Modeling of a pre-stretched dielectric elastomer: A second law of thermodynamics-based approach

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Abstract. Research in smart materials, which have fluid-like properties is playing an increasingly dominant role in the fields of aerodynamics as well as in the fluid power systems. Dielectric elastomers are the smart materials, that demonstrate the deformation behavior similar to the smart solids as well as the fluids depending on the environmental conditions. This necessitates a smart deformation approach, which may analyze the deformation of the continua, accurately. In the present paper, we report an alternative modeling method for the deformation of a continua within the framework of the second law of thermodynamics. We adopt a unified continuum mechanics approach to model the deformation of a pre-stretched dielectric elastomer with an application of the electric field. An analytical electromechanical instability model is formulated for a pre-stretched dielectric elastomer through the standard Neo-Hookean, Mooney-Rivlin, and Gent types of energy density functions. A qualitative comparison between these energy density functions is also presented based on the electromechanical instability of a pre-stretched dielectric elastomer. It is shown based on the obtained results that Gent energy density function shows the instability phenomenon with the chain stiffening effect, and the pre-stretching effect may improve the electromechanical instability of the dielectric elastomers.

1. Introduction

The growing interest in human-friendly smart material devices for engineering as well as medical field applications, and due to customer-friendly products to enhance personal convenience, has led to the rapid development of soft dielectric elastomeric actuators [1, 2, 3]. The dielectric elastomers also have fluid-like properties, which is playing an increasingly dominant role in the fields of aerodynamics as well as in the fluid power systems. The soft dielectric elastomeric actuators are the devices that exhibit stretchable, flexible, and deformable behaviors with the application of an electrical, chemical, or thermal stimulus [4, 5].

In the literature, for a dielectric material, an electrostriction phenomenon results from the effect of deformation on permittivity [4, 5]. A lot of studies [1, 2, 3] on dielectric elastomers have used the model of ideal dielectric elastomer, where the permittivity is assumed to be independent of the deformation due to the liquid-like property of the elastomer. In addition, for an electro-elastic deformation of the dielectric elastomers, the Maxwell stress tensor were readily used mostly in the literature. However, in reality the permittivity can not be assumed to be independent of the deformation. Therefore, the Maxwell stress stress tensor may lead to an conceptual inaccuracy, especially in large deformation of the dielectric elastomers. For the details discussion on the issues related to the physical objectivity of Maxwell stress tensor,
we refer [6, 7], and references therein. Accordingly, we try to overcome this physical issue by introducing a concept of an amended form of energy density function, which may also incorporate the Maxwell stress contribution for the dielectric elastomeric materials. The net stress tensor may be given by a simple formula in terms of the amended energy density function, and the same does not require the notion of the Maxwell stress tensor within the material.

Now, we may conclude that there is existing a challenging task to our research community to understand and model the electro-elastic behavior of a smart material, accurately with an application of the electro-mechanical loading. Accordingly, we try to report an alternative modeling method for the deformation of a continua within the framework of the second law of thermodynamics. We adopt a unified continuum mechanics approach to model the deformation of a pre-stretched dielectric elastomer with an application of the electric field.

2. Second law of thermodynamics-based constitutive relationships for a dielectric elastomer

The constitutive relationships are the mathematical relations, which connects the components of the stresses with the corresponding deformations. For an incompressible isotropic electro-elastic material, the constitutive relationships may be formulated through an energy density function. In an isothermal condition, a free energy density function for a class of an incompressible isotropic electro-elastic material is defined as a function of the independent field variables $F$ and $E$ as follows

$$\varphi = \varphi(F, E),$$

(1)

wherein $F$, $E$ are the deformation gradient tensor and the electric field vector, respectively. We consider $F$ and $E^l = F^T E$ as the independent variables for an electro-elastic material, then the free energy function $\phi(F, E^l)$ may be defined in the Lagrangian form as follows

$$\phi(F, E^l) = \varphi(F, F^{-1} E^l).$$

(2)

Now, an amended form of energy density function $\Omega = \Omega(F, E^l)$, which incorporates the Maxwell stress contribution for an electro-elastic material may be defined as follows

$$\Omega(F, E^l) = \rho\varphi(F, E^l) - \frac{1}{2} \epsilon_0 E^l (b^{-1} E^l),$$

(3)

wherein $\epsilon_0$ represents the electric permittivity, and $b = FF^T$ represents the left Cauchy-green deformation tensor. This amended energy function $\Omega(F, E^l)$ represents the superposition of the available forms of the electrical energy and the interaction energy. The amended energy function $\Omega(F, E^l)$ also successfully overcomes the issue related to the physical interpretation of the Maxwell stress tensor in smart material for the large deformation [8]. For the details discussion on the issues related to the physical objectivity of Maxwell stress tensor, we refer [6, 7], and references therein.

Following the Clausius-Duhem inequality principle based on the second law of thermodynamics, we may formulate the thermodynamically consistent constitutive relations for an incompressible isotropic electro-elastic material. Assuming an isothermal condition, the dissipation inequality may be written in terms of $\Omega(F, E^l)$ as follows

$$\left( T - F \frac{\partial \Omega}{\partial F} \right) : \dot{F} - \left( D^l + \frac{\partial \Omega}{\partial E} \right) : \dot{E} \geq 0,$$

(4)

wherein $T$ represents the total stress tensor, and $D^l$ represents the electric displacement vector in the Lagrangian form.
Now, the set of constitutive relations for an incompressible isotropic electro-elastic material from the above equation (4) may be written as follows

\[
\mathbf{T} = -p\mathbf{I} + \mathbf{F} \frac{\partial \Omega}{\partial \mathbf{F}}, \quad \mathbf{D}^I = -\frac{\partial \Omega}{\partial \mathbf{E}^I},
\]

(5)

wherein \( p \) is the indeterminate hydrostatic pressure arising from the incompressibility constraint.

The amended energy density function \( \Omega \) for an incompressible isotropic electro-elastic material may also be formulated in the invariant form \( \Omega(I_1, I_2, \ldots, I_6) \). These invariants may be derived from the two tensors namely, the left Cauchy green deformation tensor \( \mathbf{b} \) and \( \mathbf{E}^I \otimes \mathbf{E}^I \) as follows

\[
I_1 = \text{tr} \mathbf{b}, \quad I_2 = \frac{1}{2}[(\text{tr} \mathbf{b})^2 - \text{tr}(\mathbf{b}^2)], \quad I_3 = \det \mathbf{b} = 1, \quad I_4 = [\mathbf{E}^I \otimes \mathbf{E}^I] : \mathbf{I},
\]

\[
I_5 = [\mathbf{E}^I \otimes \mathbf{E}^I] : \mathbf{b}^{-1}, \quad I_6 = [\mathbf{E}^I \otimes \mathbf{E}^I] : \mathbf{b}^{-2}.
\]

(6)

From the relations (3), (5), and the definitions of the invariants in (6), the explicit form of \( \mathbf{T}, \mathbf{D} \) may be written as follows

\[
\mathbf{T} = -p\mathbf{I} + 2\Omega_1 \mathbf{b} + 2\Omega_2[I_1 \mathbf{b} - \mathbf{b}^2] - 2\Omega_3 \mathbf{E} \otimes \mathbf{E} - 2\Omega_6[b^{-1}\mathbf{E} \otimes \mathbf{E} + \mathbf{E} \otimes b^{-1}\mathbf{E}],
\]

\[
\mathbf{D} = -2[\Omega_4 \mathbf{b} + \Omega_5 \mathbf{I} + \Omega_6 \mathbf{b}^{-1}]\mathbf{E},
\]

(7)

wherein the notation \( \Omega_i \) represents \( \Omega_i = \frac{\partial \Omega}{\partial I_i} \) for \( i = 1, 2, 3, \ldots, 6 \). The above relations (7) represent the constitutive relations based on the second law of thermodynamics for a class of an incompressible isotropic electro-elastic material.

3. Second law of thermodynamics-based electro-mechanical instability model of a pre-stretched dielectric elastomer

Let we define the coordinates system \( (X_1, X_2, X_3) \) and \( (x_1, x_2, x_3) \) for the reference configuration \( \beta_0 \) and the current configuration \( \beta \), respectively just after applying the pre-stretching effect from the undeformed initial shape. The compliant electrodes cover the top and bottom of the elastomeric plates as shown in the Figure 1. Initially, a dielectric elastomeric actuator has the dimensions \( (L_0, B_0, H_0) \) in the original configuration. Further, as the electrical voltage is switched on in \( X_3 \) direction, the material deforms and the dimensions change to \( (L, B, H) \) in the current configuration. The original configuration was free from any mechanical and electrical load, and the reference configuration is subjected to the equi-biaxial pre-stretching loads only. However, the current configuration is subjected to a pre-stretching mechanical load as well as an electrical load applied through any voltage source.
Figure 1: An electro-elastic deformation of a pre-stretched dielectric elastomer under an equi-biaxial deformation condition.

Let the deformation field is assumed to be a homogeneous deformation field with the material consideration as an incompressible and isotropic. Therefore, the stretch in the principal directions may be defined as follows

$$\lambda_1 = \frac{l}{L}, \quad \lambda_2 = \frac{b}{B}, \quad \lambda_3 = \frac{h}{H}. \quad (8)$$

The pre-stretch values for the change in dimensions from the undeformed initial shape to the pre-stretched reference configuration are defined as follows

$$\lambda_{1p} = \frac{L}{L_0}, \quad \lambda_{2p} = \frac{B}{B_0}, \quad \lambda_{3p} = \frac{H}{H_0}. \quad (9)$$

The deformation gradient tensor $F$ and the electric field vector $E$ for the above deformation mapping may be obtained as follows

$$F = \lambda_1 e_{11} + \lambda_2 e_{22} + \lambda_3 e_{33}, \quad E = E_0 e_3. \quad (10)$$

Now, the associated stress components in the corresponding directions may be obtained from the relation $(7)_1$ as follows

$$T_{11} = -p + 2\Omega_1 \lambda_1^2 + 2\Omega_2 (\lambda_1^2 \lambda_2^2 + \lambda_2^{-2}),$$

$$T_{22} = -p + 2\Omega_1 \lambda_2^2 + 2\Omega_2 (\lambda_1^2 \lambda_2^2 + \lambda_1^{-2}),$$

$$T_{33} = -p + 2\Omega_1 \lambda_1^{-2} \lambda_2^{-2} + 2\Omega_2 (\lambda_2^{-2} + \lambda_1^{-2}) - 2\Omega_5 E_0^2 - 4\Omega_6 (\lambda_1^2 \lambda_2^2) E_0^2. \quad (11)$$

An electro-elastic deformation theory is completely general for an isotropic electro-elastic material, and admits many possible specializations. We recall the general expression of six invariants (6) related to an amended energy density function, and we may discard $I_3 = 1$ for the incompressibility constraint. Therefore, a generalized electro-elastic material model for an incompressible isotropic dielectric elastomer may be generalized using the concept of an amended
energy density function (3). The energy density function, which includes the elastic and the electrical energy contributions is defined as follows

$$\Omega = \Omega_s - \frac{\epsilon_0}{2} (k_1 I_4 + k_2 I_5), \quad (12)$$

wherein $\Omega_s$ represents the free energy contribution by pure elastic deformation part of the dielectric elastomer, and $k_1, k_2$ are the material constant parameters. The above energy density function also successfully fulfills all the fundamental criteria on the form of energy density function presented by Darrijani et al. [9].

The general expression of the electro-mechanical instability relation for a dielectric elastomeric material in an equi-biaxial deformation condition ($\lambda_1 = \lambda_2 = \lambda$ and $T_{11} = T_{22}$ with $T_{33} = 0$) may be obtained from the relations (11) and (12) as follows

$$T_{11} = T_{22} = 2\Omega_{1s}(\lambda^{2} - \lambda^{-4}) + 2\Omega_{2s}(\lambda^{4} - \lambda^{-2}) - k_2 \epsilon_0 E_0^2, \quad (13)$$

wherein the notation $\Omega_{1s}$ and $\Omega_{2s}$ represent $\Omega_{1s} = \frac{\partial \Omega_s}{\partial I_1}$ and $\Omega_{2s} = \frac{\partial \Omega_s}{\partial I_2}$, respectively. The pre-stretching effect in the above electro-mechanical instability relation (13) may be applied by introducing $T_{11} = T_{22} = T_p$ at $E_0 = 0$. Now, we obtain an EMI model expression with an amended energy function (12) from the above relation (13) with $E_0 = \phi/h$ as follows

$$\phi = \frac{H}{k_2 \epsilon_0} \sqrt{2\Omega_{1s}(\lambda^{2} - \lambda^{-8}) + 2\Omega_{2s}(1 - \lambda^{-6}) - \frac{T_p}{\lambda^4}}. \quad (14)$$

The above relation (14) represents the generalized EMI model, which is formulated within the framework of the second law of thermodynamics. This EMI model (14) may be directly applied for a qualitative comparison between Neo-Hookean, Mooney-Rivlin, and Gent energy density functions. The expressions of the Neo-Hookean, Mooney-Rivlin, and Gent energy density functions are expressed as follows

$$\Omega_{sNH} = C_1(I_1 - 3), \quad \Omega_{sMR} = C_1(I_1 - 3) + C_2(I_2 - 3), \quad \Omega_{sG} = -\frac{C_1}{2} J_m \log \left(1 - \frac{I_1 - 3}{J_m}\right). \quad (15)$$

wherein $\mu, J_m, C_1, \text{ and } C_2$ are the material parameters for the corresponding energy density function. From the formulated EMI model (14) with the pre-stretching effect, we may compare the Neo-Hookean, Mooney-Rivlin, and Gent material models by plotting the corresponding non-dimensional potential versus stretch curves for the different pre-stretching condition as shown in the Figure 2. The constant parameters values are considered as $J_m = 72, C_1 = 28 \text{ MPa}, C_2 = 18 \text{ MPa}$ for a VHB type dielectric elastomeric film existing in the literature.

4. Results and discussions

The pre-stretching effect on an electro-mechanical instability phenomenon of a dielectric elastomeric material is presented in the Figures 2 for the Neo-Hookean, Mooney-Rivlin, and Gent type energy density function. In the Figures 2, the Gent EMI model exhibits the chain stiffening behavior at large deformation. On the other hand, the Neo-Hookean, Mooney-Rivlin based EMI models do not show the chain stiffening behavior. This is because of the limiting chain extensibility parameter $J_m$, which is not presented in the Neo-Hookean and Mooney-Rivlin material models. In addition, the Neo-Hookean and Mooney-Rivlin type energy density functions show the Gaussian distribution as compared to the non-Gaussian distributed Gent
energy density function. An interesting fact may also be seen in the Figure 2(b) that the pre-stretching effect does not majorly affect the Mooney-Rivlin EMI model as compared to other Neo-Hookean and Gent based EMI models.

![Figure 2: Comparison of Neo-Hookean, Mooney-Rivlin, and Gent based electro-mechanical instability (EMI) models.](image)

For the pre-stretching effect discussion in the EMI of a dielectric elastomer, we observed that the pull-in instability phenomenon takes place, if we ignore the pre-stretching effect. However, we may notice that in the Figures 2 the pull-in instability phenomenon is suppressed or specifically saying is eliminated as we introducing the pre-stretching effect. This happen because of that an electrostriction phenomenon affects the deformation significantly. During an electro-elastic deformation, the dielectric elastomeric film squeezes with an application of electrical voltage, and this thinning effect results the elongation of polarizable dipoles presented in the dielectric elastomeric material [10, 11]. The actuation process of a dielectric elastomer stabilizes through the pre-stretching effect by increasing the stiffness. Therefore, the dielectric elastomer operates in the thinner and stiffer region to generate a continuous deformation until the breakdown state. Now, we may conclude that the EMI may be controlled by carefully selecting the dielectric elastomer with sufficiently short chain, or applying the pre-stretching effect. The obtained results are also in line with the existing literature [10, 11], and show the parallel numerical results for the dielectric elastomeric material under an electrical loading condition.
5. Conclusions
In the present paper, we formulated the constitutive relationships for a dielectric elastomeric material under an equi-biaxial deformation condition within the framework of the second law of thermodynamics. The formulated constitutive relationships are then applied to obtain the electro-mechanical instability (EMI) model for a dielectric elastomeric material. The effect of pre-stretching in a dielectric elastomer is also analyzed analytically with an application of the electrical voltage by considering different energy density function. It is shown based on the obtained results that a mechanical pre-stretch can improve the EMI phenomenon of a dielectric elastomer. The EMI phenomenon of a dielectric elastomer is totally affected by the pre-stretching effect. Finally, an EMI phenomenon is fully connected with the electrostriction phenomenon of a dielectric elastomer. The obtained numerical results for the dielectric elastomer are found in line with the existing literature.

References
[1] F. Carpi, D. De Rossi, Dielectric elastomer cylindrical actuators: electromechanical modelling and experimental evaluation, Materials Science and Engineering: C 24 (4) (2004) 555–562.
[2] M. Wissler, E. Mazza, Modeling of a pre-strained circular actuator made of dielectric elastomers, Sensors and Actuators A: Physical 120 (1) (2005) 184–192.
[3] T. Lu, S. Cheng, T. Li, T. Wang, Z. Suo, Electromechanical catastrophe, International Journal of Applied Mechanics 8 (07) (2016) 1640005.
[4] J. Xia, Y. Ying, S. H. Foulger, Electric-field-induced rejection-wavelength tuning of photonic-bandgap composites, Advanced Materials 17 (20) (2005) 2463–2467.
[5] J. Zhu, M. Kolloso, T. Lu, G. Kofod, Z. Suo, Two types of transitions to wrinkles in dielectric elastomers, Soft Matter 8 (34) (2012) 8840–8846.
[6] H.-S. Choi, I.-H. Park, W.-K. Moon, On the physical meaning of maxwell stress tensor, The Transactions of The Korean Institute of Electrical Engineers 58 (4) (2009) 725–734.
[7] C. Rinaldi, H. Brenner, Body versus surface forces in continuum mechanics: Is the maxwell stress tensor a physically objective cauchy stress?, Physical Review E 65 (3) (2002) 036615.
[8] X. Zhao, W. Hong, Z. Suo, Electromechanical hysteresis and coexistent states in dielectric elastomers, Physical review B 76 (13) (2007) 134113.
[9] H. Darijani, R. Naghdabadi, Hyperelastic materials behavior modeling using consistent strain energy density functions, Acta mechanica 213 (3-4) (2010) 235–254.
[10] X. Zhao, Z. Suo, Electrostriction in elastic dielectrics undergoing large deformation, Journal of Applied Physics 104 (12) (2008) 123530.
[11] Z. Suo, Theory of dielectric elastomers, Acta Mechanica Solida Sinica 23 (6) (2010) 549–578.