

Effective conductivity of a composite of poly-dispersed spherical particles in a linear continuum

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Rayleigh's method is used to find the electric potentials of a composite of poly-dispersed spherical particles in a linear continuum in an external electric field. Based on the solutions of potentials, analytical formula for the effective electric conductivity is derived. Based on the formula, several factors, such as the number of spherical inclusions, the spatial distribution of the spheres, the contrast ratio σ_i/σ_h (where, σ_i and σ_h are the conductivities of the spherical inclusion and the host medium, respectively) and volume fraction of the inclusions, are discussed. Our results show that at high volume fraction, the effective conductivity is also affected by the spatial distribution of the inclusions. © 2003 Kluwer Academic Publishers

1. Introduction

The transport properties of composite media have been extensively studied in recent years and used in engineering applications [1–4]. Several methods have been developed to estimate the effective physical properties of composites with two and three dimensions lattice models [5–8], such as Rayleigh's method, transformation method, Fourier series method and so on [9, 10]. On the basis of these researches, where the inclusion particles are regularly arranged, one can further investigate the effective conductivity of composites with poly-dispersed inclusions, which are very common in practical composite systems. We shall investigate the problem of the effective response of a composite with inclusion particles randomly embedded in a unit cell in a continuous matrix, and whether the effective response is related to the distribution of the particles in the matrix. In order to discuss these problems, we have to consider factors such as the volume fraction of the inclusion particles, contrast ratio σ_i/σ_h (where, σ_i and σ_h are the linear conductivity coefficients of spherical inclusion and host medium, respectively), the number of particles, the positions of particles situated in the unit cell and so on. It is well known that the volume fraction of inclusions, the geometry of particles and the contrast ratio σ_i/σ_h are important factors affecting the effective conductivity of a composite, but there is still the question of how the distribution of particles affects the effective conductivity. When an external electric field is applied to a composite, the variation of the local field at different points inside the composite is quite complex because of the particle polarization and interaction, which is related to the microstructure (or geometry of particles)

and the distribution of inclusions beside the intrinsic physical properties of the inclusion and the host. For instance, the inclusion particle size dispersion and clustering are also important factors. In the present work, we will derive the formula for the effective conductivity of a composite of poly-dispersed spherical particles randomly embedded in a linear continuum by means of Rayleigh's method and discuss the effects of particle distribution.

We apply Rayleigh's method to solve the potentials within a composite with poly-dispersed spherical particles randomly immersed in a unit cell. The advantage of this method is that it is capable of producing highly accurate numerical results for the effective response of composites by the aid of modern computers. The pioneering method was proposed by Rayleigh [11], who, in order to calculate the effective conductivity, studied in great detail the two and three dimension systems of the square array and cubic lattice. Subsequently, Rayleigh's method was developed to estimate the effective response of composites with periodic microstructure by several authors [12–14]. For example, Suen *et al.* improved this method for solving the lattice model to arbitrary order when it was used to estimate the thermal conductivity of composites [15]. Lam worked with Rayleigh's method and derived an iterative equation to calculate the effective magnetic permeability of a simple cubic lattice [16]. Gu *et al.* developed Rayleigh's method by means of Green's function and perturbation expansion to deal with the linear and nonlinear effective response of composites with periodic structure [17]. Almost all of these works are devoted to composites with regular arrangement of particles embedded in a

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lattice. The version of Rayleigh's method developed by Gu *et al.* is suitable for solving our present composite problem since the polarization of poly-dispersed random particles is considered in his model. Here, we shall make use of the same method in this study.

The structure of this paper is as follows. In Section 2, Rayleigh's identities for three-dimensional linear random composites with spherical inclusions are derived. In Section 3, a formula for effective conductivity is given, and the effects of various factors on the effective coefficients are investigated. The last section contains a brief conclusion.

2. Rayleigh's method for linear random composite

We consider the electric fields and potentials within a linear conducting composite medium in a unit cubic cell in which spherical particles are randomly suspended. We assume that the constitutive equations of current density J and local electric field E in the host and inclusion regions are

$$J_\alpha = \sigma_\alpha E, \quad \alpha = i, h. \quad (1)$$

The subscripts $\alpha = i, h$ denote the quantities in inclusion and host regions, respectively. σ denotes the conductivity. The electrostatic equations are supplemented by

$$\nabla \cdot J = 0 \quad (2)$$

$$\nabla \times E = 0 \quad (3)$$

The boundary conditions are given by the continuity of the potential and the normal component of current density across the inclusion-matrix interfaces.

Let N different inclusion spheres with different radii a_N be randomly situated inside a unit cube. The unit cube, with its contents, is replicated periodically in three directions of space to form an infinite medium. The replicated cells are labeled by subscript j . In each cubic cell, the regions of inclusion spheres and host are denoted by Ω_i and Ω_h , respectively. Suppose the origin of the coordinate system is located at the center of the k th sphere in the zeroth cell, a set of Laplace equations of the local electric potentials φ_α ($\alpha = i, h$) are given by Equations 1 and 2 since Equation 3 implies the existence of local potentials. Clearly, the general solutions of potentials in the inclusion and host regions can be directly obtained:

$$\begin{aligned} \varphi_i(\rho_k, \theta_k, \vartheta_k) &= C_0^k + \sum_{l=1}^{\infty} \sum_{m=0}^l \rho_k^l P_l^m(\cos \theta_k) [C_{l,m}^{1,k} \cos(m\vartheta_k) \\ &\quad + C_{l,m}^{2,k} \sin(m\vartheta_k)], \quad \text{in } \Omega_i \end{aligned} \quad (4)$$

$$\begin{aligned} \varphi_h(\rho_k, \theta_k, \vartheta_k) &= A_0^k + \sum_{l=1}^{\infty} \sum_{m=0}^l \{ \rho_k^l P_l^m(\cos \theta_k) [A_{l,m}^{1,k} \cos(m\vartheta_k) \\ &\quad + A_{l,m}^{2,k} \sin(m\vartheta_k)] + \rho_k^{-l-1} P_l^m(\cos \theta_k) \\ &\quad \times [B_{l,m}^{1,k} \cos(m\vartheta_k) + B_{l,m}^{2,k} \sin(m\vartheta_k)] \}, \quad \text{in } \Omega_h. \end{aligned} \quad (5)$$

where $k = 1, 2, \dots, N \cdot P_l^m(\cos \theta)$ is an associated Legendre function. The position vector \vec{r}_k of a point is expressed in spherical coordinates as $(\rho_k, \theta_k, \vartheta_k)$. The polar angles θ_k and ϑ_k are from the z -axis and the x -axis, respectively. Considering the boundary conditions of the potential and normal current density across the surface of the sphere, we can get a set of relationships between the unknown coefficients:

$$C_0^k = A_0^k$$

$$C_{l,m}^{t,k} = \frac{2l+1}{l(1-\sigma_k/\sigma_h)} \frac{B_{l,m}^{t,k}}{a_k^{2l+1}}, \quad t = 1, 2 \quad (6)$$

$$A_{l,m}^{t,k} = \left(\frac{2l+1}{l(1-\sigma_k/\sigma_h)} - 1 \right) \frac{B_{l,m}^{t,k}}{a_k^{2l+1}}, \quad t = 1, 2. \quad (7)$$

where σ_k and a_k are the conductivity coefficient and radius of the k th sphere, respectively.

If a uniform external electric field E_0 is applied to the unit cell along the z -axis, the particle polarization will induce a source term due to discontinuity of the normal electric field on the surface of the sphere. The induced surface charge density is calculated as

$$\begin{aligned} Q_k(\theta_k, \vartheta_k) &= \left(\frac{\partial \varphi_i}{\partial \rho_k} - \frac{\partial \varphi_h}{\partial \rho_k} \right)_{\rho_k=a_k} \\ &= \sum_{l=1}^{\infty} \sum_{m=0}^l \frac{2l+1}{a_k^{l+2}} P_l^m(\cos \theta_k) \\ &\quad \times [B_{l,m}^{1,k} \cos(m\vartheta_k) + B_{l,m}^{2,k} \sin(m\vartheta_k)], \\ &\quad k = 1, 2, \dots, N. \end{aligned} \quad (8)$$

Considering the applied electric field and the induced source term, we can derive another expression of the potentials in the composite medium by means of Green's function:

$$\begin{aligned} \varphi(\rho_k, \theta_k, \vartheta_k) &= -E_0 z_k + \sum_{j=0}^{\infty} \sum_{s=1}^N \frac{1}{4\pi} \int \int_{\Sigma_s} Q_s(\theta_s, \vartheta_s) \\ &\quad \times G(\vec{r}_{ksj} - \vec{r}_s) dS_s, \quad k = 1, 2, \dots, N. \end{aligned} \quad (9)$$

where $G(\vec{r}_{ksj} - \vec{r}_s) = (|\vec{r}_{ksj} - \vec{r}_s|)^{-1}$ is the general Green's function of Laplace equation in three dimensions. $\vec{r}_{ksj} = \vec{r}_k - \vec{R}_{ksj}$, where the vector \vec{R}_{ksj} is the position vector of the center of the s th sphere in the j th cell. The vector \vec{r}_s is from the center of the s th sphere to the surface of the same sphere in the j th cell. If the position vector \vec{r}_k lies in the host region, the potential can be obtained by integrating Equation 9 to obtain

$$\begin{aligned} \varphi_h(\rho_k, \theta_k, \vartheta_k) &= -E_0 z_k + \sum_{j=0}^{\infty} \sum_{s=1}^N \sum_{l=1}^{\infty} \sum_{m=0}^l \rho_{ksj}^{-l-1} P_l^m(\cos(m\theta_{ksj})) \\ &\quad \times [B_{l,m}^{1,k} \cos(m\vartheta_{ksj}) + B_{l,m}^{2,k} \sin(m\vartheta_{ksj})] \\ &\quad k = 1, 2, \dots, N. \end{aligned} \quad (10)$$

where $\rho_{ksj} = |\vec{r}_{ksj}|$.

From Equations 5 and 10, we have obtained two expressions of potentials in the host region. It is clear that, at the same position in host, the local electric fields along the z -direction obtained from Equations 5 and 10 should be equal. Using this fact, we can establish a set of Rayleigh's identities about the unknown coefficients $B_{l,m}^{1,k}$ and $B_{l,m}^{2,k}$:

$$\begin{aligned} & \sum_{l=1}^{\infty} \sum_{m=0}^l (l+m) \rho_k^{l-1} P_l^m(\cos(\theta_k)) \\ & \times [A_{l,m}^{1,k} \cos(m\vartheta_k) + A_{l,m}^{2,k} \sin(m\vartheta_k)] \\ & = -E_0 - \sum_{j=0}^{\infty} \sum_{s=1}^N \sum_{l=1}^{\infty} \sum_{m=0}^l (l-m+1) \rho_{ksj}^{-l-2} \times P_{l+1}^m \\ & \times (\cos(\theta_{ksj})) [B_{l,m}^{1,k} \cos(m\vartheta_{ksj}) + B_{l,m}^{2,k} \sin(m\vartheta_{ksj})], \\ & \quad k = 1, 2, \dots, N. \quad (11) \end{aligned}$$

where $A_{l,m}^{1,k}$ and $A_{l,m}^{2,k}$, from Equation 7, can be linearly expressed in terms of the unknown coefficients $B_{l,m}^{1,k}$ and $B_{l,m}^{2,k}$. The subscript j' denotes that the inclusion sphere with the origin of coordinates is excluded from the zeroth cell.

In order to solve the unknown coefficients from the linear Equations 11, we define and calculate the lattice sums:

$$W_{k,s}^{1,l,m}(q) = \sum_{j'=0}^{\infty} \rho_{ksj'}^{-l-2} P_{l+1}^m(\cos \theta_{ksj'}) \cos(m\vartheta_{ksj'}), \quad (12)$$

$$W_{k,s}^{2,l,m}(q) = \sum_{j'=0}^{\infty} \rho_{ksj'}^{-l-2} P_{l+1}^m(\cos \theta_{ksj'}) \sin(m\vartheta_{ksj'}), \quad (13)$$

where the point $q = (x_k, y_k, z_k)$ is in the host region with the origin of coordinates located at the center of the k th sphere in the zeroth cell.

$$\rho_{ksj} = [(x_k - x_{ksj})^2 + (y_k - y_{ksj})^2 + (z_k - z_{ksj})^2]^{1/2}.$$

where x_{ksj} , y_{ksj} and z_{ksj} are the coordinate components of the s th sphere in the j th cell under the coordinate system of the k th sphere in the zeroth cell.

$$\cos(\theta_{ksj}) = (z_k - z_{ksj})/\rho_{ksj},$$

$$\sin(\vartheta_{ksj}) = (y_k - y_{ksj})/r,$$

$$\cos(\vartheta_{ksj}) = (x_k - x_{ksj})/r,$$

where $r = [(x_k - x_{ksj})^2 + (y_k - y_{ksj})^2]^{1/2}$.

Then, Equation 11 is written as

$$\begin{aligned} & \sum_{l=1}^{\infty} \sum_{m=0}^l (l+m) P_{l-1}^m(\cos(\theta_k)) \frac{\rho_k^{l-1}}{a_k^{2l+1}} \left(\frac{2l+1}{l(1-\sigma_k/\sigma_h)} - 1 \right) \\ & \times (B_{l,m}^{1,k} \cos(m\vartheta_k) + B_{l,m}^{2,k} \sin(m\vartheta_k)) \\ & = -E_0 - \sum_{s=1}^N \sum_{l=1}^{\infty} \sum_{m=0}^l (l-m+1) (W_{k,s}^{1,l,m}(q) B_{l,m}^{1,s} \\ & + W_{k,s}^{2,l,m}(q) B_{l,m}^{2,s}), \quad \text{where } k = 1, 2, \dots, N. \quad (14) \end{aligned}$$

We should point that if the position vector \vec{r}_k is located in the inclusion region, we can obtain the same Rayleigh's identities as Equation 11 by integrating Equation 9 [17]. Thus, using the least-square method, we can invert the unknown coefficients from the linear Equation 14 after we obtain the lattice sums by choosing enough points in the medium. In calculating the effective conductivity in the next section, we shall sum up to $l=3$ in Equation 14 and the number of points q chosen is twice that of the unknown coefficients.

3. Effective conductivity

In this section, we start to derive the formula of the effective conductivity of a random composites by making use of the following formula [18]

$$\frac{1}{V} \int_v (J - \sigma_h E) dV = \langle J \rangle - \sigma_h \langle E \rangle, \quad (15)$$

where V is the volume of $\Omega_h + \Omega_i$, and $\Omega_i = \sum_{k=1}^N \Omega_k$, where Ω_k represents the region occupied by the k th sphere in unit cell. The bracket $\langle \cdot \cdot \rangle$ denotes the spatial average over the whole unit cell. For a composite medium with spherical inclusions randomly embedded in a cubic cell, the effective conductivity can be defined as

$$\langle J \rangle = \sigma_e \langle E \rangle, \quad (16)$$

where σ_e is the effective conductivity of the random composite. In order to derive the formula of the effective conductivity, substitute Equations 1 and 16 into Equation 15, thus simplifying Equation 15 as follows:

$$\frac{1}{V} \sum_{k=1}^N \int_{\Omega_k} (\sigma_k - \sigma_h) E dV = (\sigma_e - \sigma_h) \langle E \rangle. \quad (17)$$

Furthermore, we can derive the effective conductivity σ_e by using the definition $\langle E \rangle_k = \frac{1}{\Omega_k} \int_{\Omega_k} E dV$, hence

$$\sigma_e = \sigma_h + \sum_{k=1}^N f_k (\sigma_k - \sigma_h) \frac{\langle E_z \rangle_k}{\langle E_z \rangle}. \quad (18)$$

where f_k is the volume fraction of the k th sphere in the unit cell. From the electric potential in the inclusion sphere, i.e. Equation 4, the average electric field along the z -axis $\langle E_z \rangle_k$ can be calculated, giving $\langle E_z \rangle_k = -C_{1,0}^{1,k}$.

In the dilute limit, because the average electric field along the z -axis $\langle E_z \rangle$ can be regarded as equal to the external field E_0 , the formula for effective conductivity is thus

$$\sigma_e/\sigma_k = 1 - \sum_{k=1}^N f_k (\sigma_k/\sigma_h - 1) C_{1,0}^{1,k}/E_0 \quad (19)$$

To calculate the average field along the z -axis $\langle E_z \rangle$ for high concentration of inclusion spheres, in order,

we consider a spherical sample of the composite with unknown effective conductivity σ_e immersed in a homogenous matrix with conductivity σ_h . The same external electric field E_0 is applied to the composite sample along the z -axis. The averaged electric field $\langle E_z \rangle$ is given by [18]

$$\langle E_z \rangle = \frac{3E_0}{2 + \sigma_e/\sigma_h}. \quad (20)$$

Substituting Equation 20 into Equation 18, the formula for effective conductivity is derived.

$$\sigma_e/\sigma_h = \left[1 - \frac{2}{3E_0} \sum_{k=1}^N f_k(\sigma_k/\sigma_h - 1)C_{1,0}^{1,k} \right] \times \left[1 + \frac{1}{3E_0} \sum_{k=1}^N f_k(\sigma_k/\sigma_h - 1)C_{1,0}^{1,k} \right]^{-1}. \quad (21)$$

Here, we note that, Equation 19 can be obtained from Equation 21 if the volume fraction f_k is small so that $|\frac{1}{3E_0} \sum_{k=1}^N f_k(\sigma_k/\sigma_h - 1)C_{1,0}^{1,k}| \ll 1$.

Next we shall investigate the effective conductivity of random composites by means of Equation 21. To investigate the effects of various factors of the spheres on the effective conductivity, we consider the following three cases. Case (A): the number of spheres is varied for a fixed total volume fraction of inclusion spheres. For case (B): the volume fraction and the contrast ratio σ_k/σ_h are varied for fixed number and positions of spheres in the unit cell. In case (C): given the total volume fraction and the number of the inclusion spheres, the effects due to different spatial distribution are investigated. In each case, the samples are from different realizations of dispersed spheres randomly embedded in a cubic cell with edge length 100 units, which are generated by standard Monte Carlo simulation techniques.

Case (A): We discuss this case in low and higher concentration of random spheres in order to investigate the effects of the different number of inclusion spheres on the effective conductivity. In the dilute limit, the total volume fraction $f = 0.042$ is chosen. Different samples of five, ten and twenty-five spheres are used. The spheres in each sample have the same radius and conductivity. In Fig. 1, the curves of the parameters σ_e/σ_h against $\ln(\sigma_k/\sigma_h)$ are plotted. It is clear that the number of the dispersed spheres does not affect the effective conductivity. This because the polarization of the inclusions is small in dilute limit. For higher concentration, with volume fraction equals to 0.26, we have considered samples having fifteen, twenty and twenty-five spheres. Curves of σ_e/σ_h against $\ln(\sigma_k/\sigma_h)$ are shown in Fig. 2. Obviously, the effects are important when the contrast ratio σ_k/σ_h is larger than unity. The effective conductivity increases as the contrast ratio σ_k/σ_h increases, as long as the ratio is greater than unity. This indicates that the interaction due to inclusion polarization is enhanced. That is, the conductivity of the inclusions plays an important role in the effective conductivity due to particle polarization. On the other hand, the differences in the effective conductivity are very small when the parameter σ_k/σ_h is less than unity, because the interaction of particle polarization is impaired. However, as shown in Fig. 2, it is difficult to find any regularity about the effects of the different number of spheres on the effective conductivity. It may imply that the positions of the spheres in the unit cell is another important factor. That is considered in case(C).

Case (B): For fixed number and fixed positions of the spheres, the volume fraction and the contrast ratio σ_k/σ_h are varied. A sample having twenty-five spheres is employed. The plots of σ_e/σ_h against $\ln(\sigma_k/\sigma_h)$ with total volume fractions at 0.042, 0.26 and 0.43 are shown in Fig. 3. As can be seen, the volume fraction dramatically affects the effective conductivity. When the contrast parameter σ_k/σ_h is greater than unity, the effective

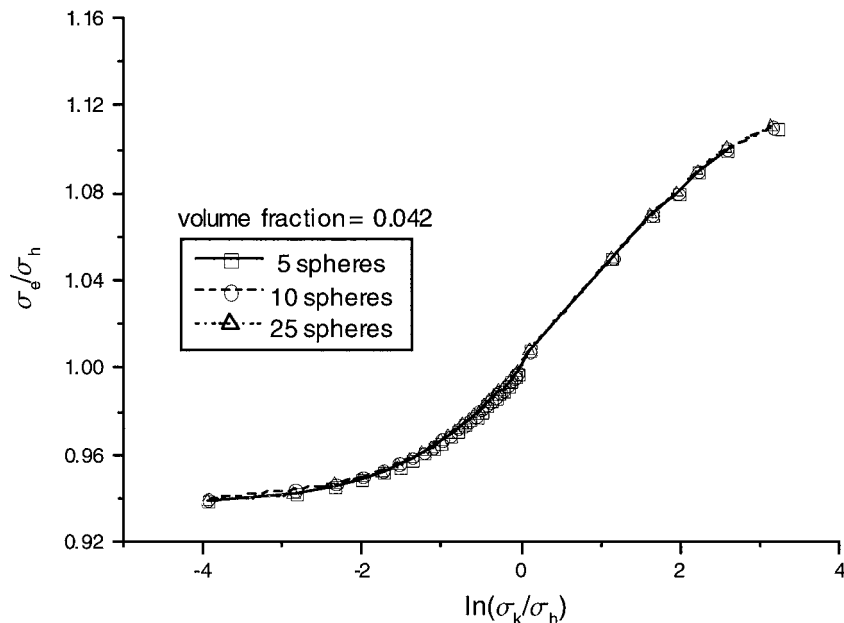


Figure 1 For different number of spheres, the effective conductivity contrast ratio σ_e/σ_h as a function of $\ln(\sigma_k/\sigma_h)$ at low volume fraction ($f = 0.042$).

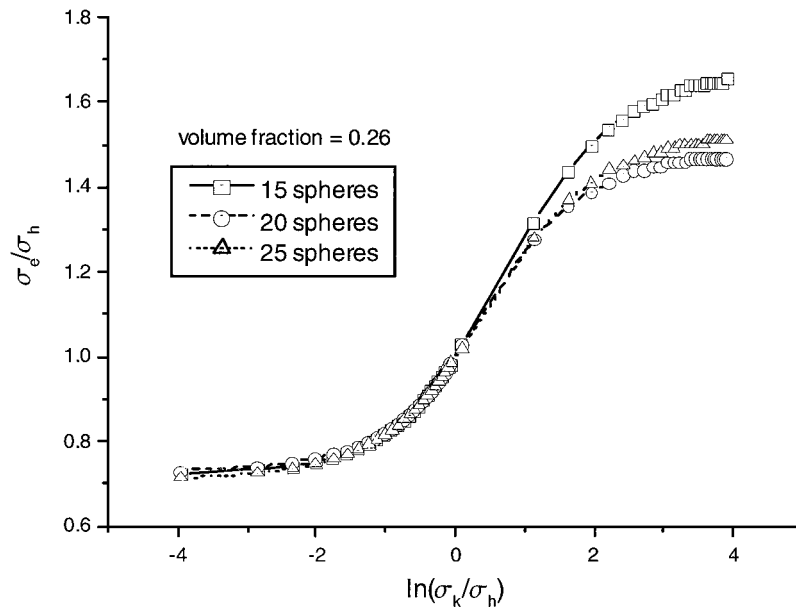


Figure 2 For different number of spheres, the effective conductivity contrast ratio σ_e/σ_h as a function of $\ln(\sigma_k/\sigma_h)$ at high volume fraction ($f = 0.26$).

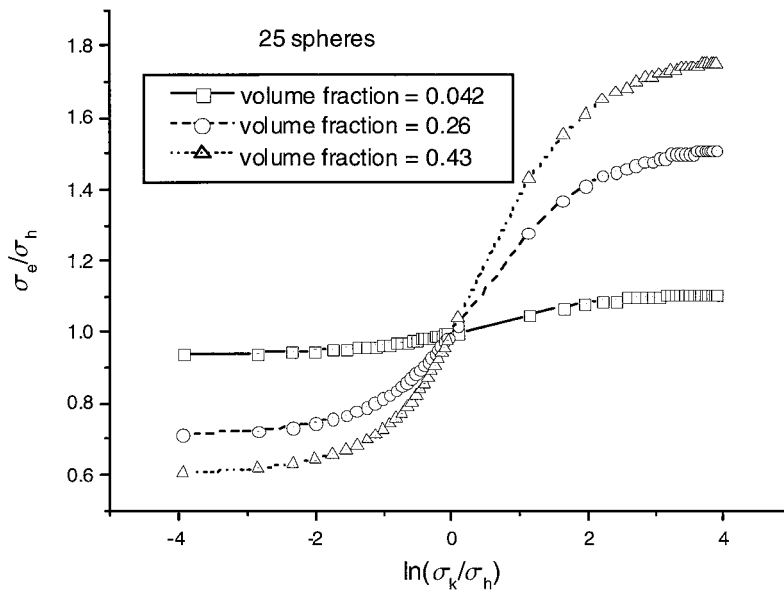


Figure 3 For different volume fractions, the effective conductivity contrast ratio σ_e/σ_h as a function of $\ln(\sigma_k/\sigma_h)$. The number of the spheres is fixed.

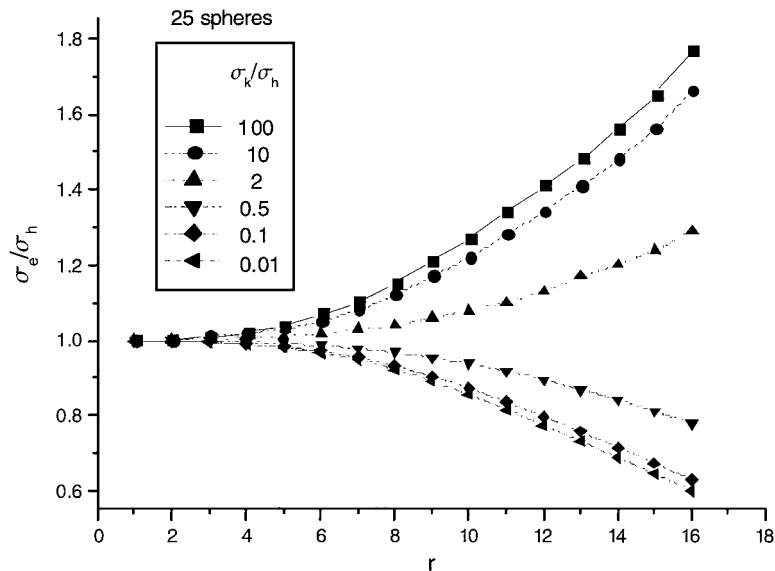


Figure 4 For different conductivity contrast ratio σ_k/σ_h , the effective conductivity contrast ratio σ_e/σ_h as a function of radius r for fixed number of spheres.

conductivity increases with the total volume fraction. As expected, the effective conductivity is less than the host conductivity σ_h if the parameter σ_k/σ_h is less than unity. In Fig. 4, curves of σ_e/σ_h against the radius r of the spheres are plotted for different contrast ratio σ_k/σ_h . Obviously, it shows that σ_k/σ_h is also an important factor.

Case (C): The spatial distribution of the spheres in the unit cell are varied, with fixed volume fraction and fixed number of the spheres. At high concentrations of the dispersed spheres, three samples, having twenty spheres with volume fraction 0.26 are considered. The plots of the effective conductivity for these spheres at different spatial distributions are shown in Fig. 5. One can see that the positions of these spheres affect the effective conductivity. From the formulae of

the lattice sums and the effective conductivity, one can see the reason. This is because the lattice sums depend on the positions of the spheres located in the unit cell, and hence give variations in the calculated values of the coefficient $C_{1,0}^{1,k}$. Thus the effective conductivity will have different values for different spatial distributions of the inclusion spheres. Theoretically, for different spatial distributions of the spheres, the intensities of the particle polarizations are different, and the values depend on the arrangement of spheres in the cubic cell. In particular, for larger contrast ratio σ_k/σ_h , the positions of the spheres produce larger effects on the effective conductivity. However, Fig. 6 shows that at low volume fraction (0.042), the dependence on spatial distribution is much less important.

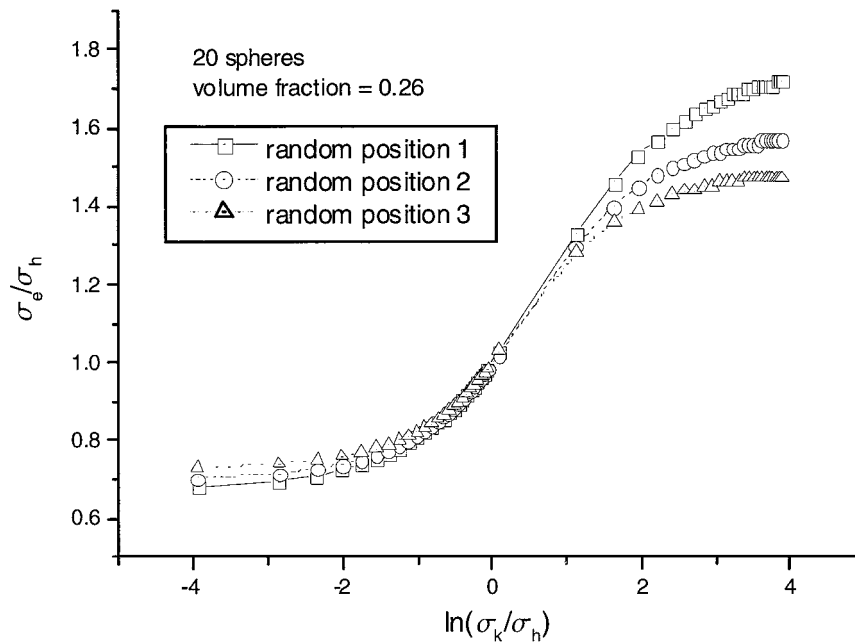


Figure 5 For different spatial distributions of twenty spheres, the effective conductivity contrast ratio σ_e/σ_h as a function of $\ln(\sigma_k/\sigma_h)$ at high volume fraction ($f = 0.26$).

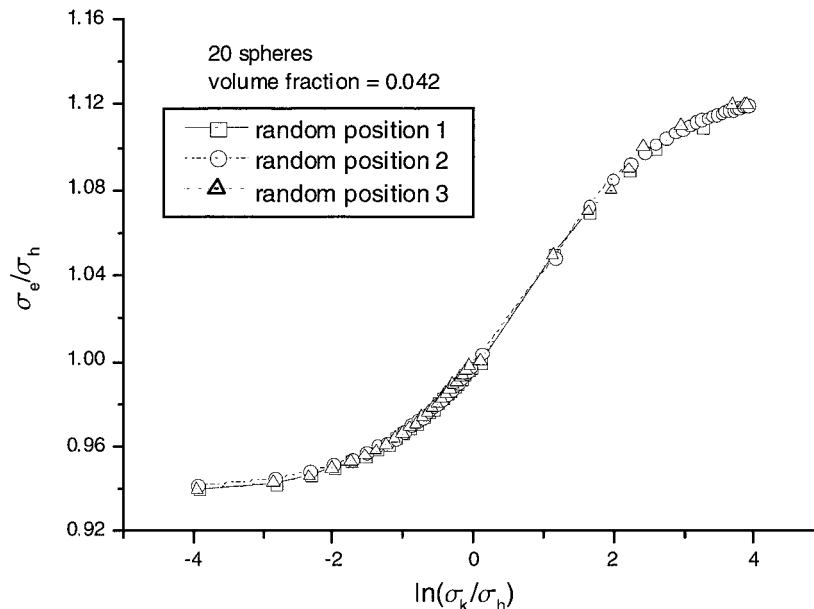


Figure 6 For different spatial distributions of twenty spheres, the effective conductivity contrast ratio σ_e/σ_h as a function of $\ln(\sigma_k/\sigma_h)$ at low volume fraction ($f = 0.042$).

4. Conclusions

Using Rayleigh's method, we have derived formula for the effective conductivity of composites with spheres randomly embedded in a linear continuum. Various factors of the poly-dispersed spheres are investigated. Our results show that the effective conductivity of the composites not only relates to the volume fraction and the conductivity of the inclusions, but also on their spatial distribution. In particular, at large conductivity contrast ratio σ_k/σ_h and high volume fractions, the derivation of the effective conductivity is larger for different spatial distribution. This may result from the intensities of the surface polarization of the inclusions. Therefore, both volume fraction and spatial distribution should be considered when we design and apply the composite material in engineering. Based on our formulae, we can also investigate the statistical properties of effective conductivities if enough samples are employed. For example, we can estimate the mean value, the mean-square-deviation and probability distribution of the effective conductivity for different spatial distribution of inclusions in the host medium.

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