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NONEQUILIBRIUM THERMODYNAMICS OF DIELECTRIC ELASTOMERS

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This paper describes an approach to construct models of dielectric elastomers undergoing dissipative processes, such as viscoelastic, dielectric and conductive relaxation. This approach is guided by nonequilibrium thermodynamics, characterizing the state of a dielectric elastomer with kinematic variables through which external loads do work, as well as internal variables that describe the dissipative processes. Within this approach, a method is developed to calculate the critical condition for electromechanical instability. This approach is illustrated with a specific model of a viscoelastic dielectric elastomer, which is fitted to stress-strain curves of a dielectric elastomer (VHB tape), measured at various strain rates. The model shows that a higher critical voltage can be achieved by applying a constant voltage for a shorter time, or by applying ramping voltage with a higher rate. A viscoelastic dielectric elastomer can attain a larger strain of actuation than an elastic dielectric elastomer.

Keywords: Viscoelasticity; dielectric elastomer; actuator; electromechanical instability.

1. Introduction

Subject to a voltage across its thickness, a membrane of a dielectric elastomer reduces in thickness and expands in area. This phenomenon is being developed as a mechanism of electromechanical transduction, of large deformation, low cost, light weight, high efficiency, and noise-free operation [Pelrine *et al.*, 2000; Ha *et al.*, 2006; Zhao and Suo, 2010]. Emerging applications include soft robotics [Bar-Cohen, 2001; Pelrine *et al.*, 2002; O'Halloran *et al.*, 2008], artificial limbs [Biddiss and Chau,

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2008], energy harvesters [Pelrine *et al.*, 2001; Koh *et al.*, 2009], Braille displays [Bar-Cohen, 2009], bio-stimulation pads [Carpi *et al.*, 2009, 2010], and adaptive optics [Beck *et al.*, 2009; Kofod *et al.*, 2009; Keplinger *et al.*, 2010].

Suo [2010] has recently reviewed the theory of dielectric elastomers. Existing models of dielectric elastomers mostly focus on elastic behavior [Anderson, 1986; Goulbourne *et al.*, 2005; McMeeking and Landis, 2005; Suo *et al.*, 2008; Kofod, 2008], although viscoelastic behavior has also been explored [Christensen, 1980; Drozdov, 1995; Bergström and Boyce, 1998; Lochmatter *et al.*, 2007; Plante and Dubowsky, 2007; Spathis and Kontou, 2008; Wissler and Mazza, 2008]. Experiments have shown that viscoelasticity can significantly affect electromechanical transduction [Palakodeti and Kessler, 2006; Plante and Dubowsky, 2006, 2007; Lochmatter *et al.*, 2007; Jhong *et al.*, 2007; Keplinger *et al.*, 2008; Ha *et al.*, 2007]. This paper uses nonequilibrium thermodynamic to guide the development of models for dielectric elastomers undergoing dissipative processes, such as viscoelastic, dielectric and conductive relaxation. A method is described to calculate the critical condition for electromechanical instability.

To illustrate the approach, a specific model is constructed for viscoelastic dielectric elastomers. The model is fitted to existing stress-strain curves for VHB measured at various strain rates [Plante and Dubowsky, 2006]. We show that the electromechanical instability is significantly affected by patterns of the applied voltage. A higher critical voltage can be achieved by applying a constant voltage for a shorter time, or by applying ramping voltage with a higher rate. Furthermore, viscoelastic elastomer can achieve larger deformation than an elastic elastomer. These findings are consistent with experimental observations [Lochmatter *et al.*, 2007; Plante and Dubowsky, 2007; Keplinger *et al.*, 2008].

2. Nonequilibrium Thermodynamics of a Dielectric Elastomer

An elastomer responds to applied loads by time-dependent, dissipative processes. Viscoelastic relaxation may result from slippage between long polymers and rotation of joints between monomers. Dielectric relaxation may result from distortion of electron clouds and rotation of polar groups. Conductive relaxation may result from migration of electrons and ions through the elastomer. This section describes an approach to construct models of dissipative dielectric elastomers, guided by nonequilibrium thermodynamics.

Figure 1 illustrates a membrane of a dielectric elastomer, sandwiched between two compliant electrodes. The electrodes have negligible electrical resistance and mechanical stiffness. In the reference state, the elastomer is subjected to no force and voltage, and is of dimensions L_1, L_2 and L_3. In the current state, at time t, the elastomer is subjected to forces P_1 and P_2, and the two electrodes are connected to a battery of voltage Φ through a conducting wire. In the current state, the dimensions of the elastomer become l_1, l_2, and l_3, the two electrodes accumulate electric charges $\pm Q$, and the Helmholtz free energy of the membrane is F.

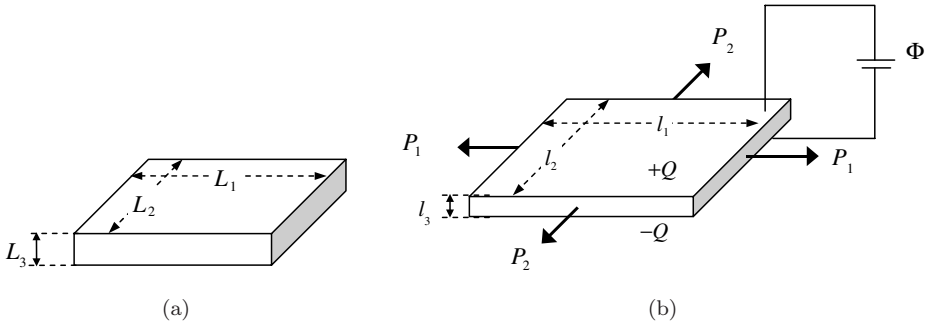

Fig. 1. Schematics of a dielectric elastomer at (a) the reference state, and (b) a current state.

When the dimensions of the membrane change by $\delta l_1, \delta l_2$, and δl_3, the mechanical forces do work $P_1\delta l_1 + P_2\delta l_2$. When a small quantity of charge δQ flows through the conducting wire, the battery does work $\Phi\delta Q$. Thermodynamics requires that the increase in the free energy should not exceed the total work done, namely,

$$\delta F \leq P_1\delta l_1 + P_2\delta l_2 + \Phi\delta Q. \tag{1}$$

For the inequality to be meaningful, the small changes are time-directed: δf means the change of the quantity f from a specific time to a slightly later time. The thermodynamic inequality (1) will guide us to construct a model of the dielectric elastomer.

Define stretches of the elastomer in the three directions by $\lambda_1 = l_1/L_1, \lambda_2 = l_2/L_2$ and $\lambda_3 = l_3/L_3$, the nominal stresses in the plane of the elastomer by $s_1 = P_1/(L_2L_3)$ and $s_2 = P_2/(L_1L_3)$, the nominal electric displacement by $\tilde{D} = Q/(L_1L_2)$, the nominal electric field $\tilde{E} = \Phi/L_3$, and the density of the Helmholtz free energy by $W = F/(L_1L_2L_3)$. Divide both sides of (1) by the volume of the membrane, $L_1L_2L_3$, and the thermodynamic inequality becomes

$$\delta W \leq s_1\delta\lambda_1 + s_2\delta\lambda_2 + \tilde{E}\delta\tilde{D}. \tag{2}$$

The dielectric elastomer is taken to be incompressible, so that $l_1l_2l_3 = L_1L_2L_3$ and $\lambda_1\lambda_2\lambda_3 = 1$. As a model of the dielectric elastomer, the free-energy density is prescribed as a function:

$$W = W(\lambda_1, \lambda_2, \tilde{D}, \xi_1, \xi_2, \ldots). \tag{3}$$

We characterize the state of a dielectric elastomer by λ_1, λ_2 and $\tilde{D}$, along with additional parameters $(\xi_1, \xi_2, \ldots)$. Inspecting (2), we note that λ_1, λ_2 and $\tilde{D}$ are the kinematic parameters through which the external loads do work. By contrast, the additional parameters $(\xi_1, \xi_2, \ldots)$ are not associated with the external loads in this way. These additional parameters describe the degrees of freedom associated with dissipative processes, and are known as internal variables. When the independent

variables change by small amounts, $(\delta\lambda_1, \delta\lambda_2, \delta\tilde{D}, \delta\xi_1, \delta\xi_2, \ldots)$, the free-energy function changes by

$$\delta W = \frac{\partial W}{\partial \lambda_1}\delta\lambda_1 + \frac{\partial W}{\partial \lambda_2}\delta\lambda_2 + \frac{\partial W}{\partial \tilde{D}}\delta\tilde{D} + \sum_i \frac{\partial W}{\partial \xi_i}\delta\xi_i. \tag{4}$$

Inserting (4) into (2), we rewrite the thermodynamic inequality as

$$\left(\frac{\partial W}{\partial \lambda_1} - s_1\right)\delta\lambda_1 + \left(\frac{\partial W}{\partial \lambda_2} - s_2\right)\delta\lambda_2 + \left(\frac{\partial W}{\partial \tilde{D}} - \tilde{E}\right)\delta\tilde{D} + \sum_i \frac{\partial W}{\partial \xi_i}\delta\xi_i \le 0. \tag{5}$$

As time moves forward, this thermodynamic inequality holds for any change in the independent variables $(\lambda_1, \lambda_2, \tilde{D}, \xi_1, \xi_2, \ldots)$. We next specify a model consistent with this inequality.

We assume that the system is in mechanical and electrostatic equilibrium, so that in (5) the factors in front of $\delta\lambda_1, \delta\lambda_2$ and $\delta\tilde{D}$ vanish:

$$s_1 = \frac{\partial W(\lambda_1, \lambda_2, \tilde{D}, \xi_1, \xi_2, \ldots)}{\partial \lambda_1}, \tag{6}$$

$$s_2 = \frac{\partial W(\lambda_1, \lambda_2, \tilde{D}, \xi_1, \xi_2, \ldots)}{\partial \lambda_2}, \tag{7}$$

$$\tilde{E} = \frac{\partial W(\lambda_1, \lambda_2, \tilde{D}, \xi_1, \xi_2, \ldots)}{\partial \tilde{D}}. \tag{8}$$

Once the free-energy function $W(\lambda_1, \lambda_2, \tilde{D}, \xi_1, \xi_2, \ldots)$ is prescribed, (6)–(8) constitute the equations of state of the dielectric elastomer.

Once the elastomer is assumed to be in mechanical and electrostatic equilibrium, the inequality (5) becomes

$$\sum_i \frac{\partial W(\lambda_1, \lambda_2, \tilde{D}, \xi_1, \xi_2, \ldots)}{\partial \xi_i}\delta\xi_i \le 0. \tag{9}$$

This thermodynamic inequality may be satisfied by prescribing a suitable relation between $(\delta\xi_1, \delta\xi_2, \ldots)$ and $(\partial W/\partial\xi_1, \partial W/\partial\xi_2, \ldots)$. For example, we will adopt a kinetic model of the type

$$\frac{d\xi_i}{dt} = -\sum_j M_{ij}\frac{\partial W(\lambda_1, \lambda_2, \tilde{D}, \xi_1, \xi_2, \ldots)}{\partial \xi_j}. \tag{10}$$

Here M_{ij} is a positive-definite matrix, which may depend on the independent variables $(\lambda_1, \lambda_2, \tilde{D}, \xi_1, \xi_2, \ldots)$.

Within this approach, the elastomer dissipates energy through the changes in the internal variables $(\xi_1, \xi_2, \ldots)$. The energy is dissipated at the rate

$$-\sum_i \frac{\partial W}{\partial \xi_i}\frac{d\xi_i}{dt} = \sum_i M_{ij}\frac{\partial W}{\partial \xi_i}\frac{\partial W}{\partial \xi_j}. \tag{11}$$

By construction, this rate is positive-definite.

3. Electromechanical Instability

Subject to a voltage, a dielectric elastomer reduces its thickness, so that the same voltage induces a higher true electric field. The positive feedback between the true electric field and the thickness may cause the elastomer to thin down drastically, resulting in electromechanical instability [Stark and Garton, 1955]. Following our recent paper on the electromechanical instability of elastic dielectric elastomers [Zhao and Suo, 2007], this section analyzes electromechanical instability of dissipative dielectric elastomers. Specifically, we consider the following scenario:

A loading program is prescribed by giving the forces and the voltage as functions of time. In response, the elastomer evolves the variables $(\lambda_1, \lambda_2, \tilde{D}, \xi_1, \xi_2, \ldots)$ in time. We seek a method to calculate the critical condition for the onset of the electromechanical instability.

Let $s_1(t)$, $s_2(t)$ and $\tilde{E}(t)$ be the prescribed program of the external loads. Differentiating the thermodynamic equations of state (6)–(8) with respect to time, we obtain that

$$\begin{bmatrix} \dfrac{ds_1}{dt} \\ \dfrac{ds_2}{dt} \\ \dfrac{d\tilde{E}}{dt} \end{bmatrix} = \begin{bmatrix} \dfrac{\partial^2 W}{\partial \lambda_1^2} & \dfrac{\partial^2 W}{\partial \lambda_1 \partial \lambda_2} & \dfrac{\partial^2 W}{\partial \lambda_1 \partial \tilde{D}} \\ \dfrac{\partial^2 W}{\partial \lambda_2 \partial \lambda_1} & \dfrac{\partial^2 W}{\partial \lambda_2^2} & \dfrac{\partial^2 W}{\partial \lambda_2 \partial \tilde{D}} \\ \dfrac{\partial^2 W}{\partial \tilde{D} \partial \lambda_1} & \dfrac{\partial^2 W}{\partial \tilde{D} \partial \lambda_2} & \dfrac{\partial^2 W}{\partial \tilde{D}^2} \end{bmatrix} \begin{bmatrix} \dfrac{d\lambda_1}{dt} \\ \dfrac{d\lambda_2}{dt} \\ \dfrac{d\tilde{D}}{dt} \end{bmatrix} + \sum_i \begin{bmatrix} \dfrac{\partial^2 W}{\partial \lambda_1 \partial \xi_i} \\ \dfrac{\partial^2 W}{\partial \lambda_2 \partial \xi_i} \\ \dfrac{\partial^2 W}{\partial \tilde{D} \partial \xi_i} \end{bmatrix} \frac{d\xi_i}{dt}. \qquad (12)$$

Because the functions $s_1(t)$, $s_2(t)$ and $\tilde{E}(t)$ are prescribed, the loading rates $ds_1/dt, ds_2/dt$ and $d\tilde{E}/dt$ are known. Furthermore, the rates of the internal variables, $d\xi_1/dt, d\xi_2/dt, \ldots$, can be expressed as functions of $(\lambda_1, \lambda_2, \tilde{D}, \xi_1, \xi_2, \ldots)$ by using the kinetic model (10). Consequently, (12) is a set of linear algebraic equations for the rates of the kinematic variables $d\lambda_1/dt, d\lambda_2/dt$ and $d\tilde{D}/dt$. Once solved, the rates $d\lambda_1/dt, d\lambda_2/dt$ and $d\tilde{D}/dt$ are expressed as functions of $(\lambda_1, \lambda_2, \tilde{D}, \xi_1, \xi_2, \ldots)$. These functions, together with the kinetic model (10), constitute a set of ordinary differential equations that simultaneously evolve in time all the independent variables $(\lambda_1, \lambda_2, \tilde{D}, \xi_1, \xi_2, \ldots)$.

The set of linear algebraic equations for the rates of the kinematic variables, (12), identify the Hessian of the free-energy function:

$$\mathbf{H} = \begin{bmatrix} \dfrac{\partial^2 W}{\partial \lambda_1^2} & \dfrac{\partial^2 W}{\partial \lambda_1 \partial \lambda_2} & \dfrac{\partial^2 W}{\partial \lambda_1 \partial \tilde{D}} \\ \dfrac{\partial^2 W}{\partial \lambda_2 \partial \lambda_1} & \dfrac{\partial^2 W}{\partial \lambda_2^2} & \dfrac{\partial^2 W}{\partial \lambda_2 \partial \tilde{D}} \\ \dfrac{\partial^2 W}{\partial \tilde{D} \partial \lambda_1} & \dfrac{\partial^2 W}{\partial \tilde{D} \partial \lambda_2} & \dfrac{\partial^2 W}{\partial \tilde{D}^2} \end{bmatrix}. \qquad (13)$$

The linear algebraic equations are solvable if and only if $\det \mathbf{H} \neq 0$. Consequently, electromechanical instability sets in when the determinant of the Hessian matrix

vanishes:

$$\det \mathbf{H} = 0. \tag{14}$$

The critical condition (14) has the same form as our previous result for elastic dielectric elastomers [Zhao and Suo, 2007]. However, the free-energy function for a dissipative dielectric elastomer depends on the kinematic variables $(\lambda_1, \lambda_2, \tilde{D})$, as well as on the internal variables $(\xi_1, \xi_2, \ldots)$. Consequently, the critical condition for the instability must be determined by simultaneously evolving in time all the independent variables $(\lambda_1, \lambda_2, \tilde{D}, \xi_1, \xi_2, \ldots)$ according to the coupled ordinary differential Eqs. (10) and (12), up to the time when $\det \mathbf{H} = 0$. Once the electromechanical instability sets in, the elastomer film thins down drastically and expands in area [Plante and Dubowsky, 2006]. The electromechanical instability is analogous to the snap-through instability [Zhao *et al.*, 2007].

4. A Viscoelastic Dielectric Elastomer

To represent a dissipative dielectric elastomer using the above approach, we need to specify a set of internal variables $(\xi_1, \xi_2, \ldots)$, and then specify the functions $W(\lambda_1, \lambda_2, \tilde{D}, \xi_1, \xi_2, \ldots)$ and $M_{ij}(\lambda_1, \lambda_2, \tilde{D}, \xi_1, \xi_2, \ldots)$. The approach is illustrated in this section by constructing a specific model of viscoelastic dielectric elastomer.

In response to an external load, an elastomer evolves toward a new state of equilibrium by various dissipative processes. The time required for each dissipative process to equilibrate is known as the relaxation time of the process. The time it takes from applying an electric field to a dielectric to the polarization of the dielectric is known as the dielectric relaxation time. The characteristic time for a viscoelastic elastomer subject to a force reaches equilibrium state is the viscoelastic relaxation time. The conductive relaxation time is related to the time for discharging a capacitor of a dielectric through the conduction of the dielectric. Experimental observations [Seki and Sato, 1995; Kestelman *et al.*, 2000; Wissler and Mazza, 2007; Johansson and Robertson, 2007; Brosseau *et al.*, 2008; Reffaee *et al.*, 2009] have shown that the dielectric relaxation-time for a dielectric elastomer is less than 0.1 ms, the viscoelastic-relaxation time is on the order of minutes, while the conductive relaxation may take hours. Here, we consider a dielectric elastomer whose various relaxation processes take place over widely different time scales: viscoelastic relaxation is much slower than dielectric relaxation, but is much faster than conductive relaxation. Consequently, to study the elastomer over a time scale in between the dielectric relaxation time and the conductive relaxation time, we may assume that dielectric relaxation has already reached equilibrium, conductive relaxation has yet to occur, and the only active dissipative process is viscoelastic relaxation.

Figure 2 illustrates a basic experiment of viscoelastic relaxation. At time zero, the elastomer is subjected to a sudden stretch. Subsequently, the stretch is held constant, while the stress is recorded as a function of time. The stress rises instantaneously, and then relaxes as a function of time. The elastomer approaches a new

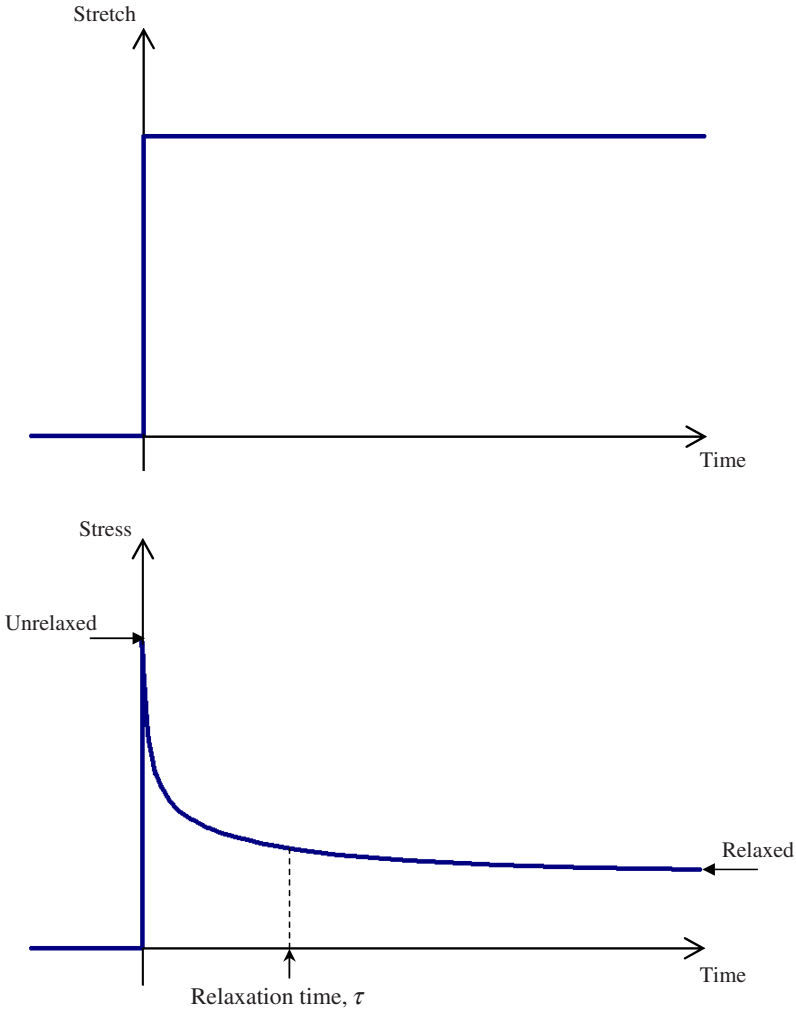

Fig. 2. Relaxation of a viscoelastic elastomer.

state of equilibrium, and the stress asymptotes to a lower level. The characteristic time for this process is known as the viscoelastic relaxation time, τ. The elastomer may relax by several viscoelastic molecular processes with distinct relaxation times. In order to focus on the main ideas, we will restrict our analysis to a single viscoelastic relaxation time.

Viscoelastic relaxation is commonly pictured with an array of springs and dashpots, known as the rheological models; please refer to Silberstein and Boyce [2010] for a recent review. Figure 3 illustrates one rheological model, including two springs and one dashpot. When this model is subjected to a sudden stretch, the unrelaxed modulus, μ_U, is represented by the sum of the stiffness of the two parallel springs.

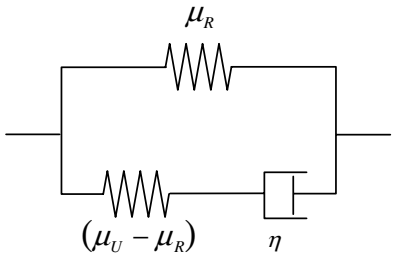

Fig. 3. A rheological model for a viscoelastic elastomer.

Over time, the viscous elongation of the dashpot gradually reduces the stress in the bottom spring. At full relaxation, the relaxed modulus, μ_R, is represented by the stiffness of the top spring. The relaxation time relates to the viscosity of the dashpot η and the stiffness of the bottom spring as

$$\tau = \eta/(\mu_U - \mu_R). \tag{15}$$

Consistent with the rheological model in Fig. 3, the elastomer may be modeled as two networks of polymers, one represented by the top spring in the model, and the other by the spring and the dashpot at the bottom. The two networks carry mechanical forces in parallel. The net deformation of both the networks are described by in-plane stretches (λ_1, λ_2). For the network represented by the string at the top, these stretches are produced by the spring. For the network represented by the spring and the dashpot at the bottom, the net stretches (λ_1, λ_2) are due to both the spring and the dashpot:

$$\lambda_1 = \lambda_1^e \xi_1, \quad \lambda_2 = \lambda_2^e \xi_2, \tag{16}$$

where $(\lambda_1^e, \lambda_2^e)$ are the stretches due to the bottom spring, and (ξ_1, ξ_2) are stretches due to the dashpot.

We represent the elasticity of each network by the neo-Hookean model, and write the free-energy function as

$$\begin{aligned} W(\lambda_1, \lambda_2, \tilde{D}, \xi_1, \xi_2) = {} & \frac{\mu_R}{2}(\lambda_1^2 + \lambda_2^2 + \lambda_1^{-2}\lambda_2^{-2} - 3) \\ & + \frac{\mu_U - \mu_R}{2}(\lambda_1^2\xi_1^{-2} + \lambda_2^2\xi_2^{-2} + \lambda_1^{-2}\lambda_2^{-2}\xi_1^2\xi_2^2 - 3) \\ & + \frac{\tilde{D}^2}{2\varepsilon}\lambda_1^{-2}\lambda_2^{-2}. \end{aligned} \tag{17}$$

In this expression, the first line represents the elastic energy of the network represented by the top spring, the second line represents the elastic energy of the network represented by the bottom spring, and the third line represents the electrostatic energy of the elastomer. We adopt the model of ideal dielectric elastomers [Zhao *et al.*, 2007], and assume that the electrostatic energy takes the same form as a dielectric liquid, with a constant permittivity ε.

Inserting (17) into (6)–(9), we obtain that

$$s_1 = \mu_R(\lambda_1 - \lambda_1^{-3}\lambda_2^{-2}) + (\mu_U - \mu_R)(\lambda_1\xi_1^{-2} - \lambda_1^{-3}\lambda_2^{-2}\xi_1^2\xi_2^2) - \lambda_1^{-3}\lambda_2^{-2}\tilde{D}^2/\varepsilon, \quad (18)$$

$$s_2 = \mu_R(\lambda_2 - \lambda_2^{-3}\lambda_1^{-2}) + (\mu_U - \mu_R)(\lambda_2\xi_2^{-2} - \lambda_2^{-3}\lambda_1^{-2}\xi_2^2\xi_1^2) - \lambda_2^{-3}\lambda_1^{-2}\tilde{D}^2/\varepsilon, \quad (19)$$

$$\tilde{E} = \lambda_1^{-2}\lambda_2^{-2}\tilde{D}/\varepsilon. \quad (20)$$

These equations constitute the equations of state of the model specified by the free-energy function (17).

We prescribe a kinetic model of the type (10). Specifically, we set $d\xi_1/dt = -\eta^{-1}\partial W/\partial\xi_1$ and $d\xi_2/dt = -\eta^{-1}\partial W/\partial\xi_2$, where η is the viscosity. Using the free-energy function (17), we write the kinetic model as

$$\frac{d\xi_1}{dt} = \frac{1}{\tau}(\lambda_1^2\xi_1^{-3} - \lambda_1^{-2}\lambda_2^{-2}\xi_1\xi_2^2), \quad (21)$$

$$\frac{d\xi_2}{dt} = \frac{1}{\tau}(\lambda_2^2\xi_2^{-3} - \lambda_1^{-2}\lambda_2^{-2}\xi_2\xi_1^2). \quad (22)$$

There is considerable flexibility in choosing kinetic models to fulfill the thermodynamic inequality (9). One choice may be deemed more suitable than others for a specific elastomer if it fits experimental data better. Our choice of the kinetic model, as represented by (21) and (22), is intended to illustrate the general approach. A more systematic search for a kinetic model for any specific dielectric elastomer is beyond our intention here.

Equations (18)–(22) are fitted to experiment data obtained from a series of uniaxial tension of VHB tested under different rates of stretch [Plante and Dubowsky, 2006]. Figure 4 compares the experimental data and the best fit. The values of fitting parameters are found to be $\mu_U = 87.8\,\text{kPa}$, $\mu_R = 22.8\,\text{kPa}$, and $\tau = 200\,\text{s}$.

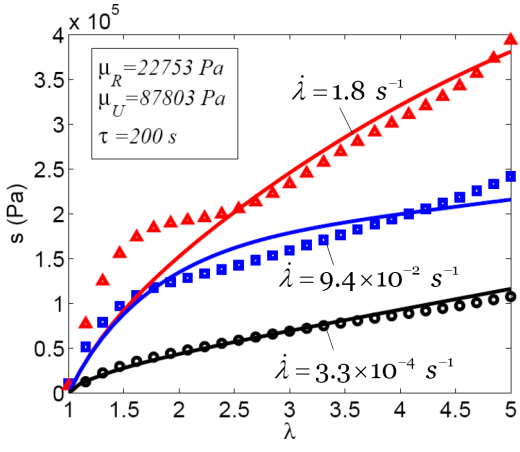

Fig. 4. The material model is fitted to experimental data of uniaxial tension of a dielectric elastomer tested under various stretching rates.

These values are consistent with the materials parameters reported in literature for VHB [Wissler and Mazza, 2007; Plante and Dubowsky, 2007; Ha *et al.*, 2007].

5. Deformation Induced by Applying a Voltage

This section applies the above model to a dielectric elastomer that is subjected to time-dependent voltage $\Phi(t)$, but not to forces. Set in (18)–(20) $\tilde{E} = \Phi(t)/L_3$, $s_1 = s_2 = 0$, $\lambda_1 = \lambda_2 = \lambda$ and $\xi_1 = \xi_2 = \xi$, giving

$$\varepsilon\tilde{E}^2 = \mu_R(\lambda^{-2} - \lambda^{-8}) + (\mu_U - \mu_R)(\lambda^{-2}\xi^{-2} - \lambda^{-8}\xi^4). \tag{23}$$

Differentiate (23) with respect to time, and we obtain that

$$\frac{d(\varepsilon\tilde{E}^2)}{dt} = [-2\lambda^{-3}(\mu_R + (\mu_U - \mu_R)\xi^{-2}) + 8\lambda^{-9}(\mu_R + (\mu_U - \mu_R)\xi^4)]\frac{d\lambda}{dt}$$
$$- (\mu_U - \mu_R)(2\lambda^{-2}\xi^{-3} + 4\lambda^{-8}\xi^3)\frac{d\xi}{dt}. \tag{24}$$

The critical condition $\det \mathbf{H} = 0$ corresponds to setting in (24) the factor in front of $d\lambda/dt$ to zero, giving

$$\lambda_c = \sqrt[3]{2}\left[\frac{\mu_R + (\mu_U - \mu_R)\xi^4}{\mu_R + (\mu_U - \mu_R)\xi^{-2}}\right]^{\frac{1}{6}}. \tag{25}$$

The kinetic model (19) becomes

$$\frac{d\xi}{dt} = \frac{1}{\tau}(\lambda^2\xi^{-3} - \lambda^{-4}\xi^3). \tag{26}$$

The voltage is applied at time $t = 0$. The dashpot does not move instantaneously, so that the initial value of the internal variable is $\xi(0) = 1$. Both the top and bottom springs deform instantaneously, and $\lambda(0)$ is determined by solving the nonlinear algebraic Eq. (23) by setting $\xi = \xi(0)$ and $\tilde{E} = \tilde{E}(0)$. Equations (24) and (26) evolve the functions $\lambda(t)$ and $\xi(t)$ up to the time when the stretch reaches the critical value, $\lambda(t) = \lambda_c$. In calculations, we set $\mu_U/\mu_R = 4$, which is suggested by fitting the experimental data, as described in Sec. 4.

Figure 5(a) illustrates a particular loading program. A voltage Φ is applied to an elastomer at $t = 0$, and is held at a constant level subsequently. Thus, the nominal electric field is prescribed at $\tilde{E} = \Phi/L_3$. For an elastic dielectric elastomer, electromechanical instability occurs when $\tilde{E} = 0.69\sqrt{\mu/\varepsilon}$, where μ is the shear modulus of the elastomer [Zhao and Suo, 2007]. For a viscoelastic dielectric elastomer, electromechanical instability will not occur if $\tilde{E} < 0.69\sqrt{\mu_R/\varepsilon}$, but will take place instantaneously if $\tilde{E} \geq 0.69\sqrt{\mu_U/\varepsilon}$. If the applied nominal electric field is within the range of $0.69\sqrt{\mu_R/\varepsilon} \leq \tilde{E} < 0.69\sqrt{\mu_U/\varepsilon}$, electromechanical instability will occur after some time.

Figure 5(b) shows the evolution of stretch of the dielectric elastomer under various applied electric fields. We adopt the notation $\tilde{E}_c = 0.69\sqrt{\mu_R/\varepsilon}$, and use it to normalize the applied electric field. At $t = 0$, the elastomer is unrelaxed, and the

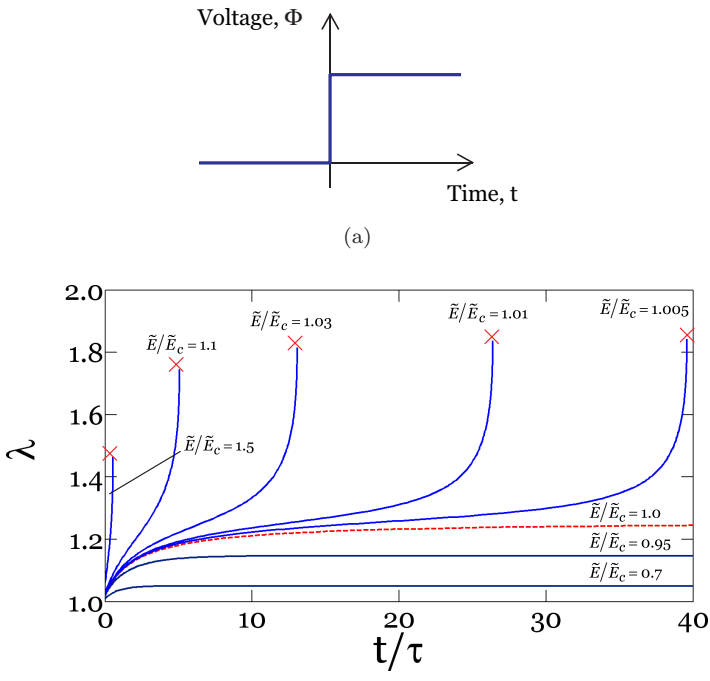

Fig. 5. Time-dependent electrical actuation of dielectric elastomer under a fixed DC voltage; (a) a voltage is suddenly applied and is subsequently held at constant, (b) the in-plane stretch as a function of time.

voltage induces a small stretch. Over time, the elastomer relaxes, and the stretch increases. For $\tilde{E} < \tilde{E}_c$, the elastomer can reach a new state of equilibrium. For $\tilde{E}_c \leq \tilde{E} < \tilde{E}_c\sqrt{\mu_U/\mu_R}$, the elastomer will suffer electromechanical instability, as marked by the crosses. The time needed to reach the instability can be read from Fig. 5(b).

As shown in Zhao and Suo [2007], the critical stretch of an elastic dielectric elastomer is $\sqrt[3]{2} \approx 1.26$. For a viscoelastic dielectric elastomer, however, a higher stretch up to 1.85 can be achieved before the electromechanical instability (Fig. 5(b)). This result is consistent with experimental observations on the increase of critical stretch due to the viscoelasticity of dielectric elastomers [Keplinger *et al.*, 2008]. Figure 5(b) also shows that a lower applied field gives a longer time to failure and a higher stretch. This finding is useful in designing more reliable dielectric elastomer actuators with larger deformation of actuation. For example, one may apply a voltage to give $\tilde{E} \approx 1.2\tilde{E}_c$ and maintain the voltage for τ (e.g., $\sim$200 s for VHB) to achieve an actuation stretch 1.65. This is much higher than the critical stretch (1.26) of an elastic dielectric elastomer.

Figure 6 illustrates another loading program: the voltage ramps up with time. We write $\tilde{E} = \tilde{E}_c kt$, where k characterizes the rate of the voltage. By solving Eqs. (24)

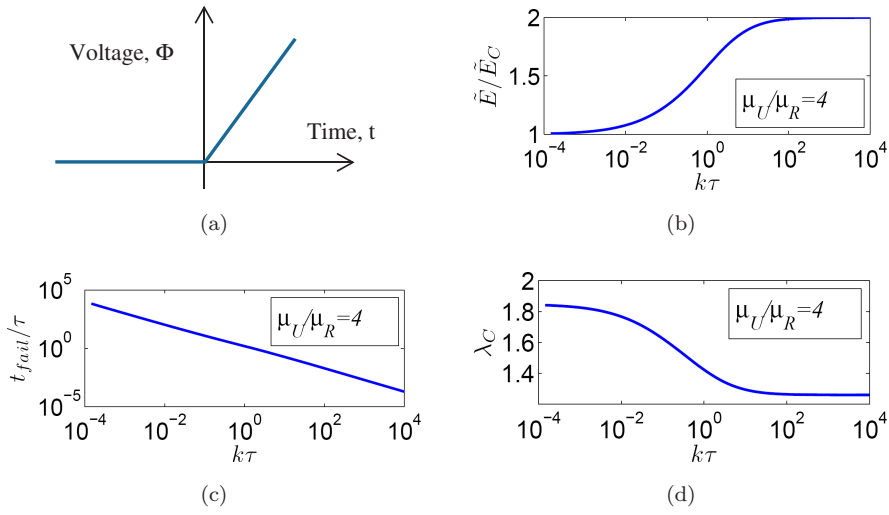

Fig. 6. (a) Applied voltage ramps up with time. (b) The critical nominal electric field. (c) The time to failure. (d) The critical stretch.

and (26) with initial conditions $\xi(0) = 1$ and $\lambda(0) = 1$, we get the evolution of λ and $\det \mathbf{H}$ with time. As $\det \mathbf{H}$ reaches 0, we record the nominal electric field $\tilde{E}$ for electromechanical instability [Fig. 6(a)] and corresponding time to failure t_{fail} [Fig. 6(b)] and critical stretch λ_C [Fig. 6(c)]. It can be seen that voltage applied at a lower rate gives a lower nominal electric field at the electromechanical instability. By selecting an appropriate ramping rate and time for the applied voltage, one can achieve a higher stretch with a viscoelastic dielectric elastomer than with an elastic dielectric elastomer.

6. Conclusions

We use nonequilibrium thermodynamics to guide the development of models of dissipative dielectric elastomers. The approach is illustrated by a specific model of a viscoelastic dielectric elastomer, which is fitted to existing experimental data. Viscoelasticity of a dielectric elastomer greatly affects electromechanical stability of the elastomer. By controlling the time-dependence of the applied voltage, one can make a viscoelastic dielectric elastomer achieve larger deformation of actuation than an elastic dielectric elastomer. The approach demonstrated here can easily be adapted to analyze homogeneous deformation of viscoelastic dielectric elastomers under other loading conditions and boundary conditions. While viscoelasticity may be represented by an array of springs and dashpots, dielectric relaxation may be represented by an array of resistors and capacitors. To model electric conduction, however, we will need to combine charge transport and large deformation, in a way similar to the theory of polyelectrolyte gels [Hong *et al.*, 2010]. It is hoped that

the approach outlined in this paper will be used in designing experiments to probe time-dependent phenomena in dielectric elastomers transducers.

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