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MICROMECHANICS PREDICTIONS OF THE EFFECTIVE ELECTROELASTIC MODULI OF PIEZOELECTRIC COMPOSITES

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Abstract—The dilute, self-consistent, Mori–Tanaka and differential micromechanics theories are extended to consider the coupled electroelastic behavior of piezoelectric composite materials. The application of each theory is based on Deeg's (1980, unpublished) rigorous three-dimensional electroelastic solution of an ellipsoidal inclusion in an infinite piezoelectric medium. Each micromechanics theory is implemented through a matrix formulation in which the effective electroelastic moduli are conveniently represented by a 9×9 matrix. As in the corresponding uncoupled elastic and electric behavior, the dilute and Mori–Tanaka schemes return explicit estimates for the effective electroelastic moduli. The self consistent method, however, returns an implicit nonlinear algebraic matrix equation for the effective electroelastic moduli. The differential scheme formally results in a set of 81 coupled nonlinear ordinary differential equations for the effective electroelastic moduli. In general, recourse to a numerical scheme is required for the self-consistent and differential theories. Numerical results are presented to illustrate the performance of each model for some typical composite microstructures and the models are compared in light of existing experimental data.

1. INTRODUCTION

Piezoelectric ceramic/polymer composites have become attractive candidates for use in transducers for underwater and biomedical imaging applications. Interest in their use is attributed to their enhanced electromechanical coupling characteristics as compared to the strong, piezoelectric ceramic and compliant (possibly piezoelectrically active) polymer constituents. Their high compliance and low density result in better acoustic impedance matching with water than PZT ceramics. Piezoelectric composites have been developed in many forms including piezoelectric ceramic fibers or particles embedded in a polymer matrix and bored, polymer filled holes in a solid piezoelectric ceramic matrix. In addition, *voided* piezoelectric ceramics and polymers have been shown to possess electroelastic properties superior to the unvoided material. In these materials, the voids are sometimes impregnated with a polymer. The three-dimensional connectivity (Newnham *et al.*, 1978) of the constituents of a piezoelectric composite may be such that a discrete phase is embedded in another serving as a matrix, or they may be combined such that each can be treated on equal footing. Improvements in electromechanical properties of piezoelectric composites, however, are not without their drawbacks. The coupled electroelastic behavior of the constituents presents a level of difficulty not present in the design and analysis of the mechanical behavior of composite materials. Further complicating factors arise from the inherent anisotropy of the piezoelectric materials. Nevertheless, a reasonable amount of theoretical work has been directed towards the study of the effective electroelastic behavior of piezoelectric composite materials.

The microstructural characterization and analysis of piezoelectric composites was launched by Newnham's (1978) *connectivity* theory. This theory is based on the combination of mechanics of materials type parallel and series models. These analyses were extended by Banno (1983) to consider discontinuous reinforcement through a *cubes* approach. A different route was taken by Smith and Auld (1991) in the analysis of continuous fiber-reinforced piezoelectric composites. In each of these approaches, however, a simplifying assumption of either a constant stress or strain component in the composite has been made which then leads to the application of Voigt or Reuss type estimates. A more rigorous treatment of the coupled electroelastic fields in a piezoelectric concentric cylinder geometry has been performed by Grekov *et al.* (1989). An interesting and unique approach has been recently taken by Zhou (1991) in which the coupled electroelastic fields in a piezoelectric composite are modeled by continuum elastic and electric multipoles. To date, though, it appears that only the work of Wang (1992) has incorporated the rigorous solution for the coupled electroelastic fields in a piezoelectric inclusion into the analysis of the behavior of piezoelectric composite materials. Wang's (1992) solution, however, is strictly valid only in the dilute limit where the interaction among reinforcement at finite concentrations is insignificant. That such a small amount of work has been done in this area is somewhat surprising considering the vast amount of corresponding research in the areas of uncoupled elastic and electric composites.

Much of the theoretical study of the behavior of elastic and electric composites has been focused on the development of *micromechanics* models. In general these are based on the assumption of statistical uniformity where recourse is made to the ergodic assumption that local details occur in any single specimen with the same frequency that they occur in a single neighborhood in an ensemble of specimens. Subject to this assumption, ensemble averages are replaced by volume averages over some representative volume which is small relative to the specimen size but large relative to the microscale and thus on average is typical of the entire composite. Those micromechanics theories which have received the most attention and use are the dilute, self-consistent, Mori–Tanaka and differential schemes. These methods have been applied with great success to a number of problems in the uncoupled mechanical and electric behavior of composites. The fundamentals of the methods are fairly well known [for example, see Mura (1987), Taya and Arsenault (1989) and Aboudi (1991)] and thus an in-depth discussion of the methods is not presented here. Recently, though, significant developments regarding theoretical aspects of these models have been provided by Benveniste *et al.* (1991). In particular, they have outlined the theoretical grounds for which the models return symmetric elastic moduli—a necessary criterion for any micromechanics model.

Common to each of these methods is the use of the well-known stress and strain concentration factors obtained through the solution of a single particle embedded in an infinite medium. To this end, recourse is usually made to Eshelby's (1957) solution for the stress and strain in an ellipsoidal inclusion. This is primarily due to two reasons: (i) under a uniform applied load, the stress and strain in the ellipsoidal inclusion are uniform, thus trivializing the otherwise complicated problem of determining the volume averaged fields, and (ii) the ellipsoidal shape allows the simulation of a wide range of microstructural geometry ranging from thin flake to continuous fiber. It appears that these micromechanics approaches have not been further applied to the coupled electroelastic behavior of piezoelectric composites. The objective of this work is precisely that. This is motivated by the reexamination of Deeg's (1980) rigorous analytical solution to piezoelectric inclusion problems by Dunn and Taya (1992) and their derivation of the piezoelectric analogs to Eshelby's tensor in elasticity. From the works of Deeg (1980) and Dunn and Taya (1992), electroelastic concentration factors are derived and utilized in the generalization of the aforementioned micromechanics models to piezoelectric composite media. Each micromechanics approach is implemented through a matrix formulation and thus the final expressions are convenient for numerical computation. The results of the proposed models indicate that the evaluation of the effective electroelastic moduli of piezoelectric composite materials can be done in a manner directly analogous to that of uncoupled elastic and electric composites.

2. RESUME OF EQUATIONS AND NOTATION

There are four representations commonly employed in the theory of linear piezoelectricity to describe the coupled interaction between the electric and elastic variables (Ikeda, 1990). Here, the elastic strain, ε_{ij}, and electric potential gradient, $\phi_{,i}$, are taken as the independent variables and are related to the stress, σ_{ij}, and electric displacement, D_i, by:

$$\sigma_{ij} = C_{ijmn}\varepsilon_{mn} + e_{nij}\phi_{,n}, \tag{1}$$

$$D_i = e_{imn}\varepsilon_{mn} - \kappa_{in}\phi_{,n}, \tag{2}$$

where C_{ijmn}, e_{nij} and κ_{in} are the elastic (measured in a constant electric field), piezoelectric (measured at a constant strain or electric field) and the dielectric (measured at a constant strain) moduli, respectively. The strain and electric field, E_i, are derivable from the elastic displacement, u_i, and electric potential as:

$$\varepsilon_{ij} = \tfrac{1}{2}(u_{i,j}+u_{j,i}), \tag{3}$$

$$E_i = -\phi_{,i}. \tag{4}$$

The complete formulation of the static theory of piezoelectricity requires eqns (1)–(4) to be supplemented with the equations of elastic equilibrium and Gauss' law of electrostatics which in the absence of body forces or free charge are given by:

$$\sigma_{ij,j} = 0, \tag{5}$$

$$D_{i,i} = 0. \tag{6}$$

In the preceding equations, conventional indicial notation is utilized where repeated subscripts are summed over the range 1–3 and a comma denotes partial differentiation.

In the solution of piezoelectric inclusion and inhomogeneity problems, it is convenient to treat the elastic and electric variables on an equal footing. To this end, the notation introduced by Barnett and Lothe (1975) is utilized and is briefly reviewed here. This notation is identical to conventional indicial notation with the exception that lower case subscripts take on the range 1–3, while CAPITAL subscripts take on the range 1–4 and repeated CAPITAL subscripts are summed over 1–4. With this notation, the elastic strain and electric field are expressed as:

$$Z_{Mn} = \begin{cases} \varepsilon_{mn}, & M = 1,2,3, \\ -\phi_{,n}, & M = 4, \end{cases} \tag{7}$$

where Z_{Mn} is derivable from U_M given by:

$$U_M = \begin{cases} u_m, & M = 1,2,3, \\ \phi, & M = 4. \end{cases} \tag{8}$$

Similarly the stress and electric displacement are represented as:

$$\Sigma_{iJ} = \begin{cases} \sigma_{ij}, & J = 1,2,3, \\ D_i, & J = 4. \end{cases} \tag{9}$$

The electroelastic moduli can then be represented as:

$$E_{iJMn} = \begin{cases} C_{ijmn}, & J,M = 1,2,3, \\ e_{nij}, & J = 1,2,3;\, M = 4, \\ e_{imn}, & J = 4;\, M = 1,2,3, \\ -\kappa_{in}, & J,M = 4. \end{cases} \tag{10}$$

The "inverse" of E_{iJMn} is defined as F_{AbiJ} and the symmetry properties of E_{iJMn} are easily derivable from those of C_{ijmn}, e_{nij} and κ_{in}. It is noted that E_{iJMn} as defined by eqn (10), is diagonally symmetric. With this shorthand notation, eqns (1) and (2) can be unified into the single equation:

$$\Sigma_{iJ} = E_{iJMn} Z_{Mn}, \tag{11}$$

or inverting:

$$Z_{Ab} = F_{AbiJ} \Sigma_{iJ}. \tag{12}$$

It is important to note that Z_{Mn}, U_M, Σ_{iJ}, E_{iJMn} and F_{AbiJ} are *not* tensors. Thus, to write corresponding equations in an alternate coordinate system, each individual tensor must be transformed by the well-known laws of tensor transformation. The resulting tensors can then be reunified into the form of eqns (7)–(10).

A matrix formulation

The key variables in eqns (7)–(10) can be represented by 9×1 column vectors (Z_{Mn} and Σ_{iJ}) and 9×9 matrices (E_{iJMn} and F_{AbiJ}) by utilizing the following mapping of adjacent indices, e.g. $(iJ) = (Ji)$ and $(Mn) = (nM)$ for J and $M \neq 4$:

$$(11) \to 1 \quad (22) \to 2 \quad (33) \to 3 \quad (23) \to 4 \quad (13) \to 5 \quad (12) \to 6 \tag{13}$$

$$(14) \to 7 \quad (24) \to 8 \quad (34) \to 9, \tag{14}$$

where for E_{iJMn} and F_{AbiJ} the first pair of indices indicates the row in the 9×9 matrix and the second the column. By inspection of eqns (7)–(12) it is seen that the indices (44) do not occur as a pair. The mappings in eqn (13) are simply those of the well-known Voigt elastic constants. Those of eqn (14) are thus a generalization of the Voigt two-index notation to include the piezoelectric and dielectric moduli. As employed here, the factor of two for the shear strains is accounted for in the 9×1 column vector $\mathbf{Z}$. This matrix formulation will be utilized throughout the subsequent analysis and will be seen to greatly simplify the resulting equations.

3. EFFECTIVE MODULI OF PIEZOELECTRIC COMPOSITES

General expressions for the effective electroelastic moduli of two-phase perfectly bonded piezoelectric composites can be derived by considering the volume average of the piezoelectric field variables σ_{ij}, ε_{ij}, D_i, $\phi_{,i}$:

$$\bar{\mathbf{\Sigma}} = c_1 \bar{\mathbf{\Sigma}}_1 + c_2 \bar{\mathbf{\Sigma}}_2, \tag{15}$$

$$\bar{\mathbf{Z}} = c_1 \bar{\mathbf{Z}}_1 + c_2 \bar{\mathbf{Z}}_2, \tag{16}$$

where an overbar denotes the volume average of a quantity and bold letters denote matrix (9×1 or 9×9) quantities. The subscripts "1" and "2" denote the two piezoelectric phases and c_i is the volume fraction of the ith phase. When considering matrix-based composites, the subscript "1" will be reserved for the matrix. In each phase $\mathbf{Z}$ and Σ are related through the matrix form of the constitutive equations (11) and (12).

Consider the two-phase composite being subjected to homogeneous elastic displacement-electric potential boundary conditions, $\mathbf{Z}^0$, i.e. $u_i(S) = e^0_{ij} x_j$ and $\phi(S) = \phi^0_{,i} x_i$ where S is the surface of the composite. By homogeneous boundary conditions it is meant that when they are applied to a homogeneous solid they result in homogeneous fields. Evaluation of the volume averaged piezoelectric fields results yields the effective electroelastic moduli, $\mathbf{E}$:

$$\bar{\mathbf{\Sigma}} = \mathbf{E} \bar{\mathbf{Z}}. \tag{17}$$

Noting that the perturbation of the strain and potential gradient fields vanishes when integrated over the domain of the entire composite (Dunn and Taya, 1992), $\bar{\mathbf{Z}}$ can be obtained as:

$$\bar{\mathbf{Z}} = \mathbf{Z}^0. \tag{18}$$

This is a generalization of the average strain theorem of elasticity [see for example Aboudi (1991)]. Use of eqns (15)–(17) and the constitutive equations in each phase then yields:

$$\mathbf{E} = \mathbf{E}_1 + c_2(\mathbf{E}_2 - \mathbf{E}_1)\mathbf{A}. \tag{19}$$

In eqn (19), **A** is the strain-potential gradient concentration matrix which relates the average strain and potential gradient in phase 2 to that in the composite, i.e.

$$\bar{\mathbf{Z}}_2 = \mathbf{A}\bar{\mathbf{Z}} = \mathbf{A}\mathbf{Z}^0. \tag{20}$$

In a similar manner, under homogeneous traction-electric displacement boundary conditions, $\mathbf{\Sigma}^0$, the effective moduli, **F**, can be obtained as:

$$\mathbf{F} = \mathbf{F}_1 + c_2(\mathbf{F}_2 - \mathbf{F}_1)\mathbf{B}, \tag{21}$$

where **B** is the stress-electric displacement concentration matrix given by:

$$\bar{\mathbf{\Sigma}}_2 = \mathbf{B}\bar{\mathbf{\Sigma}} = \mathbf{B}\mathbf{\Sigma}^0. \tag{22}$$

The estimation of **A** and **B** is thus the key to predicting the effective electroelastic moduli **E** and **F**. The approximation of **A** and **B** through the use of various micromechanics models is the subject of the subsequent section. In particular, attention will be devoted to obtaining **A** and **E** as the companion solution for **B** and **F** readily follows.

4. MICROMECHANICS MODELS

The dilute approximation

The simplest approximations of **A** and **B** are $\mathbf{A} = \mathbf{I}$ and $\mathbf{B} = \mathbf{I}$ which represent a generalization of the Voigt (1889) and Reuss (1929) approximations for the elastic moduli of composite materials, respectively. The dilute approximation is then the next simplest micromechanics approach. The key assumption made in the dilute approximation is that the interaction among the reinforcing particles in a matrix-based composite can be ignored. That the composite is assumed to be matrix-based renders the dilute approximation most applicable to 3–0 and 3–1 [in the connectivity terminology of Newnham *et al.* (1978)] piezoelectric composites. In these composites, the matrix is connected to itself in three dimensions while the reinforcing phase is self-connected in zero and one dimensions respectively. For the dilute approximation, the concentration factors **A** and **B** are obtained from the solution of the auxiliary problem of a single particle embedded in an infinite matrix. For an ellipsoidal particle, this solution has been obtained by Deeg (1980) and recently reexamined by Dunn and Taya (1992) [see also the recent paper by Wang (1992) for an alternate derivation] and is briefly outlined in Appendix A. The concentration tensor, $\mathbf{A}^{\mathrm{dil}}$, is obtained as:

$$\mathbf{A}^{\mathrm{dil}} = [\mathbf{I} + \mathbf{S}\mathbf{E}_1^{-1}(\mathbf{E}_2 - \mathbf{E}_1)]^{-1}, \tag{23}$$

where **I** is the 9×9 identity matrix and **S** is the 9×9 matrix representation of the *constraint* tensors which are the piezoelectric analog of Eshelby's tensor (Dunn and Taya, 1992). The constraint tensors, **S**, are a function of the shape of the ellipsoid and the electroelastic moduli of the matrix. Explicit expressions for **S** are given in Appendix B. It is of interest to note the similarity of eqn (23) to the corresponding result in uncoupled elasticity (Benveniste, 1987) and electrostatics of dielectrics (Hatta and Taya, 1986). Equations (19) and (23) then yield explicit expressions for the dilute predictions of effective electroelastic moduli of piezoelectric composite materials. In the case of a continuous cylindrical fiber-reinforced piezoelectric composite these agree with the results of Wang (1992). By noting that $\mathbf{E}_1$ and

$\mathbf{E}_2$ are diagonally symmetric and following an argument identical to that forwarded by Benveniste *et al.* (1991), it can be shown that $\bar{\mathbf{E}}^{dil}$ possesses diagonal symmetry for matrix-based phase piezoelectric composites.

The self-consistent method

The self-consistent scheme was originally put forward by Bruggeman (1935) in the study of the conductivity of composite materials. The application of the method to the mechanical behavior of polycrystalline and matrix based composites followed some time later (Hershey, 1954; Kröner, 1958; Budiansky, 1965; Hill, 1965). The essential assumption employed in the self-consistent method is that each particle *sees* the effective medium of as yet unknown moduli. The concentration tensor $\mathbf{A}^{sc}$ is then given by:

$$\mathbf{A}^{sc} = [\mathbf{I}+\mathbf{S}^{sc}\mathbf{E}^{-1}(\mathbf{E}_2-\mathbf{E})]^{-1}. \tag{24}$$

Upon comparison with $\mathbf{A}^{dil}$ of eqn (24), $\mathbf{A}^{sc}$ seems simple enough. When coupled with eqn (19), though, it yields an implicit algebraic 9×9 matrix equation for $\mathbf{E}$. The situation is further complicated by the fact that $\mathbf{S}^{sc}$ is itself a function of the unknown electroelastic moduli, $\mathbf{E}$. Here the superscript "sc" on $\mathbf{S}$ is used to denote that the effective electroelastic moduli $\mathbf{E}$ are used in the evaluation of $\mathbf{S}$. In general, $\mathbf{S}$ itself must be evaluated by a two-dimensional numerical integration (Dunn and Taya, 1992) and thus the evaluation of $\mathbf{E}$ becomes cumbersome. Since $\mathbf{S}$ must in general be computed numerically, $\mathbf{E}$ can be determined without *a priori* knowledge of the symmery of the effective electroelastic moduli as the material symmetry has no significant effect on the *numerical* computation of $\mathbf{S}$.

The Mori–Tanaka mean field approach

The original work of Mori and Tanaka (1973) was concerned with estimating the average internal stress in a matrix material containing precipitates with eigenstrains (transformation strains). Since then, the method has been successfully applied to many problems in the mechanics and physics of composite materials. The method was initially linked with Eshelby's (1957) equivalent inclusion method and a review of many applications in this context is given by Taya and Arsenault (1989). Recently, Benveniste (1987) re-examined the underlying assumptions of the method and reformulated it in a *direct approach*. The method has also received considerable attention from a theoretical standpoint (Weng, 1990, 1992; Benveniste *et al.*, 1991; Ferrari, 1991) and has been shown to be on strong theoretical footing for the elastic behavior of two-phase composite media. In fact, Weng (1992) showed that for a two-phase elastic composite containing ellipsoidal inclusions, the Mori–Tanaka effective moduli coincide with Willis' (1977) lower (upper) bound when the matrix is the softest (hardest) phase. More recently, Dunn and Taya (1992) applied the theory through an equivalent inclusion approach (see Appendix A) to model the coupled electroelastic behavior of piezoelectric composites. They also showed that the theory is on strong theoretical footing for two-phase piezoelectric composites with aligned inclusions. Here, the direct approach of Benveniste (1987) is utilized to obtain the concentration factor, $\mathbf{A}$, as the resulting expressions are equivalent to the original formulation of Dunn and Taya (1992).

The key assumption in the Mori–Tanaka (1973) theory is that $\mathbf{A}$ is given by the solution for a single particle embedded in an infinite matrix subjected to an applied electroelastic field equal to the as yet unknown average electroelastic field in the matrix. This solution is easily expressed as:

$$\bar{\mathbf{Z}}_2 = \mathbf{A}^{dil}\bar{\mathbf{Z}}_1, \tag{25}$$

where $\mathbf{A}^{dil}$ is given by eqn (23).

With eqns (16), (20) and (25), the concentration factor, $\mathbf{A}^{MT}$, can be written in the form as first proposed by Benveniste (1987) for elastic composites:

$$\mathbf{A}^{\mathrm{MT}} = \mathbf{A}^{\mathrm{dil}}[c_1\mathbf{I}+c_2\mathbf{A}^{\mathrm{dil}}]^{-1}. \tag{26}$$

Equations (19), (23) and (26) then provide explicit expressions for the effective electroelastic moduli of piezoelectric composites. It can be easily shown that the companion solution for the electroelastic moduli, $\mathbf{F}^{\mathrm{MT}}$ is self-consistent in that $\mathbf{F}^{\mathrm{MT}} = \mathbf{E}^{\mathrm{MT}^{-1}}$. This has been proven by Dunn and Taya (1992) who have also shown that $\mathbf{E}^{\mathrm{MT}}$ possesses diagonal symmetry and the correct limiting behavior in the low and high concentration limits for two-phase piezoelectric composites.

The differential scheme

The differential scheme has been utilized by a number of researchers to predict the effective moduli of elastic and electric composite media. The history of the method and its application to elastic composites is detailed by McLaughlin (1977) and further references can be found therein. The essence of the method is the realizable construction of the final composite from the matrix material through the successive replacement of an incremental volume of the current composite with that of the reinforcement. The method has been recently extended by Norris (1985) to allow the incremental addition of *both* phases to the current composite. The resulting effective elastic moduli are then dependent on the replacement sequence leading to the final configuration. Here the special case when one phase is incrementally added to the current material configuration is extended to piezoelectric composite media.

Following McLaughlin (1977), the removal of a volume increment ΔV of the instantaneous configuration (thus a removal of $c_2\Delta V$ of the reinforcing phase) leads to:

$$\mathrm{d}c_2 = \frac{\mathrm{d}V}{V}(1-c_2), \tag{27}$$

where V is the volume of the composite. Denoting $\mathbf{E}(c_2+\mathrm{d}c_2)$ as the effective electroelastic moduli at a reinforcement volume fraction of $c_2+\mathrm{d}c_2$, use of eqns (19) and (27) leads to:

$$\frac{\mathrm{d}\mathbf{E}}{\mathrm{d}c_2} = \frac{1}{1-c_2}(\mathbf{E}_2-\mathbf{E})\mathbf{A}^{\mathrm{dif}}, \tag{28}$$

where

$$\mathbf{A}^{\mathrm{dif}} = [\mathbf{I}+\mathbf{S}^{\mathrm{dif}}\mathbf{E}^{-1}(\mathbf{E}_2-\mathbf{E})]^{-1}. \tag{29}$$

As in the self-consistent scheme, $\mathbf{S}^{\mathrm{dif}}$ is a function of the electroelastic moduli, $\mathbf{E}$, of the instantaneous material. Formally, eqn (28) represents a set of $9\times 9 = 81$ coupled nonlinear ordinary differential equations which can be solved with the initial conditions:

$$\mathbf{E}(c_2 = 0) = \mathbf{E}_1. \tag{30}$$

Again the work of Benveniste *et al.* (1991) can be easily generalized to show that $\mathbf{E}$ predicted by the differential scheme is diagonally symmetric for two-phase matrix based piezoelectric composites.

5. RESULTS AND DISCUSSION

In the preceding section, each of the models has been developed for two-phase matrix based piezoelectric composites. Strictly speaking, they are most applicable to 3–0 and 3–1 composites where the matrix is self-connected in three dimensions and the reinforcement is self-connected in zero and one dimensions respectively. By modeling the reinforcement as ellipsoidal, though, a 2–2 composite can be simulated as the ellipsoid degenerates to a flat lamina. It appears that the differential scheme could be easily extended to treat both phases on equal footing by proceeding along the lines of Norris (1985) in which both phases

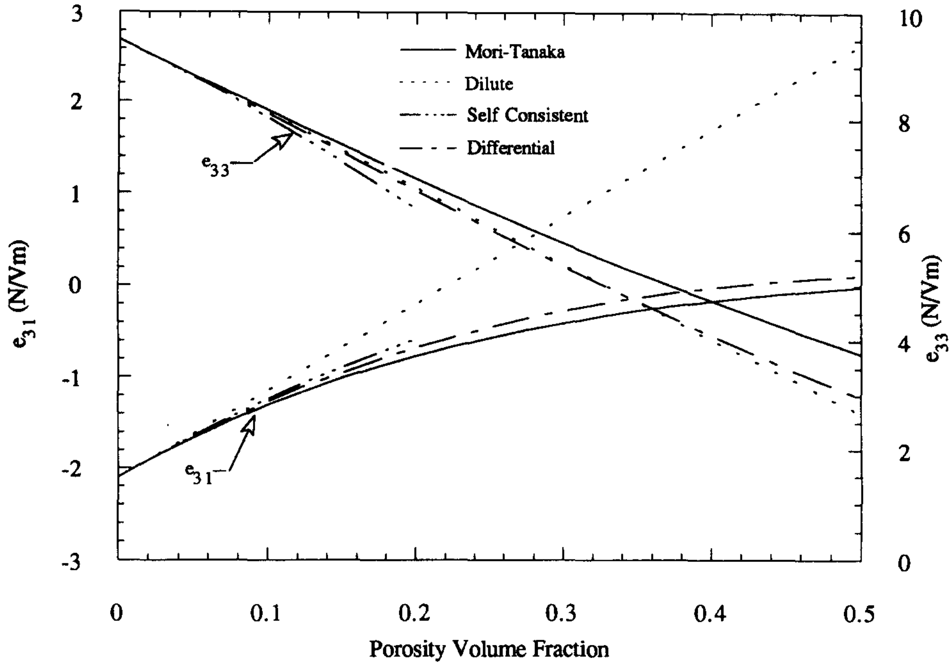

Fig. 1. Effective piezoelectric moduli ($e_{3i} = E_{9i}$) for a porous PZT-7A ceramic as a function of porosity volume fraction. The porosity is assumed to be spherical.

are incrementally added to the composite. As noted by Norris, though, the resulting moduli are then dependent on the *path* of the incremental additions which is of some theoretical concern. The self consistent scheme has traditionally received criticism for yielding unacceptable results when the difference in moduli of the constituents is large. This, unfortunately, is the case for many piezoelectric composites where the polymer phase is piezoelectrically inactive.

It is also of interest to further comment on the form of the analytical predictions yielded by each micromechanics model. The dilute and Mori–Tanaka methods yield explicit results for the moduli. Only a single numerical evaluation of $\mathbf{S}^{dil} = \mathbf{S}^{MT}$ is then required (except of course in the case where $\mathbf{S}$ can be obtained analytically as in Appendix C). The self-consistent method on the other hand yields an implicit algebraic matrix (9×9) equation for the effective moduli, resulting in recourse to a numerical scheme for their evaluation. This becomes inconvenient as in each iteration $\mathbf{S}^{sc}$ must generally be computed numerically. For the examples considered here, simple fixed point iteration was successful but the requirement of up to 25 iterations for convergence was not uncommon for various degrees of material symmetry and microsructural geometry. A numerical method such as the fourth order Runge–Kutta integration scheme must in general be used with the differential method which yields a set of ($9 \times 9 = 81$) coupled nonlinear ordinary differential equations for the effective moduli. Again, this becomes inconvenient as during each increment $\mathbf{S}^{dif}$ must generally be computed numerically. Thus from a computational standpoint, the dilute and Mori–Tanaka schemes are certainly advantageous.

Finally, the performance of each model for some typical piezoelectric composites is illustrated and the micromechanics predictions are compared to existing experimental data for fiber and particle reinforced piezoelectric composites. In Fig. 1, the piezoelectric moduli e_{33} and e_{31} [see eqn (10)] are shown as a function of porosity for a porous PZT-7A ceramic. The electroelastic moduli of the PZT-7A, and those of PZT-5 used in subsequent computations, were obtained from Berlincourt (1971) and Chan and Unsworth (1989) and are given in Table 1 where the well-known two index notation has been adopted.

In Fig. 1 it is seen that the predictions of the dilute, Mori–Tanaka and differential schemes are fairly close for e_{33} but the dilute predictions far exceed the Mori–Tanaka

Table 1. Electroelastic material properties

	C_{11} (GPa)	C_{12} (GPa)	C_{13} (GPa)	C_{33} (GPa)	C_{44} (GPa)	e_{31} (C/m^2)	e_{33} (C/m^2)	e_{15} (C/m^2)	κ_{11}† / κ_0	κ_{33}† / κ_0
PZT-7A	148	76.2	74.2	131	25.4	−2.1	9.5	9.2	460	235
PZT-5	121	75.4	75.2	111	21.1	−5.4	15.8	12.3	916	830
Epoxy	8.0	4.4	4.4	8.0	1.8	0	0	0	4.2	4.2
Polymer	3.86	2.57	2.57	3.86	0.64	0	0	0	9.0	9.0

† κ_0 = 8.85 E-12 (C^2/Nm^2) = permittivity of free space.

and differential predictions for e_{31}. The self-consistent predictions, however, are in close agreement at lower volume fractions but are not plotted past $f \approx 0.2$ as at this point other electroelastic moduli vanish. This behavior is attributed to the fact that the porosity is of vanishing stiffness and electric activity and is consistent with the behavior in the uncoupled elastic case. In particular, it is similar to the self-consistent predictions of Norris (1990) for the effective Poisson's ratio of composite reinforced randomly oriented rigid disks. In Fig. 2 similar analytical predictions for e_{31} and e_{33} are given for a composite reinforced by short (aspect ratio = 10) PZT-7A fibers in an epoxy matrix. It is of interest that even though the piezoelectric moduli of the epoxy vanish, the self-consistent estimates are in line with those of the other models and certainly seem reasonable. This suggests that it is the vanishing components of the noncoupling terms (elastic moduli and dielectric constant) which lead to the unacceptable behavior of the self-consistent scheme. In Fig. 3, the Mori–Tanaka micromechanics predictions are shown for the piezoelectric strain coefficients d_{33} and $d_h = d_{31}+d_{32}+d_{33}$ of a composite comprised of a PZT-5 matrix and elliptical soft polymer reinforcement with the generatrix of the elliptical reinforcement aligned with the x_3 axis. The strain coefficients d_{3i} are obtained from the micromechanics predictions as $-F_{9i}/F_{99}$. Three values of the aspect ratio, $\alpha = a_2/a_1$ are considered which correspond to circular fiber ($\alpha = 1$), thin ribbon ($\alpha = 10$) and lamina ($\alpha \to \infty$) reinforcement. The strain coefficient d_{33} is seen to be insensitive to the in-plane shape of the reinforcement. The hydrostatic strain coefficient, d_h, however, varies significantly with the

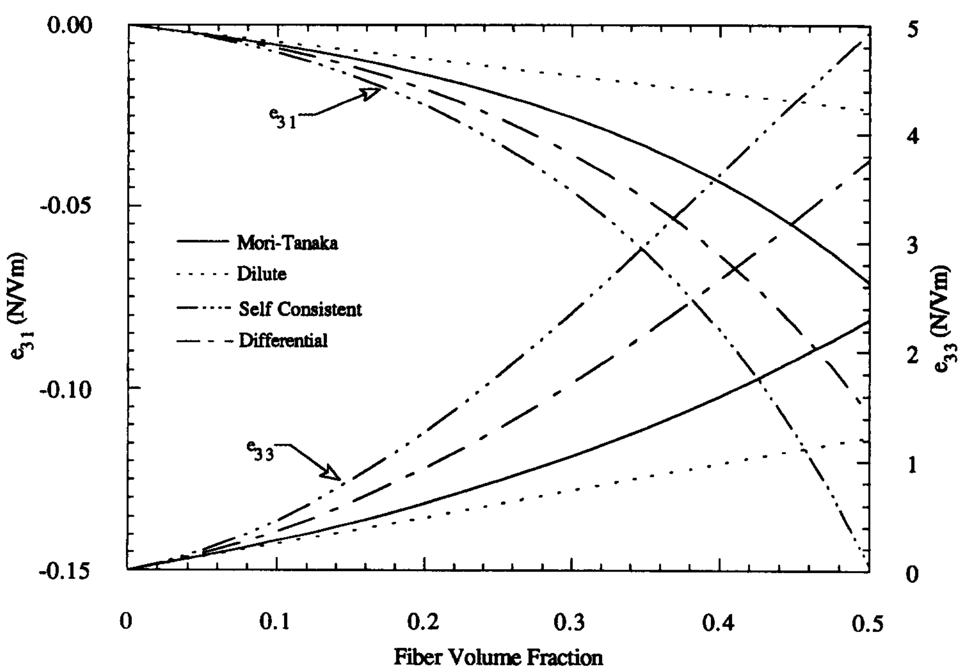

Fig. 2. Effective piezoelectric moduli ($e_{3i} = E_{9i}$) of a PZT-7A short fiber ($a_3/a_1 = 10$) reinforced epoxy composite as a function of fiber volume fraction.

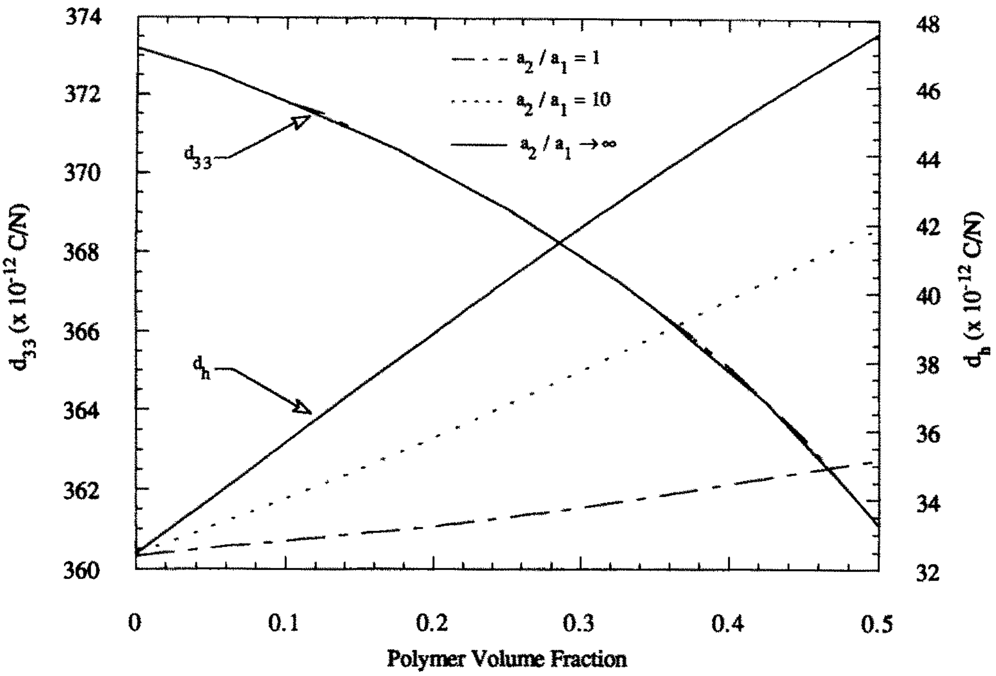

Fig. 3. Effective piezoelectric moduli ($d_{3i} = -F_{9i}/F_{99}$, $d_h = d_{31}+d_{32}+d_{33}$) of a PZT-5 ceramic reinforced by a continuous ($a_3 \rightarrow \infty$) polymer phase as a function of fiber volume fraction and the in-plane reinforcement aspect ratio, a_2/a_1.

reinforcement shape. This is attributed to the increased anisotropic electroelastic interaction resulting from the ribbon and lamina reinforcement. It is noted that when the elliptical inclusion degenerates to a two-dimensional lamina ($\alpha \rightarrow \infty$), the results for d_{33} agree with the parallel model solutions of Newnham *et al.* (1978) and Grekov *et al.* (1987).

Finally, it is of interest to compare the analytical predictions with existing experimental results for the effective electroelastic moduli of piezoelectric composites. To this end, two reinforcement geometries are considered: continuous fiber reinforcement (Chan and Unsworth, 1989) and spherical particle reinforcement (Furukawa *et al.*, 1976). In Fig. 4 it is seen that the analytical predictions of d_{33} by the micromechanics models are virtually indistinguishable for a continuous PZT-7A fiber reinforced epoxy composite. The material properties in Table 1 were used for the computations with the exception that instead of the tabulated value of Berlincourt (1971), the measured value of d_{33} of PZT-7A obtained by Chan and Unsworth (1989) was used. The micromechanics predictions are in excellent agreement with the experimental results of Chan and Unsworth (1989) for the continuous PZT-7A fiber reinforced epoxy composite. Although not shown, it is noted that the models of Grekov *et al.* (1989) and Smith and Auld (1991) are in very close agreement with the four micromechanics models. Finally, in Fig. 5 analytical predictions are compared to experimental results obtained by Furukawa *et al.* (1976) for an epoxy matrix reinforced by piezoelectric particles. The material properties used in the computations were those of PZT-5 as taken from Furukawa *et al.* (1976) and Berlincourt (1971). Also shown are the predictions of the cubes models of Banno (1983). There is seen to be a significant difference in the predictions of the models with each resulting in good agreement in the low volume fraction range. At a volume fraction of approximately 25%, the Mori–Tanaka and dilute models are in the best agreement with the experimental results. It is at about this point, however, that the Mori–Tanaka and dilute predictions begin to significantly diverge and it is expected that the Mori–Tanaka estimates will be better than the dilute estimates at higher volume fractions. The Mori–Tanaka and differential schemes are seen to be in relatively close agreement for lower volume fractions but begin to differ appreciably at volume fractions of the order of 20%. In light of the existing experimental data, though, it cannot be ascertained which prediction will be better at higher volume fractions.

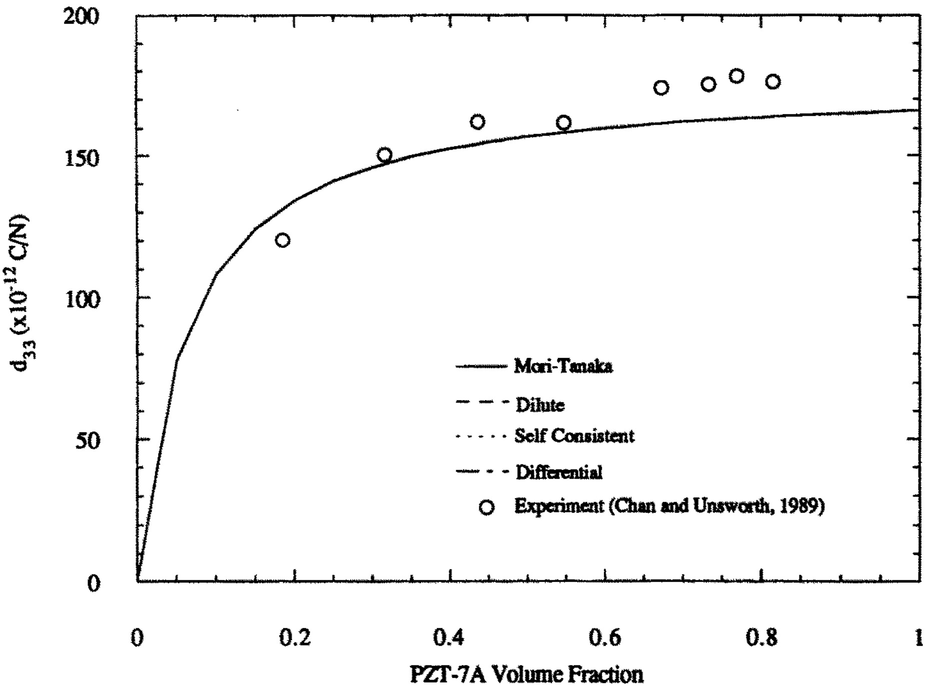

Fig. 4. Comparison of the micromechanics predictions and experimental results (Chan and Unsworth, 1989) for the effective piezoelectric moduli ($d_{33} = -F_{93}/F_{99}$) of a PZT-7A continuous fiber ($a_3/a_1 \rightarrow \infty$) reinforced epoxy composite as a function of fiber volume fraction. The micromechanics predictions are indistinguishable.

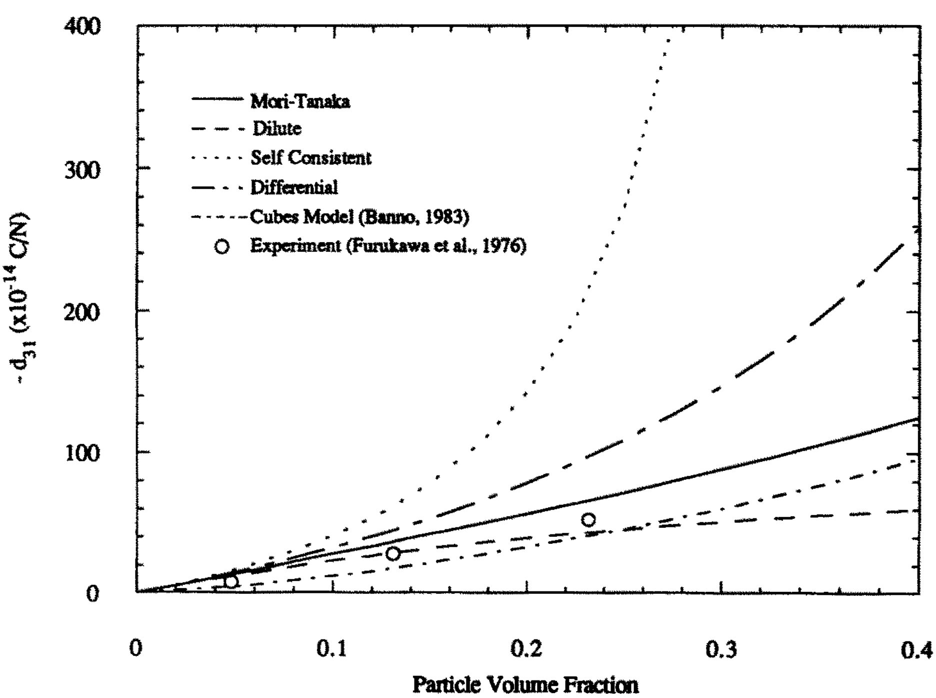

Fig. 5. Comparison of the micromechanics predictions and experimental results (Furukawa *et al.*, 1976) for the effective piezoelectric moduli ($d_{31} = -F_{91}/F_{99}$) of a PZT-5 particle ($a_3/a_1 = 1$) reinforced polymer composite as a function of particle volume fraction.

6. CONCLUSION

The rigorous analytical solution for the coupled electroelastic fields in a piezoelectric inclusion (Deeg, 1980; Dunn and Taya, 1992) has been implemented in four micromechanics models to determine the effective electroelastic moduli of a two-phase piezoelectric composite. As in the uncoupled behavior, the dilute and Mori–Tanaka schemes return explicit expressions for the effective electroelastic moduli. The self-consistent scheme, on the other hand, returns an implicit algebraic matrix equation for the effective electroelastic moduli. Application of the differential scheme formally results in a 9×9 system of coupled nonlinear ordinary differential equations for effective electroelastic moduli. The matrix formulation that is presented results in ease of implementation and generation of numerical results. The only complicating factor is that **S** must in general be evaluated numerically. The resulting matrix equations for the electroelastic moduli are completely analogous to those for the elastic moduli of two-phase composites.

Numerical results have been presented to compare the predictions of each model for a number of typical composite microstructures including a porous phase, continuous and short fiber, particle, and lamina reinforcement. When the elastic *and* piezoelectric moduli of the reinforcing phase vanish, the self-consistent scheme does not return physically acceptable results over the entire range of volume fractions. This is consistent with the corresponding self-consistent predictions for the elastic moduli of a porous anisotropic elastic solid. For the examples considered here, however, the self consistent predictions seem reasonable when *only* the piezoelectric moduli of the reinforcing phase vanish. In the case of continuous fiber reinforcement, the micromechanics predictions are indistinguishable and agree well with experimental results. The micromechanics predictions are also in good agreement with the experimental results of a piezoelectric particle reinforced polymer composite. In light of its ease of application and its good agreement with the experimental data, a strong argument can be made for the use of the Mori–Tanaka micromechanics scheme in the prediction of the electroelastic moduli of piezoelectric composite materials.

Finally, a few comments regarding items that were not considered are in order. Any prediction of the effective moduli of composite media should be judged with available bounded solutions. To this end, in uncoupled behavior, the Mori–Tanaka scheme has been shown to coincide with the upper and lower bounds (depending on which phase is the *stiffest*) for both elastic and electric composites reinforced with ellipsoidal inclusions (Weng, 1992; Hatta and Taya, 1986). Whether this is the case with regards to piezoelectric media remains an open issue. In fact, to the authors' knowledge, rigorous bounds have yet to be developed for the effective moduli of piezoelectric composites.

REFERENCES

Aboudi, J. (1991). *Mechanics of Composite Materials: A Unified Micromechanical Approach.* Elsevier, The Netherlands.
Banno, H. (1983). Recent developments of piezoelectric ceramic products and composites of synthetic rubber and piezoelectric ceramic particles. *Ferroelectrics* **50,** 3–12.
Barnett, D. M. and Lothe, J. (1975). Dislocations and line charges in anisotropic piezoelectric insulators, *Phys. stat. sol.* (*b*) **67,** 105–111.
Benveniste, Y. (1987). A new approach to the application of Mori–Tanaka's theory in composite materials. *Mech. Mater.* **6,** 147–157.
Benveniste, Y., Dvorak, G. J. and Chen, T. (1991). On diagonal symmetry of the approximate effective stiffness tensor of heterogeneous media. *J. Mech. Phys. Solids* **39,** 927–946.
Berlincourt, D. A. (1971). Piezoelectric crystals and ceramics. In *Ultrasonic Transducer Materials* (Edited by O. E. Mattiat), pp. 63–119.
Bruggeman, D. A. (1935). Berechnung verschiedener physikalischer Konstanten von heterogen Substanzen. I. Dielektrizitätskonstanten und Leitfahigkeiten der Mischkörper aus isotropen Substanzen. *Annalen der Physik* (*Leipzig*) **24,** 636.
Budiansky, B. (1965). On the elastic moduli of some heterogeneous materials. *J. Mech. Phys. Solids* **13,** 223–227.
Chan, H. L. W. and Unsworth, J. (1989). Simple model for piezoelectric ceramic/polymer 1–3 composites used in ultrasonic transducer applications. *IEEE Trans. Ultrason. Ferroelectrics and Frequency Control* **36,** 434–441.
Deeg, W. F. (1980). The analysis of dislocation, crack, and inclusion problems in piezoelectric solids. Ph.D Dissertation, Stanford University.

Dunn, M. and Taya, M. (1992). An equivalent inclusion formulation for piezoelectric composite materials. (Submitted for publication.)

Eshelby, J. D. (1957). The determination of the elastic field of an ellipsoidal inclusion, and related problems. *Proc. R. Soc. Lon.* **A241,** 376–396.

Ferrari, M. (1991). Asymmetry and the high concentration limit of the Mori–Tanaka effective medium theory. *Mech. Mater.* **11,** 251–256.

Furukawa, T., Fujino, K. and Fukada, E. (1976). Electromechanical properties in the composites of epoxy resin and PZT ceramics. *Jap. J. Appl. Phys.* **15,** 2119–2129.

Gavazzi, A. C. and Lagoudas, D. C. (1990). On the numerical evaluation of Eshelby's tensor and its application to elastoplastic fibrous composites. *Computl Mech.* **7,** 13–19.

Grekov, A. A., Kramarov, S. O. and Kuprienko, A. A. (1987). Anomalous behavior of the two-phase lamellar piezoelectric texture. *Ferroelectrics* **76,** 43–48.

Grekov, A. A., Kramarov, S. O. and Kuprienko, A. A. (1989). Effective properties of a transversely isotropic piezocomposite with cylindrical inclusions. *Ferroelectrics* **99,** 115–126.

Hatta, H. and Taya, M. (1986). Equivalent inclusion method for steady state heat conduction in composites. *Int. J. Engng Sci.* **24,** 1159–1172.

Hershev, A. V. (1954). The elasticity of an isotropic aggregate of anisotropic cubic crystals. *J. Appl. Mech.* **21,** 236–241.

Hill, R. (1965). A self-consistent mechanics of composite materials. *J. Mech. Phys. Solids* **13,** 213–222.

Ikeda, T. (1990). *Fundamentals of Piezoelectricity.* Oxford University Press, Oxford.

Kröner, E. (1958). Berechnung der elastischen konstanten des vielkristalls aus den konstanten des einkristalls. *Z. Phys.* **151,** 504–518.

McLaughlin, R. (1977). A study of the differential scheme for composite materials. *Int. J. Engng Sci.* **15,** 237–244.

Mori, T. and Tanaka, K. (1973). Average stress in matrix and average elastic energy of materials with misfitting inclusions. *Acta Metall.* **21,** 571–574.

Mura, T. (1987). *Micromechanics of Defects in Solids* (2nd Edn). Martinus Nijhoff, Dordrecht.

Newnham, R. E., Skinner, D. P. and Cross, L. E. (1978). Connectivity and piezoelectric-pyroelectric composites. *Mater. Res. Bull.* **13,** 525–536.

Norris, A. N. (1985). A differential scheme for the effective moduli of composites. *Mech. Mater.* **4,** 1–16.

Norris, A. N. (1990). The mechanical properties of platelet reinforced composites. *Int. J. Solids Structures* **26,** 663–674.

Reuss, A. (1929). Berechnung der fliessgrenze von mischkristallen auf grund der plastizitätsbedingung für einkristalle. *Z. angew. Math. Mech.* **9,** 49–58.

Smith, W. A. and Auld, B. A. (1991). Modeling 1–3 composite piezoelectrics: thickness-mode oscillations. *IEEE Transactions on Ultrasonics, Ferroelectrics and Frequency Control* **38,** 40–47.

Taya, M. and Arsenault, R. J. (1989). *Metal Matrix Composites, Thermomechanical Behavior.* Pergamon Press, Oxford.

Voigt, W. (1889). Über die Beziehung zwischen den beiden Elastizitätskonstanten isotroper Körper. *Weid. Ann.* **38,** 573–587.

Wang, B. (1992). Three-dimensional analysis of an ellipsoidal inclusion in a piezoelectric material. *Int. J. Solids Structures* **29,** 293–308.

Weng, G. (1990). The theoretical connection between Mori–Tanaka's theory and the Hashin–Shtrikman–Walpole bounds. *Int. J. Engng Sci.* **28,** 1111–1120.

Weng, G. J. (1992). Explicit evaluation of Willis' bounds with ellipsoidal inclusions. *Int. J. Engng Sci.* **30,** 83–92.

Zhou, S. (1991). A material multipole theory of elastic dielectric composites. *Int. J. Solids Structures* **28,** 423–447.

APPENDIX A. CONCENTRATION MATRIX, A

Consider a piezoelectric composite consisting of an infinite domain (D) containing an ellipsoidal particle (inhomogeneity). The ellipsoidal particle is of the same shape and aligned with the a_3 principal axis coincident with the x_3 axis. The piezoelectric particle (Ω) has electroelastic moduli $\mathbf{E}_2$ while the matrix, D-Ω, has electroelastic moduli $\mathbf{E}_1$. The composite is subjected to the far-field applied stress and electric displacement $\mathbf{\Sigma}^0$. Using the equivalent inclusion method for piezoelectric composites (Deeg, 1980; Dunn and Taya, 1992) the stress and electric displacement in the representative particle can be written as:

$$\begin{aligned}\mathbf{\Sigma} &= \mathbf{E}_2[\mathbf{Z}^0+\mathbf{Z}]\\ &= \mathbf{E}_1[\mathbf{Z}^0+\mathbf{Z}-\mathbf{Z}^*],\end{aligned} \tag{A1}$$

where $\mathbf{Z}$ represents the *perturbation* of the strain and electric fields in the particle with respect to those in the matrix and $\mathbf{Z}^*$ is the fictitious eigenfield required to ensure that the equivalency of eqn (A1) holds. In eqn (A1), $\mathbf{Z}$ and $\mathbf{Z}^*$ are related through:

$$\mathbf{Z} = \mathbf{S}\mathbf{Z}^*, \tag{A2}$$

where S is the coupled electroelastic analog of Eshelby's tensor and is defined explicitly in Appendix B. The strain and potential gradient in the inclusion (which are uniform) are given by:

$$\mathbf{Z}_2 = \mathbf{Z}^0 + \mathbf{S}\mathbf{Z}^*. \tag{A3}$$

With eqns (A1) and (A3), $\mathbf{Z}_2$ can be obtained as:

$$[\mathbf{I} + \mathbf{S}\mathbf{E}_1^{-1}(\mathbf{E}_2 - \mathbf{E}_1)]\mathbf{Z}_2 = \mathbf{Z}^0. \tag{A4}$$

From eqns (A4) and (20), $\mathbf{A}^{\text{dil}}$ is seen to be given by:

$$\mathbf{A}^{\text{dil}} = [\mathbf{I} + \mathbf{S}\mathbf{E}_1^{-1}(\mathbf{E}_2 - \mathbf{E}_1)]^{-1}. \tag{A5}$$

The stress and electric displacement concentration factor, $\mathbf{B}^{\text{dil}}$, can be easily obtained in an analogous manner.

APPENDIX B. CONSTRAINT TENSORS, S_{MnAb}

The constraint tensors, S_{MnAb}, can be expressed in terms of surface integrals over the unit sphere as (Dunn and Taya, 1992):

$$S_{MnAb} = \begin{cases} \dfrac{a_1 a_2 a_3}{8\pi} E_{iJAb} \displaystyle\int_{|\mathbf{z}|=1} \dfrac{1}{\zeta^3} [G_{mJin}(\mathbf{z}) + G_{nJim}(\mathbf{z})]\, \mathrm{d}S(\mathbf{z}), & M = 1, 2, 3, \\ \dfrac{a_1 a_2 a_3}{4\pi} E_{iJAb} \displaystyle\int_{|\mathbf{z}|=1} \dfrac{1}{\zeta^3} G_{4Jin}(\mathbf{z})\, \mathrm{d}S(\mathbf{z}), & M = 4, \end{cases} \tag{B1}$$

where $|\mathbf{z}| = 1$ is the surface of the unit sphere $\zeta = [a_1^2 z_1^2 + a_2^2 z_2^2 + a_3^2 z_3^2]^{1/2}$, and:

$$G_{MJin}(\mathbf{z}) = z_i z_n K_{MJ}^{-1}(\mathbf{z}), \tag{B2}$$

and $K_{MJ} = E_{iJMn} z_i z_n$. To perform the integration of eqns (B1), the unit sphere is parameterized as (Dunn and Taya, 1992):

$$z_1 = \frac{\xi_1}{a_1} = \frac{(1-\xi_3^2)^{1/2}\cos\theta}{a_1}, \quad z_2 = \frac{\xi_2}{a_2} = \frac{(1-\xi_3^2)^{1/2}\sin\theta}{a_2}, \quad z_3 = \frac{\xi_3}{a_3}. \tag{B3}$$

In general, for an anisotropic medium the integrals in eqns (B1) cannot be evaluated analytically. In these cases, the integration is easily performed by Gaussian quadrature. Following the work of Gavazzi and Lagoudas (1990) for elastic inclusions in an anisotropic elastic matrix, a typical term is evaluated as:

$$S_{mnab} = \frac{1}{8\pi} \sum_{p=1}^{M} \sum_{q=1}^{N} \{C_{ijab}[G_{mjin}(\theta_q, \xi_{3p}) + G_{njim}(\theta_q, \xi_{3p})] - e_{iab}[G_{m4in}(\theta_q, \xi_{3p}) + G_{n4im}(\theta_q, \xi_{3p})]\} W_{pq}, \tag{B4}$$

where p and q are the integration points over the unit sphere parameterized by ξ_3 and θ, respectively, and W_{pq} are the Gaussian weights.

APPENDIX C. CONSTRAINT TENSORS, S_{MnAb}, FOR ELLIPTICAL INCLUSIONS

For a two-dimensional elliptical inclusion ($a_3 \to \infty$, $\alpha = a_2/a_1$) in a transversely isotropic matrix, the nonzero components of the constraint tensors are explicitly obtained as:

$$S_{1111} = \frac{\alpha}{2(\alpha+1)^2}\left[\frac{2C_{11}+C_{12}}{C_{11}} + (2\alpha+1)\right],$$

$$S_{1212} = S_{2121} = S_{1221} = S_{2112} = \frac{\alpha}{2(\alpha+1)^2}\left[\frac{\alpha^2+\alpha+1}{\alpha} - \frac{C_{12}}{C_{11}}\right],$$

$$S_{1313} = S_{3131} = S_{1331} = S_{3113} = \frac{\alpha}{2(\alpha+1)},$$

$$S_{1122} = \frac{\alpha}{2(\alpha+1)^2}\left[\frac{(2\alpha+1)C_{12} - C_{11}}{C_{11}}\right],$$

$$S_{1133} = \frac{C_{13}}{C_{11}}\frac{\alpha}{\alpha+1},$$

$$S_{1143} = \frac{e_{31}}{C_{11}}\frac{\alpha}{\alpha+1},$$

$$S_{2211} = \frac{\alpha}{2(\alpha+1)^2}\left[\frac{(\alpha+2)C_{12} - C_{11}}{C_{11}}\right],$$

$$S_{2222} = \frac{\alpha}{2(\alpha+1)^2}\left[\frac{2C_{11}+C_{12}}{C_{11}} + \frac{\alpha+2}{\alpha}\right],$$

$$S_{2323} = S_{3232} = S_{2332} = S_{3223} = \frac{1}{2(\alpha+1)},$$

$$S_{2233} = \frac{C_{13}}{C_{11}}\frac{1}{\alpha+1},$$

$$S_{2243} = \frac{e_{31}}{C_{11}}\frac{1}{\alpha+1},$$

$$S_{4141} = \frac{\alpha}{\alpha+1},$$

$$S_{4242} = \frac{1}{\alpha+1}. \tag{C1}$$

For a circular cylindrical inclusion ($\alpha = 1$), the nonzero components of the constraint tensors reduce to

$$S_{1111} = S_{2222} = \frac{5C_{11}+C_{12}}{8C_{11}},$$

$$S_{1212} = S_{2121} = S_{1221} = S_{2112} = \frac{3C_{11}-C_{12}}{8C_{11}},$$

$$S_{1313} = S_{3131} = S_{1331} = S_{3113} = S_{2323} = S_{3232} = S_{2332} = S_{3223} = \tfrac{1}{4},$$

$$S_{1122} = S_{2211} = \frac{3C_{12}-C_{11}}{8C_{11}},$$

$$S_{1133} = S_{2233} = \frac{C_{13}}{2C_{11}},$$

$$S_{1143} = S_{2243} = \frac{e_{31}}{2C_{11}},$$

$$S_{4141} = S_{4242} = \tfrac{1}{2}. \tag{C2}$$

For a ribbon-like inclusion ($a_2 \gg a_1$), the nonzero components of the constraint tensors reduce to:

$$S_{1111} = \frac{3C_{11}+C_{12}}{2C_{11}}\alpha,$$

$$S_{1212} = S_{2121} = S_{1221} = S_{2112} = \frac{1}{2} - \frac{C_{11}+C_{12}}{2C_{11}}\alpha,$$

$$S_{1313} = S_{3131} = S_{1331} = S_{3113} = \frac{\alpha}{2},$$

$$S_{1122} = -\frac{C_{11}-C_{12}}{2C_{11}}\alpha,$$

$$S_{1133} = \frac{C_{13}}{C_{11}}\alpha,$$

$$S_{1143} = \frac{e_{31}}{C_{11}}\alpha,$$

$$S_{2211} = \frac{2C_{12}-C_{11}}{2C_{11}}\alpha,$$

$$S_{2222} = 1 - \frac{C_{11}-C_{12}}{2C_{11}}\alpha,$$

$$S_{2323} = S_{3232} = S_{2332} = S_{3223} = \frac{1-\alpha}{2},$$

$$S_{2233} = \frac{C_{13}}{C_{11}}(1-\alpha),$$

$$S_{2243} = \frac{e_{31}}{C_{11}}(1-\alpha),$$

$$S_{4141} = \alpha,$$

$$S_{4242} = 1-\alpha. \tag{C3}$$

The constraint tensors are represented in a 9×9 matrix format as prescribed by eqns (13) and (14). It is noted, however, that in general **S** is not diagonally symmetric and that care must be taken in the matrix representation of **S** to correctly account for the factor of two with regards to the shear strains.