Springer

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Journal of Mechanical Science and Technology 29 (1) (2015) 109~114
www.springerlink.com/content/1738-494x
DOI 10.1007/s12206-014-1217-9

Electric field induced variation of temperature and entropy in dielectric elastomers †

Liwu Liu[1,*], Jinrong Li[2], Yanju Liu[1], Jinsong Leng[2], Junqing Zhao[1] and Jianwen Zhao[3]

[1]*Department of Astronautical Science and Mechanics, Harbin Institute of Technology, No. 92 West Dazhi Street, Harbin, China*
[2]*Centre for Composite Materials, Science Park of Harbin Institute of Technology, No. 2 YiKuang Street, Harbin, China*
[3]*State Key Laboratory of Robotics and System, Harbin Institute of Technology at Weihai, Wenhua Road (W), Weihai, China*

(Manuscript Received June 11, 2014; Revised July 11, 2014; Accepted September 19, 2014)

Abstract

Dielectric elastomer is a kind of typical electro-active polymer material. Under external electric field it can produce large electrostriction deformation and possesses the advantages of high elastic energy density, super short response time, high efficiency, and so on. It is widely used in the artificial muscles, facial expressions, actuators, energy harvesters, sensors, robots and Braille display devices, and also shows huge application potential in the aerospace and intelligent bionic areas. We built the free energy of the dielectric elastomer electrical-mechanical coupling system and investigated its constitutive relation and stability behavior. Then we calculated the elastomer's critical deformation suffering from the voltage. If electrical breakdown, electromechanical instability and snap-through instability can be avoided, the large electrostriction deformation can induce adiabatic temperature change and isothermal entropy change of the dielectric elastomer. We used the entropy-temperature or electric displacement-electric field plane to describe the temperature change and entropy change of dielectric elastomer undergoing large electrostriction deformation. With the influence of temperature, we developed a temperature and deformation coupling thermodynamical free energy model to calculate the electric field induced variation of temperature and entropy in dielectric elastomers. The results should offer great help in guiding the design and fabrication of excellent actuators featuring soft dielectric elastomers.

Keywords: Dielectric elastomers; Electrostriction deformation; Entropy change; Soft; Temperature change

1. Introduction

Large deformation will be generated when an electric field is applied on thin dielectric elastomer film [1-10]. Due to their excellent properties, such as high elastic energy density, quick response, low cost, light weight, high efficiency, and easy to be processed, dielectric elastomers show great potential applications in the area of smart bionics, medical devices, aeronautics and astronautics. Many devices have been designed based on dielectric elastomers such as actuators, sensors, tactile display and energy harvesters etc. [11-25]. The main failure modes [20, 23] of dielectric elastomers include electric breakdown, material rupture, loss of tension, electromechanical instability [9-13, 15, 16, 19, 21], which will badly hinder the wide application of dielectric elastomers. Hence, a well established theoretical study on the failure mechanism of dielectric elastomers is a critical issue. Recently, the nonlinear mechanical performance [8, 28, 29], electromechanical stability [9-13, 15, 16, 19, 21], dynamical performance [7, 27, 28] and failure of application devices [26, 29] of dielectric elastomers are the hotspots of the theoretical research on electroactive soft material.

In fact, when an electrical field is applied to dielectric elastomer, it will produce a large deformation. From the stage without deformation to the large deformation, the interior dipoles have an arrangement from unordered state to ordered state, and further lead to the isothermal entropy change and adiabatic temperature change of the dielectric elastomer [30, 31].

When an electrical field is applied to a polar dielectric material, the material's polarization will change from the original dipole-disordered state to ordered state, accompanied by an entropy change and temperature change under isothermal or adiabatic condition [32, 33].

In this paper, we proposed the expression of dielectric constant relying on temperature and stretch, established the free energy function of dielectric elastomer's thermodynamic system under an adiabatic process, and deduced the constitutive relation. These simulation results should offer great help in guiding the design and fabrication of excellent actuators featuring dielectric elastomers.

*Corresponding author. Tel.: +86 451 86403269
E-mail address: liuliwu_006@163.com, yj_liu@hit.edu.cn, lengjs@hit.edu.cn
† This paper was presented at the FEOFS 2013, Jeju, Korea, June 9-13, 2013.
Recommended by Guest Editor Jung-Il Song

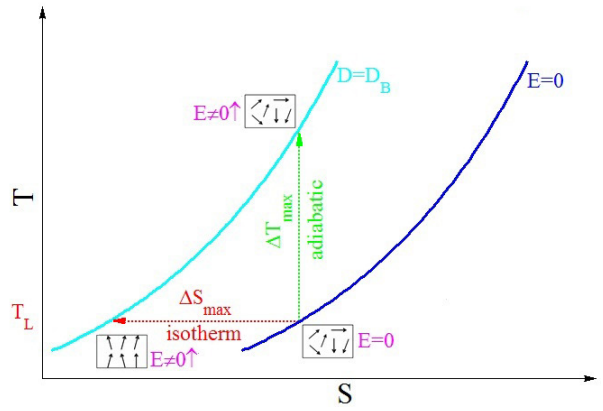

Fig. 1. The change in temperature and entropy of polar dielectrics in isothermal process and adiabatic process.

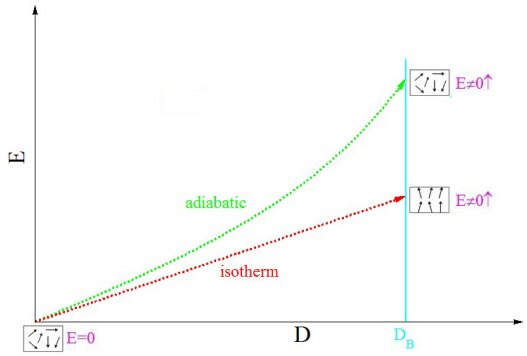

Fig. 2. The relationship between electric field and electric displacement of polar dielectrics in isothermal process and adiabatic process.

2. Fundamental theory

2.1 Preview

Fig. 1 shows the relationship of entropy and temperature, which is a group of work conjugate parameters of polar dielectric subjected to voltage. At the original temperature, without electrical field, the dipole of polar dielectric is chaotic, at this time; the entropy of the dielectric is relatively high. When subjected to voltage, the polar dielectric can reach its critical breakdown charge D_B along two kinds of routes (here we chose breakdown charge D_B as the critical value, we can also choose breakdown electric field): the isothermal process and the adiabatic process. In the isothermal process, within the increase of electric field, for the dipole orientation arrangement, the entropy of dielectric decreases. When reaching the critical breakdown charge D_B, the entropy gets to the minimum, the entropy change $\Delta S_{\max}$ is largest at the time. In the adiabatic process, within the increase of electric field, the temperature of dielectric increases too; when reaching the critical breakdown charge D_B, the temperature reaches the maximum, and the temperature change $\Delta T_{\max}$ is largest at the time. From Fig. 1 we can see, the influencing factors of polar dielectric entropy including electric field and temperature. When the temperature changes a little, we can ignore its influence. While analyzing polar dielectric within large electrocaloric effect, we need to consider the influence of temperature. Therefore, we considered the influence of temperature when building free energy. In Fig. 1, T_L is the initial temperature, $E=0$ is the initial field, D_B is the breakdown charge.

In Fig. 2, we use another group of work conjugate parameters, the electric field and the electric displacement, to describe two kinds of physical process: the isothermal process and the adiabatic process. Subjected to a voltage, the electric displacement would increase from 0 to the breakdown electric charge D_B, the constant gradient line represents the isothermal process, and the crescent gradient line represents the adiabatic process. While the gradient of the line depends on temperature, the higher the temperature reaches, the larger the gradient is. Here, we assume that D_B is a straight line, but in fact, for the influence of temperature D_B is a curve, for the influence is relatively low; we ignored the temperature's influence in the figure.

2.2 Constitutive model of dielectric elastomer

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.

$$W\left(\lambda_1,\lambda_2,T,D^{\sim}\right)=W_s\left(\lambda_1,\lambda_2,T\right)+W_e\left(\lambda_1,\lambda_2,T,D^{\sim}\right) \tag{1}$$

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

Considering the influence of temperature, a proposed thermoelastic strain energy function is

$$W_s\left(\lambda_1,\lambda_2,T\right)=\frac{1}{2}NkT\begin{bmatrix}C_1\left(\lambda_1^2+\lambda_2^2+\lambda_1^{-2}\lambda_2^{-2}-3\right)\\+C_2\left(\lambda_1^{-2}+\lambda_2^{-2}+\lambda_1^2\lambda_2^2-3\right)\end{bmatrix}+\xi\left(T\right). \tag{2}$$

Eq. (2) describes the coupling influence of temperature and stretch of the thermodynamical system. The first item in Eq. (2) is the Mooney-Rivlin elastic 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, C_1 and C_2 are the dimensionless material parameters, which are related to the material of dielectric elastomer and its application devices' structure; what's more, $C_1+C_2=1$, which is determined by experimental data.

The second term of Eq. (2) $\xi\left(T\right)$ is called the thermal contribution. It donates the thermal contribution of tempera-

ture to the free energy of the thermodynamic system.

We can describe the derivation of the thermal contribution simply. The thermal contribution can be expressed as $\xi(T)=Q(T)-TS$, where $Q(T)$ is the internal energy. As the temperature rises, internal energy $Q(T)=c_0(T-T_0)$. 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. And the expression of thermal contribution is widely applicable to the thermodynamic systems with great temperature changes.

In Jean-Mistral's experiment, it is indicated that the dielectric constant of dielectric elastomer is related to the temperature and stretch. 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, according to the research of Jean-Mistral and the Debye equation function, we propose the expression of dielectric constant $\varepsilon(T,\lambda_1,\lambda_2)$ as follows:

$$\varepsilon(T,\lambda_1,\lambda_2)=\varepsilon_r+\frac{\alpha}{T}+\beta\lambda_1\lambda_2 . \tag{3}$$

The dielectric constant is the inverse proportion function of the temperature; due to the thermal force field the dipoles reorientate, which is the molecular origin of the macroscopic dielectric permittivity, is strongly restricted. In Eq. (3), $\alpha=\frac{M\eta^2}{3\kappa\varepsilon_0}$, M is the dipole density of a dielectric elastomer, η 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.

2.3 Electric field induced variation of temperature and entropy

The elastomer is taken to be network of long polymers obeying Gaussian statistics, so that the elastic behavior of the elastomer is neo-Hookean type. 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. Consider the electrostriction of dielectric elastomers, the dielectric constant is a function of the stretches, the dielectric energy per unit volume is $\frac{\tilde{D}^2}{2\varepsilon(\lambda_1,\lambda_2)}\lambda_1^{-2}\lambda_2^{-2}$. Hence, the electric field energy density function of incompressible dielectric elastomers can be written as follows:

$$W_e\left(\lambda_1,\lambda_2,T,\tilde{D}\right)=\frac{\tilde{D}^2}{2\left[\varepsilon_r+\frac{\alpha}{T}+\beta\lambda_1\lambda_2\right]}\lambda_1^{-2}\lambda_2^{-2} . \tag{4}$$

During the fabrication process of actuators, the dielectric elastomer would be subject to pre-stretch, most commonly, unequal biaxial stretch. Therefore, in the following study, to simplify the calculation, we set $\lambda_2=p\lambda_1=p\lambda$, where 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; this can be treated as a special case of $p=1$.

Considering Eqs. (1)-(4), 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 following.

The dimensionless nominal electric displacement, the nominal electric field and the entropy of dielectric elastomer can be obtained as follows:

$$\frac{\tilde{D}}{\sqrt{NkT_0\varepsilon_r}}=\sqrt{\frac{\left[1+\frac{\alpha/T+p\beta\lambda^2}{\varepsilon_r}\right]}{[2p^{-2}+\frac{p^{-1}\beta\lambda^2}{\left[\varepsilon_r+\alpha/T+p\beta\lambda^2\right]}]}\left\{\frac{T}{T_0}C_1[(p^2+1)\lambda^6-2p^{-2}]+\frac{T}{T_0}C_2[-(p^{-2}+1)\lambda^2+2p^2\lambda^8]-\frac{s\lambda^5}{NkT_0}\right\}} \tag{5}$$

$$\frac{\tilde{E}}{\sqrt{NkT_0/\varepsilon_r}}=\sqrt{\frac{1}{[2p^{-2}+\frac{p^{-1}\beta\lambda^2}{\left[\varepsilon_r+\alpha/T+p\beta\lambda^2\right]}]\left[1+\frac{\alpha/T+p\beta\lambda^2}{\varepsilon_r}\right]}\left\{\frac{T}{T_0}C_1[(p^{-2}+p^{-4})\lambda^{-2}-2p^{-6}\lambda^{-8}]+\frac{T}{T_0}C_2[-(p^{-6}+p^{-4})\lambda^{-6}+2p^{-2}]-\frac{sp^{-4}\lambda^{-3}}{NkT_0}\right\}} \tag{6}$$

$$\frac{S}{Nk}=\frac{c_0}{Nk}\ln\frac{T}{T_0}-\frac{1}{2}\{C_1[(p^2+1)\lambda^2+p^{-2}\lambda^{-4}-3]+C_2[(p^{-2}+1)\lambda^{-2}+p^2\lambda^4-3]\}-\frac{\alpha\tilde{D}^{-2}p^{-2}\lambda^{-4}}{2T^2Nk\left[\varepsilon_r+\frac{\alpha}{T}+\beta p\lambda^2\right]^2} . \tag{7}$$

Then by substituting Eq. (5) into Eq. (7), the entropy can be expressed as a function of stretch and temperature:

$$\frac{S}{Nk}=\frac{c_0}{Nk}\ln\frac{T}{T_0}-\frac{1}{2}\left\{C_1[(p^2+1)\lambda^2+p^{-2}\lambda^{-4}-3]+C_2[(p^{-2}+1)\lambda^{-2}+p^2\lambda^4-3]\right\}-\frac{\alpha p^{-2}\{TC_1[(p^2+1))\lambda^2-2p^{-2}\lambda^{-4})]+TC_2[-(p^{-2}+1)\lambda^{-2}+2p^2\lambda^4)]-\frac{s\lambda}{Nk}\}}{2T^2\left[2p^{-2}\varepsilon_r+2p^{-2}\alpha/T+3p^{-1}\beta\lambda^2\right]}. \tag{8}$$

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. Therefore, the $E_{EMI}^{\sim}$ value is the most important issue to determine the energy allowable range.

Considering biaxial stretch that the principal planar stretch ratio is set as p, i.e., $\lambda_2=p\lambda_1=p\lambda$, according to Eq. (6), at a constant s, when

$$\Lambda=\frac{s}{NkT_0}=\frac{T}{T_0}\frac{\beta\{12C_1[(p^2+1)\lambda-5p^{-2}\lambda^{-5}]-12C_2[2(p^{-2}+1)\lambda^{-3}-p^2\lambda^3]\}-(\varepsilon_r+\alpha/T)\{C_1[32p^{-3}\lambda^{-7}-4(p+p^{-1})\lambda^{-1}]+6C_2(p^{-3}+p^{-1})\lambda^{-5}\}}{6p^{-1}\lambda^{-2}\varepsilon_r+6p^{-1}\lambda^{-2}(\alpha/T)+15\beta},$$

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. (5) and (6), the formulations of the dimensionless nominal electric displacement nominal electric field, the nominal displacement and the entropy can be induced, respectively.

$$\frac{D^{\sim}}{\sqrt{NkT_0\varepsilon_r}}=\sqrt{\frac{\left[1+\frac{\alpha/T+p\beta\lambda^2}{\varepsilon_r}\right]}{[2p^{-2}+\frac{p^{-1}\beta\lambda^2}{\left[\varepsilon_r+\alpha/T+p\beta\lambda^2\right]}]}\{\frac{T}{T_0}C_1[(p^2+1)\lambda^6-2p^{-2}]+\frac{T}{T_0}C_2[-(p^{-2}+1)\lambda^2+2p^2\lambda^8]-\Lambda\lambda^5\}} \tag{9}$$

$$\frac{E^{\sim}}{\sqrt{NkT_0/\varepsilon_r}}=\sqrt{\frac{1}{[2p^{-2}+\frac{p^{-1}\beta\lambda^2}{\left[\varepsilon_r+\alpha/T+p\beta\lambda^2\right]}]\left[1+\frac{\alpha/T+p\beta\lambda^2}{\varepsilon_r}\right]}\{\frac{T}{T_0}C_1[(p^{-2}+p^{-4})\lambda^{-2}-2p^{-6}\lambda^{-8}]+\frac{T}{T_0}C_2[-(p^{-6}+p^{-4})\lambda^{-6}+2p^{-2}]-\Lambda p^{-4}\lambda^{-3}\}} \tag{10}$$

$$\frac{S}{Nk}=\frac{c_0}{Nk}\ln\frac{T}{T_0}-\frac{1}{2}\{C_1[(p^2+1)\lambda^2+p^{-2}\lambda^{-4}-3]+C_2[(p^{-2}+1)\lambda^{-2}+p^2\lambda^4-3]\}$$
$$-\frac{\alpha p^{-2}\{TC_1[(p^2+1))\lambda^2-2p^{-2}\lambda^{-4})]+TC_2[-(p^{-2}+1)\lambda^{-2}+2p^2\lambda^4)]-\Lambda T_0\lambda\}}{2T^2\left[2p^{-2}\varepsilon_r+2p^{-2}\alpha/T+3p^{-1}\beta\lambda^2\right]}. \tag{11}$$

When the electric field 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 $E_{EMI}^{\sim}$. 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.

From the experiment result of Jean-Mistral [34] and Wissler [35], we get $\varepsilon_r=3.1834F/m$, $C_1=0.5220$, $C_2=0.4780$, $\alpha=645.4224F\cdot K/m$, $\beta=-0.0533F/m$. Other typical parameters are $C_0=1.7\times10^3J/m^2\cdot K$, $NK=3.66J/m^2\cdot K$. Then when p=0.8, 1, 1.2 respectively, the change in temperature and entropy under $E_{EMI}^{\sim}$ and without electric field is shown in Fig. 3. To obtain a maximum temperature and entropy change, dielectric elastomer application cooling devices with small p should be selected.

3. Conclusions

We first proposed the expression of dielectric constant relying on temperature and stretch, established the free energy function of dielectric elastomer's thermodynamic system under an adiabatic process, and deduced the constitutive relation. Under the condition of equal biaxial and unequal biaxial, the thermo-electromechanical stability of dielectric elastomer is studied. As a result, with the increase of temperature and the 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 variable temperature and electric field is studied. The allowable energy range of dielectric elastomer under the condition of equal and unequal biaxial stretch is

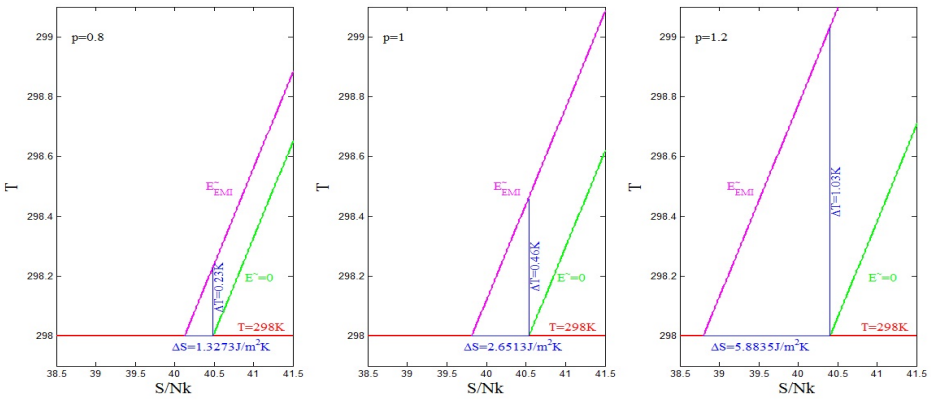

Fig. 3. The change in temperature and entropy of polar dielectrics under $E_{EMI}^{\sim}$ and without electric field.

described. Finally, the electric-induced temperature and entropy variation of dielectric elastomer is evaluated. Numerical results will offer great help in guiding the design and fabrication of dielectric elastomer application devices, such as dielectric elastomer actuators working in variable ambient temperature.

Acknowledgment

This work is supported by the National Natural Science Foundation of China (Grant No.11225211, No.11272106, No.11102052), China Postdoctoral Science Foundation (Grant No. 2012M520032) Heilongjiang Postdoctoral Fund (Grant No. LBH-Z12091) and the Fundamental Research Funds for the Central Universities (Grant No.HIT.NSRIF. 2013030).

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Liwu Liu is an associate professor in Department of Astronautical Science and Mechanics, Harbin Institute of Technology. He obtained his Ph.D. in Solid Mechanics from Harbin Institute of Technology in 2012. As a visiting scholar, he cooperated with Professor Zhigang Suo's group at Harvard University on the study of dielectric elastomer actuators for one year.

Yanju Liu is a professor in the Department of Aerospace Science and Mechanics at Harbin Institute of Technology. She obtained her Ph.D. in the field of materials from the Centre for Composite Materials and Structures of HIT. She has been involved in the research of smart materials and structures, including electro and magneto rheological fluids and their applications, electro-active polymers and their applications, and shape-memory polymers and their nanocomposites.

Jinsong Leng is the Cheung Kong Chair Professor at the Center for Composite Materials and Structures at Harbin Institute of Technology, China. His research interests include smart materials and structures, fiber-optic sensors, shape-memory polymers, electro-active polymers, structural health monitoring, morphing aircraft, deployable structures, nanocomposites, and multifunctional materials.