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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 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 $\boldsymbol{r}_n$ defined as:

$$\boldsymbol{r}_n = h\,(n_1\boldsymbol{a}_1 + n_2\boldsymbol{a}_2 + n_3\boldsymbol{a}_3) \qquad (1)$$

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	$\boldsymbol{a}_1$	$\boldsymbol{a}_2$	$\boldsymbol{a}_3$
SC	$\boldsymbol{e}_x$	$\boldsymbol{e}_y$	$\boldsymbol{e}_z$
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	$1/2(\boldsymbol{e}_x+\boldsymbol{e}_y)$	$1/2(\boldsymbol{e}_y+\boldsymbol{e}_z)$	$1/2(\boldsymbol{e}_x+\boldsymbol{e}_z)$

TABLE 1 The basic vectors $\boldsymbol{a}_1$, $\boldsymbol{a}_2$ and $\boldsymbol{a}_3$ for three cubic lattices. The vectors $\boldsymbol{e}_x$, $\boldsymbol{e}_y$ and $\boldsymbol{e}_z$ 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 $\boldsymbol{a}_1$, $\boldsymbol{a}_2$ and $\boldsymbol{a}_3$ 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:

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

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

where $\tau_0 = \boldsymbol{a}_1 \cdot [\boldsymbol{a}_2 \times \boldsymbol{a}_3]$ 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 x_1-axis, then the following relations apply:

$$x_1 = r\cos\theta, \quad x_2 = r\sin\theta\cos\varphi, \quad x_3 = r\sin\theta\sin\varphi \tag{4}$$

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:

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

$$T^i = \sum_{n=1}^{\infty}\sum_{m=0}^{m<\frac{1}{2}n} [E_{nm}^i r^{2n-1} + F_{nm}^i r^{-2n}] P_{2n-1}^{4m}(\cos\theta)\cos m\varphi \qquad (i = 2, \ldots, N) \tag{6}$$

where $P_L^M(\cos\theta)$ represents the associated Legendre polynomial of degree L and order M. Zuzovski and Brenner [7] proposed another suitable expression for the temperature inside the continuous phase:

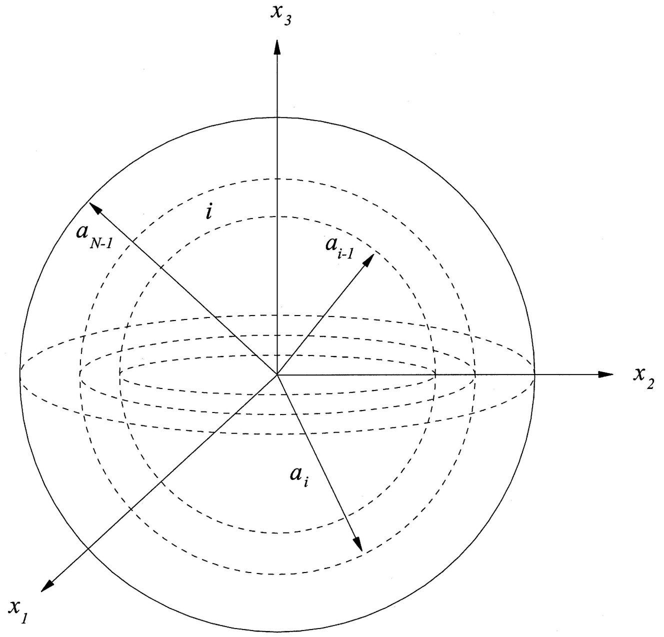

FIGURE 1 The multi-coated sphere under study

$$T^N = x_1 + \boldsymbol{G}H \tag{7}$$

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

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

where

$$\xi = x_2 + \mathrm{i}x_3, \quad \eta = x_2 - \mathrm{i}x_3 \tag{9}$$

and

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

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:

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

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:

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

Using these conditions, the following result is obtained:

$$F^i_{nm} + L^i_n a^{4n-1}_{i-1} E^i_{nm} = 0 \qquad i \geq 2 \tag{13}$$

where

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

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

$$A_{nm} = L^N_{M+1} \frac{a^{4M+3}_{N-1}}{(2M+4m+1)!} \times \sum_{J=0}^{\infty} \sum_{j=0}^{j\leq\frac{1}{2}J} \left[\frac{\lambda_1 \varepsilon_{q_1} (2p-4q_1)!}{(2J-4j+1)} S_{pq_1} + \frac{\lambda_2 \varepsilon_{q_2} (2p-4q_2)!}{(2J-4j+1)} S_{pq_2}\right] \times A_{J-2j,j} + \delta_{M0} L^N_1 a^3_{N-1} \left(1 + \frac{4\pi}{3\tau_0} A_{00}\right) \tag{15}$$

where

$$p = M+J+1, \quad q_1 = m+j, \quad q_2 = |m-j| \tag{16}$$

$$\left.\begin{array}{lll} \lambda_1 = \lambda_2 = 0.25 & \text{if} & j \neq 0, m = 0 \\ \lambda_1 = 0.5, \quad \lambda_2 = 1 & \text{if} & m = j \neq 0 \\ \lambda_1 = \lambda_2 = 0.5 & \text{otherwise} & \end{array}\right\} . \tag{17}$$

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

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

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:

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

where

$$D = -1/L^N_1 + F + c_1 L^N_2 F^{10/3} \frac{1 + c_4 L^N_3 F^{11/3}}{1 - c_2 L^N_2 F^{7/3}} + c_3 L^N_3 F^{14/3} + c_5 L^N_4 F^6 + c_6 L^N_5 F^{22/3} + O\left(F^{25/3}\right) . \tag{20}$$

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:

$$k_{\text{eff}} = 1 - \frac{3F}{-1/L^N_1 + F + c_1 L^N_2 F^{10/3}} . \tag{21}$$

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 $F \to 0$.

$$k_{\text{eff}} = 1 + 3FL^N_1 + O\left(F^2\right) . \tag{22}$$

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

$$k_{\text{eff}} = 1 + 3FL^3_1 = 1 + 3F \times \frac{(k_2-1) + (1+2k_2)}{(k_2+2) + 2(k_2-1)} \frac{\left[(k_1-k_2)/(k_1+2k_2)\right](a_1/a_2)^3}{\left[(k_1-k_2)/(k_1+2k_2)\right](a_1/a_2)^3} . \tag{23}$$

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

$$k_{\text{eff}} = 1 + 3FL^4_1 = 1 + 3F\frac{P+Q}{R+S} \tag{24}$$

where

$$\begin{aligned} P &= (k_3-1)\{(k_2+2k_3) + 2(k_2-k_3)\left[(k_1-k_2)\right. \\ &\quad \left./(k_1+2k_2)\right](a_1/a_2)^3\} \\ Q &= (1+2k_3)\left\{(k_2-k_3)(a_2/a_3)^3 + (k_3+2k_2)\left[(k_1-k_2)\right.\right. \\ &\quad \left.\left./(k_1+2k_2)\right](a_1/a_3)^3\right\} \\ R &= (k_3+2)\{(k_2+2k_3) + 2(k_2-k_3)\left[(k_1-k_2)\right. \\ &\quad \left./(k_1+2k_2)\right](a_1/a_2)^3\} \\ S &= 2(k_3-1)\left\{(k_2-k_3)(a_2/a_3)^3 + (k_3+2k_2)\left[(k_1-k_2)\right.\right. \\ &\quad \left.\left./(k_1+2k_2)\right](a_1/a_3)^3\right\} . \end{aligned} \tag{25}$$

	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}
c_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}
c_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 2 Numerical constants in explicit expression

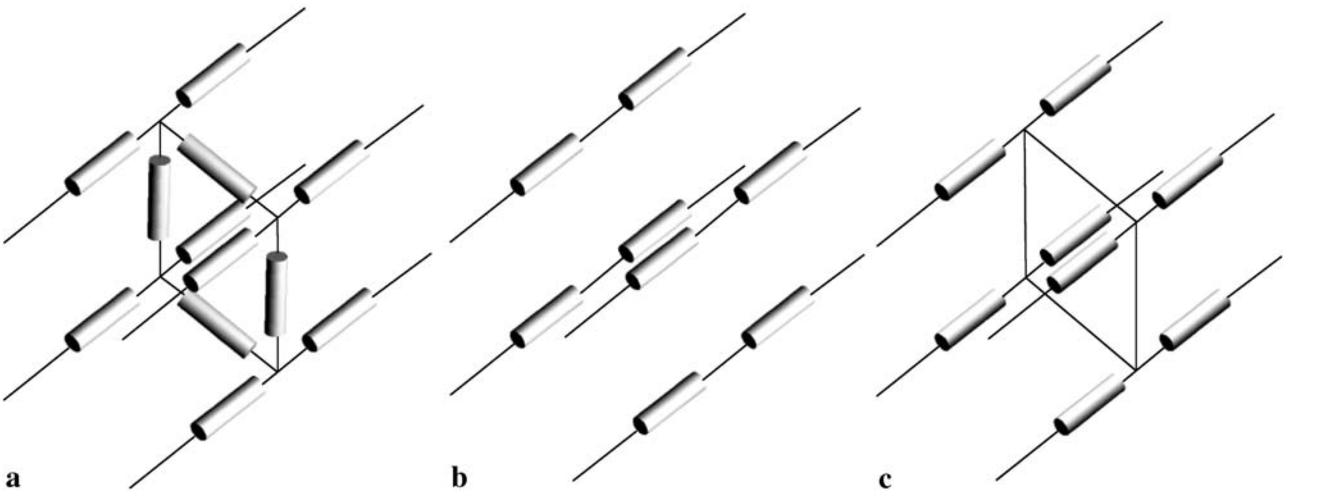

FIGURE 2 Part of the resistor network. Original (**a**) and simplified arrangements (**b, c**)

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:

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

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.

4 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_{eff}	k'_{eff}	k''_{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:

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

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:

$$K_i = \frac{1}{a_i^2 - a_{i-1}^2} \times \int_{\theta_i}^{\pi/2} \frac{a_i^2 \sin 2\theta \mathrm{d}\theta}{\sum_{j=i}^{N-1} 2a_j\sqrt{1-\left(a_i\sin\theta/a_j\right)^2}\left(1/k_j - 1/k_{j+1}\right)+1}$$
$$(i=1,\ldots,N-1) \tag{28}$$

where

$$\theta_i = \sin^{-1}\left(\frac{a_{i-1}}{a_i}\right)\ . \tag{29}$$

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

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

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 ($i = 1, \ldots, N$) are considered. For each slice, the equivalent conductivity may be specified as:

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

where

$$\left.\begin{aligned} s_j &= \tfrac{\pi}{4}\left\{a_j^2\left[1-\left(a_i\cos\theta/a_j\right)^2\right] - a_{j-1}^2\left[1-\left(a_i\cos\theta/a_{j-1}\right)^2\right]\right\} \quad \text{if } j=i,\ldots,N-1 \\ s_j &= 1/4 - \pi\left[a_{j-1}^2 - (a_i\cos\theta)^2\right] \quad \text{if } j=N \end{aligned}\right\} \tag{32}$$

and $S = 1/4$ is the non-dimensional cross-sectional area of the slice. It is then straightforward to derive the equivalent ther-

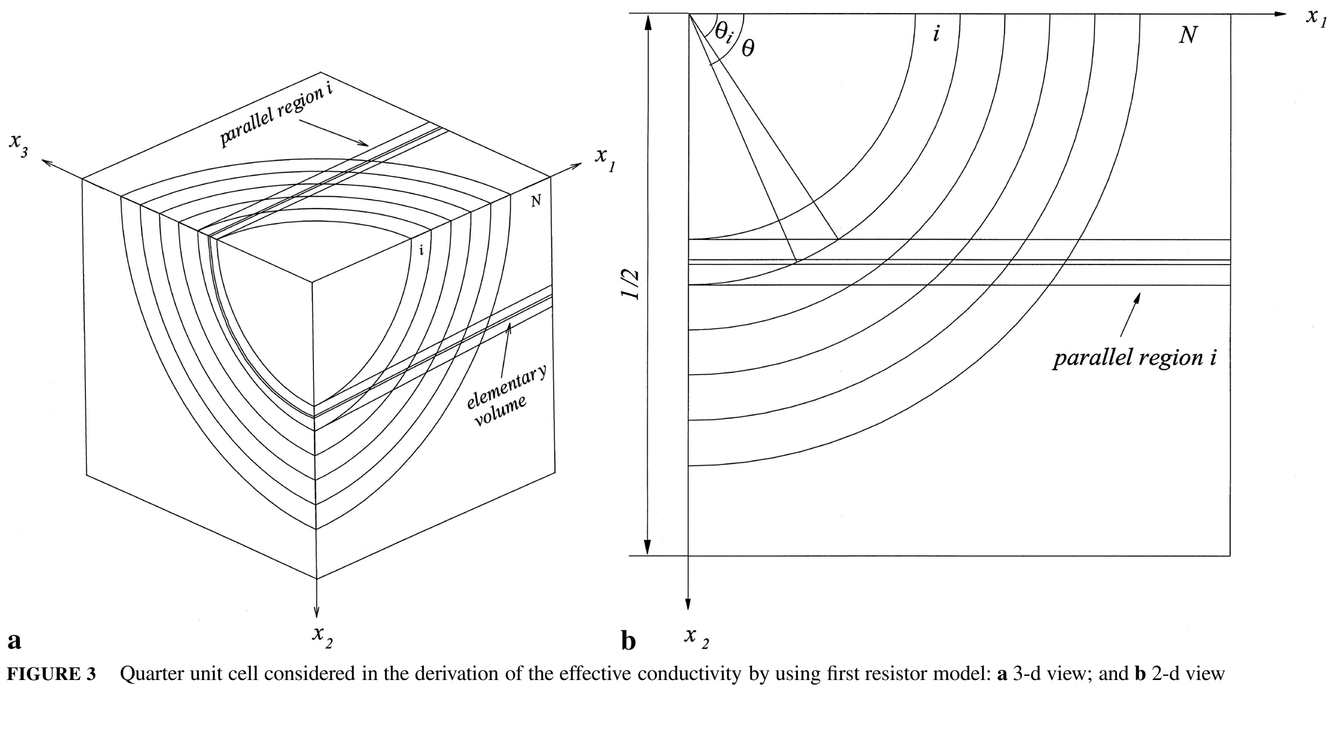

FIGURE 3 Quarter unit cell considered in the derivation of the effective conductivity by using first resistor model: **a** 3-d view; and **b** 2-d view

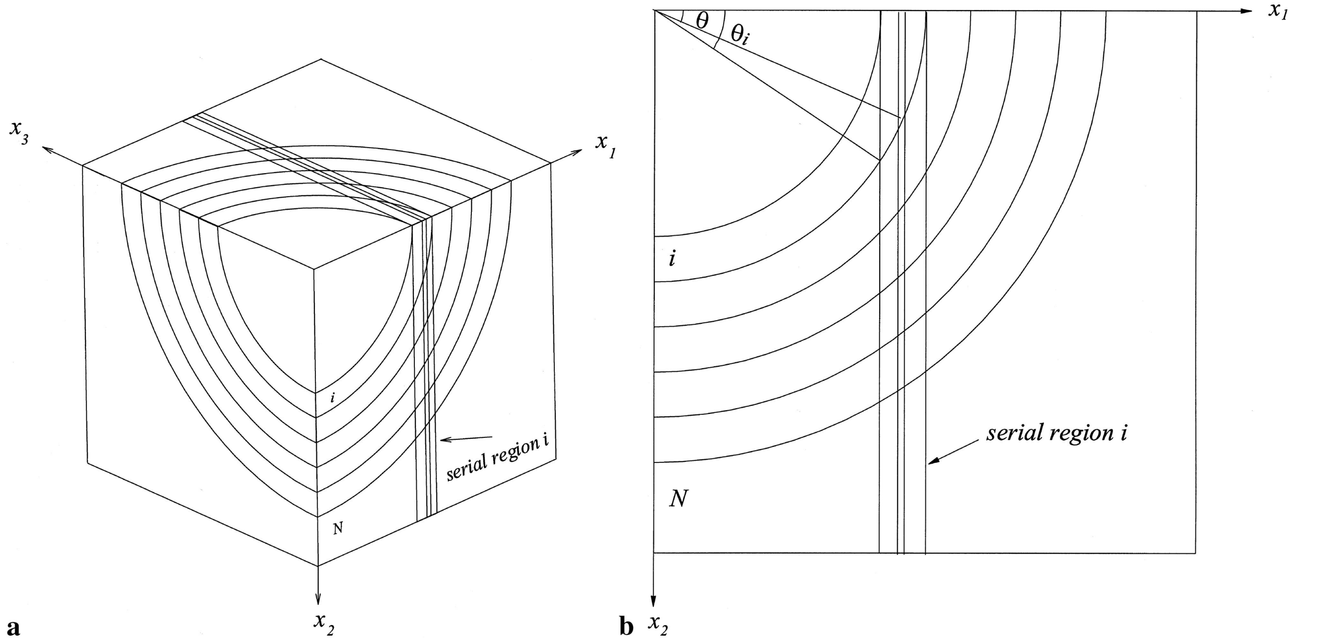

FIGURE 4 Quarter unit cell under inspection in second resistor model: **a** 3-d view; and **b** 2-d view

mal conductivity of each region as:

$$K_i = \frac{a_i - a_{i-1}}{\int_0^{\theta_i} \frac{a_i \sin\theta \mathrm{d}\theta}{\pi\left\{k_i (a_i \sin\theta)^2 + \sum_{j=i+1}^{N-1} k_j \left(a_j^2 - a_{j-1}^2\right) - \left[a_{N-1}^2 - (a_i \cos\theta)^2\right]\right\} + 1}}$$

$$(i = 1, \dots, N-1) \tag{33}$$

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:

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

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-

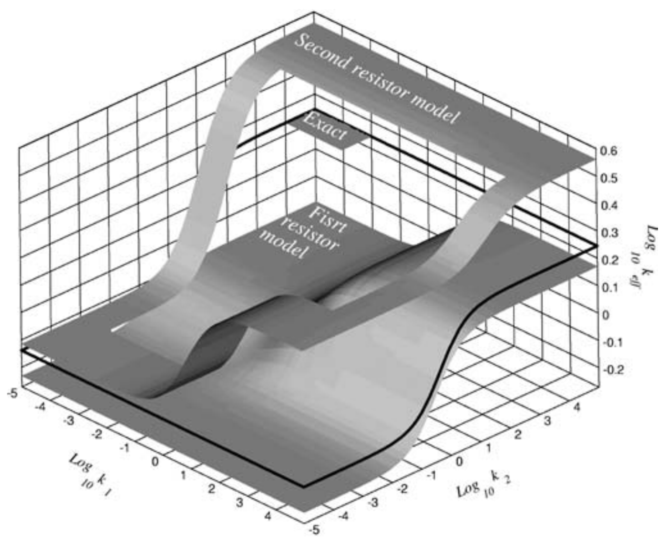

FIGURE 5 Logarithmic plot of the effective conductivity for the simple cubic array of coating spheres as a function of the conductivity of the core and shell. Despite of the accuracy, the first resistor model is unable to show the same behaviour as the exact solution when the shell is perfectly conducting.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

$$K_{1^*} = \int_0^{\pi/2} \frac{\sin(2\theta)}{1 - 2a_2 \cos\theta} = -\frac{1}{a_2} - \frac{1}{2a_2^2} \ln(1 - 2a_2)\, \mathrm{d}\theta \quad (36)$$

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

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

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

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:

$$k_{\mathrm{eff}} = \frac{1/2}{1/2 - a_2} . \quad (39)$$

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:

$$K_{1^*} = \int_0^{\pi/2} \frac{\sin\theta}{1 - \pi a_2^2 \sin^2\theta} = \frac{a_2 \pi \sqrt{1/\pi - a_2^2}\, \mathrm{d}\theta}{\arctan\left(a_2/\sqrt{1/\pi - a_2^2}\right)} \quad (40)$$

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

Also, for the first model one may find:

$$k_{\mathrm{eff}} = 1 - \pi a_2^2 . \quad (42)$$

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 ($k_1 = +\infty, k_2 = 0$ 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.

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

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

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

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.

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

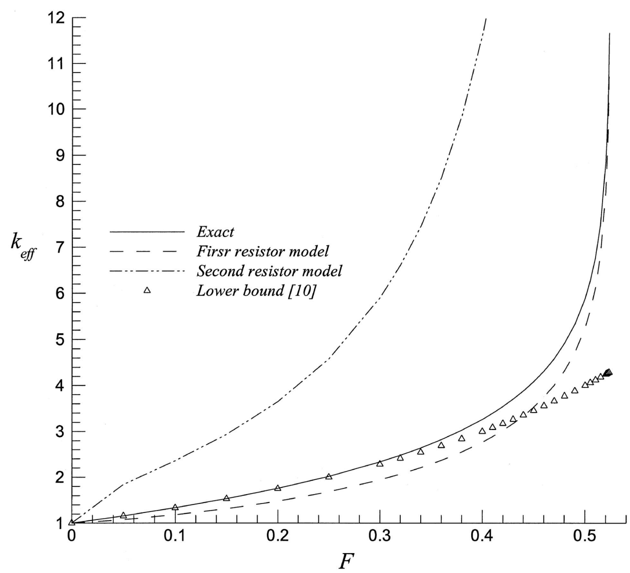

FIGURE 6 Comparison between the results of the first and second resistor models with the exact solution and the results of the lower bound given in [10] for perfectly conducting phases

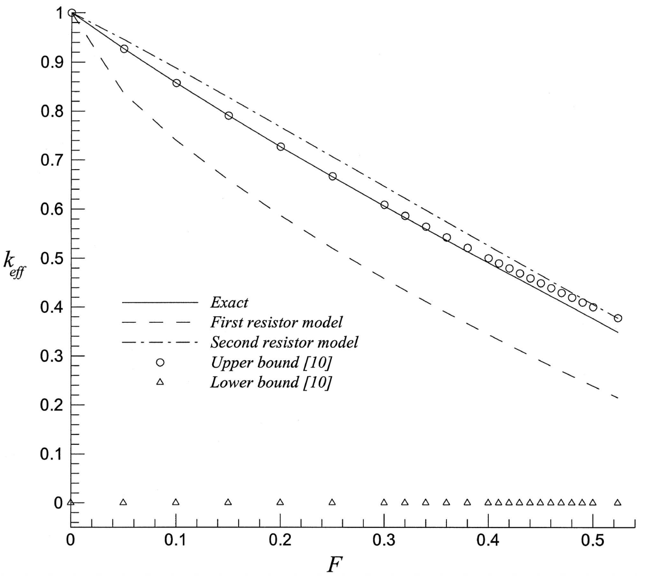

FIGURE 7 Comparison between the predictions of the first and second resistor models with the exact solution and the results of two bounds given in [10] for perfectly insulating spheres

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

2) $k_{i_{\max}} = 0$, 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 $i > i_{\max}$ and $i < i_{\max}$. The first resistor model also shows the same behaviour. For this case, (28) yields the following result:

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

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

$$L_n^{i_{\max}+1} = 1 \,. \tag{48}$$

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:

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

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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