On Electromechanical Coupling in Elastomers

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Permittivity of electroactive elastomers alters during deformation. The influence of the permittivity alterations on the electrostriction of elastomers is studied in the present work. Particularly, acrylic elastomer VHB 4910 is considered. A polarization–electric field constitutive theory is introduced accounting for the influence of mechanical deformations. The theory is used to analyze a free electrostriction of a thin elastomer plate. The elastic stress in the plate is described by various constitutive models including neo-Hookean, Yeoh, Arruda-Boyce, and Ogden. Results show that the permittivity alterations during mechanical deformation practically do not affect the process of electrostriction. [DOI: 10.1115/1.4006057]

1 Introduction

Electrostriction is a property of dielectrics that causes them to change their shape under the application of an electric field. Electrostriction in hard materials is known and used in applications for a long time [1–3]. Electrostriction in soft elastomers is a relatively new topic which is attractive in view of the versatile potential applications in sensors and actuators [4–11].

Various theories of the electromechanical coupling at large strains were formulated [12–17]. Most recently, three papers appeared [18–20] which summarized the developments of nonlinear electroelasticity taking different paths for the derivation of equations and using different notational schemes. It is noteworthy that the latter works put a special emphasis on the addition of the energy contribution of the electric field to the general expression of the Helmholtz free energy in contrast to the more traditional use of the Maxwell electric stress.

In the case where the electric permittivity of the medium is constant, both approaches based on the extended energy functions and Maxwell stresses may lead to the same results and the difference in formulation is a matter of taste. However, in the case where the electric permittivity of the medium is not constant and it depends on strains, the difference between the energy and Maxwell stress formulations becomes sound. Indeed, when the electric permittivity depending on strains is a factor in the extended energy function, then the stress tensor should depend on the derivatives of the electric permittivity with respect to strains. Such a dependence led Zhao and Suo [21], for example, to a conclusion that elastomers might exist which thicken rather than thin under an applied electric field.

In the present work we study the electrostriction of acrylic elastomer VHB 4910 by taking a conservative and simple path of coupling the elastic and Maxwell stresses enhanced with the electric permittivity of the medium depending on strains. Particularly, we consider a linear dependence of the permittivity on the first invariant of the Cauchy-Green tensor. This dependence is fitted to the results of the experimental measurements by Wissler and Mazza

[22]. The calibrated model coupled with various constitutive elastic theories, including neo-Hookean, Yeoh, Arruda-Boyce, and Ogden, is used for analysis of a free electrostriction of a thin elastomer plate. Results of the analysis show that the permittivity alterations during mechanical deformation practically do not affect the process of electrostriction.

The paper is organized as follows. The field equations are summarized in Sec. 2 and the constitutive theories are considered in Sec. 3. The electrostriction of the a thin elastomer plate is analyzed in Sec. 4 and the general conclusions are drawn in Sec. 5.

2 Field Equations

The electric field $\mathbf{E}$ and the electric displacement $\mathbf{D}$ obey the following equations of electrostatics in the absence of volumetric charges:

$$\text{curl}\,\mathbf{E} = \mathbf{0}, \quad \text{div}\,\mathbf{D} = 0 \tag{1}$$

where the differential operators are defined with respect to spatial coordinates $\mathbf{y}$.

These equations are accompanied by the conditions on interface A:

$$(\mathbf{E}_1 - \mathbf{E}_2) \times \mathbf{n} = \mathbf{0}, \quad (\mathbf{D}_2 - \mathbf{D}_1) \cdot \mathbf{n} = q_A \tag{2}$$

Here $\mathbf{n}$ designates a unit normal to the surface; $\mathbf{E}_1, \mathbf{D}_1$ and $\mathbf{E}_2, \mathbf{D}_2$ designate the electric field and electric displacement in half-spaces separated by the surface; and q_A is the surface charge.

The electric displacement and electric field are related as follows:

$$\mathbf{D} = \varepsilon_0 \mathbf{E} + \mathbf{P} \tag{3}$$

where $\varepsilon_0 = 8.854 \times 10^{-12}$ (F/m) is the permittivity of free space and $\mathbf{P}$ is the polarization vector characterizing the material properties under the imposed electric field

$$\mathbf{P} = \mathbf{P}(\mathbf{E}) \tag{4}$$

The simplest and widely used form of (4) in the case of isotropic media is the linear constitutive model

$$\mathbf{P} = \chi \varepsilon_0 \mathbf{E} \tag{5}$$

where χ is the electric susceptibility of the medium.

Substituting (5) in (3) we get

$$\mathbf{D} = \varepsilon_0 \varepsilon_r \mathbf{E} \tag{6}$$

where $\varepsilon_r = 1 + \chi$ is the dielectric constant.

Usually the dielectric constant is assumed to be an invariable material parameter. However, in the case of large deformations of elastomers this material parameter varies with strains. We postpone the description of the variation to the next section.

The electric field creates body forces $\mathbf{R}$ and couples $\mathbf{M}$ on material particles. The expressions for the electric body forces and couples vary with various theoretical frameworks [16]. However, all of them reduce to the same form in the case of electrostatics and zero distributed body charge $q = 0$,

$$\mathbf{R} = (\text{grad}\,\mathbf{E})\mathbf{P}, \quad \mathbf{M} = \mathbf{P} \times \mathbf{E} \tag{7}$$

We notice with account of (5) that the body couple equals zero: $\mathbf{M} = \mathbf{0}$.

Following Maxwell's idea for magnetism, it is convenient to present the electric body force as a divergence of the Maxwell "stress" tensor $\boldsymbol{\sigma}^M$,

$$\mathbf{R} = (\text{grad}\,\mathbf{E})\mathbf{P} = \text{div}\,\boldsymbol{\sigma}^M \tag{8}$$

Such a representation is not unique and it can be specialized as

$$\boldsymbol{\sigma}^M = \mathbf{E} \otimes (\varepsilon_0 \mathbf{E} + \mathbf{P}) - \frac{\varepsilon_0}{2} (\mathbf{E} \cdot \mathbf{E}) \mathbf{1} \tag{9}$$

or, accounting for (6),

$$\boldsymbol{\sigma}^M = \varepsilon_0 \varepsilon_r \mathbf{E} \otimes \mathbf{E} - \frac{\varepsilon_0}{2} (\mathbf{E} \cdot \mathbf{E}) \mathbf{1} \tag{10}$$

Subjected to the electric field, elastomers undergo deformations that can be effectively described by means of continuum mechanics. In continuum mechanics the atomistic or molecular structure of material is approximated by a continuously distributed set of the so-called material points or particles. The continuum material point is an abstraction that is used to designate a small (infinitesimal) representative volume of real material including many atoms and molecules. A material point that occupies position $\mathbf{x}$ in the reference configuration moves to position $\mathbf{y}(\mathbf{x})$ in the current configuration of the continuum. The deformation in the vicinity of the material point can be completely described by the deformation gradient tensor:

$$\mathbf{F} = \partial \mathbf{y} / \partial \mathbf{x} \tag{11}$$

Using the deformation gradient it is possible to introduce a convenient deformation measure—the right Cauchy-Green tensor, which is not affected by the rigid body motion

$$\mathbf{C} = \mathbf{F}^T \mathbf{F} \tag{12}$$

In the case of elastic continuum the true (Cauchy) stress is related to strain (12) with the following constitutive equation:

$$\boldsymbol{\sigma} = 2\mathbf{F} \frac{\partial \psi}{\partial \mathbf{C}} \mathbf{F}^T \tag{13}$$

where $\psi(\mathbf{C})$ is the strain energy per unit material volume in the reference configuration.

The material equilibrium equations in the electric field take form

$$\operatorname{div} \boldsymbol{\sigma} + \mathbf{R} = \mathbf{0} \tag{14}$$

Governing Eqs. (11)–(14) should be completed with boundary conditions on tractions

$$\boldsymbol{\sigma} \mathbf{n} = \bar{\mathbf{t}} \tag{15}$$

or placements

$$\mathbf{y} = \bar{\mathbf{y}} \tag{16}$$

where $\mathbf{n}$ is the unit outward normal to the surface of the continuum and barred quantities are prescribed.

Combining the elastic and Maxwell stresses it is possible to introduce the symmetric total stress

$$\tilde{\boldsymbol{\sigma}} = \boldsymbol{\sigma} + \boldsymbol{\sigma}^M \tag{17}$$

which obeys the equilibrium equation (14) without body forces

$$\operatorname{div} \tilde{\boldsymbol{\sigma}} = \mathbf{0} \tag{18}$$

3 Constitutive Theories

Proceeding with the formulation of the boundary value problem of electromechanics of elastomers, we discuss constitutive theories in the present section.

In the case of isotropic elastomers, constitutive theories in terms of invariants or principal stretches are possible. We start with the description in terms of invariants where the constitutive equations for the elastic stress can be written as follows:

$$\boldsymbol{\sigma} = -p\mathbf{1} + 2(\psi_1 + I_1 \psi_2) \mathbf{B} - 2\psi_2 \mathbf{B}^2 \tag{19}$$

where

$$\mathbf{B} = \mathbf{F} \mathbf{F}^T \tag{20}$$

is the left Cauchy-Green tensor and p is the undefined Lagrange multiplier enforcing the incompressibility condition

$$\det \mathbf{F} = 1 \tag{21}$$

Besides, we defined $\psi_a \equiv \partial \psi / \partial I_a$, where I_1 and I_2 are the principal invariants of Cauchy-Green tensors

$$
\begin{aligned}
I_1 &= \operatorname{tr} \mathbf{C} = \operatorname{tr} \mathbf{B}, \\
I_2 &= \{(\operatorname{tr} \mathbf{C})^2 - \operatorname{tr}(\mathbf{C}^2)\}/2 = \{(\operatorname{tr} \mathbf{B})^2 - \operatorname{tr}(\mathbf{B}^2)\}/2
\end{aligned}
\tag{22}
$$

It remains only to define the strain energy function (SEF) ψ. We will consider four SEFs for the acrylic elastomer VHB 4910.

First is the neo-Hookean SEF

$$\psi = c_1(I_1 - 3) \tag{23}$$

where the shear modulus is $c_1 = 0.08$ (MPa).

Second is the Yeoh SEF

$$\psi = c_1(I_1 - 3) + c_2(I_1 - 3)^2 + c_3(I_1 - 3)^3 \tag{24}$$

where the constants are $c_1 = 0.0827$ (MPa), $c_2 = -7.47 \times 10^{-4}$ (MPa), $c_3 = 5.86 \times 10^{-6}$ (MPa)—see [23] for calibration.

Third is the Arruda-Boyce SEF

$$
\begin{aligned}
\psi = \alpha \Big\{ &\frac{1}{2}(I_1 - 3) + \frac{1}{20N}(I_1^2 - 9) + \frac{11}{1050N^2}(I_1^3 - 27) \\
&+ \frac{19}{7000N^3}(I_1^4 - 81) + \frac{519}{673750N^4}(I_1^5 - 243) \Big\}
\end{aligned}
\tag{25}
$$

where the constants are $\alpha = 0.0686$ (MPa), $N = 124.88$—see [23] for calibration.

Alternatively, we can use the material description in terms of the principal stresses and stretches. In this case the constitutive equations take form

$$\sigma_1 = \lambda_1 \frac{\partial \psi}{\partial \lambda_1} - p, \quad \sigma_2 = \lambda_2 \frac{\partial \psi}{\partial \lambda_2} - p, \quad \sigma_3 = \lambda_3 \frac{\partial \psi}{\partial \lambda_3} - p \tag{26}$$

where the strain energy depends on principal stretches $\psi(\lambda_1, \lambda_2, \lambda_3)$.

We will consider the fourth SEF in Ogden's form in terms of principal stretches

$$\psi = \sum_{n=1}^{2} \frac{\mu_n}{\alpha_n} (\lambda_1^{\alpha_n} + \lambda_2^{\alpha_n} + \lambda_3^{\alpha_n} - 3) \tag{27}$$

where the constants are $\mu_1 = 0.04356$ (MPa), $\mu_2 = 0.000117$ (MPa), $\alpha_1 = 1.445$, $\alpha_2 = 4.248$—see [24] for calibration.

Besides the constitutive theories for elastic stresses, we will consider a constitutive theory for the dependence of polarization on the electric field. Specifically, we assume that (5) and (6) hold, yet the dielectric constant is a function of the invariants of the Cauchy-Green tensors defined in (22):

$$\varepsilon_r = \varepsilon_r(I_1, I_2) \tag{28}$$

We further make the simplest assumption that the dielectric parameter depends linearly on the first invariant only

$$\varepsilon_r = \beta_0 + \beta_1(I_1 - 3) \tag{29}$$

Here the second term in parentheses provides the invariable value for the dielectric parameter $\varepsilon_r = \beta_0$ in the absence of deformation $I_1 = 3$.
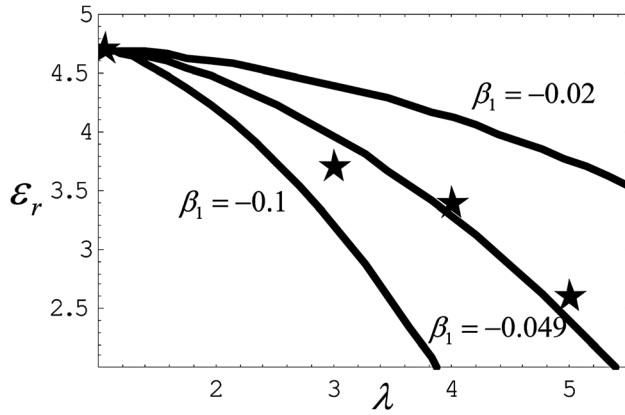
Fig. 1 Experiment (stars) versus theory (solid line) for various dielectric parameters

To fit coefficients β_0 and β_1 we used the experimental results of Wissler and Mazza [22] who measured ε_r for various magnitudes of the equal biaxial stretch, $\lambda = \lambda_1 = \lambda_2 = \lambda_3^{-1/2}$, of a thin elastomer plate. Figure 1 presents experimental points and analytical curve defined by (29) for

$$\beta_0 = 4.7, \quad \beta_1 = -0.1; \; -0.049; \; -0.02 \tag{30}$$

We will use constitutive theories defined by (23)–(25), (27), (29), and (30) in the example considered in the following section.

4 Plate Electrostriction

We consider now a typical example of electrostriction where compliant electrodes cover the top and bottom of elastomer plates (Fig. 2). When charged the plate gets thinner. We can describe the process analytically.

We assume that all fields are homogeneous and; consequently, field equations (1) and (18) are obeyed identically.

Boundary conditions can be written as follows:

$$\varepsilon_0 (\mathbf{E}_2 - \varepsilon_r \mathbf{E}_1) \cdot \mathbf{n} = q_A \tag{31}$$

$$(\mathbf{E}_1 - \mathbf{E}_2) \times \mathbf{n} = \mathbf{0} \tag{32}$$

$$(\tilde{\boldsymbol{\sigma}}_1 - \tilde{\boldsymbol{\sigma}}_2)\mathbf{n} = \mathbf{0} \tag{33}$$

where $\mathbf{E}_1, \mathbf{E}_2$ and $\tilde{\boldsymbol{\sigma}}_1, \tilde{\boldsymbol{\sigma}}_2$ are electric fields and generalized stresses inside and outside the plate accordingly.

We further present the deformation and electric fields in the form

$$\mathbf{F} = \lambda^{-1/2}(\mathbf{e}_1 \otimes \mathbf{e}_1 + \mathbf{e}_2 \otimes \mathbf{e}_2) + \lambda \mathbf{e}_3 \otimes \mathbf{e}_3 \tag{34}$$

$$\mathbf{E}_1 = E\mathbf{e}_3, \quad \mathbf{E}_2 = \mathbf{0} \tag{35}$$

where $\mathbf{e}_1, \mathbf{e}_2, \mathbf{e}_3$ are the Cartesian base vectors and the lateral stretch equals the ratio of the plate thicknesses after and before straining.
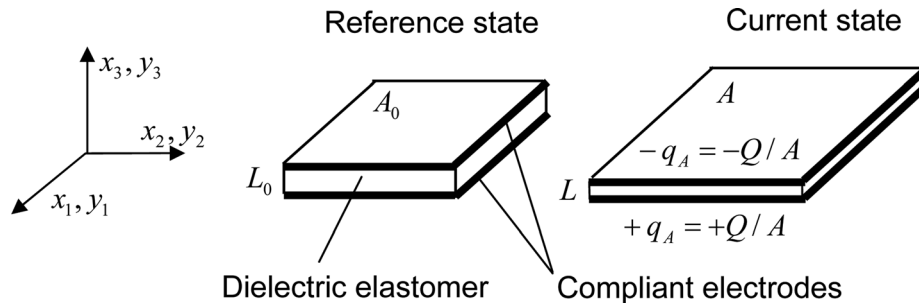
$$\lambda = \frac{L}{L_0} \tag{36}$$

We remind that the material is incompressible, and; consequently, with account of (36) we have

$$AL = A_0 L_0, \quad A = A_0/\lambda \tag{37}$$

Substituting (35) and (37) in boundary conditions (31) and (32) we get

$$E = \frac{q_A}{\varepsilon_0 \varepsilon_r} = \frac{Q}{\varepsilon_0 \varepsilon_r A} = \frac{Q\lambda}{\varepsilon_0 \varepsilon_r A_0} \tag{38}$$

Substituting (34) and (35) in (10), (19), and (17) we have

$$\tilde{\sigma}_{11} = \tilde{\sigma}_{22} = -p + 2(\psi_1 + I_1 \psi_2)\lambda^{-1} - 2\psi_2 \lambda^{-2} - \frac{\varepsilon_0}{2}\left(\frac{Q\lambda}{\varepsilon_0 \varepsilon_r A_0}\right)^2 \tag{39}$$

$$\tilde{\sigma}_{33} = -p + 2(\psi_1 + I_1 \psi_2)\lambda^2 - 2\psi_2 \lambda^4 \\ + \varepsilon_r \varepsilon_0^2 \left(\frac{Q\lambda}{\varepsilon_r \varepsilon_0 A_0}\right)^2 - \frac{\varepsilon_0}{2}\left(\frac{Q\lambda}{\varepsilon_r \varepsilon_0 A_0}\right)^2 \tag{40}$$

Assuming the stress-free deformation $\tilde{\sigma}_{11} = \tilde{\sigma}_{22} = \tilde{\sigma}_{33} = 0$ that obeys (33) and subtracting (40) from (39) we get

$$\frac{2\varepsilon_r}{\lambda^2}\{(\psi_1 + I_1 \psi_2)(\lambda^{-1} - \lambda^2) - \psi_2(\lambda^{-2} - \lambda^4)\} = \frac{Q^2}{A_0^2} \tag{41}$$

This equation allows us to calculate the lateral stretch λ and voltage $\Phi = EL$ for the given charge Q. Such calculations are presented graphically in Figs. 3–5 for the material models defined by Eqs. (23)–(25) accordingly.

In the case of the Ogden theory described in terms of the principal stresses and stretches we have modified (39) and (40) as follows:

$$\tilde{\sigma}_1 = \tilde{\sigma}_2 = -p + \lambda_1 \frac{\partial \psi}{\partial \lambda_1} - \frac{\varepsilon_0}{2}\left(\frac{Q\lambda}{\varepsilon_0 \varepsilon_r A_0}\right)^2 \tag{42}$$

$$\tilde{\sigma}_3 = -p + \lambda_3 \frac{\partial \psi}{\partial \lambda_3} + \varepsilon_r \varepsilon_0^2 \left(\frac{Q\lambda}{\varepsilon_r \varepsilon_0 A_0}\right)^2 - \frac{\varepsilon_0}{2}\left(\frac{Q\lambda}{\varepsilon_r \varepsilon_0 A_0}\right)^2 \tag{43}$$

Assuming the stress-free deformation $\tilde{\sigma}_1 = \tilde{\sigma}_2 = \tilde{\sigma}_3 = 0$ that obeys (33) and subtracting (40) from (39) we get

$$\frac{2\varepsilon_r}{\lambda^2}\left\{\lambda_1 \frac{\partial \psi}{\partial \lambda_1} - \lambda_3 \frac{\partial \psi}{\partial \lambda_3}\right\} = \frac{Q^2}{A_0^2} \tag{44}$$

Substituting (27) and (34) in (44) we have

$$\frac{2\varepsilon_r}{\lambda^2}\sum_{n=1}^{2}\mu_n(\lambda^{-\alpha_n/2} - \lambda^{\alpha_n}) = \frac{Q^2}{A_0^2} \tag{45}$$
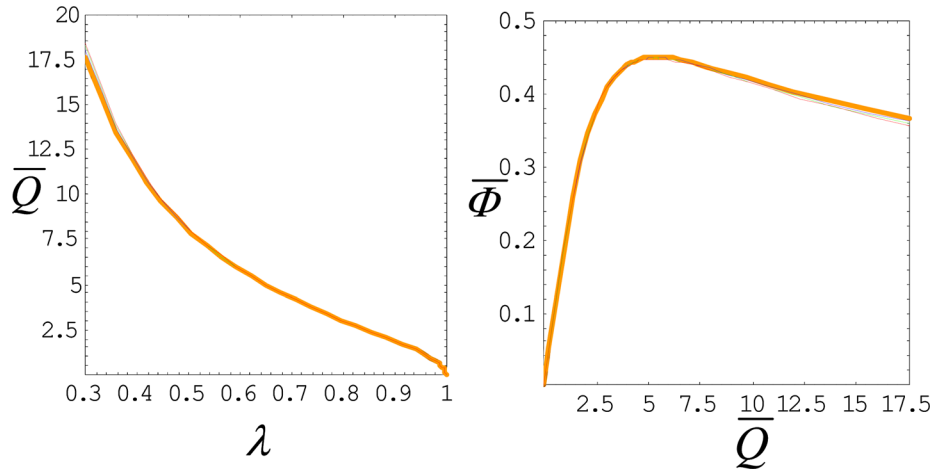

Fig. 2 Electroactive elastomer plate

Fig. 3 Dimensionless charge $\bar{Q} = Q/(A_0\sqrt{c_1})$ versus lateral stretch λ and dimensionless voltage $\bar{\Phi} = \Phi\,\varepsilon_0/(L_0\sqrt{c_1})$ for neo-Hookean model: curves for $\beta_1 = -0.1$, $\beta_1 = -0.049$, $\beta_1 = -0.02$, $\beta_1 = 0$ coincide
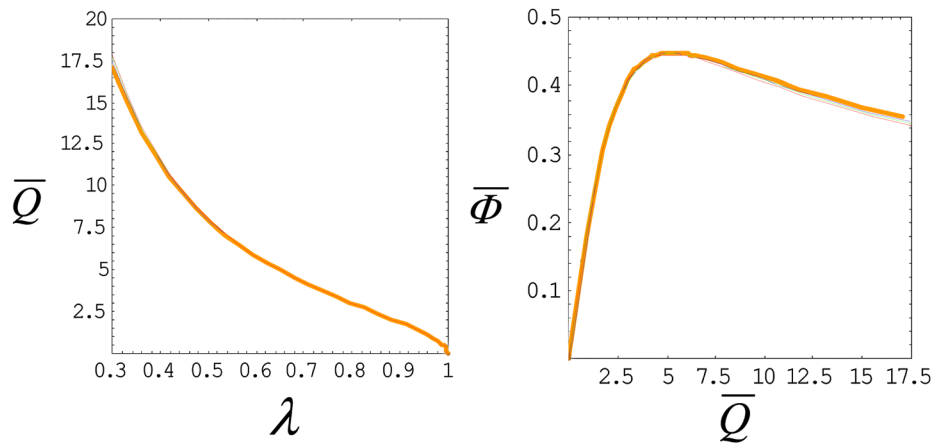

Fig. 4 Dimensionless charge $\bar{Q} = Q/(A_0\sqrt{c_1})$ versus lateral stretch λ and dimensionless voltage $\bar{\Phi} = \Phi\,\varepsilon_0/(L_0\sqrt{c_1})$ for Yeoh model: curves for $\beta_1 = -0.1$, $\beta_1 = -0.049$, $\beta_1 = -0.02$, $\beta_1 = 0$ coincide
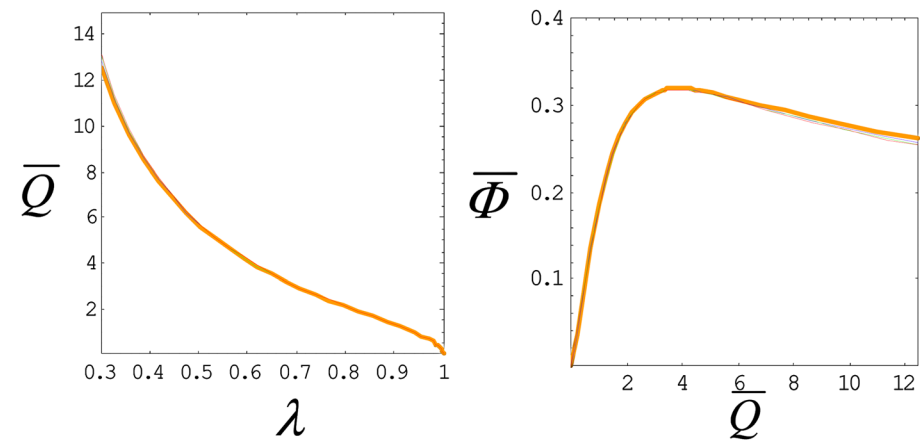

Fig. 5 Dimensionless charge $\bar{Q} = Q/(A_0\sqrt{\alpha})$ versus lateral stretch λ and dimensionless voltage $\bar{\Phi} = \Phi\,\varepsilon_0/(L_0\sqrt{\alpha})$ for Arruda-Boyce model: curves $\beta_1 = -0.1$, $\beta_1 = -0.049$, $\beta_1 = -0.02$, $\beta_1 = 0$ coincide
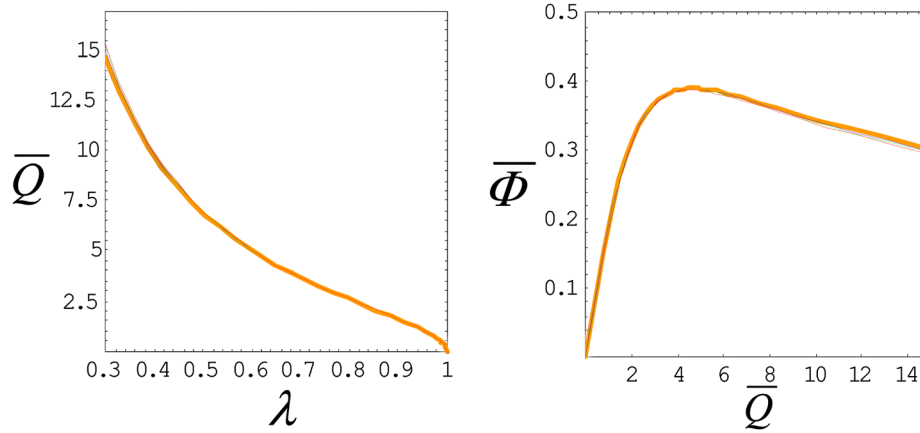
Fig. 6 Dimensionless charge $\bar{Q} = Q/(A_0\sqrt{\mu_1})$ versus lateral stretch λ and dimensionless voltage $\bar{\Phi} = \Phi\,\varepsilon_0/(L_0\sqrt{\mu_1})$ for Ogden model: curves $\beta_1 = -0.1$, $\beta_1 = -0.049$, $\beta_1 = -0.02$, $\beta_1 = 0$ coincide

This formula is presented graphically in Fig. 6

Results show that the permittivity alterations of the dielectric during mechanical deformation practically do not affect the process of electrostriction.

5 Concluding Remarks

A theory of nonlinear electroelasticity has been considered, which incorporated the experimentally calibrated dependence of the material electric permittivity on mechanical strains. The theory stemmed from the Maxwell stress concept combined with the hyperelastic stress formulations. It is shown based on the developed theory that the dependence of the material electric permittivity on mechanical strains has a minor effect on the free electrostriction of a thin elastomer plate.

It is noteworthy that the present study was limited by one material and one specific loading case. Though both the material and the loading case are of the central importance in applications, it can occur that different materials and loadings will lead to the different conclusions. We hope that our results will encourage and guide the experimentalists to examine a variety of materials and loadings.

Acknowledgment

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