Electrostatic Forces and Stored Energy for Deformable Dielectric Materials

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An isothermal energy balance is formulated for a system consisting of deformable dielectric bodies, electrodes, and the surrounding space. The formulation in this paper is obtained in the electrostatic limit but with the possibility of arbitrarily large deformations of polarizable material. The energy balance recognizes that charges may be driven onto or off of the electrodes, a process accompanied by external electrical work; mechanical loads may be applied to the bodies, thereby doing work through displacements; energy is stored in the material by such features as elasticity of the lattice, piezoelectricity, and dielectric and electrostatic interactions; and nonlinear reversible material behavior such as electrostriction may occur. Thus the external work is balanced by (1) internal energy consisting of stress doing work on strain increments, (2) the energy associated with permeating free space with an electric field, and (3) by the electric field doing work on increments of electric displacement or, equivalently, polarization. For a conservative system, the internal work is stored reversibly in the body and in the underlying and surrounding space. The resulting work statement for a conservative system is considered in the special cases of isotropic deformable dielectrics and piezoelectric materials. We identify the electrostatic stress, which provides measurable information quantifying the electrostatic effects within the system, and find that it is intimately tied to the constitutive formulation for the material and the associated stored energy and cannot be independent of them. The Maxwell stress, which is related to the force exerted by the electric field on charges in the system, cannot be automatically identified with the electrostatic stress and is difficult to measure. Two well-known and one novel formula for the electrostatic stress are identified and related to specific but differing constitutive assumptions for isotropic materials. The electrostatic stress is then obtained for a specific set of assumptions in regard to a piezoelectric material. An exploration of the behavior of an actuator composed of a deformable, electroactive polymer is presented based on the formulation of the paper. [DOI: 10.1115/1.1940661]

Introduction

The subject of electrostatics is the study of the effect of forces generated between charges. Therefore, when electric fields are present simultaneously with mechanical loading in a material, it seems obvious that the influence of electrostatic forces should be accounted for when stresses in the material are calculated. However, stresses due to electrostatic effects are often second order compared to those due to other purely mechanical effects [1] and therefore electrical forces are often neglected even as electric fields are analyzed. An example of this is the linear theory of piezoelectrics [2], where electric fields induce strain but stress due to electrostatic forces, being quadratic in electric field, is neglected. On the other hand, finite strains, nonlinear material behavior, the lack of mechanical loading, and other effects can lead to situations in which electrostatically induced stresses are comparable with forces from other sources. Such cases have been addressed for dielectric materials by several authors, beginning with the pioneering contributions of Toupin [1] and Eringen [3]. Beyond those examples given above, new reasons for the need to include the effect of electrostatically induced loading in the analysis of stress continue to emerge. For example, the analysis of cracks including the effects of cohesive zones requires a consistent treatment of electrically induced forces across cracks [4]. Actuators composed of electrically active polymers are emerging as important devices [5] and these components function due to the electrostatic forces generated in them. Thus, the question of the electrostatic contribution to stress continues to increase in importance.

As noted above, treatments of electrostatic contributions to stress have been provided previously [1,3]. In these papers, a series of electric fields is identified and added together to form the total field. One field is that which would exist if the dielectric material was not present and a second one is due to the effect of polarization charge in the material and on its surface. These two fields together compose the classical Maxwell-Faraday electric field. To this is added what is designated as the local electric field that is considered to be a function, through a constitutive law, of the material's strain and polarization. In addition, the electrostatic forces (i.e., the Maxwell stress [6], whose divergence is the electrical body force and whose surface tractions are the electrical forces per unit area acting on interfaces) are taken to have a specific relationship to the electric field and consequently the formulation is seemingly restricted. Apparently, no allowance is made for the possibility that experiments will show that for different classes of material the electrostatic forces and the Maxwell stress will have some other relationship to the electric field and the polarization than the one assumed. It should be noted that this may not be an insurmountable restriction, since it is well known that there is some arbitrariness in how stress is divided up into electrical and material contributions [1]. Therefore, any discrep-

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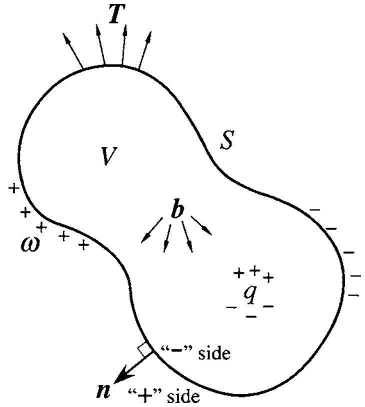

Fig. 1 A dielectric body with body forces, surfaces tractions, and free charges

ancy with experiment as far as the Maxwell stress is concerned may be compensated for by adjustment to the constitutive law for the material stress, presumably leading to nonlinear electrostrictive contributions. However, the structure just described is at the very least inconvenient due to the restrictions on the permitted Maxwell stress and due to the formalities used to construct the electric field.

In the current paper, a different formulation of the problem is presented, though one in which the basic physics is no different from that used in Refs. [1,3]. However, the structure of the electric field is taken to have a simpler form than used in Refs. [1,3] since we make no attempt to identify different contributions to it such as an external field, a depolarization field, or a local field. Instead, a single electric field is utilized throughout and it is assumed to be connected to material strain and polarization by a constitutive law. Furthermore, no general prejudgment is made of the relationship between the electrostatic forces (i.e., the Maxwell stress or alternatively the electrostatic stress) and the electric field and polarization. Instead, it is assumed that the electrostatic stress is measurable in experiments either directly through characterization of stress and electric fields or through measurement of the constitutive properties of the material. We note that our approach parallels that used by Landau and Lifschitz [6] but we do not restrict ourselves to infinitesimal elastic strains of isotropic materials and piezoelectric materials. It is our belief that this formulation is more versatile than what has been presented in the past and that it is more suitable for incorporation into modern methods such as finite element analysis.

Balance of Energy for a Deformable Dielectric Material

Consider a system consisting of dielectric materials, perfect conductors, and free space. In the current configuration, the system occupies the volume V as shown in Fig. 1. The system is considered to be isolated so that there is no interaction between electrical charges residing in the system and those outside. Formally, this implies that the volume of interest is shielded electrically from its exterior or that the extent of the system is infinite, since charges interact with each other over very long distances. However, when practical calculations with approximations are attempted, this formality can be ignored. The perimeter of the system plus interfaces within it are designated S in the current configuration. The internal interfaces separate the dielectric materials, the conductors, and free space from each other. In addition, sectors of dielectric with homogeneous or heterogeneous properties may be separated by surfaces included within S, as may sectors of free space.

Let the free charge per unit volume within V be $q(x_i,t)$ where x_i is the position of material points in the current configuration and t is time. Free charge may be placed in free space, in which case x_i is used to designate the position of the points occupied by free space in the current configuration. Furthermore, let $\omega(x_i,t)$ be the free charge per unit area on the surfaces S and define $\phi(x_i,t)$ to be the electrical potential everywhere within the system such that it is continuous everywhere in space. Note that we will consider only the electrostatic limit so that t plays the role of a load parameter as far as the electrical variables are concerned and no attempt will be made to explore Maxwell's equations relevant to the electrodynamic limit. On the other hand, time may have a real meaning as far as the deformation of material is concerned, as may be the case in strain-rate-dependent response or in the acoustic limit where inertia has to be included in the balance laws. To complete the variables to be considered in the external work statement, we include the velocity $v_i(x_i,t)$ of material points, the surface traction $T_i(x_i,t)$ defined as the force per unit area acting on S, and $b_i(x_i,t)$, which is the body force per unit volume acting at points in V. Note that the surface traction T_i and the body force b_i arise from sources other than electrical effects and, therefore, do not represent the influence of charges interacting at a distance or electrical fields exerting forces on charges. The surface traction T_i and the body force b_i and any equivalent quantity defined in the current state will be designated mechanical, though we do not make any attempt to characterize how they may arise, whether they come about by gravitational effects or other sources of force in materials. To attempt to do so in too fine a detail and make distinctions between forces that arise by electrostatic effects and forces that arise from other effects would illuminate the arbitrariness in how the designations electrical and mechanical are utilized in our scheme; e.g., consider a purely ionic solid pressing against another purely ionic solid to produce supposedly mechanical traction between them. Much of the traction between the two solids in this case will in fact be electrostatic due to the repulsion of like atoms, although some of it will also develop due to quantum exclusion effects. However, in any macroscopic treatment of this problem, this interaction between the bodies when pressed together will be represented by elasticity and the tractions thereby designated to be mechanical.

Consider the physical laws governing the electromechanical fields in the material. In the quasi-static limit, Maxwell's laws state that the electric field must be curl-free and Gauss' law states that the divergence of the electric displacement must be equal to the volume density of free charge. Therefore,

$$\in_{ijk}\frac{\partial E_j}{\partial x_k}=0 \Rightarrow E_i=-\frac{\partial \phi}{\partial x_i} \tag{1}$$

$$\frac{\partial D_i}{\partial x_i}=q \text{ in } V \tag{2}$$

$$\|D_i n_i\|=\omega \text{ on } S \tag{3}$$

Here, n_i are the Cartesian components of the unit normal to the surface S pointing from the "$-$" side of the surface out towards the "$+$" side as shown in Fig. 1, and $\in_{ijk}$ are the components of the permutation symbol. Then the notation $\|\ \|$ represents the difference or jump in the included quantity across the surface S such that

$$\|D_i\|=D_i^+-D_i^- \tag{4}$$

Furthermore, the electric displacement can be decomposed into two parts such that

$$D_i=\kappa_0 E_i+P_i \tag{5}$$

where κ_0 is the dielectric permittivity of free space and P_i are the Cartesian components of the material polarization.

Conservation of mass implies that for a given material volume

$$\frac{\mathrm{d}}{\mathrm{d}t}\int_V \rho \mathrm{d}V = 0 \Rightarrow \frac{\mathrm{d}\rho}{\mathrm{d}t} + \rho\frac{\partial v_i}{\partial x_i} = 0 \tag{6}$$

where $\rho(x_i,t)$ is the mass density of the material. The principles of conservation of linear and angular momentum are stated as

$$\int_V (b_i + b_i^E)\mathrm{d}V + \int_S (T_i + T_i^E)\mathrm{d}S = \frac{\mathrm{d}}{\mathrm{d}t}\int_V \rho v_i \mathrm{d}V \tag{7}$$

and

$$\int_V \in_{ijk} x_j(b_k + b_k^E)\mathrm{d}V + \int_S \in_{ijk} x_j(T_k + T_k^E)\mathrm{d}S = \frac{\mathrm{d}}{\mathrm{d}t}\int_V \rho \in_{ijk} x_j v_k \mathrm{d}V \tag{8}$$

Here the components of the electrical body force b_i^E and surface traction T_i^E have been introduced. These forces arise directly from electric fields acting in the material and are in addition to the mechanical body force and surface traction. Furthermore, it is assumed that the electrical body force can be derived from the Maxwell stress tensor σ_{ij}^M such that

$$b_i^E = \frac{\partial \sigma_{ji}^M}{\partial x_j} \text{ in } V \tag{9}$$

where the electrical body force is the effect of charges interacting at a distance or, equivalently, the force per unit volume arising from electric fields acting on charges. The traction relationship for Maxwell stress is then

$$T_i^E = n_j\|\sigma_{ji}^M\| \text{ on } S. \tag{10}$$

Then, in order to satisfy the principle of conservation of linear momentum for a small surface element, the Cauchy stress in the material, σ_{ij}, must balance the total surface traction such that along with Eq. (10)

$$T_i + T_i^E = -n_j\|\sigma_{ji}\| \Rightarrow T_i = -n_j\|\sigma_{ji} + \sigma_{ji}^M\| \tag{11}$$

where again $T_i(x_i,t)$ is the nonelectrical (i.e., mechanical) surface force per unit area acting on S. This statement has an equivalent meaning to Eq. (10). The Cauchy stress difference across a surface must balance both the electrical and mechanical surface tractions. Note, however, that the result in Eq. (11) illustrates why it is difficult, if not impossible, to separately measure the Cauchy and Maxwell stresses, since it shows that any traction measured by mechanical means (i.e., by the only method available) is related to their sum. Since there are no experiments that can separate the effects of the Cauchy and Maxwell stresses unambiguously [1,3,6], it is generally more profitable to consider their sum and not to try to identify them separately. The sum will be termed the total true stress.

Next, application of Eqs. (6), (9), and (11) within the principles of conservation of linear and angular momentum and recognition that the resultant integrals must be valid for any arbitrary volume yield

$$\frac{\partial \sigma_{ji}}{\partial x_j} + \frac{\partial \sigma_{ji}^M}{\partial x_j} + b_i = \rho\frac{\mathrm{d}v_i}{\mathrm{d}t} \text{ in } V \tag{12}$$

and

$$\sigma_{ji} + \sigma_{ji}^M = \sigma_{ij} + \sigma_{ij}^M \text{ in } V \tag{13}$$

Thus, for the balance of angular momentum to be satisfied, the total true stress must be symmetric. This requirement reflects the fact that moments due to mechanical body forces and inertia can be assumed, as usual, to be second order [7,8], but we must allow for the possibility that electric effects induce first-order moments, e.g., due to electric fields acting on dipoles in the material [1,3]. Since only the total true stress in the material must be symmetric, it is possible that both the Maxwell and Cauchy stress tensors can be nonsymmetric.

Now consider the rate of work by agencies external to the system. This external work rate is

$$\frac{\mathrm{d}W}{\mathrm{d}t} = \int_V b_i v_i \mathrm{d}V + \int_S T_i v_i \mathrm{d}S + \int_V \phi\frac{\mathrm{d}}{\mathrm{d}t}(q\mathrm{d}V) + \int_S \phi\frac{\mathrm{d}}{\mathrm{d}t}(\omega \mathrm{d}S) \tag{14}$$

Note that this statement conforms to the usual definition of electrical work, i.e., an increment of electrical work is given by the electric potential ϕ multiplied by the increment of charge ΔQ. For example, $\phi\Delta(q\mathrm{d}V)$ is the work done by external agencies to bring the charge $\Delta(q\mathrm{d}V)$ from infinity to the point where the potential is ϕ. The interpretation of the mechanical work done by the body forces and surface tractions is obvious.

Based on the standard continuum mechanics results for the material time derivatives of field quantities and volume and surface elements [7, pp. 211–213], it can be shown that

$$\frac{\mathrm{d}}{\mathrm{d}t}(q\mathrm{d}V) = \left(\frac{\mathrm{d}q}{\mathrm{d}t} + q\frac{\partial v_k}{\partial x_k}\right)\mathrm{d}V = \left(\frac{\partial q}{\partial t} + v_k\frac{\partial q}{\partial x_k} + q\frac{\partial v_k}{\partial x_k}\right)\mathrm{d}V \tag{15}$$

and

$$\frac{\mathrm{d}}{\mathrm{d}t}(\|D_i n_i\|\mathrm{d}S) = \left(\left\|\frac{\mathrm{d}D_i}{\mathrm{d}t} + D_i\frac{\partial v_k}{\partial x_k} - D_j\frac{\partial v_i}{\partial x_j}\right\| n_i\right)\mathrm{d}S \tag{16}$$

Then, applying these results along with the balance of linear momentum, Eqs. (11) and (12), we can write the work rate as

$$\begin{aligned}\frac{\mathrm{d}W}{\mathrm{d}t} = &\int_S \phi n_i\left\|\frac{\mathrm{d}D_i}{\mathrm{d}t} + D_i\frac{\partial v_k}{\partial x_k} - \frac{\partial v_i}{\partial x_j}D_j\right\|\mathrm{d}S + \int_V \phi\left(\frac{\partial q}{\partial t} + v_k\frac{\partial q}{\partial x_k}\right. \\ &\left. + q\frac{\partial v_k}{\partial x_k}\right)\mathrm{d}V - \int_S n_j\|\sigma_{ji} + \sigma_{ji}^M\| v_i \mathrm{d}S - \int_V \left(\frac{\partial \sigma_{ji}}{\partial x_j} + \frac{\partial \sigma_{ji}^M}{\partial x_j}\right)v_i \mathrm{d}V \\ &+ \int_V \rho\frac{\mathrm{d}v_i}{\mathrm{d}t}v_i \mathrm{d}t\end{aligned} \tag{17}$$

Use of the divergence theorem for a collection of subvolumes whose union is V and which are separated by the interfaces that collectively compose S gives

$$\begin{aligned}\frac{\mathrm{d}W}{\mathrm{d}t} = &-\int_V \frac{\partial}{\partial x_i}\left[\phi\left(\frac{\mathrm{d}D_i}{\mathrm{d}t} + D_i\frac{\partial v_k}{\partial x_k} - \frac{\partial v_i}{\partial x_j}D_j\right)\right]\mathrm{d}V \\ &+ \int_V \phi\left(\frac{\partial^2 D_i}{\partial t\partial x_i} + v_k\frac{\partial^2 D_i}{\partial x_k \partial x_i} + \frac{\partial D_i}{\partial x_i}\frac{\partial v_k}{\partial x_k}\right)\mathrm{d}V \\ &+ \int_V \frac{\partial}{\partial x_j}[(\sigma_{ji} + \sigma_{ji}^M)v_i]\mathrm{d}V - \int_V \left(\frac{\partial \sigma_{ji}}{\partial x_j} + \frac{\partial \sigma_{ji}^M}{\partial x_j}\right)v_i \mathrm{d}V \\ &+ \int_V \rho\frac{\mathrm{d}v_i}{\mathrm{d}t}v_i \mathrm{d}V\end{aligned} \tag{18}$$

where Eq. (2) has been used to dispose of the free charge density q. Equation (18) simplifies to

$$\begin{aligned}\frac{\mathrm{d}W}{\mathrm{d}t} = &\int_V \left[E_i\frac{\mathrm{d}D_i}{\mathrm{d}t} + (\sigma_{ji} + \sigma_{ji}^M - E_i D_j + E_k D_k \delta_{ij})\frac{\partial v_i}{\partial x_j}\right]\mathrm{d}V \\ &+ \int_V \rho\frac{\mathrm{d}v_i}{\mathrm{d}t}v_i \mathrm{d}V\end{aligned} \tag{19}$$

where δ_{ij} is the Kronecker delta.

Note that in regions of space without material, the electric displacement is defined to be [6]

$$D_i = \kappa_o E_i \tag{20}$$

where κ_o is the permittivity of free space. In addition, the Maxwell stress in such regions is [6]

$$\sigma_{ij}^M = \kappa_o\left(E_iE_j - \tfrac{1}{2}E_kE_k\delta_{ij}\right) \tag{21}$$

so that from Eqs. (2), (9), and (20), we find that the electrical force per unit volume is

$$b_i^E = qE_i \tag{22}$$

which is consistent with the standard result that the force on a charge is given by the charge times the electric field.

Now we recognize the material polarization P_i and use Eq. (5) to replace D_i wherever it appears in Eq. (19) to obtain

$$\frac{\mathrm{d}W}{\mathrm{d}t} = \frac{\mathrm{d}}{\mathrm{d}t}\int_V \frac{1}{2}\kappa_o E_iE_i\mathrm{d}V + \frac{\mathrm{d}}{\mathrm{d}t}\int_V \frac{1}{2}\rho v_iv_i\mathrm{d}V + \int_V\left[E_i\frac{\mathrm{d}P_i}{\mathrm{d}t} + (\sigma_{ji} + \sigma_{ji}^M - \hat{\sigma}_{ji}^M - E_iP_j + E_kP_k\delta_{ij})\frac{\partial v_i}{\partial x_j}\right]\mathrm{d}V \tag{23}$$

where

$$\hat{\sigma}_{ij}^M = \kappa_o\left(E_iE_j - \tfrac{1}{2}E_kE_k\delta_{ij}\right) \tag{24}$$

is thus the Maxwell stress at the specified electric field for free space absent any material and the second integral on the right-hand side has been obtained from the second one on the right of Eq. (19) by use of the first form of conservation of mass, Eq. (6). In the derivation of Eq. (23), the fact that

$$\kappa_o\int_V\left[E_i\frac{\mathrm{dE}_i}{\mathrm{d}t} - (E_iE_j - E_kE_k\delta_{ij})\frac{\partial v_i}{\partial x_j}\right]\mathrm{d}V = \int_V\left[\kappa_oE_i\frac{\partial E_i}{\partial t} - \hat{\sigma}_{ij}^M\frac{\partial v_i}{\partial x_j} + \frac{\partial}{\partial x_k}\left(\frac{1}{2}\kappa_oE_iE_iv_k\right)\right]\mathrm{d}V \tag{25}$$

has been used and the divergence theorem then provides

$$\int_V\left[\kappa_oE_i\frac{\partial E_i}{\partial t} + \frac{\partial}{\partial x_k}\left(\frac{1}{2}\kappa_0E_iE_iv_k\right)\right]\mathrm{d}V = \int_V \kappa_oE_i\frac{\partial E_i}{\partial t}\mathrm{d}V - \int_S \frac{1}{2}\kappa_o\|E_iE_i\|n_kv_k\mathrm{d}S \tag{26}$$

The terms on the right-hand side of Eq. (26) combine to give the first term on the right-hand side of Eq. (23). Note that in regions of free space without material, the second and third integrals on the right-hand side of Eq. (23) are both zero (given that the Cauchy stress is zero there), indicating that the first integral gives the rate of energy storage in space to permeate it with the electric field. Thus the third integral on the right-hand side of Eq. (23) is the rate at which work is stored or dissipated in the material, other than kinetic energy, which is, of course, accounted for by the first integral.

Finally, we note that Eq. (23) can be converted to a principle of virtual work; however, the details will not be emphasized here. Equation (23) is valid for the isothermal response of any electromechanical material whether the behavior is reversible or dissipative. However, the next section will focus attention on reversible material response.

Conservative Materials

In this section we formally introduce the first and second laws of thermodynamics for the electromechanical situation under consideration. Ultimately, we will specialize the results to reversible material behavior, i.e., conservative materials. In conservative materials, the work done by external agencies that is not absorbed by kinetic energy is stored in the material in the form of elastic distortion, dielectric polarization, piezoelectric response, electrostrictive behavior, electrostatic interactions, and any other recoverable energy storage mechanism that is active. The balance of energy required by the first law of thermodynamics can be stated as

$$\frac{\mathrm{d}U}{\mathrm{d}t} = \frac{\mathrm{d}W}{\mathrm{d}t} + \frac{\mathrm{d}Q}{\mathrm{d}t} \tag{27}$$

where U represents the stored internal energy of the material, the electrical energy stored by free space, and the kinetic energy of the material; $\mathrm{d}W/\mathrm{d}t$ is the external work rate derived in Eq. (14), and $\mathrm{d}Q/\mathrm{d}t$ is the rate at which heat is transferred to the system. Note that the free space in which energy is stored is not just that occupied by the body or bodies under consideration. Material subject to electric fields couple with the surrounding aether and permeate it with an electrical field. Thus, the free space being considered includes any surrounding space affected by the electric fields associated with the body or bodies. Specifically, $\mathrm{d}U/\mathrm{d}t$ and $\mathrm{d}Q/\mathrm{d}t$ are written as

$$\frac{\mathrm{d}U}{\mathrm{d}t} = \frac{\mathrm{d}}{\mathrm{d}t}\int_V \rho u\mathrm{d}V + \frac{\mathrm{d}}{\mathrm{d}t}\int_V \frac{1}{2}\kappa_oE_iE_i\mathrm{d}V + \frac{\mathrm{d}}{\mathrm{d}t}\int_V \frac{1}{2}\rho v_iv_i\mathrm{d}V \tag{28}$$

and

$$\frac{\mathrm{d}Q}{\mathrm{d}t} = \int_V \rho\dot{r}\mathrm{d}V - \int_{S_e}\hat{q}_in_i\mathrm{d}S \tag{29}$$

where u is the internal energy per unit mass, $\dot{r}$ is the heat input rate per unit mass, and $\hat{q}_i$ are the components of the heat flux vector with the positive sense directed out of the external surface S_e of the system. Note that at all points interior to the system the heat flux vector is taken to be continuous, eliminating the possibility of surfaces acting as sources of heat. Equations (23) and (27)–(29) can be combined and must hold for any arbitrary volume yielding a local form for the first law as

$$\rho\frac{\mathrm{d}u}{\mathrm{d}t} = (\sigma_{ji} + \sigma_{ji}^M - \hat{\sigma}_{ji}^M - E_iP_j + E_kP_k\delta_{ij})\frac{\partial v_i}{\partial x_j} + E_i\frac{\mathrm{d}P_i}{\mathrm{d}t} + \rho\dot{r} - \frac{\partial\hat{q}_i}{\partial x_i} \tag{30}$$

The second law of thermodynamics states that the entropy production rate must be equal to or exceed the rate of entropy input to a region, i.e.,

$$\frac{\mathrm{d}}{\mathrm{d}t}\int_V \rho s\mathrm{d}V \geqslant \int_V \frac{\rho\dot{r}}{\theta}\mathrm{d}V - \int_{S_e}\frac{\hat{q}_in_i}{\theta}\mathrm{d}S \tag{31}$$

where s is the entropy per unit mass of the material and θ is the absolute temperature. Equation (31) can be manipulated by application of the divergence theorem to the last term on the right-hand side and then required to be valid for any arbitrary volume to yield a local form of the second law as

$$\rho\frac{\mathrm{d}s}{\mathrm{d}t} \geqslant \frac{\rho\dot{r}}{\theta} - \frac{1}{\theta}\frac{\partial\hat{q}_i}{\partial x_i} + \frac{\hat{q}_i}{\theta^2}\frac{\partial\theta}{\partial x_i} \tag{32}$$

Then, defining ψ as the Helmholtz free energy per unit mass, we have

$$\psi = u - \theta s \tag{33}$$

and in combination with Eq. (30)–(33), Eq. (32) can be rewritten as

$$(\sigma_{ji} + \sigma_{ji}^M - \hat{\sigma}_{ji}^M - E_iP_j + E_kP_k\delta_{ij})\frac{\partial v_i}{\partial x_j} + E_i\frac{\mathrm{d}P_i}{\mathrm{d}t} - \rho s\frac{\mathrm{d}\theta}{\mathrm{d}t} - \rho\frac{\mathrm{d}\psi}{\mathrm{d}t} - \frac{\hat{q}_i}{\theta}\frac{\partial\theta}{\partial x_i} \geqslant 0 \tag{34}$$

For a conservative electro-active material it is assumed that in general ψ is a function of the deformation gradient, polarization, and temperature, i.e., $\psi = \psi(F_{ij}, P_i, \theta)$. Note that for material points, x_i designates their positions in the current configuration and a mapping

$$x_i = x_i(X_j,t) \tag{35}$$

associates these points at time t with their positions X_i in the reference configuration [7]. The components of the deformation gradient are given as

$$F_{ij} = \frac{\partial x_i}{\partial X_j} \tag{36}$$

Then, using

$$\frac{\mathrm{d}F_{ik}}{\mathrm{d}t} = \frac{\partial x_j}{\partial X_k}\frac{\partial v_i}{\partial x_j} \tag{37}$$

we can modify Eq. (34) to

$$\left[(\sigma_{ji} + \sigma^M_{ji} - \hat{\sigma}^M_{ji} - E_iP_j + E_kP_k\delta_{ij}) - \rho\frac{\partial\psi}{\partial F_{ik}}F_{jk}\right]\frac{\partial v_i}{\partial x_j} + \left[E_i - \rho\frac{\partial\psi}{\partial P_i}\right]\frac{\mathrm{d}P_i}{\mathrm{d}t} - \left[\rho s + \rho\frac{\partial\psi}{\partial\theta}\right]\frac{\mathrm{d}\theta}{\mathrm{d}t} - \frac{\hat{q}_i}{\theta}\frac{\partial\theta}{\partial x_i} \geqslant 0 \tag{38}$$

Following the methods of Coleman and Noll [9], we postulate that (38) must hold for every admissible process. First, consider processes where the deformation gradient, polarization, and temperature are spatially homogeneous but arbitrary functions of time. Such processes can be thought as being controlled by the appropriate applications of body forces, charge densities, and heat supply. For spatially homogeneous temperature distributions the last term on the left-hand side of (38) vanishes. If $\mathrm{d}P_i/\mathrm{d}t$ and $\mathrm{d}\theta/\mathrm{d}t$ are taken to be zero and $\partial v_i/\partial x_j$ is arbitrarily chosen, then (38) is satisfied only if

$$\sigma_{ji} + \sigma^M_{ji} - \hat{\sigma}^M_{ji} - E_iP_j + E_kP_k\delta_{ij} = \rho\frac{\partial\psi}{\partial F_{ik}}F_{jk} \tag{39}$$

If $\mathrm{d}\theta/\mathrm{d}t$ and $\partial v_i/\partial x_j$ are zero and $\mathrm{d}P_i/\mathrm{d}t$ is chosen arbitrarily, then (38) implies

$$E_i = \rho\frac{\partial\psi}{\partial P_i} \tag{40}$$

If $\partial v_i/\partial x_j$ and $\mathrm{d}P_i/\mathrm{d}t$ are zero and $\mathrm{d}\theta/\mathrm{d}t$ is arbitrarily chosen, then (38) yields

$$s = -\frac{\partial\psi}{\partial\theta} \tag{41}$$

Finally, consider spatially homogeneous and time-independent distributions of the deformation gradient and polarization, and spatially inhomogeneous but time-independent distributions of temperature. Such processes and Eq. (38) then imply the heat conduction inequality

$$-\hat{q}_i\frac{\partial\theta}{\partial x_i} \geqslant 0 \tag{42}$$

The requirements of objectivity [7] place restrictions on the forms that the constitutive laws can take. This leads us to conclude that the Helmholtz free energy per unit mass at fixed temperature must have the form

$$\psi = \psi(U_{ij},\Pi_i) \tag{43}$$

where Π_i are the components of the rotation invariant polarization defined as

$$\Pi_i = P_jR_{ji} \tag{44}$$

where R_{ji} is the orthogonal transformation arising from polar decomposition of F_{ij} into a pure deformation and a pure rotation [7] as

$$F_{ij} = R_{ik}U_{kj} \tag{45}$$

Finally, U_{ij} are the components of the right stretch tensor.

Note that the choice we have made for the functional dependence of ψ in terms of a polarization measure is not the only one possible that would satisfy objectivity. However, we find it to be of the most convenience. In general, to satisfy the requirements of objectivity, the Helmholtz free energy density can depend on any measures of polarization and strain that in turn depend only on the rotation-invariant polarization and the right stretch tensor. Since their evaluation does not require the solution of an eigenvalue problem, it is common to define the relevant polarization measure as $\mathbb{P}_i=\Pi_jU_{ji}=P_kF_{ki}$ and the strain measure as $\varepsilon_{ij}=(U_{ik}U_{kj}-\delta_{ij})/2=(F_{ki}F_{kj}-\delta_{ij})/2$. However, the form $\mathbb{P}_i$ for the polarization measure is somewhat convoluted since it is affected by both electrical polarization *and* the stretch of the material element. Hence, for the purposes of this work the polarization measure will be taken as the rotation invariant polarization Π_i, which is independent of the material stretch, and the strain measure will be taken as the Green-Lagrange strain ε_{ij} mentioned above and repeated below as

$$\varepsilon_{ij} = \tfrac{1}{2}(F_{ki}F_{kj} - \delta_{ij}) \tag{46}$$

Using these polarization and strain measures, we find more convenient forms of Eqs. (39) and (40) to be

$$E_i = \rho R_{ij}\frac{\partial\psi}{\partial\Pi_j} \tag{47}$$

$$\sigma_{ji} + \sigma^M_{ji} = \rho\frac{\partial\psi}{\partial\varepsilon_{lm}}F_{il}F_{jm} + \beta_{jikl}P_kE_l + P_jE_i - P_kE_k\delta_{ij} + \hat{\sigma}^M_{ji} \tag{48}$$

where

$$\beta_{jikl} = F_{jm}\frac{\partial R_{kn}}{\partial F_{im}}R_{ln} \tag{49}$$

The derivative of the rotation tensor is given by [10]

$$\frac{\partial R_{ik}}{\partial F_{jl}} = \frac{1}{\nu}[(V_{mm}\delta_{ij} - V_{ij})(U_{nn}\delta_{kl} - U_{kl}) - (V_{mm}R_{il} - F_{il})(V_{nn}R_{jk} - F_{jk})] \tag{50}$$

with

$$\nu = \mathrm{Det}(V_{kk}\delta_{ij} - V_{ij}) = \mathrm{Det}(U_{kk}\delta_{ij} - U_{ij}) \tag{51}$$

in which V_{ij} is the left stretch tensor given by a polar decomposition in the form [7]

$$F_{ij} = V_{ik}R_{kj} \tag{52}$$

Thus

$$\beta_{jikl} = \frac{1}{\nu}V_{jm}(V_{nn}\delta_{mp} - V_{mp})(V_{qq}\delta_{ir} - V_{ir})(\delta_{rk}\delta_{pl} - \delta_{rl}\delta_{pk}) \tag{53}$$

Furthermore, it can be shown that [10]

$$\beta_{jikl}P_kE_l + P_jE_i = \beta_{ijkl}P_kE_l + P_iE_j \tag{54}$$

and this confirms that the total true stress $\sigma_{ji}+\sigma^M_{ji}$ is symmetric as required by conservation of angular momentum. In fact, as long as the free energy density is objective, then the law of conservation of angular momentum, Eqs. (8) and (13), will be satisfied automatically [1].

We note that Eqs. (47) and (48) are together equivalent to concepts developed by Landau and Lifschitz [6] who studied these issues for infinitesimally strained isotropic elastic materials and piezoelectric systems through the use of a free energy. We believe that we have therefore placed these ideas into a more general framework.

Dielectrically Linear, Isotropic, Conservative Materials

Consider now a material in which the stored energy per unit reference volume is the sum of an isotropic function of the deformation plus a term quadratric in the polarization. Thus

$$\psi = \psi_e + \frac{1}{2\rho\kappa_0}\Pi_i\chi_{ij}^{-1}\Pi_j \tag{55}$$

where ψ_e depends only on the deformation and χ_{ij} is the electrical susceptibility of the material, which is assumed to be dependent on the strain but independent of polarization. Since the material is assumed to be isotropic, ψ_e and χ_{ij} will be functions of the invariants of the Green-Lagrange strain or, equivalently, the right or left stretch tensors [7]. Possible forms of interest for the elastic response would include neo-Hookean, Mooney-Rivlin, Blatz-Ko, and Ogden formulations [7,11–16], but allowance should be made for compressibility of the material to ensure consistency with any dilatancy that is assumed in association with the electrical behavior. The susceptibility tensor is symmetric and must be isotropic in the limit of zero strain so that the unstrained material is electrically isotropic. Due to strain χ_{ij} can become anisotropic [6].

The electric field can now be deduced from Eq. (47) to be

$$E_i = \frac{1}{\kappa_0}R_{ij}\chi_{jk}^{-1}P_lR_{lk} \tag{56}$$

which is thus a linear relationship between the components of electric field and polarization modified by the effects of straining and rotation of the material. Note that in the limit of zero strain, this relationship is consistent with what is expected for isotropic dielectrics since then the susceptibility is

$$\chi_{ij}^{-1} = \frac{1}{\tilde{\chi}}\delta_{ij} \tag{57}$$

where $\tilde{\chi}$ is the isotropic susceptibility of the unstrained material. The relationship in Eq. (56) then becomes

$$E_i = \frac{1}{\kappa_o\tilde{\chi}}P_i \tag{58}$$

The result for stress obtained from Eq. (48) is

$$\sigma_{ji} + \sigma_{ji}^M = F_{jk}\left(\rho\frac{\partial\psi_e}{\partial\varepsilon_{kl}} - \frac{\kappa_o}{2}R_{mn}\frac{\partial\chi_{np}}{\partial\varepsilon_{kl}}R_{qp}E_mE_q\right)F_{il} + \kappa_oR_{jk}\chi_{kl}R_{ml}E_mE_i - \frac{\kappa_o}{2}R_{kl}\chi_{lm}R_{nm}E_kE_n\delta_{ji} + \kappa_o\beta_{jikl}R_{km}\chi_{mn}R_{pn}E_lE_p + \hat{\sigma}_{ji}^M \tag{59}$$

where

$$\frac{\partial\rho}{\partial\varepsilon_{lm}}F_{il}F_{jm} = -\rho\delta_{ij} \tag{60}$$

has been utilized and the inverse of Eq. (56) has been used to eliminate the polarization. In Eq. (59), the term containing the derivatives of ψ_e can be considered to be the elastic stress and the remainder of the right-hand side of the expression can be taken to be the electrostatic stress as in the usage of Landau and Lifschitz [6]. However, this does not imply that the Maxwell stress and the electrostatic stress are identical, since the Cauchy stress, σ_{ij}, can have a constitutive relationship that allows it to depend quadratically on the polarization independently of whatever electrical behavior is associated with the Maxwell stress. Because of the difficulty of measuring the Cauchy and Maxwell stresses separately, it is probably impossible to resolve this question experimentally. Therefore, we will focus on the elastic stress

$$\sigma_{ji}^e = F_{jk}\rho\frac{\partial\psi_e}{\partial\varepsilon_{kl}}F_{il} \tag{61}$$

and the electrostatic stress

$$\sigma_{ji}^{es} = -\frac{\kappa_o}{2}F_{jk}R_{mn}\frac{\partial\chi_{np}}{\partial\varepsilon_{kl}}R_{qp}F_{il}E_mE_q + \kappa_0\beta_{jikl}R_{km}\chi_{mn}R_{pn}E_lE_p + \kappa_oR_{jk}\chi_{kl}R_{ml}E_iE_m - \frac{\kappa_o}{2}R_{kl}\chi_{lm}R_{nm}E_kE_n\delta_{ji} + \hat{\sigma}_{ji}^M \tag{62}$$

without attempting to separately identify the Cauchy and Maxwell stresses. Of course, the sum of the elastic and electrostatic stresses is equal to the sum of the Cauchy and Maxwell stresses and both sums are equal to the total true stress.

Linear, Isotropic, Conservative Materials at Infinitesimal Strain

For illustration, now consider the case where the strain is infinitesimal, so that to first order in the infinitesimal strains, e_{ij}, the susceptibility can be written [6]

$$\chi_{ij} = \tilde{\chi}\delta_{ij} + \frac{\kappa_1}{\kappa_o}e_{ij} + \frac{\kappa_2}{\kappa_o}e_{kk}\delta_{ij} \tag{63}$$

where κ_i are constants. With terms of higher order in strain neglected, the expression for the electric field in Eq. (56) becomes identical to Eq. (58), which is the usual relationship for isotropic dielectrics. When the strains are infinitesimal and higher order terms in strain are neglected, Eq. (59) shows that the total stress is given by

$$\sigma_{ij} + \sigma_{ij}^M = 2Ge_{ij} + \left(B - \frac{2G}{3}\right)e_{kk}\delta_{ij} + \left(\tilde{\kappa} - \frac{\kappa_1}{2}\right)E_iE_j - \frac{1}{2}(\tilde{\kappa} + \kappa_2)E_kE_k\delta_{ij} \tag{64}$$

as identified for this situation by Landau and Lifschitz [6]. In Eq. (64), G is the shear modulus, B is the bulk modulus, and $\tilde{\kappa}$ is the dielectric permittivity of the unstrained material such that

$$\tilde{\kappa} = (1 + \tilde{\chi})\kappa_o \tag{65}$$

so that the relationship between the electric displacement and the electric field during infinitesimal straining is given by

$$D_i = \tilde{\kappa}E_i \tag{66}$$

Note that, as observed by Landau and Lifschitz [6], the resulting total stress in Eq. (64) is symmetric and therefore the conservation of angular momentum is satisfied. Furthermore, consistent with the usage of Landau and Lifschitz [6], the elastic stress is

$$\sigma_{ij}^e = 2Ge_{ij} + \left(B - \frac{2G}{3}\right)e_{kk}\delta_{ij} \tag{67}$$

and the electrostatic stress is

$$\sigma_{ij}^{es} = \left(\tilde{\kappa} - \frac{\kappa_1}{2}\right)E_iE_j - \frac{1}{2}(\tilde{\kappa} + \kappa_2)E_kE_k\delta_{ij} \tag{68}$$

However, there is no implication that the electrostatic stress and the Maxwell stress are identical.

Conservative Materials that Remain Dielectrically Isotropic During Straining

Now return to the general case of arbitrarily large deformations but assume that, upon straining, the susceptibility remains isotropic and thus objectivity is assured. The susceptibility will be permitted to depend on the material density and thus is given by

$$\chi_{ij} = \chi(\rho)\delta_{ij} \tag{69}$$

Note that the material density obeys $\rho = \rho_o/\mathrm{Det}(U_{ij})$, where ρ_o is the density of the material in the reference state. Thus the dependence of the susceptibility on the density represents a contribution to the dependence of the Helmholtz free energy on the right stretch tensor, as stated in Eq. (43). Given Eq. (69), Eq. (56) then simplifies to

$$E_i = \frac{P_i}{\kappa_0 \chi(\rho)} \tag{70}$$

The total true stress from Eq. (59) is then given by

$$\sigma_{ij} + \sigma_{ij}^M = F_{ik}\frac{1}{J}\frac{\partial \psi_e}{\partial \varepsilon_{kl}}F_{jl} + \kappa\left(E_iE_j - \frac{1}{2}E_kE_k\delta_{ij}\right) + \frac{1}{2}\rho\frac{d\chi}{d\rho}\kappa_0 E_kE_k\delta_{ij} \tag{71}$$

where κ is the possibly density-dependent dielectric permittivity given by

$$\kappa = (1+\chi)\kappa_o \tag{72}$$

so that

$$D_i = \kappa E_i \tag{73}$$

To obtain Eq. (71), use was made of the fact that $\beta_{ijkl}E_kE_l=0$. As expected, the total true stress is symmetric, reflecting the fact that the balance of angular momentum is satisfied. From Eqs. (70)–(73) it can be seen that through measurement of the elastic response and the dielectric susceptibility, possibly a function of density, the properties of this type of material can be fully characterized. The combined true stress can then be calculated for a given strain and electric field. When the susceptibility remains isotropic during deformation, the electrostatic stress thus becomes

$$\sigma_{ij}^{es} = \kappa\left(E_iE_j - \frac{1}{2}E_kE_k\delta_{ij}\right) + \frac{1}{2}\rho\frac{d\chi}{d\rho}\kappa_0 E_kE_k\delta_{ij} \tag{74}$$

Now we may explore the implications of different assumptions of how the dielectric permittivity depends on the material density. If we take the susceptibility to be independent of the density (or equivalently that the stored polarization energy per unit current volume is proportional to the square of the magnitude of the polarization vector but insensitive to the density, see Eq. (55)), the electrostatic stress from Eq. (74) becomes

$$\sigma_{ij}^{es} = \kappa\left(E_iE_j - \tfrac{1}{2}E_kE_k\delta_{ij}\right) \tag{75}$$

a form commonly seen in the literature as an expression for the Maxwell stress in a dielectric [6]. The body force per unit volume associated with the electrostatic stress given in Eq. (75) is

$$\frac{\partial \sigma_{ji}^{es}}{\partial x_j} = \kappa\frac{\partial E_j}{\partial x_j}E_i = qE_i \tag{76}$$

where Eqs. (2) and (73) have been used to establish the free charge density. Thus the body force associated with this particular electrostatic stress in this particular material is the load per unit volume due to the electric field acting on the free charges, reflecting the fact that an electric field applies a unit force on a unit charge [6]. This would seem to be a reasonable choice as a postulate for the electrical body force and it is often seen as such in the literature. However, there is no implication in our approach that the electrostatic stress in Eq. (75) is the Maxwell stress, nor that the electrical body force is the expression given in Eq. (76). Indeed, there is no need to adopt any particular postulate for the electrical body force and the Maxwell stress, since the total true stress is in equilibrium with the mechanical body force whatever the form of the electrical body force and the Maxwell stress and this is all that is needed for a complete formulation of the behavior of the material in response to electric field and strain. Furthermore, the electrical body force and the Maxwell stress cannot be determined from measurements of the total true, elastic, or electrostatic stress, neither in terms of their body forces nor their interface or surface tractions.

Another possible assumption for the susceptibility is that

$$\chi = \frac{\rho\chi_1}{\rho_o} \tag{77}$$

where χ_1 is a constant. This means that the polarization energy per unit mass of the dielectric is proportional to the square of the dipole moment per unit mass, i.e.,

$$\psi_P = \frac{\rho_o}{2\kappa_0\chi_1}\left(\frac{P_k}{\rho}\right)\left(\frac{P_k}{\rho}\right) \tag{78}$$

where ψ_P is the polarization energy per unit mass of the material and the term in parentheses in Eq. (78) is clearly the dipole moment per unit mass. This relationship for the stored energy of polarization is often assumed to be the correct one for a linear, isotropic, dielectric material [1,3]. The result for the electrostatic stress from Eq. (74) is then

$$\sigma_{ij}^{es} = \kappa E_iE_j - \tfrac{1}{2}\kappa_o\chi E_kE_k\delta_{ij} = P_iE_j + \hat{\sigma}_{ij}^M \tag{79}$$

This is another form that can be found in the literature [1,3] as an expression for the Maxwell stress. The body force per unit volume arising from this electrostatic stress in this material is given by

$$\frac{\partial \sigma_{ji}^{es}}{\partial x_j} = qE_i + P_j\frac{\partial E_i}{\partial x_j} \tag{80}$$

where the free charge has been obtained from Eqs. (2), (5), and (73). The expression in Eq. (80), as a postulate for the electrical body force per unit volume, has a provenance based on molecular models for the electrical response of a lattice [3] and therefore lends credence to Eq. (79) as an expression for the Maxwell stress in this particular material. In this case, the body force is the effect of the electric field acting on free charge plus the net force acting on induced dipoles because of a gradient in the electric field. The latter effect is due to the different force magnitude on the positive and negative charges of the dipole because of the different electric field acting upon them. However, as before there is no implication in our approach that the electrostatic stress in Eq. (79) is the Maxwell stress, nor that the electrical body force is the expression given in Eq. (80). As we have emphasized already, there is no need to adopt any particular postulate for the electrical body force and the Maxwell stress.

A third possibility is that the isotropic susceptibility is inversely proportional to ρ, which leads to an electrostatic stress given by

$$\sigma_{ij}^{es} = \kappa(E_iE_j - E_kE_k\delta_{ij}) + \tfrac{1}{2}\kappa_o E_kE_k\delta_{ij} = P_iE_j - P_kE_k\delta_{ij} + \hat{\sigma}_{ij}^M \tag{81}$$

The body force per unit volume from this particular expression for the electrostatic stress in this particular material is given by

$$\frac{\partial \sigma_{ji}^{es}}{\partial x_j} = qE_i - \frac{\partial P_j}{\partial x_i}E_j \tag{82}$$

where, as before, the free charge has been obtained from Eqs. (2), (5), and (73). These forms, Eqs. (81) and (82), as a postulate for the Maxwell stress and the electrical body force, seem to be absent from the literature. However, they can be rationalized as giving a body force that accounts for the electric field acting on free charges but that also provides for an effect in which the gradient of dipole density produces a force opposite to the direction of the gradient. The latter action can be understood as being due to the electric field in association with a surplus of positive charges over negative charges (or the opposite) at a given point in the material when there is a gradient of polarization. However, as before, there is no implication in our approach that the electrostatic stress in Eq. (81) is the Maxwell stress, nor that the electrical body force is the expression given in Eq. (82). As we have emphasized already, there is no need to adopt any particular postulate for the electrical body force and the Maxwell stress.

Quasi-Linear Piezoelectric Materials

To illustrate results in the context of a piezoelectric material, we write down a stored energy given by

$$\psi = \frac{1}{2\rho_o}\varepsilon_{ij}c_{ijkl}\varepsilon_{kl} - \frac{1}{\rho}\Pi_i h_{ijk}\varepsilon_{jk} + \frac{\rho_o}{2\kappa_o\rho^2}\Pi_i\chi_{ij}^{-1}\Pi_j \tag{83}$$

where c_{ijkl} is a constant linear elasticity tensor at fixed polarization, h_{ijk} is a constant tensor of piezoelectric coefficients, and χ_{ij} is a constant susceptibility tensor denoting the response at zero strain. We note that in light of Eq. (43), ψ in Eq. (83) is objective. The choice of the form in Eq. (83) is somewhat arbitrary in the sense that the piezoelectric coefficients and the susceptibility tensor are fixed. By making such choices, we have eliminated the possibility that the susceptibility and the piezoelectric coefficients can experience a change of axiality during strain. On the other hand, in devising Eq. (83) we have utilized the notion that the stored energy per unit mass should depend on the polarization through the dipole moment per unit mass, an assumption that is fairly common in the literature.

Now we use Eqs. (47) and (48) and specialize to infinitesimal strain and rotation to obtain piezoelectric relationships in the form

$$E_i = -h_{ijk}e_{jk} + \frac{1}{\kappa_o}\chi_{ij}^{-1}P_j \tag{84}$$

and

$$\sigma_{ij} + \sigma_{ij}^M = c_{ijkl}e_{kl} - h_{kij}P_k + \tfrac{1}{2}(P_iE_j + E_iP_j) + \hat{\sigma}_{ij}^M \tag{85}$$

Note that the specialization to infinitesimal strain and rotation involves neglect of terms of higher order in strain and rotation. Complete linearization of the equations would eliminate the terms in Eq. (85) that are products of the polarization and electric field with the electric field. Note also that since the elasticity and piezoelectric coefficient tensors in Eq. (85) are symmetric on interchange of the subscripts i and j, the total true stress in Eq. (85) is symmetric, confirming that the balance of angular momentum is satisfied. As before, there is little mileage in attempting to separate the Cauchy and Maxwell stresses. Instead, we will identify the first two terms on the right-hand side of Eq. (85) to comprise the piezoelectric stress, σ_{ij}^p, and the remainder to be the electrostatic stress. Thus

$$\sigma_{ij}^p = c_{ijkl}e_{kl} - h_{kij}P_k \tag{86}$$

and

$$\sigma_{ij}^{es} = \tfrac{1}{2}(P_iE_j + E_iP_j) + \hat{\sigma}_{ij}^M \tag{87}$$

The body force per unit volume associated with this electrostatic stress in this material can be readily obtained by taking the spatial divergence of the expression in Eq. (87). However, the result is not particularly revealing. In any case, as before, there is no implication in our approach that the electrostatic stress in Eq. (87) is the Maxwell stress, nor that the electrical body force is its divergence. As we have emphasized already, there is no need to adopt any particular postulate for the electrical body force and the Maxwell stress. Instead we regard the electrostatic stress tensor in Eq. (87) simply to be an illustration of the consequences of certain constitutive assumptions embedded in Eq. (83).

Compliant Isotropic Dielectrics

Polymer dielectrics that have low shear moduli and are highly deformable to stretch ratios of order 10 have been introduced recently as actuator materials [5,11,16]. It is assumed that they are isotropic before straining and Kofod [11] has had success fitting their elastic response to a large strain, isotropic constitutive law for incompressible deformations developed by Ogden [12]. Pelrine et al. [5,16] and Kofod [11] also show that the dielectric permittivities of some of the polymers are unaffected or only slightly affected by straining, though there is evidence [17] that at least some polymers have susceptibilities that change significantly

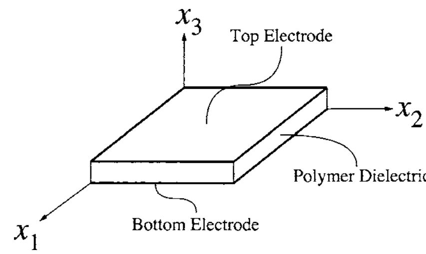

Fig. 2 A polymer dielectric actuator in the form of a slab with planar deformable electrodes

upon elastic deformation. The assumptions of incompressibility and insensitivity of the dielectric permittivity to straining are consistent with each other but not general. Therefore, we wish to study these highly deformable dielectrics within our formulation but when elastic dilatation is possible and when the permittivity depends on the strain. Ogden [14] has formulated an elasticity law for compressible materials that has a similar structure as his incompressible case; this could be used in our development and, presumably, it would model the stress-strain response of Kofod's [11] material well. However, the Ogden materials, whether incompressible or compressible, are somewhat complicated. Therefore, we will use a simpler elastic formulation, as we strive only to illustrate our approach to analyzing these materials and not to model them to a high degree of accuracy. Consequently, we will use a compressible form of the neo-Hookean material that is a variant of the Blatz-Ko [14,15] constitutive law. We will find that compressibility is essential to our ability to compute meaningful results in specific boundary value problems.

We choose also to present the results in terms of principal stresses and stretches as developed by Ogden [12,13], who also pointed out the great advantages of such an approach over formulating the results in terms of the strain tensor. In particular, it is directly applicable to the deformations of an actuator as shown in Fig. 2, which is a slab of polymer dielectric between two planar deformable electrodes [5,11,16]. For the polymer dielectric, we use a compressible generalization of the neo-Hookean formulation with an elastic strain energy density given by [14,15]

$$U_o = \frac{G}{2}(\lambda_i\lambda_i - 3J^{2/3}) + B(J - \ln J - 1) \tag{88}$$

where λ_i are the principal stretch ratios in the three orthogonal principal directions of the deformation so that the ratio of volume in the current state to volume in the reference state $J=\lambda_1\lambda_2\lambda_3 = \rho_o/\rho$. Note that the form presented in Eq. (88) is not a Blatz-Ko material per se because the term containing the bulk modulus B differs from their form [14,15]. However, our form is consistent with Blatz-Ko usage with their parameter k chosen to be unity and is used in the spirit of simplicity. Given that the principal elastic true stresses are given by [12,13]

$$\sigma_i^e = \frac{\lambda_i}{J}\frac{\partial U_o}{\partial \lambda_i} \text{ (no sum on } i) \tag{89}$$

they become

$$\sigma_i^e = G(\lambda_i^2 J^{-1} - J^{-1/3}) + B(1 - J^{-1}) \tag{90}$$

Note that for polymers B/G is usually very large compared to unity and that this effect will keep the dilatational strains very small during deformations.

Now consider the condition of the actuator shown in Fig. 2 subject to an electric field $E_3=E$ given by the voltage difference between the electrodes divided by the distance between the electrodes in the deformed state. The thickness of the actuator is very small compared to the in-plane dimensions so that a uniform electric field and stress state may be assumed in the dielectric and the effect of fringing fields at the edge can be ignored. Since we assume that the actuator is constrained so that it does not rotate and that gravitational loads are negligible, the rotation tensor $R_{ij} = \delta_{ij}$ and the actuator remains rectilinear in shape during deformation. The susceptibility is isotropic before straining and remains at least orthotropic thereafter because of the lack of shear strain. The electrostatic stress from Eq. (62) in all situations for this actuator is then given by

$$\sigma_{11}^{es} = -\left(1 + \chi_{33} + \lambda_1^2 \frac{\partial \chi_{33}}{\partial \varepsilon_{11}}\right)\frac{\kappa_o}{2}E^2$$
$$\sigma_{22}^{es} = -\left(1 + \chi_{33} + \lambda_2^2 \frac{\partial \chi_{33}}{\partial \varepsilon_{22}}\right)\frac{\kappa_o}{2}E^2 \qquad (91)$$
$$\sigma_{33}^{es} = \left(1 + \chi_{33} - \lambda_3^2 \frac{\partial \chi_{33}}{\partial \varepsilon_{33}}\right)\frac{\kappa_o}{2}E^2$$

with the shear components zero.

The electrodes are assumed to be very thin and highly compliant, as they are in practice [5,11,16], so that they offer no constraint on the deformation of the dielectric. Thus, the only mechanical loads are the tractions T_{33} applied through the electrodes and that act parallel to the x_3 axis or, alternatively, the tractions T_{11} or T_{22} applied parallel to the plane of the electrodes. In addition, the stretching of the actuator parallel to the plane of the electrodes may also be controlled kinematically.

In the first deformation we consider, the principal stretch ratios λ_1 and λ_2 are equal to each other and, consequently, the elastic stresses from Eq. (85) are

$$\sigma_{11}^{e} = \sigma_{22}^{e} = G\left(\frac{1}{\lambda_3} - \frac{1}{\lambda_1^{2/3}\lambda_3^{1/3}}\right) + B\left(1 - \frac{1}{\lambda_1^2\lambda_3}\right)$$
$$\sigma_{33}^{e} = G\left(\frac{\lambda_3}{\lambda_1^2} - \frac{1}{\lambda_1^{2/3}\lambda_3^{1/3}}\right) + B\left(1 - \frac{1}{\lambda_1^2\lambda_3}\right) \qquad (92)$$

with the shear components all zero. The case we will address first is that where the principal stretch ratios λ_1 and λ_2 are controlled but within the constraint of being equal. The result, deduced from Eq. (7), that the traction T_{33} is balanced by the sum of the σ_{33} components of the elastic and electrostatic stresses provides

$$G\left(\frac{\lambda_3}{\lambda_1^2} - \frac{1}{\lambda_1^{2/3}\lambda_3^{1/3}}\right) + B\left(1 - \frac{1}{\lambda_1^2\lambda_3}\right) + \frac{\kappa_o}{2}E^2\left(1 + \chi_{33} - \frac{\partial \chi_{33}}{\partial \varepsilon_{33}}\lambda_3^2\right) = T_{33} \qquad (93)$$

which, in principle, can be solved for λ_3. When χ_{33} is a function of strain, its derivative will be a function of λ_3 in a manner that can only be determined by experiment. Therefore, the order of Eq. (93), through the dependence of the susceptibility on the strain, is indeterminate without information from such experiments. Furthermore, if the potential difference between the electrodes is controlled, the electric field will be a function of λ_3, further complicating the equation order. Rather than pursue the solution of Eq. (93), we will consider the situation where the actuator, without mechanical load or electric field (i.e., in short circuit), is strained in the planar direction to a stretch ratio given by $\lambda_1 = \lambda_1^0$ and then an electric field applied along with blocking tractions sufficient to maintain the stretch ratios as they were before application of the field. The resulting through thickness strain before application of the electric field and traction conforms to a stretch $\lambda_3 = \lambda_3^o$ satisfying

$$G\left(\frac{\lambda_3^o}{(\lambda_1^o)^2} - \frac{1}{(\lambda_1^o)^{2/3}(\lambda_3^o)^{1/3}}\right) + B\left(1 - \frac{1}{(\lambda_1^o)^2\lambda_3^o}\right) = 0 \qquad (94)$$

and the blocking tractions required to maintain these stretch ratios are

$$T_{33} = \left[1 + \chi_{33}(\lambda_1^o,\lambda_3^o) - (\lambda_3^o)^2 \frac{\partial \chi_{33}(\lambda_1^o,\lambda_3^o)}{\partial \varepsilon_{33}}\right]\frac{\kappa_o}{2}E^2 \qquad (95)$$

A special case of this result is where all the stretch ratios are fixed at unity. The blocking traction is then

$$T_{33} = \tfrac{1}{2}(\tilde{\kappa} - \kappa_1 - \kappa_2)E^2 \qquad (96)$$

where the relevant value of the partial derivative of the susceptibility component has been deduced from Eq. (53) and, of course, Eq. (96) agrees with the result that can be obtained from the theory of Landau and Lifschitz [6], since this case is also the limit of zero infinitesimal strains.

Note that compressibility of the material is essential to our ability to calculate a meaningful result for the problem just addressed. If the polymer were exactly incompressible, then the kinematic relationships would become

$$\lambda_3 = \frac{1}{\lambda_1^2} \qquad (97)$$

and

$$\lambda_3^o = \frac{1}{(\lambda_1^o)^2} \qquad (98)$$

Then once the stretch parallel to the electrodes of the actuator has been established, applications of electric field and mechanical stress are incapable of changing the through thickness strain. Thus, actuation in the sense of thickness change for the device becomes impossible and there is no need for a blocking traction to suppress the actuation. All that applications of electric field and tractions do is to modify the hydrostatic stress in the dielectric polymer without changing the strain.

Having demonstrated that one must be careful when using an incompressible material model, we now turn to a problem that can be successfully and accurately assessed with a volume-preserving constitutive law as an approximation to the true behavior when the bulk modulus, B, is much greater than the shear modulus, G. In this problem, the actuator is first stretched in the x_2 direction with no other tractions or constraints applied and under short circuit conditions so that E is zero. This stretch ratio is held fixed thereafter so that $\lambda_2 = \lambda_2^o$ and due to incompressibility at this stage

$$\lambda_1 = \lambda_3 = \frac{1}{\sqrt{\lambda_2^o}} \qquad (99)$$

An electric field is then applied and simultaneously a traction T_{11}. In the incompressible limit, the elastic stress is given by Eq. (90) with $J=1$ and the term containing B is replaced by the negative of a pressure p. After the electric field is switched on, the stretch ratios must obey the relationship

$$\lambda_3 = \frac{1}{\lambda_1\lambda_2^o} \qquad (100)$$

and the pressure is calculated from the condition that the sum of the elastic and electrostatic stress in the σ_{33} orientation is zero because there is no traction applied in that direction. Thus

$$p = G\left[\frac{1}{(\lambda_1\lambda_2^o)^2} - 1\right] + \left[1 + \chi_{33} - \frac{1}{(\lambda_1\lambda_2^o)^2}\frac{\partial \chi_{33}}{\partial \varepsilon_{33}}\right]\frac{\kappa_o}{2}E^2 \qquad (101)$$

and it follows that the balance of forces in the σ_{11} orientation provides

$$G\left[\lambda_1^2 - \frac{1}{(\lambda_1\lambda_2^o)^2}\right] - \left\{1 + \chi_{33} + \frac{1}{2}\left[\lambda_1^2\frac{\partial \chi_{33}}{\partial \varepsilon_{11}} - \frac{1}{(\lambda_1\lambda_2^o)^2}\frac{\partial \chi_{33}}{\partial \varepsilon_{33}}\right]\right\}\kappa_o E^2 = T_{11} \qquad (102)$$

which can be solved for λ_1, in principle to determine the degree of actuation. As before, the solution is complicated by the fact that the dependence of the dielectric permittivity on the strain (i.e., on the stretch ratios) must be determined from experiment. Furthermore, if a fixed potential difference is applied between the electrodes and a fixed load provides the traction T_{11}, both the traction and the electric field are functions of λ_1 as well, further complicating the solution. A simpler situation is where T_{11} is the blocking traction which resists the effect of the electric field and maintains the stretch ratios at the levels given in Eq. (99) that prevailed prior to the application of the field. The result from Eq. (101) for this blocking traction is then

$$T_{11} = -\left\{ 1 + \chi_{33} + \frac{1}{2\lambda_2^o}\left[\frac{\partial \chi_{33}}{\partial \varepsilon_{11}} - \frac{\partial \chi_{33}}{\partial \varepsilon_{33}} \right] \right\} \kappa_o E^2 \qquad (103)$$

where the electric field and the traction are simple to establish because the thickness strain of the actuator is given by Eq. (99). The result in Eq. (103) predicts to within reasonable accuracy the results of an experiment carried out by Kofod [11]. It is notable that if the susceptibility is independent of strain, the traction predicted by Eq. (103) is compressive, reflecting the fact that application of the electric field will cause the actuator to try to expand parallel to the x_1 axis.

Discussion

The formulation presented in this paper is general and valid for materials in which the stress can be described by a local theory [7] and for which couple stresses or a director theory of materials response are not needed. In addition, the presentation is, we believe, free of unnecessary assumption in regard to the nature of the electrical body force and Maxwell stress in electrostatic systems. Instead, the expressions are presented in such a way that measurable behavior can be used to determine all the necessary functions and constants to completely describe the material constitutive behavior for reversible response. Indeed, it is not necessary to know the electrical body force or the Maxwell stress to obtain a usable constitutive law. Instead, given the assumptions of the paper regarding the constitutive law, measurements of the elastic response at zero electric field and of the material's dielectric permittivities as a function of strain will fully characterize the constitutive law. If the assumptions of the paper are incorrect in the sense that the constitutive law is more complex than that presented in Eq. (55), more extensive experiments will be needed to characterize the free energy as stated in Eq. (43), perhaps because the dielectric response involves a nonlinear dependence of the electric displacement on the electric field or that the elastic and electrostatic energy do not separate in the manner assumed in Eq. (55).

In regard to the terminology in the paper, quibbles can be raised, e.g., concerning what we call the Cauchy stress, which some workers regard to be what we have called the total true stress, i.e., the sum of what we call the elastic and electrostatic stress. However, this would be to focus on the wrong issues, because it is the formulation that is important, not the names of the terms. In any case we would not object to instructions to rename our entities, although we prefer our choice of names for what we have called the Cauchy, Maxwell, elastic, and electrostatic stresses.

Finally, we reemphasize the comments made in the Introduction that the formulation we have presented is desirable for use in a modern setting where feasibility of experiments, possible applications in finite element calculations, and the need for developments in the fracture mechanics of electromechanically loaded components have guided our thinking. We believe that our presentation is not only valid, but is attractive in the context of these needs. We have focused on conservative materials. However, all of what is developed up to and including Eq. (34) is correct for dissipative material behavior in the electrostatic limit, such as ferroelectric switching [18,19]. Thus, the formulation can and will be extended to dissipative materials in due course.

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References

[1] Toupin, R. A., 1956, "The Elastic Dielectric," J. Rational Mech. Anal., **5**, pp. 849–914.
[2] Voigt, W., 1910, *Lehrbuch der Kristallphysik*, Teubner, Leipzig.
[3] Eringen, A. C., 1963, "On the Foundations of Electroelastostatics," Int. J. Eng. Sci., **1**, pp. 127–153.
[4] Landis, C. M., and McMeeking, R. M., 2000, "Modeling of Fracture in Ferroelectric Ceramics," *Smart Materials and Structures: Active Materials: Behavior and Mechanics*, Proceedings of SPIE, edited by C. S. Lynch, Vol. 3992, pp. 176–184.
[5] Pelrine, R., Kornbluh, R., Pei, Q., and Joseph, J., 2000, "High-Speed Electrically Actuated Elastomers with Over 100% Strain," Science, **287**, pp. 836–839.
[6] Landau, L. D., and Lifschitz, E. M., 1960, *Electrodynamics of Continuous Media*, Pergamon, Oxford.
[7] Malvern, L. E., 1969, *Introduction to the Mechanics of a Continuous Medium*, Prentice-Hall, Englewood Cliffs, NJ.
[8] McMeeking, R. M., 1998, "A Maxwell Stress for Material Interactions," J. Colloid Interface Sci., **199**, pp. 187–196.
[9] Coleman, B. D., and Noll, W., 1963, "The Thermodynamics of Elastic Materials with Heat Conduction and Viscosity," Arch. Ration. Mech. Anal., **13**, pp. 167–178.
[10] Carroll, M. M., 2004, "Derivatives of the Rotation and Stretch Tensors," Math. Mech. Solids, **9**, pp. 543–553.
[11] Kofod, G., 2001, "Dielectric Elastomer Actuators," Ph.D. thesis, Department of Chemistry, Technical University of Denmark, Riso Report No. Riso-R-1286(EN).
[12] Ogden, R. W., 1972, "Large Deformation Isotropic Elasticity—On the Correlation of Theory and Experiment for Incompressible Rubberlike Solids," Proc. R. Soc. London, Ser. A, **326**, pp. 565–584.
[13] Ogden, R. W., 1972, "Large Deformation Isotropic Elasticity: On the Correlation of Theory and Experiment for Compressible Rubberlike Solids," Proc. R. Soc. London, Ser. A, **328**, pp. 567–583.
[14] Blatz, P. J., and Ko, W. L., 1962, "Application of Finite Elasticity Theory to the Deformation of Rubbery Materials," Trans. Soc. Rheol., **6**, pp. 223–251.
[15] Blatz, P. J., 1963, "Application of Finite Elastic Theory to the Behavior of Rubberlike Materials," Rubber Sci. Technol., **36**, pp. 1459–1496.
[16] Pelrine, R., Kornbluh, R., Heydt, J. J. R., Pei, Q., and Chiba, S., 2000, "High-Field Deformation of Elastomeric Dielectrics for Actuators," Mater. Sci. Eng., C, **C11**, pp. 89–100.
[17] Shkel, Y. M., and Klingenberg, D. J., 1996, "Material Parameters for Electrostriction," J. Appl. Phys., **80**, pp. 4566–4572.
[18] Cocks, A. C. F., and McMeeking, R. M., 1999, "A Phenomenological Constitutive Law for the Behavior of Ferroelectric Ceramics," Ferroelectrics, **228**, pp. 219–228.
[19] Landis, C. M., 2002, "Fully Coupled, Multi-Axial, Symmetric Constitutive Laws for Polycrystalline Ferroelectric Ceramics," J. Mech. Phys. Solids, **50**, pp. 127–152.