



An improved visco-hyperelastic model charactering the electromechanical behaviour of dielectric polymers

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ARTICLE INFO

Article history:

Received 12 January 2018

Received in revised form

21 June 2018

Accepted 23 June 2018

Available online 3 July 2018

Keywords:

Dielectric polymer

Visco-hyperelastic model

Electromechanical behaviour

ABSTRACT

Dielectric polymers can achieve large, reliable deformation in response to an external electric field and have attracted significant interest as actuators and transducers. The electromechanical performance depends on the interaction between the visco-hyperelastic behaviour of materials and the Maxwell stress caused by the electric field. However, the mechanisms of complex structures, such as a surface-treated and filler-reinforced space system, are still not completely understood. A mechanical model for evaluating the surface effect and electromechanical performance is first proposed on the basis of visco-hyperelastic theory and statistical mechanics. Then, several factors affecting the geometry and material properties of complex structures are quantitatively investigated. The model will contribute to the development of dielectric polymers because its theoretical predictions are in agreement with existing experimental data.

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1. Introduction

Dielectric elastomers have been increasingly studied in recent years for possible applications in the so-called electro-active polymer actuators, sensors and generators [1,2]. A dielectric elastomer actuator (DEA) consists of a thin membrane of polymer sandwiched between two compliant electrodes. DEAs shrink in thickness and expand in area when a voltage is applied across the electrodes [3]. The strain of DEA induced by the external voltage is limited by multiple modes of film failure, such as material rupture and electrical breakdown [4,5].

Studies of basic mechanical properties show that time have a negligible influence on the stress-strain relationship, such as stress relaxation and creep [6,7]. However, the electromechanical properties are more complicated because they are affected by the combination of an electric field (influenced by frequency and voltage) and a mechanical field (influenced by stiffness, time and temperature) [8]. Pre-stretching has a great influence on stiffness and electromechanical behaviour of DEAs [9–11]. Several advanced hyperelastic models show reasonable agreement with the experimental results obtained for equal biaxial pre-stretch and unequal biaxial pre-stretch experiment, such as the Yeoh, Ogden and

Arruda-Boyce strain energy potential [12–15]. These models are functions of stretch ratio and on the basis of isotropy of materials, generally. These models show poor predictive capabilities of DEAs under pure shear pre-stretch conditions when using the same parameters related to equal biaxial pre-stretch or unequal biaxial pre-stretch conditions. This lower actuator performance in pure shear pre-stretch conditions may be explained by the deviation of the film external boundary, which will pull the edges inward, thereby decreasing the effective area and increasing the real thickness [16]. The difference between ideal pure shear and a real shear condition is discussed by considering different dimensions of origin under several pre-stretch levels in this work.

Beyond the effect of external surface mentioned above, internal surfaces also reveal non-negligible effect on the electromechanical behaviour. In recent years, conductive fillers including carbon nanotube, graphite, metal particles and conductive oxides are added to the polymer matrix to develop high-performance composites (high dielectric permittivity and low loss tangent) [17–19]. Among those particles, CaCu₃Ti₄O₁₂ (CCTO) shows a high dielectric permittivity which is almost not changing in a large range of temperature and frequency, larger than 10000 [20]. This reinforcement can be explained by two main reasons: the physical properties of filler; the redistribution of polymer chain in the interphase due to chemical action [21,22].

The macro mechanical properties have found originate from the microstructure of the polymers including spatial distribution,

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interfacial reaction and chemical modification [23–26]. Based on an analysis of the micro structure, several multi scale models have been proposed, such as the 3 chain model, 8 chain model and tetrahedral model [27–29]. Beyond these, the hyperelastic models aimed to capture the limiting stretch of polymer chain are always used to evaluating mechanical behaviour of soft polymer [30–32]. Statistical mechanics method has been widely used in these models to charactering the distribution of polymer chains. However, the effect of surface is seldom investigated.

In this paper, the basic concept of polymer chain is defined: a chain system with alternating chain length or chain number is used to connect the distribution law and the geometry of the material. Then, a statistics mechanical motivated constitutive model is constructed, presented and used, and the numerical simulation results are compared with some experiments in the existing literature.

2. Improved visco-hyperelastic model

2.1. Viscoelastic theory and electromagnetic theory

2.1.1. Strain energy potential and viscoelasticity

The mechanical property of soft materials (large strain elastic; hyperelastic) can be characterized using strain energy potential W . A classic model presented by Yeoh is chosen on account of its applicability to describe the electromechanical properties comparing with the Arruda-Boyce and Ogden models [13,14]. The Yeoh model for the strain energy potential uses three parameters is described as follows [12]:

$$W = C_1(I_1 - 3) + C_2(I_1 - 3)^2 + C_3(I_1 - 3)^3 \quad (1)$$

where I_1 is the first invariant of the left Cauchy-Green deformation tensor which has relation with the stretch ratio in three directions, $I_1 = \lambda_x^2 + \lambda_y^2 + \lambda_z^2$. The nominal stress is given by the derivative of the strain energy potential:

$$\bar{\sigma}_i = \frac{\partial W}{\partial \lambda_i} - \frac{1}{\lambda_i} p_{hy}, \quad i = x, y \text{ and } z \quad (2)$$

where $\bar{\sigma} = \sigma/\lambda$ and p_{hy} are the nominal stress and hydrostatic pressure, respectively.

As has been demonstrated widely, most polymers show viscoelastic behaviours such as stress relaxation and creep. A time function describing the time dependence of the mechanical response of the material is used combining the strain energy potential. The time function is given as [14].

$$g(t) = 1 - \sum_{k=1}^n g_k \left(1 - \exp\left(-\frac{t}{t_k}\right) \right) \quad (3)$$

where g_k and t_k are parameters that characterize the relaxation behaviour.

2.1.2. Basic equations of electromagnetism

When a test charge q is introduced in an electric field \mathbf{E} , the charge has the potential to move along the direction of \mathbf{E} , which indicates that a force is applied on q :

$$\mathbf{f} = q\mathbf{E} \quad (4)$$

where \mathbf{f} is the electrostatic force. The relationship between the charge on \mathbf{X} and electric field \mathbf{E} can be expressed as

$$\frac{\partial E_i}{\partial X_j} = \frac{\partial E_j}{\partial X_i}, \quad \frac{\partial E_i}{\partial X_i} = \frac{q}{\varepsilon \varepsilon_0} \quad (5)$$

where ε_0 is the vacuum permittivity, and ε is the dielectric constant for a given material. Inserting Eq. (5) into Eq. (4), we can obtain

$$f_i = \frac{\partial}{\partial X_j} \left(\varepsilon \varepsilon_0 E_i E_j - \frac{\varepsilon \varepsilon_0}{2} E_k E_k \delta_{ij} \right) \quad (6)$$

Eq. (6) is derived from equilibrium equation in continuum mechanics. Generally,

$$\sigma_{ij}^{el} = \varepsilon \varepsilon_0 E_i E_j - \frac{\varepsilon \varepsilon_0}{2} E_k E_k \delta_{ij} \quad (7)$$

is known as the Maxwell stress. When we consider a voltage U applied on the surface of a rectangular DEA, Maxwell stress σ_{ij}^{el} is determined by the applied voltage U and can be expressed as follows:

$$\sigma_{zz}^{el} = \varepsilon \varepsilon_0 E_z^2 = \varepsilon \varepsilon_0 \left(\frac{U}{l_z} \right)^2 = \varepsilon \varepsilon_0 \lambda_x^2 \lambda_y^2 \left(\frac{U}{L_z} \right)^2 \quad (8)$$

where $l_z = \lambda_z L_z$ is the thickness of the elastomer membrane after deformation, U is the applied voltage. In view of the work in Ref. [9], dielectric constant is affected by the stretch ratio in x-y plane: λ_x and λ_y . The correct expression is shown as

$$\varepsilon(\lambda_x, \lambda_y) = 4.7 \left(1 + a(\lambda_x + \lambda_y - 2) + b(\lambda_x + \lambda_y - 2)^2 + c(\lambda_x + \lambda_y - 2)^3 \right) \quad (9)$$

where a , b and c are the coefficients.

2.2. Kelvin-Voigt model

To exhibit the visco-hyperelastic behaviour, we start with Kelvin-Voigt model [33,34] which combining a parallel spring-damper element (E_{par} and η_{par}) and a spring element (E_{ser}), as shown in Fig. 1a. The cube (side length l) composed of Kelvin-Voigt model can be regard as a finite element, as shown in Fig. 1b. Generally, the finite elements are assembled into a specific configuration $L_x \times L_y \times L_z$ in three directions, $i = x, y$ and z (Fig. 1c). The following assumptions are considered in the numerical simulation:

The materials are isotropic and homogeneous at the reference state: the side length l is negligible comparing with the real dimension.

The materials are incompressible during the deformation process: volume V is constant.

The materials are low weight, which means that the effect of gravity can be neglected considering the slow movements.

The mechanical constitutive behaviour of Kelvin-Voigt model is introduced according to [34], which means that the relation between stress and strain can be summarized as follows:

$$(E_{ser} + E_{par})\sigma + \eta_{par} \frac{d}{dt} \sigma = E_{ser} E_{par} \varepsilon + E_{ser} \eta_{par} \frac{d}{dt} \varepsilon \quad (10a)$$

$$\sum_{k=0}^m p_k \frac{d^k \sigma}{dt^k} = \sum_{k=0}^n q_k \frac{d^k \varepsilon}{dt^k} \quad (10b)$$

where p and q are the material parameters. Eq. (10) is the general expression for a standard Kelvin-Voigt model, and the modified

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