Constitutive Modeling of Electrostrictive Polymers using a Hyperelasticity-Based Approach

A.W. Richards and G.M. Odegard[1]

Department of Mechanical Engineering – Engineering Mechanics
Michigan Technological University
Houghton, MI 49931

Abstract

The use of constitutive equations to describe the electro-mechanical behavior of electrostrictive materials began over 100 years ago. While these equations have been used to model a host of ceramic-based and polymer-based electroactive materials, a fully-characterized model has not yet been developed to predict the response of transversely-isotropic polymer electrostrictives. A constitutive model is developed within a thermodynamic and hyperelastic framework that incorporates the transversly-isotropic material symmetry that is present in many polymer-based electrostrictives. The resulting constitutive model is characterized for three electrostrictive polymer systems using empirical data that is available in the literature. The model has a relatively simple functional form that is easily adaptable to other polymer electrostrictive material systems.

Keywords: Electroactive materials, Electrostriction, MEMS, Polyurethane Elastomers

1. Introduction

Polymer-based electrostrictive materials have become a focal point of research for applications that require large magnitudes of actuation and significant weight-savings [1-23]. These applications range from micro-electro-mechanical systems (MEMS) to artificial muscles. The use of these materials in engineering applications requires accurate and flexible constitutive relations to relate loads, deformation, electric displacement, and applied electrical field.

In the late 19th century and the early part of the 20th century, several studies reported a relationship between the applied electric field, stress field, and strain field for cylindrical condensers [24-33]. In the middle portion of the 20th century, many scholars studied the broader applications of electrostrictive materials, and established more specific definitions of electrostrictive behavior. In 1941, Stratton [34] defined electrostriction as simply the elastic deformation of a dielectric under the forces exerted by an electrostatic field. However, numerous authors [35-42] generally agreed that electrostrictive materials exhibit a constitutive behavior that can be described in a thermodynamic framework. More recently, many studies employed this thermodynamic framework for electrostrictive materials [43-56].

The objective of this study is to establish a hyperelastic framework for modeling polymer-based

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[1] Corresponding author

electrostrictive materials with transverse-isotropic material symmetry. The resulting constitutive model is characterized for three electrostrictive polymer systems using empirical data that is available in the literature. The thermodynamic framework of the modeling approach is established first followed by the characterization of material parameters.

2. Kinematics and Balance Laws

Consider a region of a material manifold $\mathfrak{R}$ embedded in a three-dimensional Euclidean space with its volume enclosed by the surface $\partial\mathfrak{R}$. A material point of the region in its reference state is located by its rectangular coordinate vector $\mathbf{X}$ at time $t = 0$ whose components are taken with respect to the mutually perpendicular basis set $e = \{\mathbf{e}_1, \mathbf{e}_2, \mathbf{e}_3\}$. For any time $t > 0$, the region deforms to the spatial configuration $\mathfrak{R}_t$ with surface $\partial\mathfrak{R}_t$. The coordinate of the material point in the spatial configuration is given by the vector $\mathbf{x}$. The coordinates in the reference and spatial configurations are related by

$$\mathbf{x} = \chi(\mathbf{X}, t) \tag{1}$$

The deformation gradient tensor is given by

$$F_{iK} = \frac{\partial x_i}{\partial X_K} \tag{2}$$

The right Cauchy-Green deformation tensor is defined as $\mathbf{C} = \mathbf{F}^T \mathbf{F}$, whose eigenvalues are the squares of the principal stretches associated with the deformation of Equation (1). The region is subjected to an electric field vector denoted in $\mathfrak{R}$ as the Lagrangian electric field $\mathbf{E}$.

Over $\mathfrak{R}$, the mass balance is

$$\dot{\rho} = 0 \tag{3}$$

where the superposed dot indicates a material derivative, and ρ is the mass density of the material. The balance of linear momentum, assuming static conditions and no body forces, is

$$Div\,\mathbf{FS} = \mathbf{o} \tag{4}$$

where Div is the divergence operator of with respect to the reference configuration, $\mathbf{S}$ is the second Piola-Kirchhoff stress tensor, and $\mathbf{o}$ is the null vector. The angular momentum balance of the system is simply the proof of the symmetry of the second Piola-Kirchhoff stress tensor

$$\mathbf{S} = \mathbf{S}^T \tag{5}$$

These three balance principles can be easily established using standard techniques [57, 58]. The energy balance in the reference configuration, which includes the energy of the electric field, is [39, 59-61]

$$-\rho \dot{U} + (1/2)\mathbf{S}:\dot{\mathbf{C}} + \mathbf{E}\cdot\dot{\mathbf{D}} + Div\,\mathbf{Q} + \rho h = 0 \tag{6}$$

where U is the specific internal energy, $\mathbf{D}$ is the Lagrangian electric displacement vector, $\mathbf{Q}$ is the heat flux vector, and h is the volumetric thermal heat source. It has been shown that $\mathbf{E}$ and $\mathbf{D}$ are work conjugates for a deformable dielectric [62]. The second law of thermodynamics for the reference configuration is [59]

$$\rho \dot{\eta} - \frac{1}{\theta} Div\,\mathbf{Q} + \left(\frac{1}{\theta^2}\right)\mathbf{Q}\cdot\text{Grad}\,\theta - \frac{\rho h}{\theta} \geq 0 \tag{7}$$

where θ is the temperature and $Grad$ is the gradient function with respect to the reference configuration. The free energy of the systems is defined as [59]

$$\psi = U - \theta\eta - \left(\frac{1}{\rho}\right)\mathbf{E}\cdot\mathbf{D} \tag{8}$$

The Clausius-Duhem inequality is established by substitution of Equations (6) and (8) into (7)

$$-\rho\left(\dot{\theta}\eta + \dot{\psi}\right) + (1/2)\mathbf{S}:\dot{\mathbf{C}} - \dot{\mathbf{E}}\cdot\mathbf{D} + \left(\frac{1}{\theta}\right)\mathbf{Q}\cdot\text{Grad}\,\theta \geq 0 \tag{9}$$

Therefore, Equations (3), (4), (5), and (6) constitute eight scalar field equations for 22 scalar quantities (ρ, $\mathbf{S}$, $\mathbf{x}$, $\mathbf{D}$, ψ, θ, $\mathbf{E}$, and η with h prescribed). Therefore, 14 constitutive equations are required to completely describe the electromechanical behavior of the material.

The appropriate boundary conditions of the electric field and the electric displacement vector are important in the solution of boundary value problems. It has been shown [63] that in the absence of surface charges the normal component of the electric displacement and the tangent component of the electric field with respect to $\partial\mathfrak{R}$ must be continuous across $\partial\mathfrak{R}$.

3. Constitutive Modeling

Similar to Eringen [59], it is assumed that the thermodynamic forces for $\mathfrak{R}$ are $\mathbf{C}$, $\mathbf{E}$, and θ. Therefore, the remaining parameters are thermodynamic fluxes, and these are used to establish the constitutive relations

$$\mathbf{S} = \hat{\mathbf{S}}(\mathbf{C},\mathbf{E},\theta)$$
$$\mathbf{D} = \hat{\mathbf{D}}(\mathbf{C},\mathbf{E},\theta)$$
$$\eta = \hat{\eta}(\mathbf{C},\mathbf{E},\theta)$$
$$\Psi = \hat{\Psi}(\mathbf{C},\mathbf{E},\theta)$$

(10)

where $\Psi = \rho\psi$, and use has been made of the Principle of Equipresence [57, 58], in which a variable present as an independent variable in one constitutive equation is present in all other constitutive equations. Expanding Equation (10)$_4$

$$\dot{\Psi} = \frac{\partial \hat{\Psi}(\mathbf{C},\mathbf{E},\theta)}{\partial \mathbf{C}} : \dot{\mathbf{C}} + \frac{\partial \hat{\Psi}(\mathbf{C},\mathbf{E},\theta)}{\partial \mathbf{E}} \cdot \dot{\mathbf{E}} + \frac{\partial \hat{\Psi}(\mathbf{C},\mathbf{E},\theta)}{\partial \theta} \dot{\theta}$$

(11)

Substitution of Equation (11) into (9) yields

$$\left[\frac{1}{2}\mathbf{S} - \frac{\partial \hat{\Psi}(\mathbf{C},\mathbf{E},\theta)}{\partial \mathbf{C}}\right] : \dot{\mathbf{C}} - \left[\mathbf{D} + \frac{\partial \hat{\Psi}(\mathbf{C},\mathbf{E},\theta)}{\partial \mathbf{E}}\right] \cdot \dot{\mathbf{E}} - \left[\frac{\partial \hat{\Psi}(\mathbf{C},\mathbf{E},\theta)}{\partial \theta} + \rho\eta\right]\dot{\theta} + \left(\frac{1}{\theta}\right)\mathbf{Q}\cdot\mathrm{Grad}\,\theta \geq 0 \quad (12)$$

In general, this modified form of the Clausius-Duhem inequality must hold for arbitrary values of the thermodynamic forces $\dot{\mathbf{C}}$, $\dot{\mathbf{E}}$, and $\dot{\theta}$. Therefore,

$$\mathbf{S} = 2\frac{\partial \hat{\Psi}(\mathbf{C},\mathbf{E},\theta)}{\partial \mathbf{C}}$$
$$\mathbf{D} = -\frac{\partial \hat{\Psi}(\mathbf{C},\mathbf{E},\theta)}{\partial \mathbf{E}}$$
$$\rho\eta = -\frac{\partial \hat{\Psi}(\mathbf{C},\mathbf{E},\theta)}{\partial \theta}$$
$$\left(\frac{1}{\theta}\right)\mathbf{Q}\cdot\mathrm{Grad}\,\theta \geq 0$$

(13)

These equations represent general forms of the constitutive equations. The specific form of $\hat{\Psi}$ depends on the assumptions of material symmetry and behavior.

Whereas many electroactive materials are crystalline (e.g. piezoelectrics), most electrostrictive polymers are composed of amorphous regions that contain small electroactive crystallites attached to polymer chains [17-21]. A schematic of the molecular structure of a typical electrostrictive polymer is shown in Figure 1. Although the crystallites themselves are electroactive, the overall material cannot exhibit a bulk electrostrictive effect unless the crystallites are aligned using a biasing field during processing so that their electroactively-induced strains all act in the same direction with respect to an applied electric field. A schematic of the molecular structure of a polarized electrostrictive polymer is shown in Figure 2. As shown

in Figure 2, the direction of polarization of the electrostrictive polymer creates an axis of symmetry. Along this axis, the crystallites are oriented normal to the axis of symmetry. Each crystallite is oriented at a statistically random angle about the axis of symmetry. For a bulk sample of the electrostrictive polymer, the statistical orientation of the crystallites results in reflectional and rotational symmetries with respect to the axis of symmetry (Figure 2). With these material symmetries, the bulk electrostrictive polymer is classified as a transversely isotropic material [64]. It will be henceforth assumed that the axis of symmetry of $\mathfrak{R}$ is aligned with the $\mathbf{e}_1$ basis vector.
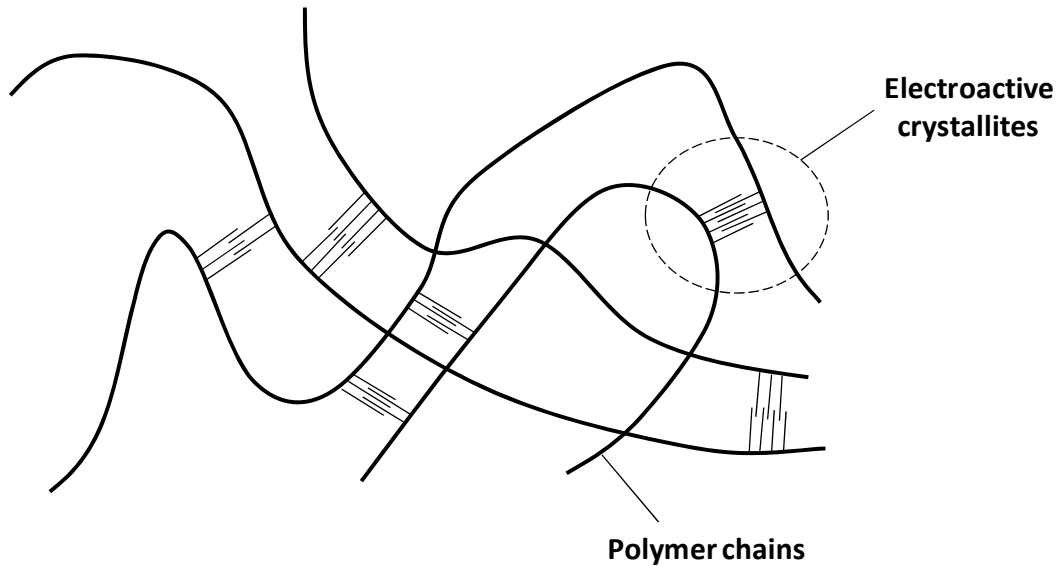

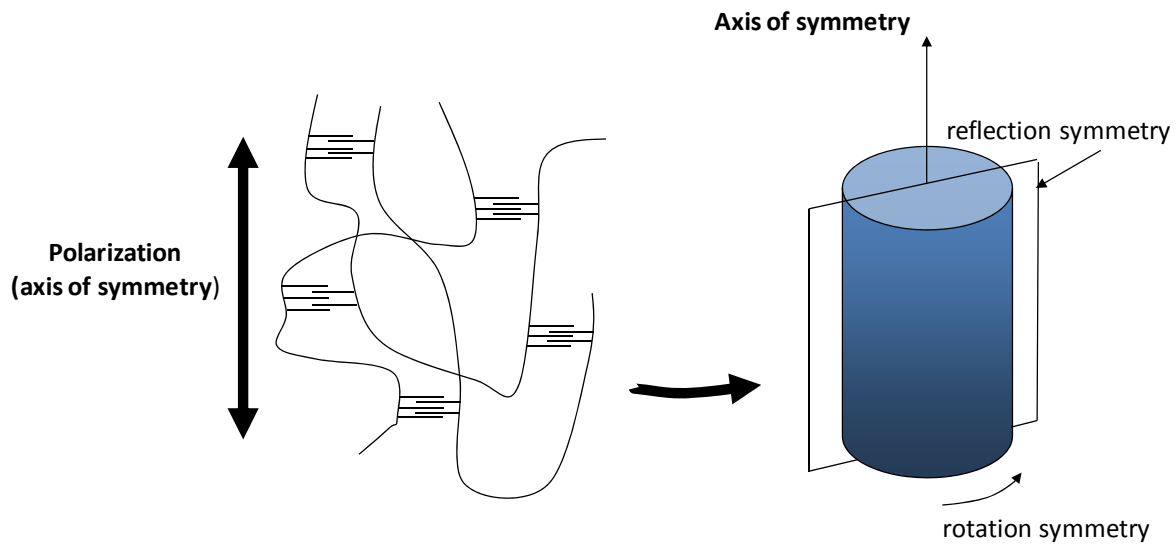*Figure 1 – Schematic of the molecular chains in an unpolarized electrostrictive polymer*

Figure 2 – Molecular structure of a polarized electrostrictive polymer (left) and the resulting bulk-level material symmetry (right)

For the given material symmetry and symmetry axis vector, a set of scalar invariants for $\mathbf{C}$ include $I_1 = tr(\mathbf{C})$, $I_2 = 1/2\left[(\text{tr } \mathbf{C})^2 - \text{tr}(\mathbf{C}^2)\right]$, $I_3 = det\,\mathbf{C}$, $I_4 = C_{11}$, and $I_5 = C_{12}^{\,2} + C_{13}^{\,2}$, where $tr(\bullet)$ denotes the trace operator [64]. The scalar invariants for the vector $\mathbf{E}$ are E_1 and $E_2^2 + E_3^2$. Scalar invariants for the coupling of $\mathbf{C}$ and $\mathbf{E}$ are $E_\alpha C_{1\alpha}$, $E_\alpha C_{\alpha\beta} C_{1\beta}$, and $E_\alpha C_{\alpha\beta} E_\beta$ where subscripts α and β have values of 2 and 3 and obey the summation convention. Together with the scalar invariant θ for temperature, a total of 11 invariants can be used to formulate the functional form of the constitutive equation $(10)_4$.

To simplify this process of establishing a functional form of the constitutive equation $(10)_4$, some additional assumptions need to be made regarding the material behavior. First, it has been shown [21] that while electroactive responses tend to be linear with θ, dielectric responses appear to be nonlinear with θ. Second, it has been observed that electrostrictive materials do not exhibit a reverse electroactive effect [36]. That is, an electric displacement is not generated when a mechanical load is applied to the material in the absence of an electric field, as is observed in piezoelectric materials. Third, residual electric displacements are observed for electroactive polymers that have been polarized [21]. The polarization of the material results in a non-zero $\mathbf{D}$ vector when the electric field vanishes. Fourth, electrostrictive materials exhibit a response that depends on E_1^2, while piezoelectric materials exhibit a dependence on E_1. Fifth, electrostrictive polymers tend to be elastomeric, and thus incompressible. Finally, no electroactive response is expected for the case in which $E_1 = 0$. With these assumptions and the invariants given above, the free energy density is postulated to be

$$\Psi = A_1 + A_2 + A_3 + A_4 \tag{14}$$

where A_1, A_2, A_3, and A_4 are the elastic, electrostrictive, dielectric, and residual electric displacement responses, respectively. The elastic response is given by

$$A_1 = \frac{1}{2}(\alpha_1 + \alpha_2 T)(I_1 - 3) + \frac{1}{2}(\alpha_3 + \alpha_4 T)(I_2 - 3) + \frac{1}{2}\alpha_5 T I_1 - \frac{1}{2}p(I_3 - 1) \tag{15}$$

The electrostrictive response is

$$A_2 = -\frac{1}{2}(\alpha_6 + \alpha_7 T)E_1^2(I_4 - 1) \tag{16}$$

The dielectric response is

$$A_3 = -\frac{1}{2}(\alpha_8 + \alpha_9 T + \alpha_{10}T^2)E_1^2 - \frac{1}{2}(\alpha_{11} + \alpha_{12}T + \alpha_{13}T^2)(E_2^2 + E_3^2) \tag{17}$$

The residual electric displacement is established with

$$A_4 = -D_0 E_1 \tag{18}$$

In Equations (15) - (17), $\alpha_1, \alpha_2, \ldots, \alpha_{13}$ are material parameters, D_0 is the residual electric displacement, and T is the change in the ambient temperature T_0 resulting in the current absolute temperature θ $(\theta = T_0 + T)$. The first two terms on the right-hand side of Equation (15) are the Mooney-Rivlin response of the material (with temperature-dependent material properties), the third term introduces the thermal expansion coefficient, α_5, and the last term enforces incompressibility with the Lagrange multiplier p. Substitution of Equations (14) - (18) into $(13)_1$ and $(13)_2$ yields the constitutive equations for the stress tensor,

$$\mathbf{S} = \left[(\alpha_1 + \alpha_2 T) + I_1 (\alpha_3 + \alpha_4 T) + \alpha_5 T \right] \mathbf{I} - (\alpha_3 + \alpha_4 T) \mathbf{C} - p I_3 \mathbf{C}^{-1} - (\alpha_6 + \alpha_7 T) E_1^2 (\mathbf{e}_1 \otimes \mathbf{e}_1) \tag{19}$$

and the electric displacement vector components

$$\begin{aligned}
D_1 &= (\alpha_6 + \alpha_7 T) E_1 (I_4 - 1) + (\alpha_8 + \alpha_9 T + \alpha_{10} T^2) E_1 + D_0 \\
D_2 &= (\alpha_{11} + \alpha_{12} T + \alpha_{13} T^2) E_2 \\
D_3 &= (\alpha_{11} + \alpha_{12} T + \alpha_{13} T^2) E_3
\end{aligned} \tag{20}$$

where $\mathbf{I}$ is the identity tensor. Note the nonlinear dependence of E_1 and I_4 on D_1 in Equation $(20)_1$. The stress components S_{11}, S_{22}, S_{33}, S_{23}, S_{13}, and S_{12} are expected to vanish in the undeformed state with no applied electrical field and temperature change $(\mathbf{C} = \mathbf{I}, I_1 = 3, I_4 = 1, \mathbf{E} = \mathbf{o}, T = 0)$. Under this condition, Equation (19) reveals

$$p = \alpha_1 + 2\alpha_3 \tag{21}$$

To simplify these expressions, the following quantities are defined

$$\begin{aligned}
\beta_1(T) &= \alpha_1 + \alpha_2 T \\
\beta_2(T) &= \alpha_3 + \alpha_4 T \\
\beta_3(T) &= \alpha_5 T \\
\beta_4(T) &= \alpha_6 + \alpha_7 T \\
\beta_5(T) &= \alpha_8 + \alpha_9 T + \alpha_{10} T^2 \\
\beta_6(T) &= \alpha_{11} + \alpha_{12} T + \alpha_{13} T^2
\end{aligned} \tag{22}$$

which are clearly temperature-dependent material properties, which are necessary to model with polymer materials below the glass transition temperature. Substitution of Equation (22) into Equations (19) - (20) results in a modified form of the constitutive equations for stress,

$$\mathbf{S} = \left[\beta_1 + I_1\beta_2 + \beta_3 \right] \mathbf{I} - \beta_2\mathbf{C} - pI_3\mathbf{C}^{-1} - \beta_4 E_1^2 \left(\mathbf{e}_1 \otimes \mathbf{e}_1 \right) \tag{23}$$

and electrical displacement

$$
\begin{aligned}
D_1 &= \beta_4 E_1 \left(I_4 - 1 \right) + \beta_5 E_1 + D_0 \\
D_2 &= \beta_6 E_2 \\
D_3 &= \beta_6 E_3
\end{aligned}
\tag{24}
$$

where the temperature arguments associated with the temperature-dependent material parameters have been removed for brevity. Equations (23) - (24) describe the general three-dimensional electromechanical behavior of an incompressible electrostrictive polymer material. Equation (14) can be expressed in terms of the temperature-dependent parameters of Equation (22)

$$
\begin{aligned}
\Psi = &\frac{1}{2}\beta_1\left(I_1 - 3\right) + \frac{1}{2}\beta_2\left(I_2 - 3\right) + \frac{1}{2}\beta_3 I_1 - \frac{1}{2}p\left(I_3 - 1\right) \\
&- \frac{1}{2}\beta_4 E_1^2\left(I_4 - 1\right) - \frac{1}{2}\beta_5 E_1^2 - \frac{1}{2}\beta_6\left(E_2^2 + E_3^2\right) - P_0 E_1
\end{aligned}
\tag{25}
$$

Equation (25) is in a form that can be conveniently used to determine the response for specific electrostrictive polymer materials, which is demonstrated in the next section.

4. Material parameters

To demonstrate the functionality of the proposed constitutive model, the material parameters need to be quantitatively established for a sampling of electrostrictive polymer materials. Using data from the literature [18], the values of some of the material constants in Equations (23) - (24) have been determined at room temperature for three electrostrictive polyurethane material systems: PS 2000, PS 1000, and PM 2000. Details on these material systems may be found elsewhere [18]. The electromechanical properties of the materials were established by equating the mechanical components of the free energy density given by Equation (25) with an equivalent expression found elsewhere [65], with which the material parameters from the literature [18] have been characterized. The equating of the free energies was performed for four different boundary conditions: shear deformation with no applied electric field, axial elongation with no applied electric field, transversely-applied electric field, and axial elongation with an applied electric field. It is important to note that the energy expression from the literature [65] was not established in a large-deformation hyperelastic framework, so the applied deformations in the current analysis were relatively small.

For each prescribed deformation and electric field, material parameters were determined by minimizing the sum of the squares of the errors between the free energies from the proposed

model and those found in the literature. The material parameters were iteratively updated to minimize the differences in the free energies. As a result, the proposed model was characterized for the three material systems. The applied isochoric deformations and electric fields were specifically chosen to efficiently determine the material parameters in the constitutive equations given by Equations (23) - (24). The details of each deformation are given below.

4.1 Shear deformation with no applied electric field

For the case of simple shear deformation at the ambient temperature and no applied electric field $(\mathbf{E} = \mathbf{o})$, Equation (1) is

$$
\begin{aligned}
x_1 &= X_1 + \gamma X_2 \\
x_2 &= X_2 \\
x_3 &= X_3
\end{aligned}
\tag{26}
$$

where γ is the shear strain. The components of $\mathbf{C}$ with respect to the basis set are $C_{11} = C_{33} = 1$, $C_{22} = 1 + \gamma^2$, $C_{12} = \gamma$, and $C_{23} = C_{13} = 0$. Based on these assumptions, Equation (25) simplifies to

$$
\Psi = \frac{1}{2}(\beta_1 + \beta_2)\gamma^2
\tag{27}
$$

Together with the proceeding applied deformation, comparison of the strain energy function from the literature [65] with the corresponding material properties [18] for deformations in the range of $\gamma = 0 \rightarrow 0.02$ established the values of β_1 and β_2 shown in Table 1.

Table 1 – Isotheral electromechanical material parameters for electrostrictive polymers at room temperature

	β_1 (MPa)	β_2 (MPa)	β_3 (MPa)	β_4 (MPa)m^2/V^2	β_5 (MPa)m^2/V^2	β_6 (MPa)m^2/V^2
PS 2000	11.2	1.6	0.0	-2.1(10^{-16})	5.4(10^{-17})	5.4(10^{-17})
PS 1000	6.3	0.9	0.0	1.6(10^{-15})	6.4(10^{-17})	6.4(10^{-17})
PM 2000	2.2	0.3	0.0	8.4(10^{-16})	6.9(10^{-17})	6.9(10^{-17})

4.2 Axial elongation with no applied electric field

For the case of a uniaxial tension along the $\mathbf{e}_1$ axis at the ambient temperature with no applied electric field, Equation (1) is

$$x_1 = \lambda X_1$$
$$x_2 = \frac{1}{\sqrt{\lambda}} X_2 \qquad\qquad (28)$$
$$x_3 = \frac{1}{\sqrt{\lambda}} X_3$$

where λ is the principal stretch along the $\mathbf{e}_1$ axis. The components of $\mathbf{C}$ with respect to the basis set are $C_{11} = \lambda^2$, $C_{22} = C_{33} = 1/\lambda$, and $C_{23} = C_{13} = C_{12} = 0$. Based on these assumptions, Equation (25) simplifies to

$$\Psi = \frac{1}{2}\beta_1\left(\lambda^2 + \frac{2}{\lambda} - 3\right) + \frac{1}{2}\beta_2\left(2\lambda + \frac{1}{\lambda^2} - 3\right) \qquad (29)$$

With the assumption that $\beta_1/\beta_2 = 7$ [66], the sum of the squares of the error where minimized between Equations (27) and (29) and the free energy found from the literature [65] for the range of $1 \le \lambda < 1.02$. The resulting values of β_1 and β_2 are listed in Table 1 for all three polymer systems. These two material parameters represent the hyperelastic response of the electrostrictive polymers in the absence of an electric field.

4.3 Transversely-applied electric field

For the case of no deformation at the ambient temperature and an applied electric field of $\mathbf{E} = E\mathbf{e}_2 + E\mathbf{e}_3$, Equation (25) simplifies to

$$\Psi = -\beta_6 E^2 \qquad (30)$$

Comparison of Equation (30) with the small-deformation free-energy equation [65] reveals that β_6 is the permittivity in the $\mathbf{e}_2$ and $\mathbf{e}_3$ directions, which are perpendicular to the axis of symmetry. If it is assumed that permittivity in the $\mathbf{e}_1$ direction is the same, then it is apparent from Equation (25) that $\beta_5 = \beta_6$. From the data on the three polymer systems [18], the resulting values of β_5 and β_6 are shown in Table 1.

4.4 Axial elongation with applied electric field

For the case of uniaxial tension along the $\mathbf{e}_1$ axis at the ambient temperature and an applied electric field of $\mathbf{E} = E\mathbf{e}_1$ with no initial electric displacement, Equation (1) is

$$x_1 = \lambda X_1$$

$$x_2 = \frac{1}{\sqrt{\lambda}} X_2 \qquad\qquad (31)$$

$$x_3 = \frac{1}{\sqrt{\lambda}} X_3$$

where λ is the principal stretch along the $\mathbf{e}_1$ axis. The components of $\mathbf{C}$ with respect to the basis set are $C_{11} = \lambda^2$, $C_{22} = C_{33} = 1/\lambda$, and $C_{23} = C_{13} = C_{12} = 0$. For this condition, Equation (25) simplifies to

$$\Psi = \frac{1}{2}\beta_1\left(\lambda^2 + \frac{2}{\lambda} - 3\right) + \frac{1}{2}\beta_2\left(2\lambda + \frac{1}{\lambda^2} - 3\right) - \frac{1}{2}\beta_4 E^2\left(\lambda^2 - 1\right) - \frac{1}{2}\beta_5 E^2 \qquad (32)$$

Although the assumption of no initial electric displacement is not realistic, it is necessary here for illustrative purposes because the formulation from the literature [18] did not include it. Therefore, for an accurate characterization of the current model using data from the model of ref [18], the initial electric displacement must be set to zero. Because the material parameters β_1, β_2, β_5, and β_6 have been determined with the preceding loading conditions, the only unknown parameter in Equation (32) is the material parameter β_4 (β_3 is zero-valued at room temperature) which serves as a electro-mechanical coupling parameter, which is analogous to the piezoelectric constant in piezoelectric materials. For each material, the value of β_4 was determined by comparing the free energies from Equation (32) and from the literature [18], and minimizing the sum of the squares of the errors between the two energies my iteratively adjusting the β_4 parameter. The energies were compared for a deformation range of $1 \le \lambda < 1.02$ and an applied electric field of $E = 1\,GV$. The resulting values of β_4 for the three electrostrictive polymers are listed in Table 1.

5. Conclusions

The characterization of the model in this paper demonstrates its functional simplicity. The scalar free-energy density expression of Equation (25) and the resulting constitutive relationships of Equations (23) and (24) have a relatively simple form and are very powerful in that they can describe the large-deformation behavior of the material as a function of temperature, applied deformations, and applied electric fields. Although a limited number of material parameters were established for the three electrostrictive elastomers, a full set of electro-thermal-mechanical data on these materials could be used to fully characterize the model.

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