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Thermo-electro-mechanical instability of dielectric elastomers

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Abstract
We here propose a new method to analyze the thermo-electro-mechanical instability of dielectric elastomers. The equilibrium equations in this thermodynamic system at different temperatures are initially established. We then obtained the critical nominal electric field and the critical stretch under various mechanical and thermal loads, involving the effects of different stretch regimes on the system stability, i.e. the equal-biaxial stretch, the unequal-biaxial stretch and the thickness elongation. Finally, numerical results showed that as the temperature increases, the critical nominal electric field and the stretch of the dielectric elastomer are strengthened, which consequently stabilize the system. The results provide guidance to the design and synthesis of dielectric elastomer-based devices, especially for those operating at various temperatures.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Dielectric elastomers are a subgroup of electroactive polymer materials that are capable of undergoing very large deformations while subject to an electric field, benefiting from attractive advantages such as large deformation, high elastic energy density, excellent conversion efficiency and high responsive speed [1, 3–10]. Such properties have enabled a variety of applications including actuators, sensors, energy harvests, refreshable Braille displays, active tactile displays, medical devices and space robotics [1–10].

The dielectric elastomer actuators have a relatively high failure rate [2], which greatly prevents practical application of dielectric elastomer materials [4, 5]. The types of failure of dielectric elastomer include the pull-in electro-mechanical instability, material strength rupture and dielectric strength [3, 4, 11, 13–27, 30–41, 43–46, 50]. Among these failure modes, electro-mechanical instability is the most concerned failure type, especially when the dielectric elastomer actuates in the absence of pre-stretch [2, 13, 27]. The research on dielectric elastomers' failure and nonlinear electro-mechanical stability has been an interesting topic in recent years [10–41, 43, 44]. Zhao and Suo proposed a general method using the free energy function of dielectric elastomers to analyze their electro-mechanical stability. Their theoretical study proved that the critical electric fields for electro-mechanical stability of dielectric elastomers can be improved by pre-stretch. The critical electric fields evaluated by the method are consistent with experimental results [10]. Díaz-Calleja's and Suo's groups investigated the electro-mechanical stable domain of dielectric elastomer [15, 22, 23]. The influence of different material models such as the

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neo-Hookean model [11, 15, 18, 20, 22–26, 38], Ogden model [21], Mooney–Rivlin model [14, 16, 19] and Arruda–Boyce model [13, 27] on the electro-mechanical stability of dielectric elastomer is then investigated by Suo's, Norrisa's and Liu's groups. Bifurcation and chaos in a thermodynamic system of dielectric elastomer was investigated by Díaz-Calleja, Zhu, Cai and Suo [11, 26]. Zhao and Suo proposed the programmable designing method of dielectric elastomer actuator [25]. Also, the electro-mechanical stability of dielectric elastomer undergoing homogeneous or inhomogeneous large deformation has been investigated by Zhao and He [18, 24]. The failure of dielectric elastomer will cause negative influence on its applications. It therefore highlights the research on the failure model and designing guidance.

In the research mentioned above, the dielectric elastomer is assumed to be in an isothermal environment. In this paper, we involved the influence of temperature on the thermodynamic system of dielectric elastomer, established the nonlinear thermodynamics equilibrium equation, and analyzed its thermo-electro-mechanical instability. We presented three groups of conjugated variable: nominal electric field and displacement, nominal stress and stretch, nominal volume entropy and temperature. We investigate the effects of three loading conditions, an equal-biaxial stretch, an unequal-biaxial stretch and a thickness elongation, on the thermo-electro-mechanical instability of dielectric elastomers' membranes. Besides, we also calculated the critical nominal electric field and critical stretch ratio under different parameters. The results show that along with the increase in temperature and material constant ratio α ($C_2 = \alpha C_1$, where C_1 and C_2 are material constants), or the decrease in the ratio between principal planar stretches, and nominal stress, the critical nominal electric field will be developed and the systemic thermo-electro-mechanical performance will be improved as well, which implies that its thermo-electro-mechanical stability is enhanced.

2. Thermo-electro-mechanical coupling system

We consider membranes of lengths L_1, L_2 and L_3 in a homogeneous state of deformation and of electrification, as illustrated in figure 1, in a thermo-electro-mechanical system of dielectric elastomers, in a reference state at temperature T_0 and in the absence of applied mechanical forces and voltage. The system is in a current state with a temperature T, subject to mechanical forces F_1, F_2 in the plane principal directions and subject to voltage U between the two electrodes. The dielectric elastomer membrane deforms to $\lambda_1 L_1$, $\lambda_2 L_2$ and $\lambda_3 L_3$, and the magnitude of the electric charge on either electrode is Q, where λ_1, λ_2 and λ_3 are the principal stretches and S is the entropy of the dielectric elastomer. The dielectric elastomer is taken to be incompressible, $\lambda_3 = 1/(\lambda_1\lambda_2)$.

We define the nominal stresses by the mechanical forces divided by the area of the elastomer in the undeformed state, $\sigma_1^{\sim} = \frac{F_1}{L_2L_3}$, $\sigma_2^{\sim} = \frac{F_2}{L_1L_3}$, the nominal electric field by the voltage in the deformed state divided by the thickness of the elastomer in the undeformed state $E^{\sim} = \frac{U}{L_3}$ and define the nominal electric displacement as the charge on an electrode in the deformed state divided by the area of the electrode in the undeformed state, $D^{\sim} = \frac{Q}{L_1L_2}$, the nominal entropy density by total entropy divided by the volume of the elastomer in the undeformed state, $s^{\sim} = \frac{S}{L_1L_2L_3}$. In contrast, the true stresses are defined as the mechanical force divided by the area of the elastomer in the current state, $\sigma_1 = \frac{F_1}{\lambda_2\lambda_3L_2L_3}$, $\sigma_2 = \frac{F_2}{\lambda_1\lambda_3L_1L_3}$. The true electric field is defined as the voltage divided by the thickness of the elastomer in the current state, $E = \frac{U}{\lambda_3L_3}$, and the true electric displacement is defined as the charge divided by the area of the electrode in the deformed state, $D = \frac{Q}{\lambda_1\lambda_2L_1L_2}$, the true entropy density by total entropy divided by the volume of the elastomer in the deformed state, $s = \frac{S}{\lambda_1\lambda_2\lambda_3L_1L_2L_3}$. For an incompressible dielectric elastomer, $s^{\sim} = s$.

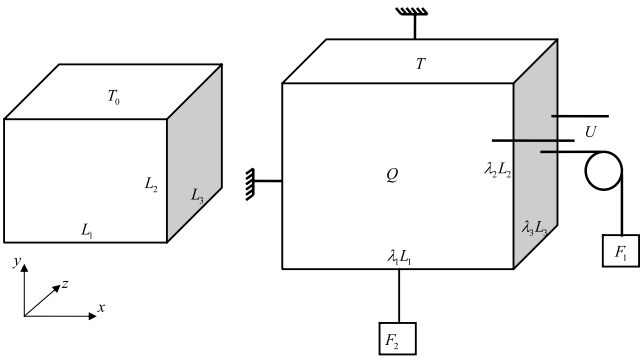

Figure 1. The thermo-electro-mechanical coupling system of dielectric elastomers.

Assuming that dielectric elastomer is incompressible, the free energy function is a function of the stretch, the nominal electric displacement and the current temperature. The dielectric elastomer, the battery and the weights, constitute the thermodynamic system [11], characterized by four generalized coordinates λ_1, λ_2, $D^{\sim}$, T and four control parameters F_1, F_2, U and S. When the generalized coordinates vary by small amounts, $\delta\lambda_1$, $\delta\lambda_2$, $\delta D^{\sim}$ and δT, the free energy of the thermodynamics system varies by

$$\begin{aligned} \mathrm{d}W(\lambda_1, \lambda_2, T, D^{\sim}) = {} & \frac{\partial W(\lambda_1, \lambda_2, D^{\sim}, T)}{\partial \lambda_1}\,\mathrm{d}\lambda_1 \\ & + \frac{\partial W(\lambda_1, \lambda_2, D^{\sim}, T)}{\partial \lambda_2}\,\mathrm{d}\lambda_2 + \frac{\partial W(\lambda_1, \lambda_2, D^{\sim}, T)}{\partial D^{\sim}}\,\mathrm{d}D^{\sim} \\ & - \frac{\partial W(\lambda_1, \lambda_2, D^{\sim}, T)}{\partial T}\,\mathrm{d}T. \end{aligned} \tag{1}$$

Thermodynamics indicates that a stable equilibrium state should minimize $W(\lambda_1, \lambda_2, D^{\sim}, T)$. Therefore, in a dielectric elastomer thermo-electro-mechanical coupling system, the nominal stress, $\sigma_1^{\sim}(\lambda_1, \lambda_2, T, D^{\sim})$, $\sigma_2^{\sim}(\lambda_1, \lambda_2, T, D^{\sim})$, the nominal electric field, $E^{\sim}(\lambda_1, \lambda_2, T, D^{\sim})$, the nominal entropy per unit volume, $s^{\sim}(\lambda_1, \lambda_2, T, D^{\sim})$, can be expressed respectively by the following equations

$$\sigma_1^{\sim}(\lambda_1, \lambda_2, T, D^{\sim}) = \frac{\partial W(\lambda_1, \lambda_2, D^{\sim}, T)}{\partial \lambda_1} \tag{2}$$

$$\sigma_2^{\sim}(\lambda_1, \lambda_2, T, D^{\sim}) = \frac{\partial W(\lambda_1, \lambda_2, D^{\sim}, T)}{\partial \lambda_2} \tag{3}$$

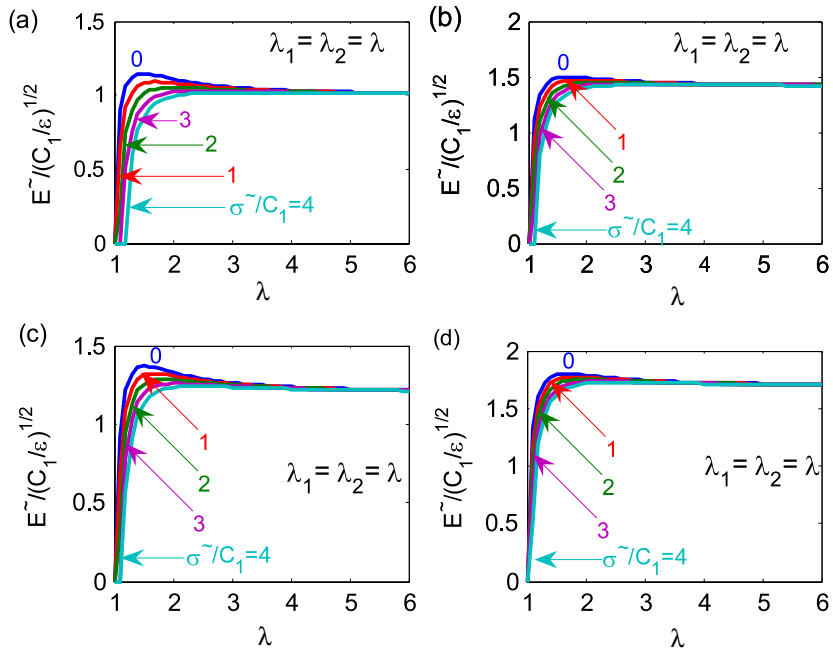

Figure 2. The relationship between nominal electric field and the stretch of the thermo-electro-mechanical coupling system of dielectric elastomers under specialized load conditions, namely $\lambda_1 = \lambda_2 = \lambda$. (a) $T = T_0, \alpha = 1$, (b) $T = T_0, \alpha = 2$, (c) $T = 1.44T_0, \alpha = 1$, (d) $T = 1.44T_0, \alpha = 2$.

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

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

The corresponding true stresses, the true electric field and the true entropy are

$$\sigma_1(\lambda_1, \lambda_2, T, D^{\sim}) = \lambda_1 \frac{\partial W(\lambda_1, \lambda_2, D^{\sim}, T)}{\partial \lambda_1} \tag{6}$$

$$\sigma_2(\lambda_1, \lambda_2, T, D^{\sim}) = \lambda_2 \frac{\partial W(\lambda_1, \lambda_2, D^{\sim}, T)}{\partial \lambda_2} \tag{7}$$

$$E(\lambda_1, \lambda_2, T, D^{\sim}) = \lambda_1\lambda_2 \frac{\partial W(\lambda_1, \lambda_2, D^{\sim}, T)}{\partial D^{\sim}} \tag{8}$$

$$s(\lambda_1, \lambda_2, T, D^{\sim}) = -\frac{\partial W(\lambda_1, \lambda_2, D^{\sim}, T)}{\partial T}. \tag{9}$$

And the Hessian matrix of the dielectric elastomer thermo-electro-mechanical coupling system is

$$H = \begin{bmatrix} \frac{\partial^2 W(\lambda_1,)}{\partial \lambda_1^2} & \frac{\partial^2 W(\lambda_1,\lambda_2,)}{\partial \lambda_1 \partial \lambda_2} & \frac{\partial^2 W(\lambda_1,D^{\sim},)}{\partial \lambda_1 \partial D^{\sim}} & \frac{\partial^2 W(\lambda_1,T,)}{\partial \lambda_1 \partial T} \\ \frac{\partial^2 W(\lambda_1,\lambda_2,)}{\partial \lambda_1 \partial \lambda_2} & \frac{\partial^2 W(\lambda_2,)}{\partial \lambda_2^2} & \frac{\partial^2 W(\lambda_2,D^{\sim},)}{\partial \lambda_2 \partial D^{\sim}} & \frac{\partial^2 W(\lambda_2,T,)}{\partial \lambda_2 \partial T} \\ \frac{\partial^2 W(\lambda_1,D^{\sim},)}{\partial \lambda_1 \partial D^{\sim}} & \frac{\partial^2 W(\lambda_2,D^{\sim},)}{\partial \lambda_2 \partial D^{\sim}} & \frac{\partial^2 W(D^{\sim},)}{\partial D^{\sim 2}} & \frac{\partial^2 W(D^{\sim},T,)}{\partial D^{\sim} \partial T} \\ \frac{\partial^2 W(\lambda_1,T,)}{\partial \lambda_1 \partial T} & \frac{\partial^2 W(\lambda_2,T,)}{\partial \lambda_2 \partial T} & \frac{\partial^2 W(D^{\sim},T,)}{\partial D^{\sim} \partial T} & \frac{\partial^2 W(T,)}{\partial T^2} \end{bmatrix}. \tag{10}$$

To obtain a stable state for the dielectric elastomer thermo-electro-mechanical coupling system (DE-TEM-CS), the determinant of the Hessian matrix should be positive. When the DE-TEM-CS reaches the critical point, $\det(H) = 0$. Solving the determinant $\det(H) = 0$ of equation (10), we obtain the critical thermo-electro-mechanical stability parameters of dielectric elastomer, such as the critical nominal electric field $E^{\sim}_{\max}(\lambda_1, \lambda_2, T, D^{\sim})$, the critical true electric field $E_{\max}(\lambda_1, \lambda_2, T, D^{\sim})$; the critical nominal stress $\sigma_c^{\sim}(\lambda_1, \lambda_2, T, D^{\sim})$ and the critical true stress $\sigma_c(\lambda_1, \lambda_2, T, D^{\sim})$; the critical nominal entropy per unit volume $s_c^{\sim}(\lambda_1, \lambda_2, T, D^{\sim})$ and the critical true entropy per unit volume $s_c(\lambda_1, \lambda_2, T, D^{\sim})$.

3. Constitutive relation

Due to the dielectric elastomer's incompressibility and considering the effect of the temperature on the dielectric elastomer thermodynamic system, the free energy function can be expressed as follows [10–41]:

$$W(\lambda_1, \lambda_2, \lambda_1^{-1}\lambda_2^{-1}, T, D^{\sim}) = \xi(\lambda_1, \lambda_2, \lambda_1^{-1}\lambda_2^{-1}, T) + \eta(\lambda_1, \lambda_2, \lambda_1^{-1}\lambda_2^{-1}, D^{\sim}). \tag{11}$$

In equation (11), $W(\lambda_1, \lambda_2, \lambda_1^{-1}\lambda_2^{-1}, T, D^{\sim})$ is the free energy of the DE-TEM-CS as a function of the three generalized coordinates, the stretch, λ_1 and λ_2, the current temperature, T, and the nominal electric displacement, $D^{\sim}$. $\xi(\lambda_1, \lambda_2, \lambda_1^{-1}\lambda_2^{-1})$ is the thermo-elastic strain energy, $\eta(\lambda_1, \lambda_2, \lambda_1^{-1}\lambda_2^{-1}, D^{\sim})$ is the electric field energy. In the thermodynamics system, to simplify this work but without losing its physical picture, we take the following assumptions: the system is an adiabatic process, the dielectric elastomers are ideal hyperelastic elastomers, the electrode is the ideal fluid and compliant, the temperature varies linearly. Therefore, the thermo-elastic strain energy can be expressed as

$$\xi(\lambda_1, \lambda_2, \lambda_1^{-1}\lambda_2^{-1}, T) = \frac{T}{2T_0}[C_1(\lambda_1^2 + \lambda_2^2 + \lambda_1^{-2}\lambda_2^{-2} - 3) + C_2(\lambda_1^{-2} + \lambda_2^{-2} + \lambda_1^2\lambda_2^2 - 3)] + c_0\left[(T - T_0) - T\ln\frac{T}{T_0}\right]. \tag{12}$$

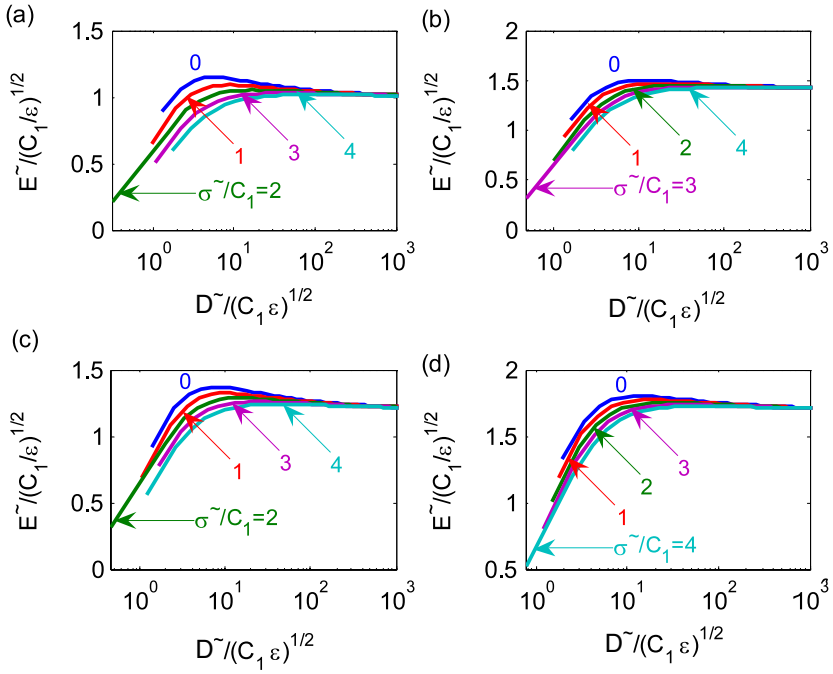

Figure 3. The relationship between the nominal electric field and the nominal electric displacement of the thermo-electro-mechanical coupling system of dielectric elastomers under specialized load conditions, namely $\lambda_1 = \lambda_2 = \lambda$. (a) $T = T_0$, $\alpha = 1$, (b) $T = T_0$, $\alpha = 2$, (c) $T = 1.44T_0$, $\alpha = 1$, (d) $T = 1.44T_0$, $\alpha = 2$.

In the right-hand side of equation (12), the first item is the thermal hyperelastic energy. The second item is the thermal contribution [29]. We employ a Mooney–Rivlin model with two material constants to describe the hyperelastic performances (Mooney and Rivlin assumed the rubbery polymer was isotropic, so that the strain energy is only a function of stretch in both directions, thus we have the model: $W = C_1(\lambda_1^2+\lambda_2^2+\lambda_1^{-2}\lambda_2^{-2}-3)+C_2(\lambda_1^{-2}+\lambda_2^{-2}+\lambda_1^2\lambda_2^2-3)$. According to the experimental validation, this model is able to capture the stress–strain behavior when the strain is under 200%. Since our critical stretch is under 200%, so that we select this model in our study [42]), where C_1, C_2 are material constants determined by experiments. T_0 is the reference temperature. c_0 is the specific heat for dielectric elastomers.

The material is assumed to be an ideal dielectric elastomer, where the dielectric behavior of the elastomer is taken to be liquid-like, unaffected by deformation. The true electric field and the true electric displacement are in linear relation. The electric field energy is [10, 11]

$$\eta(\lambda_1, \lambda_2, \lambda_1^{-1}\lambda_2^{-1}, D^{\sim}) = \frac{D^{\sim 2}}{2\varepsilon}\lambda_1^{-2}\lambda_2^{-2} \tag{13}$$

ε is the relative permittivity of the dielectric elastomer, $\varepsilon = 3.5 \times 10^{-11}$ F m^{-1} [13, 23].

Substituting equations (11)–(13) into (2)–(5), we obtain the nominal stresses, the nominal electric field and the nominal entropy per unit volume of dielectric elastomer as

$$\sigma_1^{\sim}(\lambda_1, \lambda_2, T, D^{\sim}) = \frac{T}{T_0}[C_1(\lambda_1 - \lambda_1^{-3}\lambda_2^{-2}) + C_2(\lambda_1\lambda_2^2 - \lambda_1^{-3})] - \frac{D^{\sim 2}}{\varepsilon}\lambda_1^{-3}\lambda_2^{-2} \tag{14}$$

$$\sigma_2^{\sim}(\lambda_1, \lambda_2, T, D^{\sim}) = \frac{T}{T_0}[C_1(\lambda_2 - \lambda_1^{-2}\lambda_2^{-3}) + C_2(\lambda_2\lambda_1^2 - \lambda_2^{-3})] - \frac{D^{\sim 2}}{\varepsilon}\lambda_1^{-2}\lambda_2^{-3} \tag{15}$$

$$E^{\sim}(\lambda_1, \lambda_2, D^{\sim}) = \frac{D^{\sim}}{\varepsilon}\lambda_1^{-2}\lambda_2^{-2} \tag{16}$$

$$s^{\sim}(\lambda_1, \lambda_2, T) = c_0 \ln\frac{T}{T_0} - \frac{1}{T_0}\left[\frac{C_1}{2}(\lambda_1^2+\lambda_2^2+\lambda_1^{-2}\lambda_2^{-2}-3) + \frac{C_2}{2}(\lambda_1^{-2}+\lambda_2^{-2}+\lambda_1^2\lambda_2^2-3)\right]. \tag{17}$$

Correspondingly, the true stresses, the true electric field and the true entropy are

$$\sigma_1(\lambda_1, \lambda_2, T, D^{\sim}) = \frac{T}{T_0}[C_1(\lambda_1^2 - \lambda_1^{-2}\lambda_2^{-2}) + C_2(\lambda_1^2\lambda_2^2 - \lambda_1^{-2})] - \frac{D^{\sim 2}}{\varepsilon}\lambda_1^{-2}\lambda_2^{-2} \tag{18}$$

$$\sigma_2(\lambda_1, \lambda_2, T, D^{\sim}) = \frac{T}{T_0}[C_1(\lambda_2^2 - \lambda_1^{-2}\lambda_2^{-2}) + C_2(\lambda_1^2\lambda_2^2 - \lambda_2^{-2})] - \frac{D^{\sim 2}}{\varepsilon}\lambda_1^{-2}\lambda_2^{-2} \tag{19}$$

$$E(\lambda_1, \lambda_2, D^{\sim}) = \frac{D^{\sim}}{\varepsilon}\lambda_1^{-1}\lambda_2^{-1} \tag{20}$$

$$s(\lambda_1, \lambda_2, T) = c_0 \ln\frac{T}{T_0} - \frac{1}{T_0}\left[\frac{C_1}{2}(\lambda_1^2+\lambda_2^2+\lambda_1^{-2}\lambda_2^{-2}-3) + \frac{C_2}{2}(\lambda_1^{-2}+\lambda_2^{-2}+\lambda_1^2\lambda_2^2-3)\right]. \tag{21}$$

4. Thermo-electro-mechanical instability

When a mechanical force field, together with an electric field and a thermal field are applied to a layer of a dielectric elastomer, the membrane reduces in thickness and expands in area. As the electric field, the mechanical force field and the thermal field increase, the elastomer thins down, so that the same voltage will induce an even higher electric field. The positive feedback may cause the elastomer to thin down drastically, resulting in an even larger electric field. When the electric field exceeds the critical value, the electrical breakdown takes place and the actuator fails. This process is called here thermo-electro-mechanical instability.

The Hessian matrix of dielectric elastomer thermo-electro-mechanical coupling system can be expressed as

$$H = \begin{bmatrix} \frac{\partial \sigma_1^{\sim}(\lambda_1,\lambda_2,T,D^{\sim})}{\partial \lambda_1} & \frac{\partial \sigma_1^{\sim}(\lambda_1,\lambda_2,T,D^{\sim})}{\partial \lambda_2} & \frac{\partial \sigma_1^{\sim}(\lambda_1,\lambda_2,T,D^{\sim})}{\partial D^{\sim}} & \frac{\partial \sigma_1^{\sim}(\lambda_1,\lambda_2,T,D^{\sim})}{\partial T} \\ \frac{\partial \sigma_2^{\sim}(\lambda_1,\lambda_2,T,D^{\sim})}{\partial \lambda_1} & \frac{\partial \sigma_2^{\sim}(\lambda_1,\lambda_2,T,D^{\sim})}{\partial \lambda_2} & \frac{\partial \sigma_2^{\sim}(\lambda_1,\lambda_2,T,D^{\sim})}{\partial D^{\sim}} & \frac{\partial \sigma_2^{\sim}(\lambda_1,\lambda_2,T,D^{\sim})}{\partial T} \\ \frac{\partial E^{\sim}(\lambda_1,\lambda_2,D^{\sim})}{\partial \lambda_1} & \frac{\partial E^{\sim}(\lambda_1,\lambda_2,D^{\sim})}{\partial \lambda_2} & \frac{\partial E^{\sim}(\lambda_1,\lambda_2,D^{\sim})}{\partial D^{\sim}} & \frac{\partial E^{\sim}(\lambda_1,\lambda_2,D^{\sim})}{\partial T} \\ \frac{\partial s^{\sim}(\lambda_1,\lambda_2,T)}{\partial \lambda_1} & \frac{\partial s^{\sim}(\lambda_1,\lambda_2,T)}{\partial \lambda_2} & \frac{\partial s^{\sim}(\lambda_1,\lambda_2,T)}{\partial D^{\sim}} & \frac{\partial s^{\sim}(\lambda_1,\lambda_2,T)}{\partial T} \end{bmatrix} \quad (22)$$

where

$$\frac{\partial \sigma_1^{\sim}(\lambda_1,\lambda_2,T,D^{\sim})}{\partial \lambda_1} = \frac{T}{T_0}[C_1(1+3\lambda_1^{-4}\lambda_2^{-2}) + C_2(\lambda_2^2+3\lambda_1^{-4})] + 3\frac{D^{\sim 2}}{\varepsilon}\lambda_1^{-4}\lambda_2^{-2}$$

$$\frac{\partial \sigma_1^{\sim}(\lambda_1,\lambda_2,T,D^{\sim})}{\partial \lambda_2} = \frac{\partial \sigma_2^{\sim}(\lambda_1,\lambda_2,T,D^{\sim})}{\partial \lambda_1} = 2\frac{T}{T_0}(C_1\lambda_1^{-3}\lambda_2^{-3} + C_2\lambda_1\lambda_2) + 2\frac{D^{\sim 2}}{\varepsilon}\lambda_1^{-3}\lambda_2^{-3}$$

$$\frac{\partial \sigma_1^{\sim}(\lambda_1,\lambda_2,T,D^{\sim})}{\partial D^{\sim}} = \frac{\partial E^{\sim}(\lambda_1,\lambda_2,D^{\sim})}{\partial \lambda_1} = -2\frac{D^{\sim}}{\varepsilon}\lambda_1^{-3}\lambda_2^{-2}$$

$$\frac{\partial \sigma_1^{\sim}(\lambda_1,\lambda_2,T,D^{\sim})}{\partial T} = \frac{\partial s^{\sim}(\lambda_1,\lambda_2,T)}{\partial \lambda_1} = \frac{1}{T_0}[C_1(\lambda_1-\lambda_1^{-3}\lambda_2^{-2}) + C_2(\lambda_1\lambda_2^2-\lambda_1^{-3})]$$

$$\frac{\partial \sigma_2^{\sim}(\lambda_1,\lambda_2,T,D^{\sim})}{\partial \lambda_2} = \frac{T}{T_0}[C_1(1+3\lambda_1^{-2}\lambda_2^{-4}) + C_2(\lambda_1^2+3\lambda_2^{-4})] + 3\frac{D^{\sim 2}}{\varepsilon}\lambda_1^{-2}\lambda_2^{-4}$$

$$\frac{\partial \sigma_2^{\sim}(\lambda_1,\lambda_2,T,D^{\sim})}{\partial D^{\sim}} = \frac{\partial E^{\sim}(\lambda_1,\lambda_2,D^{\sim})}{\partial \lambda_2} = 2\frac{D^{\sim}}{\varepsilon}\lambda_1^{-2}\lambda_2^{-3}$$

$$\frac{\partial \sigma_2^{\sim}(\lambda_1,\lambda_2,T,D^{\sim})}{\partial T} = \frac{\partial s^{\sim}(\lambda_1,\lambda_2,T)}{\partial \lambda_2} = \frac{1}{T_0}[C_1(\lambda_2-\lambda_1^{-2}\lambda_2^{-3}) + C_2(\lambda_2\lambda_1^2-\lambda_2^{-3})]$$

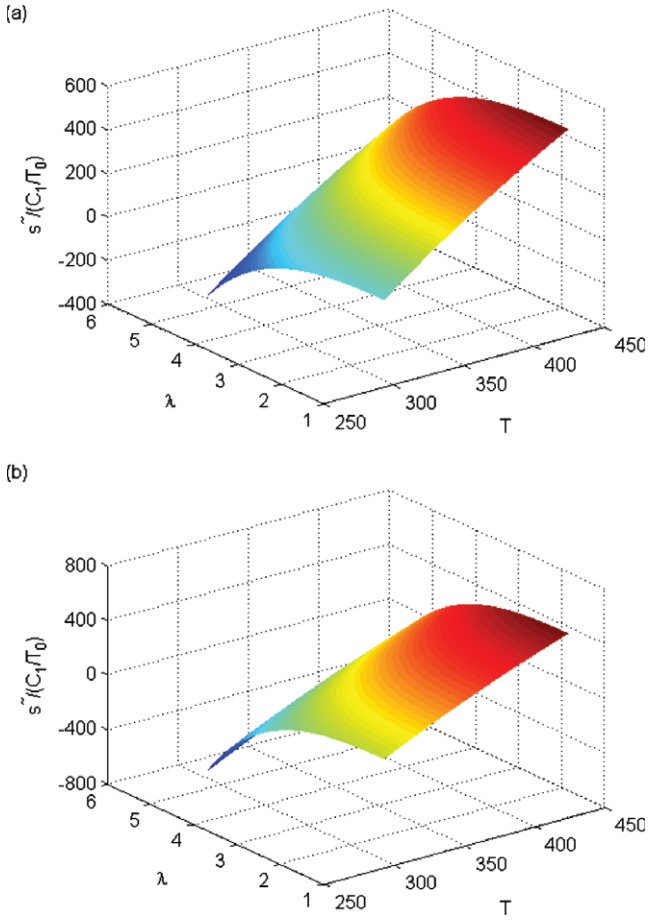

Figure 4. Entropy of the thermo-electro-mechanical coupling system of dielectric elastomers at different temperatures and stretches under specialized load conditions, namely $\lambda_1 = \lambda_2 = \lambda$: (a) $\alpha = 1$; (b) $\alpha = 2$.

$$\frac{\partial E^{\sim}(\lambda_1,\lambda_2,D^{\sim})}{\partial D^{\sim}} = \frac{1}{\varepsilon}\lambda_1^{-2}\lambda_2^{-2}$$

$$\frac{\partial E^{\sim}(\lambda_1,\lambda_2,D^{\sim})}{\partial T} = \frac{\partial s^{\sim}(\lambda_1,\lambda_2,T)}{\partial D^{\sim}} = 0$$

$$\frac{\partial s^{\sim}(\lambda_1,\lambda_2,T)}{\partial T} = \frac{c_0}{T}.$$

Consider equation (22), by solving $\det(H) = 0$, we obtain $E_{\max}^{\sim}(\lambda_1,\lambda_2,D^{\sim})$, $E_{\max}(\lambda_1,\lambda_2,D^{\sim})$, $\sigma_c^{\sim}(\lambda_1,\lambda_2,T,D^{\sim})$, $\sigma_c(\lambda_1,\lambda_2,T,D^{\sim})$, $s_c^{\sim}(\lambda_1,\lambda_2,T)$ and $s_c(\lambda_1,\lambda_2,T)$.

4.1. Effect of equal-biaxial stretch on thermo-electro-mechanical instability

In order to investigate the thermo-electro-mechanical instability of a dielectric elastomer, we postulate that $C_2 = \alpha C_1$, where α is a material constant ratio [14, 16, 19]. Evidently, the material constant ratios are different for different dielectric elastomer materials (such as BJB TC-A/B-C, 3M VHB 4905/4910, VHB 4910-HDDA, VHB 4905-TMPTMA CF19-2186 silicone, Dow Corning HS3 silicone, Nusil CF19-2186 silicone, Dow Corning Sylgard 186 [3–5]).

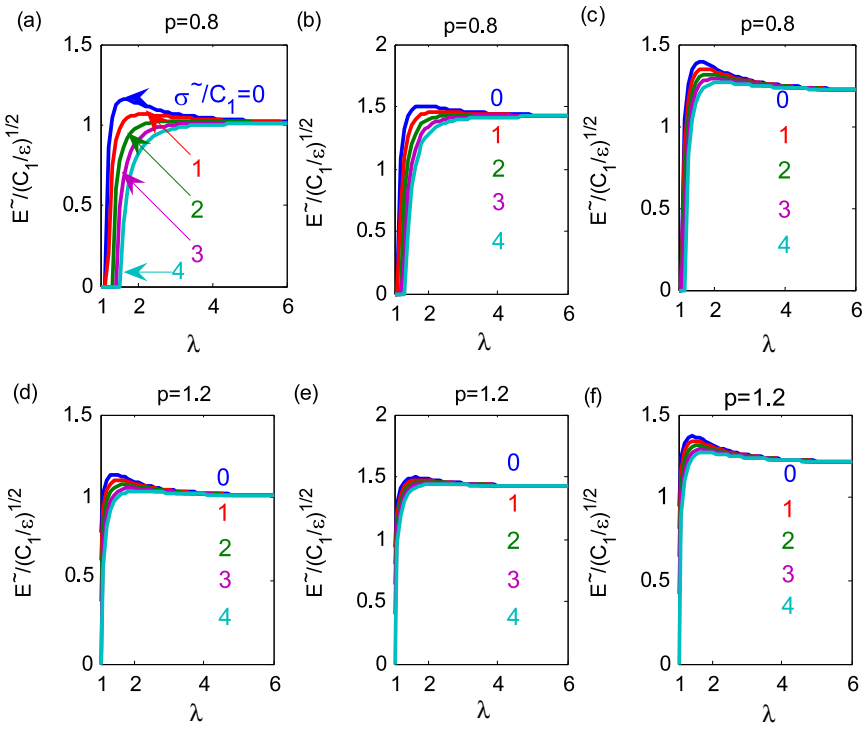

Figure 5. The relationship between nominal electric field and the stretch of the thermo-electro-mechanical coupling system of dielectric elastomers under specialized load conditions, namely $\lambda_2 = p\lambda_1 = p\lambda$. (a) $T = T_0$, $\alpha = 1$, $p = 0.8$, (b) $T = T_0$, $\alpha = 2$, $p = 0.8$, (c) $T = 1.44T_0$, $\alpha = 1$, $p = 0.8$, (d) $T = T_0$, $\alpha = 1$, $p = 1.2$, (e) $T = T_0$, $\alpha = 2$, $p = 1.2$, (f) $T = 1.44T_0$, $\alpha = 1$, $p = 1.2$.

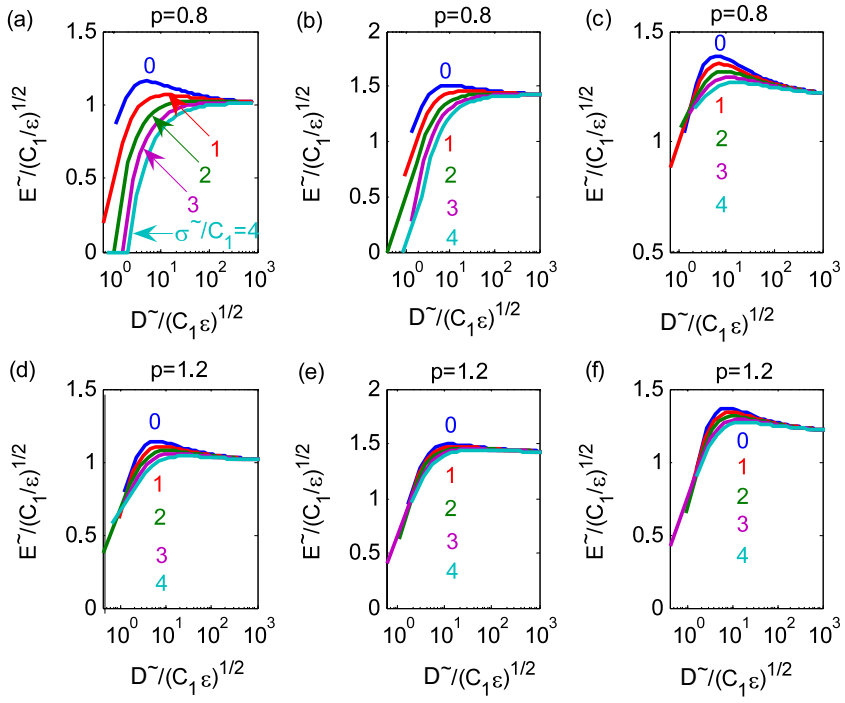

Figure 6. The relationship between the nominal electric field and nominal electric displacement of the thermo-electro-mechanical coupling system of dielectric elastomers under specialized load conditions, namely $\lambda_2 = p\lambda_1 = p\lambda$. (a) $T = T_0$, $\alpha = 1$, $p = 0.8$, (b) $T = T_0$, $\alpha = 2$, $p = 0.8$, (c) $T = 1.44T_0$, $\alpha = 1$, $p = 0.8$, (d) $T = T_0$, $\alpha = 1$, $p = 1.2$, (e) $T = T_0$, $\alpha = 2$, $p = 1.2$, (f) $T = 1.44T_0$, $\alpha = 1$, $p = 1.2$.

Subsequently, we study the effect of equal-biaxial stretch on the thermo-electro-mechanical instability of dielectric elastomers. Let $\lambda_2 = \lambda_1 = \lambda$, according to the equations (14)–(17), the nominal electric field, the nominal electric displacement and the nominal entropy per unit volume can be derived as follows:

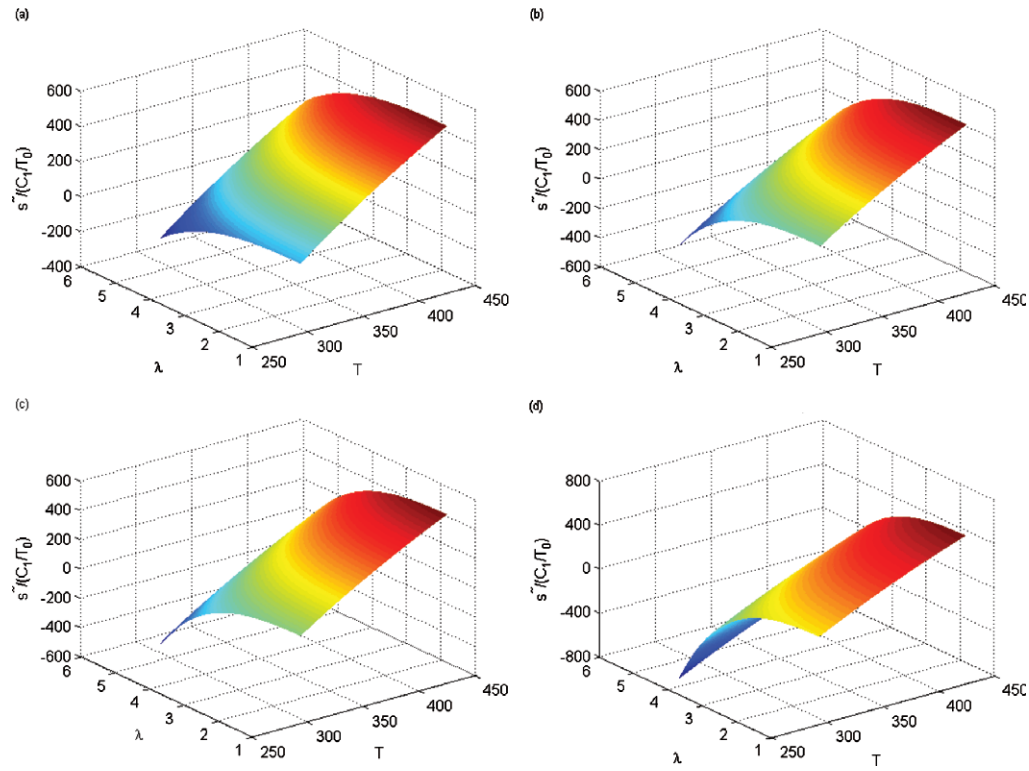

Figure 7. Entropy of the thermo-electro-mechanical coupling system of dielectric elastomers at different temperatures and stretches under specialized load conditions, namely $\lambda_2 = p\lambda_1 = p\lambda$. (a) $\alpha = 1$, $p = 0.8$, (b) $\alpha = 2$, $p = 0.8$, (c) $\alpha = 1$, $p = 1.2$, (d) $\alpha = 2$, $p = 1.2$.

$$\frac{D^{\sim}}{\sqrt{C_1\varepsilon}} = \sqrt{\frac{T}{T_0}[(\lambda^6 - 1) + \alpha(\lambda^8 - \lambda^2)] - \frac{\sigma^{\sim}}{2C_1}\lambda^5} \tag{23}$$

$$\frac{E^{\sim}}{\sqrt{C_1/\varepsilon}} = \sqrt{\frac{T}{T_0}[(\lambda^{-2} - \lambda^{-8}) + \alpha(1 - \lambda^{-6})] - \frac{\sigma^{\sim}}{2C_1}\lambda^{-3}} \tag{24}$$

$$\frac{s^{\sim}}{C_1/T_0} = \frac{c_0}{C_1/T_0}\ln\frac{T}{T_0} - \frac{1}{2}[(2\lambda^2 + \lambda^{-4} - 3) + \alpha(2\lambda^{-2} + \lambda^4 - 3)]. \tag{25}$$

In the following analysis session, representative parameters are selected, $T_0 = 293$ K, $T_{\max} = 423$ K [4], $c_0 = 1.7 \times 10^6$ J km^{-3}, $C_1 = 0.33 \times 10^6$ Pa, and hence dimensionless parameter $\frac{c_0}{C_1/T_0} = 1.5 \times 10^3$.

Under equal-biaxial stretch conditions and different temperatures, the relationships among dimensionless parameters of the dielectric elastomer thermo-electro-mechanical coupling system are presented in figures 2–4, namely the relationship between nominal electric field and nominal electric displacement, the relationship between nominal electric field and stretch and the relationship among entropy, temperature and stretch.

Figure 2 plots the $\frac{E^{\sim}}{\sqrt{C_1/\varepsilon}}$ as a function of λ when $\alpha = 1, 2$ and $T = T_0$, $1.44T_0$, respectively. In each curve, with the increase in stretch λ, the nominal electric field $\frac{E^{\sim}}{\sqrt{C_1/\varepsilon}}$ increases. When the critical nominal electric field is reached, the nominal electric field decreases and approaches a constant value.

Figure 3 shows the relationship between $\frac{E^{\sim}}{\sqrt{C_1/\varepsilon}}$ and $\frac{D^{\sim}}{\sqrt{C_1\varepsilon}}$ when $\alpha = 1, 2$ and $T = T_0$, $1.44T_0$, respectively. In each curve, $\frac{\sigma^{\sim}}{C_1}$ takes different values such as 0–4 and $E^{\sim}$ reaches its peak values. Before $E^{\sim}$ reaches its peak value, the Hessian matrix is positive definite while after the peak value, the Hessian matrix is negative definite. Simply speaking, in the peak point, $\det(H) = 0$. As the value $\frac{\sigma^{\sim}}{C_1}$ increases, the nominal electric field decreases.

Figure 4 illustrates the entropy of a dielectric elastomer thermo-electro-mechanical coupling system of different dielectric elastomer materials ($\alpha = 1, 2$) under the loading condition as λ_1 and T. As shown in figure 4, the entropy of the thermodynamics system decreases with the increase in stretch or with the decrease in temperature.

4.2. Effect of unequal-biaxial stretch on thermo-electro-mechanical instability

Unequal-biaxial experiments on dielectric elastomers are common [41]. Furthermore, what we need in many cases is the biaxial stretching in accordance with the specific ratio between two axes. The equal-biaxial stretch is a specific case of unequal-biaxial stretch and many dielectric elastomer devices operate under different conditions [47, 48], for instance, the apex of an inflating membrane and an expanding balloon [6, 26, 49]. As a matter of fact, an equal-biaxial stretch plane is an ideal case [47, 48] that cannot be satisfied perfectly in experiments. Therefore we have to consider the biaxial stretching in accordance with the specific ratio between two axes, namely unequal-biaxial. Let $\lambda_2 = p\lambda_1$, with $p >$

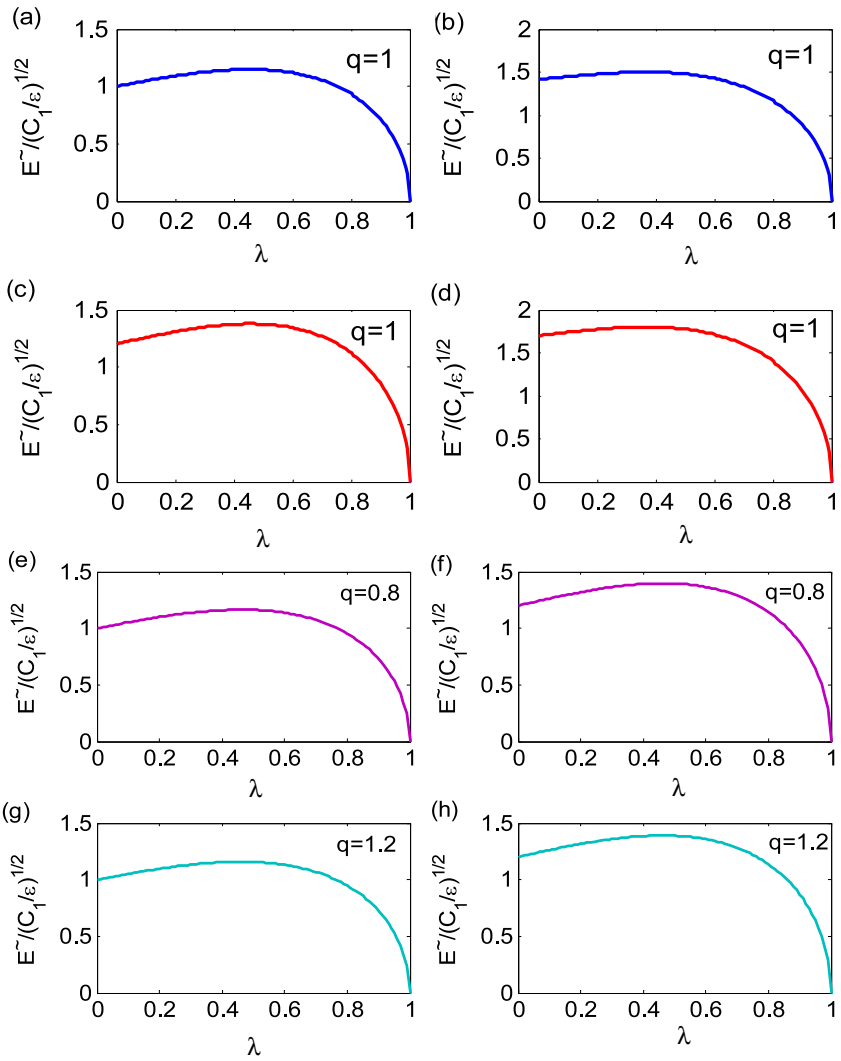

Figure 8. The relation between nominal electric field and the stretch of the thermo-electro-mechanical coupling system of dielectric elastomers under specialized load conditions, namely $\lambda_3 = \lambda$, $\lambda_1 = q\lambda^{-1/2}$ and $\lambda_2 = \lambda^{-1/2}/q$. (a) $T = T_0$, $\alpha = 1$, $q = 1$, (b) $T = T_0$, $\alpha = 2$, $q = 1$, (c) $T = 1.44T_0$, $\alpha = 1$, $q = 1$, (d) $T = 1.44T_0$, $\alpha = 2$, $q = 1$, (e) $T = T_0$, $\alpha = 1$, $q = 0.8$, (f) $T = 1.44T_0$, $\alpha = 1$, $q = 0.8$, (g) $T = T_0$, $\alpha = 1$, $q = 1.2$, (h) $T = 1.44T_0$, $\alpha = 1$, $q = 1.2$.

0, which is the ratio between principal planar stretches, and $p = 1$ recovers the equal-biaxial condition. To simplify the formulation, let $\lambda_1 = \lambda$, then the nominal electric field, the nominal electric displacement and the nominal entropy per unit volume can be derived as follows:

$$\frac{D^{\sim}}{\sqrt{C_1\varepsilon}} = \left\{\frac{T}{2T_0}\{[(1+p^2)p^2\lambda^6 - 2] + \alpha[2p^4\lambda^8 - (p^2+1)\lambda^2]\} - \frac{\sigma^{\sim}}{2C_1}p^2\lambda^5\right\}^{1/2} \tag{26}$$

$$\frac{E^{\sim}}{\sqrt{C_1/\varepsilon}} = \left\{\frac{T}{2T_0}\{[(p^{-2}+1)\lambda^{-2} - 2p^{-4}\lambda^{-8}] + \alpha[2 - (p^{-2}+1)\lambda^{-6}]\} - \frac{\sigma^{\sim}}{2C_1}p^{-2}\lambda^{-3}\right\}^{1/2} \tag{27}$$

$$\frac{s^{\sim}}{C_1/T_0} = \frac{c_0}{C_1/T_0}\ln\frac{T}{T_0} - \frac{1}{2}\{[(1+p^2)\lambda^2 + p^{-2}\lambda^{-4} - 3] + \alpha[(1+p^{-2})\lambda^{-2} + p^2\lambda^4 - 3]\}. \tag{28}$$

The thermodynamic performance of dielectric elastomer at different temperatures under the unequal-biaxial stretch condition is described in figures 5–7. Figures 5–7 show the relationship among the stretch and the nominal electric field, the nominal electric displacement and the nominal electric field, the temperature and the entropy of a dielectric elastomer with different values of the material constant ratio α, the ratio between principal planar stretches p, the temperature T/T_0 and the nominal stress $\sigma^{\sim}/C_1$, respectively.

From figures 5 and 6, with the increase in T/T_0 and α, or the decrease in p and $\sigma^{\sim}/C_1$, the critical nominal electric field of a dielectric elastomer thermo-electro-mechanical system is improved. That is to say the system becomes more stable. For figure 5, with the increase in stretch, the nominal electric field increases. When the critical nominal electric field is reached, the nominal electric field decreases and approaches a constant value. Clearly, the critical stretch of a dielectric

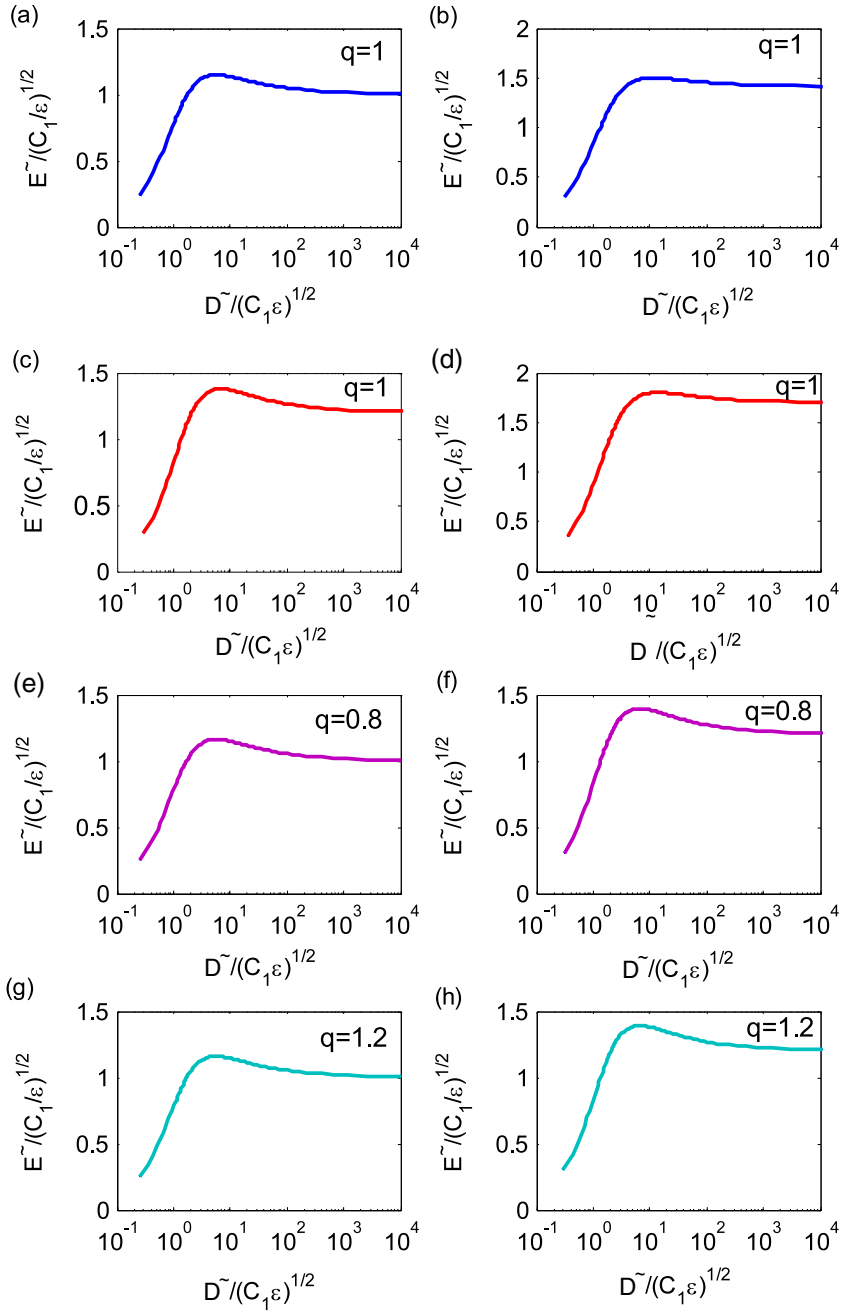

Figure 9. The relationship between the nominal electric field and nominal electric displacement of the thermo-electro-mechanical coupling system of dielectric elastomers under specialized load conditions, namely $\lambda_3 = \lambda$, $\lambda_1 = q\lambda^{-1/2}$ and $\lambda_2 = \lambda^{-1/2}/q$. (a) $T = T_0$, $\alpha = 1$, $q = 1$, (b) $T = T_0$, $\alpha = 2$, $q = 1$, (c) $T = 1.44T_0$, $\alpha = 1$, $q = 1$, (d) $T = 1.44T_0$, $\alpha = 2$, $q = 1$, (e) $T = T_0$, $\alpha = 1$, $q = 0.8$, (f) $T = 1.44T_0$, $\alpha = 1$, $q = 0.8$, (g) $T = T_0$, $\alpha = 1$, $q = 1.2$, (h) $T = 1.44T_0$, $\alpha = 1$, $q = 1.2$.

elastomer increases along with the increase in the temperature. However, with the increase in p and α, the critical stretch decreases. These results coincide with a recent conclusion on the instability of dielectric elastomer when unequal-biaxial stretched [41].

The critical nominal electric field and critical stretch are listed as follows. When $T/T_0 = 1$, $\alpha = p = 1$ and $\sigma^{\sim}/C_1 = 0$, the critical nominal electric field is $1.15E^{\sim}/\sqrt{C_1/\varepsilon}$, the corresponding critical stretch is 1.47. Let $T/T_0 = 1.44$, the critical nominal electric field and the corresponding critical stretch are $1.38E^{\sim}/\sqrt{C_1/\varepsilon}$ and 1.49, respectively. When $p = 1.2$, the critical nominal electric field and the critical stretch are $1.13E^{\sim}/\sqrt{C_1/\varepsilon}$ and 1.41, respectively.

In figure 7, the ratio between principal planar stretch values are 0.8 and 1.2, respectively. Evidently, along with the increase in temperature, the entropy of a dielectric elastomer thermo-electro-mechanical coupling system increases. At a specified temperature, the system entropy decreases with the increase in stretch. Along with the materials constant ratio α increases and the entropy decreases.

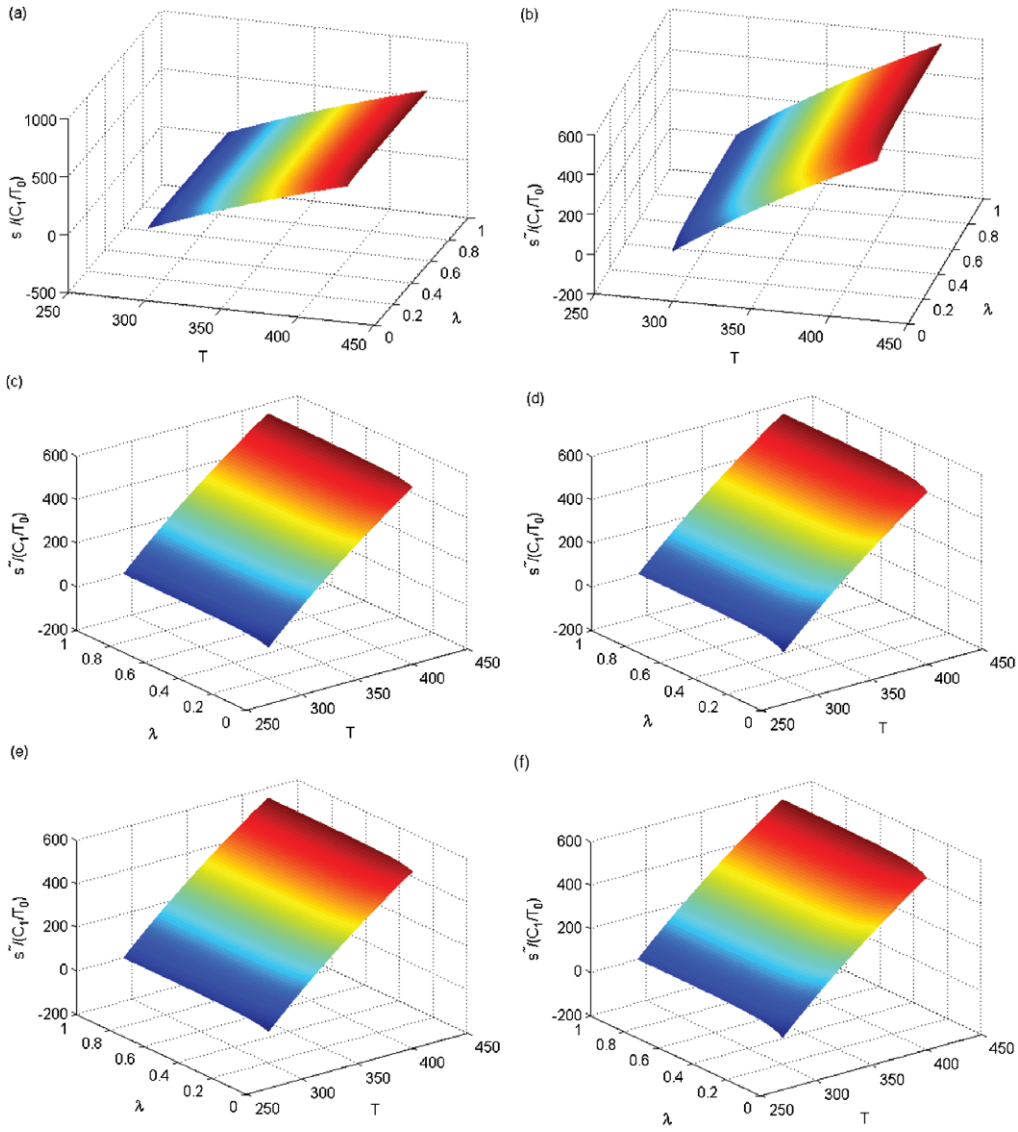

Figure 10. Entropy of the thermo-electro-mechanical coupling system of dielectric elastomers at different temperatures and stretches under specialized load conditions, namely $\lambda_3 = \lambda$, $\lambda_1 = q\lambda^{-1/2}$ and $\lambda_2 = \lambda^{-1/2}/q$. (a) $\alpha = 1$, $q = 0.8$, (b) $\alpha = 2$, $q = 0.8$, (c) $\alpha = 1$, $q = 1$, (d) $\alpha = 2$, $q = 1$, (e) $\alpha = 1$, $q = 1.2$, (f) $\alpha = 2$, $q = 1.2$.

4.3. Effect of thickness direction stretch on thermo-electro-mechanical instability

We next focus on the effect of thickness direction stretch on the thermo-electro-mechanical instability of dielectric elastomers. Supposing $\lambda_3 = \lambda$, if the boundary of dielectric elastomer membrane is free, consider the equibiaxial planar stretch case, $\lambda_1 = \lambda_2 = \lambda^{-1/2}$. Consider the unequal-biaxial planar stretch case, $\lambda_1 = q\lambda^{-1/2}$ and $\lambda_2 = \lambda^{-1/2}/q$, according to equations (11)–(13), the free energy of a thermodynamic system can be given by

$$W(\lambda, T, D^{\sim}) = \frac{T}{2T_0}\{C_1[(q^2+q^{-2})\lambda^{-1}+\lambda^2-3] + C_2[(q^2+q^{-2})\lambda+\lambda^{-2}-3]\} + c_0\left[(T-T_0) - T\ln\frac{T}{T_0}\right] + \frac{D^{\sim 2}}{2\varepsilon}\lambda^2. \tag{29}$$

Applying a similar method of investigation, we express the non-dimensional nominal electric displacement, the non-dimensional nominal electric field and the non-dimensional nominal entropy as:

$$\frac{D^{\sim}}{\sqrt{C_1\varepsilon}} = \left\{\frac{T}{T_0}\left\{\left[\frac{(q^2+q^{-2})}{2}\lambda^{-3}-1\right] + \alpha\left[\lambda^{-4} - \frac{(q^2+q^{-2})}{2}\lambda^{-1}\right]\right\} + \frac{\sigma^{\sim}}{C_1}\lambda^{-1}\right\}^{1/2} \tag{30}$$

$$\frac{E^{\sim}}{\sqrt{C_1/\varepsilon}} = \left\{\frac{T}{T_0}\left\{\left[\frac{(q^2+q^{-2})}{2}\lambda-\lambda^4\right] + \alpha\left[1 - \frac{(q^2+q^{-2})}{2}\lambda^3\right]\right\} + \frac{\sigma^{\sim}}{C_1}\lambda^3\right\}^{1/2} \tag{31}$$

$$\frac{s^{\sim}}{C_1/T_0} = \frac{c_0}{C_1/T_0}\ln\frac{T}{T_0} - \frac{1}{2}\{[(q^2+q^{-2})\lambda^{-1}+\lambda^2-3] + \alpha[(q^2+q^{-2})\lambda+\lambda^{-2}-3]\}. \tag{32}$$

Under different temperatures and considering the free or constrained boundary condition, figures 8–10 describe the relationship among thermodynamics parameters of dielectric elastomers in the thickness stretch direction, including the nominal electric field and the stretch, the nominal electric field and the nominal electric displacement, the entropy and the temperature. Here, we only consider the stress free state, namely $\frac{\sigma}{C_1} = 0$.

Figure 8 plots the voltage-induced deformation of dielectric elastomer under different temperatures. In each case, with the decrease of stretch from 1 to 0, the voltage increases to the critical value firstly, and then decreases to a constant value. This variation trend is coincident with the former conclusion, which results from the consideration of stretch in the thickness direction.

Figure 9 illustrates the voltage-induced charge of dielectric elastomers under different temperatures. In each curve, with the increase of charge, the voltage increases to the critical value firstly, and then declines to a constant level. Notice that in the current work we do not invoke the polarization saturation of dielectric elastomer for it falls out of the range of this temperature or voltage load [31].

Figure 10 shows the variation trend of dielectric elastomer entropy under different temperatures and stretches. With the increase of temperature and stretch, the entropy of the dielectric elastomer thermo-electro-mechanical coupling system increases. This variation trend of the entropy is coincident with the former conclusion.

5. Conclusions

In this paper, the equilibrium equations to describe the thermodynamics performance of dielectric elastomer were established by considering different temperatures. Based on these equations, the systemic thermodynamical instability was studied. The effects of the equal-biaxial stretch, the unequal-biaxial stretch and the thickness elongation on the thermo-electro-mechanical instability of dielectric elastomers are investigated. The critical nominal electric field and the critical stretch of the system under different material constant ratios and various temperatures are obtained. The results indicate that for the dielectric elastomer, the critical nominal electric field and the critical stretch increases with the increase in temperature and the system received an enhancement in its stabilization. However, along with the increase in the ratio between principal planar stretches and the unequal-biaxial stretch, the critical nominal electric field decreases and thermo-electro-mechanical stability is reduced. The results provide guidance in seeking for dielectric elastomer-based transducers with an improved performance.

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