A Fully Coupled Constitutive Model for Electrostrictive Ceramic Materials

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ABSTRACT: A three-dimensional, electromechanical constitutive law has been formulated for electrostrictive ceramic materials. This fully coupled, phenomenological model relates the key state variables of stress, strain, electric field, polarization and temperature in a set of compact nonlinear equations. The direct and converse electrostrictive effects are modeled by assuming that the electrically induced strain depends on second-order polarization terms. In addition, a simple empirical relationship for the dielectric behavior is used to model the saturation of the induced polarization with increasing electric field.

Unlike previous electrostrictive constitutive laws based on polynomial expansions, this constitutive law depends on a manageable number of material constants. As an example, material constants for the model were determined from induced strain and dielectic data for a relaxor-ferroelectric based on lead magnesium niobate, $Pb(Mg_{1/3}Nb_{2/3})O_3$-$PbTiO_3$-$BaTiO_3$ (PMN-PT-BT). Finally, predictions of the material's mechanical behavior under constant electric field and its electrical behavior under constant applied stress are made.

INTRODUCTION

Two important components of an active smart materials system are the sensors and actuators. Sensors are used by the system to detect changes in the environment, while actuators are used in a feedback loop to respond to those changes. Devices constructed from piezoelectric and electrostrictive ceramics are excellent candidates for both these roles under a variety of applications. The most common ceramic materials used for actuators and sensors today are piezoelectrics based on lead zirconiate titanate, $Pb(Zr,Ti)O_3$. As actuators, these materials can convert electrical energy into small but accurate displacements with a fast response time. Compared with electromagnetic actuators, they are more compact, consume less power, and are less prone to overheating (Uchino, 1986). In the sensor role, electromechanical ceramics can detect minute displacements by converting mechanical work into electrical energy. Typically, these sensors are packaged in polymer composite plates and used in underwater hydrophone and medical applications (Li and Sottos, 1994). Collocated devices have also been developed that combine the sensor and actuator functions into a single piezoelectric element (Dosch, Inman and Garcia, 1992). Recently considerable research has been devoted to the relatively new electrostrictive ceramics, and several electrostrictive actuator designs have been proposed (Nakajima et al., 1985; Uchino, 1986; Winzer, Shankar and Ritter, 1989). These materials generally offer higher electrically induced strain with lower hysteresis than their piezoelectric counterparts, but with the penalties of complicated electromechanical behavior and temperature dependency. Also Hom et al. (1994) have shown that the electromechanical coupling efficiency for electrostrictors is comparable to the coupling efficiency for piezoelectrics.

Piezoelectrics and electrostrictors both belong to a class of ionic crystals known as ferroelectrics. Ferroelectrics consist of subvolumes, called domains, that have a uniform, permanent, reorientable polarization. Since the direction of polarization for each domain is randomly oriented, the crystal itself has no net bulk polarization. Above a characteristic temperature, the Curie temperature, a ferroelectric undergoes a transition where the spontaneous polarization disappears (Landau and Lifshitz, 1960; Nye, 1985). Piezoelectricity is induced in a ferroelectric ceramic by applying a high electric field at elevated temperatures during manufacture. This process, called poling, partially aligns the polar axes of the domains to create a macroscopic polarization in the crystal. The resulting poled-piezoelectric will deform when subjected to an electric field and polarize when mechanically stressed. For low electric fields [<0.1 MV/m for $Pb(Zr,Ti)O_3$], the electrically-induced strain response for a piezoelectric is proportional to the applied field. The electric field, in turn, is proportional to the induced polarization. At higher AC fields [>0.6–1.1 MV/m peak-peak for $Pb(Zr,Ti)O_3$ depending on the formulation], significant electromechanical hysteresis occurs as the domains grow and their boundaries (walls) move. This hysteresis can create servo-displacement control problems in piezoelectric actuator devices.

Electrostriction is also a coupled electromechanical ef-

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fect; however, the induced strain is approximately proportional to the square of the induced polarization. Therefore the same deformation occurs when the field is reversed, in contrast to piezoelectricity. Materials with very large polarizations, such as the relaxor-ferroelectrics, can exhibit large electrostrictive strains (Somolenski et al., 1961; Cross, 1986; Winzer, Shankar and Ritter, 1989). The cation distributions in relaxor-ferroelectric are partially disordered, so a mixture of pyroelectric and paraelectric microphases exists over a wide temperature range. As a result, the spontaneous polarization is not suddenly lost at a specific Curie temperature, but slowly decays with increasing temperature. The dielectric hysteresis disappears before the spontaneous polarization, so significant electrostriction with minimal hysteresis is possible both above and below the nominal transition temperature. The most promising relaxor-ferroelectric materials for actuator devices are based on lead magnesium niobate, $Pb(Mg_{1/3}Nb_{2/3})O_3$ (PMN), or its solid solutions with lead titanate, $Pb(Mg_{1/3}Nb_{2/3})O_3$-$PbTiO_3$ (PMN-PT). PMN-based materials operating above the nominal transition temperature typically have high electrically-induced strains ($\sim 0.1\%$) and low hysteresis ($<5\%$) over moderate electric fields (~ 1 MV/m peak-peak).

A constitutive theory is essential for predicting the reliability and performance of any electroceramic device. When the constitutive theory is incorporated into a finite element code, structural analysis of the device design can be performed to identify critical stress areas. Also, finite element analysis can be performed to predict the response of the device when embedded in complicated composite structures. Piezoelectrics have been in use for over 50 years, and their electromechanical behavior is commonly modeled by a linear constitutive law formulated by Voight (1910). This phenomenological model assumes that the polarization and stress depend linearly on the total strain and the electric field.

Conversely, electrostrictive materials are relatively new and complicated in behavior. Consequently, nonlinear constitutive models for electrostrictors are not as mature as models for piezoelectrics. The Voight model can be extended to nonlinear electrostrictive behavior by adding second-order polynomial terms for the electric field (Nye, 1985; Damjanovic and Newnham, 1992). As shown in Figure 1, this approximation is reasonable at low electric fields when dielectric behavior is approximately linear. However, at high electric fields (>1 MV/m), the polarization begins to saturate, and higher-order terms must be added to the model. Experimental testing of PMN (Pilgrim et al., 1992) indicates that the electrically induced strain still depends on the square of polarization even at high electric fields. These results indicate that a constitutive theory based on polarization may model electrostriction better than a theory based on electric field. Devonshire (1954) formulated a phenomenological model of ferroelectricity that used polarization, stress and temperature as independent state variables and included an electrostrictive effect. Suo (1991) used Devonshire's theory to model an isotropic relaxor-ferroelectric. Suo's model assumed that induced strain depends only on second-order polarization terms, while the electric field depends on a polynomial expansion of the polarization.

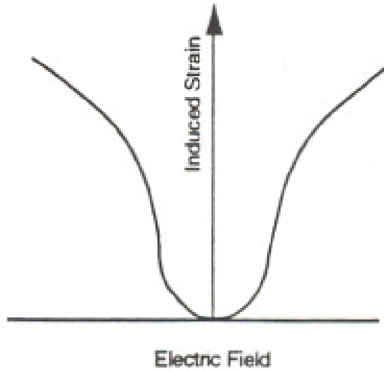

Figure 1. A typical electrostrictive response (induced strain versus applied field) for a relaxor-ferroelectric above the Curie temperature.

In this paper, Suo's general approach was followed to formulate a constitutive model for relaxor-ferroelectrics that uses polarization and strain as the independent state variables. However, the material's nonlinear dielectric behavior was modeled with an explicit function that simulates polarization saturation. The model was then used to predict the mechanical behavior of the relaxor-ferroelectric under constant electric field and its electrical behavior under constant stress.

CONSTITUTIVE LAW FORMULATION

A constitutive theory for ferroelectrics is a relationship between the key state variables of polarization, **P**; electric field, **E**; strain, ϵ; and stress, σ. The strain in the crystal is defined by

$$\epsilon_{ij} = \frac{1}{2}\left(\frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i}\right) \tag{1}$$

where **x** denotes the Cartesian coordinates of a material point and **u** is that point's mechanical displacement from its undeformed position. Here, compressed matrix notation is adopted, where a single index replaces the two indices for stress and strain. Thus, the normal strains are

$$\epsilon_1 \equiv \epsilon_{11}, \quad \epsilon_2 \equiv \epsilon_{22}, \quad \epsilon_3 \equiv \epsilon_{33} \tag{2}$$

while the shear strains are

$$\epsilon_4 \equiv 2\epsilon_{12}, \quad \epsilon_5 \equiv 2\epsilon_{23}, \quad \epsilon_6 \equiv 2\epsilon_{13} \tag{3}$$

Similarly, the normal stresses and shear stresses are defined by

$$\sigma_1 \equiv \sigma_{11}, \quad \sigma_2 \equiv \sigma_{22}, \quad \sigma_3 \equiv \sigma_{33}$$

$$\sigma_4 \equiv \sigma_{12}, \quad \sigma_5 \equiv \sigma_{23}, \quad \sigma_6 \equiv \sigma_{13} \tag{4}$$

A set of phenomenological constitutive relations can be formulated from Devonshire's Theory of Ferroelectrics (1954), which describes a relaxor-ferroelectric's macroscopic behavior. From thermodynamic considerations, Devonshire proposed that a Helmholtz free energy, A, exists that is solely a function of polarization, strain and temperature, T (i.e., $A = A(\mathbf{P},\epsilon,T)$). The remaining state variables are related to A by,

$$S = -\frac{\partial A}{\partial T} \tag{5}$$

$$\sigma_p = \frac{\partial A}{\partial \epsilon_p} \tag{6}$$

$$E_i = \frac{\partial A}{\partial P_i} \tag{7}$$

where S is entropy. Once a form of A has been selected for a particular material, it can be inserted into Equations (4), (5) and (6) to define the constitutive law.

Using a set of empirical assumptions, A will now be derived for an isothermal polycrystalline relaxor-ferroelectric above the Curie transition temperature. These assumptions are: (1) the electrically induced strain only depends on second-order polarization terms, (2) the elastic modulus of the crystal is independent of polarization and temperature, (3) the stress-free dielectric behavior can be expressed by a closed-form equation, and (4) the material is initially isotropic. Clearly, these assumptions must be validated by experimental testing.

The formulation of the electrostrictive model begins by considering the mechanical behavior of a relaxor-ferroelectric. The first assumption, that the electrically induced strain ϵ^E, depends only on second-order polarization terms, means that,

$$\epsilon_p^E = Q_{pij} P_i P_j \tag{8}$$

where the electrostrictive coefficients, $\mathbf{Q}$, are material parameters that must be measured.** Electrical testing of PMN has shown that $\mathbf{Q}$ is relatively independent of temperature (Uchino et al., 1980; Pilgrim et al., 1992). The total strain of the ceramic is the sum of the elastic strain, the electrically induced strain and the thermally induced strain. Therefore, the stress in the relaxor ferroelectric is

$$\sigma_p = C_{pq}(\epsilon_q - \epsilon_q^E - \alpha_q(T - T_o)) \tag{9}$$

where $\mathbf{C}$ and α are the elastic stiffness and thermal expansion coefficients of the material, respectively. T_o is the reference temperature of the ceramic. As mentioned previously, the second assumption for this model is that the stiffness of the crystal does not depend on its polarization or temperature. Equations (6), (8) and (9) can be combined to obtain,

$$\frac{\partial A}{\partial \epsilon_q} = C_{pq}(\epsilon_q - Q_{qij} P_i P_j - \alpha_q(T - T_o)) \tag{10}$$

Integration of Equation (10) with respect to strain yields

$$A = \frac{C_{pq}\epsilon_p\epsilon_q}{2} - C_{pq}Q_{pij}\epsilon_q P_i P_j - C_{pq}\epsilon_q\alpha_p(T - T_o) + f(\mathbf{P},T) \tag{11}$$

where f is a function of polarization and temperature.

Now consider the dielectric behavior of the relaxor-ferroelectric. Electrical testing shows that the dielectric behavior of relaxor-ferroelectrics is nonlinear (Pilgrim et al., 1992). At low fields the polarization is approximately proportional to the applied electric field. However, at high fields, the induced polarization saturates, where the saturation value is the spontaneous polarization. A possible mechanism for this dielectric behavior can be explained as follows. The individual crystals are divided into domains in which a uniform permanent dipole moment is embedded in the atomic lattice. Since the crystals are randomly oriented, the macroscopic or net polarization of the polycrystal is initially zero. An applied electric field induces the permanent dipoles to rotate and stretch towards the direction of the field resulting in a net polarization of the polycrystal. However, as the field is increased, the lattice structure prevents the complete alignment or further elongation of the dipoles, so the macroscopic polarization eventually saturates. The third assumption of the constitutive model is that this *stress-free* dielectric behavior can be described by the hyperbolic tangent function, i.e.,

$$|\mathbf{P}| = P_s \tanh(k|\mathbf{E}|) \tag{12}$$

where P_s is the spontaneous polarization, k is a new material constant, and $|\mathbf{P}|$ and $|\mathbf{E}|$ are the magnitudes of the polarization and electric field, respectively. Both P_s and k depend on the temperature of the crystal. Zhang and Rodgers (1993) originally proposed Equation (12) in order to describe the dielectric behavior of piezoelectrics with hysteresis. Assuming the material is isotropic, the *stress-free* behavior described by Equation (12) can be rewritten as,

$$E_i = \frac{1}{k}\operatorname{arctanh}\left(\frac{|\mathbf{P}|}{P_s}\right)\frac{P_i}{|\mathbf{P}|} \tag{13}$$

Equations (7) and (11) can be used to obtain

**In this paper, Einstein summation is used on all repeated indices.

$$E_i = -2C_{pq}Q_{qij}\epsilon_p P_j + \frac{\partial f}{\partial P_i} \tag{14}$$

Also the total strain under stress-free conditions is the sum of the electrically induced strain given by Equation (8) and the thermally induced strain. The combination of Equations (8), (9), (13) and (14) results in

$$\frac{\partial f}{\partial P_i} = 2C_{pq}Q_{qij}P_j(Q_{pkl}P_kP_l + \alpha_p(T - T_o)) + \frac{1}{k}\operatorname{arctanh}\left(\frac{|\mathbf{P}|}{P_s}\right)\frac{P_i}{|\mathbf{P}|} \tag{15}$$

Integration of Equation (15) with respect to polarization yields

$$f = C_{pq}Q_{qij}P_iP_j\left(\frac{Q_{pkl}}{2}P_kP_l + \alpha_p(T - T_o)\right) + \frac{1}{2k}\left[|\mathbf{P}|\ln\left(\frac{P_s + |\mathbf{P}|}{P_s - |\mathbf{P}|}\right) + P_s\ln\left(1 - \left(\frac{|\mathbf{P}|}{P_s}\right)^2\right)\right] + \frac{C_{qp}}{2}\alpha_q\alpha_p(T - T_o)^2 \tag{16}$$

By combining Equations (11) and (16), A can be defined as

$$A = \frac{C_{pq}}{2}(\epsilon_q - \epsilon_q^r - \alpha_q(T - T_o)) \times (\epsilon_p - \epsilon_p^r - \alpha_p(T - T_o)) + \frac{1}{2k}\left[|\mathbf{P}|\ln\left(\frac{P_s + |\mathbf{P}|}{P_s - |\mathbf{P}|}\right) + P_s\ln\left(1 - \left(\frac{|\mathbf{P}|}{P_s}\right)^2\right)\right] \tag{17}$$

Finally using Equations (7) and (17), the dielectric behavior of the relaxor-ferroelectric under mechanical load is given by

$$E_i = -2C_{pq}Q_{pij}(\epsilon_q - Q_{qkl}P_kP_l - \alpha_q(T - T_o))P_j + \frac{1}{k}\operatorname{arctanh}\left(\frac{|\mathbf{P}|}{P_s}\right)\frac{P_i}{|\mathbf{P}|} \tag{18}$$

Equations (9) and (18) together form the constitutive laws that define the electromechanical behavior of a relaxor-ferroelectric. The first term on the right side of Equation (18) is the converse electrostrictive effect; through this term the state of stress affects the dielectric behavior. In this sense the equations are fully coupled, since the stress state also depends on the polarization through Equation (9).

Assuming that the relaxor-ferroelectric is isotropic significantly simplifies the material constants, **C**, α and **Q**. For isotropic materials, the elastic stiffness is given by

$$\mathbf{C} = \frac{Y}{(1 + \nu)(1 - 2\nu)}\begin{bmatrix} 1-\nu & \nu & \nu & 0 & 0 & 0 \\ \nu & 1-\nu & \nu & 0 & 0 & 0 \\ \nu & \nu & 1-\nu & 0 & 0 & 0 \\ 0 & 0 & 0 & \frac{(1-2\nu)}{2} & 0 & 0 \\ 0 & 0 & 0 & 0 & \frac{(1-2\nu)}{2} & 0 \\ 0 & 0 & 0 & 0 & 0 & \frac{(1-2\nu)}{2} \end{bmatrix} \tag{19}$$

where Y and ν are the Young's modulus and Poisson's ratio, respectively. α is given by

$$\boldsymbol{\alpha} = [\,\alpha \quad \alpha \quad \alpha \quad 0 \quad 0 \quad 0\,] \tag{20}$$

For isotropic behavior, the non-zero elements of **Q** are related by

$$\begin{gathered} Q_{111} = Q_{222} = Q_{333} \\ Q_{122} = Q_{133} = Q_{211} = Q_{233} = Q_{311} = Q_{322} \\ Q_{412} = Q_{523} = Q_{613} = 2(Q_{111} - Q_{122}) \end{gathered} \tag{21}$$

All other components of **Q** are zero, so only two independent components, Q_{111} and Q_{112}, actually exist.

The electrostrictive material model presented in this paper depends on a limited number of parameters: Y, ν, α, Q_{111}, Q_{122}, P_s and k. However, P_s and k depend on temperature, so the model can only be used for the temperature at which those two constants are valid. The model's material parameters are determined by electrical and mechanical tests. Y and ν are measured by mechanical testing of the crystal under applied compression and zero electric field (Cao and Evans, 1993). The strain response during testing is purely elastic since the material will not polarize without an electric field. The stress-strain results are used to determine the elastic stiffness. α is determined by standard thermal expansion tests.

Q_{111}, Q_{122}, P_s and k are measured by electrical testing of small pellets of material (Pilgrim et al., 1992; Hom et al., 1994). After electroding, the stress-free pellet is subjected to an applied electric field. A piezoresistive strain gage bonded to the electroded surface measures the transverse strain, while an eddy current sensor measures the longitudinal strain from a metal target bonded to the surface of the electrode. Measurement of the induced polarization can be performed with the integrating capacitor technique used by Sawyer and Tower (1930).

Table 1. Material parameters for a PMN-PT-BT relaxor-ferroelectric at 5°C.

Y (GPa)	Q_{111} $\times 10^{-1}$ m^4/C^2	Q_{122} $\times 10^{-2}$ m^4/C^2	P_s (C/m^2)	k (m/MV)
97.0	0.133	−0.606	0.259	1.16

In order to illustrate how the material constants are determined, the results of electromechanical testing by Hom et al. (1994) on a PMN-PT-BT based relaxor-ferroelectric are now considered. The actual composition of this material, designated b250077, is a ternary system that contains 7.7% $PbTiO_3$ (PT) with PMN as the base and 2.5% $BaTiO_3$ (BT) as a dopant (percentages by volume). b250077 has a nominal transition temperature of 18°C, and it was tested by Hom et al. at 5°C using the techniques described above. The material parameters measured by Hom et al. are listed in Table 1.

Figure 2 shows the dielectric behavior measured when a 2 MV/m AC electric field was applied at 1 Hz. P_s and k were determined from this data using a least squares fit. Figure 2 also shows the model's behavior [Equation (12)] with the curve fitted parameters. Considering it is only a two-parameter fit, the empirical model matches the experiment extremely well. Q_{111} and Q_{122} were determined by curve fitting the electrically induced strain versus polarization data to Equation (8); the results are shown in Table 1. Figure 3 shows the measured induced strain versus electric field and the corresponding behavior predicted by the model. The model agrees well with the experiment even at high fields.

PREDICTED ELECTROMECHANICAL BEHAVIOR

The electrostrictive constitutive laws presented in the previous section represent a set of implicit equations for a relaxor-ferroelectric's state of polarization and strain. Given

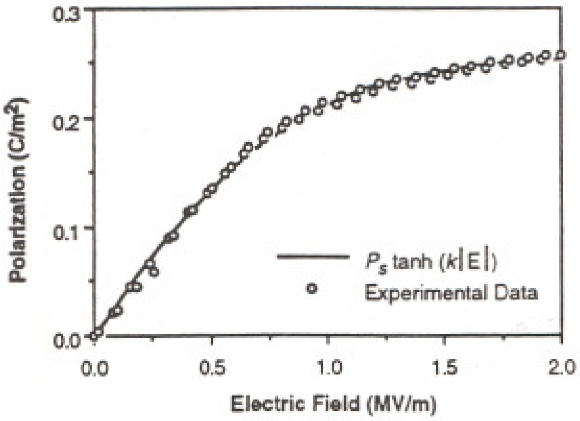

Figure 2. Induced polarization versus applied electric field for PMN-PT-BT under stress-free conditions. The test temperature is 5°C and the applied AC field frequency is 1 Hz. The solid line is Equation (12) with P_s = 0.259 C/m² and k = 1.160 m/MV.

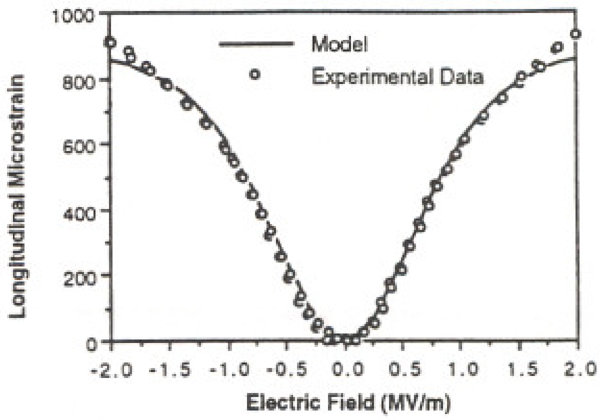

Figure 3. Induced strain versus applied electric field for PMN-PT-BT under stress-free conditions. The test temperature is 5°C and the applied AC field frequency is 1 Hz. The solid line is the behavior predicted by the model.

the electric field and stress, Equations (9) and (18) can be solved numerically. In this section, the solutions of those equations are presented for two simple cases: the dielectric behavior under constant prestress and the mechanical behavior under constant electric field. The electrostrictive material constants used in this exercise correspond to the b250077 PMN-PT-BT relaxor-ferroelectric at 5°C described in the previous section.

Electric Behavior under Constant Stress

The electric behavior of an electrostrictor under mechanical load is important when the device is being used for sensor applications. As an illustration, consider a relaxor-ferroelectric under a compressive prestress, where the mechanical load, σ, and the applied electric field, E, are aligned in the same direction. Using Equations (9) and (18), the induced polarization, P, is implicitly defined by

$$E = -2Q_{111}\sigma P + \frac{1}{k}\,\mathrm{arctanh}\left(\frac{P}{P_s}\right) \tag{22}$$

where E and σ are known quantities. Using the material constants in Table 1, Equation (22) was solved with the Newton-Raphson iteration technique. The results of this calculation for various levels of prestress are shown in Figure 4.

The calculation indicates how the relaxor-ferroelectric would perform as stress detecting sensor. As shown in Figure 4, the compressive prestress can be measured by the drop in polarization. However the applied bias field must be chosen carefully in order to make this measurement. If no electric field is applied, the crystal will not polarize. Conversely, if too much bias is applied the polarization will saturate. Clearly there is an optimum bias field, where the device's sensitivity is highest.

The electrically induced strain can be computed from Equation (8), and Figure 5 shows that strain for different

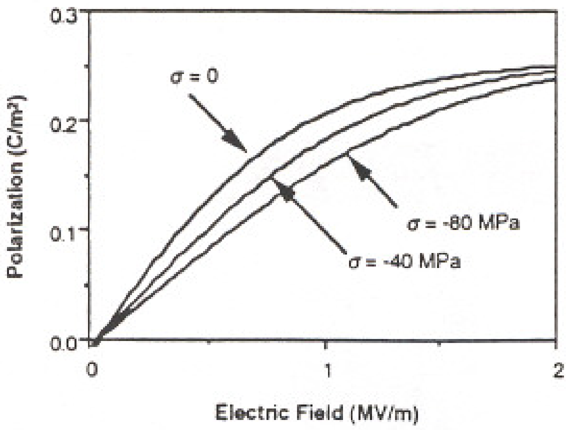

Figure 4. Predicted dielectric response for PMN-PT-BT at 5°C under compressive prestress.

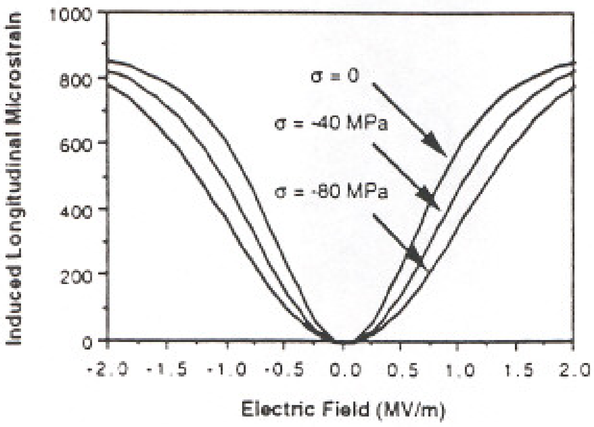

Figure 5. Predicted induced strain response for PMN-PT-BT at 5°C under compressive prestress.

levels of prestress. Significant drops in induced strain are predicted with increasing compressive prestress, particularly near 1 MV/m. Indeed, an 80 MPa compressive stress reduces the induced strain at 1 MV/m by 40%. Prediction of this phenomenon is important for actuator design, since a surrounding structure will compress embedded devices when they are activated.

Mechanical Behavior under Constant Electric Field

Experimental testing has shown that the stiffness of an electrostrictive actuator can actually change with the applied electric field (Nakajima et al., 1985). This observation appears at first to contradict the assumption that stiffness is constant. As an example, consider the behavior of an electrostrictor that has a constant electric field applied and then is compressed in the same direction as that field. For this case, Equations (9) and (18) can be combined to obtain

$$E = -2Q_{111}Y(\epsilon - Q_{111}P^2)P + \frac{1}{k}\operatorname{arctanh}\left(\frac{P}{P_s}\right) \quad (23)$$

Given the applied field and the total strain, ϵ, the polarization can be computed implicitly from Equation (23). This calculation was done numerically using Newton-Raphson iteration, then the reaction stress, σ, was computed directly from Equation (9). Figure 6 shows the results for various applied fields.

The calculation shows that when $E < 1.5$ MV/m the "apparent" modulus of the material decreases with increasing electric field. Since the field is constant, the increasing compressive stress causes a decrease in the induced polarization by the converse effect. This drop in polarization in turn causes a drop in the electrically induced strain. As a result, the modulus appears to decrease. In this specific example, the effective modulus of the b250077 relaxor-ferroelectric at 5°C decreases 21% when a 1 MV/m field is applied. At higher fields ($E > 2$ MV/m), the polarization saturates and the converse effect is limited. Any drop in

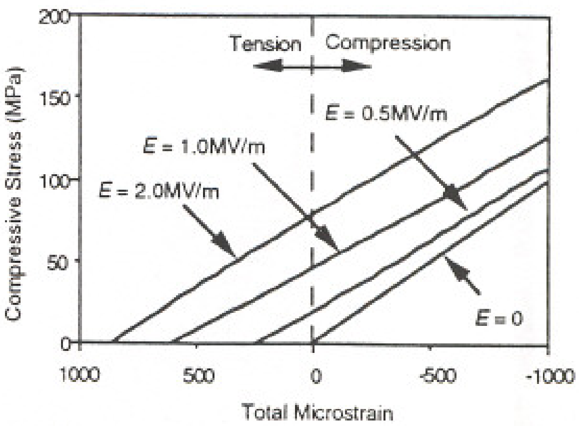

Figure 6. Predicted mechanical response for PMN-PT-BT at 5°C under fixed electric field.

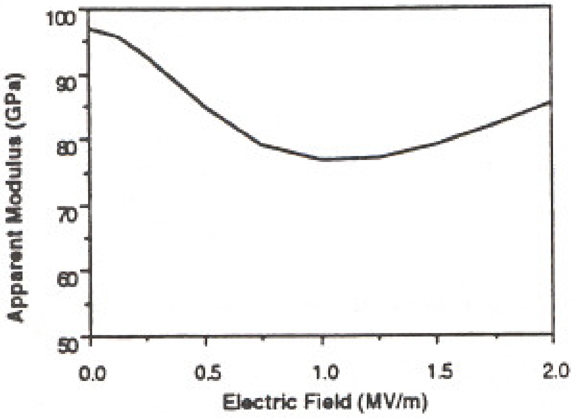

***Figure 7.** Predicted "apparent" modulus for PMN-PT-BT at 5°C under constant electric field.*

polarization is relatively small, so the "apparent" modulus returns to the actual modulus of the material, Y. A least-squares fit of the stress-strain curves in Figure 6 was performed to determine the "apparent" modulus of the material under electric field. Figure 7 shows the predicted modulus values from that procedure.

DISCUSSION

The electrostrictive constitutive model described in this paper is empirically based, and it agrees with the observed stress-free behavior of PMN-PT. However the general assumptions of the model still need to be validated by more complicated electromechanical testing. Towards that goal, the electrical behavior under constant prestress and the mechanical behavior under constant field are currently being measured for PMN-PT. The experimental results will be compared with the predictions made in the previous section, and reported on at a later date.

Finally, the electrostrictive constitutive model has also been incorporated in a nonlinear finite element code. This code will be used in the future to predict the stress state in multilayered electrostrictive actuators and to predict the performance of electrostrictive devices in composite structures. The development and results of the finite element analysis will also be reported at a later date.

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