

A MATERIAL MULTIPOLE THEORY OF ELASTIC DIELECTRIC COMPOSITES†

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Abstract—This article is concerned with the study of the overall mechanical and electrical properties of elastic dielectric composites by using the concept of material multipoles. In particular, by developing a statistical continuum material multipole theory, the effects of the microstructure of the inhomogeneities on the overall properties of the composites can be derived. In this theory, inhomogeneities are modelled as point material-induced multipoles. The macroscopic fields obtained from the ensemble average of the microscopic fields in the composite with statistically-distributed inhomogeneities in a uniform matrix are described by statistical continuum material multipoles in the matrix. This theory, in comparison with classical effective medium theory, has the possibility of attacking rather complicated problems such as, for instance, electromagnetoelastic composites. It is shown that the statistical anisotropy and shape effects of microscopic ellipsoidal particles and their orientations on the overall effective properties of dielectric composites may be obtained in an explicit form. Also, the macroscopic constitutive relations of elastic dielectric composites and their macroscopic material parameters accounting for electroelastic interaction may be derived with the use of this statistical continuum multipole theory.

1. INTRODUCTION

The study of composite materials has received considerable attention in recent years. To provide some theoretically-predictable models for composite materials in various practical applications, certain assumptions to simplify the problems are usually introduced due to the complexity of the composite materials. For instance, deformation effects are neglected in studying overall electromagnetic properties of composites, or electromagnetic effects are ignored in studying effective mechanical properties of composites (Kröner, 1959; Hashin and Shtrikman, 1962; Beran, 1968; Miller, 1969; Jeffrey, 1973; McCoy and Beran, 1976; Van Beek, 1967; Hale, 1976; Christensen, 1979; Willis, 1983; Zhou and Hsieh, 1986). The various approaches and models based on these assumptions may work well for many applications but they may fail for others in which both electromagnetic and deformation fields and their coupling effects are important. Such examples, even for homogeneous materials, are numerous, for instance, piezoelectric ceramics and superconducting materials, etc. (Nelson, 1979; Maugin, 1984; Moon, 1984; Zhou *et al.*, 1986; Zhou and Hsieh, 1988). Difficulties are, however, faced in attempts to apply the classical effective medium theory to study the overall response of electromagnetic deformable composites. For instance, in the case of using the well-known self-consistent scheme in the classical effective medium theory, the overall effective constitutive relation for the composite has to be preassumed as a known form with unknown effective material constants to be determined. Such a procedure will, however, fail if one does not even know the form of the overall macroscopic constitutive relation of the composite, which, in general, is the case with electromagnetic deformable composites. The overall macroscopic constitutive relations and overall material parameters of such composites have to be derived from both the knowledge of specific material microstructures and the microscopic material properties of the composites.

In this article, the electroelastic coupling phenomena and their influences on the overall behaviors of elastic dielectric composites will, therefore, be studied by introducing a statistical continuum material multipole theory (Zhou, 1987). In this theory, a composite is modelled as a medium composed of a large number of inhomogeneous particles distributed in a uniform matrix, and the particles are assumed to be firmly bonded with the matrix at their interfaces. The microstructure of the composite is supposed to be of a

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random nature so that a statistical approach is adopted. The inhomogeneous particles are modelled as point-material-induced multipoles and the macroscopic fields from the ensemble average of the microscopic fields in the composite are described by statistical continuum material multipoles in a uniform matrix.

To study quantitatively the overall properties of elastic dielectric composite materials, a basic solution for an ellipsoidal elastic inhomogeneity with electric polarization in an infinite elastic dielectric medium is first derived in Section 2, which shows that classical Eshelby's elastic solution (Eshelby, 1961) is modified by the presence of electric-elastic interaction. The overall behaviors of the elastic dielectric composites are then studied by the statistical continuum material multipole theory. It will be shown that the overall macroscopic constitutive relations of the elastic dielectric composites as well as their overall macroscopic material parameters accounting for the electroelastic interaction effect can be derived. Some quantitative calculations on problems with statistical anisotropy, shape effect and electric-elastic interaction are given for dilute composites.

2. ANALYSIS OF AN ELLIPSOIDAL INHOMOGENEITY IN ELASTIC DIELECTRICS

It is well known that classical Eshelby's solutions on elastic inhomogeneous inclusions are of fundamental importance in studying the overall properties of various elastic composite materials [see, for instance, Eshelby (1961) and Mura (1982)]. In this section, we shall deal with the problem of an electrically-polarized ellipsoidal inhomogeneous elastic particle embedded in an infinite elastic dielectric medium. In the case of mechanical equilibrium, one has the following balance equation:

$$\nabla \cdot \mathbf{t} + \mathbf{f} = 0 \quad (1)$$

where \mathbf{t} denotes the Cauchy stress tensor and \mathbf{f} the body force.

If the medium is purely elastic and can be characterized by the following linear Hooke's law

$$t_{ij}(\mathbf{x}) = C_{ijkl}u_{k,l}(\mathbf{x}) \quad (2)$$

with C_{ijkl} being the elastic moduli and \mathbf{u} the mechanical displacement vector field, one may find with the aid of the method of Green's function that the mechanical displacement field \mathbf{u} due to the body force \mathbf{f} may be obtained as

$$u_i = u_i^0 + \int_{V'} f_j(\mathbf{x}') G_{ij}(\mathbf{x} - \mathbf{x}') d\mathbf{x}' \quad (3)$$

where G_{ij} is the elastic Green's function, which, for an isotropic medium, can be written as

$$G_{ij} = \frac{1}{16\pi\mu(1-\nu)|\mathbf{x} - \mathbf{x}'|} \left\{ (3-4\nu)\delta_{ij} + \frac{(x_i - x'_i)(x_j - x'_j)}{|\mathbf{x} - \mathbf{x}'|^2} \right\} \quad (4)$$

where μ and ν are respectively the elastic shear modulus and Poisson's ratio of the medium. If the material medium is not elastic and it is characterized by the following relation:

$$t_{ij}(\mathbf{x}) = C_{ijkl}u_{k,l}(\mathbf{x}) + t_{ij}^{\text{inc}}(\mathbf{x}) \quad (5)$$

where t_{ij}^{inc} denotes the inelastic part of the stress, we may find formally the displacement field \mathbf{u} which can be expressed with the use of the elastic Green's function as

$$u_i = u_i^0 + \int_{V_\varepsilon} f_j(\mathbf{x}') G_{ij}(\mathbf{x} - \mathbf{x}') d\mathbf{x}' + \int_{V_\varepsilon} t_{jk}^{ine}(\mathbf{x}') G_{i,jk}(\mathbf{x} - \mathbf{x}') d\mathbf{x}'. \quad (6)$$

Using above results, we may now study the problem of elastic deformation at a point \mathbf{x} due to the electric forces acting on an electric dipole centered at \mathbf{x}' in an infinite elastic homogeneous medium under the exertion of an electric field \mathbf{E} . By using the method of Green's functions, the elastic displacement field may be found as

$$u_i(\mathbf{x}) = G_{ij} \left(\mathbf{x} - \mathbf{x}' - \frac{d}{2} \mathbf{n}' \right) f_j^+ + G_{ij} \left(\mathbf{x} - \mathbf{x}' + \frac{d}{2} \mathbf{n}' \right) f_j^- \quad (7)$$

in which \mathbf{n}' is a unit vector of the direction of the electric dipole and d is the distance between two point electric charges which constitute the electric dipole. \mathbf{f}^+ and \mathbf{f}^- are the point electric body forces given by

$$\mathbf{f}^+ = q\mathbf{E} \left(\mathbf{x}' + \frac{d}{2} \mathbf{n}' \right) \quad \text{and} \quad \mathbf{f}^- = -q\mathbf{E} \left(\mathbf{x}' - \frac{d}{2} \mathbf{n}' \right) \quad (8)$$

where q denotes the point body charge. For an ideal point electric dipole \mathbf{p} , resulting from the limiting process of letting the distance between the two charge decrease indefinitely and at the same time letting the amount of charge increase in such a way that the product $\mathbf{p} = q d\mathbf{n}'$ remains a constant vector, the displacement, eqn (7), becomes

$$u_i(\mathbf{x}) = G_{ij}(\mathbf{x} - \mathbf{x}') p_j E_{i,j}(\mathbf{x}') - G_{i,jk}(\mathbf{x} - \mathbf{x}') p_k E_i(\mathbf{x}') \quad (9)$$

where the electric field has been assumed to be smooth enough such that all the high-order terms $O(d^2)$ vanish during the limiting process.

Equation (9) shows that the elastic field caused by such an electric dipole may be modelled as that generated by an induced point elastic monopole at \mathbf{x}' defined by

$$P_{jk} = p_k E_j(\mathbf{x}') \quad (10)$$

and an induced point body force at \mathbf{x}' defined by

$$f_j = p_k E_{j,k}(\mathbf{x}') \quad (11)$$

which vanishes for a uniform electric field.

Considering now an inhomogeneous particle with a continuum dielectric polarization \mathbf{P} embedded in an elastic dielectric matrix medium, which is subject to certain external electric and mechanical loads (denoted by \mathbf{E}^0 and \mathbf{u}^0), one may find that

$$E_i = E_i^0 + \int_{V_i} P_k^e(\mathbf{x}') G_{,ki}^e(\mathbf{x} - \mathbf{x}') d\mathbf{x}' \quad (12)$$

and

$$u_i = u_i^0 + \int_{V_i} f_j(\mathbf{x}') G_{ij}(\mathbf{x} - \mathbf{x}') d\mathbf{x}' - \int_{V_i} P_{jk}(\mathbf{x}') G_{i,jk}(\mathbf{x} - \mathbf{x}') d\mathbf{x}' \quad (13)$$

where V_i is the volume of the particle, and G^e and G_{ij} are respectively the electric and elastic Green's functions for the infinite matrix medium. The superscript e is used to identify quantities which are related to electricity.

Here, one has assumed that the size of the particle is large enough compared with molecular sizes so that it can be treated as a continuum, and that the dielectric polarization of the particle is of the form

$$\mathbf{P} = \varepsilon_0 \chi^* \mathbf{E} + \mathbf{P}^0 \quad (14)$$

where \mathbf{P}^0 denotes the spontaneous polarization, and χ^* the dielectric susceptibility of the particle. It is also assumed that the dielectric polarization of the matrix material is so small that the electric body force acting on the matrix may be neglected.

It can be seen from eqns (12) and (13) that the effect of such an inhomogeneous particle embedded in an elastic dielectric matrix medium may be modelled by a distribution of continuum electric dipole defined by

$$P_k^c = P_k^0 + \Delta\varepsilon_{kj} E_j \quad (15)$$

and the body force by

$$f_j = P_k E_{j,k} \quad (16)$$

and the continuum elastic monopole by

$$P_{jk} = P_k E_j - \Delta C_{jkmn} u_{m,n} \quad (17)$$

where $\Delta\varepsilon$ and ΔC denote the perturbation values of the material properties between the particle and the matrix, which, for the isotropic materials, read

$$\Delta\varepsilon_{ij} = (\varepsilon^* - \varepsilon) \delta_{ij} \quad (18)$$

where $\varepsilon [= \varepsilon_0(1 + \chi)]$ and $\varepsilon^* [= \varepsilon_0(1 + \chi^*)]$ are respectively the permittivity of the matrix and of the particle, and

$$\Delta C_{ijkl} = (\lambda^* - \lambda) \delta_{ij} \delta_{kl} + (\mu^* - \mu) (\delta_{ik} \delta_{jl} + \delta_{il} \delta_{jk}) \quad (19)$$

where λ , μ and λ^* , μ^* are respectively Lamé's elastic constants of the matrix and of the particle.

In order to determine these induced continuum electric and elastic multipoles, one has to solve eqns (12) and (13), which are in general coupled since the elastic deformation depends on the electric field which is dependent of the orientation of the particle which is, in turn, affected by the elastic deformation. As a first approximation, we may ignore the influence of the small change of the orientation of the particle, due to elastic deformation, on the electric field. Thus, for uniform external electric and elastic strain fields, we find that the electrically-induced body force vanishes and that the electric and elastic fields within the particle are also uniform, and they become respectively

$$E_i = \left(\delta_{ik} + \frac{\Delta\varepsilon}{\varepsilon} L_{ik}^c \right)^{-1} \left(E_k^0 - \frac{1}{\varepsilon} L_{km}^c P_m^0 \right) \quad (20)$$

and

$$e_{ij} = (\delta_{im} \delta_{jn} + L_{ijkl} \Delta C_{jkmn})^{-1} (e_{mn}^0 + L_{mnik} E_l P_k) \quad (21)$$

where e_{ij} denotes the infinitesimal elastic strain tensor, and \mathbf{L}^c and \mathbf{L} denote respectively the electric and elastic depolarization tensors defined by

$$L_{ik}^e = \frac{1}{4\pi} \int_{S_j} \frac{x'_i n'_k}{|x'|^3} dS' \quad (22)$$

and

$$L_{ijkl} = -\frac{1}{2} \int_{S_j} (G_{il,j} + G_{jl,i}) n'_k dS' \quad (23)$$

which are constant tensors in the case of the particle being of ellipsoidal shape. Here, S_j is the surface of the particle, and \mathbf{n}' the outer normal vector of the surface S_j . For an isotropic spherical particle, the electric and elastic depolarization tensors may be found explicitly as

$$L_{ik}^e = \frac{1}{3} \delta_{ik} \quad (24)$$

and

$$L_{ijkl} = \frac{1}{30\mu} \left[\frac{9\kappa + 18\mu}{3\kappa + 4\mu} (\delta_{kj} \delta_{il} + \delta_{ik} \delta_{jl}) - \frac{6\kappa + 2\mu}{3\kappa + 4\mu} \delta_{ij} \delta_{kl} \right] \quad (25)$$

and the electric and elastic strain fields in the spherical particle may be derived explicitly as

$$E_i = \frac{3\epsilon}{3\epsilon + \Delta\epsilon} \left(E_i^0 - \frac{1}{3\epsilon} P_i^0 \right) \quad (26)$$

and

$$e_{kk} = \frac{3\kappa + 4\mu}{3\kappa + 4\mu + 3\Delta\lambda + 2\Delta\mu} e_{kk}^0 + \frac{E_k P_k}{3\kappa + 4\mu + 3\Delta\lambda + 2\Delta\mu} \quad (27)$$

where sum is made over the suffix k , and for $i \neq j$

$$e_{ij} = \frac{5\mu(3\kappa + 4\mu)}{5\mu(3\kappa + 4\mu) + \Delta\mu(6\kappa + 12\mu)} e_{ij}^0 + \frac{(3\kappa + 6\mu)(E_i P_j + E_j P_i)}{10\mu(3\kappa + 4\mu) + \Delta\mu(12\kappa + 24\mu)} \quad (28)$$

in which $\kappa = \lambda + 2\mu/3$ is the elastic bulk modulus of the matrix, \mathbf{P} the dielectric polarization in the particle given by eqn (14), and \mathbf{E} the electric field in the particle given by eqn (26). It is shown that for an ellipsoidal elastic inhomogeneous particle with electric polarization embedded in an infinite elastic dielectric medium, Eshelby's classical solution is modified by the presence of an additional term on the right-hand side of eqn (21) due to the electroelastic interaction.

The stress fields within or outside the particle may be found by noting eqns (5), (6) and (13) as

$$t_{ij}(\mathbf{x}) = C_{ijkl} u_{k,l}(\mathbf{x}) + (\Delta C_{ijkl} u_{k,l}(\mathbf{x}) - P_j(\mathbf{x}) E_i(\mathbf{x})) \gamma(\mathbf{x}) \quad (29)$$

where the indicative function $\gamma(\mathbf{x})$ is defined by

$$\gamma(\mathbf{x}) = \begin{cases} 1, & \mathbf{x} \text{ in } V_l \\ 0, & \text{otherwise.} \end{cases} \quad (30)$$

The antisymmetric part of the elastic deformation field in the particle may be studied by introducing the antisymmetric deformation tensor defined by $2\omega = \nabla \mathbf{u} - (\nabla \mathbf{u})^T$. After some calculations, one can obtain

$$\omega_{i,j} = \omega_{i,j} + L''_{ijkl} (\Delta C_{ikmn} e_{mn} - P_k E_l) \quad (31)$$

where the tensor L'' is defined by

$$L''_{ijkl} = \frac{1}{2} \int_{S_l} (G_{il,j'} - G_{jl,i'}) n'_k dS' \quad (32)$$

which, in the case of a spherical particle, reads

$$L''_{ijkl} = \frac{1}{6\mu} (\delta_{ik}\delta_{lj} - \delta_{il}\delta_{kj}) \quad \text{in } V_l. \quad (33)$$

For a spherical particle, one can deduce explicitly from eqn (31) that

$$\nabla \times \mathbf{u} = \nabla \times \mathbf{u}^0 + \frac{\varepsilon}{\mu(3\varepsilon + \Delta\varepsilon)} \mathbf{P}^0 \times \mathbf{E}^0 \quad (34)$$

which gives the proportional relation between the rotation vector and the electric moment caused by the spontaneous polarization of the particle. It is shown that the rotation vector of the spherical particle is independent of the elastic properties of the particle, and it is also a constant vector within the particle under uniform external loads. It should be noticed that formula (34) may only be used to predict small rotations of the particle since the solution is derived from a small (infinitesimal) deformation theory. For large rotations of the particle, one needs a finite deformation theory.

It is shown that a basic solution of an ellipsoidal elastic inhomogeneity with electric polarization embedded in an infinite elastic dielectric medium can be obtained with the use of the multipole approach. It is found that under the exertion of uniform electric and elastic loadings, the elastic strain and electric field in the ellipsoidal elastic inhomogeneity with electric polarization are both uniform and can be determined in general by eqns (20) and (21). The inhomogeneous particle can also be rotated uniformly due to the electric moment acting on the inhomogeneity with spontaneous polarization, as shown in eqn (34). It is shown that Eshelby's classical results for an ellipsoidal elastic inhomogeneity embedded in an infinite elastic medium are modified in the case of elastic dielectrics by the presence of the electric-elastic interaction.

3. STATISTICAL CONTINUUM MATERIAL MULTIPOLES

It has been shown that an inhomogeneity embedded in a matrix medium may be modelled as continuum material multipoles in a uniform matrix medium. Composite materials may, however, be composed of many such inhomogeneities. In addition, though some of the composites may have a regular structure such as laminated media in which the material properties can be well defined periodically, there is, however, a large class of composite materials in which the microstructure is so complex that it is hardly feasible to define its material properties at each point. It is more likely, instead, that only a certain amount of statistical information on the microstructure of the composites is available. Such composites are, for instance, dispersion-strengthened, particle-reinforced, and chopped-fiber-reinforced materials. To study this class of composite materials with large numbers of randomly-distributed inhomogeneous particles, a statistical approach will, therefore, be adopted and the concepts of statistical continuum material multipoles will be introduced in this section.

We shall start with the generalization of the concept of statistical continuum elastic multipoles (Zhou and Hsieh, 1986) to the case where some random parameters accounting for the microstructure of the body-force array are required. Then, concepts of statistical continuum electric and magnetic multipoles may be considered similarly.

In analogy with the Gibbs ensemble used in classical statistical mechanics, we may imagine a great number of independent samples identical in the sense of having the same macroscopic elastic property, the same geometric shape and subject to the same number of point body-force arrays, but varying in an undetermined manner in the distributions and orientations of these point body-forces from sample to sample (Fig. 1).

The total elastic displacement fields in a given sample due to these M body-force arrays can be written in general as

$$u_i(\mathbf{x}; \mathbf{x}^1, \dots, \mathbf{x}^M, \omega^1, \dots, \omega^M) = \sum_{\alpha=1}^M \sum_{k=0}^{\infty} \frac{(-1)^k}{k!} P_{j_1, \dots, j_k}^{\alpha} G_{ij, s_1, \dots, s_k}(\mathbf{x}, \mathbf{x}^{\alpha}) \quad (35)$$

where \mathbf{x}^{α} is a random variable (varying with different samples) which indicates the center position of the α th point body-force array, and ω^{α} is a random parameter (probably several parameters) characterizing the microstructure of the body-force array. G_{ij} is the elastic Green's function of the sample, which is supposed to be an infinite elastic medium. $P_{j_1, \dots, j_k}^{\alpha}$ is the elastic multipole moment of order k for the α th point body-force array. For force arrays in self-equilibrium, we have $P_j^{\alpha} = 0$. If the point body forces are permanent in the sense that they are independent of each other, we may write

$$P_{j_1, \dots, j_k}^{\alpha} = P_{j_1, \dots, j_k}^{\alpha}(\mathbf{x}^{\alpha}, \omega^{\alpha}) = \sum_{\beta=1}^{N(\alpha)} f_j^{\beta}(\mathbf{x}^{\alpha}, \omega^{\alpha}) d_{r_1}^{\beta}(\mathbf{x}^{\alpha}, \omega^{\alpha}) \dots d_{r_k}^{\beta}(\mathbf{x}^{\alpha}, \omega^{\alpha}) \quad (36)$$

where $N(\alpha)$ denotes the number of the point body-forces in the α th point body-force array and $d^{\beta}(\mathbf{x})$ denotes the β th position vector from \mathbf{x}^{α} to its corresponding position of the point body-force $f^{\beta}(\mathbf{x})$. If the point body-forces are of the induced type so that they are dependent on each other due to interaction among themselves, we shall, in general, have

$$P_{j_1, \dots, j_k}^{\alpha} = \sum_{\beta=1}^{N(\alpha)} f_j^{\beta}(\mathbf{x}^{\alpha}; \mathbf{x}^1, \dots, \mathbf{x}^M, \omega^1, \dots, \omega^M) d_{r_1}^{\beta} \dots d_{r_k}^{\beta} \quad (37)$$

which means that the elastic multipoles modelling the α th point body-force array are dependent of other body-force arrays, their positions and distributions etc.

With the aid of the joint-probability density function $f(\Omega^1, \dots, \Omega^M)$ with $\Omega^{\alpha} = (\mathbf{x}^{\alpha}, \omega^{\alpha})$, the ensemble average of elastic fields due to the randomly-distributed point body-force arrays may be expressed as

$$\langle u_i \rangle(\mathbf{x}) = \sum_{k=0}^{\infty} \frac{(-1)^k}{k!} \int_V \bar{P}_{j_1, \dots, j_k}(\mathbf{x}') G_{ij, s_1, \dots, s_k}(\mathbf{x}, \mathbf{x}') d\mathbf{x}' \quad (38)$$

where we have introduced the statistical continuum elastic multipole of order k , defined by

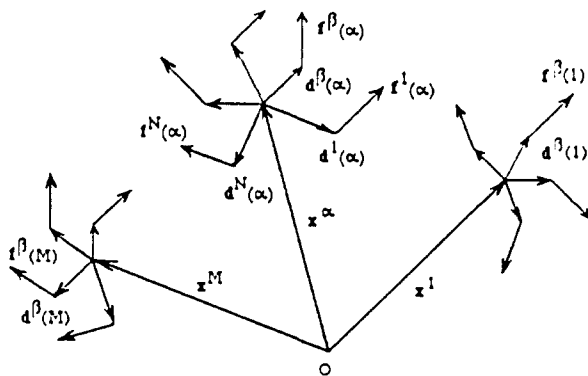


Fig. 1. A distribution of point body-force arrays.

$$\bar{P}_{j_1 \dots j_k}(\mathbf{x}') = \sum_{\alpha=1}^M \int_{\Gamma} F_{j_1 \dots j_k}^{\alpha}(\Omega') d\Gamma' \tag{39}$$

in which

$$F_{j_1 \dots j_k}^{\alpha} = \underbrace{\int_{\Omega} \dots \int_{\Omega}}_{M-1} f(\Omega^1, \dots, \Omega^M) P_{j_1 \dots j_k}^{\alpha} d\Omega^1 \dots d\Omega^{\alpha-1} d\Omega^{\alpha+1} \dots d\Omega^M \tag{40}$$

where Ω is a defined space, $\Omega = V \times \Gamma$, in which V is the volume of the material body and Γ is a parameter space, $\omega^{\alpha} \in \Gamma$.

In the case of permanent point body-force arrays, eqn (39) may be reduced to

$$\bar{P}_{j_1 \dots j_k}(\mathbf{x}') = \int_{\Gamma} \rho(\Omega') P_{j_1 \dots j_k}(\Omega') d\Gamma' \tag{41}$$

where

$$\rho(\Omega') = \sum_{\alpha=1}^M W_{\alpha}(\Omega') \tag{42}$$

with

$$W_{\alpha} = \underbrace{\int_{\Omega} \dots \int_{\Omega}}_{M-1} f(\Omega^1, \dots, \Omega^M) d\Omega^1 \dots d\Omega^{\alpha-1} d\Omega^{\alpha+1} \dots d\Omega^M. \tag{43}$$

The physical meaning of the function $\int_{\Gamma} \rho(\Omega') d\Gamma'$ may be explained as the number of the elastic multipoles per unit volume since one has

$$\int_{\Omega} \rho(\Omega') d\Omega' = \int_V \left(\int_{\Gamma} \rho(\Omega') d\Gamma' \right) d\mathbf{x}' = M. \tag{44}$$

It is shown that the ensemble-average elastic fields due to a statistically-discrete distribution of point body-force arrays may be modelled as the elastic fields due to a distribution of statistical continuum elastic multipoles as defined in eqn (39) or eqn (41).

Similarly, for M randomly-distributed point body-charge arrays in a dielectric body, we find that the ensemble average of the electric field due to the randomly-distributed point body-charge arrays may be expressed as

$$\langle E_i \rangle(\mathbf{x}) = \sum_{k=0}^{\infty} \frac{(-1)^{k+1}}{k!} \int_V \bar{P}_{j_1 \dots j_k}^c(\mathbf{x}') G_{i, j_1 \dots j_k}^c(\mathbf{x}, \mathbf{x}') d\mathbf{x}' \tag{45}$$

where we have introduced the statistical continuum electric multipole of order k , defined by

$$\bar{P}_{j_1 \dots j_k}^c(\mathbf{x}') = \sum_{\alpha=1}^M \int_{\Gamma} F_{j_1 \dots j_k}^{\alpha c}(\Omega') d\Gamma' \tag{46}$$

in which

$$F_{s_1 \dots s_k}^{cz} = \underbrace{\int_{\Omega} \dots \int_{\Omega}}_{M-1} f(\Omega^1, \dots, \Omega^M) P_{s_1 \dots s_k}^{cz} d\Omega^1 \dots d\Omega^{z-1} d\Omega^{z+1} \dots d\Omega^M \quad (47)$$

with $P_{s_1 \dots s_k}^{cz}$ being the electric multipole moment of order k for the α th point body-charge array, defined by

$$P_{s_1 \dots s_k}^{cz} = \sum_{\beta=1}^{N(\alpha)} q^{\beta}(\mathbf{x}^z; \mathbf{x}^1, \dots, \mathbf{x}^M, \omega^1, \dots, \omega^M) d_{s_1}^{\beta} \dots d_{s_k}^{\beta} \quad (48)$$

If the α th point body-charge array is self-electrically neutral, we have $P^{cz} = 0$. In particular, if the point body-charge arrays are of permanent type, i.e.

$$P_{s_1 \dots s_k}^{cz} = P_{s_1 \dots s_k}^c(\mathbf{x}^z, \omega^z) = \sum_{\beta=1}^{N(\alpha)} q^{\beta}(\mathbf{x}^z, \omega^z) d_{s_1}^{\beta}(\mathbf{x}^z, \omega^z) \dots d_{s_k}^{\beta}(\mathbf{x}^z, \omega^z) \quad (49)$$

then the statistical continuum electric multipole of order k may be expressed simply by

$$\bar{P}_{s_1 \dots s_k}^c(\mathbf{x}') = \int_{\Gamma} \rho(\Omega') P_{s_1 \dots s_k}^c(\Omega') d\Gamma' \quad (50)$$

where $\rho(\Omega')$ is given by eqn (42).

4. STATISTICAL CONTINUUM MULTIPOLE MODELLING OF MATERIAL COMPOSITES

To illustrate the use of statistical continuum material multipoles, we shall consider, in this section, the statistical continuum electric and elastic multipole modelling of elastic dielectric composite materials with a large number of M statistically-distributed inhomogeneous particles with electrical polarization. All particles will be assumed to have the same shape, the same size and the same strength of spontaneous electric polarization but they can have different orientations. Other types of material composites may be studied similarly. To formulate the problem mathematically, one has several possibilities with regard to the composite specimen shape and boundary conditions. To avoid using the concept of an "infinite" specimen as well as the convergence difficulty (Jeffrey, 1973), we shall consider a finite spherical specimen, which is assumed to be perfectly embedded in an infinite homogeneous elastic dielectric medium with the same material properties as the matrix of the composite (see Fig. 2).

The problem of interest to us is to find the overall properties of such an elastic dielectric composite. The geometrical arrangement of the inhomogeneities in a given sample of the composite is specified by the indicative functions

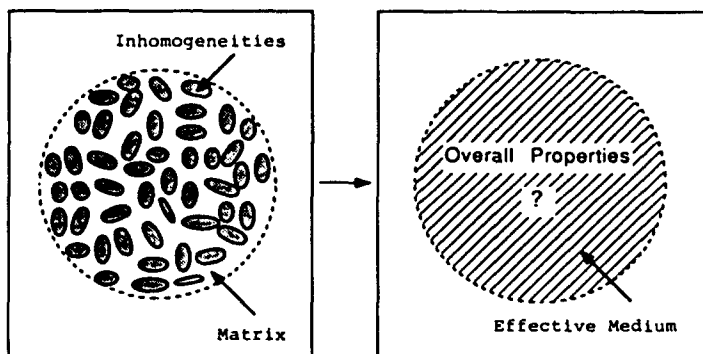


Fig. 2. A material composite of spherical shape.

$$\gamma^x(\mathbf{x}) = \begin{cases} 1, & \mathbf{x} \text{ in } V_x \\ 0, & \text{otherwise} \end{cases} \tag{51}$$

where V_x is the volume of the x th inhomogeneity ($x = 1, 2, \dots, M$). The dimension of the inhomogeneities will be always assumed to be much smaller than the dimension of the composite specimen, but much larger than the size of the molecule (or atom) such that the inhomogeneous particle may be treated as a continuum with permittivity ϵ^* and elastic moduli C^* . With the aid of results given in Section 2, the microscopic electric and elastic fields in the composite under the action of external fields (\mathbf{E}^0 and \mathbf{u}^0) may be written as

$$E_i = E_i^0 + \sum_{x=1}^M \int_{V_x} (P_k^{0(x)} + \Delta\epsilon_{ki} E_j) G_{ki}^c(\mathbf{x} - \mathbf{x}') d\mathbf{x}' \tag{52}$$

and

$$u_i = u_i^0 + \sum_{x=1}^M \left[\int_{V_x} f_j^{(x)} G_{ij}(\mathbf{x} - \mathbf{x}') d\mathbf{x}' - \int_{V_x} P_{jk}^{(x)} G_{ij,k}(\mathbf{x} - \mathbf{x}') d\mathbf{x}' \right]. \tag{53}$$

By noting the geometrical arrangements and the orientations of the inhomogeneities, eqns (52) and (53) can be further expressed as

$$E_i(\mathbf{x}; \Omega^1, \dots, \Omega^M) = E_i^0 + \sum_{x=1}^M \int_{V_x} (P_k^{0(x)} + \Delta\epsilon_{ki} E_j^{(x)}) G_{ki}^c(\mathbf{x} - \mathbf{x}^x - \mathbf{y}^x) d\xi \tag{54}$$

and

$$u_i(\mathbf{x}; \Omega^1, \dots, \Omega^M) = u_i^0 + \sum_{x=1}^M \int_{V_x} f_j^{(x)} G_{ij}(\mathbf{x} - \mathbf{x}^x - \mathbf{y}^x) d\xi - \sum_{x=1}^M \int_{V_x} P_{jk}^{(x)} G_{ij,k}(\mathbf{x} - \mathbf{x}^x - \mathbf{y}^x) d\xi \tag{55}$$

in which $\mathbf{P}^{0(x)}$, $\mathbf{E}^{(x)}$, $\mathbf{f}^{(x)}$ and $\mathbf{P}^{(x)}$ are respectively the permanent electric polarization, the electric field, the induced electric body force and the induced elastic dipole density defined in the x th inhomogeneity, which are, in general, dependent on the geometrical arrangements of all other inhomogeneities due to interaction among themselves.

Here, we introduce the notation $\Omega^x = (\mathbf{x}^x, \theta^x, \psi^x, \omega^x)$ and

$$\mathbf{y}^x = \mathbf{Q}^x(\theta^x, \psi^x, \omega^x) \cdot \boldsymbol{\xi} \tag{56}$$

where \mathbf{x}^x is the position vector of the gravitational center and \mathbf{Q}^x the orientation tensor of the x th inhomogeneous ellipsoid, which is an orthogonal tensor, defined by

$$(\mathbf{Q}_i^x) = \begin{pmatrix} \cos \psi^x \cos \omega^x - \cos \theta^x \sin \psi^x \sin \omega^x & -\cos \psi^x \sin \omega^x - \cos \theta^x \sin \psi^x \cos \omega^x & \sin \psi^x \sin \theta^x \\ \cos \omega^x \sin \psi^x + \cos \theta^x \cos \psi^x \sin \omega^x & -\sin \psi^x \sin \omega^x + \cos \theta^x \cos \psi^x \cos \omega^x & -\cos \psi^x \sin \theta^x \\ \sin \theta^x \sin \omega^x & \cos \omega^x \sin \theta^x & \cos \theta^x \end{pmatrix} \tag{57}$$

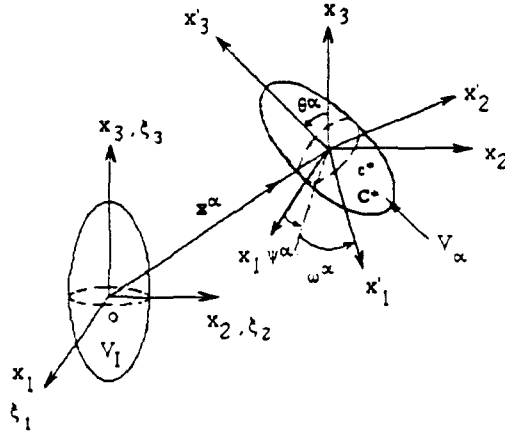


Fig. 3. Phase geometry of the composite.

where θ^α , ψ^α and ω^α are the Euler angles of the α th inhomogeneity (see Fig. 3).

In particular, for inhomogeneities with an ellipsoidal shape of revolution, we may let $\omega^\alpha = 0$ and the orientation tensor is reduced to

$$(Q_{ij}^\alpha) = \begin{pmatrix} \cos \psi^\alpha & -\cos \theta^\alpha \sin \psi^\alpha & \sin \theta^\alpha \sin \psi^\alpha \\ \sin \psi^\alpha & \cos \theta^\alpha \cos \psi^\alpha & -\sin \theta^\alpha \cos \psi^\alpha \\ 0 & \sin \theta^\alpha & \cos \theta^\alpha \end{pmatrix}. \tag{58}$$

For spherical inhomogeneous particles, we have simply $Q_{ij}^\alpha = \delta_{ij}$, which is the Kronecker delta. By a Taylor expansion of the Green's function, eqns (54) and (55) may be written as

$$E_i(\mathbf{x}; \Omega^1, \dots, \Omega^M) = E_i^0 - \sum_{\alpha=1}^M \sum_{k=1}^{\infty} \frac{(-1)^k}{k!} P_{s_1 \dots s_k}^{(e)(\alpha)}(\Omega^1, \dots, \Omega^M) G_{is_1 \dots s_k}^e(\mathbf{x} - \mathbf{x}^\alpha) \tag{59}$$

$$u_i(\mathbf{x}; \Omega^1, \dots, \Omega^M) = u_i^0 + \sum_{\alpha=1}^M f_j^{(\alpha)}(\Omega^1, \dots, \Omega^M) G_{ij}(\mathbf{x} - \mathbf{x}^\alpha) + \sum_{\alpha=1}^M \sum_{k=1}^{\infty} \frac{(-1)^k}{k!} P_{j_1 \dots j_k}^{(z)}(\Omega^1, \dots, \Omega^M) G_{ij_1 \dots j_k}(\mathbf{x} - \mathbf{x}^\alpha) \tag{60}$$

which shows that given discretely-distributed inhomogeneities may be modelled by a discrete distribution of the induced electric and elastic multipoles, defined respectively by

$$P_{s_1 \dots s_k}^{(e)(\alpha)} = k \int_{V_I} (P_{s_1}^{(0)(\alpha)} + \Delta \epsilon_{j s_1} E_j^{(\alpha)}) y_{s_2}^\alpha \dots y_{s_k}^\alpha d\xi \quad (k = 1, 2, \dots) \tag{61}$$

and

$$f_j^{(\alpha)} = \int_{V_I} P_n^{(z)} E_{j,n}^{(\alpha)} d\xi \tag{62}$$

$$P_{j_1 \dots j_k}^{(z)} = -k \int_{V_I} \left[\Delta C_{mnj_1} u_{m,n}^{(\alpha)} - P_{s_1}^{(z)} E_{j_1}^{(\alpha)} - \frac{1}{k} P_n^{(z)} E_{j_1 n}^{(\alpha)} y_{s_1}^\alpha \right] y_{s_2}^\alpha \dots y_{s_k}^\alpha d\xi \tag{63}$$

for $k = 1, 2, \dots$, where $y_{s_2}^\alpha \dots y_{s_k}^\alpha = 1$ when $k = 1$. It is also shown that the total resultant

charge (the zeroth order of the electric multipole) of the point-charge arrays modelling the inhomogeneities vanishes, since these inhomogeneities are self-electrically neutral. The induced body-force term due to the electric and elastic interaction given by eqn (62), in general, does not vanish. If, however, the microscopic electric field inside the particles is uniform, this term will be zero.

Considering now an ensemble of large numbers of such samples, the ensemble average of the electric field and the elastic displacement field in the composite may be expressed by

$$\begin{aligned} \langle E_i \rangle(\mathbf{x}) &= \int_{\Omega} \cdots \int_{\Omega} f(\Omega^1, \dots, \Omega^M) E_i(\mathbf{x}; \Omega^1, \dots, \Omega^M) d\Omega^1 \dots d\Omega^M \\ &= E_i^0 - \sum_{k=1}^{\infty} \frac{(-1)^k}{k!} \int_{V_R} \bar{P}_{j_1 \dots j_k}^e(\mathbf{x}') G_{,i j_1 \dots j_k}^e(\mathbf{x}, \mathbf{x}') d\mathbf{x}' \end{aligned} \tag{64}$$

$$\begin{aligned} \langle u_i \rangle(\mathbf{x}) &= \int_{\Omega} \cdots \int_{\Omega} f(\Omega^1, \dots, \Omega^M) u_i(\mathbf{x}; \Omega^1, \dots, \Omega^M) d\Omega^1 \dots d\Omega^M \\ &= u_i^0 + \int_{V_R} \bar{J}_j(\mathbf{x}') G_{ij}(\mathbf{x}, \mathbf{x}') d\mathbf{x}' + \sum_{k=1}^{\infty} \frac{(-1)^k}{k!} \int_{V_R} \bar{P}_{j_1 \dots j_k}(\mathbf{x}') G_{i j_1 \dots j_k}(\mathbf{x}, \mathbf{x}') d\mathbf{x}' \end{aligned} \tag{65}$$

where $f(\Omega^1, \dots, \Omega^M)$ is the M -point joint probability density function, and $\Omega^x \in \Omega$ which is a defined space, $\Omega = V_R \times \Gamma$, in which $\Gamma = (0, \pi) \times (0, 2\pi) \times (0, 2\pi)$ is a parameter space and V_R the volume of the composite specimen.

Equations (64) and (65) show that the macroscopic ensemble average behavior of such M statistically-distributed inhomogeneous particles may be modelled overall by a distribution of induced statistical continuum electric and elastic multipoles defined respectively by

$$\bar{P}_{j_1 \dots j_k}^e(\mathbf{x}') = \sum_{\alpha=1}^M \int_{\Gamma} Z_{j_1 \dots j_k}^{e(\alpha)}(\Omega') d\Gamma' \tag{66}$$

and

$$\bar{J}_j(\mathbf{x}') = \sum_{\alpha=1}^M \int_{\Gamma} Z_j^{(\alpha)}(\Omega') d\Gamma' \tag{67}$$

$$\bar{P}_{j_1 \dots j_k}(\mathbf{x}') = \sum_{\alpha=1}^M \int_{\Gamma} Z_{j_1 \dots j_k}^{(\alpha)}(\Omega') d\Gamma' \tag{68}$$

where

$$Z_{j_1 \dots j_k}^{e(\alpha)} = \underbrace{\int_{\Omega} \cdots \int_{\Omega}}_{M-1} f(\Omega^1, \dots, \Omega^M) P_{j_1 \dots j_k}^{e(\alpha)} d\Omega^1 \dots d\Omega^{\alpha-1} d\Omega^{\alpha+1} \dots d\Omega^M \tag{69}$$

and

$$Z_j^{(\alpha)} = \underbrace{\int_{\Omega} \cdots \int_{\Omega}}_{M-1} f(\Omega^1, \dots, \Omega^M) f_j^{(\alpha)}(\Omega^1, \dots, \Omega^M) d\Omega^1 \dots d\Omega^{\alpha-1} d\Omega^{\alpha+1} \dots d\Omega^M \tag{70}$$

$$Z_{j_1 \dots j_k}^{(\alpha)} = \underbrace{\int_{\Omega} \cdots \int_{\Omega}}_{M-1} f(\Omega^1, \dots, \Omega^M) P_{j_1 \dots j_k}^{(\alpha)} d\Omega^1 \dots d\Omega^{\alpha-1} d\Omega^{\alpha+1} \dots d\Omega^M \tag{71}$$

in which $d\Omega^{\alpha-1} = 1$ when $\alpha = 1$ and $d\Omega^{\alpha+1} = 1$ when $\alpha = M$.

Some special cases may be of interest. If the electric and elastic multipoles modelling the α th inhomogeneity do not explicitly depend on the geometrical positions and orientations of its surrounding inhomogeneities, i.e.

$$P_{s_1 \dots s_k}^{e(\alpha)} = P_{s_1 \dots s_k}^e(\Omega^\alpha) \quad (72)$$

and

$$f_j^{(\alpha)} = f_j(\Omega^\alpha) \quad (73)$$

$$P_{j s_1 \dots s_k}^{(\alpha)} = P_{j s_1 \dots s_k}(\Omega^\alpha) \quad (74)$$

eqns (66), (67) and (68) may be reduced to

$$\bar{P}_{s_1 \dots s_k}^e(\mathbf{x}') = \int_{\Gamma} \rho(\Omega') P_{s_1 \dots s_k}^e(\Omega') d\Gamma' \quad (75)$$

and

$$\bar{f}_j(\mathbf{x}') = \int_{\Gamma} \rho(\Omega') f_j(\Omega') d\Gamma' \quad (76)$$

$$\bar{P}_{j s_1 \dots s_k}(\mathbf{x}') = \int_{\Gamma} \rho(\Omega') P_{j s_1 \dots s_k}(\Omega') d\Gamma' \quad (77)$$

where the function $\rho(\Omega')$ is defined in eqn (42). Examples of such cases may be dilute suspensions or systems in which the microscope electric and elastic fields in the particles may be solved by using the self-consistent scheme approximation. Other cases, such as systems suitable for the pair interaction approximation or the nearest interaction approximation, may also exist and eqns (66)–(68) can also be simplified. It is seen that reasonable approximations may not only simplify the many-body interaction problem, but may also make it possible to obtain the necessary statistical information in many cases.

5. EFFECTIVE PROPERTIES OF COMPOSITES WITH RANDOM MICROSTRUCTURES

The problem of interest in this section is to see how the proposed statistical continuum multipole approach can be used to find explicitly the effective properties of the composite and the statistical anisotropy and shape effects of the microstructures on the overall properties of composites with randomly-distributed inhomogeneities. For simplicity, we shall study elastic dielectric composites in which the electroelastic interaction effect is ignored. The problem of finding the effective permittivity and the effective elastic properties of the composite is thus fully separated. We may now consider the following microscopic constitutive relations

$$\mathbf{D} = \left(\varepsilon + \sum_{\alpha=1}^M \Delta \varepsilon \gamma^\alpha \right) \cdot \mathbf{E} \quad (78a)$$

and

$$\mathbf{t} = \left(\mathbf{C} + \sum_{\alpha=1}^M \Delta \mathbf{C} \gamma^\alpha \right) : \mathbf{e} \quad (78b)$$

where γ^α is the indicative function of the α th inhomogeneous particle. The ensemble average of relations (78a) and (78b) then gives

$$\langle \mathbf{D} \rangle = \boldsymbol{\varepsilon} \cdot \langle \mathbf{E} \rangle + \Delta \boldsymbol{\varepsilon} \cdot \left\langle \sum_{\alpha=1}^M \gamma^\alpha \mathbf{E} \right\rangle \tag{79a}$$

and

$$\langle \mathbf{t} \rangle = \mathbf{C} : \langle \mathbf{e} \rangle + \Delta \mathbf{C} : \left\langle \sum_{\alpha=1}^M \gamma^\alpha \mathbf{e} \right\rangle. \tag{79b}$$

Due to the linearity of the problems, the microscopic electric and elastic fields in the α th inclusion ($\alpha = 1, 2, \dots, M$) may be expressed by

$$E_i^{(\alpha)}(\boldsymbol{\xi}; \Omega^1, \dots, \Omega^M) = T_{ij}^\alpha(\boldsymbol{\xi}; \Omega^1, \dots, \Omega^M) E_j^0, \quad \boldsymbol{\xi} \in V_\alpha \tag{80a}$$

and

$$e_{ij}^{(\alpha)}(\boldsymbol{\xi}; \Omega^1, \dots, \Omega^M) = A_{ijkl}^\alpha(\boldsymbol{\xi}; \Omega^1, \dots, \Omega^M) e_{kl}^0, \quad \boldsymbol{\xi} \in V_\alpha \tag{80b}$$

where tensors \mathbf{T}^α and \mathbf{A}^α , in general, are unknown beforehand and have to be determined by solving the equations of microscopic electric and elastic fields.

With the use of electric and elastic multipole modelling, the microscopic electric and elastic fields in the composite may be written respectively as

$$E_i(\mathbf{x}; \Omega^1, \dots, \Omega^M) = F_{ij}(\mathbf{x}; \Omega^1, \dots, \Omega^M) E_j^0 \tag{81a}$$

and

$$e_{ij}(\mathbf{x}; \Omega^1, \dots, \Omega^M) = B_{ijkl}(\mathbf{x}; \Omega^1, \dots, \Omega^M) e_{kl}^0 \tag{81b}$$

in which

$$F_{ij} = \delta_{ij} + \sum_{\beta=1}^M R_{ij}^\beta \tag{82a}$$

and

$$B_{ijkl} = \delta_{ik} \delta_{jl} + \sum_{\beta=1}^M Y_{ijkl}^\beta \tag{82b}$$

where

$$R_{ij}^\beta = \sum_{k=1}^k \frac{(-1)^{k-1}}{(k-1)!} G_{im_1 \dots m_k}^\alpha(\mathbf{x} - \mathbf{x}^\beta) \int_{V_\beta} \Delta \varepsilon_{p_1} T_{pj}^\beta y_{s_2}^\beta \dots y_{s_k}^\beta d\xi \tag{83a}$$

and

$$Y_{ijkl}^\beta = \sum_{k=1}^k \frac{(-1)^{k-1}}{(k-1)!} g_{lm_1 \dots m_k}(\mathbf{x} - \mathbf{x}^\beta) \int_{V_\beta} \Delta C_{p_1 m_1} A_{p_1 q_1 k l}^\beta y_{s_2}^\beta \dots y_{s_k}^\beta d\xi \tag{83b}$$

with

$$g_{lm_1 \dots m_k}(\mathbf{x} - \mathbf{x}^\beta) = \frac{1}{2} [G_{m, \beta_1 \dots \beta_k}(\mathbf{x} - \mathbf{x}^\beta) + G_{m, \beta_1 \dots \beta_k}(\mathbf{x} - \mathbf{x}^\beta)]. \tag{84}$$

The effective properties of the composite can thus be determined by comparing its definition

$$\langle \mathbf{D} \rangle = \boldsymbol{\varepsilon}^{\text{eff}} \cdot \langle \mathbf{E} \rangle \tag{85a}$$

and

$$\langle \mathbf{t} \rangle = \mathbf{C}^{\text{eff}} : \langle \mathbf{e} \rangle \quad (85b)$$

with eqns (79a,b), which leads to the result

$$\varepsilon_{ij}^{\text{eff}} = \varepsilon_{ij} + \Delta \varepsilon_{ip} \left\langle \sum_{\alpha=1}^M i^{\alpha} T_{pk}^{\alpha} \right\rangle \langle F_{kj} \rangle^{-1} \quad (86a)$$

and

$$C_{ijkl}^{\text{eff}} = C_{ijkl} + \Delta C_{ijpq} \left\langle \sum_{\alpha=1}^M i^{\alpha} A_{pqmn}^{\alpha} \right\rangle \langle B_{mnl} \rangle^{-1}. \quad (86b)$$

Obviously to perform the ensemble average, the details of the statistical information on the microstructure of the composite have to be given. In addition, to complete the calculation, one has to find the microscopic electric and elastic fields in the particles, which seems to be impossible in practice because of the complicated many-body interaction problems as well as their statistical character. Some physically reasonable assumptions are, therefore, needed to simplify the problem and make it solvable. For instance, for small-concentration suspensions, a first-order approximation can be made by neglecting interaction between particles, where classical solutions of a single inhomogeneous particle embedded in an infinite medium may be used [see Eshelby (1961), Mura (1982) and Böttcher (1973)]. Higher-order approximations accounting for some interaction effects have also been proposed as, for instance, the nearest interaction model of using a solution of a pair of interactive particles [see Jeffrey (1973, 1974) and McCoy and Beran (1976)] and the classical self-consistent scheme [see Hershey (1954) and Hashin (1968)]. For weakly-inhomogeneous composites, theories of bounds and classical perturbation methods have been used quite successfully [see Hashin and Shtrikman (1962), Beran (1968), Hori (1973) and Willis (1977)].

In this section, we shall use the statistical continuum multipole approach to study the effective properties of an elastic and dielectric composite with M statistically-distributed identical inhomogeneous elastic and dielectric ellipsoidal particles with elastic moduli \mathbf{C}^* and permittivity ε^* embedded in a homogeneous matrix with elastic moduli \mathbf{C} and permittivity ε . To see how the statistical continuum multipole approach works, we consider a dilute suspension system, i.e. the effect of interaction between particles may be neglected. Under the assumption of the statistical homogeneity but not necessarily statistical isotropic on the geometric arrangement of the inhomogeneous particles, the probability density function (see Section 3) can be introduced as

$$\rho(\Omega') = M \rho^*(\Gamma') / V_R \quad (87)$$

in which

$$\rho^*(\Gamma') = \sum_{\alpha=1}^M W_{\alpha}^*(\Gamma') / M \quad (88)$$

where

$$W_{\alpha}^*(\Gamma') = \underbrace{\int_{\Gamma} \cdots \int_{\Gamma}}_{M-1} w(\Gamma^1, \dots, \Gamma^M) d\Gamma^1 \cdots d\Gamma^{\alpha-1} d\Gamma^{\alpha+1} \cdots d\Gamma^M \quad (89)$$

with the normalization condition

$$\int_{\Gamma} \dots \int_{\Gamma} w(\Gamma^1, \dots, \Gamma^M) d\Gamma^1 \dots d\Gamma^M = 1. \tag{90}$$

In the dilute approximation, the microscopic electric and elastic strain fields in the α th ellipsoidal inclusion (particle) may be found, as a single inhomogeneous ellipsoid with a certain orientation in an infinite matrix subjected to the external electric field \mathbf{E}^0 and the external strain field \mathbf{e}^0 , to be

$$E_m^{(\alpha)} = Q_{mi}(\Gamma^\alpha) Q_{nj}(\Gamma^\alpha) T_{ij} E_n^0 \tag{91a}$$

and

$$e_{mn}^{(\alpha)} = Q_{mi}(\Gamma^\alpha) Q_{nj}(\Gamma^\alpha) Q_{pk}(\Gamma^\alpha) Q_{ql}(\Gamma^\alpha) A_{ijkl} e_{pq}^0 \tag{91b}$$

where T_{ij} and A_{ijkl} are the transformation tensors that characterize respectively the electric and elastic field in an ellipsoidal particle with its axes coincide with a chosen coordinate system (see Fig. 3) in which $\theta = 0, \psi = 0$ and $\omega = 0$. They are both known constant tensors [see Stratton (1941) and Mura (1982)].

The electric and elastic multipoles defined by eqns (61)–(63) can thus be found by inserting eqn (91). For instance, the electric dipole is found as

$$P_m^{e(\alpha)} = V_t \Delta \epsilon_{mk} Q_{ki}(\Gamma^\alpha) Q_{nj}(\Gamma^\alpha) T_{ij} E_n^0 \tag{92}$$

where one has set $\mathbf{P}^{0(\alpha)} = 0$ for inhomogeneous particles without spontaneous polarization, and the elastic monopole is found as

$$P_{st}^{e(\alpha)} = -V_t \Delta C_{stmn} Q_{mi}(\Gamma^\alpha) Q_{nj}(\Gamma^\alpha) Q_{pk}(\Gamma^\alpha) Q_{ql}(\Gamma^\alpha) A_{ijkl} e_{pq}^0 \tag{93}$$

where only the elastic contribution is considered.

An interesting result in the case of statistical homogeneity is found; since the statistical continuum electric and elastic multipoles defined in eqns (75)–(77) are independent of \mathbf{x}' in the dilute approximation, eqn (64) can be reduced to

$$\langle E_m \rangle = E_m^0 - \frac{1}{\epsilon} \bar{P}_i^e L_{im}^e \tag{94}$$

where $\bar{\mathbf{P}}^e$ is the statistical continuum electric dipole moment given by

$$\bar{P}_k^e = f \Delta \epsilon \left[\int_{\Gamma} \rho^*(\Gamma') Q_{ki}(\Gamma') Q_{nj}(\Gamma') d\Gamma' \right] T_{ij} E_n^0 \tag{95}$$

with $f = MV_I/V_R$ being the volume fraction of inhomogeneous particles in the composite and L^e the electric depolarizing tensor given by

$$L_{im}^e = -\epsilon \int_{V_R} G_{,im}(\mathbf{x} - \mathbf{x}') d\mathbf{x}' = \frac{1}{3} \delta_{im} \tag{96}$$

for \mathbf{x} inside the spherical volume V_R . Here, the matrix and the particle are both assumed to be isotropic for simplicity.

Also, from eqn (65), one can find the ensemble average of the elastic strain fields in the composite by

$$\langle e_{ij} \rangle = e_{ij}^0 + L_{ijkl} \bar{P}_{lk} \tag{97}$$

where \bar{P}_{lk} is the statistical continuum elastic monopole moment given by

$$\bar{P}_{ik} = -f \Delta C_{ikmn} \left[\int_{\Gamma} \rho^*(\Gamma') Q_{mi}(\Gamma') Q_{nj}(\Gamma') Q_{ps}(\Gamma') Q_{qt}(\Gamma') d\Gamma' \right] A_{ijkl} e_{pq}^0 \quad (98)$$

and L_{ijkl} is the elastic depolarization tensor defined by

$$L_{ijkl} = -\frac{1}{2} \int_{S_R} (G_{il,j} + G_{jl,i}) n'_k dS' \quad (99)$$

which can be given explicitly from eqn (25) for isotropic elastic matrix media.

Now, comparing eqn (94) with eqn (81a), we find

$$\langle F_{mk} \rangle = \delta_{mk} - \frac{\Delta \epsilon f}{3\epsilon} T_{ij} \int_{\Gamma} \rho^*(\Gamma') Q_{mi}(\Gamma') Q_{kj}(\Gamma') d\Gamma'; \quad (100)$$

and, comparing eqn (97) with eqn (81b), we get

$$\langle B_{ijkl} \rangle = \delta_{ip} \delta_{jq} - f L_{ijkl} \Delta C_{klmn} \left[\int_{\Gamma} \rho^*(\Gamma') Q_{mi}(\Gamma') Q_{nj}(\Gamma') Q_{ps}(\Gamma') Q_{qt}(\Gamma') d\Gamma' \right] A_{ijkl}. \quad (101)$$

Furthermore, by noting eqns (80a) and (91a), we find

$$\left\langle \sum_{z=1}^M \gamma^z T_{ki}^z \right\rangle = f \int_{\Gamma} \rho^*(\Gamma') T_{ij} Q_{ki}(\Gamma') Q_{lj}(\Gamma') d\Gamma'; \quad (102)$$

and, by noting eqns (80b) and (91b), we get

$$\left\langle \sum_{z=1}^M \gamma^z A_{mnpq}^z \right\rangle = f \int_{\Gamma} \rho^*(\Gamma') A_{ijkl} Q_{mi}(\Gamma') Q_{nj}(\Gamma') Q_{pk}(\Gamma') Q_{ql}(\Gamma') d\Gamma'. \quad (103)$$

Thus, we have obtained, in general, analytical expressions, eqn (86) and eqns (100)–(103), for the determination of the effective permittivity and of the effective elastic moduli of a statistically-anisotropic dielectric and elastic composite in the dilute suspension approximation, provided that the probability density function $\rho^*(\Gamma')$ of the statistical distribution of orientations of the particles is given.

For simplicity as well as for the similarity of the dielectric and elastic problems, we shall now consider a concrete example in which we would like to find the effective permittivity of a (rigid) dielectric composite with M statistically-distributed identical inhomogeneous rigid ellipsoidal particles. We assume that the dielectric composite contains M ellipsoidal inhomogeneous particles which are statistically distributed in a uniform matrix. The orientations of the particles are assumed to be characterized by the Gaussian distribution

$$\rho^*(\Gamma') = C \exp(-\sigma^2 |\Gamma' - \Gamma^0|^2) \quad (104)$$

where σ is a statistical parameter determined and controllable by the method of manufacture of the composite, and C is the constant determined by the following normalization condition

$$\int_{\Gamma} \rho^*(\Gamma') d\Gamma' = 1. \quad (105)$$

According to Stratton (1941), we may write explicitly

$$Q_{ki}(\Gamma') Q_{lj}(\Gamma') T_{ij} = \sum_{i=1}^3 \lambda_i Q_{ki}(\Gamma') Q_{lj}(\Gamma') \quad (106)$$

where

$$\lambda_i = \left(1 + abc \frac{\Delta \epsilon}{2\epsilon} B_i \right)^{-1} \quad (107)$$

in which

$$B_1 = \int_0^x \frac{ds}{(s+a^2)^{3/2} \sqrt{(s+b^2)(s+c^2)}} \quad (108)$$

and

$$B_2 = \int_0^x \frac{ds}{(s+b^2)^{3/2} \sqrt{(s+a^2)(s+c^2)}} \quad (109)$$

and

$$B_3 = \int_0^x \frac{ds}{(s+c^2)^{3/2} \sqrt{(s+a^2)(s+b^2)}} \quad (110)$$

where a , b and c are the axes of the ellipsoid.

It can then be seen from eqns (86a), (100) and (102) that the shape effect of the inhomogeneous particles on the overall electric permittivity of the composite is taken into account in the parameters λ_i ($i = 1, 2, 3$), while the effect of the statistical orientations of the particles is taken into account by the orientation tensor $\mathbf{Q}(\Gamma')$ as well as the probability density function $\rho^*(\Gamma')$. In the case of spherical inhomogeneous particles, the problem is much simplified and we have

$$\lambda_1 = \lambda_2 = \lambda_3 = \frac{3\epsilon}{3\epsilon + \Delta\epsilon} \quad (111)$$

and

$$\langle F_{kj} \rangle = \frac{3\epsilon + (1-f)\Delta\epsilon}{3\epsilon + \Delta\epsilon} \delta_{kj} \quad (112)$$

and

$$\left\langle \sum_{x=1}^M \gamma^x T_{kj}^x \right\rangle = \frac{3\epsilon f}{3\epsilon + \Delta\epsilon} \delta_{kj}. \quad (113)$$

The effective permittivity of a composite with dilute spherical inhomogeneous particles can thus be obtained as

$$\epsilon^{\text{eff}} = \epsilon \frac{2\epsilon + \epsilon^* + 2f\Delta\epsilon}{2\epsilon + \epsilon^* - f\Delta\epsilon} \quad (114)$$

which is in accordance with the Rayleigh mixture formula (Rayleigh, 1892) obtainable by using a volume-average method. The equivalence of the ensemble average and the volume average is then proved in this case. It should be noticed, however, that real physical systems very rarely satisfy the ergodic hypothesis (Kröner, 1986). The result also shows that the derivation of the Rayleigh mixture formula for a composite with statistically-homogeneous distributed dilute spherical particles is independent of the assumption of statistical isotropy for the composite provided that one ignores the interaction among particles.

To see explicitly the shape effect of inhomogeneous particles on the overall effective properties of a dielectric composite, let us consider ellipsoidal particles (with $a = b$). If one

assumes that all directions of the orientations of the particles are equally probable, the probability of the particle orientation being within the range $(\theta', \theta' + d\theta')$ and $(\psi', \psi' + d\psi')$ then reads

$$\rho^*(\Gamma') d\Gamma' = \frac{1}{4\pi} \sin \theta' d\theta' d\psi'. \tag{115}$$

Using eqns (58), (86a), (100) and (102), the effective permittivity of the dielectric composite can thus be derived as

$$\epsilon^{\text{eff}} = \epsilon \frac{9\epsilon + 2f\Delta\epsilon(2\lambda_1 + \lambda_3)}{9\epsilon - f\Delta\epsilon(2\lambda_1 + \lambda_3)} \tag{116}$$

in which the shape effect parameters λ_1 and λ_2 are given by eqns (107)–(110) with $a = b$. For $c \gg a$, one has the rod or needle type of particle. For $c \ll a$ and close to zero, one has the disc type of particles. For $c = a$, one gets the spherical particles. The numerical results of the effective permittivity for different types of particle shapes are shown in Fig. 4.

It is shown that for fully-random orientations of ellipsoidal particles, the ensemble-average behavior of a dielectric composite displays an isotropic property. If now the orientations of the particles ($a = b$) have a preferred direction, say in the direction of the x_1 -axis ($\theta^0 = 0$), and if their statistical distribution is represented by a Gaussian distribution (104), one can then write

$$\rho^*(\Gamma') d\Gamma' = C \exp(-\sigma^2 \theta'^2) \sin \theta' d\theta' d\psi' \tag{117}$$

where σ is a statistical parameter characterizing the standard deviation of θ' and C is a constant determined by eqn (105), which can be written as

$$C = \left[2\pi \int_0^\pi \exp(-\sigma^2 \theta'^2) \sin \theta' d\theta' \right]^{-1}. \tag{118}$$

After some calculations, we can obtain

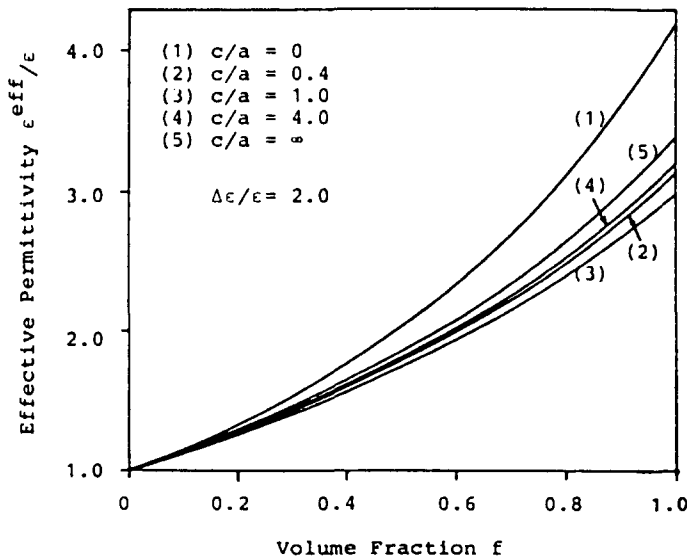


Fig. 4. Shape effect of ellipsoidal inhomogeneous particles on the effective permittivity of the dielectric composite.

$$(\epsilon_{ij}^{\text{eff}}) = \epsilon \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & \eta \end{pmatrix} \quad (119)$$

where the dimensionless parameter η is defined by

$$\eta = \frac{(3\epsilon + 2\Delta\epsilon f A_2)(3\epsilon - 2\Delta\epsilon f A_1)}{(3\epsilon + 2\Delta\epsilon f A_1)(3\epsilon - 2\Delta\epsilon f A_2)} \quad (120)$$

with the constants A_1 and A_2 being given respectively by

$$A_1 = \pi \int_0^\pi C \exp(-\sigma^2 \theta^2) (\lambda_1 (1 + \cos^2 \theta) + \lambda_3 \sin^2 \theta) \sin \theta \, d\theta \quad (121)$$

and

$$A_2 = 2\pi \int_0^\pi C \exp(-\sigma^2 \theta^2) (\lambda_1 \sin^2 \theta + \lambda_3 \cos^2 \theta) \sin \theta \, d\theta. \quad (122)$$

It is shown that the overall effective permittivity of the composite can be anisotropic, provided that the orientations of the microellipsoidal particles statistically have a preferred direction. In the considered (transversely isotropic) case, the macroscopic effective permittivities are found to have two independent constants and they are given in eqn (119). The dependence of some numerical values of the macroscopic anisotropic parameter η on the statistical parameter σ and the shape of the microparticles is shown in Fig. 5.

6. OVERALL BEHAVIOR OF ELASTIC DIELECTRIC COMPOSITES

This section is concerned with the study of the overall behavior of an elastic dielectric composite, in which some effects of the interaction between the electric field and elastic field are taken into account. The constitutive relations characterizing the overall behavior of the elastic dielectric composite will be derived with the use of the statistical continuum multipole

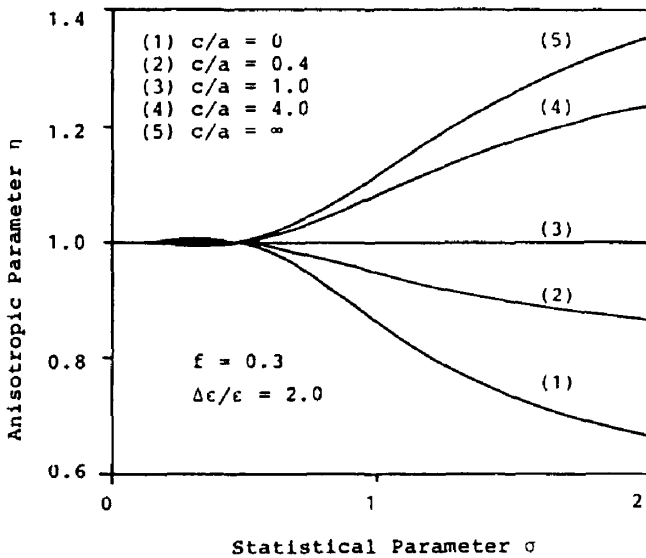


Fig. 5. Statistical anisotropy and shape effects of inhomogeneous particles on the effective permittivities of the composite.

approach, in which no pre-assumptions on the macroscopic constitutive relations of the composite are needed. This result thus makes the statistical continuum multipole approach superior to the classical effective medium theory where the effective constitutive relations of composites have to be pre-assumed. To make a distinction, we shall call the derived governing equations for the overall behavior of the elastic dielectric composites the overall macroscopic constitutive relations and call their coefficients the overall macroscopic material coefficients rather than the effective constitutive relations and the effective material coefficients defined in effective medium theory.

Consider an elastic dielectric composite with M randomly-distributed elastic dielectric inhomogeneous particles. The material properties of the matrix and the particles are supposed to be the same as those described in Section 2. With the use of the result given in Section 2, we may find that the ensemble-average electric polarization and elastic stress fields can be expressed in the following form

$$\langle P_i \rangle = \epsilon_0 \chi \langle E_i \rangle + \epsilon_0 \Delta \chi \left\langle \sum_{\alpha=1}^M \gamma^\alpha E_i \right\rangle \quad (123)$$

and

$$\langle t_{ij} \rangle = \lambda \langle e_{kk} \rangle \delta_{ij} + 2\mu \langle e_{ij} \rangle + \left\langle \sum_{\alpha=1}^M \gamma^\alpha (\Delta \lambda e_{kk} \delta_{ij} + 2\Delta \mu e_{ij} - P_i E_j) \right\rangle. \quad (124)$$

To perform the ensemble average, the detailed solution of the microscopic fields has to be obtained. For simplicity, we consider an elastic dielectric composite with dilute suspension of spherical inhomogeneous particles and make the assumptions of statistical homogeneity and statistical isotropy for the composite. To first order, the interaction between particles is ignored for the dilute system, and the influence of the small change of particle orientations caused by small elastic deformation on the overall electric field is neglected. In such a case, after some calculations, we can obtain

$$\langle \mathbf{P} \rangle = \epsilon_0 \chi^{\text{mac}} \langle \mathbf{E} \rangle \quad (125)$$

where the overall macroscopic dielectric susceptibility can be written as

$$\chi^{\text{mac}} = \chi + \frac{3\epsilon f \Delta \chi}{3\epsilon + (1-f)\Delta \epsilon} \quad (126)$$

which is in accordance with the classical Rayleigh mixture formula for the effective dielectric susceptibility of composites with dilute spherical particles in the rigid-body approximation.

The ensemble-average strain field may be written as

$$\langle e_{ij} \rangle = e_{ij}^0 + L_{ijkl} \bar{P}_{kl} \quad (127)$$

where the statistical continuum elastic monopole moment $\bar{\mathbf{P}}$ may be found from eqns (63), (74) and (77) as

$$\bar{P}_n = - \frac{(3\kappa + 4\mu)f}{3\kappa + 4\mu + 3\Delta \kappa} \left\{ 3\Delta \kappa e_n^0 - \frac{(3\epsilon)^2 \epsilon_0 \chi^*}{[3\epsilon(1-f)\Delta \epsilon]^2} \langle E_i \rangle \langle E_i \rangle + \frac{3\epsilon \Delta \epsilon - \epsilon_0 \chi^*}{(3\epsilon + \Delta \epsilon)^2} (p^0)^2 \right\} \quad (128)$$

where the sum is over the suffix i , and for $i \neq j$

$$\bar{P}_{ij} = -\frac{5\mu(3\kappa+4\mu)f}{5\mu(3\kappa+4\mu)+(6\kappa+12\mu)\Delta\mu} \left\{ 2\Delta\mu e_{ij}^0 - \frac{(3\varepsilon)^2 \varepsilon_0 \chi^*}{[3\varepsilon+(1-f)\Delta\varepsilon]^2} \langle E_i \rangle \langle E_j \rangle \right\}. \quad (129)$$

By noting eqns (25) and (94), we finally arrive at the result that the overall macroscopic stress in an elastic dielectric composite with dilute spherical particles in the absence of permanent electric polarizations may be written as

$$\langle t_{ij} \rangle = (\kappa^{\text{mac}} - \frac{2}{3}\mu^{\text{mac}}) \langle e_{nn} \rangle \delta_{ij} + 2\mu^{\text{mac}} \langle e_{ij} \rangle - \left[\frac{1}{3}(\xi - \tau) \langle P_n \rangle \langle E_n \rangle \delta_{ij} + \tau \langle P_j \rangle \langle E_i \rangle \right] \quad (130)$$

where the overall macroscopic elastic moduli read

$$\kappa^{\text{mac}} = \kappa + \frac{(3\kappa+4\mu)f\Delta\kappa}{3\kappa+4\mu+3(1-f)\Delta\kappa} \quad (131)$$

and

$$\mu^{\text{mac}} = \mu + \frac{5\mu(3\kappa+4\mu)f\Delta\mu}{5\mu(3\kappa+4\mu)+6\Delta\mu(1-f)(\kappa+2\mu)} \quad (132)$$

which are in accordance with classical results for the effective elastic moduli of an elastic composite with dilute spherical particles in the case of no electric forces [see, for instance, Christensen (1979)].

By taking into account the electroelastic interaction, it is found from eqn (130) that there are two new dimensionless macroscopic electroelastic parameters responsible for the overall behavior of an elastic dielectric composite material, defined by

$$\xi = \frac{f\chi^*}{\left[1 + \frac{3\Delta\kappa(1-f)}{3\kappa+4\mu} \right] \left[1 + (1-f) \frac{\Delta\varepsilon}{3\varepsilon} \right]^2 \chi^{\text{mac}}} \quad (133)$$

and

$$\tau = \frac{f\chi^*}{\left[1 + \frac{6\Delta\mu(1-f)(\kappa+2\mu)}{5\mu(3\kappa+4\mu)} \right] \left[1 + (1-f) \frac{\Delta\varepsilon}{3\varepsilon} \right]^2 \chi^{\text{mac}}} \quad (134)$$

where f denotes the volume fraction of the particles. The numerical results of the two parameters are shown respectively in Figs 6a and 6b for some combinations of different two phase materials.

The equations obtained, (125) and (130), may be used approximately as the overall macroscopic constitutive relations of the composite, provided that applied electric and mechanical loads do not have significant variations within any representative volume which is macroscopically small compared with the total volume of the composite, but is microscopically large enough to contain many particles. However, when treating composites with loads having significant variations within the scale length of several particles, such as cracked composites, the effect of high-order multipoles has to be, in general, taken into account. In addition, if the particles in the composites have spontaneous electrical polarizations $p^0 \mathbf{n}^z$ ($z = 1, 2, \dots, M$), a stress term $\beta_0 (p^0)^2 \delta_{ij}$ with

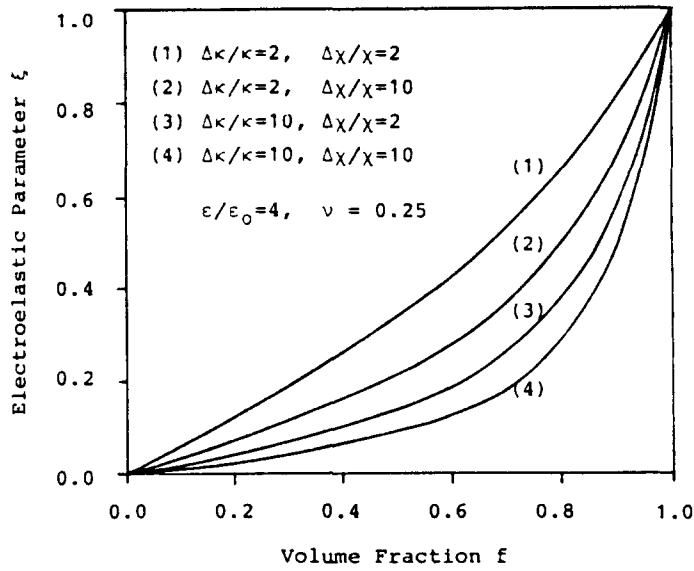


Fig. 6a. Macroscopic electroelastic parameter ξ and its dependence on the volume fraction of the particles and the microscopic material properties.

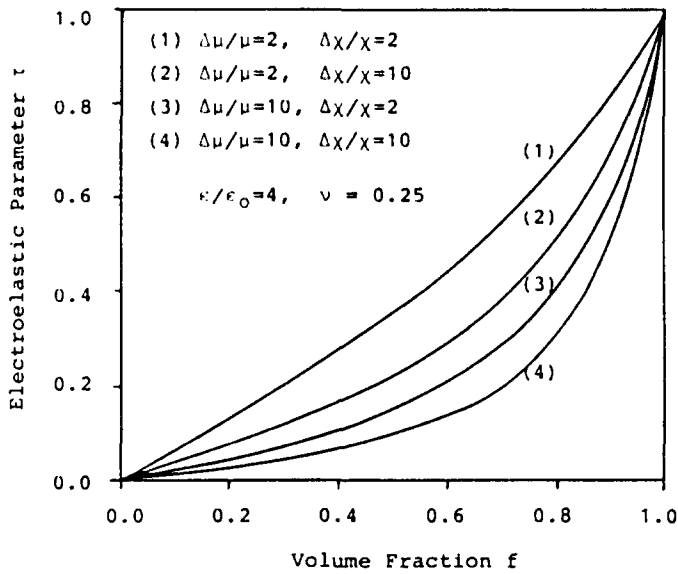


Fig. 6b. Macroscopic electroelastic parameter τ and its dependence on the volume fraction of the particles and the microscopic material properties.

$$\beta_0 = \frac{f(3\epsilon + \Delta\epsilon - \epsilon_0\chi^*)}{3 \left[1 + \frac{3\Delta\kappa(1-f)}{3\kappa + 4\mu} \right] (3\epsilon + \Delta\epsilon)^2} \quad (135)$$

is found to be present in eqn (130). This stress term exists even in the absence of external electric and mechanical loads, and, therefore, may be considered as the internal stress in the composite. The presence of the internal stress is physically understandable since the internal electric field caused by all permanent electric dipole moments of the particles will generate electric forces and torques on the particles, which must be balanced by mechanical forces in equilibrium state. The presented example shows the advantage of the statistical continuum multipole model in comparison with the classical effective medium theory where

the formal effective macroscopic constitutive relations for the composite have to be assumed beforehand, which, however, as shown, may be unknown for complex electromagnetic deformable composite materials. Similar results may also be obtained for certain elastic magnetic composite materials due to the analogy between electric polarization and magnetization.

7. CONCLUSIONS

A statistical continuum material multipole theory has been developed in this article to treat problems of elastic dielectric composites with large numbers of statistically-distributed inhomogeneous elastic dielectric particles. A basic solution accounting for the electroelastic interaction of an ellipsoidal inhomogeneous particle with electric polarization in an infinite elastic dielectric medium is first derived, which modifies the classical Eshelby's elastic solution for an ellipsoidal elastic inhomogeneity in an elastic medium by the presence of the electroelastic interaction. With the use of the solution, the overall macroscopic constitutive relations for elastic dielectric composites as well as their overall macroscopic material parameters accounting for the electroelastic interaction are then obtained. It is found that if the mechanical behaviors of the matrix and the inhomogeneous particles are both assumed to be described microscopically by Hooke's laws, the overall macroscopic mechanical constitutive relation of the elastic dielectric composite can, however, be of non-Hookean form. Furthermore, overall internal stresses may exist in an elastic dielectric composite with inhomogeneous particles having permanent electric dipole moments due to the effects of electric forces and torques acting on the microscopic particles. Illustratively, the statistical anisotropy and shape effects of ellipsoidal inhomogeneous particles and the effect of their orientations on the overall effective properties of the elastic and dielectric composites have also been studied and formulated explicitly with the use of the statistical continuum multipole approach. This theory thus presents its advantages of uniformity, generality and possibility of treating more complicated interaction and electromagnetoelastic coupling phenomena in composite materials.

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