Thermoelectromechanical stability of dielectric elastomers undergoing temperature variation

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ABSTRACT

In this paper, the influence of both temperature and deformation on dielectric constant is considered during the establishment of free energy function of dielectric elastomers. A constitutive model of the thermodynamic systems undergoing adiabatic process is derived to study its thermoelectromechanical stability. The relations between different work conjugated parameters of dielectric elastomer are theoretically described, including the relations between nominal electric field and nominal electric displacement, entropy and temperature. Under different temperatures and electric fields, the allowable energy range of dielectric elastomer is calculated. Furthermore, the electric-induced variation of dielectric elastomer's temperature and entropy is also studied under various principal planar stretch ratios. These simulation results should offer assistances in guiding the design and fabrication of excellent actuators featuring dielectric elastomers.

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1. Introduction

Large deformation will be generated when an electric field is applied on a thin dielectric elastomer film (Pelrine et al., 2000; Brochu and Pei, 2010; O'Halloran et al., 2008; Zhang et al., 2010). Due to its excellent properties, such as high elastic energy density, fast response, low cost, light weight, high efficiency, and easy to be processed, dielectric elastomers show great application potential in the area of smart bionics, medical devices, aeronautics and astronautics. Various devices based on dielectric elastomers such as actuators, sensors, tactile display and energy harvesters etc. have also been designed (Gallone et al., 2004, 2010; Carpi et al., 2008, 2010a; Keplinger et al., 2010; Kollosche and Kofod, 2010; Plante and Dubowsky, 2006; Adrian Koh et al., 2011; Liu et al., 2010b). The principal failure modes of the dielectric elastomers include electric breakdown,

material rupture, loss of tension, electromechanical instability (Adrian Koh et al., 2011), would definitely hinder their wide applications. Hence, a well established theoretical model on the failure mechanism of dielectric elastomers is important. Recently, the nonlinear mechanical performance (Suo et al., 2008; Liu et al., 2009b,c), electromechanical stability (Liu et al., 2009d,e; Zhao et al., 2007, 2008b; Zhou et al., 2008; Liu et al., 2008, 2010a,b,c; Kong et al., 2011; Li et al., 2012), dynamical performance (Zhao et al., 2011; Zhu et al., 2010b) and failure of application devices (Adrian Koh et al., 2011; Moscardo et al., 2008; Liu et al., 2014) of dielectric elastomers are hotspots in all theoretical researches on electroactive materials.

Zhao and Suo have proposed an analytical method based on thermodynamics and continuum mechanics to predict the electromechanical stability of dielectric elastomers (Zhao and Suo, 2007). Based on the method, the electromechanical instability of Mooney–Rivlin type dielectric elastomers and the allowable area of the energy harvester were studied (Liu et al., 2009a, 2010a). The stable area of dielectric elastomers was investigated by research groups of

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Díaz-Calleja and Suo respectively (Díaz-Calleja et al., 2008; Adrian Koh et al., 2009). The bifurcation and chaos performance of dielectric elastomer's thermodynamic system was also studied by Zhu et al. (2010a). The electromechanical stability of dielectric elastomer undergoing homogeneous and inhomogeneous large deformation was investigated by Zhao and Suo (2008a) and He et al. (2009). Suo et al. studied electromechanical stability of dielectric elastomers of interpenetrating networks (Suo and Zhu, 2009). Zhao and Suo proposed the theory of dielectric elastomers capable of giant deformation for actuation (Zhao and Suo, 2010). Besides, the constitutive relation and electromechanical instability of different dielectric elastomer actuators, such as planar, rolled, tubular and hemispheric actuators, were also studied by Zhao and Suo (2007), Moscardo et al. (2008), Zhu et al. (2010c). However, the influence of temperature to dielectric elastomers has not been considered in the above mentioned theoretical analyses. Recent experiments indicate that the temperature plays a considerable role in the dielectric property of dielectric elastomers (Jean-Mistral et al., 2010). In Suo's research group, a model was proposed to show that the snap-through instability is markedly affected by both the extension limit of polymer chains and the polarization saturation of dipoles. The model is essentially useful for the future research of high-performance dielectric elastomer transducers (Li et al., 2011a; Liu et al., 2012a,b). In addition, some potential applications of stretchable dielectric elastomers are predicted by Carpi et al. (2010b). In Zhuo's research group, Li proposed a model to study the polarization mode, and characterize the behavior when the dipolar alignment is constrained by deformation. The conditional polarization would modify the final state as well as the path to instability during actuation due to the mechanism of induced electrostriction, indicating a new route to optimize the performance of elastic dielectrics (Li et al., 2011b).

Suo reviews the theory of dielectric elastomers developed within continuum mechanics and thermodynamics. The theory couples large deformation and electric potential, and describes nonlinear and nonequilibrium behavior. It also enables the finite element method to simulate transducers of realistic configurations, and suggests alternative routes to achieve giant voltage-induced deformation (Suo, 2010).

In this paper, a constitutive model of the thermodynamic systems under adiabatic process is deduced in order to study the thermoelectromechanical stability of dielectric elastomers. The allowable energy range of dielectric elastomer's thermodynamic system is predicted and the electric-induced changes of dielectric elastomer's temperature and entropy are also evaluated.

2. Fundamental theory

After being uniformly coated with compliant electrodes on the both surfaces and applied a voltage across, a thin dielectric elastomer film will endure shrinkage in thickness and expansion in area. As a result of increased temperature, the material's modulus will decrease and then generate faster deformation during its thickness reduction and area expanding. The decreased thickness of dielectric elastomer film results in a higher electric filed in the material. Under the coupling effect of thermal, electric and mechanical fields, such positive feedback continues. When the induced electric filed surpass the critical filed, the dielectric elastomer film will breakdown, resulting in the thermoelectromechanical instability.

As we know, a dielectric elastomer is actually a network of polymer chains. Each polymer chain consists of many monomers. The polymer chains are crosslinked by covalent bonds. The covalent bonds give solid-like behavior to the rubber. If these crosslinks are removed, the rubber becomes a polymer melt of liquid form. In fact, a dielectric elastomer is very similar to liquid at the level of monomers, where the scales are not large enough yet to include any crosslink. Like liquids, the polymers are densely packed and it is difficult to change the rubber's volume. Also like liquids, the polymers can move relative to one another, which contributes to dielectric elastomers' strong abilities in shape alternation (Liu et al., 2011a). For this
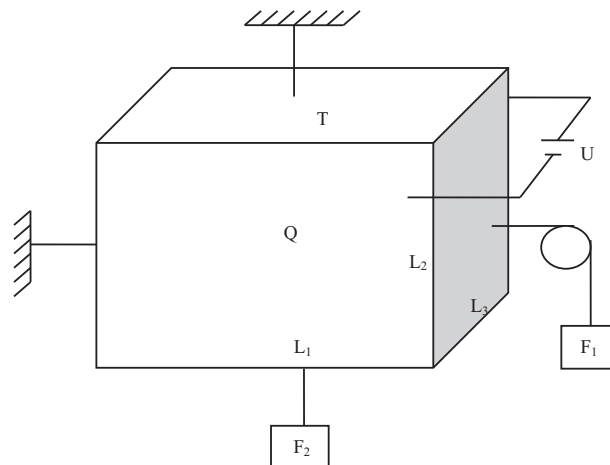

Fig. 1. Thermoelectromechanical coupling system of dielectric elastomer.

Table 1
The typical parameters of dielectric elastomer.

p	α (F/m K)	β (F/m)	ε_r	Nk (J/kg K)	c_0 (J/kg K)
0.9		−0.0592			
1	−0.0048	−0.0533	5.54	3.66	1.7×10^3
1.1		−0.0484			

reason, most dielectric elastomers can be taken as incompressible materials.

When a dielectric elastomer undergoes large deformation suffering different physical fields, the change in the shape of the elastomer is much larger than that of the volume. Consequently, the elastomer is often taken to be
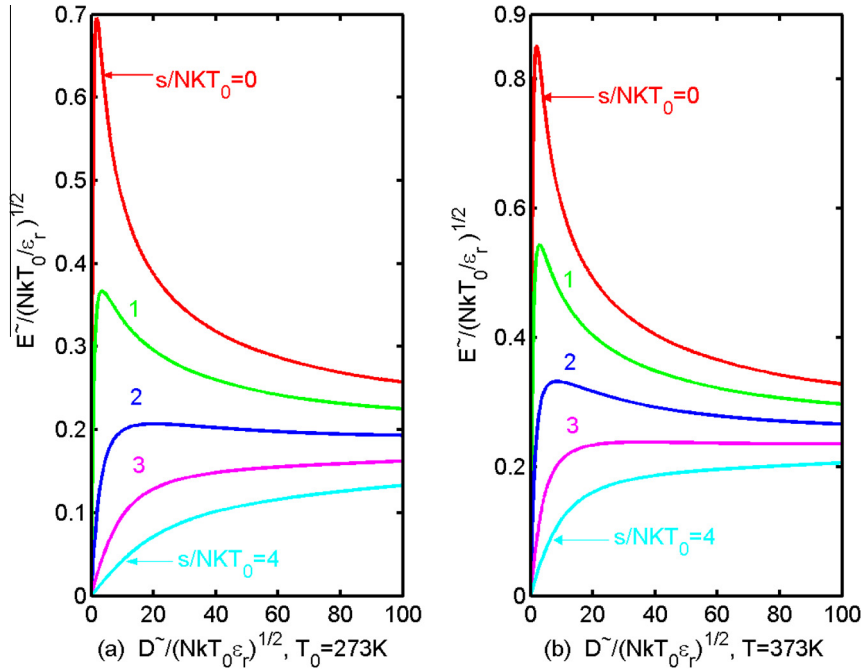
Fig. 2. Nominal electric field vs. nominal electric displacement of dielectric elastomer under different temperatures.
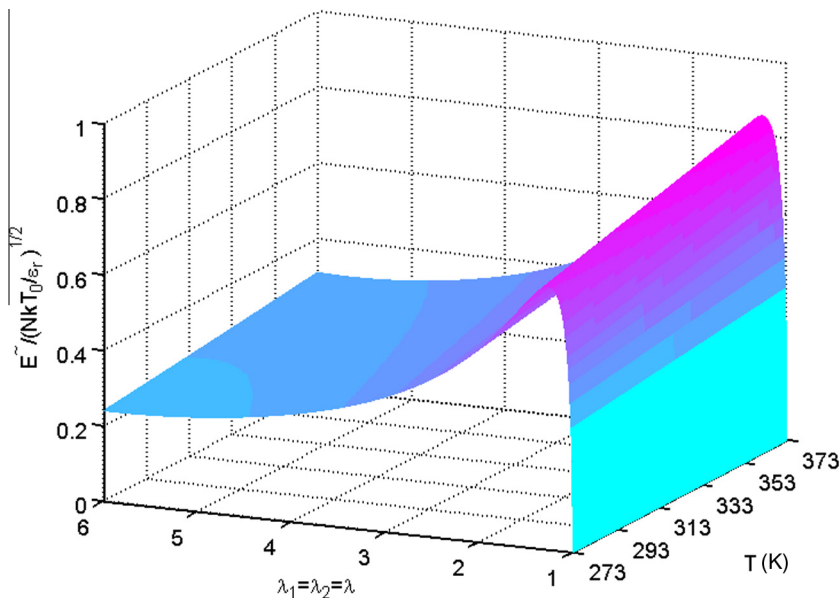
Fig. 3. Nominal electric field of dielectric elastomer under different temperatures and stretches.
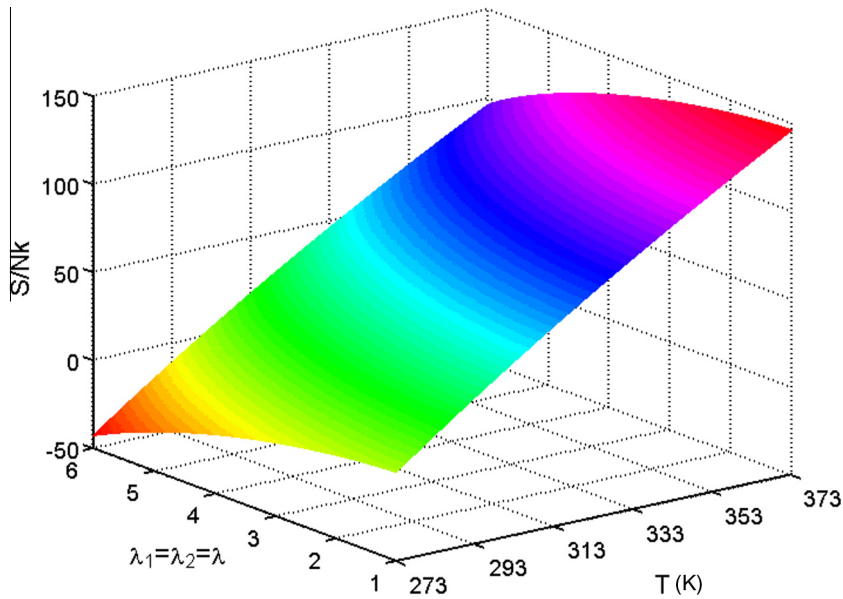
Fig. 4. Entropy of dielectric elastomer under different temperatures and stretches.
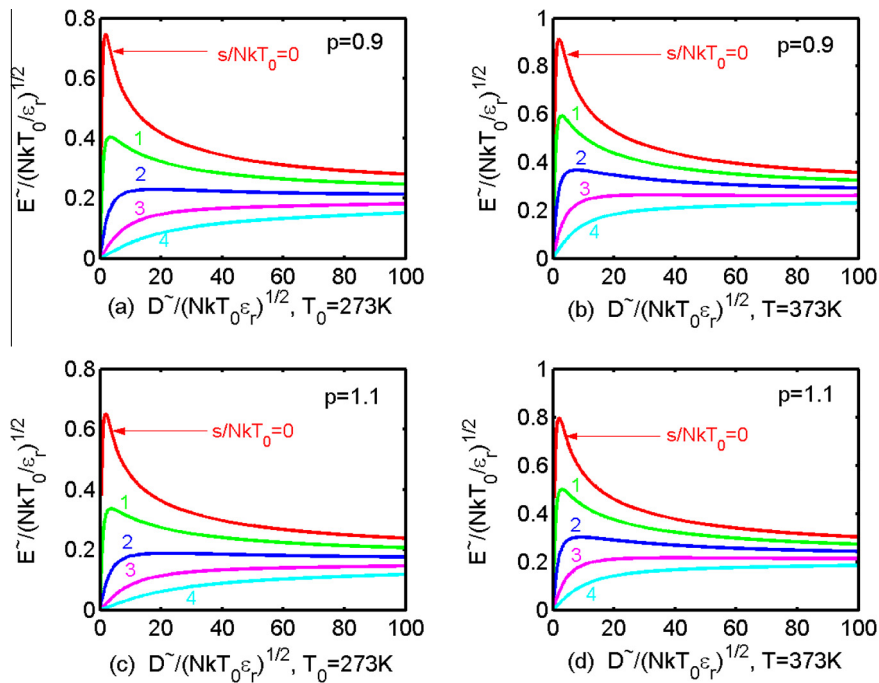

Fig. 5. Nominal electric field and nominal electric displacement of dielectric elastomer under different stretches.

incompressible, that is, the volume of the material remains unchanged during the deformation.

As is shown in Fig. 1, for the isotropic, homogeneous and incompressible thermoelastic solid, the free energy function can be expressed as a function with four variables, λ_1, λ_2, $D^\sim$ and T, namely $W(\lambda_1, \lambda_2, D^\sim, T)$, where λ_1 and λ_2 are stretches in the principal planar directions, $D^\sim$ donates

nominal electric displacement, T represents current temperature. Under the coupling effect of thermal, electric and mechanical fields, the small variation of the four independent variables are $d\lambda_1$, $d\lambda_2$, $dD^\sim$ and dT respectively. The variation of free energy of dielectric elastomer's thermo-electromechanical coupling system can be expressed as following
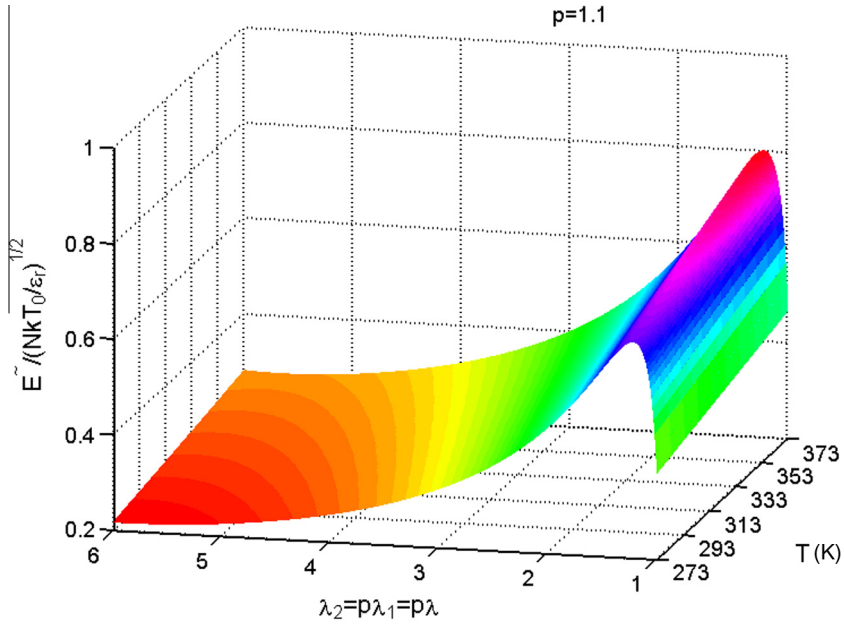
Fig. 6. Nominal electric field of dielectric field under different temperatures and stretches.
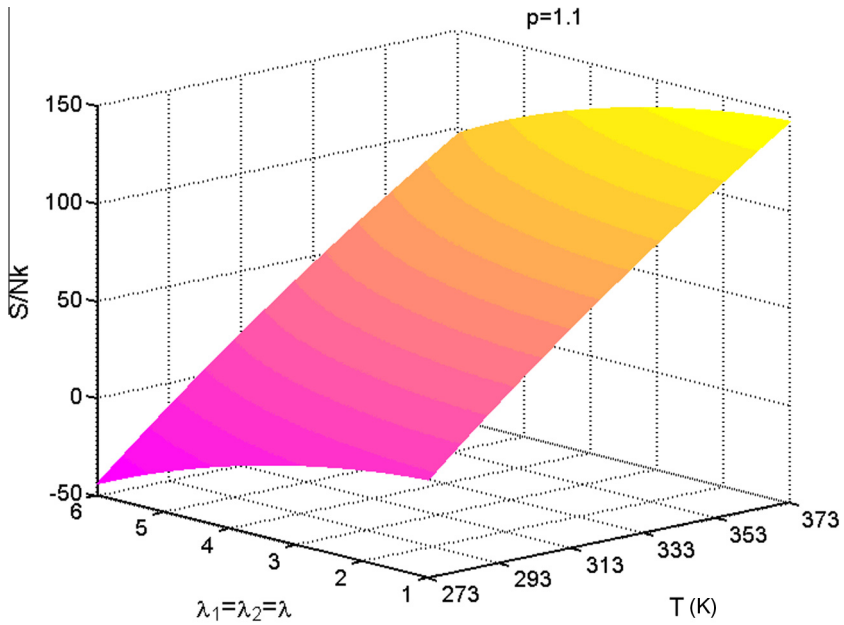

Fig. 7. The entropy of dielectric elastomer under different temperatures and stretches.

$$dW = \frac{\partial W(\lambda_1, \lambda_2, D^\sim, T)}{\partial \lambda_1} d\lambda_1 + \frac{\partial W(\lambda_1, \lambda_2, D^\sim, T)}{\partial \lambda_2} d\lambda_2$$
$$+ \frac{\partial W(\lambda_1, \lambda_2, D^\sim, T)}{\partial D^\sim} dD^\sim - \frac{\partial W(\lambda_1, \lambda_2, D^\sim, T)}{\partial T} dT \quad (1)$$

Therefore, the nominal stress, nominal electric field and entropy are

$$s_1 = \frac{\partial W(\lambda_1, \lambda_2, D^\sim, T)}{\partial \lambda_1} \quad (2)$$

Table 2
Critical nominal electric field and critical stretch of dielectric elastomer under different temperatures and stretches.

p	Critical nominal electric field		Critical stretch	
	273 K	373 K	273 K	373 K
0.9	$0.747\sqrt{NkT_0/\varepsilon_r}$	$0.913\sqrt{NkT_0/\varepsilon_r}$	1.22	
1	$0.695\sqrt{NkT_0/\varepsilon_r}$	$0.851\sqrt{NkT_0/\varepsilon_r}$	1.26	
1.1	$0.652\sqrt{NkT_0/\varepsilon_r}$	$0.799\sqrt{NkT_0/\varepsilon_r}$	1.31	

$$s_2 = \frac{\partial W(\lambda_1, \lambda_2, D^{\sim}, T)}{\partial \lambda_2} \tag{3}$$

$$E^{\sim} = \frac{\partial W(\lambda_1, \lambda_2, D^{\sim}, T)}{\partial D^{\sim}} \tag{4}$$

$$S = -\frac{\partial W(\lambda_1, \lambda_2, D^{\sim}, T)}{\partial T} \tag{5}$$

$$W(\lambda_1, \lambda_2, T, D^{\sim}) = U(\lambda_1, \lambda_2, T) + V(\lambda_1, \lambda_2, T, D^{\sim}) \tag{6}$$

where $U(\lambda_1, \lambda_2, T)$ is thermoelastic energy per unit volume. $V(\lambda_1, \lambda_2, T, D^{\sim})$ is electric filed energy density.

In Jean-Mistral's experiment, it is indicated that the dielectric constant of dielectric elastomer is related with the temperature and stretch (Jean-Mistral et al., 2010). Therefore, in the following study, a model of electric field energy density function with variable dielectric constant is introduced to analyze the mechanical performance and electromechanical stability of dielectric elastomers. Considering the influence of temperature and stretch, we propose the expression of dielectric constant $\varepsilon(T, \lambda_1, \lambda_2)$ as following:

$$\varepsilon(T, \lambda_1, \lambda_2) = \varepsilon_r + \alpha(T - T_0) + \beta \lambda_1 \lambda_2 \tag{7}$$

where ε_r is the relative dielectric constant of dielectric elastomer under the reference temperature without deformation, α is a phenomenology parameter, T_0 is the temperature of referenced state, β is the coefficient of electrostriction.

Expression (7) is the result of the Taylor expansion of $\varepsilon(T, \lambda_1, \lambda_2)$. The dielectric constant model we suggested only considers the simplest circumstances, and we ignore the influence of high order term of the temperature. That is to say, the dielectric constant in this paper is only taken to be a linear function of temperature. This assumption is reasonable as in many researchers' experimental studies, the dielectric constant of insulating oil liquid is linear with the temperature variation, and also the ideal silicon rubber dielectric elastomer could be taken as ideal liquid dielectric (Hmmer et al., 2002). Therefore, in the research below, all the parameters (α, β and so on) are studied base on the Jean-Mistral's experiment (Jean-Mistral et al., 2010).

Of course, we can also use model with more clear physical significance to describe the temperature's influence to dielectric constant. According to the research of Jean-Mistral and the Debye equation function (Jean-Mistral et al., 2010), we propose the expression of dielectric constant $\varepsilon(T, \lambda_1, \lambda_2)$ as follow:

$$\varepsilon(T, \lambda_1, \lambda_2) = \varepsilon_r + \frac{\alpha'}{T} + \beta' \lambda_1 \lambda_2 \tag{8}$$

The dielectric constant is the inverse proportion function of the temperature. Due to the thermal force field, the dipoles reorientation, which is the molecular origin of the macroscopic dielectric permittivity, is strongly restricted. In Eq. (8), $\alpha' = \frac{N\mu^2}{3\kappa\varepsilon_0}$, N is the dipole density of a dielectric elastomer, μ is the dipole moment of a dielectric elastomer, κ is the Boltzmann constant, ε_0 is the dielectric permittivity of a vacuum, β' is the coefficient of electrostriction a dielectric elastomer.

The influence of temperature on the permittivity of acrylic dielectric elastomers was studied in detail by Sheng et al. First, it was experimentally observed that the permittivity changed in demonstrating a peak value as the temperature alternation. Theoretical studies then showed that the Cole–Cole equation was a better choice to describe the temperature induced permittivity increase, while the permittivity decrease after the peak value could be well captured by Debye equation. Furthermore, the mathematical model for the permittivity of VHB 4910 was established, which would provide assistances for the further studies of the electromechanical stability of dielectric elastomers (Sheng et al., 2013).

The elastomer is taken to be network of long polymers obeying the Gaussian statistics, so that the elastic behavior of the elastomer is neo-Hookean. For an ideal dielectric elastomer, the dielectric energy per unit volume is $\frac{D^2}{2\varepsilon}$, and the dielectric constant ε is a constant independent of deformation (Zhu et al., 2010b). Considering the electrostriction of dielectric elastomers, the dielectric constant is a function of the stretches, the dielectric energy per unit volume is $\frac{D^2}{2\varepsilon(\lambda_1, \lambda_2)}\lambda_1^{-2}\lambda_2^{-2}$ (Suo, 2010). Hence, the electric field energy density function of incompressible dielectric elastomers can be written as follows (Zhao and Suo, 2007)

$$V(\lambda_1, \lambda_2, T, D^{\sim}) = \frac{D^{\sim 2}}{2[\varepsilon_r + \alpha(T - T_0) + \beta \lambda_1 \lambda_2]}\lambda_1^{-2}\lambda_2^{-2} \tag{9}$$

Considering the influence of temperature, a proposed thermoelastic strain energy function is Horgan and Saccomandi (2006)

$$U(\lambda_1, \lambda_2, T) = \frac{1}{2}NkT(\lambda_1^2 + \lambda_2^2 + \lambda_1^{-2}\lambda_2^{-2} - 3)$$
$$+ c_0\left[(T - T_0) - T\ln\frac{T}{T_0}\right] \tag{10}$$

Equation (9) describes the coupling influence of temperature and stretch of the thermodynamical system. The first item in Eq. (10) is the neo-Hookean thermoelastic energy. N is the number of polymer chains per unit volume of dielectric elastomer, kT is the temperature in units of energy and $NkT = \mu(T)$. Here $\mu(T)$ is the temperature related shear modulus under infinitesimal deformation. The second item donotes the thermal contribution of temperature to the free energy of the entire thermodynamic system, c_0 is the specific heat of dielectric elastomers.

The second term of Eq. (10) $c_0\left[(T - T_0) - T\ln\frac{T}{T_0}\right]$ is called the thermal contribution. We can describe the derivation of the thermal contribution simply as followings (Liu et al., 2011b), the thermal contribution can be expressed as $\xi(T) = Q(T) - TS$, where $Q(T)$ is the internal energy (Horgan and Saccomandi, 2006). As the temperature rises, internal energy follows $Q(T) = c_0(T - T_0)$. According to the relation of specific heat for constant volume and entropy, $c_0 = T\frac{\partial S}{\partial T}$, we obtain $S = c_0\ln\frac{T}{T_0}$, where c_0 is the specific heat of polar dielectric, T_0 and T are the temperatures in reference and current states respectively. Therefore, the thermal contribution of the thermodynamic system related to temperature is $\xi(T) = c_0[(T - T_0) - T\ln\frac{T}{T_0}]$, which reflects the effect of temperature on the free energy of polar dielectric thermo-electric coupling system.
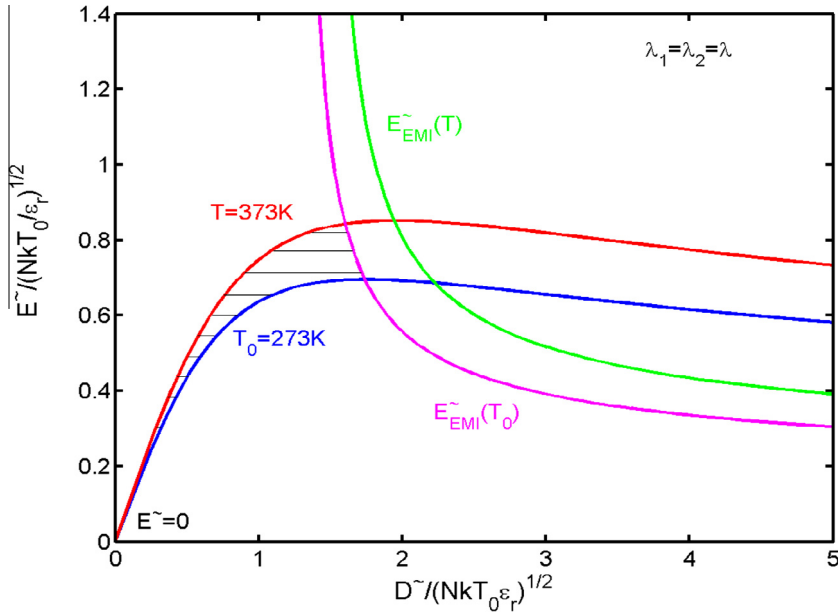
Fig. 8. The allowable energy range of dielectric elastomer in plane of nominal electric field and nominal electric displacement.
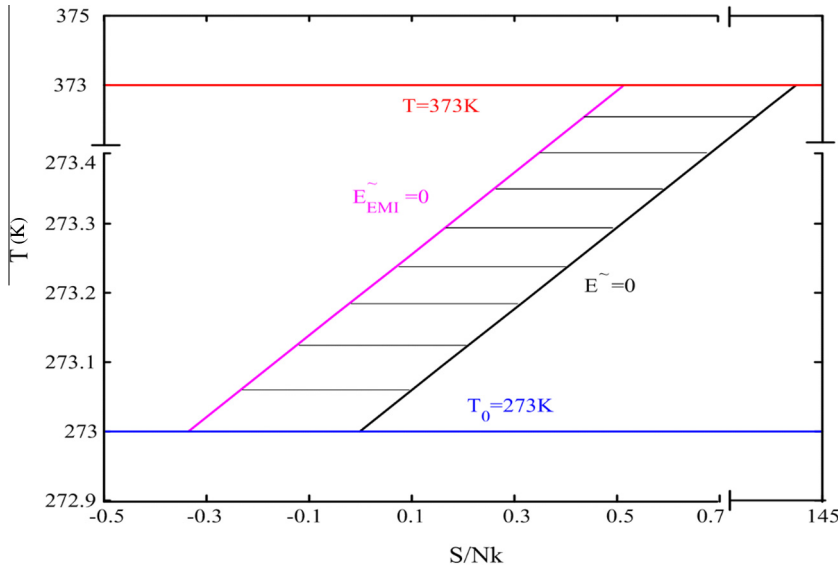

Fig. 9. The allowable energy range of dielectric elastomer in plane of temperature and entropy.

And the expression of thermal contribution is widely applicable to the thermodynamic systems with great temperature changes.

3. Constitutive model of dielectric elastomers

Considering equations (2)–(6), the nominal stress of dielectric elastomer's thermodynamic system in the two planar principal directions, the nominal electric field in the thickness direction and the entropy are obtained respectively, as is shown in the followings

$$s_1 = NkT(\lambda_1 - \lambda_1^{-3}\lambda_2^{-2}) - \frac{D^{\sim 2}\lambda_1^{-3}\lambda_2^{-2}}{[\varepsilon_r + \alpha(T - T_0) + \beta\lambda_1\lambda_2]}$$
$$- \frac{D^{\sim 2}\beta\lambda_1^{-2}\lambda_2^{-1}}{2[\varepsilon_r + \alpha(T - T_0) + \beta\lambda_1\lambda_2]^2} \tag{11}$$

$$s_2 = NkT(\lambda_2 - \lambda_1^{-2}\lambda_2^{-3}) - \frac{D^{\sim 2}\lambda_1^{-2}\lambda_2^{-3}}{[\varepsilon_r + \alpha(T - T_0) + \beta\lambda_1\lambda_2]}$$
$$- \frac{D^{\sim 2}\beta\lambda_1^{-1}\lambda_2^{-2}}{2[\varepsilon_r + \alpha(T - T_0) + \beta\lambda_1\lambda_2]^2} \tag{12}$$
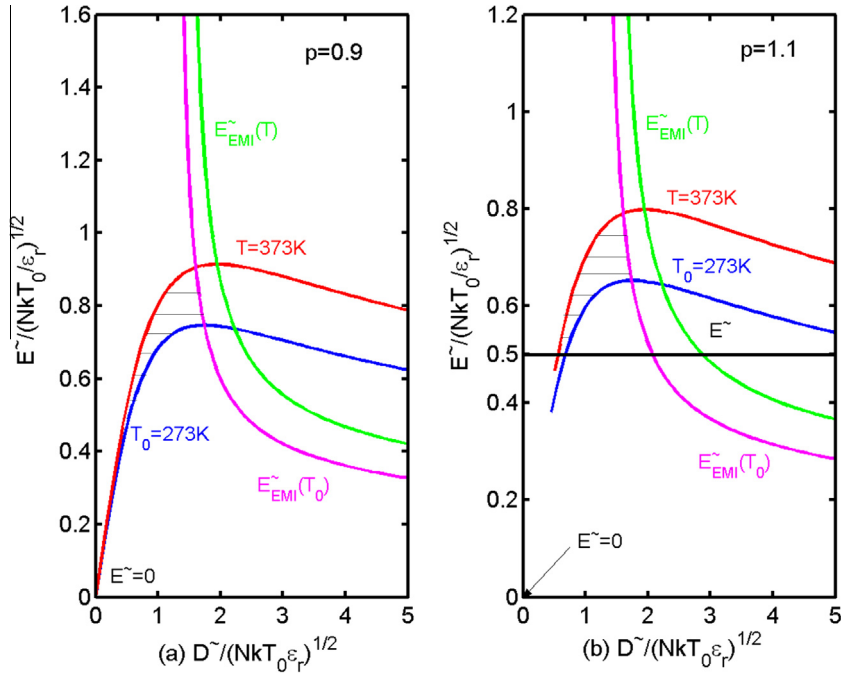
Fig. 10. The allowable energy range of dielectric elastomer in plane of nominal electric field and nominal electric displacement.
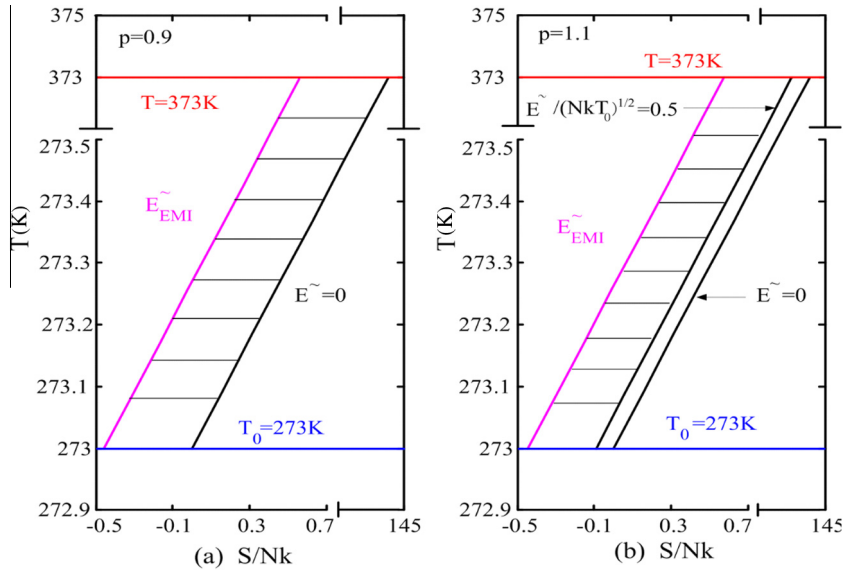

Fig. 11. The allowable energy range of dielectric elastomer in plane of temperature and entropy.

Table 3
Entropy variation and allowable energy range under different temperatures.

p	Entropy variation (J/kg K) 273–373 K	Allowable energy range (J/kg) 273–373 K
0.9	1.67	167
1	1.22	122
1.1	1.29	129

$$E^{\sim} = \frac{1}{[\varepsilon_r + \alpha(T - T_0) + \beta\lambda_1\lambda_2]} D^{\sim} \lambda_1^{-2} \lambda_2^{-2} \tag{13}$$

$$S = c_0 \ln\frac{T}{T_0} - \frac{1}{2} Nk(\lambda_1^2 + \lambda_2^2 + \lambda_1^{-2}\lambda_2^{-2} - 3)$$

$$+ \frac{\alpha D^{\sim 2} \lambda_1^{-2} \lambda_2^{-2}}{2[\varepsilon_r + \alpha(T - T_0) + \beta\lambda_1\lambda_2]^2} \tag{14}$$

Fig. 1 represents the schematic graph of the thermo-electromechanical system of dielectric elastomer. In Fig. 1, mechanical load F_1 and F_2 are applied. Before the deformation, dielectric elastomer's dimensions in these two directions are L_1 and L_2 respectively. Dimensions after deformation will be $\lambda_1 L_1$ and $\lambda_2 L_2$ respectively. The nominal stress of dielectric elastomer in the planar direction are $s_1 = F_1/L_2 L_3$, $s_2 = F_2/L_1 L_3$. The true stress of dielectric elastomer are $\sigma_1 = F_1/\lambda_2 \lambda_3 L_2 L_3$, $\sigma_2 = F_2/\lambda_1 \lambda_3 L_1 L_3$. Therefore, the relation between true stress and nominal stress are $\sigma_1 = s_1/\lambda_2 \lambda_3$, $\sigma_2 = s_2/\lambda_1 \lambda_3$. Similarly, the nominal electric field of dielectric elastomer is defined as $E^{\sim} = \frac{U}{L_3}$. The nominal electric displacement is defined as $D^{\sim} = \frac{Q}{L_1 L_2}$. Thus, the corresponding true electric field of dielectric elastomer can be expressed as $E = \frac{U}{\lambda_3 L_3} = \frac{E^{\sim}}{\lambda_3}$, while the true electric displacement is $D = \frac{Q}{\lambda_1 L_1 \lambda_2 L_2} = \frac{D^{\sim}}{\lambda_1 \lambda_2}$.

Based on the above mentioned definitions, the true stress and true electric field can be given respectively as

$$\sigma_1 = NkT(\lambda_1^2 - \lambda_1^{-2}\lambda_2^{-2}) - \frac{D^{\sim 2}\lambda_1^{-2}\lambda_2^{-2}}{[\varepsilon_r + \alpha(T - T_0) + \beta\lambda_1\lambda_2]}$$
$$- \frac{D^{\sim 2}\beta\lambda_1^{-1}\lambda_2^{-1}}{2[\varepsilon_r + \alpha(T - T_0) + \beta\lambda_1\lambda_2]^2} \tag{15}$$

$$\sigma_2 = NkT(\lambda_2^2 - \lambda_1^{-2}\lambda_2^{-2}) - \frac{D^{\sim 2}\lambda_1^{-2}\lambda_2^{-2}}{[\varepsilon_r + \alpha(T - T_0) + \beta\lambda_1\lambda_2]}$$
$$- \frac{D^{\sim 2}\beta\lambda_1^{-1}\lambda_2^{-1}}{2[\varepsilon_r + \alpha(T - T_0) + \beta\lambda_1\lambda_2]^2} \tag{16}$$

$$E = \frac{1}{[\varepsilon_r + \alpha(T - T_0) + \beta\lambda_1\lambda_2]}D^{\sim}\lambda_1^{-1}\lambda_2^{-1} \tag{17}$$

4. Thermoelectromechanical stability of dielectric elastomers

During the fabrication process of actuators, the dielectric elastomers are subjected to pre-stretch, most commonly, unequal biaxial stretch. Therefore, in the following study, to simplify the calculation, we set $\lambda_2 = p\lambda_1 = p\lambda$. Here p is the ratio between principal planar stretches of dielectric elastomer. Sometimes, this pre-stretch could be an equal biaxial one, such as while making the spherical devices featuring isotropic dielectric elastomers (Fox and Goulbourne, 2008). This can be treated as a special case of $p = 1$.

According to equations (11)–(14), the dimensionless nominal electric displacement, the nominal electric field and the entropy of dielectric elastomer can be obtained as followings

$$\frac{D^{\sim}}{\sqrt{NkT_0\varepsilon_r}} = \sqrt{\frac{\left[1 + \frac{\alpha(T-T_0)+p\beta\lambda^2}{\varepsilon_r}\right]}{\left[p^{-2} + \frac{p^{-1}\beta\lambda^2}{2[\varepsilon_r+\alpha(T-T_0)+p\beta\lambda^2]}\right]}\left[\frac{T}{T_0}(\lambda^6 - p^{-2}) - \frac{s_1\lambda^5}{NkT_0}\right]} \tag{18}$$

$$\frac{E^{\sim}}{\sqrt{NkT_0/\varepsilon_r}}$$
$$= \sqrt{\frac{1}{\left[p^{-2} + \frac{p^{-1}\beta\lambda^2}{2[\varepsilon_r+\alpha(T-T_0)+p\beta\lambda^2]}\right]\left[1 + \frac{\alpha(T-T_0)+p\beta\lambda^2}{\varepsilon_r}\right]}\left[\frac{T}{T_0}(p^{-4}\lambda^{-2} - p^{-6}\lambda^{-8}) - \frac{s_1 p^{-4}\lambda^{-3}}{NkT_0}\right]} \tag{19}$$

$$\frac{S}{Nk} = \frac{c_0}{Nk}\ln\frac{T}{T_0} - \frac{1}{2}[(1 + p^2)\lambda^2 + p^{-2}\lambda^{-4} - 3]$$
$$+ \frac{\alpha D^{\sim 2}p^{-2}\lambda^{-4}}{2Nk[\varepsilon_r + \alpha(T - T_0) + p\beta\lambda^2]^2} \tag{20}$$

Then by submitting Eq. (18) into Eq. (20), the entropy can be expressed as a function of stretch and temperature:

$$\frac{S}{Nk} = \frac{c_0}{Nk}\ln\frac{T}{T_0} - \frac{1}{2}[(1 + p^2)\lambda^2 + p^{-2}\lambda^{-4} - 3]$$
$$+ \frac{\alpha p^{-2}}{[2\varepsilon_r + 2\alpha(T - T_0) + 3p^{-1}\beta\lambda^2]}\left\{T(\lambda^2 - p^{-2}\lambda^{-4}) - \frac{s\lambda}{Nk}\right\} \tag{21}$$

Further, to investigate the relation of entropy and nominal electric field, we submit Eq. (13) into Eq. (20),

$$\frac{S}{Nk} = \frac{c_0}{Nk}\ln\frac{T}{T_0} - \frac{1}{2}[(1 + p^2)\lambda^2 + p^{-2}\lambda^{-4} - 3]$$
$$+ \frac{\alpha E^{\sim 2}p^2\lambda^4}{2Nk} \tag{22}$$

By referring to Jean-Mistral's experimental research (Jean-Mistral et al., 2010) of polyacrylate very-high-bond (VHB) dielectric elastomer, some material parameters used for the simulation are shown in Table 1. The representative value of α and β shown in Table 1 are calculated according to experimental results. Further, an initial temperature of $T_0 = 273$ K and a maximum temperature of $T_{\max} = 373$ K are also selected to precede the analysis. These are also the temperatures under which the dielectric elastomer was experimentally investigated by Jean-Mistral et al.

Based on Eqs. (18) and (19), under the condition of equal biaxial condition ($p = 1$), the relation of nominal electric field and nominal electric displacement under different temperature is presented in Fig. 2, and the nominal electric field under different temperatures and stretches is described in Fig. 3. Clearly, on each curve, the voltage first increases, reaches a peak, and then decreases. With the increase of temperature, the critical nominal electric field increases, namely the thermoelectromechanical stability increases. The entropy of dielectric elastomer under different temperatures and stretches are presented in Fig. 4. With the increase of temperature and decrease of stretch, the entropy increases.

Similarly, under unequal biaxial state ($p \neq 1$), the relation among different performance parameters of dielectric elastomer's thermodynamic system is given in Figs. 5–7. As shown in Fig. 5, with increase of stretch, the critical nominal electric field of dielectric elastomer decreases. From Fig. 6, it can be seen that with the increase of stretch, critical stretch decreases.

Table 2 summarizes certain values of critical nominal electric field and critical stretch of dielectric elastomer under different temperatures and stretches. As an example, in the state of equal biaxial stretch, the critical nominal electric field of dielectric elastomer is 0.695 $\sqrt{NkT_0/\varepsilon_r}$ and 0.851 $\sqrt{NkT_0/\varepsilon_r}$ when the temperatures are 273 and 373 K respectively. The critical stretch is 1.26, which is the same as that obtained by Zhao and Suo (2007).
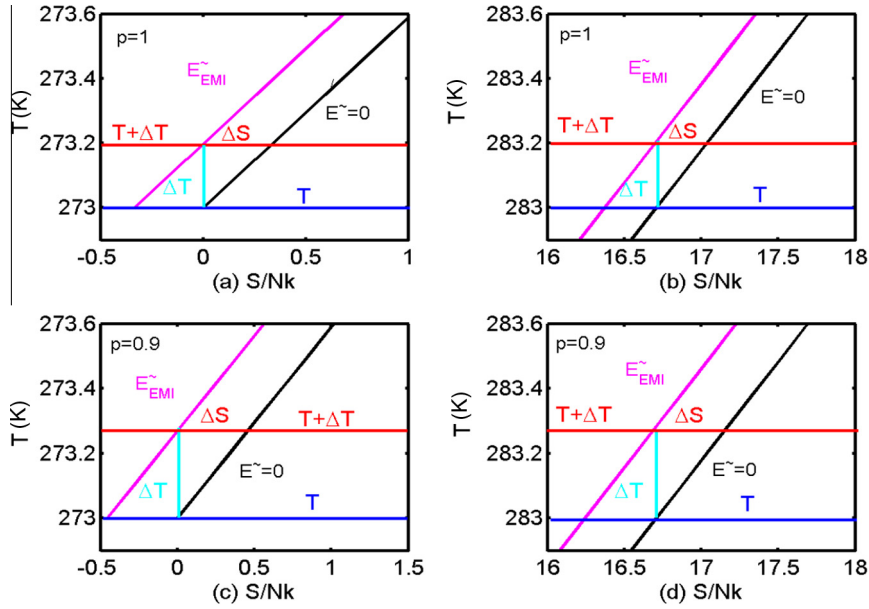
Fig. 12. The electric-induced temperature and entropy variation of dielectric elastomer.
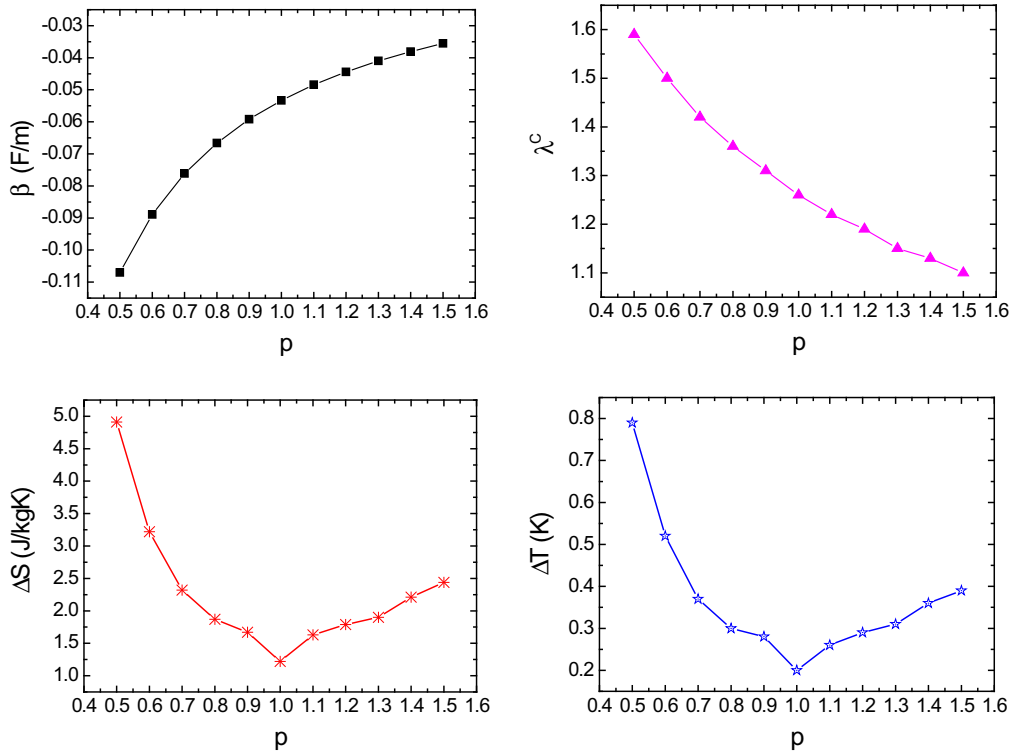

Fig. 13. Temperature and entropy variation of dielectric elastomer under different stretches.

5. Allowable energy range of dielectric elastomers under the coupling effect of electric field and temperature

The allowable energy range of dielectric elastomer is determined by the critical conditions which the elastomers could encounter during their working process. The main failure mode of soft materials such as dielectric elastomer is electromechanical instability (Zhao and Suo, 2010). Therefore, the $E^{\sim}_{EMI}$ value is the most important issue to determine the allowable energy range.

Considering biaxial stretch that the principle planar stretch ratio is set as p, i.e. $\lambda_2 = p\lambda_1 = p\lambda$, according to Eq. (19), at a constant s, when $\frac{s_1}{NkT_0} = \frac{T}{T_0} \frac{\{12\beta+4[\varepsilon_r+\alpha(T-T_0)]\}\lambda-p^{-2}\{30\beta+16[\varepsilon_r+\alpha(T-T_0)]\}\lambda^{-5}}{15\beta+6[\varepsilon_r+\alpha(T-T_0)]}$, the function $E^\sim(\lambda, s)$ reaches its maximum value. This maximum nominal electric field corresponds to the critical voltage for the onset of the electromechanical instability. By submitting it into Eqs. (18) and (19), the formulations of the dimensionless nominal electric displacement nominal electric field, the nominal displacement and the entropy can be induced respectively.

$$\frac{D^\sim}{\sqrt{NkT_0\varepsilon_r}} = \sqrt{\frac{\left[1+\frac{\alpha(T-T_0)+p\beta\lambda^2}{\varepsilon_r}\right]}{\left[p^{-2}+\frac{p^{-1}\beta\lambda^2}{2[\varepsilon_r+\alpha(T-T_0)+p\beta\lambda^2]}\right]}\left\{\frac{T}{T_0}\frac{3\beta+2[\varepsilon_r+\alpha(T-T_0)]}{15\beta+6[\varepsilon_r+\alpha(T-T_0)]}(\lambda^6+5p^{-2})\right\}} \quad (23)$$

$$\frac{E^\sim}{\sqrt{NkT_0/\varepsilon_r}} = \sqrt{\frac{1}{\left[p^{-2}+\frac{p^{-1}\beta\lambda^2}{2[\varepsilon_r+\alpha(T-T_0)+p\beta\lambda^2]}\right]\left[1+\frac{\alpha(T-T_0)+p\beta\lambda^2}{\varepsilon_r}\right]}\left\{\frac{T}{T_0}\frac{3\beta+2[\varepsilon_r+\alpha(T-T_0)]}{15\beta+6[\varepsilon_r+\alpha(T-T_0)]}(p^{-4}\lambda^{-2}+5p^{-6}\lambda^{-8})\right\}} \quad (24)$$

$$\frac{S}{Nk} = \frac{c_0}{Nk}\ln\frac{T}{T_0} - \frac{1}{2}[(1+p^2)\lambda^2+p^{-2}\lambda^{-4}-3]$$
$$+ \frac{T\alpha(p^{-2}\lambda^2+5p^{-4}\lambda^{-4})}{[2\varepsilon_r+2\alpha(T-T_0)+3p\beta\lambda^2]}$$
$$\times \frac{3\beta+2[\varepsilon_r+\alpha(T-T_0)]}{15\beta+6[\varepsilon_r+\alpha(T-T_0)]} \quad (25)$$

The allowable energy range on the plane of dielectric elastomer's nominal electric field and nominal electric displacement is the encircled range by four curves, which reflect the four critical conditions as initial temperature T_0, maximum temperature T, zero electric field $E^\sim = 0$, as well as electromechanical instability $E^\sim_{EMI}(T_0)$. Due to the fact that the dielectric elastomer gets more stable with the increasing temperature, the EMI curve under the lower temperature, i.e. initial temperature is selected as the controlling curve. Further, based on Eqs. (21) and (25), the allowable energy range on the plane of dielectric elastomer's temperature and entropy can also be determined.

Figs. 8 and 9 show the allowable energy range (the hatching range) in terms of nominal electric field vs. displacement and temperature vs. entropy respectively. Here, equal biaxial stretch is considered, i.e. $p = 1$, and the critical stretch value is 1.26 obtained from the EMI analysis. Similarly, the allowable energy ranges of dielectric elastomer undergoing unequal biaxial stretches in which $p = 0.9$ and 1.1 are plotted in Figs. 10 and 11. These results would be of great help in analyzing and understanding the physical process of dielectric elastomer undergoing different temperatures and electric fields.

Table 3 illustrates the entropy variation and allowable energy range under different conditions. For the condition of equal biaxial stretch, namely $\lambda_1 = \lambda_2 = \lambda$, the entropy variation is 1.22 J/kgK and the range is 122 J/kg. Evidently when dielectric elastomer undergoes equal biaxial stretch, allowable energy range is minimum.

6. Electric-induced temperature and entropy variation of dielectric elastomers

Applying an electric field to polar dielectric materials, represented by ferroelectric polymer materials such as P (VDF-TrFE) or P (VDF-TrFE-CFE), large variation in temperature and entropy will be induced due to the electrocaloric effect, which shows potential in designing and fabrication of cooling devices (Neese et al., 2008; Lu and Zhang, 2009). In this session, the temperature and entropy change of dielectric elastomer under high electric field will be evaluated.

When the electric filed applied on dielectric elastomer increases from a low voltage, the temperature and the entropy variation will be generated. The plane of work conjugated parameters entropy-temperature is applied to demonstrate this process. In this plane, the allowable range of dielectric elastomer's thermoelectromechanical system is encircled by four curves T, $T + \Delta T$, $E^\sim$ and $E^\sim_{EMI}$. In the allowable range, each point in a plane represents a specified temperature and voltage state of dielectric elastomer, each curve represents a temperature and voltage variation process, and each cycle represents a possible thermodynamics energy cycle. The maximum energy cycle process can be described in Fig. 12 since the temperature variation has reached a maximum value.

As shown in Fig. 12, for example, under the condition of equal biaxial, when the referenced temperatures are 273 and 283 K respectively, the temperature changes of dielectric elastomer are 0.2 and 0.2 K, the entropy changes are 1.22 and 1.23 J/kgK respectively. Under the condition of unequal biaxial ($p = 0.9$), the temperature changes of dielectric elastomer are 0.28 and 0.28 K, the entropy changes are 1.67 and 1.70 J/kgK respectively.

Under different stretch ratios in the principal plane, Fig. 13 represents the coefficient of electrostriction, the critical stretch, and the temperature and entropy variation of dielectric elastomer with electric field applied. Clearly, when $0.5 \leqslant p < 1$, with the increase of stretch ratio in the principal plane, the temperature and entropy variation decreases. To obtain a maximum temperature and entropy change, dielectric elastomer application cooling devices with small p should be selected.

7. Conclusion

In this paper, we firstly proposed the expression of dielectric constant relying on temperature and stretch, established free energy function of dielectric elastomer's

thermodynamic system under an adiabatic process, and deduced constitutive relation. Under equal and unequal biaxial conditions, the thermoelectromechanical stability of dielectric elastomer was studied. As a result, with the increase of temperature and decrease of the ratio between principal planar stretches, the stability of dielectric elastomer materials or structures increases. Then, the thermodynamic performance of dielectric elastomer undergoing various temperatures and electric fields is studied. The allowable energy range of dielectric elastomer under the condition of equal and unequal biaxial stretches is described. Finally, the electric-induced temperature and entropy variation of dielectric elastomer is evaluated. When $0.5 \leqslant p < 1$, with the decrease of the ratio between principal planar stretches, the temperature and the entropy variation increases. Numerical results will offer great help in guiding the design and fabrication of dielectric elastomer application devices, such as dielectric elastomer actuators operating in variable ambient temperatures.

Acknowledgments

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