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EFFECTIVE CONDUCTIVITIES OF ISOTROPIC PARTICULATE COMPOSITES**

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EQUIVALENT INHOMOGENEITY METHOD FOR EVALUATING THE EFFECTIVE CONDUCTIVITIES OF ISOTROPIC PARTICULATE COMPOSITES

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The problem of calculating the effective conductivity of isotropic composite materials with periodic or random arrangements of spherical particles is revisited by using the equivalent inhomogeneity method. The approach can be viewed as an extension of classical Maxwell's methodology. It is based on the idea that the effective conductivity of the composite material can be deduced from the effect of the cluster embedded in an infinite space on the far-fields. The key point of the approach is to precisely account for the interactions between all the particles in the cluster that represent the composite material in question. It is done by using a complete, multipole-type analytical solution for the problem of an infinite isotropic matrix containing a finite cluster of isotropic spherical particles, regarded as the finite cluster model of particulate composite. The effective conductivity of the composite is evaluated by applying the "singular-to-singular" re-expansion formulae and comparing the far-field asymptotic behavior with the equivalent inhomogeneity solution. The model allows one to adequately capture the influence of the micro-structure of composite material on its overall properties.

Numerical realization of the method is simple and straightforward. Comparison of the numerical results obtained by the proposed approach with those available in literature (both for periodic and random arrangements) demonstrate its accuracy and numerical efficiency.

1. Introduction

The problem of evaluating the effective conductivities (thermal, electrical, etc.) of particulate composites with spherical particles is well studied, especially for the case of periodic arrangements of particles. In the latter case, following the direction outlined by Lord Rayleigh [Rayleigh 1892], and the techniques developed in the series of follow up papers [Runge 1925; de Vries 1952; Meredith and Tobias 1960; Zuzovsky and Brenner 1977; McPhedran and McKenzie 1978; McKenzie et al. 1978; O'Brien 1979; Sangani and Acrivos 1983], the complete, multipole-type analytical solutions are obtained for three cubic arrays of identical spheres. The basis concept of the method is to directly account for triple periodicity by enforcing periodic conditions on the boundary of a unit cell and to use local expansions for the fields (e.g., temperature) in terms of solid spherical harmonics. Zuzovsky and Brenner [1977] and Sangani and Acrivos [1983] also used a special representation for the temperature as a spatially periodic function.

The conduction through a random suspension of spheres is also relatively well studied. Jeffrey [1973] used the method suggested in [Batchelor 1972; Batchelor and Green 1972], which accounts only for a pair-wise interaction between the particles and employs an information about the particles distribution in the form of simple probability density functions. In [Sangani and Yao 1988; Bonnecaze and Brady 1990;

1991; Zinchenko 1994], the problem of a random dispersion was treated as a triple-periodic problem with random arrangement of the particles in a cubic unit cell. Sangani and Yao [1988] employed a method of multiple expansion developed in [Zuzovsky and Brenner 1977; Sangani and Acrivos 1983], while Bonnecaze and Brady [1990; 1991] used a "Stokesian dynamics"-like approach. Zinchenko [1994] used the boundary integral equation method with the triply periodic Green function and the approximations of the boundary temperature by a series of surface spherical harmonics. In the latter paper an efficient iterative algorithm is constructed to obtain the solution for suspensions with arbitrary conductivity ratio. A different approach was suggested by Kim and Torquato [1990; 1991], who developed a generalized Brownian motion simulation technique.

There is also an extensive literature related to effective medium theories and variational bounds (see [Milton 2002; Torquato 2002] for reviews). In [Batchelor and O'Brien 1977; Suen et al. 1979; Zinchenko 1998] asymptotic relations are suggested for obtaining the effective conductivities of materials with closely packed highly conducting particles. The latest developments include the extension of Rayleigh's method to include the effects of interfacial resistance [Lu and Lin 1996; Cheng and Torquato 1997] and coating. (For the latter problem mostly approximate methods are available; see, e.g., [Lu and Lin 1996; Hashin 2001; Sevostianov and Kachanov 2007].) Moosavi et al. [2003], extended the method of [Zuzovsky and Brenner 1977] to predict the effective thermal properties of a periodic array of multi-coated spheres.

Although those developments have been influenced by the pioneering work of Maxwell [1892], his original methodology was based on a somewhat different concept. Maxwell, who studied electrical conductivity of materials, suggested that a cluster of spherical particles affects the fields at large distances from the cluster in the same way as an equivalent sphere whose conductivity is equal to the effective one. Maxwell did not account for the interactions between the particles, and therefore, he concluded that the method would only be valid for materials with low volume fractions c of particles. In other words, Maxwell's solution was that of order c. Hasselman and Johnson [1987] extended Maxwell's methodology on the case of the composites with interfacial thermal barrier resistance. In [Lu and Lin 1996; Lu 1998] this was used it to estimate the effective conductivities of aligned spheroids dispersions with perfect and imperfect interfaces. Maxwell's methodology was recently extended to multi-phase composites in [McCartney and Kelly 2008; McCartney 2010]. All these papers employed the original Maxwell's methodology that does not account for the interactions between the particles.

In [Mogilevskaya et al. 2010], the Maxwell's concept of the finite cluster was generalized to evaluate the effective transverse properties of linearly elastic, multi-phase unidirectional composites. The key point of the approach was to precisely account for the interactions between all the fibers in the cluster that represents the composite material in question. The same approach was adopted in [Pyatigorets and Mogilevskaya 2011] to evaluate the effective transverse mechanical properties of transversely isotropic viscoelastic composites and in [Koroteeva et al. 2010] to solve the problem of evaluating the effective thermal conductivity of isotropic porous materials. It was demonstrated that the approach allows to adequately capture the influence of the micro-structure of composite material on its overall properties.

In the present paper the approach based on Maxwell's concept of the finite cluster is employed to evaluate the effective thermal conductivity of isotropic suspensions with highly conducting particles that are either in perfect contact with the matrix or possess an interface resistance. The cluster is embedded into an infinite matrix subjected to the prescribed temperature gradient at infinity. The interactions
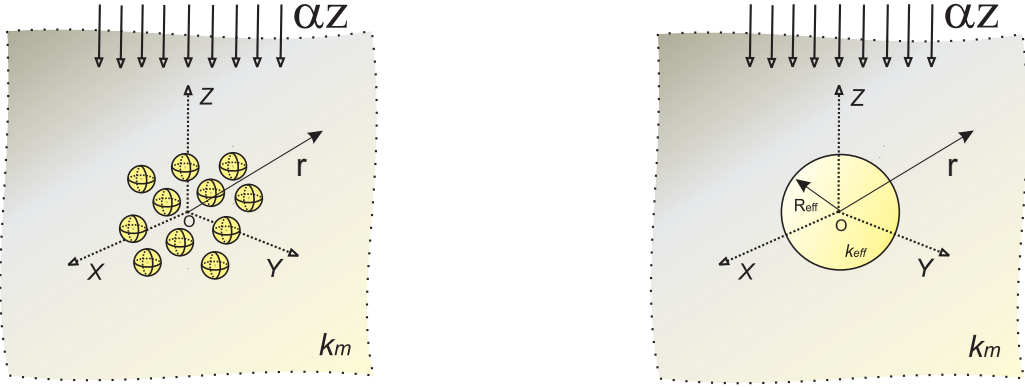
Figure 1. Left: a cluster of spherical particles. Right: an equivalent inhomogeneity.

among all the particles in the cluster are precisely accounted for by using a complete, multipole-type analytical solution for the problem. The effective conductivity of the composite is evaluated by applying the singular-to-singular re-expansion formulae and comparing the far-field asymptotic behavior with the equivalent inhomogeneity solution. The results obtained by the proposed approach are compared with those available in literature both for periodic and random arrangements of particles.

2. The finite cluster model

Consider the two problems shown in Figure 1. The left-hand side of the figure contains a cluster of N non-overlapping spherical particles arranged in an isotropic pattern representative of the composite material. The cluster is embedded in an infinite matrix with the conductivity k_m, and the temperature gradient $T = \alpha z$ is applied at infinity. The particles may have different radii and conductivities, but here, for simplicity, we assume that the particles have the same radii $a_p = 1$ and conductivities k_p.

The particles are either in perfect contact with the matrix or possess an interface resistance. The interface conditions, in the local spherical coordinate system $(O_p, r_p, \theta_p, \varphi_p)$ of the p-th particle ($p = 1, \ldots, N$), are as follows ($r_p = a_p$):

(a) perfect thermal contact:
$$T_p = T_m; \quad k_p \frac{\partial T_p}{\partial r_p} = k_m \frac{\partial T_m}{\partial r_p} \tag{1}$$

(b) interface resistance:
$$\frac{T_p - T_m}{\widetilde{R}} = -k_p \frac{\partial T_p}{\partial r_p}; \quad k_p \frac{\partial T_p}{\partial r_p} = k_m \frac{\partial T_m}{\partial r_p} \tag{2}$$

where T_p, T_m are the temperatures in the p-th particle and matrix, respectively, and $\widetilde{R}$ can be defined through the limiting process described in [Torquato and Rintoul 1995].

Figure 1, right, contains only one (equivalent) inhomogeneity/particle of radius R_{eff} and center O, which coincides with the origin of global coordinate system (O, r, θ, φ) used in both problems. The inhomogeneity is in perfect contact with the matrix and its conductivity coincides with the effective conductivity k_{eff} of the composite material. The choice of the radius of the equivalent inhomogeneity
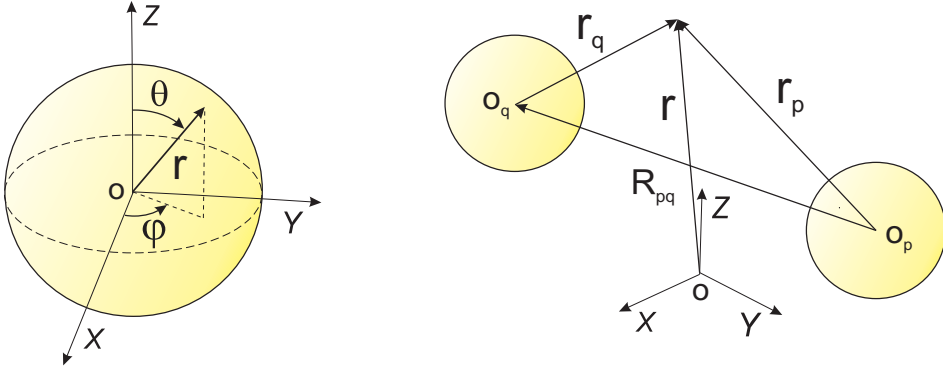
Figure 2. Coordinate systems: local (left) and global (right).

R_{eff} is natural and follows from the definition of the volume fraction c:

$$R_{\text{eff}} = (N/c)^{1/3} \tag{3}$$

Because only isotropic materials are considered in this work, the choice of point O is also natural. For the periodic composites it is located at the center of a unit cell; for random composites it is located at the center of the cube, in which the random arrangement of the particles is generated.

The basic idea of the method is that the temperatures at large distances ($r \gg R_{\text{eff}}$) are the same for the cluster and for the equivalent inhomogeneity. In [Koroteeva et al. 2010] these temperatures are compared using discrete set of points at finite distances away from the cluster. In the present paper the assumption that the temperatures have the same asymptotic behavior is employed, in this case the choice of the center O of the cluster is not even important.

3. Solutions for the two problems

Both problems are governed by Laplace's equation $\Delta T = 0$. The exact expression for the temperature in the matrix for the equivalent inhomogeneity problem in the local spherical coordinate system (O, r, θ, φ) (see Figure 2, left) has the form

$$T_m^*(r) = T_{\text{far}}(r) + B_{10}^* Y_1^0(r) \tag{4}$$

where r is a radius vector, $T_{\text{far}}(r) = \alpha r \cos \theta$, $Y_1^0(r)$ is a solid spherical harmonic (see the Appendix), and B_{10}^* is given by

$$B_{10}^* = -\alpha R_{\text{eff}}^3 \frac{k_{\text{eff}} - k_m}{k_{\text{eff}} + 2k_m} \tag{5}$$

The problem of a finite cluster can be solved using a complete, multipole-type semi-analytical solution. The main steps of the solution can be described as follows. Using the superposition principle, the temperature field inside the matrix $T = T^{(0)}$ can be represented in the form

$$T^{(0)}(r) = T_{\text{far}}(r) + \sum_{q=1}^{N} T_{\text{dist}}^{(q)}(r_q) \tag{6}$$

where $T_{\text{far}} = \alpha r \cos\theta$, while $T_{\text{dist}}^{(q)}(\boldsymbol{r}_q)$ is the disturbance of the field caused by the q-th particle, and the meaning of $\boldsymbol{r}_q$ is clear from Figure 2, right.

The temperature $T_{\text{dist}}^{(q)}(\boldsymbol{r}_q)$ is represented in the form

$$T_{\text{dist}}^{(q)}(\boldsymbol{r}_q) = \sum_{t=1}^{\infty} \sum_{s=-t}^{t} A_{ts}^{(q)} Y_t^s(\boldsymbol{r}_q), \tag{7}$$

where the $A_{ts}^{(q)}$ are unknown complex coefficients.

Applying the re-expansion formula (S2R decomposition, see (28) in the Appendix), the temperature $T^{(0)}(\boldsymbol{r})$ can be expressed in terms of the spherical coordinate system associated with the p-th particle as follows

$$T^{(0)}(\boldsymbol{r}_p) = \alpha[z_c^{(p)} + r_p y_1^0(\boldsymbol{r}_p)] + \sum_{t=1}^{\infty} \sum_{s=-t}^{t} A_{ts}^{(p)} Y_t^s(\boldsymbol{r}_p) + \sum_{t=0}^{\infty} \sum_{s=-t}^{t} a_{ts}^{(p)} y_t^s(\boldsymbol{r}_p), \tag{8}$$

where $z_c^{(p)}$ is the z-th coordinate of the center of the p-th particle.

$$a_{ts}^{(p)} = (-1)^{t+s} \sum_{\substack{q=1 \\ q \neq p}}^{N} \sum_{k=1}^{\infty} \sum_{l=-k}^{k} A_{kl}^{(q)} Y_{k+t}^{l-s}(\boldsymbol{R}_{qp}), \tag{9}$$

where $\boldsymbol{R}_{\mathbf{pq}} = \boldsymbol{r}_p - \boldsymbol{r}_q$ (Figure 2, right).

The temperature inside the p-th particle is represented by the series

$$T^{(p)}(\boldsymbol{r}_p) = \sum_{t=0}^{\infty} \sum_{s=-t}^{t} D_{ts}^{(p)} y_t^s(\boldsymbol{r}_p), \tag{10}$$

where $D_{ts}^{(p)}$ are unknown complex coefficients.

The fluxes $k_p \partial T_p / \partial r_p$, $k_m \partial T_m / \partial r_p$ can be represented by the series similar to those given by Eqs. (8)-(10) by using differentiation rules (25)-(27) in the Appendix and the properties of spherical harmonics. Substituting the series expressions into the boundary conditions (1) (case of perfect thermal contact) or (2) (case of interface resistance) and using some algebra, an infinite set of linear equations can be obtained:

$$\lambda_p \frac{(t-s)! \, (t+s)!}{(a_p)^{2t+1}} A_{ts}^{(p)} + (-1)^{t+s} \sum_{\substack{q=1 \\ q \neq p}}^{N} \sum_{k=1}^{\infty} \sum_{l=-k}^{k} A_{kl}^{(q)} Y_{k+t}^{l-s}(R_{qp}) = -\alpha \delta_{t1} \delta_{s0}. \tag{11}$$

Here $t = 1, 2, \ldots, \; |s| \leq t, \; p = 1, \ldots, N$, and

$$\lambda_p = \frac{\omega_p + (1 + 1/t)(1 + t\gamma)}{\omega_p - 1 - t\gamma}, \tag{12}$$

where $\omega_p = k_p/k_m$, and γ equals 0 for the case of a perfect thermal contact and $\tilde{R}k_p/a_p$ for the case of interface resistance. The number of unknowns in (11) can be reduced by taking into account the equation

$$A_{k,-l}^{(q)} = (-1)^l \overline{A_{kl}^{(q)}},$$

which follows directly from (23) in the Appendix and the fact that the temperature is a real-valued quantity. The resulting system can be truncated by adopting the maximum value of $t = t_{\max}$ and solved in terms of the unknown coefficients $A_{ts}^{(p)}$.

4. Evaluation of the effective conductivity of the material

The temperature $T_{\text{dist}}(\boldsymbol{r})$ in expression (7) can also be re-expanded in a series of spherical harmonics in terms of the spherical coordinate system associated with some sphere with the center O and radius $\|\boldsymbol{r}\|$:

$$T_{\text{dist}}(\boldsymbol{r}) = \sum_{q=1}^{N} T_{\text{dist}}^{(q)}(\boldsymbol{r}_q) = \sum_{t=1}^{\infty} \sum_{s=-t}^{t} B_{ts} Y_t^s(\boldsymbol{r}) \tag{13}$$

Assuming that $\|\boldsymbol{r}\| \geq R_{\text{eff}}$ and using the re-expansion formula (30) from the Appendix, we obtain

$$B_{10} = \sum_{q=1}^{N} A_{10}^{(q)} \tag{14}$$

Using Equations (4)–(6) and (13)–(14) and assuming that the far-field asymptotic behavior of $T^{(0)}$ in (6) is the same as that of $T_m^*(\boldsymbol{r})$ in (4), the coefficient B_{10} can be equated with the coefficient B_{10}^*:

$$B_{10} = B_{10}^* \tag{15}$$

The effective conductivity k_{eff} can then be found from (5). We obtain

$$k_{\text{eff}} = k_m \frac{1 - 2A_{10}^*}{1 + A_{10}^*}, \tag{16}$$

where we have set

$$A_{10}^* = \frac{c}{\alpha} \langle A_{10}^{(p)} \rangle, \tag{17}$$

with

$$\langle A_{10}^{(p)} \rangle = \frac{1}{N} \sum_{p=1}^{N} A_{10}^{(p)} \tag{18}$$

The analytical approach to find k_{eff} allows one to note several interesting facts. It can be seen from expressions (16)–(18) that the effective conductivity k_{eff} is independent of the choice of the center O of the cluster. In addition, it is independent on the value of the temperature gradient at infinity, as the coefficients $A_{10}^{(p)}$ are directly proportional to the value of α (it follows from Eq. (11)). Expressions (16)-(18) involve only the average of the first coefficients from series (7) but, in our model, even those coefficients contain the information about the particles interactions as they are obtained from a complete, multipole-type semi-analytical solution and they are expressed via all the coefficients $A_{ts}^{(q)} (q = 1, \ldots, N, \ q \neq p)$ of Eq. (11). In case of non-interacting particles, the coefficients $A_{10}^{(p)}$ are the same for all particles (see Eqs. (11)-(12) with $a_p = 1$). They can be written as

$$A_{10}^{(p)} = -\alpha \frac{k_p - k_m(1 + \gamma)}{k_p + 2k_m(1 + \gamma)}$$

When the interface conditions are those of perfect thermal contact ($\gamma = 0$), Eq. (16) reduces to classical Maxwell's estimate for the effective conductivity

$$k_{\text{eff}} = k_m \frac{k_p(1+2c) + 2k_m(1-c)}{k_p(1-c) + k_m(2+c)} \tag{19}$$

while for $\gamma \neq 0$ (interface resistance), it reduces to the following expression obtained by in [Hasselman and Johnson 1987]

$$k_{\text{eff}} = k_m \frac{2c[k_p - k_m(1+\gamma)] + k_p + 2k_m(1+\gamma)}{c[k_m(1+\gamma) - k_p] + k_p + 2k_m(1+\gamma)} \tag{20}$$

It is interesting to note that estimate (19) coincides with those by Mori–Tanaka method [Markov 2000, Eq. 5.46] and the Hashin–Shtrikman formula [1962] for coated sphere assemblage [Milton 2002, Eq. 7.7]. Additionally, Equation (20) has the same form as the estimate by the generalized self-consistent model [Lee et al. 2006, Eq. 28].

5. The results

It was demonstrated in [Koroteeva et al. 2010] that the approach based on the equivalent inhomogeneity method allows to adequately capture the influence of the micro-structure of porous material on its overall properties. Here we employed it to evaluate the effective thermal conductivity of isotropic suspensions with conducting particles that are either in perfect contact with the matrix or possess an interface resistance. The maximum number of spherical harmonics used in the numerical simulations reported below was $t_{\max} = 15$ for periodic arrays and $t_{\max} = 11$ for random arrays.

Periodic composites, perfect thermal contact. Consider three cubic arrays of identical particles: the simple cubic array (SC), the body-centered cubic array (BCC), and the face-centered array (FCC) (see Figure 3), and assume that $\alpha = k_p/k_m = 10$.

Table 1 compares our results with those of [Sangani and Acrivos 1983; Cheng and Torquato 1997]. Sangani and Acrivos's results are obtained by using their expression (31), which is of order $O(c^9)$. The coefficients involved in the expression are given by [Sangani and Acrivos 1983, Table 1]. Cheng and Torquato's results [1997] are also of order $O(c^9)$. They are obtained by using expression (3.1) in their paper with the coefficients presented in their Table 1. We have also compared our results with those
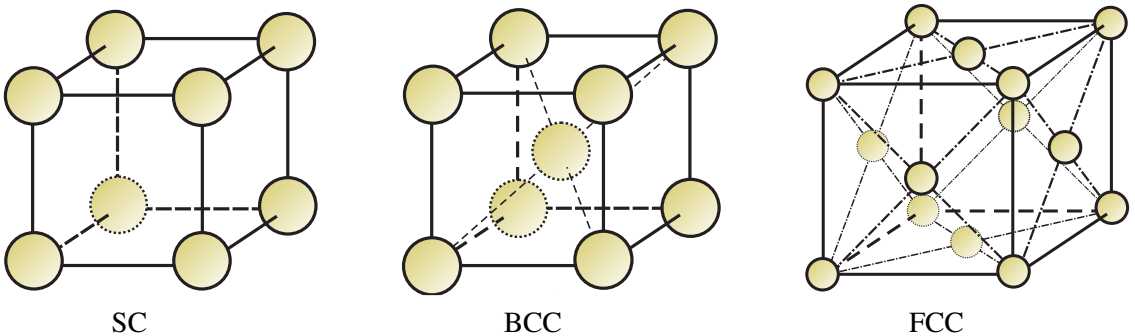

Figure 3. The three cubic arrays: simple (SC), body-centered (BCC), and face-centered (FCC).

$c =$	0.1	0.2	0.3	0.4	0.45	0.5	0.6	0.65	0.7
k_{eff}/k_m for simple cubic array									
Finite cluster model ($N = 8$)	1.24	1.53	1.89	2.35	2.65	3.04			
Finite cluster model ($N = 27$)	1.24	1.53	1.89	2.37	2.69	3.13			
Repetitive unit cell	1.24	1.53	1.89	2.36	2.69	3.14			
[Sangani and Acrivos 1983]	1.24	1.53	1.89	2.36	2.68	3.09			
[Cheng and Torquato 1997]	1.24	1.53	1.89	2.36	2.66	3.05			
Maxwell, (19)	1.24	1.53	1.87	2.29	2.53	2.80			
Differential scheme	1.26	1.59	2.03	2.59	2.92	3.30			
[Hashin and Shtrikman 1962]	1.65	2.34	3.08	3.86	4.28	4.71			
k_{eff}/k_m for BCC array									
Finite cluster model ($N = 9$)	1.24	1.53	1.88	2.32		2.89	3.69	4.26	
Finite cluster model ($N = 35$)	1.24	1.53	1.88	2.33		2.92	3.79	4.44	
Repetitive unit cell	1.24	1.53	1.87	2.30		2.84	3.62	4.19	
[Sangani and Acrivos 1983]	1.24	1.53	1.87	2.30		2.84	3.60	4.12	
[Cheng and Torquato 1997]	1.24	1.53	1.87	2.30		2.84	3.58	4.06	
Maxwell, (19)	1.24	1.53	1.87	2.29		2.80	3.45	3.85	
Differential scheme	1.26	1.59	2.03	2.59		3.30	4.19	4.72	
[Hashin and Shtrikman 1962]	1.65	2.34	3.08	3.86		4.71	5.61	6.09	
k_{eff}/k_m for FCC array									
Finite cluster model ($N = 14$)	1.24	1.53	1.88	2.32		2.90	3.71		5.05
Finite cluster model ($N = 63$)	1.24	1.53	1.88	2.32		2.90	3.73		5.13
Repetitive unit cell	1.24	1.53	1.87	2.29		2.83	3.56		4.69
[Sangani and Acrivos 1983]	1.24	1.53	1.87	2.29		2.82	3.50		4.45
[Cheng and Torquato 1997]	1.24	1.53	1.87	2.29		2.83	3.55		4.60
Maxwell, (19)	1.24	1.53	1.87	2.29		2.80	3.45		4.32
Differential scheme	1.26	1.59	2.03	2.59		3.30	4.19		5.29
[Hashin and Shtrikman 1962]	1.65	2.34	3.08	3.86		4.71	5.61		6.58

Table 1. Normalized effective conductivity k_{eff}/k_m for the simple (top), body-centered (middle) and face-centered (bottom) cubic arrays.

obtained by direct solution of a triple-periodic problem that employs the concept a repetitive unit cell (RUC) (the "direct substitution method", according to the terminology of Sangani and Acrivos). The latter solution also involves a series of spherical harmonics to approximate the temperature on every sphere in the unit cell, and the value of t_{max} was set to be $t_{\text{max}} = 25$. For the case of a simple cubic array, the RUC results are in perfect agreement with the Brownian motion simulation data in [Kim and Torquato 1991, Table II]. We also include estimates by the differential scheme [Markov 2000, Eq. 10.42] and the classical results of Maxwell given in (19). The latter estimate coincides with those by Mori–Tanaka method, the
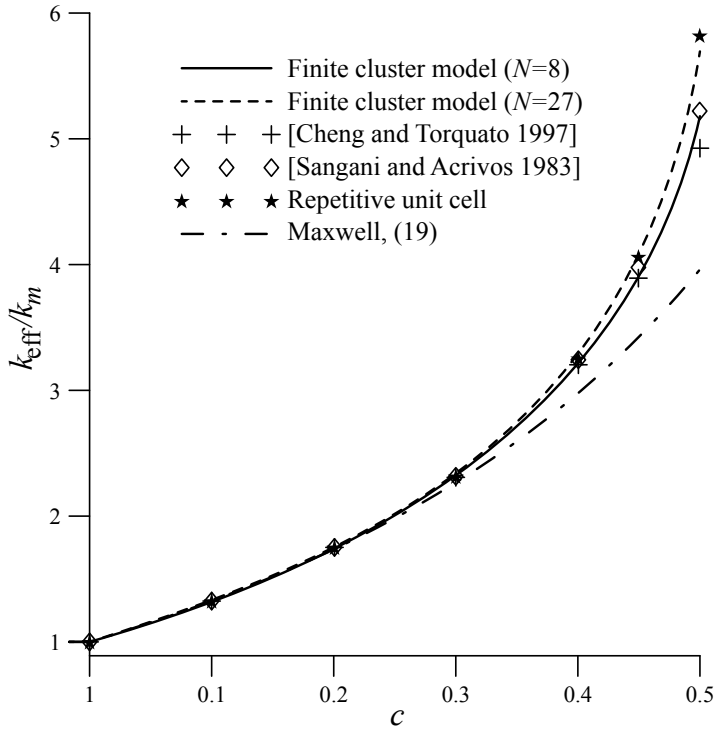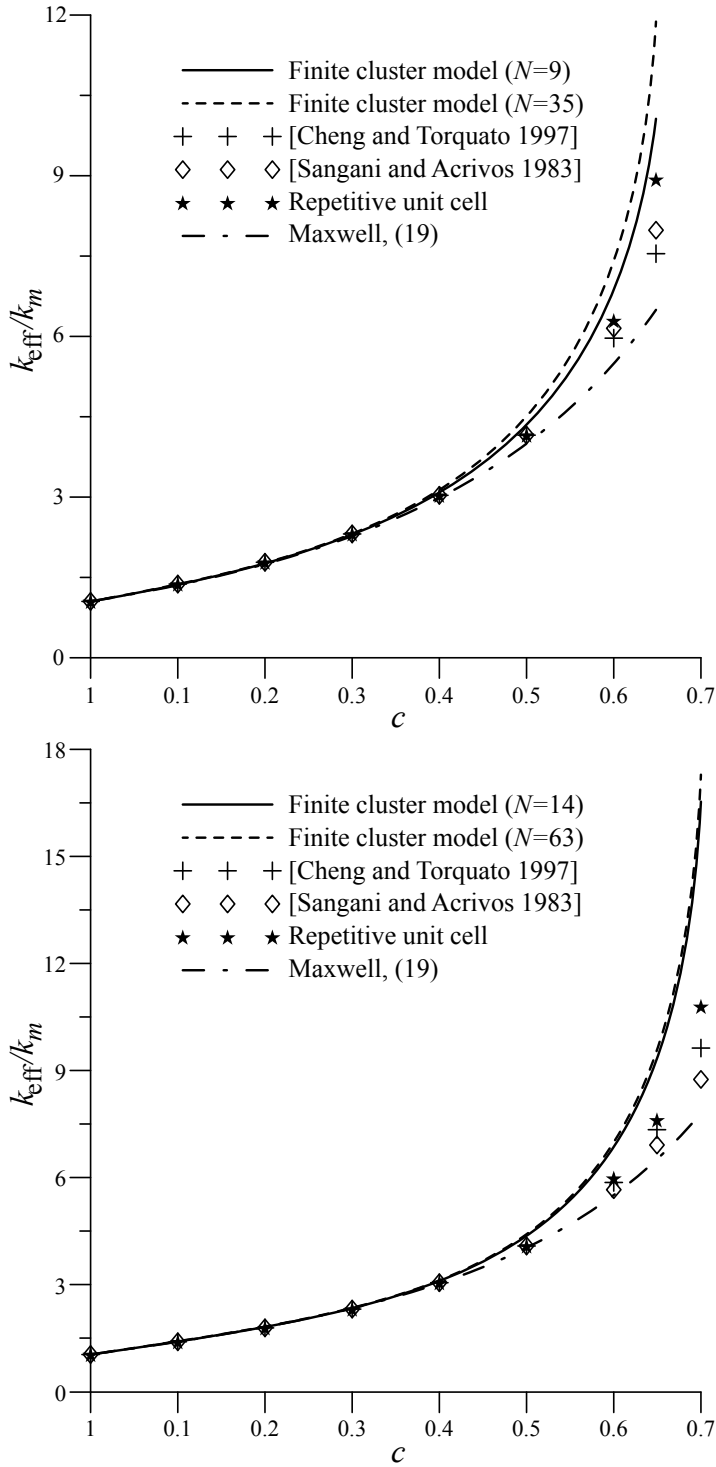
Figure 4. Normalized effective conductivity versus volume fraction for the simple cubic case; $k_p/k_m = 1000$.

Hashin–Shtrikman formula for coated sphere assemblage and with the lower bounds of [Hashin and Shtrikman 1962]. Hashin and Shtrikman's upper bounds are included in the tables (last line).

The results for the SC and BCC lattices (top and middle in Table 1) are in good agreement with the those obtained by using a RUC concept and with the results of [Sangani and Acrivos 1983; Cheng and Torquato 1997] (especially for the SC case). However for the FCC lattice, the results agree well only for volume fractions $c < 0.6$. Classical Maxwell estimate underpredicts the values of k_{eff}, while the estimate by the differential scheme overpredicts those values.

A similar trend can be observed on Figures 4 and 5, where the results for the periodic arrays are presented for more severe case, $\alpha = k_p/k_m = 1000$. One can also notice that for the case of very high volume fractions, there is a significant scatter in the previously available results. As in the previous case, the results for the SC lattice are in excellent agreement with the RUC results.

Random composites: perfect thermal contact. We apply our approach to isotropic materials with random arrangement of highly conducting particles ($k_p/k_m = 1000$). To generate the cluster with a random distribution of particles, the method of molecular dynamics of growing particles is used [Sangani and Yao 1988]. To have representative data set, the simulation was executed 30 times with $N = 40$ particles generated in every run. In the case of near-touching spheres, the minimal gap between particles 0.02 was allowed. Our results are compared with those from [Kim and Torquato 1991; Bonnecaze and Brady 1990], who gave the estimated for the case of $\alpha = \infty$.
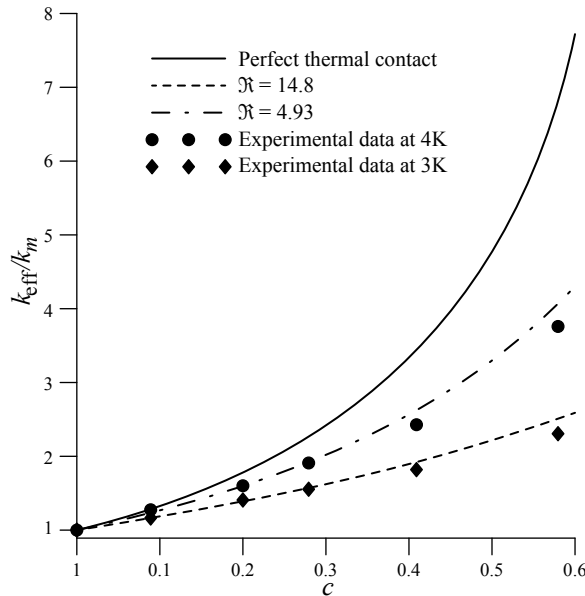
Figure 5. Normalized effective conductivity versus volume fraction for the BCC (top) and FCC (bottom) cases; $k_p/k_m = 1000$.

		k_{eff}/k_m					
	$c =$	0.1	0.2	0.3	0.4	0.5	0.6
FCM	$(N = 40)$	1.34	1.80	2.44	3.40	4.76	7.84
[Kim and Torquato 1991]	$(\alpha = \infty)$	1.34	1.83	2.48	3.42	4.78	8.32
[Bonnecaze and Brady 1990]	$(\alpha = \infty)$	1.35	1.82	2.53	3.59	4.97	8.85
Maxwell	$(\alpha = 1000)$	1.33	1.75	2.28	2.99	3.98	5.47

Table 2. Normalized effective conductivity (random array).

The results presented in Table 2 show that, as expected, our estimates are slightly below those obtained by Kim and Torquato as well as Bonnecaze and Brady for the superconducting spheres ($\alpha = \infty$). It can also be seen that the classical Maxwell estimate underpredicts the value of the effective conductivity for larger volume fractions.

Random composites: interface resistance. In this case the materials with random distribution of particles which possess interfacial resistance are considered. We obtained the results for a cluster of $N = 20$ particles with conductivities $\alpha = k_p/k_m = 1000$, and then compare them to the experimental data of Araujo and Rosenberg [1976], who measured the effective thermal conductivity of random dispersions of metallic spheres in an epoxy matrix. The same data were used by Torquato and Rintoul [1995] to compare their bounds. In Araujo and Rosenberg's experiment, the resistance at the boundaries of particles appears at low temperatures (less than 20 K) due to an acoustic mismatch. As one can see from Figure 6, the estimates of our approach agree well with the experimental results. Comparing with the

Figure 6. Normalized effective conductivity versus volume fraction: comparison with experimental data ($k_p/k_m = 1000$, $\mathfrak{R} = a_p/\widetilde{R}$).

results for the perfectly bonded particles, one can see a noticeable drop in the effective conductivity due to interfacial resistance.

6. Discussion and summary

In this paper, the approach based on the concept of the equivalent inhomogeneity is employed to evaluate the effective thermal conductivities of isotropic suspensions with highly conducting particles that are either in perfect contact with the matrix or possess an interface resistance. The approach can be viewed as an extension of the classical Maxwell's methodology, but, with the interactions between the particles in the cluster precisely accounted for. The approach has been tested on a number of benchmark solutions for suspensions with highly conducting particles. The numerical results indicate that, in the latter case, the classical Maxwell methodology accurately predict the effective thermal conductivity only for relatively low volume fractions. In the case of high volume fractions, the interactions between the particles must be taken into account to obtain better estimates. The approach is especially attractive for study the materials with random statistically homogeneous micro-structure, as it does not require the concept of a repetitive unit cell, which itself violates the notion of randomness. The analytical solution of the cluster problem is also simpler than the solution of the corresponding triple-periodic problem. In the present paper, the representative clusters for random composites were generated inside the cube, but they could also be generated inside the sphere (the influence of the shape and the size of the cluster will be the topic of future research). The method could be useful for estimation of the overall properties of the material based on the small samples (obtained by e.g., scanning electron micrograph, X-ray micro-tomography, etc). The approach can be extended on the case of macroscopically anisotropic materials containing isotropic particles, if the concept of an equivalent anisotropic inhomogeneity is adopted.

Appendix: solid harmonics

We adopt the following representation of the solid spherical harmonics (see [Kushch 1985]):

$$y_t^s(\boldsymbol{r}) = \frac{r^t}{(t+s)!}\chi_t^s(\theta,\varphi) \text{ and } Y_t^s(\boldsymbol{r}) = \frac{(t-s)!}{r^{t+1}}\chi_t^s(\theta,\varphi) \tag{21}$$

where $y_t^s(\boldsymbol{r})$ and $Y_t^s(\boldsymbol{r})$ are the regular and singular solid spherical harmonics, respectively, and $\boldsymbol{r} = (x, y, z)$ is the radius vector. Here,

$$\chi_t^s(\theta,\varphi) = P_t^s(\cos\theta)\exp(is\varphi) \tag{22}$$

are the surface spherical harmonics, $t \geq 0$, $|s| \leq t$, $i = \sqrt{-1}$, and $P_t^s(\cos\theta)$ are the associated Legendre polynomials [Hobson 1955].

The normalization adopted in (21) is convenient in many aspects.

$$y_t^{-s}(\boldsymbol{r}) = (-1)^s \overline{y_t^s(\boldsymbol{r})}, \quad Y_t^{-s}(\boldsymbol{r}) = (-1)^s \overline{Y_t^s(\boldsymbol{r})} \tag{23}$$

For $t = 1$ we get

$$y_1^0(\boldsymbol{r}) = z, \quad y_1^1(\boldsymbol{r}) = \frac{1}{2}(x+iy), \quad y_1^1(\boldsymbol{r}) = -\frac{1}{2}(x-iy). \tag{24}$$

and the following differentiation rule can be used:

$$D_1 y_t^s = y_{t-1}^{s-1}, \quad D_2 y_t^s = -y_{t-1}^{s+1}, \quad D_3 y_t^s = y_{t-1}^s; \tag{25}$$
$$D_1 Y_t^s = Y_{t+1}^{s-1}, \quad D_2 Y_t^s = -Y_{t+1}^{s+1}, \quad D_3 Y_t^s = Y_{t+1}^s;$$

where D_i are the differential operators

$$D_1 = \left(\frac{\partial}{\partial x} - i \frac{\partial}{\partial y} \right), \quad D_2 = \overline{D_1} = \left(\frac{\partial}{\partial x} + i \frac{\partial}{\partial y} \right), \quad D_3 = \frac{\partial}{\partial z}. \tag{26}$$

Operators D_2 and D_3 also satisfy

$$(D_2)^s (D_3)^{t-s} \left(\frac{1}{r} \right) = (-1)^t Y_t^s (\boldsymbol{r}) \tag{27}$$

The solid spherical harmonics can be re-expanded in terms of spherical coordinates of a particular local coordinate system using the re-expansion formulae. Let $\boldsymbol{r}_p = \boldsymbol{R}_{pq} + \boldsymbol{r}_q$ (Figure 2, left). In our notations, the re-expansions take the simplest possible form:
singular to regular (S2R):

$$Y_t^s(\boldsymbol{r}_p) = \sum_{k=0}^{\infty} \sum_{l=-k}^{k} (-1)^{k+l} Y_{t+k}^{s-l}(\boldsymbol{R}_{pq}) y_k^l(\boldsymbol{r}_q), \quad \|\boldsymbol{r}_q\| < \|\boldsymbol{R}_{pq}\|; \tag{28}$$

regular to regular (R2R):

$$y_t^s(\boldsymbol{r}_p) = \sum_{k=0}^{t} \sum_{l=-k}^{k} y_{t-k}^{s-l}(\boldsymbol{R}_{pq}) y_k^l(\boldsymbol{r}_q); \tag{29}$$

singular to singular (S2S):

$$Y_t^s(\boldsymbol{r}_p) = \sum_{k=t}^{\infty} \sum_{l=-k}^{k} (-1)^{t+k+s+l} y_{k-t}^{s-l}(\boldsymbol{R}_{pq}) Y_k^l(\boldsymbol{r}_q), \quad \|\boldsymbol{r}_q\| > \|\boldsymbol{R}_{pq}\|. \tag{30}$$

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