum_{n=1}^{\infty} E_{n}^{1} r^{2n-1} \cos(2n-1)\theta \qquad r \le a_{1}, \qquad (5a)$$

$$V^{i} = \sum_{n=1}^{\infty} [E_{n}^{i} r^{2n-1} + F_{n}^{i} r^{-2n+1}] \cos(2n-1)\theta \qquad a_{i-1} \le r \le a_{i} (i=2,\dots,N-1),$$
(5b)

$$V^{N} = \sum_{n=1}^{\infty} [E_{n}^{N} r^{2n-1} + F_{n}^{N} r^{-2n+1}] \cos(2n-1)\theta \quad r \ge a_{n-1},$$
 (5c)

where E_n^N and F_n^N are unknown coefficients that are to be determined.

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At all the surfaces between the layers, the potential and normal component of the electric displacement are continuous, i.e.,

$$\begin{vmatrix} V^{1} = V^{2} \\ \varepsilon_{1} \frac{\partial V^{1}}{\partial r} = \varepsilon_{2} \frac{\partial V^{2}}{\partial r} \end{cases} \qquad r = a_{1}, \qquad (6a)$$

$$\begin{vmatrix} V^{i-1} = V^{i} \\ \varepsilon_{i-1} \frac{\partial V^{i-1}}{\partial r} = \varepsilon_{i} \frac{\partial V^{i}}{\partial r} \\ r = a_{i-1}(i = 2, \dots, N-1), \quad (6b)$$

$$\begin{vmatrix} V^{N-1} = V^{N} \\ \varepsilon_{N-1} \frac{\partial V^{N-1}}{\partial r} = \varepsilon_{N} \frac{\partial V^{N}}{\partial r} \qquad r = a_{N-1}. \end{aligned}$$
(6c)

By applying Eqs. (6) E_n^i and F_n^i can be related to each other as:

$$F_n^i + L_n^i a_{i-1}^{4n-2} E_n^i = 0 \quad (i \ge 2)$$
(7)

where

$$L_n^i = \frac{\varepsilon_{i-1,i} - 1 + (\varepsilon_{i-1,i} + 1)L_n^{i-1}(a_{i-2}/a_{i-1})^{4n-2}}{\varepsilon_{i-1,i} + 1 + (\varepsilon_{i-1,i} - 1)L_n^{i-1}(a_{i-2}/a_{i-1})^{4n-2}}.$$
(8)

To derive E_n^N and F_n^N , one may follow the method of Zuzovski and Brenner.¹³ These authors derived another relation for the potential through the matrix in one unknown A_n . Comparing the resultant relation with Eq. (5c), allows two linear equations to be found relating E_n^N and F_n^N to A_n . These two equations yield a set of linear equations in the unknowns A_n with the help of Eq. (7)

$$\frac{A_n}{a_{N-1}^{4n+2}L_{n+1}^N} = \sum_{J=1}^{\infty} \left(\frac{2n+2J-1}{2n+1}\right) S_{2n+2J}A_{J-1} + \delta_{n0} , \qquad (9)$$

where δ_{n0} represents the Kronecker delta (1 for n = 0 otherwise 0) and S_n are constants characteristic of the array. After finding E_n^N and F_n^N , other coefficients can be found by using Eqs. (6).

The effective dielectric constant can be calculated using the following formula

$$\varepsilon_{\text{eff}} = 1 + 2\pi A_0 \,, \tag{10}$$

where A_0 can be derived by solving the system of linear algebraic equations obtained from Eq. (9). As a solution in an explicit form, we present the following simple formula, which gives reasonable results in very low volume fractions

$$\varepsilon_{\text{eff}} = 1 - \frac{2F}{-1/L_1^N + F}.$$
 (11)

In order to obtain a more accurate expression one can use the methods outlined by Manteufel and Todreas. 14

4. Theoretical Prediction

As can be seen from Eq. (9), a_{N-1} and L_n^N play important roles in the response of the system to the applied field. Due to the form of the mathematical expression of L_n^N [i.e., Eq. (8)], for some cases, there may exist a different number and series of dispersed layers that provide following relation

$$a_{N^*-1}^{4n+2}L_{n+1}^{N^*} \equiv a_{N-1}^{4n+2}L_{n+1}^N \tag{12}$$

and, as a result, the response of the system in these situations would be the same. For example, when the coating layer i is perfectly conducting or insulating, L_n^{i+1} would be 1 and -1, respectively. This means that the layers under the layer i will have no effect on the calculation of L_n^N . Thus, there are infinite selections for the number and property of the layers under the layer i.

The particular cases occur when

$$\varepsilon_{i-1} + \varepsilon_i = 0 \quad (2 \le i \le N). \tag{13}$$

From Eq. (8) we find the following

$$L_n^{i+1} = \frac{P+Q}{R+S} \quad (1 \le i \le N-1)$$
(14)

where

$$\begin{cases} P = (\varepsilon_{i,i+1} - 1) \left[\varepsilon_{i-1,i} + 1 + (\varepsilon_{i-1,i} - 1) L_n^{i-1} \left(\frac{a_{i-2}}{a_{i-1}} \right)^{4n-2} \right] \\ Q = (\varepsilon_{i,i+1} + 1) \left[\varepsilon_{i+1,i} - 1 + (\varepsilon_{i-1,i} + 1) L_n^{i-1} \left(\frac{a_{i-2}}{a_{i-1}} \right)^{4n-2} \right] \left(\frac{a_{i-1}}{a_i} \right)^{4n-2} \\ R = (\varepsilon_{i,i+1} + 1) \left[\varepsilon_{i-1,i} + 1 + (\varepsilon_{i-1,i} - 1) L_n^{i-1} \left(\frac{a_{i-2}}{a_{i-1}} \right)^{4n-2} \right] \\ S = (\varepsilon_{i,i+1} - 1) \left[\varepsilon_{i-1,i} - 1 + (\varepsilon_{i-1,i} + 1) L_n^{i-1} \left(\frac{a_{i-2}}{a_{i-1}} \right)^{4n-2} \right] \left(\frac{a_{i-1}}{a_i} \right)^{4n-2} \end{cases}$$
(15)

Substituting condition (13) into Eq. (14) gives the following statement

$$L_n^{i+1} = \frac{\varepsilon_{i-1,i+1} - 1 + (\varepsilon_{i-1,i+1} + 1)L_n^{i-1}(a_{i-2}/a_i)^{4n-2}(a_i/a_{i-1})^{2(4n-2)}}{\varepsilon_{i-1,i+1} + 1 + (\varepsilon_{i-1,i+1} - 1)L_n^{i-1}(a_{i-2}/a_i)^{4n-2}(a_i/a_{i-1})^{2(4n-2)}}.$$
 (16)

If two layers, i-1 and i, have the same dielectric constant, which is equal to ε_{i-1} , one may derive the following

$$L_n^{i+1} = \frac{\varepsilon_{i-1,i+1} - 1 + (\varepsilon_{i-1,i+1} + 1)L_n^{i-1}(a_{i-2}/a_i)^{4n-2}}{\varepsilon_{i-1,i+1} + 1 + (\varepsilon_{i-1,i+1} - 1)L_n^{i-1}(a_{i-2}/a_i)^{4n-2}}.$$
(17)

A comparison of Eqs. (16) and (17) shows that the field through the continuous phase would be the same if layer i had the same property as layer i - 1 and all

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the layers under layer i - 1 were magnified by a factor of $(a_i/a_{i-1})^4$. Layer i - 1 experiences two changes. Magnification by occupying the place of layer i and reduction due to the extension of layer i - 2. Thus, this case can be materialized only if

$$a_{i-1} > \sqrt{a_i a_{i-2}}$$
 (18)

The inspection of L_n^{i+1} in condition (13) revealed part of the results. Let us now derive an expression for L_n^i . From Eq. (8) after substituting Eq. (13) and developing a relation for L_n^{i-1} analogous to Eq. (8), the following equation can be derived

$$L_{n}^{i} = \frac{\varepsilon_{i-2,i} - 1 + (\varepsilon_{i-2,i} + 1)L_{n}^{i-2}(a_{i-3}/a_{i-1})^{4n-2}(a_{i-1}/a_{i-2})^{4n-2}}{\varepsilon_{i-2,i} + 1 + (\varepsilon_{i-2,i} - 1)L_{n}^{i-2}(a_{i-3}/a_{i-1})^{4n-2}(a_{i-1}/a_{i-2})^{4n-2}} \times \left(\frac{a_{i-1}}{a_{i-2}}\right)^{4n-2} \quad (2 \le i \le N).$$

$$(19)$$

When two layers, i - 1 and i - 2, have the same dielectric constant equal to ε_{i-2} , the result for L_n^i would be

$$L_n^i = \frac{\varepsilon_{i-2,i} - 1 + (\varepsilon_{i-2,i} + 1)L_n^{i-2}(a_{i-3}/a_{i-1})^{4n-2}}{\varepsilon_{i-2,i} + 1 + (\varepsilon_{i-2,i} - 1)L_n^{i-2}(a_{i-3}/a_{i-1})^{4n-2}}.$$
(20)

A comparing of Eqs. (19) and (20) indicates that there is another equivalent system in which all the layers (i = 1, ..., i - 3) have been extended by a factor of $(a_{i-1}/a_{i-2})^4$. The layer i-1 now has the property equal to that of layer i-2 and its outer radius is $a_{i-1} \times (a_{i-1}/a_{i-2})$. This system can be materialized if

$$a_{i-1} < \sqrt{a_i a_{i-2}} \,. \tag{21}$$

Some conclusions can be drawn on the basis of the above relations. When i = 2 in Eq. (13), the second equivalent system cannot occur, but there is always an equivalent system. When $3 \le i \le N-1$, both equivalent systems can occur, although not simultaneously because of the limitations dictated in Eqs. (18) and (21), which contravene each other. Therefore, finding the equivalent system in this situation has been warranted except when $a_{i-1} = \sqrt{a_i a_{i-2}}$. If i = N, the first equivalent system has no meaning and the second one can occur if $a_{i-1} < \sqrt{a_{i-2}/2}$.

Since Eq. (13) expresses resonance between two successive layers, one may consider different cases in which several pair layers satisfy relation (13), i.e.,

$$\varepsilon_{i-1} + \varepsilon_i = 0 \quad (i = j, k, l, \ldots).$$
 (22)

The behavior of the system in these states can be readily detected by successively applying the methods declared above. For example, we consider the following case

$$\varepsilon_{i-1} + \varepsilon_i = 0 \quad (i = 2, \dots, N-1).$$
⁽²³⁾

Successively employing the methods ultimately indicates that the field inside the matrix would not change if the multi-coated cylinders were replace by solid cylinders

of radius a_{N-1} and dielectric constant ε_1 . Therefore it can be shown that L_n^N can be simplified into the following form

$$L_n^N = \frac{\varepsilon_1 - 1}{\varepsilon_1 + 1} \tag{24}$$

and as a result, the potential inside the matrix would be

$$V^{N} = \sum_{n=1}^{\infty} F_{n}^{N} \left[\frac{1 + \varepsilon_{1}}{(1 - \varepsilon_{1})a_{N-1}^{4n-2}} r^{2n-1} + r^{-2n+1} \right] \cos(2n-1)\theta.$$
 (25)

From the above result, it can be understood that in this situation, the sign of ε for the layers between layers 1 and N(1 < i < N) can be arbitrarily chosen and all the cases yield the same response.

When all the layers (i = 2, ..., N) fulfil condition (13) and N is an odd number, the system behaves like a solid medium with the dielectric constant of the matrix.

5. Numerical Verification

Numerical simulation is nowadays a well-developed tool for inspecting the response of systems. Although the unit cell of the periodic structures has been largely simulated,¹⁵ they are mostly in two-phase with positive transport properties.

For given geometry and solid volume fractions, the Matlab PDE toolbox was utilized and the Laplace equation solved for a unit cell of the system using the finite element method. The unit cell consists of matrix and dispersed layers (core and coating layers). A potential gradient of unit magnitude was applied externally along the x_1 -axis and other external boundaries were insulated. At the surfaces between the layers, continuity conditions (6) were implemented. By using solutionadaptive refinement, one can add cells where they are needed in the mesh, thus enabling the features of the potential field to be better resolved. Based on the theoretical findings, the three considerable cases were studied numerically.

- Figure 2(a) shows a unit cell of a system consisting of three-coated cylinders. The dielectric constants of the core and coating layers were selected to be +2.5, +5, +2 and -2 and the radiuses 0.15, 0.2, 0.275 and 0.33, respectively. In order to construct the equivalent system [Fig. 2(b)], second and third coating layers were joined together and considered as one unit layer with a dielectric constant equal to +2. Also, the core and first coating layers are magnified by the factor $(0.33/0.275)^4$. Therefore, the radiuses of the layers are $a_1^* = 0.216$, $a_2^* = 0.288$ and $a_3^* = 0.33$. The distributions of the induced fields are given in the figures show that although the fields inside the dispersed layers of the systems are completely different, they are exactly the same through the matrix.
- Now we consider the system of Fig. 2(a) with one alteration. In order to satisfy condition (21), the radius of the third coating layer has been extended to the value 0.4 as is shown in Fig. 3(a). Based on the predicted scheme for the second equivalent system, we assemble a two-coated cylinder [Fig. 3(b)] with radiuses



Fig. 2. Equipotential contours inside the unit cell of the first case. The original system (a) and equivalent system (b).

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(b)

Fig. 3. Equipotential contours inside the unit cell of the second case. The original system (a) and equivalent system (b).



Fig. 4. Induced potential field inside the layers of the unit cell in third case. The original (a) and equivalent system (b).



(b)

Fig. 5. Induced potential fields inside the layers of a simple system composed of a two-coated cylinder and arbitrary boundaries. The original (a) and equivalent system (b). Note that the potential fields outside the first coating layers are equal. This is also correct for the third case.

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of $a_1^* = 0.28359375$, $a_2^* = 0.378125$ and $a_3^* = 0.4$. The dielectric constants of the layers are +2.5 for the core, +5 for the first coating layer and -2 for the second coating layer. It is clear from the figures that the potential fields outside a_2^* are equal.

• In third case, a two-coated cylinder has been considered as depicted in Fig. 4(a). The dielectric constants of the core and coating layers are -2, +2 and -2, respectively. In the next system [Fig. 4(b)], the two-coated cylinder is replaced by a solid cylinder with the same dielectric constants as the core of the original system. Both systems have the same total volume fraction. Again, by applying the same boundary conditions, the fields through the matrix of both systems are the same.

To further investigate this subject, we consider a simple system consisting of a two-coated cylinder (with details explained in the third case above) covered by another material of unit dielectric constant. Arbitrary boundary and boundary conditions were selected and implemented on the system as is represented in Fig. 5(a). Figure 5(b) shows that changing the dielectric constant of the first coating layer to -2 causes no disturbance in the field through the matrix. Therefore, disordered structures can also be partially resonant. This fact had been predicted by Nicorovici *et al.*^{5,6} and the numerical investigation shows the same result.

6. Summary

Inspection of the resonant behavior of the multi-coated structures exposed new results in this field. When the sum of the dielectric constants of two successive layers is equal to zero, a series of layers can be magnified and the magnification is not limited to one layer. The ratio of magnification for the layers detailed. We also explained that, for every resonant state, there may be two types of equivalent system, although only one of them may be of any physical significance. All the numerical investigations were in accordance with the theoretical predictions.

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The effective conductivity of composite materials with cubic arrays of multi-coated spheres

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ABSTRACT Two premeditated resistor models have been developed and tested for the prediction of the effective thermal conductivity of a periodic array of multi-coated spheres embedded in a homogeneous matrix of unit conductivity. The results have been compared and evaluated with the exact solution, as obtained by extending a method originally devised by Zuzovski and Brenner. The results for the two models were found to yield bounds for the exact solution. For some situations, the model results match well with the exact solution, but in other cases the results for one of the models could deviate from the exact solution.

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1 Introduction

Many studies have been conducted on calculating the effective thermal conductivity of a composite medium consisting of a periodic array of spheres embedded in an isotropic matrix. The basic work for the solid spheres has been outlined by Maxwell [1], who inspected the effective conductivity of a dilute spherical dispersion. Rayleigh [2] described the polarization of each sphere in an external field by an infinite set of multipole moments and gave a relation of low order for a simple array of spheres. His solution was corrected later by Runge [3] and improved by Meredith and Tobias [4]. Maxwell's theory was extended by Jeffrey [5] to higher particle concentrations. McPhedran et al. [6] modified Rayleigh's method to overcome a non-absolutely convergent series involved in the solution. Zuzovski and Brenner [7] presented another method that avoids the problems encountered in Rayleigh's original method. Sangani and Acrivos [8] modified the Zuzovski-Brenner method to circumvent the tedious algebra encountered in the calculations.

Runge [4] developed Rayleigh's method to coated elements where the geometry was composed of an array of tubes having the same core material as the matrix. Lurie and Cherkaev [9] showed that the bounds on the effective conductivity derived by Hashin and Shtrikman [10] for three-phase

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$$\mathbf{r}_n = h \left(n_1 \mathbf{a}_1 + n_2 \mathbf{a}_2 + n_3 \mathbf{a}_3 \right) \tag{1}$$

composites are realizable for coated sphere assemblages. Yu et al. [11] derived a relation for the effective properties of coated spheres in the dilute limit. Nicorovici et al. [12] extended Rayleigh's method for a simple cubic array of coated spheres and inspected the behaviour of the solution as a function of the properties of the core and shell of the sphere. Lu and Song [13] and Lu [14] developed a boundary collocation scheme to compute the effective conductivity of a simple array of multi-coated spheres and derived a relation for the effective conductivity of the random array of coated and multi-coated spheres which is correct to $O(F^2)$ where *F* is the total volume fraction.

In this report, we extend the Zuzovski–Brenner method to multi-coated spheres. We also present a scheme for deriving the effective conductivity of the system using two resistor models and compare the results with the exact solution. It should be noted that the formulation and the results for the thermal conductivity could be applied exactly to the seven other associated transport properties listed by Batchelor [15]. (These properties include electrical conductivity, dielectric permittivity, magnetic permeability, mobility, permeability of a porous medium, modulus of torsion in a cylindrical geometry and effective mass in bubbly flow.)

This paper is organized as follows. The next section describes the geometry under study. Section 3 reports the details of one approach [7] for calculating the effective conductivity of a cubic array of multi-coated spheres embedded in a homogeneous matrix of unit conductivity, and presents the exact solution for this system. Section 4 develops two resistor models for deriving the conductivity of the system. In Sect. 5, the results of the two resistor models are compared and evaluated with the exact solution. Finally, Sect. 6 summarizes the key findings of the study.

2 Geometric description

Consider a homogenous matrix with unit conductivity surrounding an array of composite spheres with a topology based upon the well-known cubic lattices, i.e. the simple cubic (SC) lattice, the body-centered cubic (BCC) lattice and the face-centered cubic (FCC) lattice. Each lattice point of a cubic array can be described by a lattice vector \mathbf{r}_n defined as:

	a_1	a ₂	a ₃
SC	e_x	e _y	<i>e</i> _z
BCC	$1/2(\boldsymbol{e}_x + \boldsymbol{e}_y - \boldsymbol{e}_z)$	$1/2(-\boldsymbol{e}_x+\boldsymbol{e}_y+\boldsymbol{e}_z)$	$1/2(\boldsymbol{e}_x-\boldsymbol{e}_y+\boldsymbol{e}_z)$
FCC	$1/2(e_x + e_y)$	$1/2(e_y + e_z)$	$1/2(e_x + e_z)$

TABLE 1 The basic vectors a_1 , a_2 and a_3 for three cubic lattices. The vectors e_x , e_y and e_z form an orthonormal basis in space

where *h* is the characteristic length to express all the distances in dimensionless form and n_1 , n_2 and n_3 are arbitrary integers. The three basic vectors a_1 , a_2 and a_3 belonging to a SC, BCC and FCC lattice are given in Table 1.

The radius of the core is determined by a_1 and the other coating layers of the multi-coated sphere are a_2, \ldots, a_{N-1} respectively, as shown in Fig. 1. The conductivity ratio between the phase i - 1 and i is assigned as $k_{i-1,i}$. With this consideration, the volume fraction occupied by the core and coating layers can take the following values:

$$f_i = \frac{4}{3\tau_0} \pi \left(a_i^3 - a_{i-1}^3 \right) \qquad (i = 1, \dots, N-1)$$
(2)

$$F = \sum_{i=1}^{N-1} f_i \tag{3}$$

where $\tau_0 = a_1 \cdot [a_2 \times a_3]$ refers to the non-dimensional volume of the unit cell, and has values of 1, 1/2 and 1/4 for SC, BCC and FCC lattices respectively. Note that $a_0 = 0$ is used here and in the subsequent relations in order to reduce the number of mathematical notations. If φ is an azimuthal angle measured from the plane of x_1x_2 and θ is a polar angle measured from the following relations apply:

$$x_1 = r \cos \theta, \quad x_2 = r \sin \theta \cos \varphi, \quad x_3 = r \sin \theta \sin \varphi$$
 (4)

3 The exact solution

The symmetry of the geometry makes the solution of the problem independent of the applied external gradient temperature, which is assumed to occur along the x_1 -axis. Since the arrays are periodic, we just study a unit cell of the systems. For the unit cell located at the origin, considering the general solution of the Laplace equation in spherical coordinates (r, θ , φ) and following Rayleigh [2], the temperature inside the layers may be given by:

$$T^{1} = \sum_{n=1}^{\infty} \sum_{m=0}^{m<\frac{1}{2}n} E_{nm}^{1} r^{2n-1} P_{2n-1}^{4m} (\cos \theta) \cos m\varphi$$
(5)

$$T^{i} = \sum_{n=1}^{\infty} \sum_{m=0}^{m<\frac{1}{2}n} [E^{i}_{nm}r^{2n-1} + F^{i}_{nm}r^{-2n}]P^{4m}_{2n-1}(\cos\theta)\cos m\varphi$$

(*i* = 2, ..., *N*) (6)

where $P_L^M(\cos\theta)$ represents the associated Legendre polynomial of degree L and order M. Zuzovski and Brenner [7]



FIGURE 1 The multi-coated sphere under study

proposed another suitable expression for the temperature inside the continuous phase:

$$T^N = x_1 + GH \tag{7}$$

where G is the differential operator, whose preferred form for a cubic array of spheres later was derived by Sangani and Acrivos [8] as:

$$G = \sum_{M=0}^{\infty} \sum_{m=0}^{m \le \frac{1}{2}M} \frac{2^{4m-1}}{(2n+1)!} A_{nm} \frac{\partial^{2n+1}}{\partial x_1^{2n+1}} \\ \times \left\{ \left(\frac{\partial}{\partial \xi}\right)^{4m} + \left(\frac{\partial}{\partial \eta}\right)^{4m} \right\} \quad (M = n+2m)$$
(8)

where

$$\xi = x_2 + ix_3, \ \eta = x_2 - ix_3 \tag{9}$$

and

$$H = \frac{1}{r} - \sigma + \frac{2\pi}{3}r^{2} + \sum_{n=2}^{\infty} \sum_{m=0}^{m \le \frac{1}{2}n} \frac{\epsilon_{m} (2n - 4m)!}{(2n + 4m)!} S_{nm} r^{2n} P_{2n}^{4m} (\cos \theta) \cos m\varphi$$
(10)

where ε_m represents the Neumann symbol (1 for m = 0 and 2 otherwise). The calculated values of constant array σ for the three cubic arrays [16] are 2.837297, 3.639233 and 4.584862 for SC, BCC and FCC respectively. The lattice sums have the following form:

$$S_{lm} = \sum_{n=1}^{\infty} |\mathbf{r}_n|^{-(2l+1)} P_{2l}^{4m} (\cos \theta_n) \cos m\varphi_n$$
(11)

where their values may be calculated by direct summation or in some region of *l* and *m* by approximate correlation [6, 17]. Note that θ_n and φ_n are polar and azimuthal angles measured from the lattice point *n*.

The unknown coefficients A in (8) can be determined by implementing boundary conditions at the surface of the core and coating layers:

$$T^{i-1} = T^i, \quad k_{i-1,i}\partial T^{i-1}/\partial n = \partial T^i/\partial n \quad r = a_{i-1}.$$
(12)

Using these conditions, the following result is obtained:

$$F_{nm}^{i} + L_{n}^{i} a_{i-1}^{4n-1} E_{nm}^{i} = 0 \qquad i \ge 2$$
(13)

where

т i

$$L_{n}^{i} = \frac{(2n-1)\left(k_{i-1,i}-1\right) + \left[2n\left(k_{i-1,i}+1\right)-1\right]L_{n}^{i-1}\left(a_{i-2}/a_{i-1}\right)^{4n-1}}{\left[(2n-1)\left(k_{i-1,i}+1\right)+1\right] + 2n\left(k_{i-1,i}-1\right)L_{n}^{i-1}\left(a_{i-2}/a_{i-1}\right)^{4n-1}}.$$
(14)

Calculating T^N by using the above expressions for H and G and comparing the resulting relation with (5), two linear equations can be found relating E_{nm}^N and F_{nm}^N to A_{nm} . These two equations combined with (13) yield a set of linear equations in the unknowns A_{nm} :

$$A_{nm} = L_{M+1}^{N} \frac{a_{N-1}^{4M+3}}{(2M+4m+1)!} \times \sum_{J=0}^{\infty} \sum_{j=0}^{j \le \frac{1}{2}J} \left[\frac{\lambda_1 \varepsilon_{q_1} (2p - 4q_1)!}{(2J - 4j + 1)} S_{pq_1} + \frac{\lambda_2 \varepsilon_{q_2} (2p - 4q_2)!}{(2J - 4j + 1)} S_{pq_2} \right] \times A_{J-2j,j} + \delta_{M0} L_1^N a_{N-1}^3 \left(1 + \frac{4\pi}{3\tau_0} A_{00} \right)$$
(15)

where

$$p = M + J + 1, \quad q_1 = m + j, \quad q_2 = |m - j|$$
 (16)

$$\lambda_{1} = \lambda_{2} = 0.25 \quad \text{if} \quad j \neq 0, m = 0 \\ \lambda_{1} = 0.5, \ \lambda_{2} = 1 \quad \text{if} \quad m = j \neq 0 \\ \lambda_{1} = \lambda_{2} = 0.5 \quad \text{otherwise}$$
 (17)

The effective thermal conductivity can be calculated using the following formula:

$$k_{\rm eff} = 1 + 4\pi A_{00}/\tau_0 \tag{18}$$

3.1 The explicit expression

As outlined by Manteufel and Todreas [18] different methods can be used to derive an explicit expression. By using linear truncation, Nicorovici et al. [12] have developed a relation of low order for the simple cubic lattice of coated spheres. For the truncation order L = 4, the solution of (15) is given as follows:

$$k_{\rm eff} = 1 - \frac{3F}{D} \tag{19}$$

where

$$D = -1/L_1^N + F + c_1 L_2^N F^{10/3} \frac{1 + c_4 L_3^N F^{11/3}}{1 - c_2 L_2^N F^{7/3}} + c_3 L_3^N F^{14/3} + c_5 L_4^N F^6 + c_6 L_5^N F^{22/3} + O(F^{25/3}) .$$
(20)

. . . .

The numerical constants are given in [8] and have been represented here in Table 2. These constants were verified as part of the current work. In (19), if the terms containing $c_4 - c_6$ are neglected, the formula of truncation order L = 3 can be obtained. By taking c_2 to c_6 equal to zero, the expression of the second-order approximation (L = 2), analogous to that of Lord Rayleigh [2] for the simple cubic lattice of solid spheres, will be obtained:

$$k_{\rm eff} = 1 - \frac{3F}{-1/L_1^N + F + c_1 L_2^N F^{10/3}} \,. \tag{21}$$

To test the correctness of (19) for multi-coated spheres, the effective thermal conductivity of coated and doubly coated spheres in the dilute limit was derived. Examination of (19) shows that it can be reduced to the following relation when $F \rightarrow 0$.

$$k_{\rm eff} = 1 + 3FL_1^N + O\left(F^2\right) \,. \tag{22}$$

Extending (22) for coated spheres gives the following result:

$$k_{\text{eff}} = 1 + 3FL_1^3 = 1 + 3F$$

$$\times \frac{(k_2 - 1) + (1 + 2k_2)}{(k_2 + 2) + 2(k_2 - 1)} \frac{[(k_1 - k_2) / (k_1 + 2k_2)] (a_1/a_2)^3}{[(k_1 - k_2) / (k_1 + 2k_2)] (a_1/a_2)^3}.$$
(23)

Likewise, the expression for doubly coated spheres is as follows:

$$k_{\rm eff} = 1 + 3FL_1^4 = 1 + 3F\frac{P+Q}{R+S}$$
(24)

where

$$P = (k_3 - 1) \{ (k_2 + 2k_3) + 2 (k_2 - k_3) [(k_1 - k_2) / (k_1 + 2k_2)] (a_1/a_2)^3 \}$$

$$Q = (1 + 2k_3) \{ (k_2 - k_3) (a_2/a_3)^3 + (k_3 + 2k_2) [(k_1 - k_2) / (k_1 + 2k_2)] (a_1/a_3)^3 \}$$

$$R = (k_3 + 2) \{ (k_2 + 2k_3) + 2 (k_2 - k_3) [(k_1 - k_2) / (k_1 + 2k_2)] (a_1/a_2)^3 \}$$

$$S = 2 (k_3 - 1) \{ (k_2 - k_3) (a_2/a_3)^3 + (k_3 + 2k_2) [(k_1 - k_2) / (k_1 + 2k_2)] (a_1/a_3)^3 \}.$$
(25)

	SC	BCC	FCC
c_1	1.3047	1.29×10^{-1}	7.529×10^{-2}
c_2	4.054×10^{-1}	7.642×10^{-1}	-7.410×10^{-1}
<i>c</i> ₃	7.231×10^{-2}	2.569×10^{-1}	4.195×10^{-2}
C4	2.305×10^{-1}	-4.129×10^{-1}	6.966×10^{-1}
C5	1.526×10^{-1}	1.13×10^{-2}	2.31×10^{-2}
c ₆	1.05×10^{-2}	5.62×10^{-3}	9.14×10^{-7}

TABLE 2 Numerical constants in explicit expression



4

FIGURE 2 Part of the resistor network. Original (a) and simplified arrangements (b, c)

Setting $a_3 = 1$ in (25) will reduce (23) and (24) to the same equations given by Yu et al. [11, 19] for the effective properties of coated and doubly coated spheres in the dilute limit.

3.2 The effect of azimuthal terms for a simple cubic array

The simple cubic array is of interest since it permits spheres to come closer to touching for low-volume-fraction systems as compared to any other arrangement. McPhedran et al. [6] have inspected the effect of azimuthal terms for a simple array of solid spheres. Due to the very small differences between the results, it was not clear whether considering azimuthal terms causes these differences or whether they are just numerical artifacts. This problem was again studied by Sangani and Acrivos [8], but due to the lack of convergence these authors were not able to obtain reliable results. Table 3 compares the calculated values of effective thermal conductivity for the most sensitive case of perfectly conducting spheres (meaning that the core and all coating layers are perfectly conducting) with the resulting values without azimuthal terms and with those obtained by McPhedran et al. [6]. For this calculation, the effective conductivity without azimuthal terms can be obtained by reducing (14) to the following expression:

$$\frac{A_{n,0}}{a_{N-1}^{4n+3}L_{n+1}^N} = \sum_{J=1}^{\infty} \binom{2n+2J}{2n+1} S_{2n+2J,0}A_{J-1,0} + \delta_{n,0} .$$
(26)

Mathematica 3.0 was used to solve the set of linear algebraic equations obtained from (15) and (26). 50 zonal unknowns and 50 azimuthal unknowns were considered in solving (15) and (26). All elements of the right-hand-side column vector have values of zero except for the first position, which has a value of unity. Multiplying the matrix of coefficients in the column vector of the results and comparing with the right-hand-side vector tests the correctness of the solution. The effective conductivity may be directly calculated from (18). It is clear from the results that there is a real (although small) effect associated with the azimuthal terms.

Resistor modeling

The unit cell can be considered to consist of an infinite series of resistors (Fig. 2a). Hsu et al. [20, 21] assumed for two-phase composite materials that all the resistors in the direction normal to the applied heat flow have infinite resistance. Thus they suggested a simplified configuration as shown in Fig. 2b. Another simplified configuration can be considered in which the resistors in the direction normal to the applied heat flow are perfectly conducting resistors, as shown in Fig. 2c. In the discussion that follows, the effective heat conductivity of the system will be derived using both methods, and the accuracy of the results and behaviour of the solutions will be compared with the exact solution. Since similar results could be expected for three cubic arrays, here only the simple cubic array is investigated.

4.1 *First resistor model*

Suppose that the unit cell is divided to N parallel regions, each composed of infinite infinitesimal parallel elementary volumes in annular cylinder geometry. Due to the symmetry of the problem and for the sake of simplicity, only a quarter of the unit cell needs to be considered, as depicted in Fig. 3a,b. An expression for the thermal conductivity of each element in these parallel regions (κ) can be

F	$k_{\rm eff}$	$k'_{ m eff}$	$k_{\rm eff}''$
0.30	2.3329	2.3326	2.333
0.40	3.2626	3.2612	3.262
0.50	5.8913	5.8875	5.891
0.510	6.7664	6.7623	_
0.520	8.8688	8.8644	_
0.523	11.671	11.666	_

TABLE 3 Effective thermal conductivity for a simple cubic array of perfectly conducting solid spheres. k'_{eff} corresponds to the solution without considering azimuthal terms and k''_{eff} are results of McPhedran et al. [6]

derived:

$$\frac{1/2}{\kappa} = \sum_{j=i}^{N-1} \frac{a_j \sqrt{1 - (a_i \sin \theta / a_j)^2 - a_{j-1} \sqrt{1 - (a_i \sin \theta / a_{j-1})^2}}}{k_j}$$
$$+ \frac{1}{2} - a_{N-1} \sqrt{1 - (a_i \sin \theta / a_{N-1})^2}$$
$$= \sum_{j=i}^{N-1} a_j \sqrt{1 - (a_i \sin \theta / a_j)^2} \left(\frac{1}{k_j} - \frac{1}{k_{j+1}}\right) + \frac{1}{2}$$
$$(i = 1, \dots, N-1) .$$
(27)

Clearly, for the last region, the thermal conductivity is equal to the unit value. The next step is to determine the relation for the equivalent thermal conductivity of each region (K_i) . Using (27) and remembering that all elements are in parallel, the following result is obtained:

$$K_{i} = \frac{1}{a_{i}^{2} - a_{i-1}^{2}} \times \int_{\theta_{i}}^{\pi/2} \frac{a_{i}^{2} \sin 2\theta d\theta}{\sum_{j=i}^{N-1} 2a_{j} \sqrt{1 - (a_{i} \sin \theta/a_{j})^{2} (1/k_{j} - 1/k_{j+1}) + 1}}$$
$$(i = 1, \dots, N-1)$$
(28)

where

$$\theta_i = \sin^{-1} \left(\frac{a_{i-1}}{a_i} \right) \,. \tag{2}$$

With the above procedures, the effective conductivity of the unit cell can be derived to be:

$$k_{\rm eff} = \sum_{i=1}^{N-1} \pi \left[a_i^2 - a_{i-1}^2 \right] K_i + 1 - \pi a_{N-1}^2 \tag{30}$$

4.2 Second resistor model

As shown in Fig. 4a,b, in the second method the unit cell is divided into infinite slices normal to the applied field. As for the first method, an infinite number of cells in N serial regions (i = 1, ..., N) are considered. For each slice, the equivalent conductivity may be specified as:

$$\kappa = \sum_{i=i}^{N} K_j s_j / S \quad (i = 1, \dots, N-1)$$
(31)

where

$$s_{j} = \frac{\pi}{4} \left\{ a_{j}^{2} \left[1 - \left(a_{i} \cos \theta / a_{j} \right)^{2} \right] -a_{j-1}^{2} \left[1 - \left(a_{i} \cos \theta / a_{j-1} \right)^{2} \right] \right\} \text{ if } j = i, \dots, N-1$$

$$s_{j} = 1/4 - \pi \left[a_{j-1}^{2} - \left(a_{i} \cos \theta \right)^{2} \right] \text{ if } j = N$$
(32)





FIGURE 3 Quarter unit cell considered in the derivation of the effective conductivity by using first resistor model: a 3-d view; and b 2-d view



FIGURE 4 Quarter unit cell under inspection in second resistor model: a 3-d view; and b 2-d view

mal conductivity of each region as:

$$K_{i} = \frac{a_{i} - a_{i-1}}{\int_{0}^{\theta_{i}} \frac{a_{i} \sin \theta d\theta}{\pi \left\{ k_{i}(a_{i} \sin \theta)^{2} + \sum_{j=i+1}^{N-1} k_{j} \left(a_{j}^{2} - a_{j-1}^{2} \right) - \left[a_{N-1}^{2} - (a_{i} \cos \theta)^{2} \right] \right\} + 1}}$$

(*i* = 1, ..., *N* - 1) (33)

where

$$\theta_i = \cos^{-1} \left(\frac{a_{i-1}}{a_i} \right) \,. \tag{34}$$

Consequently, the effective conductivity of the system is given as follows:

$$k_{\rm eff} = \frac{1}{\sum_{i=1}^{N-1} \frac{2(a_i - a_{i-1})}{K_i} + 1 - 2a_{N-1}}$$
(35)

5 Results and discussion

Figure 5 compares the resulting effective thermal conductivity values from the exact solution for the simple array of coated spheres with those obtained from resistor models. The total volume fraction (F) was selected to be 0.2 ($f_1 = 0.1$ and $f_2 = 0.1$) and the exact solution was obtained using (19), which should yield reasonable results in this limit [6]. All integrals in (28) and (33) were calculated using the Gauss-Legendre integration technique [22]. The results show that the first method always underestimates the exact solution except when the conductivity of the core and shell is equal to the matrix conductivity. This result is consistent with the finding of Hsu et al. [21] in their comparison of results for the effective thermal conductivity of non-touching solid circular cylinders. In reality, the infinite resistance assumption of the resistors in the normal direction to the applied field introduces a lower bound for the effective conductivity

of the system. The accuracy of the solution depends on the conductivity and volume fraction of the phases and geometry under consideration. In contrast to the first approach, the second method overestimates the solution except in the situation mentioned for the first resistor model. Thus, another bound is introduced for the conductivity of the system.

Of particular interest are the situations where the core and shell tend to their limiting conductivity values $(0, +\infty)$. The errors for other cases cannot be much higher than the error for the limiting cases, as is evident from Fig. 5. Thus, the follow-



FIGURE 5 Logarithmic plot of the effective conductivity for the simple cubic array of coating spheres as a function of the conductivity of the core and shell. Despite of the accuracy, the first resistor model is unable to show the same behaviour as the exact solution when the shell is perfectly conducting. The same condition applies for the second resistor model when the shell is perfectly insulating

ing section discusses the ability of the models to predict these limiting cases.

Relatively good results can be expected from the first model for the case when both the core and shell have a large conductivity value. Here the accuracy of the solution is tested for a perfectly conducting core and coating layer. Since this case is equal to an array of perfectly conducting solid spheres of radius $a_{1*} = a_2$, it is easier to investigate the solution for the alternative system. For this case the effective thermal conductivity can be derived as follows:

$$K_{1*} = \int_{0}^{\pi/2} \frac{\sin(2\theta)}{1 - 2a_2\cos\theta} = -\frac{1}{a_2} - \frac{1}{2a_2^2}\ln(1 - 2a_2) \,\mathrm{d}\theta \quad (36)$$

$$k_{\rm eff} = -\frac{\pi}{2} \ln \left(1 - 2a_2 \right) - \left(\pi a_2^2 + \pi a_2 - 1 \right) \,. \tag{37}$$

As was predicted by Batchelor and O'Brien [23], for nearly touching perfectly conducting spheres, the effective thermal conductivity satisfies the following relation:

$$k_{\rm eff} = -C_1 \ln (1 - 2a_2) - C_2 \qquad (a_2 \to 1/2)$$
 (38)

where C_1 and C_2 are two positive constants. The constant C_1 was derived by Batchelor and O'Brien [23] to be $\pi/2$, and the second constant has been calculated by Sangani and Acrivos [8] to be about 0.69. The first resistor model gives the same value for C_1 but the suggested value for C_2 is equal to 1.356, which is almost twice the value given by Sangani and Acrivos [8]. For this situation, the second resistor model provides the following simple expression:

$$k_{\rm eff} = \frac{1/2}{1/2 - a_2} \,. \tag{39}$$

Figure 6 compares the results of the two-resistor model with the data obtained by exact solution and the results of the lower bound given by Hashin and Shtrikman [10].

Since dispersed phases have small conductivity values, a second limiting case can be presented for such dispersed systems. The results of this system have been calculated for the condition where the core and shell are perfectly insulating (an array of perfectly insulating solid sphere of radius placed in a matrix of unit conductivity). It can be seen from Fig. 5 that the second model predicts more accurate results in this situation. From (33) and (35), the following relations for the second model can be derived:

$$K_{1*} = \int_{0}^{\pi/2} \frac{\sin\theta}{1 - \pi a_2^2 \sin^2\theta} = \frac{a_2 \pi \sqrt{1/\pi - a_2^2} \,\mathrm{d}\theta}{\arctan\left(a_2/\sqrt{1/\pi - a_2^2}\right)} \tag{40}$$

$$k_{\mathrm{eff}} = \frac{\pi \sqrt{1/\pi - a_2^2}}{1 - \pi a_2^2 + 1 - \pi a_2^2}$$

$$k_{\rm eff} = \frac{1}{2 \arctan\left(a_2/\sqrt{1/\pi - a_2^2}\right) + (1 - 2a_2) \pi \sqrt{1/\pi - a_2^2}}$$
(41)

Also, for the first model one may find:

$$k_{\rm eff} = 1 - \pi a_2^2 \,. \tag{42}$$

A comparison between the exact solution with the predicted results based on the two resistor model and two bounds given in [10] for the case of perfectly insulating spheres is shown in Fig. 7.

The results of the resistor models for two rested positions $(k_1 = +\infty, k_2 = 0 \text{ and } k_1 = 0, k_2 = +\infty)$ are examined in a different way. As can be seen from (15) L_n^N plays an important role in the response of the system to the applied field. Due to the shape of mathematical expression (14), for some cases a different series of dispersed phases may exist that gives the same value for L_n^N . Nicorovici et al. [12] have studied the behaviors of coated spheres. The following discussion considers the case of multi-coated spheres for three scenarios.

1)
$$k_{i^*-1} = k_{i^*}$$
 $(2 \le i^* \le N - 1)$. (43)

After two successive applications of (13) for i^* and $i^* + 1$, the resulting expression for $L_n^{i^*+1}$ is:

$$L_{n}^{i^{*}+1} = \frac{\left(2n-1\right)\left(k_{i^{*}-1,i^{*}+1}-1\right)}{\left[2n\left(k_{i^{*}-1,i^{*}+1}+1\right)-1\right]L_{n}^{i^{*}-1}\left(a_{i^{*}-2}/a_{i^{*}}\right)^{4n-1}}{\left[\left(2n-1\right)\left(k_{i^{*}-1,i^{*}+1}+1\right)+1\right]} + 2n\left(k_{i^{*}-1,i^{*}+1}-1\right)L_{n}^{i^{*}-1}\left(a_{i^{*}-2}/a_{i^{*}}\right)^{4n-1}}$$
(44)

Therefore L_n^N will not change if these two phases are unified and considered as a unique phase with volume fraction $4\pi \left(a_{i^*}^3 - a_{i^*-2}^3\right)/3\tau_0$. If the core and all coating layers have the same conductivity, the problem can be reduced to the simple array of solid spheres of radius a_{N-1} immersed in a matrix of unit conductivity.

$$L_n^N = \frac{k_1 - 1}{k_1 + 2n/(2n - 1)} \,. \tag{45}$$



FIGURE 6 Comparison between the results of the first and second resistor models with the exact solution and the results of the lower bound given in [10] for perfectly conducting phases



FIGURE 7 Comparison between the predictions of the first and second resistor models with the exact solution and the results of two bounds given in [10] for perfectly insulating spheres

It can be shown from (27) and (31) that both resistor models also demonstrate the same behaviour.

2) $k_{i_{\text{max}}} = 0$, where i_{max} is the largest coating layer which has conductivity equal to zero. For this case (44) reduces to a simplified expression:

$$L_n^{i_{\max}+1} = -\frac{2n-1}{2n} \,. \tag{46}$$

This result implies that the phases under i_{max} have no effect in calculating the and in fact layer i_{max} cuts the relation between the phases $i > i_{\text{max}}$ and $i < i_{\text{max}}$. The first resistor model also shows the same behaviour. For this case, (28) yields the following result:

$$K_i = 0 \qquad (i \le i_{\max}) \ . \tag{47}$$

Thus, these phases play the same role as for the exact solution. In contrast, it can be seen from (33) that the second model does not demonstrate equal behaviour and the results of this model have more errors in this situation.

3) $k_{i_{\text{max}}} = +\infty$, where i_{max} is the largest coating layer which is perfectly conducting. Consequently,

$$L_n^{i_{\max}+1} = 1. (48)$$

Thus, this phase makes the phases under the phase i_{max} irrelevant in the response of the system to the applied field. In this situation, the second resistor model illustrates similar behaviour. All the region conductivities become infinite:

$$K_i = +\infty \qquad (i \le i_{\max}) \ . \tag{49}$$

Therefore, they do not have any effect on the calculation of the effective thermal conductivity (i.e. (35)). However, the first resistor model is unable to predict this behaviour and the error tends to increase for this condition.

6 Concluding remarks

The method devised by Zuzovski and Brenner [7] has been extended to enable the calculation of the effective conductivity of systems of multi-coated spheres consisting of SC, BCC and FCC unit cells. An explicit expression for the effective thermal conductivity to $O(F^9)$ has been presented. Two resistor models have been developed to obtain alternative solutions to the problem. By inspection of the results of the resistor model and comparison with the results of the exact solution, it was found that the results of these models constitute two bounds for the exact solution. Some scenarios were investigated to identify and discuss situations in which one of these models predicts better results.

The use of a more complex method of analysis, such as combinational theory, should help to improve the accuracy of the calculations and provide a more thorough explanation of the physical behaviors of the system. The development of a method based on combinational theory and its application to the system studied in this work will be the focus of a future investigation in this area.

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III

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The effective conductivity of three-phase composite materials with circular cylindrical inclusions

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Abstract

We extend the Rayleigh method for the calculation of the effective conductivity to threephase composite materials. The materials under study consist of two types of circular cylinders in a periodic arrangement embedded in a matrix. Highly accurate values for lattice sums were obtained using algorithms which have been recently developed. A series of explicit formulations, which are used to facilitate the calculation of the effective conductivity of the composites under study, are reported. We also perform a series of numerical calculations to study the behavior of these composites.

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1. Introduction

Multi-phase heterogeneous systems can be found in a wide range of practical processes and are of considerable technological importance. Colloidal dispersions, emulsions, solid rocket propellant, oil-filled porous rocks, concrete, reinforced materials are a few examples of such systems. To handle these systems optimally one needs to know how they behave at the macroscopic level; therefore a great deal of interest has been focused on relating the microstructural and macroscopic properties of these systems, such as the effective thermal, or electrical, conductivity [1,2], dielectric permittivity [3,4] and permeability of a porous medium [5].

One common type of these systems consists of a matrix and a series of dispersed phases. Of these systems those with circular cylinders were among the first to be studied by researchers for deriving the effective conductivity, as is the case in Rayleigh's studies [1]. Rayleigh took into account a rectangular array of circular cylinders and showed that to completely relate the microstructural and macroscopic properties, the effect of interaction between the cylinders should be taken into account. This fact was later extensively used for obtaining more accurate analytical relations for calculating the effective conductivity of these composite systems [6,7]. Recently, some attempts have been made to generalize the study by developing formulations for multi-phase cases [8]. Clearly, this is an important task since the behavior demonstrated by multi-phase systems can be completely different from that understood on the basis of two-phase systems and further investigation is necessary.

In this research, we are concerned with the problem of calculating the effective conductivity of three-phase periodic structures composed of two types of circular cylinders. The unit cell of the composites under study is a rectangular cylinder with a circular cylinder of type one at the center and a circular cylinder of type two at each corner, as depicted in figure 1. The phases can be solid or stagnant fluid. The configuration of the geometry selected for study makes it possible to construct many periodic structures, which can be widely found in literature. In what follows, we first develop the Rayleigh method to the three-phase composites. Then, we verify the algorithm, comparing our results with some existing results. Finally, we inspect the behavior of the systems. We shall use terms and notations appropriate to the case of thermal conduction for convenience. It is worth noting that the results of this study can be applied to many transport properties besides thermal or electrical conductivity [9,10].



Figure 1. The unit cell of the three-phase composites under study. Cylinders of the same type have a distance equal to a unity in the *x*-direction and *b* in the *y*-direction.

2. Governing equations

Suppose that the origin of cartesian coordinates has been placed at the centre of a cylinder of type one in a unit cell of the system in which the x- and y-axes are parallel to the sides of the unit cell (see figure 1). Furthermore, assume that the matrix of the composite under study has unit conductivity and the periodicity of the system in the y-direction is equal to a unity for greater generality. Applying these conditions, we denote the conductivity of the cylinders by k_1 and k_2 , the radiuses a_1 and a_2 and the volume fractions f_1 and f_2 for the cylinders of type one and two, respectively. Also the periodicity of the system in the x-direction will be denoted by b.

For the case $b \neq 1$, the effective conductivity of the system in the *x*-direction (parallel) would be different from that of the *y*-direction (perpendicular) and one should calculate both these conductivities. In order to simplify the presentation without loss of generality herein, we only consider the parallel direction. The necessary comments for the perpendicular direction shall be outlined in a separate section.

Let us apply a uniform temperature gradient of unit magnitude externally to the system along the *x*-axis in the negative direction. By taking the center of any cylinder of type i (i = 1 or 2) as the origin of polar coordinates (r, q), the temperature within that cylinder and outside it through the matrix can be given as

$$T_{i}(r,\boldsymbol{q}) = C_{0,i} + \sum_{n=1}^{\infty} C_{2n-1,i} r^{2n-1} \cos(2n-1)\boldsymbol{q}$$
(1)

$$T_{m,i}(r,\boldsymbol{q}) = A_{0,i} + \sum_{n=1}^{\infty} \left(A_{2n-1,i} r^{2n-1} + B_{2n-1,i} r^{-2n+1} \right) \cos\left(2n-1\right) \boldsymbol{q} , \qquad (2)$$

where the set of coefficients $C_{2n-1,i}$, $A_{2n-1,i}$ and $B_{2n-1,i}$ are unknowns to be determined. The periodicity of the system implies that these coefficient be exactly the same for all cylinders of the same type. $C_{0,i}$ and $A_{0,i}$ express the average of the temperature within the cylinder and outside it, respectively. Thus, they are exactly the same only for cylinders of the same type placed in a column normal to the applied field. In equations (1) and (2), also note that the cosines of the even multiples have been ignored. This is because of anti-symmetric behavior of the temperature around q = p/2, where q is measured from the parallel direction. At the surface of the cylinders, the temperature and the normal component of heat flux are continuous, i.e.,

$$T_i = T_{m,i}, \quad k_i \frac{\partial T_i}{\partial r} = \frac{\partial T_{m,i}}{\partial r} \quad r = a_i$$
 (3)

By applying the above-mentioned boundary conditions, we can obtain

$$A_{2n-1,i} = \frac{B_{2n-1,i}}{g_i a_i^{4n-2}} \tag{4}$$

 $C_{2n-1,i} = \frac{B_{2n-1,i}}{\mathbf{c} \cdot a_{\cdot}^{4n-2}},$

where

$$\boldsymbol{g}_{i} = \frac{1-k_{i}}{1+k_{i}} \qquad \boldsymbol{c}_{i} = \frac{1-k_{i}}{2}$$
(6)

(5)

For the case of non-conducting cylinders, $k_i = 0$, we have $g_i = 1$ and $c_i = 0.5$. Also, applying perfectly conducting cylinders, $k_i = \infty$, implies that $g_i = -1$ and $c_i = -\infty$. The unknown coefficients still cannot be determined since the relations given in equations (4) and (5) do not provide a complete set of equations in terms of the unknowns; we require a further series of relations between the coefficients. For this purpose, we employ Rayleigh's strategy which is based on the fact that at any point the temperature may be regarded due to external sources and multiple sources placed at the center of the cylinders. By examination of temperature function (2) written for a cylinder, we find that terms with radius raised to a positive power cannot be due to sources placed at the center of that cylinder since they increase when *r* increases; therefore they stem from the external field and sources originated from the center of other cylinders. As a result, we can write

$$A_{0,i} + \sum_{n=1}^{\infty} A_{2n-1,i} r^{2n-1} \cos(2n-1) \boldsymbol{q} = x + \sum_{j \neq 0} \sum_{n=1}^{\infty} \frac{B_{2n-1,i}}{r_{j,i}^{2n-1}} \cos(2n-1) \boldsymbol{q}_{j,i} + \sum_{j} \sum_{n=1}^{\infty} \frac{B_{2n-1,2-\boldsymbol{d}_{12}}}{r_{j,2-\boldsymbol{d}_{12}}^{2n-1}} \cos(2n-1) \boldsymbol{q}_{j,2-\boldsymbol{d}_{12}},$$
(7)

where $r_{j,i}$ and $q_{j,i}$ are measured from the center of cylinder *j* situated in the array of cylinders of type *i*. As is specified in equation (7), in the sum over the cylinders of type *i*, all the cylinders, except the cylinder under study (*j*=0), should be taken into account but in the sum over the cylinders of the other type, all the cylinders are to be considered without any exception. The above expression can be considered as the real part of the following relation:

$$A_{0,i} + \sum_{n=1}^{\infty} A_{2n-1,i} \left[x - \mathbf{x}_{0,i} + i(y - \mathbf{h}_{0,i}) \right]^{2n-1} = x + iy + \sum_{j \neq 0} \sum_{n=1}^{\infty} B_{2n-1,i} \left[x - \mathbf{x}_{0,i} - \mathbf{x}_{j,i} + i(y - \mathbf{h}_{0,i} - \mathbf{h}_{j,i}) \right]^{-2n+1} + \sum_{j} \sum_{n=1}^{\infty} B_{2n-1,2-\mathbf{d}_{12}} \left[x - \mathbf{x}_{0,i} - \mathbf{x}_{j,2-\mathbf{d}_{12}} + i(y - \mathbf{h}_{0,i} - \mathbf{h}_{j,2-\mathbf{d}_{12}}) \right]^{-2n+1},$$
(8)

where $\mathbf{x}_{j,i}$ and $\mathbf{h}_{j,i}$ are the coordinates of cylinder *j* of type *i* in the coordinate system (*x*, *y*). Now we perform successive differentiation with respect to *x* on both sides of the above equation and evaluate the results at the center of the cylinder under study $(\mathbf{x}_{0,i}, \mathbf{h}_{0,i})$. After applying equation (4), the process yields the following set of linear algebraic equations in the unknowns $B_{2n-1,i}$ (*i* = 1,2):

$$\frac{B_{2n-1,i}}{\boldsymbol{g}_{i}a_{i}^{4n-2}} + \sum_{m=1}^{\infty} \binom{2n+2m-3}{2n-1} S_{2n+2m-2,1}B_{2m-1,i} + S_{2n+2m-2,2}B_{2m-1,2-\boldsymbol{d}_{12}} = \boldsymbol{d}_{n1}, \qquad (9)$$

where $S_{2l,i} = \sum_{j \neq 0} (\mathbf{x}_{j,i} + i\mathbf{h}_{j,i})^{-2l}$ are lattice sums over cylinders of type *i*. Solving equation (9)

and using equations (4) and (5) we can obtain all the unknown coefficients and as result the temperature functions. Since considering $B_{2n-1,i}$ (i = 1,2) for a sufficiently large *n* has no significant effect on the values of the temperature functions, in practice, the system of equations (9) is truncated.

3. Determining the effective conductivity of the system

Based on Fourier's law, the effective conductivity of the system can be derived using the following formula [11]:

$$\left\langle \boldsymbol{F} \right\rangle = -k_{e} \left\langle \nabla T \right\rangle \tag{10}$$

where $\langle \mathbf{F} \rangle = (1/V_{cell}) \int_{V_{cell}} \mathbf{F} dV$ and $\langle \nabla T \rangle = (1/V_{cell}) \int_{V_{cell}} \nabla T dV$ are the average heat flux and temperature gradient over the unit cell, respectively. To proceed, let us decompose the average heat flux as the following:

$$\left\langle \boldsymbol{F} \right\rangle = \frac{1}{V_{cell}} \left[\int_{V_1} \boldsymbol{F} \, dV + \int_{V_2} \boldsymbol{F} \, dV + \int_{V_m} \boldsymbol{F} \, dV \right] \tag{11}$$

whence

$$\left\langle \boldsymbol{F} \right\rangle = \frac{1 - k_1}{V_{cell}} \int_{V_1} \nabla T_1 dV + \frac{1 - k_2}{V_{cell}} \int_{V_2} \nabla T_2 dV - \frac{1}{V_{cell}} \int_{V_{cell}} \nabla T dV \quad , \tag{12}$$

where V_1 , V_2 and V_m are the volumes of cylinders of type one and two and the matrix placed in the unit cell, respectively. After performing the integrals (see appendix A), from equation (12) we may find

$$\left\langle \boldsymbol{F} \right\rangle = \frac{2\boldsymbol{p}B_{1,1}}{V_{cell}} \mathbf{i} + \frac{2\boldsymbol{p}B_{1,2}}{V_{cell}} \mathbf{i} \cdot \left\langle \nabla T \right\rangle$$
(13)

Taking into account that $\langle \nabla T \rangle = \mathbf{i}$ and $V_{cell} = b$, the final result for the effective conductivity from equations (13) and (10) can be given as

$$k_{e} = 1 - 2\boldsymbol{p} (B_{1,1} + B_{1,2}) / b = 1 - 2f_{1}\boldsymbol{g}_{1}A_{1,1} + 2f_{2}\boldsymbol{g}_{2}A_{1,2}$$
(14)

As can be seen from equation (14), knowing $B_{1,1}$ and $B_{1,2}$ is enough for obtaining the effective conductivity of the system. The term $2p(B_{1,1} + B_{1,2})/b$ was produced because of the presence of the inclusions in the matrix. It can be positive (impairing case), negative (enhancing case) or equal to zero. For the case in which N types of cylinders are placed in the unit cell, the effective conductivity of the system can be obtained using

$$k_{e} = 1 - 2\mathbf{p} \sum_{i=1}^{N} B_{1,i} / V_{cell}$$
(15)

4. The effective conductivity for the perpendicular direction

For deriving the effective conductivity in this direction in order to make it similar with the foregone relations, we rotate the system through an angle of $\mathbf{a} = \mathbf{p}/2$. If we follow the above-mentioned procedures for the parallel direction, we get

$$\frac{B'_{2n-1,i}}{\boldsymbol{g}_{i}a_{i}^{4n-2}} + \sum_{m=1}^{\infty} \binom{2n+2m-3}{2n-1} S'_{2n+2m-2,1}B'_{2m-1,i} + S'_{2n+2m-2,2}B'_{2m-1,2-\boldsymbol{d}_{i2}} = \boldsymbol{d}_{n1}, \quad (16)$$

where $S'_{2l,i}$ are lattice sums over cylinders of type *i* in this new position. It can be proved that $S'_{2l,i} = (-1)^l S_{2l,i}$ for l > 1 and also $S'_{2,i} = 2\mathbf{p}/b - S_{2,i}$ (see Refs. [6] and [12]). The effective conductivity of the system may be obtained using the similar formula as (14), i.e.,

$$k'_{e} = 1 - 2\mathbf{p} \left(B'_{1,1} + B'_{1,2} \right) / b \tag{17}$$

Since the composite under study is a 2-D structure, through a methodology (see appendix B) we can show that the effective conductivity in the perpendicular direction has been linked to that in the parallel direction using the well-know reciprocal theorem of Keller [13-15], i.e.,

$$k_{e}(k_{1},k_{2},1) \times k_{e}'(1/k_{1},1/k_{2},1) = 1$$
(18)

5. Explicit solutions

For low-volume fractions or when the conductivity of the cylinders is small, considering a few numbers of $B_{2n-1,1}$ and $B_{2n-1,2}$ in the process of solving (9) may yield reasonable results for $B_{1,1}$ and $B_{1,2}$ and as a result, based on equation (14), for the effective conductivity. It is more useful that, within these boundaries, we manage to derive an explicit relation for the effective conductivity of the system. Based on the method used for truncating (for example square or triangular manner) and on the number of the unknowns taken into account, one may obtain different expressions. If we truncate equation (9) in a triangular manner and keep only the coefficient $B_{1,i}$ and $B_{3,i}$ (i = 1, 2), we find

$$\frac{B_{1,1}}{g_{1}a_{1}^{2}} + S_{2,1}B_{1,1} + S_{2,2}B_{1,2} + 3S_{4,1}B_{3,1} + 3S_{4,2}B_{3,2} = 1$$

$$\frac{B_{3,1}}{g_{1}a_{1}^{6}} + S_{4,1}B_{1,1} + S_{4,2}B_{1,2} = 0$$

$$\frac{B_{1,2}}{g_{2}a_{2}^{2}} + S_{2,1}B_{1,2} + S_{2,2}B_{1,1} + 3S_{4,1}B_{3,2} + 3S_{4,2}B_{3,1} = 1$$

$$\frac{B_{3,2}}{g_{2}a_{2}^{6}} + S_{4,1}B_{1,2} + S_{4,2}B_{1,1} = 0$$
(19)

Deriving $B_{1,1}$ and $B_{1,2}$ from the above equation and using equation (14), we can obtain the effective conductivity of the system in an explicit relation, i.e.,

$$k_{e} = 1 - \frac{2f_{1}}{(\boldsymbol{l}_{1}\boldsymbol{l}_{2} - \boldsymbol{x}_{1}\boldsymbol{x}_{2})/(\boldsymbol{l}_{2} - \boldsymbol{x}_{2})} - \frac{2f_{2}}{(\boldsymbol{l}_{1}\boldsymbol{l}_{2} - \boldsymbol{x}_{1}\boldsymbol{x}_{2})/(\boldsymbol{l}_{1} - \boldsymbol{x}_{1})},$$
(20)

with

$$\boldsymbol{l}_{i} = \frac{1}{\boldsymbol{g}_{i}} + c_{1}f_{i} - c_{2}\boldsymbol{g}_{i}f_{i}^{4} - c_{3}\boldsymbol{g}_{2-\boldsymbol{d}_{i2}}f_{i}f_{2-\boldsymbol{d}_{i2}}^{3}$$
(21)

$$\mathbf{x}_{i} = c_{4}f_{i} - c_{5}\left(\mathbf{g}_{i}f_{i}^{4} + \mathbf{g}_{2-\mathbf{d}_{12}}f_{i}f_{2-\mathbf{d}_{12}}^{3}\right),$$
(22)

where $c_1 = S_{2,1}b/p$, $c_2 = 3(b/p)^4 S_{4,1}^2$, $c_3 = 3(b/p)^4 S_{4,2}^2$, $c_4 = S_{2,2}b/p$ and $c_5 = 3S_{4,1}S_{4,2}(b/p)^4$. One may leave the higher orders and obtain a simpler relation

$$k_{e} = 1 - \frac{2f_{1}}{w/u_{2}} - \frac{2f_{2}}{w/u_{1}},$$
(23)

where

$$\boldsymbol{w} = (\mathbf{1}/\boldsymbol{g}_1 + c_1 f_1) (\mathbf{1}/\boldsymbol{g}_2 + c_1 f_2) - c_4^2 f_1 f_2$$
(24)

$$\boldsymbol{u}_{i} = 1/\boldsymbol{g}_{i} + (c_{1} - c_{4})f_{i}$$
(25)

For the case of uniform cylinders $(f_1 = f_2 = f^*, g_1 = g_2 = g^*)$ from equation (20), we obtain the following formula:

$$k_{e} = 1 - \frac{2F}{1/g^{*} + F - dg^{*}F^{4}}, \qquad (26)$$

where $F = f_1 + f_2 = 2f^*$ is the total volume fraction and $d_1 = 3(b/\mathbf{p})^4 (S_{4,1} + S_{4,2})^2 / 16$. We have listed c_1, \ldots, c_5 for the case $b = \sqrt{3}$ in table 1, calculating highly accurate values for lattice sums using integral representation technique [16]. For this case, d = 0 as we expected [6]. Note that equation (20) can also be applied for the perpendicular direction if we calculate the coefficients for this direction. In table 1 we have also reported these values.

Table 1. The calculated values for $c_1, ..., c_5$ used in the analytical formula (20) 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	
<i>c</i> ₂	1.310523128	1.310523128	
<i>c</i> ₃	1.310523128	1.310523128	
<i>c</i> ₄	1.812981866	0.187018134	
c_5	-1.310523128	-1.310523128	

6. Results and discussions

Before starting the discussion on the results of the three-phase system, it is helpful to verify the validity of the extension to the three-phase system.

Table 2. The calculated lattice sums over cylinders of type one $(S_{n,1})$ and two $(S_{n,2})$ for the case $b = \sqrt{3}$ for $n \le 40$. The fourth column shows that the sum of these two lattice sums is zero for cases $n \ne 2$ and $n \ne 6m$ $(m = 1, \dots, \infty)$.

п	${S}_{n,1}$	<i>S</i> _{<i>n</i>,2}	$S_n = S_{n,1} + S_{n,2}$
2	0.3392133718630	3.2883853566054	3.6275987284684
4	2.1744038488973	-2.1744038488973	0.0
6	-2.0154171446150	-3.8476145488104	-5.8630316934253
8	2.0262994706141	-2.0262994706141	0.0
10	-1.9919685438111	1.9919685438111	0.0
12	2.0041914554050	4.0054485162926	6.0096399716977
14	-1.9990818777902	1.9990818777902	0.0
16	2.0003051627592	-2.0003051627592	0.0
18	-1.9999213768630	-3.9997969795075	-5.9997183563705
20	2.0000338688335	-2.0000338688335	0.0
22	-1.9999887079801	1.9999887079801	0.0
24	2.0000041209456	4.0000075266341	6.0000116475798
26	-1.9999987455894	1.9999987455894	0.0
28	2.0000004181529	-2.0000004181529	0.0
30	-1.9999998662038	-3.9999997212317	-5.9999995874356
32	2.000000464611	-2.000000464611	0.0
34	-1.9999999845130	1.9999999845130	0.0
36	2.000000052497	4.0000000103247	6.000000155744
38	-1.99999999982792	1.99999999982792	0.0
40	2.000000005736	-2.000000005736	0.0

We perform a series of calculations for two-phase composites with uniform cylinders arranged either in square (b = 1) or hexagonal orders $(b = \sqrt{3})$ and then compare the results with those reported by Perrins et al. [6]. The two-phase cases can be constructed from the three-phase one simply by applying $f_1 = f_2$ and $k_1 = k_2$. For this purpose highly accurate values for lattice sums over cylinders of type one and two were derived and equation (9) (i=1,2) was solved numerically using LU decomposition method [17]. Taking into account 100 unknowns of $B_{2n-1,1}$ and $B_{2n-1,2}$ gives us a measure of obtaining accurate results for all the volume fractions and conductivities considered (see Ref. [6]). Table 2 shows a part of the calculated lattice sums used in the procedure of the solutions. In figure 2 the results are compared for both the square and hexagonal arrays for the most challenging case, i.e., the case of perfectly conducting cylinders. As can be seen for all the values of volume fractions, the results of two studies are in excellent agreement.



Figure 2. The effective conductivity of two-phase composites with mono-sized perfectly conducting circular cylinders arranged in square and hexagonal orders. *F* shows the volume fraction of the cylinders. The solid lines are the results of Perrins et al. [6] and the dotted lines are those obtained by solving the governing equations of the three-phase composites.

Figure 3 shows a typical result for the effective conductivity of the system for both the parallel and perpendicular directions. The volume fractions are $f_1 = 0.4$ and $f_2 = 0.4$, and the periodicity in the x-direction was supposed to be $b = \sqrt{3}$. For deriving the conductivity of the system in the perpendicular direction we can either solve equation (16) and apply equation (17) or, alternatively, use the Keller theorem for this purpose. Through a careful examination of this figure, it appears that increasing or decreasing the conductivity of the both types of cylinders may enhance or diminish the conductivity of the system, respectively, which is obvious and remains correct for both directions. Furthermore, the system demonstrates higher effective conductivity in the perpendicular direction. This behavior is a consequence of the

rectangular shape of the unit cell which provides a more (less) important role for the cylinders with lower conductivity in the parallel (perpendicular) direction.



Figure 3. The contours of the effective conductivity in the parallel and perpendicular directions for the case of equal sized cylinders. $f_1 = 0.4$, $f_2 = 0.4$ and $b = \sqrt{3}$.

Interestingly, for the case of mono-sized cylinders with $k_1 = \infty$ and $k_2 = 0$, increasing the volume fraction of the cylinders causes the conductivity of the system to approach zero in the parallel direction and approach infinity in the perpendicular direction (see table 3). When perfectly insulating cylinders touch each other, they form a barrier which prevents heat flow in the parallel direction (Note that for the case of spherical inclusions this behavior does not hold true since heat can pass through the gaps between spheres). This behavior can also be observed for all systems for which b > 1. For the case b = 1, however, the system is isotropic and the same results can be expected for both directions. For this case, the same type cylinders are not able to touch each other and a limited value for the effective conductivity of the system is simply a unity. This result can be confirmed using the Keller theorem as follows: Since the system is isotropic and interchange between the material of the cylinders keeps the system unchanged, using equation (18) we can get

$$k_{e}(k,1/k,1) \times k_{e}'(1/k,k,1) = k_{e}(k,1/k,1) \times k_{e}(k,1/k,1) = 1$$
(27)

Table 3. The effective conductivity in the parallel and perpendicular directions, with respect to the total volume fraction (*F*). The cylinders are mono-sized, $k_1 = 0$, $k_2 = \infty$ and $b = \sqrt{3}$ (b > 1).

F	k_{e}	k'_{e}
0.1	0.983871	1.01639
0.2	0.937008	1.06723
0.3	0.863599	1.15794
0.4	0.769417	1.29969
0.5	0.660207	1.51468
0.6	0.539863	1.85232
0.7	0.408730	2.44660
0.8	0.261159	3.82908
0.9	3E-7	19.253
$\boldsymbol{p}/(2\sqrt{3})$	0	-

Considering k = 0 proves our case. Sculgasser [18] has shown that in a three-phase system with interchangeable phases (see figure 4), when one of the phases has conductivity equal to k and the two remaining phases are perfectly conducting and non-conducting, the effective conductivity of the system is k. From the above results, it is clear that it is not necessary for the first phase to be interchangeable, and it can simply be a matrix.



Figure 4. The three-phase structure investigated by Schulgasser et al. [18]. All the phases are interchangeable.

For the case b = 1 with non-equal sized cylinders, if $f_i \leq \frac{\mathbf{p}}{4} (\sqrt{2} - 1)^2$, $f_{2-\mathbf{d}_{i2}}$ can be increased freely to the touching value limit and the effective conductivity of the system can approach infinity or zero, depending on the conductivity of the touching cylinders.

Figure 5 shows the results of the effective conductivity for a system with a lower total volume fraction, i.e., $f_1 = 0.4$ and $f_2 = 0.2$. *b* as before is equal to $\sqrt{3}$. Comparing with figure 4 we can see that the case $k_1 = k_2 = 1$ is the only situation in which both systems for the given conductivities present the same effective conductivity. In this situation $g_1 = g_2 = 0$, which leads to $B_{1,1} = B_{1,2} = 0$, and as a result, $k_e = 1$. Figures 3 and 5 also show that having cylinders with conductivities equal to the conductivity of the matrix is not the only condition for the effective conductivity to be equal to the conductivity of the matrix. In fact, this case is a special state of the following general situation:

$$B_{1,1} + B_{1,2} = f_1 g_1 A_{1,1} + f_2 g_2 A_{1,2} = 0$$
(28)

The importance of the situation $k_1 = k_2 = 1$ ($B_{1,1} = B_{1,2} = 0$) is that it is independent of the values of f_1 , f_2 and b, and for all theses situations, we would find that $k_e = 1$, which is physically obvious. This behavior does not hold for the other values of the conductivities.



Figure 5. The contours of the effective conductivity in the parallel and perpendicular directions for the case of unequal sized cylinders. $f_1 = 0.4$, $f_2 = 0.2$ and $b = \sqrt{3}$.

7. Conclusion

The effective conductivity of three-phase composites with circular cylindrical inclusions in a periodic arrangement was derived by extending a method put forwarded by Lord Rayleigh [1]. Considering the recent development in the fast and accurate calculation of lattice sums

[16,19], such an extension can be used efficiently for calculating the conductivity of the system. A study of the bahvior of the composites revealed that they may exhibit unexpected results in particular states. The structure considered in this study was an ideal one, but the results can be useful for understanding the interplay between microstructures and the effective property of real multi-phase fiber composites and specifically those which can be approximated with the use of periodic structures and circular cylindrical inclusions. Also, the results provide a helpful resource in the process of testing and developing well-known classical numerical methods such as the boundary element method [20] or other proposed schemes in calculating the effective conductivity of multi-phase composite materials.

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Appendix A

We would like to calculate the following term:

$$\left\langle \boldsymbol{S} \right\rangle_{i} = \frac{1 - k_{i}}{V_{cell}} \int_{V_{i}} \nabla T_{i} dV \tag{A-1}$$

By using Green's first identity [21], the above relation becomes

$$\langle S \rangle_i = \frac{1 - k_i}{V_{cell}} \int_{S_i} T_i n dS ,$$
 (A-2)

where s_i is the surface of the cylinder of type *i* in the unit cell and *n* expresses the unit outward normal vector to the surface. Taking into account the temperature function given in equation (1) and after using the orthogonality properties of trigonometric functions, we can obtain

$$\langle S \rangle_{i} = \frac{1 - k_{i}}{V_{cell}} \int_{0}^{2p} a_{i}^{2} C_{1,i} \cos^{2} q \, dq \, \mathbf{i} = \frac{p a_{i}^{2} (1 - k_{i})}{V_{cell}} C_{1,i} \, \mathbf{i} = \frac{2p B_{1,i}}{V_{cell}} \mathbf{i}$$
(A-3)

Appendix B

Here extending the procedure given by Perrins et al. [6] we prove Keller's theorem for the system under study as follows: Considering that reversing the conductivity of the phases only makes the sign of \mathbf{g}_i (i = 1,2) negative and applying the mentioned property of lattice sums in section 4, from equation (16), we may write

$$\frac{(-1)^{n}B'_{2n-1,i}}{\boldsymbol{g}_{i}a_{i}^{4n-2}} + \sum_{m=1}^{\infty} \binom{2n+2m-3}{2n-1} S_{2n+2m-2,1}(-1)^{m}B'_{2m-1,i} + (-1)^{m}S_{2n+2m-2,2}B'_{2m-1,2-\boldsymbol{d}_{i2}} = \left[1-2\boldsymbol{p}\left(B'_{1,i}+B'_{1,2-\boldsymbol{d}_{i2}}\right)/b\right]\boldsymbol{d}_{n1}$$
(B-1)

Using equation (17) and comparing the above relation with equation (9), we find that

$$\frac{B'_{2n-1,i}}{k'_{e}(1/k_{1},1/k_{2},1)} = (-1)^{n} B_{2n-1,i}$$
(B-2)

Writing the above relation for n = 1 and using again equation (17) we have

$$k'_{e}(1/k_{1},1/k_{2},1) = 1 + 2p(B_{1,1} + B_{1,2})k'_{e}(1/k_{1},1/k_{2},1)/b$$
(B-3)

Applying equation (14) gives

$$k_e(k_1, k_2, 1) \times k'_e(1/k_1, 1/k_2, 1) = 1$$
 (B-4)

IV

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Critical resistance for multi-phase composite materials

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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 $R \le k - 1$, 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. © 2003 Elsevier Science B.V. All rights reserved.

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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. Start-ing with the work of Maxwell [1] and Rayleigh [2], who considered the problem of calculating the effec-tive 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.

* Corresponding author. E-mail address: ali.moosavi@lut.fi (A. Moosavi). 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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Fig. 1. The structure of the three-phase composite under study.

18 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 22 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.

26 Here, we extend the discussion to multi-phase com-27 posite materials. In order to simplify the presentation, let us consider a three-phase system that con-28 29 sists of a matrix and two types of circular cylindri-30 cal inclusions that are arranged in rectangular order 31 with periodicities equal to a unity in the y-direction and b in the x-direction, as depicted in Fig. 1. Let 32 33 us assume that a uniform field of magnitude E_{ext} has 34 been applied along the x-axis in the negative direction. At the surface of any cylinder of type i (i = 1, 2), 35 36 we may consider a dimensionless interfacial resis-37 tance [15], R_i , and express the boundary conditions 38 as follows:

43 where a and k represent the radius and the relative 44 conductivity of cylinders, respectively. T shows the 45 temperature function and *m* refers to the matrix. Us-46 ing the Rayleigh method for the purpose of solving the 47 Laplace equation through the system provides a sys-48 tem of algebraic equations in which the *n*th equation

$$(n = 1, \dots, \infty)$$
 of the set reads 49

$$\frac{B_{2n-1}^{i}}{\gamma_{2n-1}^{i}a_{i}^{4n-2}} + \sum_{m=1}^{\infty} \binom{2n+2m-3}{2n-1}$$
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$$\times \left(S_{2n+2m-2}^{1}B_{2m-1}^{i}+S_{2n+2m-2}^{2}B_{2m-1}^{2-\delta_{i2}}\right) = E_{\text{ext}}\delta_{n1}, \qquad \overset{53}{54}$$

$$(2)$$
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are the lattice sums 56
represents the Kro. 57

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where B_{2n-1}^i are unknowns, S_{2n}^i [1] over cylinders of type *i*, δ_{ij} represents the Kronecker delta (1 for i = j, otherwise 0) and γ_{2n-1}^{i} , which can be referred to as multipolar polarizabilities, are of the form

$$\gamma_{2n-1}^{i} = \frac{1 - k_i + R_i(2n-1)}{1 + k_i + R_i(2n-1)}.$$
(3)

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

$$k_e = 1 - \frac{2\pi (B_1^1 + B_1^2)}{bE_{\text{ext}}}$$
(4)

or more generally, for the case of N types of cylinders in the unit cell as $k_e = 1 - 2\pi \sum_{i=1}^{N} B_1^i / (bE_{\text{ext}})$. The values of B_1^1 and B_1^2 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_1^2 in (4), we can obtain an analytical relation for the effective conductivity, which can be applied to low-volume fractions, i.e.,

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

with

$$\lambda_i = \frac{1}{\gamma_1^i} + c_1 f_i - c_2 \gamma_3^i f_i^4 - c_3 \gamma_3^{2-\delta_{i2}} f_i f_{2-\delta_{i2}}^3, \quad (6)$$

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

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 conduc-95 tivity in the presence of the interfacial resistance. The 96

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The calculated values for c_1, \ldots, c_5 used in the analytical for-2 mula (5) for determining the effective conductivity in the parallel 3 and perpendicular directions for the case $b = \sqrt{3}$ 4

	Parallel	Perpendicular
<i>c</i> ₁	0.187018134	1.812981866
c2	1.310523128	1.310523128
c3	1.310523128	1.310523128
c_4	1.812981866	0.187018134
c5	-1.310523128	-1.310523128



Fig. 2. The contours of the effective conductivity with respect to the interfacial resistance of the cylinders. $f_1 = 0.4$, $f_2 = 0.2$, $k_1 = 101$, 32 $k_2 = 101$ and $b = \sqrt{3}$. As can be seen for the cases in which the 33 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.

38 volume fractions are $f_1 = 0.4$ and $f_2 = 0.2$, the con-39 ductivities of the cylinders were assumed to be $k_1 =$ 101, $k_2 = 101$, and the periodicity in the parallel di-40 rection is $b = \sqrt{3}$. The effective conductivity was de-41 42 rived numerically by solving Eq. (2) and then applying 43 Eq. (4). 100 unknowns of B_{2n-1}^{i} (*i* = 1, 2) were con-44 sidered in the process of the solution. As can be seen, 45 the effective conductivity of the system can be highly 46 affected, and based on the values of R_1 and R_2 , the 47 system may yield a conductivity outside the limit of 48 the conductivity of the phases. Providing that B_1^1 +

 $B_1^2 = 0$, the system is subject to situations in which 49 the effect of the inclusions can be neglected ($k_e = 1$ 50 in Fig. 2). Of these states, the case $B_1^1 = B_1^2 = 0$ is of 51 particular interest. This case occurs when $R_i = k_i - 1$ 52 (i = 1, 2), and as a result based on Eq. (3) the dipole 53 polarizabilities (γ_1^1, γ_1^2) are zero. Obtaining a value of 54 zero for the dipole polarizabilities means that the state 55 of the system resembles that of a system of perfect in-56 terfaces which is made up of cylinders the conductivity 57 of which is equal to a unity. Therefore, we may expect 58 that the effective conductivity would be independent 59 of the volume fractions and the direction of the calcu-60 lation of the effective conductivity. 61

While the effect of both types of cylinders can be 62 exactly neglected when $R_i = k_i - 1$ (i = 1, 2), in 63 general the effect of the inclusions of type i (i = 164 or 2) cannot be neglected when only $R_i = k_i - 1$. 65 In other words, systems with $k_i = 1$, $R_i = 0$ (two-66 phase system) and k_i , $R_i = k_i - 1$ (three-phase sys-67 tem) are not equivalent when $R_{2-\delta_{i2}} \neq k_{2-\delta_{i2}} - 1$. 68 Although for both cases, B_1^i is zero and the effec-69 tive conductivity can be calculated simply by using 70 $k_e = 1 - 2\pi B_1^{2-\delta_{i2}}/(bE_{\text{ext}})$ but for the imperfect in-71 72 terface case, the terms B_{2n-1}^i (n > 1) are not zero 73 and are present in the procedure of the calculation of 74 $B_1^{2-\delta_{i2}}$, as is evident in Eq. (2). For the perfect inter-75 face case, all the terms of B_{2n-1}^i (n > 1) are zero and 76 do not affect the value of $B_1^{2-\delta_{i2}}$. This means that the 77 field distributions inside the matrix for the two- and 78 three-phase systems may be dissimilar. The degree of 79 discrepancy can only be numerically determined and 80 depends on the geometrical considerations, resistance 81 and conductivity of the inclusions. Fig. 3 shows a com-82 parison between the effective conductivity of the two 83 systems for a series of given data. As can been seen, 84 the results for the three-phase system underestimate 85 the conductivity of the two-phase system. The reason 86 can be understood when considering that for the three-87 phase case $\gamma_{2n-1}^i > 0$ (n > 1), while for the two-phase 88 one they are zero. Increasing f_i increases the error, 89 since the higher-order terms play an important role 90 in the response of the system. In the dilute limit, the 91 systems can be used equivalently. This is also evident 92 from Eq. (5), as ignoring the higher orders for the case 93 $\gamma_1^i = 0$ gives $k_e = 1 - 2f_{2-\delta_{i2}}/(1 + c_1\gamma_1^{2-\delta_{i2}})$, which 94 is the conductivity of the system, which neglects the 95 effect of the type-*i* inclusions. 96

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Fig. 3. The effective conductivity of the two- and three-phase 18 systems. For the three-phase system, $k_1 = 101$, $k_2 = 101$, $R_1 = 10$, 19 $R_2 = k_2 - 1 = 100$ 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.

23 By reducing R_i from the value $k_i - 1$, we may 24 obtain a situation in which the three-phase system 25 exactly gives the effective conductivity of the two-26 phase one. The expectation of finding such a situation 27 stems from the fact that cylinders the conductivity of which is greater than that of the matrix, boost 28 29 the conductivity of the system. Fig. 4 reports such an interfacial resistance for type-two cylinders as 30 31 a function of the interfacial resistance of type-one 32 cylinders for another series of given data. As can be 33 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 34 cell, by extending Eqs. (2) and (4), it can be shown 35 36 that when $R_i = k_i - 1$ (i = 1, ..., N), the effect 37 of all types of cylinders can be neglected and the 38 system simply behaves like a homogeneous system 39 with the conductivity of the matrix. In other cases, the resistance, in which the effect of type-*i* cylinders in the 40 direction of the calculation the effective conductivity 41 42 can be neglected, may be estimated by $R_i < k_i - 1$.

43 In summary, the effective conductivity of a multi-44 phase composite material that is made up of a periodic 45 structure in the presence of the interfacial resistance 46 was studied. The situations in which the effect of one 47 or all types of cylinders can be neglected due to the 48 interfacial resistance were explained. The structure



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 $b = \sqrt{3}$.

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considered in this study was composed of circular cylinders in a periodic arrangement, but when the results given in Refs. [11-14] 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).

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V

An investigation into behavior of multi-phase composite materials: coated systems

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Abstract

This paper reports a series of investigations into the effective transport properties of composite materials with coated inclusions. The geometry of the inclusions under study are circular cylindrical, elliptical cylindrical and spherical. The method we use for this study extends the theorems or exact solutions given for two-phase systems. It is shown that only in a few cases coated composites exhibit the same behavior.

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1. Introduction

Determining the response of composite systems to an imposed field has long been of interest from both the theoretical and applied standpoints. The problem arises when studying a variety of phenomena in physics [1-3], mathematics [4,5], biology [6] and engineering [7]. In many cases it is necessary to coat the inclusions; one such case is for the purpose of increasing the wettability and adhesion of the inclusions and the matrix. When the enhancing effect of the inclusions is not sufficient, a coating layer may modify the behaviour of the system. For protection from chemical reactions, a suitable coating layer can also be applied. In some cases, the coating layer is unwanted but occurs because of many problems in the production processes. For instance, due to a mismatch in thermal expansion, a coating layer with a third material may appear between the inclusions and the matrix. In some cases, modelling a system as a coated one may provide reasonable results, even though the system is not exactly coated. This is the case for wet wood for which the moisture is considered to be a coating layer. Therefore, extending the formulations to coated systems and inspecting behavior of these systems may have many implications.

In this research, we consider coated composite systems and inspect some of the behaviour of these systems. The formulations have been given in the context of dielectric constant for greater generality [8].

2. Analysis

Consider a statistically homogeneous composite material that consists of a random array of cylinders with arbitrary cross-sections embedded in a host medium. The dielectric constants of the layers are e_i (i = 1,2) where i = 1 stands for the core and i=2 for the matrix. Mendelson [9] has shown that Keller's reciprocal theorem [10] is valid for the system. Therefore we can write the following :

$$\boldsymbol{e}_{eff}^{x}\left(\boldsymbol{e}_{1},\boldsymbol{e}_{2}\right) \times \boldsymbol{e}_{eff}^{y}\left(1/\boldsymbol{e}_{1},1/\boldsymbol{e}_{2}\right) = 1$$

$$(1)$$

where \mathbf{e}_{eff}^{x} and \mathbf{e}_{eff}^{y} are the effective dielectric constants in the direction of the principal axes x and y, respectively. The mathematical basis for the proof of this behaviour is the observation that any two-dimensional divergence-free field, when rotated locally at each point by 90°, produces a curl-free field and vice versa [11]. If we develop this result to multiply coated systems we obtain

$$\boldsymbol{e}_{eff}^{x}\left(\boldsymbol{e}_{1},\ldots,\boldsymbol{e}_{N}\right) \times \boldsymbol{e}_{eff}^{y}\left(1/\boldsymbol{e}_{1},\ldots,1/\boldsymbol{e}_{N}\right) = 1$$

$$\tag{2}$$

where N stands for the matrix(see also Lu [12] and Fel et al. [13]). Considering the following general rule

$$\boldsymbol{e}_{eff}^{y}\left(\boldsymbol{a}_{1}\boldsymbol{e}_{1},\ldots,\boldsymbol{a}_{l}\boldsymbol{e}_{l},\ldots,\boldsymbol{a}_{N}\boldsymbol{e}_{N}\right)=\boldsymbol{a}_{l}\boldsymbol{e}_{eff}^{y}\left(\boldsymbol{a}_{1,l}\boldsymbol{e}_{1},\ldots,\boldsymbol{e}_{l},\ldots,\boldsymbol{a}_{N,l}\boldsymbol{e}_{N}\right)$$
(3)

where $\mathbf{a}_{i,i} = \mathbf{a}_i / \mathbf{a}_i$, we may also derive

$$\boldsymbol{e}_{eff}^{x}\left(\boldsymbol{e}_{1},\ldots,\boldsymbol{e}_{N}\right)\times\boldsymbol{e}_{eff}^{y}\left(\boldsymbol{e}_{2}\boldsymbol{e}_{3}\cdots\boldsymbol{e}_{N},\boldsymbol{e}_{1}\boldsymbol{e}_{3}\cdots\boldsymbol{e}_{N},\ldots,\boldsymbol{e}_{1}\boldsymbol{e}_{2}\cdots\boldsymbol{e}_{N-1}\right)=\boldsymbol{e}_{1}\boldsymbol{e}_{2}\cdots\boldsymbol{e}_{N} \qquad (4)$$

If the medium is statistically isotropic, relation (2) can be rewritten as

$$\boldsymbol{e}_{eff}\left(\boldsymbol{e}_{1},\ldots,\boldsymbol{e}_{N}\right)\times\boldsymbol{e}_{eff}\left(1/\boldsymbol{e}_{1},\ldots,1/\boldsymbol{e}_{N}\right)=1$$
(5)

The behaviour expressed in Eq. (2) and the resultant relations adhere to the following general facts that can be applied to any coated composites. When one layer is perfectly insulating or conducting, the layers under this layer will have no affect in the transport property of the system. Secondly, if two layers have the same dielectric constant, they can be considered to be a unit layer.

For composites consisting of a random array of singly coated circular cylinders, Nicorovici et al. [14] proved that, by coating the cores with the material of the core or its negative, the same effective dielectric constant may be achieved. This behaviour can be formulated as

$$\boldsymbol{e}_{eff}^{x}\left(\boldsymbol{e}_{1},\boldsymbol{e}_{1},\boldsymbol{e}_{2}\right) \times \frac{1}{\boldsymbol{e}_{eff}^{x}\left(\boldsymbol{e}_{1},\pm\boldsymbol{e}_{1},\boldsymbol{e}_{2}\right)} = 1$$

$$(6)$$

$$\boldsymbol{e}_{eff}^{y}\left(\boldsymbol{e}_{1},\boldsymbol{e}_{1},\boldsymbol{e}_{2}\right) \times \frac{1}{\boldsymbol{e}_{eff}^{y}\left(\boldsymbol{e}_{1},\pm\boldsymbol{e}_{1},\boldsymbol{e}_{2}\right)} = 1$$
(7)

Our attempt [15] to extend the results to multiply coated cylinders showed that coating the cores with a series of layers, which have the same property as the core or its negative, may yield the same response to the applied field. For example, the following doubly coated systems all have exactly the same dielectric constant:

$$\begin{array}{l}
 e_{1}, e_{1}, e_{1}, e_{2} \\
 e_{1}, -e_{1}, e_{1}, e_{2} \\
 e_{1}, e_{1}, -e_{1}, e_{2} \\
 e_{1}, -e_{1}, -e_{1}, e_{2} \\
 \end{array}$$
(8)

This means that

$$\boldsymbol{e}_{eff}^{x}\left(\boldsymbol{e}_{1},\boldsymbol{e}_{1},\cdots,\boldsymbol{e}_{1},\boldsymbol{e}_{2}\right)\times\frac{1}{\boldsymbol{e}_{eff}^{x}\left(\boldsymbol{e}_{1},\pm\boldsymbol{e}_{1},\cdots,\pm\boldsymbol{e}_{1},\boldsymbol{e}_{2}\right)}=1$$
(9)

$$\boldsymbol{e}_{eff}^{y}(\boldsymbol{e}_{1},\boldsymbol{e}_{1},\cdots,\boldsymbol{e}_{1},\boldsymbol{e}_{2}) \times \frac{1}{\boldsymbol{e}_{eff}^{y}(\boldsymbol{e}_{1},\pm\boldsymbol{e}_{1},\cdots,\pm\boldsymbol{e}_{1},\boldsymbol{e}_{2})} = 1$$
(10)

Using relation (2) we may also find that

$$\boldsymbol{e}_{eff}^{x}\left(\boldsymbol{e}_{1},\boldsymbol{e}_{1},\cdots,\boldsymbol{e}_{1},\boldsymbol{e}_{2}\right)\times\boldsymbol{e}_{eff}^{y}\left(1/\boldsymbol{e}_{1},\pm1/\boldsymbol{e}_{1},\cdots,\pm1/\boldsymbol{e}_{1},1/\boldsymbol{e}_{2}\right)=1$$
(11)

The proof of the behaviour outlined in Eqs. (9) and (10) is different from that given for Eqs. (1) and (2) and is based on deriving the effective dielectric constant of

the system. Herein, we briefly explain the method for the composites with square array of multiply coated circular cylinders. The effective dielectric constant of the system can be calculated using $\mathbf{e}_{eff} = 1 - 2\mathbf{p}B_1^N$ which is the result of Green's theorem [16]. B_1^N is an unknown that can be obtained by solving the following set of linear equations [17]:

$$\boldsymbol{x}_{2n-1}^{N}B_{2n-1}^{N} + \sum_{m=1}^{\infty} \frac{(2n+2m-3)!}{(2m-2)!(2n-1)!} S_{2n+2m-2}B_{2m-1}^{N} = \boldsymbol{d}_{n1}$$
(12)

where d_{n1} represents the Kronecker delta (1 for n=1, otherwise 0), S_n are the lattice sums [16,17] and a_i is the radius of layer *i*. The coefficients \mathbf{x}_{2n-1}^N can be obtained by successively applying the following procedure which is the result of implementing the boundary conditions between the layers:

$$\mathbf{x}_{2n-1}^{i} = \frac{(\mathbf{e}_{i} - \mathbf{e}_{i-1}) + (\mathbf{e}_{i} + \mathbf{e}_{i-1}) \mathbf{x}_{2n-1}^{i-1} a_{i-1}^{4n-2}}{(\mathbf{e}_{i} + \mathbf{e}_{i-1}) + (\mathbf{e}_{i} - \mathbf{e}_{i-1}) \mathbf{x}_{2n-1}^{i-1} a_{i-1}^{4n-2}} \times \frac{1}{a_{i-1}^{4n-2}}$$
(13)

For all the cases given in (8), relation (13) can be reduced to $\mathbf{x}_{2n-1}^4 = (\mathbf{e}_2 + \mathbf{e}_1)/[(\mathbf{e}_2 - \mathbf{e}_1)a_3^{4n-2}]$ which is exactly equal to that given for solid cylinders of radius a_3 . Since the other parameters remain unchanged, all the cases yield the same B_1 and, as a result, the same dielectric constant can be expected for all of them. We have numerically verified the above theorems. Our numerical investigations show that it is not necessary for the system to be composed only of coated cylinders. Other geometries, which do not interrupt coated cylinders, can also be included in the system.

The above bring up the question as to whether one can expect the same behaviour for all composites with cylindrical inclusions. Among a few potential geometries for analytical inspection, we rather consider elliptical inhomogenities [18] since the solution is given based on the Rayleigh method. Because of the anisotropy, we are concerned with two series of relations for x, i.e.

$$\mathbf{x}_{n}^{x,2} = \frac{\boldsymbol{e}_{2} \cosh(n\boldsymbol{m}_{1}) + \boldsymbol{e}_{1} \sinh(n\boldsymbol{m}_{1})}{(\boldsymbol{e}_{2} - \boldsymbol{e}_{1}) \sinh(2n\boldsymbol{m}_{1})} \times \frac{1}{(c/2)^{2n} e^{n\boldsymbol{m}_{1}}}$$
(14)

$$\boldsymbol{x}_{n}^{y,2} = \frac{\boldsymbol{e}_{1} \cosh(n\boldsymbol{m}_{1}) + \boldsymbol{e}_{2} \sinh(n\boldsymbol{m}_{1})}{(\boldsymbol{e}_{2} - \boldsymbol{e}_{1}) \sinh(2n\boldsymbol{m}_{1})} \times \frac{1}{(c/2)^{2n} e^{n\boldsymbol{m}_{1}}}$$
(15)

where (m,q) are elliptic cylindrical coordinates and c is the distance between the two foci. Here we develop the solution to a multiply coated system. The results can be expressed as

$$\boldsymbol{x}_{n}^{x,i} = \frac{e^{-n\boldsymbol{m}_{i-1}} \left(\boldsymbol{e}_{i} - \boldsymbol{e}_{i-1}\right) + 2\boldsymbol{x}_{n,x}^{i-1} (c/2)^{2n} \left[\boldsymbol{e}_{i} \cosh(n\boldsymbol{m}_{i-1}) + \boldsymbol{e}_{i-1} \sinh(n\boldsymbol{m}_{i-1})\right]}{2e^{-n\boldsymbol{m}_{i-1}} \left[\boldsymbol{e}_{i} \sinh(n\boldsymbol{m}_{i-1}) + \boldsymbol{e}_{i-1} \cosh(n\boldsymbol{m}_{i-1})\right] + 2\boldsymbol{x}_{n,x}^{i-1} (c/2)^{2n} \sinh(2n\boldsymbol{m}_{i-1}) \left(\boldsymbol{e}_{i} - \boldsymbol{e}_{i-1}\right)} \times \frac{1}{(c/2)^{2n} e^{n\boldsymbol{m}_{i-1}}}$$
(16)

$$\mathbf{x}_{n}^{y,i} = \frac{e^{-n\mathbf{m}_{i-1}}(\mathbf{e}_{i} - \mathbf{e}_{i-1}) + 2\mathbf{x}_{n,y}^{i-1}(c/2)^{2n}[\mathbf{e}_{i}\sinh(n\mathbf{m}_{i-1}) + \mathbf{e}_{i-1}\cosh(n\mathbf{m}_{i-1})]}{2e^{-n\mathbf{m}_{i-1}}[\mathbf{e}_{i}\cosh(n\mathbf{m}_{i-1}) + \mathbf{e}_{i-1}\sinh(n\mathbf{m}_{i-1})] + 2\mathbf{x}_{n,y}^{i-1}(c/2)^{2n}\sinh(2n\mathbf{m}_{i-1})(\mathbf{e}_{i} - \mathbf{e}_{i-1})} \times \frac{1}{(c/2)^{2n}e^{n\mathbf{m}_{i-1}}}$$
(17)

Using the relations $\lim_{\substack{c \to 0 \\ \mathbf{m}_{l} \to \infty}} ce^{\mathbf{m}_{l}}/2 = a_{i}$ and $\lim_{\substack{c \to 0 \\ \mathbf{m}_{l} \to \infty}} ce^{-\mathbf{m}_{l}}/2 = 0$, one can show that in the limit, both $\mathbf{x}_{n}^{x,i}$ and $\mathbf{x}_{n}^{y,i}$ will be reduced exactly to the corresponding relation given for circular cylinders (i.e. Eq. (13)). But substituting the relations $\mathbf{e}_{i} = -\mathbf{e}_{i-1}$ ($i = 2, \dots, N-1$) for an arbitrary c and \mathbf{m} does not reduce $\mathbf{x}_{n}^{x,i}$ and $\mathbf{x}_{n}^{y,i}$ to the shape of the formulations given for a two-phase system (i.e. Eqs. (14) and (15)). Therefore, the above-mentioned behaviour for circular cylinders cannot be applied for all coated composites with cylindrical inclusions. In figures 1 (a, b) this fact has been verified. As can be seen, changing the dielectric constant of the coating layer from $\mathbf{e}_{2} = \mathbf{e}_{1}$ to $\mathbf{e}_{2} = -\mathbf{e}_{1}$ alters the field through the matrix. The equivalent system for the case $\mathbf{e}_{2} = -\mathbf{e}_{1}$ can only be numerically determined.



Fig. 1. The equipotential contours inside the unit cell of a system composed of singly coated elliptical cylinders. A potential gradient of unit magnitude was applied externally along the *x*-axis and other external boundaries were insulated. (a) $\boldsymbol{e}_2 = \boldsymbol{e}_1$ (b) $\boldsymbol{e}_2 = -\boldsymbol{e}_1$.
Now we consider composites with coated spherical inclusions. The behavior given in Eq. (2) is not valid for these composites [19]. Also, it has been shown that relations (6) and (7) are not applicable for composites with spherical inclusions [14]. The equivalent system again can only be numerically determined. However, in the dilute limit, where we can assume that there is no interaction between the spheres, cases $\mathbf{e}_2 = -0.5\mathbf{e}_1$ and $\mathbf{e}_2 = \mathbf{e}_1$ give the same response to the applied field. If we develop the solution given for the solid spheres [20] to multiply coated spheres, the corresponding \mathbf{x}_n^i can be obtained as

$$\boldsymbol{x}_{2n-1}^{i} = \frac{2n(\boldsymbol{e}_{i} - \boldsymbol{e}_{i-1}) + \boldsymbol{x}_{2n-1}^{i-1} [(2n-1)\boldsymbol{e}_{i-1} + 2n\boldsymbol{e}_{i}] a_{i-1}^{4n-1}}{[2n\boldsymbol{e}_{i-1} + (2n-1)\boldsymbol{e}_{i}] + \boldsymbol{x}_{2n-1}^{i-1} (2n-1)(\boldsymbol{e}_{i} - \boldsymbol{e}_{i-1}) a_{i-1}^{4n-1}} \times \frac{1}{a_{i-1}^{4n-1}}$$
(19)

By substituting $\mathbf{e}_i = -0.5\mathbf{e}_{i-1}$ $(i = 3, \dots, N-1)$ for n=1 in Eq. (19) one can show that the above-mentioned behaviour for composites with singly coated spherical inclusions cannot be extended to multiply coated spheres. We also argue that applying the above conditions for multiply coated systems means that, for infinitely coated layers, the property of the outermost coating layer tends to be zero. As mentioned before, this layer has the property of impeding the effects of the other coating layers in the dielectric constant of the system. Therefore, the dielectric constant of the system consisting of spheres with the property of the core.

3. Conclusion

The behaviour of coated composites in a viewpoint can be divided into two main groups of which one is applicable to every coated system and the other to those which are valid just for one or a series of coated systems. The restrictions in the behaviour outlined in this research were the dimension, concentration and geometry.

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