Influence of the Temperature and Deformation-Dependent Dielectric Constant on the Stability of Dielectric Elastomers

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ABSTRACT: The performance of dielectric elastomers is affected by temperature and deformation, but very few models account for the effect of both factors. Recent experiments showed that the dielectric constant of the most widely used dielectric elastomer varies significantly with respect to temperature and deformation. We propose a model to study the influence of changes in dielectric constant on the electromechanical stability of dielectric elastomer. The model predicts the stability criteria with physical interpretation of its dependence on dielectric constant and electrostrictive coefficient. The numerical simulation indicates that the stress due to electrostriction relies on the temperature and deformation dependent dielectric constant and contributes to enhance the stability of the dielectric elastomer. © 2012 Wiley Periodicals, Inc. J. Appl. Polym. Sci. 000: 000–000, 2012

INTRODUCTION

Dielectric elastomer (DE) is emerging electroactive material and DE can produce large electromechanical deformation under a voltage. It can deform with an area strain well exceed 300%, which has been designed for diverse applications, including soft robots, optical devices, space applications, and energy generators.[1-6] It is generally sandwiched between two compliant electrodes and used as a stretchable capacitor.

However, under conventional operating circumstances, the practical application of dielectric elastomer is limited by the electromechanical instabilities,[7-9] including pull-in instability and snap-through instability. As voltage increases, the positive feedback between a higher electric field and a thinner elastomer may result in pull-in instability.[10,11] This pull-in instability usually occurs before the electrical breakdown with a maximum thickness deformation of 30–40%.[12,13] After the pull-in instability, if the dielectric elastomer can be stiffened at moderate deformation, it may snap to a thinner state close to its compression limit, exhibiting and resulting in snap-through instability.[14] This phenomenon was interpreted as two electromechanical co-existent states owing to a nonconvex free-energy function of the dielectric elastomer, which leads to a discontinuous phase transition.[11] Recently, the process of this electromechanical phase transition under various conditions has been discussed in detail,

which can significantly improve the stability in dielectric elastomer without electrical breakdown.[15]

In fact, both experiment and theory have proved that large stable deformation induced by voltage is possible, as long as the pull-in and snap-through instabilities are suppressed, e.g., by prestretch,[12,16] by nonlinear polarization,[17] by using swollen dielectric organogel[18] or by using interpenetrating network.[19] Zhao and Suo[20] proposed a method to study the electromechanical instability by involving the deformation dependent electrostriction. They showed that the deformation-dependent dielectric constant could suppress the electromechanical instability. Consequently, the deformation-dependent electrostriction is then investigated further in the stability of DE.[13,21]

In the past investigation, DE is assumed to operate in an isothermal environment. Recently, Liu et al.[22] studied the thermo-electro-mechanical instability of dielectric elastomers by employing a Mooney–Rivlin model, but assumed that dielectric constant is not affect by deformation. In fact, experimental results on the most widely used dielectric elastomer (VHB 4910, 3M) displayed a strong relation between the dielectric constant and area prestretch at different temperatures.[23,24] Therefore, the electrostrictive stress functioning as electrostriction will be generated as a result of the variation of dielectric constant[9] and consequently affect the actuation and the stability of DE. To the
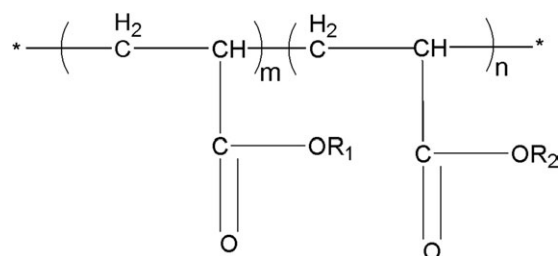
Figure 1. Chemical structures of the VHB 4910 acrylic elastomer.

authors' best knowledge, however, there is very few modeling work on the impact of temperature on electromechanical instabilities, including electrostrictive effect in a dielectric elastomer actuator undergoing large deformation has not been presented.

In this article, we aim to study the temperature and deformation-dependent dielectric constant and its influence on the electromechanical stabilities of DE. With the proposed free energy model, we are able to characterize the actuation and investigate the conditions of instability of DE undergoing large thermoelastic deformation. As the dielectric constant changes with respect to deformation and temperature, the model shows that the Maxwell stress and the electrostrictive stress contribute to the voltage-induced deformation simultaneously.

EXPERIMENTAL

Effect of Temperature and Deformation on Dielectric Constant

The molecular chain in the dielectric elastomer comprises numbers of polar monomer units in its backbone and side chains. It is obvious that most chains crosslink with each other via covalent bonds, forming a three-dimensional network which enables the dielectric elastomer to undergo large deformation. At a stress free or a relative small deformation state, the dipoles are free to align with an external electric field and the dipole polarization may saturate when all the dipoles are perfectly aligned. Consequently, the covalent bonds have inappreciable influence on polarization of the monomers. This behavior of ideal polarization is similar to polymer melt at small stretch. However, as the chain is further stretched to large deformation, the dipoles composing the backbone are most likely oriented in the direction of the force, and partly lose their freedom of alignment at other directions, while the rotations of dipoles in the side chains are less likely affected by the force. If an electric field is applied, let us say, normal to the in-plane stretch deformation, the dipole orientation polarization of the elastomer will be lower than that in the undeformed state, hence the dielectric constant decreases.[13] The chemical structure of the most widely used VHB 4910 is shown in Figure 1.[25] However, the chemical name of VHB 4910 is obscure due the commercial secret, where "n, m" in Figure 1 is the number of rigid links composing a single chain and is not known. In recent experiment, Jean-Mistral et al.[24] stretched membranes of a VHB 4910 elastomer by an equal prestretch in the planar directions at different temperatures and fitted the dielectric constant as a function of the prestretch in the statement of electrostriction[12,20]

$$\varepsilon(\lambda) = a\lambda_p^2 + c. \qquad (1)$$

where λ_p is the prestretch of the elastomer, a and c are parameters at different temperatures. In writing eq. (1), we have assumed that the dielectric elastomer is isotropic initially. When the electric field is applied in the direction of thickness, by symmetry, the coefficient of electrostriction is the same in the plane perpendicular to the direction of the electric field.[20]

When the temperature varies, the thermal expansion coefficient does not affect the thickness of the structures as its value of VHB 4910 is considerably low. In addition, VHB 4910 presents almost a permanent dipole moment due to its asymmetrical structure.[24] It is shown that the effect of temperature on dielectric constant is relatively significant and can be explained by a simple Debye model for gas[24]

$$\varepsilon = \varepsilon_\infty + N\mu^2/3\varepsilon_0 kT. \qquad (2)$$

where ε_∞ is the limiting value of the dielectric constant at high frequencies, N is the dipole density, μ is the dipole moment, and k is the Boltzmann constant.

The molecules have more thermal energy at higher temperature and therefore, the amplitude of random thermal motion is greater. This means that the range of deviation from a perfect alignment with the electric field is greater. Therefore, the molecules are less closely aligned with each other, and the dielectric constant reduces with increasing temperature. According to their experimental data[24] at different temperatures and prestretches in Figure 2, we fit a 3D function which is

$$\varepsilon = \varepsilon(\lambda, T) = a\lambda^2 + b/T + c, \qquad (3)$$

With $a = -0.053$, $b = 638$, and $c = 3.024$. Here a is defined as the electrostrictive coefficient and is in agreement with the value
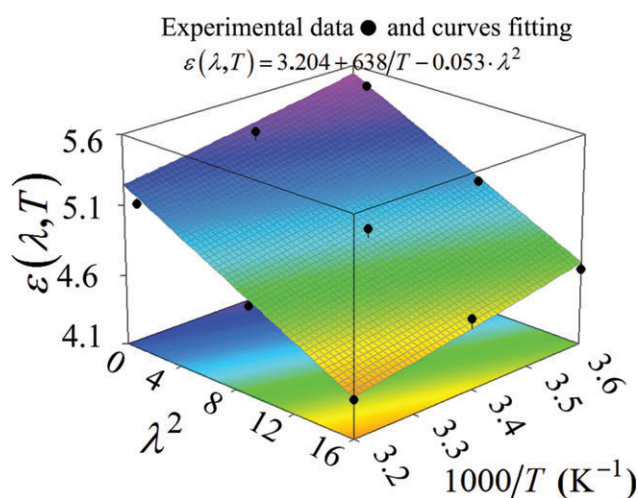

Figure 2. Experimentally measured dielectric constant[24] and 3D curves fitting result as a function of area prestretch and temperature. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]
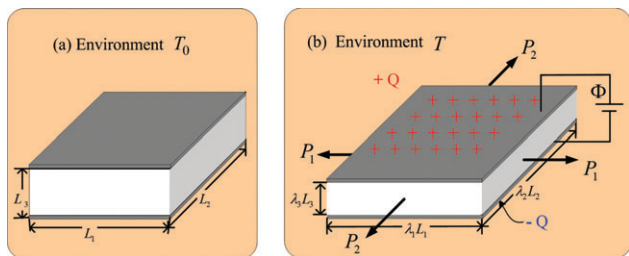
Figure 3. Schematic of a dielectric elastomer in (a) undeformed state; (b) deformed state, subject to forces and an electric voltage with an environment of a fixed temperature T. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

(-0.053) in a previous report.[20] To simplify the mathematical expressions, we introduce $B_T = b/T + c$.

EQUATIONS OF STATE FOR DE

Free Energy Model of Dielectric Elastomer Undergoing Large Deformation

To study the effect of temperature and deformation dependent dielectric constant on electromechanical instability of DE, we focus on a widely used configuration where a membrane of DE sandwiched between two electrodes[1,2] (see Figure 3). In the undeformed state, the membrane is of dimensions L_1, L_2, and L_3. In the current state, the membrane is subjected to tension forces P_1, P_2 and an electric voltage Φ with an environment of a fixed temperature T. Electrons flow through the external circuit, each surface of the membrane gains electric charge, Q, and dimensions of the DE become $\lambda_1 L_1$, $\lambda_2 L_2$, and $\lambda_3 L_3$, where λ_1, λ_2, and λ_3 are the stretches. Define the nominal electric field by $E = \Phi/L_3$, and the nominal electric displacement by $D = Q/(L_1 L_2)$, the nominal stresses by $s_1 = P_1/(L_2 L_3)$ and $s_2 = P_2/(L_1 L_3)$. The elastomer is taken to be incompressible and the three stretches are no longer independent, so that $\lambda_3 = \lambda_1^{-1}\lambda_2^{-1}$. We regard λ_1 and λ_2, along with D and T, as four independent variables that describe the state of the elastomer.

The dielectric elastomer, the mechanical forces, the thermal force and the voltage constitute a thermodynamic system. The free energy model of the system can be expressed as[22,26]:

$$W = \frac{T}{T_0} W_s(\lambda_1, \lambda_2) + \rho_0 c_0 \left[T - T_0 - T \ln\left(\frac{T}{T_0}\right) \right] + \frac{D^2}{2\varepsilon_0 \varepsilon(T, \lambda_1, \lambda_2)} \lambda_1^{-2}\lambda_2^{-2}. \quad (4)$$

The first term in the right-hand side of eq. (4) is the thermoelastic energy, where $W_s(\lambda_1, \lambda_2)$ is the isothermal-free energy density due to stretch at the reference temperature T_0.[27,28] The second term is the purely thermal contribution, where the parameters ρ_0 and c_0 denote the mass density and the specific heat capacity.[28] The third term is the dielectric energy, where $\varepsilon_0 = 8.85 \times 10^{-12}$ F/m is the dielectric constant of the vacuum, and $\varepsilon(T, \lambda_1, \lambda_2) = \varepsilon$ is the temperature and deformation dependence of dielectric constant of the polymer.

Thermodynamics dictates that a stable equilibrium state should minimize the free energy of the system.[29] The eq. (4) is minimized when the current state of the system is a state of stable equilibrium. Consequently, the nominal stress, nominal electric field and specific entropy can be expressed via the partial derivative of free energy function W as

$$s_1 = \frac{\partial W}{\partial \lambda_1} = \frac{T}{T_0}\frac{\partial W_s(\lambda_1, \lambda_2)}{\partial \lambda_1} - \frac{\lambda_1^{-3}\lambda_2^{-2}D^2}{\varepsilon_0\varepsilon} - \frac{\lambda_1^{-2}\lambda_2^{-2}D^2}{2\varepsilon_0\varepsilon^2}\frac{\partial\varepsilon}{\partial\lambda_1}, \quad (5)$$

$$s_2 = \frac{\partial W}{\partial \lambda_2} = \frac{T}{T_0}\frac{\partial W_s(\lambda_1, \lambda_2)}{\partial \lambda_2} - \frac{\lambda_1^{-2}\lambda_2^{-3}D^2}{\varepsilon_0\varepsilon} - \frac{\lambda_1^{-2}\lambda_2^{-2}D^2}{2\varepsilon_0\varepsilon^2}\frac{\partial\varepsilon}{\partial\lambda_2}, \quad (6)$$

$$E = \frac{\partial W}{\partial D} = \frac{D}{\varepsilon_0\varepsilon}\lambda_1^{-2}\lambda_2^{-2}, \quad (7)$$

Once the elastic energy $W_s(\lambda_1, \lambda_2)$ is specified for the incompressible dielectric elastomer, the three eqs., (5)–(7), constitute the equations of state.

To be specific, we consider the elastomer subject to equal biaxial stresses $s_1 = s_2 = s$. We assume that the dielectric is isotropic, so that the two in-plane stretches are equal, which we denote as $\lambda_1 = \lambda_2 = \lambda$. Because of incompressibility, the stretch in the direction of thickness is given by $\lambda_3 = \lambda^{-2}$.

DE exhibits significant stiffening on approaching the limiting stretch.[9] To take into account the effect of extension limit, we use the Gent model[27,30]

$$W_s(\lambda) = -\frac{\mu J_m(1 + \theta((T - T_0)/T_0))}{2} \ln\left(1 - \frac{2\lambda^2 + \lambda^{-4} - 3}{J_m(1 + \theta((T - T_0)/T_0))}\right), \quad (8)$$

where μ is the isothermal infinitesimal shear modulus at reference temperature T_0, J_m is a dimensionless parameter related to the limiting stretch and θ is a parameter that takes into account the modifications of this maximum average chain length to reflect dependence on the temperature.[27]

In the following calculations, we take $\theta = 0$ as the special case proposed by Bilgili et al.[28] With eqs. (4) and (8), the equilibrium conditions for DE undergoing equal biaxial ($\lambda_1 = \lambda_2 = \lambda$) deformation become

$$s = \frac{\partial W}{\partial \lambda} = 2\mu \frac{T}{T_0}\frac{\lambda - \lambda^{-5}}{1 - \frac{2\lambda^2 + \lambda^{-4} - 3}{J_m}} - \frac{2D^2}{\varepsilon_0\varepsilon}\lambda^{-5} - \frac{D^2}{2\varepsilon_0\varepsilon^2}\cdot\frac{\partial\varepsilon}{\partial\lambda}\lambda^{-4}, \quad (9)$$

$$\frac{E}{\sqrt{\mu/\varepsilon_0}} = \frac{D}{\sqrt{\mu\varepsilon_0}}\cdot\frac{\lambda^{-4}}{\varepsilon}. \quad (10)$$

Equations (9) and (10) constitute a complete set of equations of state for the specific material model of dielectric elastomers, which we use in the following analysis to study the effect of temperature and deformation dependence of dielectric constant on the actuation and the electromechanical instability in dielectric elastomer.

RESULTS AND DISCUSSION

Electromechanical Instability

By substituting eq. (3) into eqs. (9) and (10), we obtain that
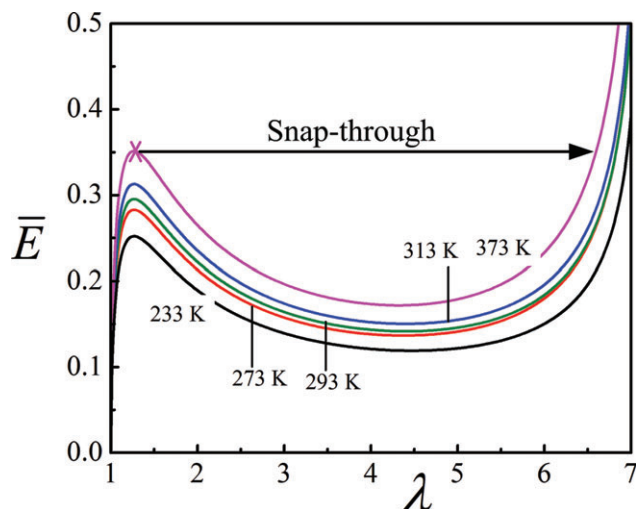
Figure 4. Nominal electric field-stretch curves at various temperatures. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

$$\frac{E}{\sqrt{\mu/\varepsilon_0}} = \frac{D}{\sqrt{\mu\varepsilon_0}} \cdot \frac{\lambda^{-4}}{a\lambda^2 + B_T}, \qquad (11)$$

$$\frac{s}{\mu} = 2\frac{T}{T_0}\frac{\lambda - \lambda^{-5}}{1 - \frac{2\lambda^2 + \lambda^{-4} - 3}{J_m}} - \frac{3a\lambda^2 + 2B_T}{\left(a\lambda^2 + B_T\right)^2} \cdot \frac{D^2}{\varepsilon_0\mu}\lambda^{-5}. \qquad (12)$$

To be specific, we consider a dielectric elastomer is subject to no mechanical force $s = 0$. In this condition, combining eqs. (11) and (12), we write the equilibrium state in the dimensionless form:

$$\frac{E}{\sqrt{\mu/\varepsilon_0}} = \sqrt{\frac{T}{T_0}\frac{2\left(\lambda^{-2} - \lambda^{-8}\right)}{1 - \frac{2\lambda^2 + \lambda^{-4} - 3}{J_m}} \cdot \frac{1}{3a\lambda^2 + 2B_T}}, \qquad (13)$$

$$\frac{D}{\sqrt{\mu\varepsilon_0}} = \sqrt{\frac{T}{T_0}\frac{2\left(\lambda^{6} - 1\right)}{1 - \frac{2\lambda^2 + \lambda^{-4} - 3}{J_m}} \cdot \frac{\left(a\lambda^2 + B_T\right)^2}{3a\lambda^2 + 2B_T}}. \qquad (14)$$

In plotting these equations and in the analysis of what follows, we normalize the nominal electric field as $\bar{E} = E/\sqrt{\mu/\varepsilon_0}$, the

nominal electric displacement as $\bar{D} = D/\sqrt{\mu\varepsilon_0}$ and take $T_0 = 300$ K, $J_m = 100$,[17,26] $a = -0.053$ and $B_T = 3.204 + 638/T$ as representative values for VHB 4910 dielectric elastomer.

The condition of eq. (13) is plotted in Figure 4. We mark the critical point for pull-in instability by cross and snap-through instability by an arrow in Figure 4. As the temperature increases from 233 to 373 K, the critical value of the nominal electrical field increases. The maximum values for $\bar{E}$ are, respectively, about 0.26 at 233 K, 0.30 at 273 K, 0.31 at 293 K, 0.33 at 313 K and 0.37 at 373 K. As the dielectric constant of the polymer is temperature and deformation dependent, $\partial\varepsilon(T, \lambda)/\partial\lambda < 0$ for all temperatures, usually known as electrostrictive effect, therefore, the third term in eq. (9) can partially reduce the squeezing deformation from the Maxwell stress and offsets the positive feedback between the true electric field and the thickness, contributing to improve the pull-in instability.

After the pull-in instability, the electric field rises again with the stretch, because the chains approach the limiting stretches and then stiffen steeply. As marked in Figure 4, the snap-through instability is linked to inhomogeneous deformation and may lead to electromechanical phase transition.[15] This phase transition corresponds to two states of deformation, thin state with small stretch and thick state with relatively large stretch. Under special conditions, the two states of the membrane may coexist. This can be achieved by wrinkling the regions in the thin state.[7]

To further illustrate the physical idea of the effect of temperature and deformation on electromechanical instabilities in dielectric elastomer, we explore the consequence of this model by plotting the nominal electric field-stretch curves for various values of the electrostrictive coefficients under different temperatures in Figure 5. At 300 K in Figure 5(a), for $a = 0$, the elastomer possesses deformation-independent dielectric constants, which corresponds to the ideal dielectric studied previously[11] and the critical stretch is about $\lambda \approx 1.26$. For a typical VHB 4910, $a = -0.053$, the nominal electric field reaches a peak value of 0.297 at a bigger stretch $\lambda \approx 1.27$. Especially at $a = -0.7$ in Figure 5(a), the local peak disappears and large stretch may generate, resulting in a monotonic electric field-stretch curve and enabling the elastomer to survive the pull-in and snap-through instabilities.
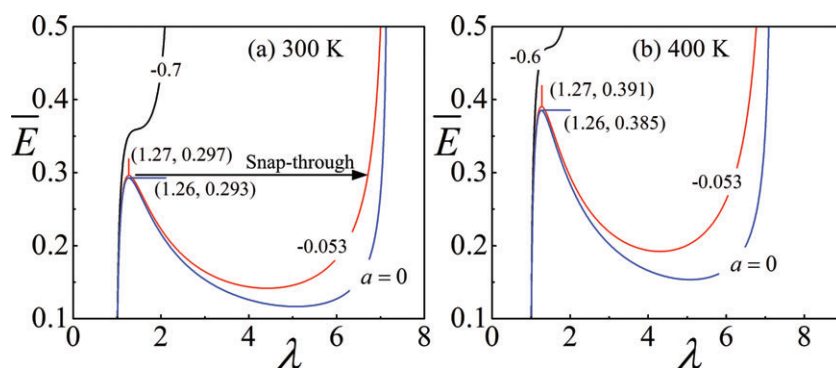

Figure 5. The effect of the electrostrictive coefficient on the electromechanical instability at different temperatures (a) 300 K and (b) 400 K. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

At a high temperature of 400 K in Figure 5(b), the local peak vanishes for $a = -0.6$. As temperature increases from 300 to 400 K, the maxima for the electric fields are, respectively, $\bar{E}_{300} = 0.293$ and $\bar{E}_{400} = 0.385$ for $a = 0$, $\bar{E}_{300} = 0.297$ and $\bar{E}_{400} = 0.391$ for $a = -0.053$. Compared with Figure 5(a), shows that the critical value of the nominal electrical field at the onset of instability increases with the increasing of temperature. That is to say, as the temperature increases, the electromechanical stability of the elastomer can be improved and large stretch can be obtained. This result is consistent with experimental observations on the increase of critical stretch and electromechanical responses due to the increasing temperature.[31] At lower temperatures the VHB 4910 becomes very stiff and the strain response can be significantly reduced.[32]

The critical stretch at the onset of the pull-in instability can be obtained from eq. (13).

Figure 6 plots the critical stretch (λ_c) corresponding to pull-in instability with respect to electrostrictive coefficient at two temperatures. As a decreases, the critical value of the stretch increases. When $a \leq -0.636$ for $T = 300$ K and when $a \leq -0.551$ for $T = 400$ K, the pull-in instability is eliminated. The increase of temperature increases the critical value of the electrostrictive coefficient to suppress the pull-in instability.

The curve of temperature dependent critical electrostrictive coefficient to suppress the electromechanical instability is plotted in Figure 7. The critical electrostrictive coefficient decreases with the decrease of temperature. As shown in Figure 7, two regions, stable and unstable state in electromechanical deformation, are separated by the curve. Above the curve, the voltage-stretch character of the elastomer is nonmonotonous and the local pull-in instability peak will take place, so that the system is electromechanical unstable; below the curve, the negative electrostrictive coefficient is sufficient for the elastomer to survive the electromechanical instability and generate large stretch. That is to say, a variation of dielectric constant with tempera-
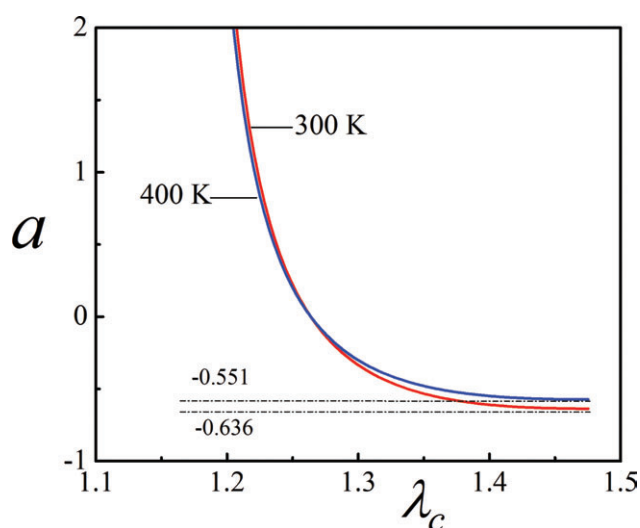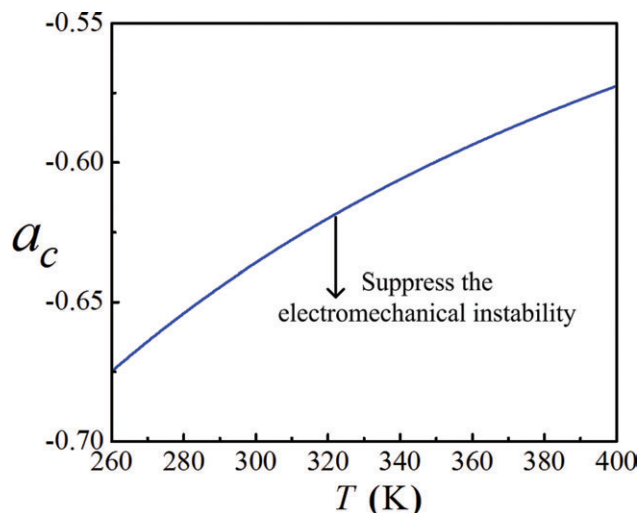

Figure 7. The critical electrostrictive coefficient to suppress the electromechanical instability with different temperatures. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

ture and deformation can make the dielectric elastomer to work in a new stable state of electromechanical actuation and generate larger deformation.

The dielectric constant of DE is a function of temperature and deformation. When temperature and deformation are changed, the variation of dielectric constant can result in electrostrictive effect. The electrostrictive effect induced by the stretch with the changing of temperature would modify the electric field–stretch curve and would eventually suppress the pull-in instability. Unlike the Maxwell stress, the existence of the temperature and deformation dependent electrostrictive effect generates electrostrictive stress, which thickens the elastomer as a result of the elongation of dipoles alignment, a fact that restrains the positive feedback between the electric field and the thickness. So that an enhancement of stability will be obtained and a new stable state of large stretch will be generated.

CONCLUSIONS

In summary, we proposed a model to characterize the influence of temperature and deformation-dependent dielectric constant on electromechanical instability and electrostrictive effect in dielectric elastomers. The model shows that electrostriction caused by the variation of dielectric constant could enhance the actuation stability of dielectric elastomer. A moderate negative electrostrictive coefficient will restrain the pull-in and snap-through instabilities. Simultaneously, the increase of temperature improves the critical value of electrostrictive coefficient for eliminating the pull-in and snap-through instabilities. This study offers a method to improve the electromechanical instability of DE and may be used to guide the design of new high-performance DE material.

Figure 6. The effect of the electrostrictive coefficient on the critical stretch at two temperatures 300 and 400 K. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

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