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ON THE EFFECTIVE ELECTROELASTIC PROPERTIES OF PIEZOELECTRIC COMPOSITES CONTAINING SPATIALLY ORIENTED INCLUSIONS

WEN-SHYONG KUO Department of Textile Engineering, Feng Chia University, Taichung 407, Taiwan, Republic of China

and

JIN H. HUANG

Department of Mechanical Engineering, Feng Chia University, Taichung 407, Taiwan, Republic of China

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Abstract—Presented here is an analytical method for determining the effective electroelastic properties of piezoelectric composites containing spatially oriented inclusions. Both inclusion and matrix are assumed to be linearly piezoelastic. The three-dimensional anisotropic inclusion method has been developed, and the explicit expressions for the electrostatic tensors analogous to the Eshelby tensors for elastic ellipsoidal inclusions have been obtained. With these tensors, analytical expressions for the electroelastic fields of the piezoelectric composite can be established. Based upon the Mori-Tanaka mean field theory to account for the interaction between the inclusions and matrix, the effective electroelastic properties (elastic, piezoelectric and dielectric constants) of the composites are expressed analytically in terms of phase properties, orientation angles, volume fraction and shape. Numerical results have been given for a BaTiO₃/PZT-5H composite. The results indicate that the proposed methods yield identical results whether a traction-electric displacement or an elastic displacement-electric field is prescribed on the composite boundary is self-consistency. The electromechanical coupling due to the piezoelastic effect is found to be strongly affected by both the volume fraction and the orientation of inclusions. (C) 1997 Elsevier Science Ltd.

1. INTRODUCTION

Combining two or more distinct constituents, piezoelectric composite materials can take the advantages of each constituent and have superior electromechanical coupling characteristics to monolithic piezoelectric materials. These materials have been developed in many forms including secondary-phase piezoelectric ceramic inclusions embedded in a polymer matrix and polymer filled secondary-phase piezoelectric inclusions in a solid piezoelectric ceramic matrix. The secondary-phase piezoelectric inclusions in the matrix of composites could be continuous fibers, short fibers, holes, voids, or dispersed particles. More recently, piezoelectric composites are extensively used as transducers for sonar projector, underwater and medical ultrasonic imaging applications. This may explain why piezoelectric composite materials constitute an important branch of the recently emerging technologies of modern engineering materials.

The introduction of inclusions into base media leads to the material being anisotropic, complicated and, in some situations, even detrimental to the performance of the composite. Most models are based on the assumption that the crystalline directions of the matrix coincide with the principal axes of an inclusion. Thus, the effect of the derivation of the inclusion axis from the crystal axis merits further investigation. That is, the electroelastic response must be clearly analyzed so that the influence of material parameters and orientation angles can be understood.

In the study of the overall behavior of a composite material containing high concentrations of inclusions, the Mori-Tanaka mean field theory is generally believed to be one of the most powerful methods. Based upon the equivalent inclusion method (Eshelby, 1957), Mori and Tanaka (1973) first introduced the concept of average stress in a matrix material containing precipitates with eigenstrain (stress-free tranformation strain). Since then, this theory has been successfully applied to obtain elastic behavior and effective properties of composite materials (see, for example, Taya and Mura, 1981; Mura, 1987; Benveniste, 1987; Mori and Wakashima, 1990; Weng, 1990, 1992; Huang *et al.*, 1993; Dunn and Taya, 1993; Benveniste, 1993a, b, 1994 among others). Motivated by these rigorous works, we set out to determine the overall stress-strain relation of a piezoelectric composite, in which inclusions are randomly distributed in a piezoelectric matrix. The inclusions and the matrix are assumed to be perfectly bonded, without any sliding, void nucleation, or growth on their interfaces. All the phases are taken to be transversely isotropic and may possess distinctly different electroelastic moduli.

First, by modeling the disturbed strain and electric field induced by an electromechanical load as eigenfields, the anisotropic inclusion method is extended to the case of the inherently anisotropic coupled behavior of a piezoelectric composite. Then, a unified explicit expression for the coupled electroelastic Eshelby tensors of a piezoelectric ellipsoidal inclusion in a transversely isotropic medium is obtained. Finally, based on the Mori– Tanaka mean field theory, the use of the resulting tensors in the analysis of the overall electroelastic behavior of a composite containing spatially oriented inclusions is further studied. The effective electroelastic modili of two-phase piezoelectric composite materials are obtained analytically and the piezoelastic coupling effect is examined numerically.

2. ELECTROELASTIC ESHELBY TENSORS

Consider a sufficiently large piezoelectric composite D with elastic constants C_{ijmn} , piezoelectric constants e_{imn} and dielectric constants κ_{in} , which tends to undergo eigenstrain (or stress-free transformation strain) ε_{ab}^* and eigenelectric field (or electric displacementfree transformation electric field) E_b^* in an ellipsoidal inclusion defined by

$$\Omega: \frac{x_1^2}{a_1^2} + \frac{x_2^2}{a_2^2} + \frac{x_3^2}{a_3^2} \leqslant 1, \tag{1}$$

where a_1 , a_2 and a_3 are the semiaxes of the ellipsoid. The assumption that the shape of the inclusion is ellipsoidal enables one to treat composite reinforcement geometries ranging from thin flake to continuous fiber reinforcement. Suppose that the crystalline directions of the inclusion are assumed to be coincident with the principal axes of the inclusion, and both the piezoelectric matrix and piezoelectric inclusions are considered to be linearly piezoelastic. When the eigenstrain ε_{ab}^* and the eigen electric field E_b^* in the inclusion are uniform, the induced strain ε_{am} and electric field E_n inside Ω can be expressed as

$$\varepsilon_{mn} = S_{mnab}\varepsilon_{ab}^{*} - S_{mn4b}E_{b}^{*},$$

$$E_{n} = S_{4n4b}E_{b}^{*} - S_{4nab}\varepsilon_{ab}^{*},$$
(2)

where

$$S_{mnab} = \frac{1}{8\pi} [C_{ijab}(G_{mjin} + G_{njim}) - e_{iab}(G_{m4in} + G_{n4im})],$$

$$S_{mn4b} = \frac{1}{8\pi} [e_{bij}(G_{mjin} + G_{njim}) + \kappa_{ib}(G_{m4in} + G_{n4im})],$$

$$S_{4nab} = \frac{1}{4\pi} (C_{ijab}G_{4jin} - e_{iab}G_{44in}),$$

$$S_{4n4b} = \frac{1}{4\pi} (e_{bij}G_{4jin} + \kappa_{ib}G_{44in}).$$
(3)

The expression G_{MJin} (hereafter lowercase latin subscripts range from 1 to 3, while uppercase subscripts range from 1 to 4) appearing in eqn (3) has been given by Huang and Yu (1994):

$$G_{MJin} = \int_{-1}^{1} \int_{0}^{2\pi p} N_{MJ}(\bar{\xi}) D^{-1}(\bar{\xi}) \bar{\xi}_i \bar{\xi}_n \, \mathrm{d}\theta \, \mathrm{d}\bar{\zeta}_3, \tag{4a}$$

where

$$a_1 \bar{\xi}_1 = \zeta_1, \quad a_2 \bar{\xi}_2 = \zeta_2, \quad a_3 \bar{\xi}_3 = \zeta_3,$$

$$\zeta_1 / \zeta = \bar{\zeta}_1, \quad \zeta_2 / \zeta = \bar{\zeta}_2, \quad \zeta_3 / \zeta = \bar{\zeta}_3, \quad \zeta = (\zeta_1^2 + \zeta_2^2 + \zeta_3^2)^{1/2},$$

$$\bar{\zeta}_1 = (1 - \bar{\zeta}_3^2)^{1/2} \cos \theta, \quad \bar{\zeta}_2 = (1 - \bar{\zeta}_3^2)^{1/2} \sin \theta,$$
(4b)

with $N_{MJ}(\bar{\xi})$ and $D(\bar{\xi})$ being the cofactor and the determinant of the 4×4 matrix $L_{iMJn}\xi_i\xi_n$, respectively. Here $L_{iJMn} = C_{ijmn}$ as $J, M \leq 3$; $L_{iJMn} = e_{nij}$ as $J \leq 3$ and M = 4; $L_{iJMn} = e_{mn}$ as J = 4, $M \leq 3$; $L_{iJMn} = -\kappa_{in}$ as J, M = 4. Note that $N_{MJ}(\bar{\xi}) = N_{JM}(\bar{\xi})$ due to the symmetry of the electroelastic moduli L_{iMJn} .

The corresponding stress σ_{ij} and electric displacement D_i inside the inclusion due to a uniform eigenfield in Ω can be derived from the constitutive equation for a piezoelectric material given by

$$\sigma_{ij} = C_{ijmn}(S_{mnab}\varepsilon_{ab}^* - S_{mn4b}E_b^* - \varepsilon_{mn}^*) - e_{nij}(S_{4n4b}E_b^* - S_{4nab}\varepsilon_{ab}^* - E_n^*),$$

$$D_i = e_{imn}(S_{mnab}\varepsilon_{ab}^* - S_{mn4b}E_b^* - \varepsilon_{mn}^*) + \kappa_{in}(S_{4n4b}E_b^* - S_{4nab}\varepsilon_{ab}^* - E_n^*).$$
(5)

If the generalized Voight two-index notation (Huang, 1995a) is adopted, eqn (5) can be expressed in matrix form as

$$\sigma = [\mathbf{C}] \cdot ([S_{mnab}]\varepsilon^* - [S_{mn4b}]\mathbf{E}^* - \varepsilon^*) - [\mathbf{e}]^{\mathsf{T}} \cdot ([S_{4n4b}]E^* - [S_{4nab}]\varepsilon^* - E^*),$$

$$\mathbf{D} = [\mathbf{e}] \cdot ([S_{mnab}]\varepsilon^* - [S_{mn4b}]\mathbf{E}^* - \varepsilon^*) + [\kappa] \cdot ([S_{4n4b}]E^* - [S_{4nab}]\varepsilon^* - E^*),$$
(6)

where the preceding superscript T denotes matrix transpose and

$$\sigma = [\sigma_{11}, \sigma_{22}, \sigma_{33}, \sigma_{23}, \sigma_{13}, \sigma_{12}]^{\mathrm{T}}, \quad \mathbf{D} = [D_1, D_2, D_3]^{\mathrm{T}},$$

$$\varepsilon^* = [\varepsilon_{11}^*, \varepsilon_{22}^*, \varepsilon_{33}^*, \gamma_{23}^*, \gamma_{13}^*, \gamma_{12}^*]^{\mathrm{T}}, \quad \mathbf{E}^* = [E_1^*, E_2^*, E_3^*]^{\mathrm{T}}, \quad (7)$$

in which γ_{ii}^* is engineering shear eigenstrain.

For a transversely isotropic piezoelectric material with x_3 being the symmetric axis, the stiffness matrix **[C]**, the piezoelectric matrix **[e]** and the dielectric matrix **[\kappa]** are, respectively, written in the following forms:

$$[\mathbf{C}] = \begin{bmatrix} C_{11} & C_{12} & C_{13} & 0 & 0 & 0 \\ & C_{11} & C_{13} & 0 & 0 & 0 \\ & & C_{33} & 0 & 0 & 0 \\ & & & C_{44} & 0 & 0 \\ & & & & C_{66} \end{bmatrix},$$

$$[\mathbf{e}] = \begin{bmatrix} 0 & 0 & 0 & 0 & e_{15} & 0 \\ 0 & 0 & 0 & e_{15} & 0 & 0 \\ e_{31} & e_{31} & e_{33} & 0 & 0 & 0 \end{bmatrix}, \quad [\mathbf{\kappa}] = \begin{bmatrix} \kappa_{11} & 0 & 0 \\ 0 & \kappa_{11} & 0 \\ 0 & 0 & \kappa_{33} \end{bmatrix}.$$
 (8)

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For an ellipsoidal inclusion embedded in anisotropic piezoelectric media, the electroelastic Eshelby tensor S_{MnAb} has been given by Huang (1995b) and can also be written in the following matrix forms:

$$[S_{mnab}] = \begin{bmatrix} S_{11} & S_{12} & S_{13} & 0 & 0 & 0 \\ S_{21} & S_{22} & S_{23} & 0 & 0 & 0 \\ S_{31} & S_{32} & S_{33} & 0 & 0 & 0 \\ 0 & 0 & 0 & S_{44} & 0 & 0 \\ 0 & 0 & 0 & 0 & S_{55} & 0 \\ 0 & 0 & 0 & 0 & 0 & S_{66} \end{bmatrix}, \quad [S_{mn4b}] = \begin{bmatrix} 0 & 0 & S_{19} \\ 0 & 0 & S_{29} \\ 0 & 0 & S_{39} \\ 0 & S_{48} & 0 \\ S_{57} & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix},$$
$$[S_{4nab}] = \begin{bmatrix} 0 & 0 & 0 & 0 & S_{75} & 0 \\ 0 & 0 & 0 & S_{84} & 0 & 0 \\ S_{91} & S_{92} & S_{93} & 0 & 0 & 0 \end{bmatrix}, \quad [S_{4n4b}] = \begin{bmatrix} S_{77} & 0 & 0 \\ 0 & S_{88} & 0 \\ 0 & 0 & S_{99} \end{bmatrix}, \quad (9)$$

where

$$\begin{split} S_{11} &= S_{1111} = \frac{1}{4\pi} (C_{11}G_{1111} + C_{12}G_{1212} + C_{13}G_{1313} + e_{31}G_{1413}), \\ S_{12} &= S_{1122} = \frac{1}{4\pi} (C_{12}G_{1111} + C_{11}G_{1212} + C_{13}G_{1313} + e_{31}G_{1413}), \\ S_{13} &= S_{1313} = \frac{1}{4\pi} (C_{13}G_{1111} + C_{13}G_{1212} + C_{33}G_{1313} + e_{33}G_{1413}), \\ S_{19} &= S_{1143} = \frac{1}{4\pi} (e_{31}G_{1111} + e_{31}G_{1212} + e_{33}G_{1313} - \kappa_{33}G_{1413}), \\ S_{21} &= S_{2211} = \frac{1}{4\pi} (C_{11}G_{1212} + C_{12}G_{2222} + C_{13}G_{2323} + e_{31}G_{2432}), \\ S_{22} &= S_{2222} = \frac{1}{4\pi} (C_{12}G_{1212} + C_{11}G_{2222} + C_{13}G_{2323} + e_{31}G_{2423}), \\ S_{23} &= S_{2233} = \frac{1}{4\pi} (C_{13}G_{1212} + C_{13}G_{2222} + C_{33}G_{2323} - \kappa_{33}G_{2423}), \\ S_{29} &= S_{2243} = \frac{1}{4\pi} \{e_{31}(G_{1212} + G_{2222}) + e_{33}G_{2323} - \kappa_{33}G_{2423}\}, \\ S_{31} &= S_{3311} = \frac{1}{4\pi} (C_{11}G_{1313} + C_{12}G_{2323} + C_{13}G_{3333} + e_{31}G_{3433}), \\ S_{32} &= S_{3322} = \frac{1}{4\pi} (C_{12}G_{1313} + C_{11}G_{2323} + C_{13}G_{3333} + e_{31}G_{3433}), \\ S_{33} &= S_{3333} = \frac{1}{4\pi} \{e_{31}(G_{1313} + G_{2323}) + C_{33}G_{3333} - \kappa_{33}G_{3433}\}, \\ S_{39} &= S_{3343} = \frac{1}{4\pi} \{e_{31}(G_{1313} + G_{2323}) + e_{33}G_{3333} - \kappa_{33}G_{3433}\}, \\ S_{44} &= S_{2323} = S_{2332} = S_{3223} = S_{3223} = S_{3223} = S_{3232} \\ &= \frac{1}{8\pi} \{C_{44}(G_{2233} + 2G_{2323} + G_{3322}) + e_{15}(G_{2423} + G_{3422})\}, \\ \end{cases}$$

$$\begin{aligned} S_{48} &= S_{2342} = \frac{1}{8\pi} \{ e_{15}(G_{2233} + 2G_{2323} + G_{3322}) - \kappa_{11}(G_{2423} + G_{3422}) \}, \\ S_{55} &= S_{1313} = S_{1313} = S_{3113} = S_{3131} \\ &= \frac{1}{8\pi} \{ C_{44}(G_{1133} + 2G_{1313} + G_{3311}) + e_{15}(G_{1413} + G_{3411}) \}, \\ S_{57} &= S_{1341} = \frac{1}{8\pi} \{ e_{15}(G_{1133} + 2G_{1313} + G_{3311}) - \kappa_{11}(G_{1413} + G_{3411}) \}, \\ S_{66} &= S_{1212} = S_{1221} = S_{2112} = S_{2121} = \frac{1}{8\pi} C_{66}(G_{1122} + 2G_{1212} + G_{2211}), \\ S_{75} &= S_{4113} = S_{4131} = \frac{1}{4\pi} \{ C_{44}(G_{4113} + G_{4311}) + e_{15}G_{4411} \}, \\ S_{77} &= S_{4141} = \frac{1}{4\pi} \{ e_{15}(G_{4113} + G_{4311}) - \kappa_{11}G_{4411} \}, \\ S_{84} &= S_{4223} = S_{4232} = \frac{1}{4\pi} \{ C_{44}(G_{4223} + G_{4322}) + e_{15}G_{4422} \}, \\ S_{88} &= S_{4242} = \frac{1}{4\pi} \{ e_{15}(G_{4223} + G_{4322}) - \kappa_{11}G_{4323} + e_{31}G_{4433}), \\ S_{92} &= S_{4322} = \frac{1}{4\pi} \{ C_{13}(G_{4113} + C_{12}G_{4223} + C_{13}G_{4333} + e_{31}G_{4433}), \\ S_{93} &= S_{4333} = \frac{1}{4\pi} \{ C_{13}(G_{4113} + G_{4223}) + C_{33}G_{4333} - \kappa_{33}G_{4433} \}, \\ S_{99} &= S_{4343} = \frac{1}{4\pi} \{ e_{11}(G_{4113} + G_{4223}) + e_{33}G_{4333} - \kappa_{33}G_{4433} \}. \end{aligned}$$

The electroelastic Eshelby tensors given in the above equation and their transformations, as listed in Appendix, are the key ingredients necessary for determining the effective electroelastic properties of a piezoelectric composite containing spatially oriented inclusions in the subsequent sections.

3. THREE-DIMENSIONAL TRANSFORMATION

As mentioned before, the results in the previous section are based upon the assumption that the principal axes of the ellipsoid coincide with the crystalline directions of the piezoelectric matrix. In practice, it is very difficult to maintain a perfect alignment and the orientation distribution is dominated by the processing conditions. We therefore in this section turn to the associated problem involving a spatially oriented inclusion. In this case the general framework for the perfectly aligned inclusion problem in the previous section is still valid. The only change is to take the components of each tensor in the principal axes of the ellipsoid.

For a spatially oriented inclusion in a generally anisotropic medium, its orientation can be described by three Euler angles θ , ϕ and ω as depicted in Fig. 1. Let \mathbf{u}_1 , \mathbf{u}_2 and \mathbf{u}_3 be the unit vectors in the (x_1, x_2, x_3) coordinate and $\mathbf{\bar{u}}_1$, $\mathbf{\bar{u}}_2$ and $\mathbf{\bar{u}}_3$ be the unit vectors in the $(\bar{x}_1, \bar{x}_2, \bar{x}_3)$ coordinate. The relations between these two sets of unit vectors can be expressed as



$$\begin{cases} \mathbf{\tilde{u}}_1 \\ \mathbf{\tilde{u}}_2 \\ \mathbf{\tilde{u}}_3 \end{cases} = [\lambda] \begin{cases} \mathbf{u}_1 \\ \mathbf{u}_2 \\ \mathbf{u}_3 \end{cases},$$
(11)

where $[\lambda]$ is the direction cosine matrix. According to the above definitions of the rotational angles, the matrix is then given by

$$[\lambda] = \begin{bmatrix} mpr - ns & npr + ms & -qr \\ -mps - nr & mr - nps & qs \\ mq & nq & p \end{bmatrix},$$
(12)

where $m = \cos(\theta)$, $n = \sin(\theta)$, $p = \cos(\phi)$, $q = \sin(\phi)$, $r = \cos(\omega)$ and $s = \sin(\omega)$. With this transformation, the constitutive eqn (6) for a transversely isotropic piezoelectric composite defined in the $(\bar{x}_1, \bar{x}_2, \bar{x}_3)$ coordinate can be written as

$$\bar{\sigma} = [\mathbf{\bar{C}}] \cdot ([\bar{S}_{mnab}]\bar{\varepsilon}^* - [\bar{S}_{mn4b}]\mathbf{\bar{E}}^* - \bar{\varepsilon}^*) - [\mathbf{\bar{e}}]^{\mathrm{T}} \cdot ([\bar{S}_{4n4b}]\bar{E}^* - [\bar{S}_{4nab}]\bar{\varepsilon}^* - \bar{E}^*),$$

$$\mathbf{\bar{D}} = [\mathbf{\bar{e}}] \cdot ([\bar{S}_{mnab}]\bar{\varepsilon}^* - [\bar{S}_{mn4b}]\mathbf{\bar{E}}^* - \bar{\varepsilon}^*) + [\mathbf{\bar{\kappa}}] \cdot ([\bar{S}_{4n4b}]E^* - [\bar{S}_{4nab}]\bar{\varepsilon}^* - \bar{E}^*).$$
(13)

Throughout this paper the overbar denotes quantities associated within the principal directions $(\bar{x}_1, \bar{x}_2, \bar{x}_3)$ of the inclusion.

It is important to note that [C], [e] and $[S_{MnAb}]$ in eqn (6) as well as $[\tilde{\mathbf{C}}]$, $[\bar{\mathbf{e}}]$ and $[\bar{S}_{MnAb}]$ in the foregoing equations are not tensors. Thus, tensor tranformation laws cannot be directly applied to relate $[\bar{\mathbf{C}}]$, $[\bar{\mathbf{e}}]$ and $[\bar{S}_{MnAb}]$. One possible approach is to use the transformation laws for \bar{C}_{ijmn} , \bar{e}_{imn} and \bar{S}_{MnAb} first and then adopt the generalized Voigt notation to obtain $[\bar{\mathbf{C}}]$, $[\bar{\mathbf{e}}]$ and $[\bar{S}_{MnAb}]$. Complete explicit transformations of a general orthotropic case have been obtained in this work; however, the results are too lengthy to be listed herein. Shown in the Appendix are the transformed electroelastic Eshelby tensors for a transversely isotropic case.

4. EFFECTIVE ELECTROELASTIC MODULI

Before proceeding, a few notations used in the subsequent developments are introduced. The usual summation convention applies to repeated Latin subscripts. Lowercase latin subscripts range from 1 to 3, while uppercase subscripts range from 1 to 4. For example $T_J U_J = T_j U_j + T_4 U_4$, where j = 1-3. With this notation, the elastic, piezoelectric and dielectric moduli of a piezoelectric material are denoted as (Barnett and Lothe, 1975)

$$L_{iJMn} = \begin{cases} C_{ijmn} & J, M \leq 3, \\ e_{nij} & J \leq 3; & M = 4, \\ e_{imn} & J = 4; & M \leq 3, \\ -\kappa_{in} & J, M = 4. \end{cases}$$
(14)

Similarly, elastic displacement and electric potential, U_M , stress-electric displacement, Σ_{iJ} and elastic strain-electric potential gradient, Z_{Mn} , are conveniently represented as

$$U_{M} = \begin{cases} u_{m} & M \leq 3, \\ \phi & M = 4, \end{cases} \quad \Sigma_{iJ} = \begin{cases} \sigma_{ij} & J \leq 3, \\ D_{i} & J = 4, \end{cases} \quad Z_{Mn} = \begin{cases} \varepsilon_{mn} & M \leq 3, \\ \phi_{,n} = -E_{n} & M = 4, \end{cases}$$
(15)

where u_m and ϕ are the elastic displacement and electric potential, respectively.

Now, consider a sufficiently large two-phase piezoelectric composite D consists of spatially oriented ellipsoidal inhomogeneities Ω (= $\Omega_1 + \Omega_2 + \cdots + \Omega_N$) with electroelastic constants \bar{L}^*_{iJMn} and volume fraction f. The surrounding peizoelectric matrix is denoted by $D-\Omega$ and has electroelastic constants L_{iJMn} measured in the crystalline directions (x_1, x_2, x_3) of the matrix. To deal with such a piezoelectric composite with randomly oriented inclusions, the Mori-Tanaka mean field theory is employed to predict the effective elastic, piezoelectric and dielectric constants of the composite. An advantage of using this theory is that the resulting moduli satisfy self-consistency. The present analysis to follow in the traction and displacement prescribed conditions also proves this point.

4.1. Traction-electric displacement prescribed

Let a piezoelectric composite be subjected to a far-field traction and electric displacement, $\Sigma_{i,J}^{0}n_{i}$, on the boundary with outward unit normal vector n_{i} . In the absence of the piezoelectric inhomogeneity, the strain and electric field, Z_{Mn}^{0} , distributes uniformly. The existence of the inhomogeneity Ω_{k} provides disturbance in local fields of both the matrix and the kth inhomogeneity. The averages of these quantities are expressed by $\langle \Sigma_{iJ}^{m} \rangle$ and $\langle \Sigma_{iJ}^{\Omega} \rangle$ for the matrix and the kth inhomogeneity respectively. Hereafter the curly bracket $\langle \rangle$ over a field variable denotes its value obtained by volume averaging over the entire composite domain D and the superscripts 'm' and '\Omega' denote quantities in the matrix and the inhomogeneity, respectively.

Since the volume average of the disturbance portion of the stress and electric displacement vanishes, i.e.

$$\int_{D} \bar{\Sigma}_{iJ} \, \mathrm{d} \mathbf{x} = 0, \tag{16}$$

we have

$$(1-f)\langle \bar{\Sigma}_{iJ}^{m} \rangle + f\langle \bar{\Sigma}_{iJ}^{\Omega} \rangle = 0, \qquad (17a)$$

where

$$\bar{\Sigma}_{iJ}^{m} = \begin{cases} \bar{\sigma}_{iJ}^{m} = \lambda_{ip}\lambda_{jq}\sigma_{pq}^{m} & J \leq 3, \\ \bar{D}_{i}^{m} = \lambda_{ip}D_{p}^{m} & J = 4, \end{cases} \quad \bar{\Sigma}_{ij}^{\Omega} = \begin{cases} \bar{\sigma}_{iJ}^{\Omega} = \lambda_{ip}\lambda_{jq}\sigma_{pq}^{\Omega} & J \leq 3, \\ \bar{D}_{i}^{\Omega} = \lambda_{ip}D_{p}^{\Omega} & J = 4, \end{cases}$$
(17b)

with λ_{ip} being the direction cosine between the two coordinates $(\bar{x}_1, \bar{x}_2, \bar{x}_3)$ and (x_1, x_2, x_3)

as defined in eqn (12). The average disturbed stress and electric displacement in the matrix and the kth inhomogeneity can, respectively, be written as

$$\langle \bar{\Sigma}_{iJ}^m \rangle = \bar{L}_{iJMn} \langle \bar{Z}_{Mn}^m \rangle \quad \text{in } D - \Omega,$$
 (18)

$$\langle \bar{\Sigma}_{if}^{\Omega_k} \rangle = \bar{L}_{iJMn}^* (\langle \bar{Z}_{Mn}^m \rangle + \langle \bar{Z}_{Mn} \rangle) \quad \text{in } \Omega_k,$$
⁽¹⁹⁾

where $\langle \bar{Z}_{Mn}^n \rangle$ are the average strain and electric field in the matrix, $\langle \bar{Z}_{Mn} \rangle$ the average disturbance of the otherwise uniform strain and electric field in Ω_k . Since all inhomogeneities are of the same shape with the same material properties, the average value over Ω_k is identical with that over Ω_s , namely, $\langle \bar{\Sigma}_{ik}^{\Omega_k} \rangle = \langle \bar{\Sigma}_{ik}^{\Omega_k} \rangle$.

When the piezoelectric composite is subjected to the uniform far-field mechanical load and electric displacement, $\bar{\Sigma}_{iJ}^0$, the average stress and electric displacement in the inhomogeneities can be expressed as

$$\bar{\Sigma}_{iJ}^{0} + \langle \bar{\Sigma}_{iJ}^{\Omega} \rangle = \bar{L}_{iJMn}^{*} (\bar{Z}_{Mn}^{0} + \langle \bar{Z}_{Mn}^{m} \rangle + \bar{Z}_{Mn}), \qquad (20a)$$

where

$$\tilde{\Sigma}^{0}_{iJ} = \begin{cases} \bar{\sigma}^{0}_{ij} = \lambda_{ip}\lambda_{jq}\sigma^{0}_{pq} & J \leq 3, \\ \bar{D}^{0}_{i} = \lambda_{ip}D^{0}_{p} & J = 4, \end{cases} \quad \vec{Z}^{0}_{Mn} = \begin{cases} \bar{e}^{0}_{mn} = \lambda_{mp}\lambda_{nq}e^{0}_{pq} & M \leq 3, \\ -\bar{E}^{0}_{n} = -\lambda_{np}E^{0}_{p} & M = 4. \end{cases}$$
(20b)

In the derivation of eqn (20a), $\langle \vec{Z}_{Mn} \rangle = \vec{Z}_{Mn}$ in Ω has been used since the applied electromechanical load is uniform and the inhomogeneity is ellipsoidal (Huang and Yu, 1994).

By means of the equivalent inclusion method (Eshelby, 1957), the stress and electric displacement in the inhomogeneity can be simulated by those in an equivalent inclusion with the electroelastic constants of the matrix and a fictitious eigenstrain and eigenelectric field, \mathbb{Z}_{Mn}^{*} . Therefore, eqn (20a) can be written as

$$\bar{\Sigma}_{iJ}^{0} + \langle \bar{\Sigma}_{iJ}^{\Omega} \rangle = \bar{L}_{iJMn}^{*} (\bar{Z}_{Mn}^{0} + \langle \bar{Z}_{Mn}^{m} \rangle + \bar{Z}_{Mn})$$

$$= \bar{L}_{iJMn} (\bar{Z}_{Mn}^{0} + \langle \bar{Z}_{Mn}^{m} \rangle + \bar{Z}_{Mn} - \bar{Z}_{Mn}^{*}),$$
(21)

where the disturbed field \bar{Z}_{Mn} can be related to the fictitious eigenfield \bar{Z}_{Mn}^* by

$$\bar{Z}_{Mn} = \bar{S}_{MnAb} \bar{Z}_{Ab}^*, \tag{22}$$

in which \bar{S}_{MnAb} is the transformed electroelastic Eshelby tensor.

Then, the average disturbance of stress and electric displacement in the inhomogeneity can be written by substituting eqn (22) into (21) as

$$\langle \bar{\Sigma}_{ij}^{\Omega} \rangle = \bar{L}_{iJMn} \langle \bar{Z}_{Mn}^{m} \rangle + \bar{L}_{iJMn} (\bar{S}_{MnAb} - I_{MnAb}) \bar{Z}_{Ab}^{*}, \tag{23}$$

where I_{MnAb} are the fourth-order and the second-order identity tensors, namely,

$$I_{MnAb} = \begin{cases} (\delta_{ma}\delta_{nb} + \delta_{mb}\delta_{na})/2 & M, A \leq 3\\ \delta_{nb} & M, A = 4,\\ 0 & \text{otherwise.} \end{cases}$$
(24)

Combining eqns (17), (19) and (23) leads to

$$\langle \bar{Z}_{Mn}^{m} \rangle = -f(\bar{S}_{MnAb} - I_{MnAb})\bar{Z}_{Ab}^{*}.$$
(25)

Substitution of eqn (25) into (18) and (23), respectively, yields

$$\langle \bar{\Sigma}_{iJ}^m \rangle = -f \bar{L}_{iJMn} (\bar{S}_{MnAb} - I_{MnAb}) \bar{Z}_{Ab}^*, \qquad (26)$$

$$\langle \bar{\Sigma}_{iJ}^{\Omega} \rangle = (1 - f) \bar{L}_{iJMn} (\bar{S}_{MnAb} - I_{MnAb}) \bar{Z}_{Ab}^{*}.$$
⁽²⁷⁾

The equivalent eigenstrain and eigenelectric field, \bar{Z}^*_{Ab} , are solved by substituting eqn (25) into the equivalency eqn (21). Thus, we have

$$\bar{Z}_{Ab}^{*} = -\bar{U}_{AbiJ}^{-1}(\bar{L}_{iJMn}^{*} - \bar{L}_{iJMn})\bar{Z}_{Mn}^{0}, \qquad (28)$$

where \bar{U}_{AbiJ}^{-1} is the inverse of \bar{U}_{iJAb} given by

$$\bar{U}_{iJAb} = (\bar{L}^*_{iJMn} - \bar{L}_{iJMn})[(1-f)\bar{S}_{MnAb} + fI_{MnAb}] + \bar{L}_{iJAb}.$$
(29)

The overall strain and electric field, denoted by $\langle \bar{Z}_{Mn}^c \rangle$, of the piezoelectric composite can then be obtained as the weighted average of that over each phase:

$$\langle \bar{Z}^{c}_{Mn} \rangle = \frac{1}{V} \Biggl[\int_{D-\Omega} (\bar{Z}^{0}_{Mn} + \langle \bar{Z}^{m}_{Mn} \rangle) \, \mathrm{d}\mathbf{x} + \int_{\Omega} (\bar{Z}^{0}_{Mn} + \langle \bar{Z}^{\Omega}_{Mn} \rangle) \, \mathrm{d}\mathbf{x} \Biggr], \tag{30}$$

where V denotes the volume of the entire composite.

Substituting eqns (22) and (25) into the above equation results in the following equation:

$$\langle \bar{Z}^c_{Mn} \rangle = \bar{Z}^0_{Mn} + f \bar{Z}^*_{Mn}. \tag{31}$$

with the equivalent eigenfields Z_{Mn}^* obtained from eqn (28), the overall strain and electric field $\langle \bar{Z}_{Mn}^c \rangle$ readily follow as

$$\langle \bar{Z}^c_{Mn} \rangle = \bar{E}^{-1}_{MniJ} \bar{\Sigma}^0_{iJ}, \qquad (32)$$

where \bar{E}_{MniJ}^{-1} is the effective electroelastic compliance of the piezoelectric composite and is defined as

$$\bar{E}_{MniJ}^{-1} = [I_{MnAb} - f\bar{U}_{MnqR}^{-1}(\bar{L}_{qRAb}^* - \bar{L}_{qRAb})]\bar{L}_{AbiJ}^{-1}.$$
(33)

4.2. Elastic displacement-electric field prescribed

When the piezoelectric composite is subjected to a far-field applied elastic displacement and electric field, Z_{Mn}^{0} , a disturbed strain and electric field due to the presence of the piezoelectric inhomogeneities result in each phase. The volume average of the disturbed strain and electric field must vanish which yields

$$(1-f)\langle \bar{Z}_{Mn}^{m}\rangle + f\langle \bar{Z}_{Mn}^{\Omega}\rangle = 0, \qquad (34)$$

where $\langle Z_{Mn}^{m} \rangle$ and $\langle \overline{Z}_{Mn}^{\Omega} \rangle$ represent the average strain and electric field in the matrix and the inhomogeneities, respectively. Then, the average stress and electric displacement in the inhomogeneities can be obtained by the equivalent inclusion method. To solve the equivalent eigen field \overline{Z}_{Mn}^{*} , the following equations

$$\langle \bar{Z}_{Mn}^{m} \rangle = -f \bar{S}_{MnAb} \bar{Z}_{Ab}^{*},$$

$$\langle \bar{Z}_{Mn}^{\Omega} \rangle = \langle \bar{Z}_{Mn}^{m} \rangle + \bar{Z}_{Mn},$$
 (35)

are substituted into the equivalency eqn (21) which can then be expressed as

$$\bar{Z}_{Ab}^{*} = -\bar{V}_{AbiJ}^{-1}(\bar{L}_{iJMn}^{*} - \bar{L}_{iJMn})\bar{Z}_{Mn}^{0}, \qquad (36)$$

where \bar{V}_{AbiJ}^{-1} is the inverse of \bar{V}_{iJAb} defined by

$$\bar{V}_{iJAb} = (1 - f)(\bar{L}^*_{iJMn} - \bar{L}_{iJMn})\bar{S}_{MnAb} + \bar{L}_{iJAb}.$$
(37)

The overall stress and electric displacement, $\langle \bar{\Sigma}_{ij}^c \rangle$, of the piezoelectric composite is defined in a manner similar to that used in eqn (30) as

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$$\langle \bar{\Sigma}_{ij}^c \rangle = \frac{1}{V} \left[\int_{D-\Omega} (\bar{\Sigma}_{ij}^0 + \langle \bar{\Sigma}_{ij}^m \rangle) \, \mathrm{d}\mathbf{x} + \int_{\Omega} (\bar{\Sigma}_{ij}^0 + \langle \bar{\Sigma}_{ij}^\Omega \rangle) \, \mathrm{d}\mathbf{x} \right].$$
(38)

Substituting eqns (22) and (35) into (38), followed by some manipulation yields

$$\langle \bar{\Sigma}_{iJ}^c \rangle = \bar{L}_{iJMn} (\bar{Z}_{Mn}^0 - f \bar{Z}_{Mn}^*).$$
(39)

Furthermore, several steps of straightforward manipulation after substitution of eqn (35) into (39) result in

$$\langle \bar{\Sigma}_{iJ}^c \rangle = \bar{E}_{iJMn} \bar{Z}_{Mn}^0, \tag{40}$$

where the effective electroelastic moduli \bar{E}_{iJMn} is obtained as

$$\bar{E}_{iJMn} = \bar{L}_{iJAb} [I_{AbMn} + f \bar{V}_{AbqR}^{-1} (\bar{L}_{qRMn}^* - \bar{L}_{qRMn})].$$
(41)

It can be shown that the effective electroelastic stiffness \bar{E}_{iJMn} given by eqn (28) and compliance \bar{E}_{MniJ}^{-1} given by eqn (33) are reciprocal to each other, namely, $\bar{E}_{MniJ}^{-1}\bar{E}_{iJAb} = I_{MnAb}$. This verifies that the proposed method to compute effective electroelastic moduli satisfies self-consistency. It is also of interest to examine the behavior of the present model for the two-phase piezoelectric composite in the low (dilute) and high concentration limits. As $f \to 0$, \bar{E}_{MniJ}^{-1} reduces to \bar{L}_{MniJ}^{-1} , while \bar{E}_{MniJ}^{-1} becomes $\bar{L}_{MniJ}^{*}^{-1}$ as $f \to 1$. In a similar manner, \bar{E}_{iJMn} can be shown to be \bar{L}_{iJMn} and \bar{L}_{iJMn}^{*} in the limits $f \to 0$ and $f \to 1$, respectively.

5. NUMERICAL EXAMPLES

In this section, analytical predictions of the effective electroelastic moduli for PZT-5 material reinforced by BaTiO₃ continuous fibers $(a_1 = a_2, a_3 \rightarrow \infty)$ are calculated using the proposed method. Similar to the study of the electroelastic Eshelby tensors in the earlier section, both the matrix and the inhomogeneities are considered to be transversely isotropic with x_3 as a symmetry axis. The electroelastic material properties of the constituents used in the numerical computation are given below (Huang, 1995a):

BaTiO₃ continuous fiber :

$$C_{11}^{*} = 166 \text{ GPa}, C_{33}^{*} = 162 \text{ GPa}, C_{44}^{*} = 43 \text{ GPa}, C_{12}^{*} = 77 \text{ GPa}, C_{13}^{*} = 78 \text{ GPa},$$
$$e_{31}^{*} = -4.4 \text{ Cm}^{-2}, e_{33}^{*} = 18.6 \text{ Cm}^{-2}, e_{15}^{*} = 11.6 \text{ Cm}^{-2},$$
$$\kappa_{11}^{*} = 11.2 \times 10^{-9} \text{ C}^{2} \text{ N}^{-1} \text{m}^{-2}, \\ \kappa_{33}^{*} = 12.6 \times 10^{-9} \text{ C}^{2} \text{ N}^{-1} \text{m}^{-2},$$

PZT-5H matrix:

$$C_{11} = 126 \text{ GPa}, C_{33} = 117 \text{ GPa}, C_{44} = 35.3 \text{ GPa}, C_{12} = 55 \text{ GPa}, C_{13} = 53 \text{ GPa},$$
$$e_{31} = -6.5 \text{ Cm}^{-2}, e_{33} = 23.3 \text{ Cm}^{-2}, e_{15} = 17.0 \text{ Cm}^{-2},$$
$$\kappa_{11} = 15.1 \times 10^{-9} \text{ C}^2 \text{ N}^{-1} \text{m}^{-2}, \kappa_{33} = 13.0 \times 10^{-9} \text{ C}^2 \text{ N}^{-1} \text{m}^{-2}.$$

The electroelastic Eshelby tensors for a continuous fiber in transversely isotropic piezoelectric solids have been given by Huang and Yu (1994) as

$$S_{1111} = S_{2222} = \frac{5C_{11} + C_{12}}{8C_{11}}, \quad S_{1122} = S_{2211} = \frac{3C_{12} - C_{11}}{8C_{11}},$$
$$S_{1133} = S_{2233} = \frac{C_{13}}{2C_{11}}, \quad S_{1143} = S_{2243} = \frac{e_{31}}{2C_{11}},$$
$$S_{1212} = S_{1221} = S_{2112} = S_{2121} = \frac{3C_{11} - C_{12}}{8C_{11}},$$



Fig. 2. The composite normal modulus \bar{E}_{1111} against f and ϕ .

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

$$S_{4141} = S_{4242} = \frac{1}{2}.$$
 (42)

The transformation of the above tensors can be easily performed as listed in the Appendix.

It is noted that, in mapping a tensor into a matrix through the generalized Voigt twoindex notation, care should be taken in accounting for the shear strain terms, i.e. the factor of two. Thus, the inversion of a fourth-order tensor is not a standard matrix manipulation. A special scheme for the fourth-order inversion must be developed which is briefly introduced here. First, with the notation, a 9 by 9 matrix is constructed for the given fourthorder tensor. The matrix element in columns 4 to 6 is two times their corresponding tensor component. The 9 by 9 matrix is inverted and is then used to map the corresponding inverse tensor. In mapping the matrix back to the corresponding tensor, each element in columns 4 to 6 is divided by 2 to obtain the tensor element. With this scheme, \bar{U}_{MnqR}^{-1} in eqn (33) and \bar{V}_{AbqR}^{-1} in eqn (41) can be evaluted, followed by the results of the compliance \bar{E}_{Mnid}^{-1} and the stiffness \bar{E}_{iJMn} , respectively. The obtained compliance tensor from eqn (33) is inverted to compare with the stiffness tensor from eqn (41); of course both approaches are found to yield identical results.

The rotation θ with respect to the axis 3, is irrelevant to the composite properties since both the inclusion and the matrix are transversely isotropic and the inclusion is a continuous fiber (cylinder). As a result, the Eshelby tensors are symmetric in the 1–2 plane. Thus the resulting composite behavior is only a function of ϕ , ω and f. In addition, to simplify the discussion, representative results at $\omega = 0$ are evaluated in examining the effect of orientation and piezoelectric coupling on the problem. The composite normal moduli \vec{E}_{1111} and \vec{E}_{2222} are shown in Figs 2 and 3, respectively, with the inclusion content f varied from 0 to 1 and ϕ varied from 0° to 90°. As f increases, the influence of ϕ diminishes and at f = 1 the composite is a monolithic BaTiO₃ material. Figure 4 shows the shear modulus \vec{E}_{1212} , indicating that the influence of ϕ is stronger, and the nonlinear relation between f and \vec{E}_{1212} is more prominent than that between f and \vec{E}_{1111} and \vec{E}_{2222} . Figures 5 and 6 are the results for \vec{E}_{3411} (e_{31} of the composite) and \vec{E}_{1441} (κ_{11} of the composite), respectively.

Figures 7, 8 and 9 display the piezoelectric coupling effect on elastic moduli. The moduli have been normalized by the corresponding results without taking into account piezoelectric effects. By prescribing that all e_{ij} in both the inclusion and matrix vanish, the



Fig. 4. The composite shear modulus \vec{E}_{1212} against f and ϕ .

obtained composite stiffness constants are denoted by a symbol ('). For the material system studied, the piezoelectric effect is found to enhance \vec{E}_{1111} and reduce \vec{E}_{2222} . For both the moduli, the highest coupling effect occurs at $\phi = 45^{\circ}$ and about f = 0.45, while the effect naturally vanishes at f = 0 and 1 as well as at $\phi = 0^{\circ}$ and 90°. For the shear modulus \vec{E}_{1212} , the coupling effect is found to be even more prominent, and the highest coupling occurs at $\phi = 45^{\circ}$ and f = 1.0.

It is noted, as pointed out by Weng (1992), that when the moduli of the matrix are larger (smaller) than those of inhomogeneities, the effective elastic moduli \vec{E}_{ijmn} lie on Willis' (1977) upper (lower) bound. Therefore, in the absence of piezoelectric coupling, the present prediction for the effective elastic constants of this two-phase composite are the lower bounds, while the effective dielectric constants \vec{E}_{i44n} are the upper bounds on the effective



Fig. 5. The composite piezoelectric constant \vec{E}_{3411} against f and ϕ .



Fig. 6. The composite dielectric constant \overline{E}_{1441} against f and ϕ .

moduli. However, the bounded solutions analogous to those of Hashin and Shtrikman's (1963) ones for the effective electroelastic moduli E_{iJMn} of a piezoelectric composite have not appeared in the literature yet. Although the results of Eshelby tensors and the resulting composite moduli are given only for long fibers in this paper, the proposed analytical model is able to treat multiphase composites with ellipsoidal reinforcements, and thus the influence of inclusion shape on the resulting piezoelectric behavior can also be analyzed by using the theoretical basis developed in the present article (Huang and Kuo, 1996).

6. CONCLUDING REMARKS

The rigorous anisotropic inclusion method has been extended to derive the electroelastic Eshelby tensors for an ellipsoidal piezoelectric inclusion in an infinite matrix.



Fig. 7. Effect of piezoelastic coupling on $\bar{E}_{(11)}$.



Fig. 8. Effect of piezoelastic coupling on \vec{E}_{2222} .

Explicit expressions have been obtained for the electroelastic Eshelby tensors for spheroidal inclusions in a transversely isotropic matrix. With the expressions for the electroelastic Eshelby tensors in hand, a micromechanics model has been developed to predict the effective electroelastic moduli of a composite reinforced with piezoelectric inhomogeneities. The effect of inclusion-inclusion interaction at high concentrations has been taken into account, using the Mori–Tanaka mean field theory. The model proposed in the present paper is able to analyze the effect of inclusion shape on the composite piezoelastic behavior, thus applicable to a much wider range of composite microstructural geometry. Finally, an illustrated example of a BaTiO₃/PZT-5H composite has been given. The results indicate that the piezoelastic coupling enhances \vec{E}_{1111} and \vec{E}_{1212} while it reduces \vec{E}_{2222} for the composite



Fig. 9. Effect of piezoelastic coupling on \bar{E}_{1212} .

studied. It is also found that the coupling effect is determined by both the orientation and the inclusion content. It should be noted that the analytical expressions for the effective elastic constants of this two-phase composite are the lower bounds, while the effective dielectric constants are the upper bounds on the effective moduli. The determination of these bounds should be of interest to pursue, although this could be a challenging work due to the coupled behavior and the inherent anisotropy of piezoelectric materials.

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APPENDIX

Transformed electroelastic Eshelby tensors

Listed herein are the electroelastic Eshelby tensors for a continuous fiber $(a_1 = a_2, a_3 = \infty)$ with the axis-3 being the longitudinal direction. Short-handed symbols are used. Due to symmetry in the 1-2 plane, we have following relations for nonzero terms:

$$S_{11} = S_{22} = S_{1111} = S_{2222},$$

$$S_{12} = S_{1122} = S_{2211},$$

$$S_{13} = S_{23} = S_{1133} = S_{2233},$$

$$S_{44} = S_{55} = S_{1313} = S_{1331} = S_{3113} = S_{3131} = S_{2323} = S_{2332} = S_{3223} = S_{3232},$$

$$S_{66} = S_{1212} = S_{1221} = S_{2112} = S_{2121},$$

$$S_{77} = S_{88} = S_{4141} = S_{4242},$$

$$S_{19} = S_{29} = S_{1143} = S_{2243}.$$
(A1)

In representing the electroelastic Eshelby tensors for a general orthotropic case, we find that using the following three constants enables the transformation to be greatly simplified.

$$Y_{1} = S_{22} + S_{33} - S_{23} - S_{32} - 4S_{44},$$

$$Y_{2} = S_{13} + S_{31} - S_{13} - S_{31} - 4S_{55},$$

$$Y_{3} = S_{11} + S_{22} - S_{12} - S_{21} - 4S_{66}.$$
 (A2)

For the case studied, the tensors are transversely isotropic (eqns (42) and (A1)) with the axis-3 being a symmetric axis. Then, from the foregoing equation, we have

$$Y_1 = Y_2 = \frac{-3C_{11} + C_{12} - 4C_{13}}{8C_{11}},$$

$$Y_3 = 0.$$
 (A3)

Employing the transformation laws for S_{MnAb} with the help of eqn (A3), the components of the transformed electroelastic Eshelby tensors can be written concisely in the following forms:

$$\begin{split} & S_{11} = S_{11}(p^2r^2 + s^2) - Y_1q^2r^2(s^2 - p^2r^2), \\ & S_{12} = S_{23}q^2s^2 + S_{32}q^2r^2 + S_{12}p^2 + Y_1q^4r^2s^2, \\ & S_{13} = S_{23}p^2 + S_{32}q^2r^2 + S_{12}q^2s^2 + Y_1p^2q^2r^2, \\ & S_{14} = S_{23}pqs + S_{12}pqs + Y_1pq^3r^2s, \\ & S_{15} = S_{11}pqr/2 - (S_{23} - S_{32})pqr(s^2 - r^2)/2 + Y_1pqr(1 - 2q^2r^2)/2, \\ & S_{16} = S_{11}q^2rs/2 - (S_{23} - S_{32})q^2rs/2 + Y_1q^2rs(1 - 2q^2r^2)/2, \\ & S_{17} = -S_{19}qr(1 - q^2r^2), \\ & S_{18} = S_{19}qs(1 - q^2r^2), \\ & S_{19} = S_{19}p(1 - q^2r^2), \\ & S_{22} = S_{11}(p^2s^2 + r^2) - Y_1q^2s^2(p^2s^2 + r^2), \\ & S_{23} = S_{12}q^2r^2 + S_{23}p^2 + S_{32}q^2s^2 + Y_1p^2q^2s^2, \end{split}$$

$$\begin{split} & S_{24} = -S_{11}pqs/2 + (S_{23} - S_{32})pqs/2 - Y_1p^3qs^3, \\ & S_{25} = -S_{23}pqr + S_{12}pqr - Y_1pq^3rs^2, \\ & S_{26} = S_{11}q^2rs/2 - (S_{23} - S_{32})q^2rs/2 + Y_1q^2rs(1 - 2q^2s^2)/2, \\ & S_{27} = -S_{19}qr(1 - q^2s^2), \\ & S_{28} = S_{19}qs(1 - q^2s^2), \\ & S_{33} = S_{11}q^2 - Y_1p^2q^2, \\ & S_{34} = -S_{11}pqs/2 + (S_{13} - S_{31})pqs/2 + Y_1pqs(p^2 - q^2)/2, \\ & S_{35} = S_{11}pqr/2 - (S_{13} - S_{31})pqs/2 + Y_1pqr(-p^2 + q^2)/2, \\ & S_{36} = -S_{23}q^2rs/2 + S_{12}q^2rs - Y_1p^2q^2rs, \\ & S_{37} = -S_{19}q^3r, \\ & S_{38} = S_{19}q^3, \\ & S_{39} = S_{19}qq^2, \\ & S_{44} = S_{44}(p^2r^2 + s^2) + S_{66}q^2r^2 + Y_1p^2q^2s^2, \\ & S_{45} = -S_{44}q^2rs + S_{66}q^2rs - Y_1p^2q^2rs, \\ & S_{46} = -S_{44}qqrs + S_{66}qqr - Y_1pq^3rs^2, \\ & S_{47} = S_{19}pq^2rs, \\ & S_{48} = -S_{19}pq^2s^2, \\ & S_{55} = S_{44}(p^2s^2 + r^2) + S_{66}q^2s^2 + Y_1p^2q^2r^2, \\ & S_{56} = S_{44}pqs - S_{66}pqs + Y_1pq^3r^2s, \\ & S_{57} = -S_{19}pq^2rs, \\ & S_{59} = S_{19}p^2qr, \\ & S_{58} = S_{19}pq^2rs, \\ & S_{59} = S_{19}p^2qr, \\ & S_{56} = S_{44}q^2 + S_{66}p^2 + Y_1q^4r^2s^2, \\ & S_{57} = -S_{19}q^3r^2s, \\ & S_{59} = S_{19}p^2rs, \\ & S_{59} = S_{19}pq^2rs, \\ & S_{59} = S_{19}pq^2$$

For \vec{S}_{ij} where i > j, simply replace S_{ij} by S_{ji} in the above expressions.

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(A4)