

Viscoelectroelastic behavior of heterogeneous piezoelectric solids

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We study the viscoelectroelastic behavior of heterogeneous piezoelectric solids, focusing on the connection between heterogeneity and coupled mechanical and electrical relaxations. Our approach is based on the existence of a correspondence between quasistatic viscoelectroelasticity and static piezoelectricity when linear constitutive response exists. We couple this correspondence principle with micromechanics models to predict the overall behavior of heterogeneous piezoelectric solids in terms of microstructural details. We devote specific attention to a class of two-phase materials consisting of a lossless piezoelectric phase embedded in a lossy (mechanically and electrically) matrix and obtain closed form expressions for the effective complex electroelastic moduli. Numerical results are presented and discussed, and qualitative agreement with experiment is observed. © 2001 American Institute of Physics. [DOI: 10.1063/1.1337595]

I. INTRODUCTION

Widespread practical applications of piezoelectric solids in numerous technological fields have fueled the understanding of many of their physical properties. A set of properties that has received little study, though, are the complex elastic, dielectric, and piezoelectric constants. Actually, the first two have received considerable attention in uncoupled elastic and dielectric solids and many excellent texts exist.¹⁻³ Of course the complex elastic and dielectric constants are associated with mechanical and electrical losses in the material, respectively, and in linear theoretical treatments, alternative, but equivalent, representations exist. These include the loss tangent and quality factor, for both elastic and electric losses.

Complex piezoelectric constants, however, have received far less attention in the literature, but they present many interesting features. Their complex parts do not necessarily represent losses, and they are influenced by both the mechanical and dielectric losses and their coupling through the piezoelectric effect. The rigorous thermodynamic treatment of complex piezoelectric constants (their existence, constraints on properties, etc.) has existed for quite some time,⁴⁻⁷ and a handful of observations exist. In most of the observed cases, the source of the complex piezoelectric constants was heterogeneity at some length scale. These include observed Debye-type relaxations in ferroelectric single crystals that have been attributed to ferroelectric domain wall motion,⁸ and relaxation in polycrystalline ceramics due to motion of 90° domain walls.⁹⁻¹² Complex piezoelectric constants have also been observed in PVDF piezoelectric polymers where they are attributed to the strong interactions between amorphous and highly oriented crystalline phases.¹³ At a higher length scale, they have been observed in bone,¹⁴ and in piezoelectric ceramic/polymer matrix composites.¹⁵⁻¹⁷ In bone, they are strongly influenced by the highly heterogeneous microstructure. Their role in polymer matrix compos-

ites is especially significant because of the extensive applications of such materials in transducer applications. In underwater sonar applications, temperature changes can significantly affect mechanical and electric losses in the polymer phase, which in turn impact the complex piezoelectric constants in a nontrivial way. For example, due to the interaction between the piezoelectric effect of the ceramic and the elastic and dielectric relaxation of the polymer, piezoelectric relaxation will be produced in the composite (demonstrated by complex piezoelectric constants), even though neither phase shows coupled elastic and dielectric relaxation.

Although experimental studies of complex piezoelectric constants in heterogeneous media are limited, corresponding modeling efforts are rarer, even though substantial effort has been directed toward micromechanical modeling of elastic, dielectric, and piezoelectric constants of heterogeneous solids in the absence of losses. The latter include, in recent years, efforts toward estimating the effective moduli of the piezoelectric composites in terms of microstructural details and constituent properties;¹⁸⁻²⁶ establishing exact connections between different components of the effective moduli and internal electroelastic fields;²⁷⁻³³ and establishing rigorous upper and lower bounds on the effective moduli.³⁴⁻³⁷

In this work we study the effect of relaxation in the phases of a heterogeneous solid on the overall response of the solid. Our approach follows along similar lines as Hashin,^{38,39} Weng and Li,⁴⁰ and Dunn⁴¹ regarding viscoelastic composites. We do not consider details of the origin of relaxation in individual phases, but instead we assume these relaxations can be represented by complex moduli. Our focus is then on the interaction of these phases and how that affects the overall behavior of the heterogeneous solid. In Sec. II we introduce the basic equations and notation of *viscoelectroelasticity*. The correspondence between the quasistatic viscoelectroelastic problem and the static piezoelectric problem is established in Sec. III, where the complex electroelastic moduli are introduced. The effective complex electroelastic moduli for heterogeneous materials are studied in Sec. IV

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using the correspondence principle. First, simple microstructural models are developed to try to identify the key phenomena, then more realistic composite microstructures are analyzed using the Mori–Tanaka mean field approach.⁴² Finally, numerical results are presented and compared with measurements in Sec. V.

II. BASIC EQUATIONS AND NOTATION

Uncoupled elastic and dielectric relaxation of materials has been studied extensively and integral constitutive equations have been used to express the dependence of the current material response on the mechanical and electric loading history. In a similar manner, the constitutive equations for the linear viscoelectroelastic effect, where the elastic and dielectric relaxation are coupled, through the piezoelectric effect, can be written as

$$\begin{aligned} \sigma_{ij}(\mathbf{x}, t) &= \int_{-\infty}^t \left[C_{ijkl}(\mathbf{x}, t - \tau) \frac{d\varepsilon_{kl}(\mathbf{x}, \tau)}{d\tau} \right. \\ &\quad \left. - e_{ijk}(\mathbf{x}, t - \tau) \frac{dE_k(\mathbf{x}, \tau)}{d\tau} \right] d\tau, \\ D_i(\mathbf{x}, t) &= \int_{-\infty}^t \left[e_{ikl}(\mathbf{x}, t - \tau) \frac{d\varepsilon_{kl}(\mathbf{x}, \tau)}{d\tau} \right. \\ &\quad \left. + \kappa_{ik}(\mathbf{x}, t - \tau) \frac{dE_k(\mathbf{x}, \tau)}{d\tau} \right] d\tau, \end{aligned} \tag{1}$$

where σ_{ij} and ε_{ij} are stress and strain tensors, respectively; D_i and E_k are the electric displacement and field, respectively; C_{ijkl} , e_{ijk} , and κ_{ik} are elastic, piezoelectric, and dielectric moduli, respectively; \mathbf{x} is the position vector of a material point (in general all the quantities are regarded as functions of position); t and τ are the current and a reference time, respectively. In general, each component of C_{ijkl} , e_{ijk} , and κ_{ik} may have different time dependence. It is observed from the constitutive equations that (i) elastic and electric relaxations are fully coupled through the piezoelectric relaxation moduli e_{ijk} ; (ii) by ignoring the piezoelectric relaxation, the familiar constitutive equations for uncoupled mechanical and dielectric relaxations are recovered; (iii) the stress and electric displacement depend on the strain and electric field history. These characteristics distinguish the quasistatic viscoelectroelastic effect from the static piezoelectric effect, as well as from uncoupled mechanical and dielectric relaxations.

In the analysis that follows, it is convenient to treat the elastic and electric fields on equal footing. To this end, the notation introduced by Barnett and Lothe⁴³ for piezoelectric analysis is utilized. This notation is identical to conventional indicial notation with the exception that lowercase subscripts take on the range 1→3, while uppercase subscripts take on the range 1→4 and repeated uppercase subscripts are summed over 1→4. With this notation, the field variables take the following forms:

$$\Sigma_{iJ}(\mathbf{x}, t) = \begin{cases} \sigma_{ij}(\mathbf{x}, t) \\ D_i(\mathbf{x}, t) \end{cases}, \quad Z_{Ji}(t) = \begin{cases} \varepsilon_{ij}(\mathbf{x}, t) \\ -E_i(\mathbf{x}, t) \end{cases}, \tag{2}$$

$$U_J(t) = \begin{cases} u_j(\mathbf{x}, t), & J = 1, 2, 3 \\ \phi(\mathbf{x}, t), & J = 4, \end{cases}$$

and the electroelastic moduli can be written as

$$E_{iJKl}(\mathbf{x}, t - \tau) = \begin{cases} C_{ijkl}(\mathbf{x}, t - \tau), & J, K = 1, 2, 3 \\ e_{ijl}(\mathbf{x}, t - \tau), & K = 4, \quad J = 1, 2, 3 \\ e_{ikl}(\mathbf{x}, t - \tau), & J = 4, \quad K = 1, 2, 3 \\ -\kappa_{il}(\mathbf{x}, t - \tau), & J = 4, \quad K = 4. \end{cases} \tag{3}$$

With the electroelastic fields and relaxation moduli written in this abbreviated notation, the constitutive equations (1) can be rewritten as

$$\Sigma_{iJ}(\mathbf{x}, t) = \int_{-\infty}^t E_{iJKl}(\mathbf{x}, t - \tau) \frac{dZ_{Kl}(\mathbf{x}, \tau)}{d\tau} d\tau. \tag{4}$$

Equation (4) reduces to the static piezoelectric constitutive equations when the electroelastic relaxation moduli do not depend on time, i.e., when there is no relaxation.

The equilibrium and gradient equations for viscoelectroelastic solid with a distribution of body force and electric charge are

$$\Sigma_{iJ,i}(\mathbf{x}, t) + \rho_J(\mathbf{x}, t) = 0 \tag{5}$$

and

$$Z_{Ji}(\mathbf{x}, t) = U_{J,i}(\mathbf{x}, t), \tag{6}$$

where ρ_J is the body force when J equals to 1–3, and electric charge when J equals 4; the subscript comma is used to denote the partial differentiation with respect to the corresponding coordinate. The problem now is to solve the electroelastic fields modeled by Eqs. (4)–(6), and subjected to the following boundary conditions:

$$\Sigma_{iJ}(\mathbf{x}, t) = \Sigma_{iJ}^0(t) \quad \text{on } S_1 \tag{7a}$$

and

$$U_J(\mathbf{x}, t) = U_J^0(\mathbf{x}, t) \quad \text{on } S_2, \tag{7b}$$

where S_1 and S_2 combined to form the surface of the solid. Note that now all the field variables are functions of time.

III. CORRESPONDENCE PRINCIPLE AND COMPLEX MODULI

A. Viscoelectroelastic correspondence principle

Consider the Laplace transform of a function $f(\mathbf{x}, t)$, $\hat{f}(\mathbf{x}) = \int_0^\infty f(\mathbf{x}, t) e^{-st} dt$, where s is the transform variable. Taking the Laplace transform of Eqs. (4)–(7) yields

$$\hat{\Sigma}_{iJ}(\mathbf{x}) = s \hat{E}_{iJKl}(\mathbf{x}) \hat{Z}_{Kl}(\mathbf{x}), \tag{8}$$

$$\hat{\Sigma}_{iJ,i}(\mathbf{x}) + \hat{\rho}_J(\mathbf{x}) = 0, \tag{9}$$

$$\hat{Z}_{Ji}(\mathbf{x}) = \hat{U}_{J,i}(\mathbf{x}), \tag{10}$$

$$\hat{\Sigma}_{iJ}(\mathbf{x}) = \hat{\Sigma}_{iJ}^0 \quad \text{on } S_1, \tag{11}$$

$$\hat{U}_J(\mathbf{x}) = \hat{U}_J^0(\mathbf{x}) \quad \text{on } S_2. \tag{12}$$

In the context of linear viscoelasticity $s\hat{E}_{iJKl}$ are called the *transform domain* (TD) moduli; we adopt the same terminology here. The form of the set of Eqs. (8)–(12) is identical to the set of equations for linear static piezoelectricity when the transforms of the viscoelectroelastic variables are associated with the corresponding static electroelastic variables, and $s\hat{E}_{iJKl}$ is associated with the static electroelastic moduli E_{iJKl} . For the problem modeled by Eqs. (4)–(7), it then follows that the *Laplace-transformed viscoelectroelastic solution is obtained directly from the solution of the corresponding static electroelastic problem by replacing static electroelastic moduli E_{iJKl} with the transform domain moduli $s\hat{E}_{iJKl}$* . The final solution can be realized by inverting the transformed solution. This is the *viscoelectroelastic correspondence principle*. This correspondence principle reveals that static piezoelectric solutions can be easily converted to quasistatic viscoelectroelastic solutions. This correspondence principle is directly analogous to correspondence principles in linear elasticity and electrostatics of dielectrics. It is applicable to both homogeneous and heterogeneous materials.

B. Complex electroelastic moduli

Assume that the viscoelectroelastic solid is subjected to a sinusoidal strain and electric field

$$Z_{Ji}(\mathbf{x},t) = Z_{Ji}(\mathbf{x})e^{i\omega t}, \tag{13}$$

where ω is the circular frequency and $i = \sqrt{-1}$. Substituting Eq. (13) into Eq. (4), and making the change of variables $p = t - \tau$ yields

$$\Sigma_{iJ}(\mathbf{x},t) = i\omega Z_{Kl}(\mathbf{x})e^{i\omega t} \int_0^\infty E_{iJKl}(\mathbf{x},p)e^{-i\omega p} dp. \tag{14}$$

Let

$$E_{iJKl}^c(\mathbf{x},i\omega) = i\omega \int_0^\infty E_{iJKl}(\mathbf{x},p)e^{-i\omega p} dp, \tag{15}$$

where the superscript c is used to explicitly denote complex quantities, Eq. (14) can be recast as

$$\Sigma_{iJ}(\mathbf{x},t) = E_{iJKl}^c(\mathbf{x},i\omega)Z_{Kl}(\mathbf{x},t). \tag{16}$$

Because Eq. (16) formally resembles the static piezoelectric constitutive equation, the E_{iJKl}^c are called *complex electroelastic moduli*. E_{iJKl}^c can be separated into real and imaginary parts:

$$E_{iJKl}^c(\mathbf{x},i\omega) = E_{iJKl}^r(\mathbf{x},i\omega) + iE_{iJKl}^i(\mathbf{x},i\omega), \tag{17}$$

where the superscripts r and i denote the real and imaginary parts of the complex moduli. The real and imaginary parts of the complex moduli are related to the piezoelectric enthalpy stored and dissipated by the viscoelectroelastic solid and, thus, are called the *storage moduli* and *loss moduli*, respectively. By choosing a different set of independent variables, an alternative representation of the complex electroelastic moduli related to internal energy can also be obtained. The ratio of loss moduli to storage moduli is called the *loss tangent*.

Let us now consider the transform domain moduli,

$$s\hat{E}_{iJKl}(\mathbf{x}) = s \int_0^\infty E_{iJKl}(\mathbf{x},t)e^{-st} dt. \tag{18}$$

Comparing Eq. (18) to Eq. (15) shows that the functional dependence of the complex electroelastic moduli $E_{iJKl}^c(i\omega)$ on frequency $i\omega$ is precisely the same as the functional dependence of transform domain moduli $s\hat{E}_{iJKl}$ on the transform variable s . Therefore, we have

$$E_{iJKl}^c(\mathbf{x},i\omega) = i\omega \hat{E}_{iJKl}(\mathbf{x},i\omega). \tag{19}$$

From the correspondence principle established in Sec. III A, it is clear that there is also a correspondence between the complex electroelastic moduli $E_{iJKl}^c(i\omega)$ and the static electroelastic moduli of piezoelectric solid.

IV. THE EFFECTIVE COMPLEX ELECTROELASTIC MODULI OF HETEROGENEOUS SOLIDS

We have considered arbitrary loading, material properties, and field variables in Secs. III A and III B. Now we sharpen our focus and turn our attention to heterogeneous viscoelectroelastic solids subjected to sinusoidal electroelastic loading, so that Eq. (16) and the complex electroelastic moduli are applicable. Consider a multiphase composite with piecewise homogeneity described by

$$E_{iJKl}^c(\mathbf{x},t) = \sum_r E_{iJKl}^c(t)|_r \Phi_r(\mathbf{x}), \tag{20}$$

where the subscript r and the symbol $|_r$ are used to denote properties associated with phase r . $\Phi_r(\mathbf{x})$ is a characteristic function describing the topology of the microstructure of the heterogeneous material such that

$$\Phi_r(\mathbf{x}) = \begin{cases} 1, & \mathbf{x} \in r \\ 0, & \mathbf{x} \notin r. \end{cases} \tag{21a}$$

It has the following property:

$$\langle \Phi_r(\mathbf{x})P(\mathbf{x}) \rangle = c_r P_r, \tag{21b}$$

where $\langle \cdot \rangle = (1/V) \int_V (\cdot) dV$ denotes an average over the volume of heterogeneous material V ; $P(\mathbf{x})$ is any integrable material property that varies over the microstructure, and P_r is the value of that property in phase r ; c_r is the volume fraction of phase r and satisfies $\sum_r c_r = 1$. If the characteristic length scale of heterogeneity is much less than the wavelength of the applied loading, the effective constitutive equation can be expressed in term of the effective complex electroelastic moduli $E_{iJKl}^*(i\omega)$,

$$\langle \Sigma_{iJ}(\mathbf{x},t) \rangle = E_{iJKl}^*(i\omega) \langle Z_{Kl}(\mathbf{x},t) \rangle. \tag{22}$$

The correspondence between the complex electroelastic moduli $E_{iJKl}^c(i\omega)$ and the TD moduli $i\omega \hat{E}_{iJKl}(i\omega)$, Eq. (19), combined with the viscoelectroelastic correspondence principle established in Sec. III, leads to a correspondence between the effective complex electroelastic moduli of heterogeneous viscoelectroelastic solids and the effective static electroelastic moduli of piezoelectric solids. That is, *the effective complex electroelastic moduli of a heterogeneous ma-*

terial are found by replacing the constituent static electroelastic moduli by the constituent complex electroelastic moduli in the relationships between the effective static electroelastic moduli and the constituent static electroelastic moduli of the heterogeneous piezoelectric solid. This correspondence principle can be coupled with any exact results or with a micromechanics model to estimate the effective complex moduli of heterogeneous viscoelectroelastic solids. In Secs. IV A and IV B we focus on two-phase composites consisting of one phase that is piezoelectric and lossless (e.g., a piezoelectric ceramic) and another phase that is not piezoelectric, but is both elastically and electrically lossy (e.g., a polymer).

A. One-dimensional model systems

To demonstrate the basic ideas, we first consider two one-dimensional models; the constitutive equations of the composite are given by

$$\sigma = C^c \varepsilon - e^c E, \quad D = e^c \varepsilon + \kappa^c E. \tag{23}$$

In the following analysis, we assume that phase 1 is the uncoupled viscoelastic material with volume fraction c_1 , elastic constant $C_1^c = C_1^r + iC_1^i$, and dielectric constant $\kappa_1^c = \kappa_1^r + i\kappa_1^i$; and phase 2 is the lossless piezoelectric material with volume fraction c_2 , elastic constant C_2 , piezoelectric

constant e_2 , and dielectric constant κ_2 , all being real. In the following we will refer to phase 1 as the *viscoelastic material* and phase 2 as the *piezoelectric material*, recognizing that each possesses the properties just outlined.

First consider the case where the viscoelastic and piezoelectric materials are arranged in parallel and electric and/or elastic loading is applied parallel to the layering. In this configuration, the strain ε and electric field E in both phases are equal and equal to the average strain and average electric field in the composite, respectively. As a result, each component of the effective moduli of the composite is given exactly by the linear rule of mixtures. Notable in this case is the fact that the composite exhibits no piezoelectric relaxation.

Now, consider the case where the viscoelastic and piezoelectric materials are arranged in series and electric and/or elastic loading is applied perpendicular to the layering. In this configuration, the stress σ and electric displacement D in both phases are equal and equal to the average stress and average electric displacement in the composite, respectively. The average strain and average electric field in the composite can be straightforwardly determined from the volume average of the strain and electric field in the constituents which leads to the following expressions for the effective moduli of the composite:

$$\begin{aligned} C^r &= \frac{C_1^r(c_1 e_2^2 + c_1 C_2 \kappa_2 + c_2 C_2 \kappa_2^r)}{c_1^2 e_2^2 + (c_1 C_2 + c_2 C_1^r)(c_1 \kappa_2 + c_2 \kappa_1^r)}, & C^i &= \frac{c_1 [(c_1 e_2^2 + c_2 C_2 \kappa_1^r + c_1 C_2 \kappa_2)^2 C_1^i - c_2^2 C_1^{r2} e_2^2 \kappa_1^i]}{[c_1^2 e_2^2 + (c_1 C_2 + c_2 C_1^r)(c_1 \kappa_2 + c_2 \kappa_1^r)]^2}, \\ e^r &= \frac{c_2 C_1^r \kappa_1^r e_2}{c_1^2 e_2^2 + (c_1 C_2 + c_2 C_1^r)(c_1 \kappa_2 + c_2 \kappa_1^r)}, & e^i &= \frac{c_1 c_2 e_2 [c_2 C_1^{r2} \kappa_2 \kappa_1^i + c_1 C_1^r (e_2^2 + C_2 \kappa_2) \kappa_1^i + \kappa_1^r (c_1 e_2^2 + c_2 C_2 \kappa_1^r + c_1 C_2 \kappa_2) C_1^i]}{[c_1^2 e_2^2 + (c_1 C_2 + c_2 C_1^r)(c_1 \kappa_2 + c_2 \kappa_1^r)]^2}, \\ \kappa^r &= \frac{\kappa_1^r (c_1 e_2^2 + c_1 C_2 \kappa_2 + c_2 C_1^r \kappa_2)}{c_1^2 e_2^2 + (c_1 C_2 + c_2 C_1^r)(c_1 \kappa_2 + c_2 \kappa_1^r)}, & \kappa^i &= \frac{c_1 \{c_1^2 e_2^4 \kappa_1^i + (c_1 C_2 + c_2 C_1^r)^2 \kappa_2 \kappa_1^i + e_2^2 [2c_1 (c_1 C_2 + c_2 C_1^r) \kappa_2 \kappa_1^i - c_2^2 \kappa_1^{r2} C_1^i]\}}{[c_1^2 e_2^2 + (c_1 C_2 + c_2 C_1^r)(c_1 \kappa_2 + c_2 \kappa_1^r)]^2}. \end{aligned} \tag{24}$$

As is apparent from Eqs. (24), piezoelectric loss results from the interaction between piezoelectric coupling of the piezoelectric material and the elastic and dielectric losses of the viscoelastic material. This is shown by dependence of e^i on the product of e_2 and C_1^i or κ_1^i of the polymer. Since κ_1^i is negative while C_1^i is positive, e^i may change sign with respect to change in the volume fraction (note that the sign convention for dielectric loss moduli is different from the usual choice). It can also be concluded from Eq. (24) that the dielectric loss moduli κ^i and elastic loss moduli C^i of the composite may be larger than the corresponding constants in uncoupled elastic composites due to the piezoelectric coupling. These phenomena are shown graphically in Fig. 1, where representative numerical results are presented in terms of a map of the real versus imaginary parts of the constants as the volume fraction of the piezoelectric material increases from zero to unity. In the calculation, we have used $C_1^c = 6.37 + 0.157i$ GPa, $\kappa_1^c / \kappa_0 = 4.431 - 0.0585i$, $C_2 = 111$

GPa, $\kappa_2 / \kappa_0 = 830$, and $e_2 = 15.8$ C²/m. Also shown in Fig. 1 are corresponding results for an uncoupled composite; these calculations were carried out using the same equations but with $e_2 = 0$.

B. Three-dimensional multiphase composites

Turning to more realistic microstructures, let us consider the polymer-based piezoelectric composites, where the polymer is elastic, electrically and elastically lossy, and isotropic, and the reinforcement is piezoelectric, lossless, and transversely isotropic. The x_3 direction is assumed to be the unique axis for both the ceramic reinforcement and the composite. Owing to the interaction between the viscoelastic and piezoelectric phases, the composite demonstrates the viscoelectroelastic effect represented by the effective complex electroelastic moduli. Our goal is to establish the connections between the effective complex electroelastic moduli of

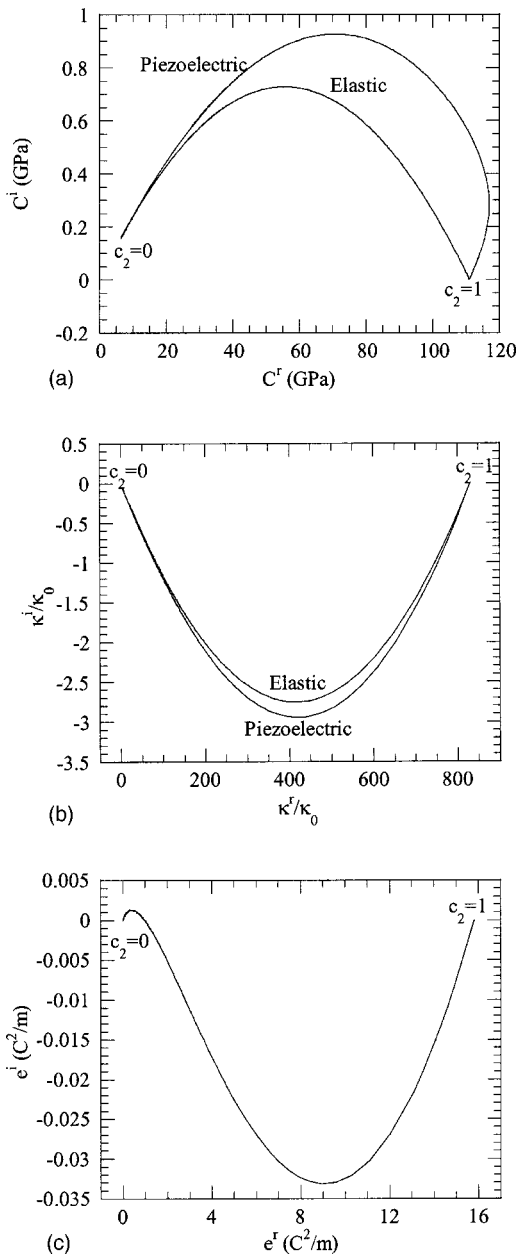


FIG. 1. Storage-loss map of electroelastic moduli of one-dimensional series composite: (a) elastic constant; (b) dielectric constant; (c) piezoelectric constant.

polymer-based composites and the microstructure. We do so by applying the correspondence principle along with the Mori–Tanaka mean-field approach.⁴² We obtained closed form expressions for the effective complex moduli. Details of the Mori–Tanaka approach are given by Mura⁴⁴ and Nemat-Nasser and Hori.⁴⁵ Its application to piezoelectric ma-

terials can be found in Dunn and Taya.²³ The basic concept of the Mori–Tanaka approach is to approximate the actual average electroelastic fields in the reinforcement by the electroelastic fields that exist in a single reinforcement embedded in an infinite matrix, subjected to a uniform electroelastic field at the boundary equal to the as yet unknown average field in the matrix. The average field in a single reinforcement can be obtained using Eshelby’s equivalent inclusion concept.⁴⁶ The necessary auxiliary problem for piezoelectric media has been solved by Dunn and Taya²² and Dunn and Wienecke.⁴⁷ With this combination of the correspondence principle and the Mori–Tanaka approach, the effective complex electroelastic moduli for a composite consisting of an ellipsoidal piezoelectric phase embedded in a piezoelectric matrix can be expressed as

$$E_{iJKl}^* = E_{iJMn}^c|_1 + c_2(E_{iJMn}^c|_2 - E_{iJMn}^c|_1)A_{Mnkl}^{MT}, \quad (25)$$

where

$$A_{Mnkl}^{MT} = A_{MnOp}^{dil} [c_1 I_{klop} + c_2 A_{klop}^{dil}]^{-1} \quad (26)$$

and

$$A_{Mnkl}^{dil} = [I_{klmn} + S_{klop} E_{opij}^{-1} |_1 (E_{iJMn}^c|_2 - E_{iJMn}^c|_1)]^{-1}. \quad (27)$$

The Eshelby tensor \mathbf{S} is a function of electroelastic moduli of matrix and aspect ratio of the inclusion, and has been tabulated in Dunn and Taya²² and Dunn and Wienecke.⁴⁷ The results of Eq. (25) simplify considerably for the special cases of fibrous or laminated piezoelectric composites where one phase is piezoelectric, but lossless, and the other is nonpiezoelectric, but lossy. The following equations summarize the effective complex electroelastic moduli for such fibrous and laminated composites. In the equations, the notation $|_i$ ($i = 1, 2$) denotes the phase ($1 =$ matrix, $2 =$ reinforcement) and for simplification we use the Hill moduli $k = (C_{11} + C_{12})/2$ and $m = (C_{11} - C_{12})/2$.

Fibrous composites:

$$k^r = \frac{k_1^r k_2 + c_2 m_1^r k_2 + c_1 k_1^r m_1^r}{c_1 k_2 + c_2 k_1^r + m_1^r},$$

$$k^i = \frac{c_1 [c_2 (k_2 - k_1^r)^2 m_1^i + (k_2 + m_1^r)^2 k_1^i]}{(c_1 k_2 + c_2 k_1^r + m_1^r)^2},$$

$$m^r = \frac{m_1^r [(1 + c_2) k_1^r m_2 + 2 m_1^r m_2 + c_1 k_1^r m_1^r]}{c_1 k_1^r m_2 + 2 c_1 m_1^r m_2 + (1 + c_2) k_1^r m_1^r + 2 c_2 m_1^r{}^2},$$

$$\begin{aligned} \frac{m^i}{c_1} &= \frac{2 c_2 (m_2 - m_1^r)^2 m_1^r{}^2}{[c_1 k_1^r m_2 + 2 c_1 m_1^r m_2 + (1 + c_2) k_1^r m_1^r + 2 c_2 m_1^r{}^2]^2} k_1^i \\ &+ \frac{(1 + c_2) k_1^r{}^2 m_1^r{}^2 + 2 m_2 k_1^r m_1^r (c_1 k_1^r + 2 m_1^r) + m_2^2 [(1 + c_2) k_1^r{}^2 + 4 (k_1^r + m_1^r) m_1^r]}{[c_1 k_1^r m_2 + 2 c_1 m_1^r m_2 + (1 + c_2) k_1^r m_1^r + 2 c_2 m_1^r{}^2]^2} m_1^i, \end{aligned}$$

$$\begin{aligned}
 C_{13}^r &= 2C_{12}^r|_1 + \frac{2c_2C_{11}^r|_1(C_{13}|_2 - 2C_{12}^r|_1)}{c_1k_2 + c_2k_1^r + m_1^r}, \\
 C_{13}^i &= 2C_{12}^i|_1 + 2c_2 \left[\frac{(C_{13}|_2 - 2C_{12}^i|_1)C_{11}^i|_1 - C_{11}^r|_1C_{12}^i|_1}{c_1k_2 + c_2k_1^r + m_1^r} - \frac{C_{11}^r|_1(C_{13}|_2 - 2C_{12}^r|_1)(c_2k_1^i + m_1^i)}{(c_1k_2 + c_2k_1^r + m_1^r)^2} \right], \\
 C_{33}^r &= 2c_1C_{11}^r|_1 + c_2C_{33}|_2 - \frac{c_1c_2(C_{13}|_2 - 2C_{12}^r|_1)^2}{c_1k_2 + c_2k_1^r + m_1^r}, \\
 C_{33}^i &= 2c_1C_{11}^i|_1 + \left[\frac{4c_1c_2(C_{13}|_2 - 2C_{12}^r|_1)C_{12}^i|_1}{c_1k_2 + c_2k_1^r + m_1^r} + \frac{2c_1c_2(C_{13}|_2 - 2C_{12}^r|_1)^2(c_2k_1^i + m_1^i)}{(c_1k_2 + c_2k_1^r + m_1^r)^2} \right], \\
 C_{44}^r &= m_1^r + \frac{2c_1c_2m_1^r(e_{15}|_2^2 - C_{44}|_2\kappa_{11}|_2 - m_1^r\kappa_{11}|_2) + (1+c_2)(C_{44}|_2 - m_1^r)\kappa_{11}^r|_1}{c_1^2(e_{15}|_2^2 - C_{44}|_2\kappa_{11}|_2) + (1-c_2^2)(C_{44}|_2\kappa_{11}^r|_1 + \kappa_{11}|_2m_1^r) + (1+c_2)^2m_1^r\kappa_{11}^r|_1}, \\
 C_{44}^i &= m_1^i + 2c_2b^r \frac{c_1e_{15}|_2^2 + [c_1\kappa_{11}|_2 + (1+c_2)\kappa_{11}^r|_1](C_{44}|_2 - 2m_1^r)}{b^{r^2} + b^{i^2}} m_1^i + \frac{2(1+c_2)bm_1^r(C_{44}|_2 - m_1^r)\kappa_{11}^i|_1}{b^{r^2} + b^{i^2}} \\
 &\quad - 2c_2m_1^r \frac{c_1e_{15}|_2^2 + [c_1\kappa_{11}|_2 + (1+c_2)\kappa_{11}^r|_1](C_{44}|_2 - 2m_1^r)}{b^{r^2} + b^{i^2}} b^i, \\
 e_{31}^r &= \frac{2c_2e_{31}|_2C_{11}^r|_1}{c_1k_2 + c_2k_1^r + m_1^r}, \quad e_{31}^i = \frac{2c_2e_{31}|_2C_{11}^i|_1}{c_1k_2 + c_2k_1^r + m_1^r} + \frac{2c_2C_{11}^r|_1e_{31}^r|_1(c_2k_1^i + m_1^i)}{(c_1k_2 + c_2k_1^r + m_1^r)^2}, \\
 e_{33}^r &= c_2 \left[e_{33}|_2 - \frac{c_1e_{31}|_2(C_{13}|_2 - 2C_{12}^r|_1)}{c_1k_2 + c_2k_1^r + m_1^r} \right], \\
 e_{33}^i &= c_1c_2e_{31}|_2 \left[\frac{2C_{12}^i|_1}{c_1k_2 + c_2k_1^r + m_1^r} + \frac{(C_{13}|_2 - 2C_{12}^r|_1)(c_2k_1^i + m_1^i)}{(c_1k_2 + c_2k_1^r + m_1^r)^2} \right], \\
 e_{15}^r &= \frac{4c_2e_{15}|_2m_1^r\kappa_{11}^r|_1}{c_1^2(e_{15}|_2^2 - C_{44}|_2\kappa_{11}|_2) + (1-c_2^2)(C_{44}|_2\kappa_{11}^r|_1 + \kappa_{11}|_2m_1^r) + (1+c_2)^2m_1^r\kappa_{11}^r|_1}, \\
 e_{15}^i &= - \frac{4c_2e_{15}|_2\kappa_{11}^r|_1[(1-c_2^2)(C_{44}|_2\kappa_{11}^i|_1 + \kappa_{11}|_2m_1^i) + (1+c_2)^2(\kappa_{11}^r|_1m_1^i + m_1^r\kappa_{11}^i|_1)]}{[c_1^2(e_{15}|_2^2 - C_{44}|_2\kappa_{11}|_2) + (1-c_2^2)(C_{44}|_2\kappa_{11}^r|_1 + \kappa_{11}|_2m_1^r) + (1+c_2)^2m_1^r\kappa_{11}^r|_1]^2} \\
 &\quad + \frac{4c_2e_{15}|_2(\kappa_{11}^r|_1m_1^i + m_1^r\kappa_{11}^i|_1)}{c_1^2(e_{15}|_2^2 - C_{44}|_2\kappa_{11}|_2) + (1-c_2^2)(C_{44}|_2\kappa_{11}^r|_1 + \kappa_{11}|_2m_1^r) + (1+c_2)^2m_1^r\kappa_{11}^r|_1}, \\
 \kappa_{11}^r &= \kappa_{11}^r|_1 + 2c_2\kappa_{11}^r|_1 \frac{c_1(e_{15}|_2^2 + C_{44}|_2\kappa_{11}|_2 - C_{44}|_2\kappa_{11}^r|_1) + (1+c_2)(\kappa_{11}|_2 - \kappa_{11}^r|_1)m_1^r}{c_1^2(e_{15}|_2^2 - C_{44}|_2\kappa_{11}|_2) + (1-c_2^2)(C_{44}|_2\kappa_{11}^r|_1 + \kappa_{11}|_2m_1^r) + (1+c_2)^2m_1^r\kappa_{11}^r|_1}, \\
 \kappa_{11}^i &= \kappa_{11}^i|_1 - 2c_2\kappa_{11}^r|_1 \frac{c_1e_{15}|_2^2 + [c_1C_{11}|_2 + (1+c_2)m_1^r](\kappa_{11}|_2 - \kappa_{11}^r|_1)}{b^{r^2} + b^{i^2}} b^i + \frac{2(1+c_2)c_2b\kappa_{11}^r|_1(\kappa_{11}|_2 - \kappa_{11}^r|_1)m_1^i}{b^{r^2} + b^{i^2}} \\
 &\quad + 2c_2b^r \frac{c_1e_{15}|_2^2 + [c_1C_{44}|_2 + (1+c_2)m_1^r](\kappa_{11}|_2 - 2\kappa_{11}^r|_1)}{b^{r^2} + b^{i^2}} \kappa_{11}^i|_1, \\
 \kappa_{33}^r &= c_1\kappa_{11}^r|_1 + c_2\kappa_{33}|_2 + \frac{c_1c_2e_{31}|_2^2}{c_1k_2 + c_2k_1^r + m_1^r}, \quad \kappa_{33}^i = c_1\kappa_{11}^i|_1 - \frac{c_1c_2e_{31}|_2^2(c_2k_1^i + m_1^i)}{(c_1k_2 + c_2k_1^r + m_1^r)^2},
 \end{aligned}$$

where

$$b^r = c_1^2e_{15}|_2^2 + [c_1C_{44}|_2 + (1+c_2)m_1^r][c_1\kappa_{11}|_2 + (1+c_2)\kappa_{11}^r|_1],$$

$$\frac{b^i}{1+c_2} = [c_1 C_{44|2} + (1+c_2)m_1^r] \kappa_{11|1}^i + [c_1 \kappa_{11|2} + (1+c_2)\kappa_{11|1}^r] m_1^i.$$

Laminated composites:

$$k^r = -c_2 \frac{(e_{31|2} e_{33|2} + C_{13|2} \kappa_{33|2})(C_{13|2} - C_{13}^r) + (e_{31|2} - e_{31}^r)(C_{13|2} e_{33|2} - C_{33|2} e_{31|2})}{e_{33|2}^2 + C_{33|2} \kappa_{33}^2}$$

$$- \frac{c_1(C_{13}^r - 2C_{12|1}^r)C_{12|1}^r}{C_{11|1}^r} + c_1 k_1^r + c_2 k_2,$$

$$k^i = c_1 k_1^i + \frac{c_2(e_{31|2} e_{33|2} + C_{13|2} \kappa_{33|2})C_{13}^i + (C_{13|2} e_{33|2} - C_{33|2} e_{31|2})e_{31}^i}{e_{33|2}^2 + C_{33|2} \kappa_{33}^2}$$

$$- 2c_1(C_{13}^r - 2C_{12|1}^r) \left[\frac{\kappa_{11|1}^r C_{12|1}^r + C_{12|1}^r \kappa_{11}^i}{2\kappa_{11}^r C_{11|1}^r} + \frac{\kappa_{11|1}^r C_{12|1}^r (\kappa_{11|1}^r C_{11|1}^i + C_{11|1}^r \kappa_{11|1}^i)}{2\kappa_{11}^r C_{11|1}^r{}^2} \right],$$

$$m^r = c_1 m_1^r + c_2 m_2, \quad m^i = c_1 m_1^i,$$

$$C_{13}^r = \frac{c_2[e_{33}^r(C_{13|2} e_{33|2} - C_{33|2} e_{31|2}) + C_{33}^r(e_{31|2} e_{33|2} + C_{13|2} \kappa_{33|2})]}{e_{33|2}^2 + C_{33|2} \kappa_{33|2}} + \frac{c_1 C_{12|1}^r C_{33}^r}{C_{11|1}^r},$$

$$C_{13}^i = \frac{c_2[(C_{13|2} e_{33|2} - C_{33|2} e_{31|2})e_{33}^i + (e_{31|2} e_{33|2} + C_{13|2} \kappa_{33|2})C_{33}^i]}{e_{33|2}^2 + C_{33|2} \kappa_{33|2}} + \frac{c_1 C_{12|1}^r C_{33}^r}{C_{11|1}^r}$$

$$+ c_1 C_{33}^r C_{12|1}^r \left(\frac{\kappa_{11|1}^i}{C_{11|1}^r \kappa_{11|1}^r} - \frac{\kappa_{11|1}^r C_{11|1}^i + C_{11|1}^r \kappa_{11|1}^i}{C_{11|1}^r{}^2 \kappa_{11|1}^r} \right),$$

$$C_{33}^r = \frac{c_1}{a^r \kappa_{11|1}^r} + \frac{c_2 C_{33|2}}{a^r (e_{33|2}^2 + C_{33|2} \kappa_{33|2})},$$

$$C_{33}^i = - \frac{c_1(\kappa_{11|1}^r a^i + a^r \kappa_{11}^i)}{a^{r2} \kappa_{11|1}^r{}^2} - \frac{c_2 a^i C_{33|2}}{a^{r2} (e_{33|2}^2 + C_{33|2} \kappa_{33|2})},$$

$$C_{44}^r = \frac{C_{44|2} m_1^r}{c_1 C_{44|2} + c_2 m_1^r}, \quad C_{44}^i = \frac{c_1 C_{44|2} m_1^i}{(c_1 C_{44|2} + c_2 m_1^r)^2},$$

$$e_{31}^r = \frac{c_2[\kappa_{33}^r(C_{33|2} e_{31|2} - C_{13|2} e_{33|2}) + e_{33}^r(e_{31|2} e_{33|2} + C_{13|2} \kappa_{33|2})]}{e_{33|2}^2 + C_{33|2} \kappa_{33|2}} + \frac{c_1 C_{12|1}^r e_{33}^r}{C_{11|1}^r},$$

$$e_{31}^i = \frac{c_2[(C_{33|2} e_{31|2} - C_{13|2} e_{33|2})\kappa_{33}^i + (e_{31|2} e_{33|2} + C_{13|2} \kappa_{33|2})e_{33}^i]}{e_{33|2}^2 + C_{33|2} \kappa_{33|2}} + \frac{c_1(C_{12|1}^r e_{33}^i + e_{33}^r C_{12|1}^r)}{C_{11|1}^r} - \frac{c_1 C_{12|1}^r e_{33}^r C_{11|1}^i}{C_{11|1}^r{}^2},$$

$$e_{33}^r = \frac{c_2 e_{33|2}}{a^r (e_{33|2}^2 + C_{33|2} \kappa_{33|2})},$$

$$\kappa_{11}^i = c_1 \kappa_{11|1}^i + \frac{e_{15}^{r2} C_{44}^i - 2C_{44}^r e_{15}^r e_{15}^i}{C_{44}^r{}^2},$$

$$e_{33}^i = - \frac{c_2 e_{33|2} a^i}{a^{r2} (e_{33|2}^2 + C_{33|2} \kappa_{33|2})},$$

$$\kappa_{33}^r = \frac{c_1}{2a^r C_{11|1}^r} + \frac{c_2 \kappa_{33|2}}{a^r (e_{33|2}^2 + C_{33|2} \kappa_{33|2})},$$

$$e_{15}^r = \frac{c_2 e_{15|2} C_{44}^r}{C_{44|2}}, \quad e_{15}^i = \frac{c_2 e_{15|2} C_{44}^i}{C_{44|2}},$$

$$\kappa_{33}^i = c_1 \frac{-a^r \kappa_{11|1}^r C_{11|1}^i - \kappa_{11|1}^r C_{11|1}^r a^i}{2C_{11|1}^r{}^2 a^{r2} \kappa_{11|1}^r}$$

$$\kappa_{11}^r = c_1 \kappa_{11|1}^r + c_2 \kappa_{11|2} + \frac{c_2 e_{15|2}{}^2}{C_{44|2}} - \frac{e_{15}^{r2}}{C_{44}^r{}^2},$$

$$- \frac{c_2 \kappa_{33|2} a^i}{a^{r2} (e_{33|2}^2 + C_{33|2} \kappa_{33|2})}$$

TABLE I. Electroelastic moduli of PZT-5A and epoxy. Units: C_{ij} , GPa; e_{ij} , C²/m; κ_0 (permittivity of free space)= 8.85×10^{-12} C²/Nm².

	C_{11}^r	C_{11}^i	C_{12}^r	C_{12}^i	κ_{11}^r/κ_0	κ_{11}^i/κ_0	
Epoxy	6.37	0.034	3.58	-0.012	4.43	-0.059	
PZT-5A	121	0	75.2	0	916	0	
	C_{13}	C_{33}	C_{44}	e_{31}	e_{33}	e_{15}	κ_{33}/κ_0
Epoxy	3.58	6.37	1.40	0	0	0	4.43
PZT-5A	75.2	111	21.1	-5.4	15.8	12.3	830

where

$$a^r = \frac{c_2 e_{33|2}^2}{(e_{33|2}^2 + C_{33|2} \kappa_{33|2})^2} + \left(\frac{c_2 \kappa_{33|2}}{e_{33|2}^2 + C_{33|2} \kappa_{33|2}} + \frac{c_1}{2C_{11|1}^r} \right) \times \left(\frac{c_2 C_{33|2}}{e_{33|2}^2 + C_{33|2} \kappa_{33|2}} + \frac{c_1}{\kappa_{11|1}^r} \right),$$

$$a^i = \left(\frac{c_2 C_{33|2}}{e_{33|2}^2 + C_{33|2} \kappa_{33|2}} + \frac{c_1}{\kappa_{11|1}^r} \right) \frac{c_1 C_{11|1}^i}{2C_{11|1}^r} - \frac{2c_1 c_2 \kappa_{33|2} \kappa_{11|1}^i}{2\kappa_{11|1}^r (e_{33|2}^2 + C_{33|2} \kappa_{33|2})} - \frac{c_1 \kappa_{11|1}^i}{2C_{11|1}^r \kappa_{11|1}^r}.$$

V. NUMERICAL RESULTS AND DISCUSSION

To demonstrate the applicability of the general theory, we present numerical results for PZT-5A reinforced epoxy composites. The material moduli of PZT-5A and epoxy are summarized in Table I. In order to study the influence of the piezoelectric effect, we also present the effective complex moduli of composites with pure elastic reinforcement, for which the piezoelectric constants are assumed to be zero. Both fibrous composites and laminates are considered. In the following we loosely refer to the phases as the *ceramic* and the *polymer*, although more specifically we mean the piezoelectric phase and the nonpiezoelectric phase.

It is important to note that $\sum_{ij} Z_{ji}$ represents the piezoelectric enthalpy rather than the internal energy with the current constitutive representation, thus it is not necessarily positive. The loss tangents, representing the imaginary part of the piezoelectric enthalpy, thus are also not necessarily positive. The overall internal energy dissipated, which is related to the piezoelectric enthalpy, however, is always positive.

A. Fibrous composites

Figure 2(a) shows the complex moduli C_{11} , C_{12} , C_{13} , and C_{33} of piezoelectric and elastic fibrous composites. These are shown in the form of maps of the real versus imaginary parts of the moduli; other representations can be easily constructed from these results. These constants are identical in the piezoelectric and elastic composites. This is because in their definition they are associated with the uniform far-field strains $\epsilon_{11} = \epsilon_{22}$ and ϵ_{33} . These strains are uncoupled from the electric field in the polymer matrix due to the fibrous configuration because they only induce electric

displacement D_3 in piezoelectric ceramic, which has no effect on the electric field in polymer matrix. Therefore, the stress state in piezoelectric composite under these strains is identical to that in the elastic composite. The loss moduli of

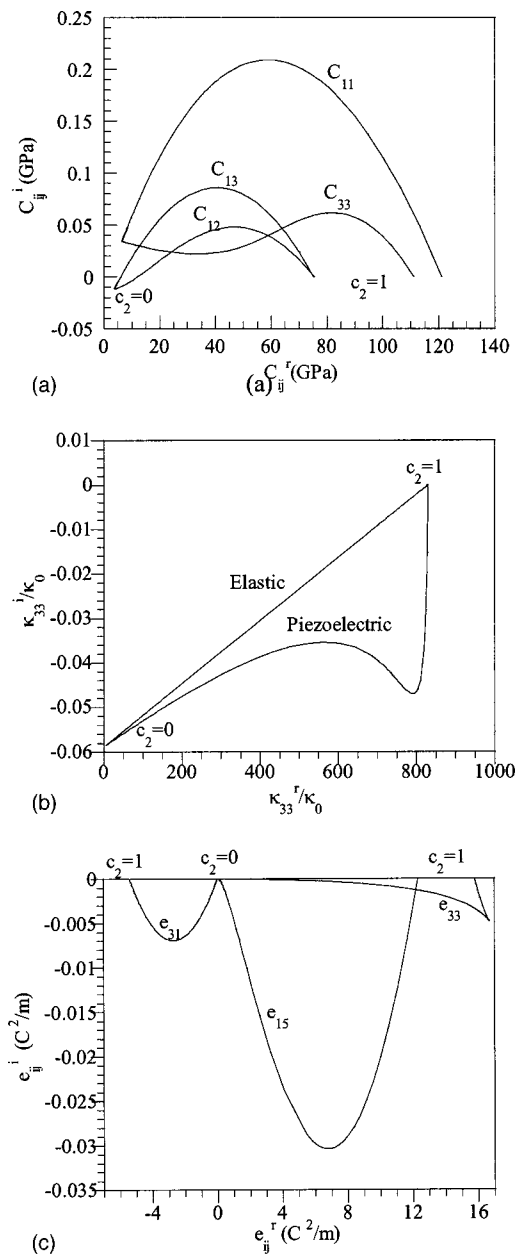


FIG. 2. Storage-loss map of electroelastic moduli of fibrous composites: (a) elastic moduli; (b) dielectric modulus; (c) piezoelectric moduli.

the composites are much higher than those of matrix in some fiber volume-fraction regions, and exhibit a maximum at high volume fractions. This is because the ceramic is much stiffer than the polymer, and has zero loss moduli. It is also noted that the loss tangents of C_{12} and C_{13} change signs from negative to positive with the increase of volume fraction of fiber. The negative loss tangent of C_{12} of the polymer suggests that $\sigma_{11}\epsilon_{22}$ produces rather than dissipates energy. However, the overall energy dissipated for the deformation ϵ_{22} , $(\sigma_{11} + \sigma_{22})\epsilon_{22}$, remains positive. The addition of elastic or piezoelectric fibers changes the signs of loss tangent of C_{12} and C_{13} , because for the fibrous configuration, the lateral deformation of polymer is constrained by the ceramic fibers.

Figure 2(b) shows the complex modulus for the dielectric constant κ_{33} of the piezoelectric and elastic composites. Due to the fibrous configuration, the average electric field E_3 in the elastic composite, the polymer matrix, and the ceramic fiber are identical. Thus the effective κ_{33} in the elastic composite obeys the rule of mixtures. For piezoelectric composites, the storage modulus is approximately given by rule of mixture for the following reason. An applied far-field E_3 will induce σ_{11} and σ_{22} in the piezoelectric ceramic, which will then introduce σ_{11} and σ_{22} , as well as the corresponding strains ϵ_{11} , ϵ_{22} , and ϵ_{33} , in the polymer matrix due to the continuity of traction between the phases. In order to maintain overall zero strain in the composite, negative strains $-\epsilon_{11}$, $-\epsilon_{22}$, and $-\epsilon_{33}$ are also introduced in the ceramic. However, the strain ϵ_{33} must be continuous across fibrous boundary and thus must remain zero. Therefore, the deformations ϵ_{11} and ϵ_{22} in the polymer and ceramic phases are also constrained, and have limited effect on electric displacement D_3 . As a result the storage modulus of the piezoelectric composite does not deviate significantly from a linear variation with respect to the fiber volume fraction. The loss modulus of the piezoelectric composite is higher than that of elastic composite due to the additional contribution from the elastic loss in the polymer, and shows two local peaks. The first peak at high volume fraction is because the addition of the polymer phase to the piezoelectric ceramic introduces dielectric loss in the composite that does not exist in the piezoelectric ceramic. With further addition of the polymer phase, the volume average of the dielectric loss due to the piezoelectric coupling decreases, and the loss modulus also decreases. Eventually it will reach the point where the elastic loss from the polymer dominates, and that is the reason for the second peak.

Figure 2(c) shows the complex piezoelectric constants e_{31} , e_{33} , and e_{15} of the fibrous piezoelectric composite. Peaks occur in the loss moduli at high volume fractions for all three piezoelectric constants because the loss moduli must be zero at volume fractions of zero and unity. Between these two limits, the interaction between the piezoelectric effect of the fibers and the relaxation of the polymer results in the nonzero piezoelectric loss moduli of the composite. The losses in e_{31} and e_{33} are purely due to the elastic loss in the polymer, and thus, they have a positive loss tangent. Again, this is because the far-field strains they are associated with, ϵ_{11} and ϵ_{33} , are uncoupled from the electric field in polymer

matrix due to the fibrous configuration, as discussed for Fig. 1(a). Thus the dielectric loss in the polymer has no effect on the piezoelectric loss in this particular deformation mode. The loss tangent of e_{15} changes sign with an increase in the volume fraction. This is because the piezoelectric constant e_{15} is associated with the far-field strain ϵ_{13} , which will induce an electric displacement D_1 in the piezoelectric ceramic. Since D_1 must remain continuous across fiber-matrix interface, D_1 , as well as E_1 , will be induced in the polymer matrix. Thus both the elastic and dielectric losses in the polymer, which have opposite loss tangents, contribute to the piezoelectric loss associated with e_{15} . It is also observed that there is a peak in storage modulus of e_{33} . This is because $D_3 = e_{33}\epsilon_{33} + e_{11}(\epsilon_{11} + \epsilon_{22})$, where e_{33} is positive and e_{11} is negative. When a far-field ϵ_{33} is applied, ϵ_{11} and ϵ_{22} in the ceramic will have signs opposite to ϵ_{33} due to the Poisson effect, thus the initial addition of polymer will enhance the average D_3 , and leads to a higher effective e_{33} .

B. Laminated composites

Figure 3(a) shows the complex modulus C_{13} of the piezoelectric and elastic laminates, respectively. In the piezoelectric laminate, the storage and loss moduli of C_{13} are smaller than those in the elastic laminate. This is because C_{13} is associated with the far-field strain component ϵ_{33} , which introduces a positive D_3 in the ceramic. Because D_3 is continuous across phase boundaries in the laminate, a positive D_3 is also induced in the matrix by ϵ_{33} , which in turn, induces an electric field in the matrix. A negative E_3 is then induced in the ceramic to maintain the overall zero electric field boundary condition, which thus induces stress σ_{11} in the ceramic: $\sigma_{11} = C_{13}\epsilon_{33} - e_{31}E_3$. Because e_{31} is negative, the stress σ_{11} , and the corresponding storage and loss moduli of C_{13} , are lower in the piezoelectric laminate. This also explains the negative peak in the loss modulus at high volume fractions, since both elastic and dielectric losses in polymer, which have opposite signs, contribute to the loss tangent of C_{13} .

Figure 3(b) shows the complex modulus for the dielectric constant κ_{11} of the piezoelectric and elastic laminates. Due to the laminate configuration, the average electric field E_1 in the elastic composite, polymer matrix, and ceramic fiber are identical. Thus the effective κ_{11} in the elastic composite obeys the rule of mixtures. The storage and loss moduli of the piezoelectric composite, however, both show peaks at high ceramic volume fractions due to the piezoelectric coupling. This is because when a positive electric field E_1 is applied to the composite, negative stresses σ_{13} will be induced in the piezoelectric ceramic, which will again introduce negative σ_{13} in the polymer due to the continuity of traction, and correspondingly a negative strain ϵ_{13} . Thus a positive strain ϵ_{13} must be induced in the piezoelectric ceramic to maintain the overall zero strain boundary condition. This positive strain ϵ_{13} will give extra electric displacement besides the contribution from E_1 , $D_1 = e_{15}\epsilon_{13} + \kappa_{11}E_1$, thus effectively enhancing the effective storage and loss moduli of κ_{11} at high ceramic volume fractions.

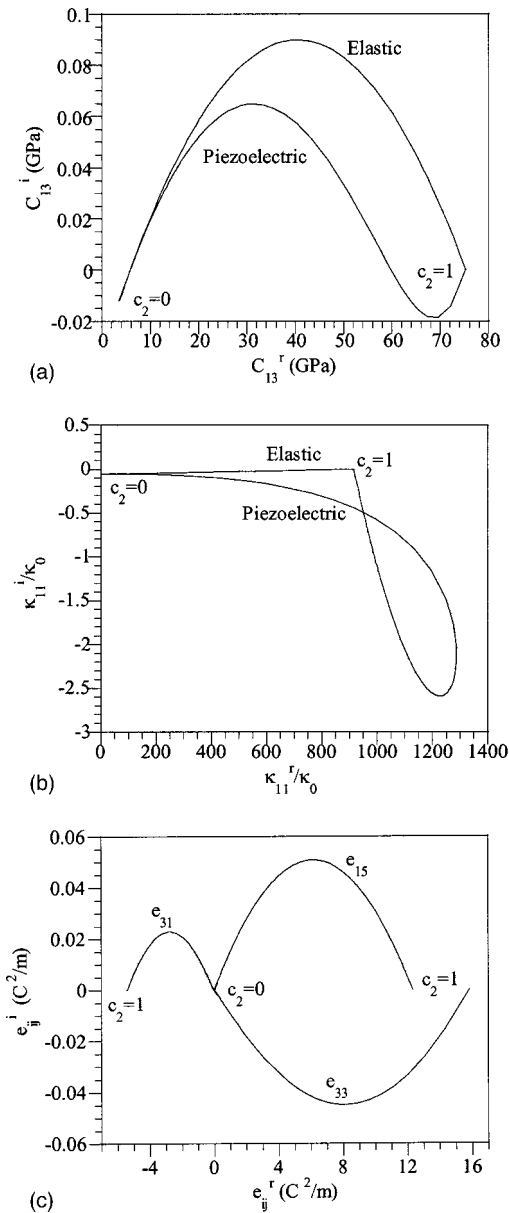


FIG. 3. Storage-loss map of electroelastic moduli of laminated composites: (a) elastic moduli; (b) dielectric modulus; (c) piezoelectric moduli.

Figure 3(c) shows the complex moduli for the piezoelectric constants e_{31} , e_{33} , and e_{15} for piezoelectric laminate. e_{15} has a positive loss tangent because only the elastic loss of the polymer, which has a positive loss tangent, contributes to it. This is because for the laminate configuration, the strain ϵ_{13} is uncoupled from the electric field in the polymer. Both elastic and dielectric losses contribute to the loss moduli of e_{31} and e_{33} , which are apparently dominated by dielectric loss since they both have negative loss tangents.

C. Comparison with experiment

In an attempt to validate the theory to some degree, we fabricated and tested a series of PZT-5A reinforced epoxy laminates with different PZT-5A volume fractions. PZT-5A thin sheets were obtained from Morgan Matroc, Inc., and the epoxy is FM 73 film adhesive provided by CYTEC Engineered Materials, Inc., with Young’s modulus $E_1^r = 2.72$

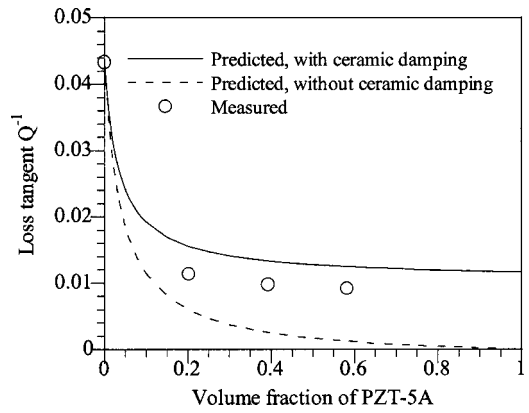


FIG. 4. Loss modulus of E_{11} of PZT-5A reinforced epoxy laminate as a function of PZT-5A volume fraction.

GPa, and $Q^{-1} = E_1^i/E_1^r = 0.043$.⁴⁸ The complex moduli for Young’s modulus E_{11} of the piezoelectric laminates was measured by Ledbetter and Kim,⁴⁸ using a three-component Marx oscillator.^{49,50} Figure 4 shows the loss modulus for Young’s modulus E_{11} of laminate as a function of volume fraction of PZT-5A. The open circles are measurements and the lines are predictions. The dashed line is a prediction for the case where mechanical and dielectric losses of the ceramic phase were assumed to be zero, and only the losses in the polymer phase are considered. In this case the theoretical prediction underestimates the measurements, although it agrees qualitatively. The solid line is a prediction where losses in the ceramic phase are considered, albeit approximately. To effect this calculation, we assumed the loss moduli of the PZT-5A are given by the elastic and piezoelectric loss moduli of Motorola 3203HD PZT (see Table II) reported by Mukherjee and Sherrit,⁵¹ and the dielectric loss moduli of PZT-5A reported by Morgan Matroc.⁵² The elastic loss moduli are scaled by a common factor 0.75 to match

$$Q_M = \frac{\text{Re}(C_{11} - C_{13}^2/C_{33})}{\text{Im}(C_{11} - C_{13}^2/C_{33})} = 75,$$

while the dielectric loss moduli are determined from $Q_E = \kappa_{33}^r/\kappa_{33}^i = 50$, both given by Morgan Matroc⁵² for thin disk PZT-5A. With these considerations, the theoretical predictions show better agreement with experiment, but overestimate the loss tangent of the composite. This is probably because we do not accurately know all the components of the loss moduli of PZT-5A.

TABLE II. Loss moduli used in the calculation for PZT-5A. Units: C_{ij} , GPa; e_{ij} , C^2/m ; κ_0 (permittivity of free space) = $8.85 \times 10^{-12} C^2/Nm^2$.

C_{11}^i	C_{12}^i	C_{13}^i	C_{33}^i	C_{44}^i
0.885	0.061	0.203	0.710	0.634
κ_{11}^i/κ_0	κ_{33}^i/κ_0	e_{31}^i	e_{33}^i	e_{15}^i
-18.3	-16.6	0.574	0.27	-0.291

VI. CONCLUSION

We studied the viscoelectroelastic behavior of heterogeneous piezoelectric solids. Emphasis was placed on the connection between the heterogeneous microstructure and the coupled mechanical and electrical relaxations. We make extensive use of the existence of a correspondence between quasistatic viscoelectroelasticity and static piezoelectricity when linear constitutive response exists, and this correspondence is discussed in detail. We coupled this correspondence principle with micromechanics models to predict the overall behavior of heterogeneous piezoelectric solids in terms of microstructural details. We devoted specific attention to a class of two-phase materials consisting of a lossless piezoelectric phase embedded in a lossy (mechanically and electrically) matrix and obtained closed-form expressions for the effective complex electroelastic moduli. Numerical results are presented and discussed, and qualitative agreement with limited measurements for a piezoelectric laminate were observed.

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