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Hysteresis in electroactive polymers

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ABSTRACT

In this paper we present a model for the description of rate-independent hysteresis in Electroactive Polymers (EAPs). Our analysis is based on a model proposed by the authors for the description of damage and healing effects in polymeric materials and on a variational formulation for the resulting electromechanical equilibrium problem. The analysis of the class of equibiaxial strain, relevant for many actuation and energy harvesting devices, evidences that the model is effective and computationally efficient. The model shows a significant agreement with important experimental phenomena observed in EAP devices.

1. Introduction

EAPs devices are typically constituted by a thin, dielectric elastomeric film which is sandwiched between two compliant electrodes, attached to its surfaces. When a voltage is applied, due to Coulomb forces acting on the electrodes, the polymeric layer is compressed along the thickness direction with a resulting area expansion. This response is at the basis of technological applications of electroactive polymers as actuators and harvesting devices exhibiting important features such as lightweight, small size, low-cost, flexibility and fast response.

The advances in the design of new highly deformable polymeric materials is currently a very promising challenge for several applications of EAPs, ranging from robotics, to biomedical applications and to energy harvesting systems (see *e.g.*, Carpi et al., 2008). As a result EAPs attracted the attention of researchers of many different fields, ranging from Material Science (optimal material design), Physics, Electrical Engineering (control systems and energy harvesting) and technologists, Continuum Mechanics (non-linear electromechanical behavior, constitutive modeling).

From a theoretical point of view, many studies regarded the analysis of the highly nonlinear behavior leading to the occurrence of instabilities, localization and history-dependent effects observed in EAP applications. Particularly important in this framework are the analysis of the so called pull-in instability (De Tommasi et al., 2010b; Pelrine et al., 2000), wrinkling instabilities of the film (De Tommasi et al., 2011, 2012; Plante and Dubowsky, 2007), deformation localization (De Tommasi et al., 2013a; Puglisi and Zurlo, 2012; Zurlo, 2013; Puglisi, 2008), influence of electrode stiffness (De Tommasi et al., 2014; Carpi et al., 2003), dielectric breaking phenomena (Bozlar et al., 2012).

In this work we focus our attention on the hysteretic mechanical dissipation. This phenomenon may represent an important drawback for EAP applications *e.g.* for the resulting energy loss in the case of energy harvesting devices (York et al., 2010; Huang et al., 2013; Zhao et al., 2007) and for the uncertainty of the electromechanical response of control systems (Rizzello et al., 2013; Truong et al., 2013). The observed dissipation in the EAP devices can be addressed to several phenomena, ranging from rate-dependent viscous effects (Hong, 2011), current leakage (Plante and Dubowsky, 2007; Foo et al., 2012), rate-independent mechanical effects (Eddiai et al., 2012). In this paper attention is focused on this last effect, which represents the main source of dissipation for several technological applications (York et al., 2010).

With the aim to describe the occurrence of damage and hysteresis in EAP membranes, in this work we consider the model for hysteresis in amorphous materials recently proposed in De Tommasi et al. (2006); De Tommasi et al. (2008) within the variational approach adopted by the authors in De Tommasi et al. (2013c) to analyze the electromechanical equilibrium of EAP sheets.

In order to develop a theory for the material optimization of the EAP device (Yang et al., 2012; York et al., 2010), the target of this papers to show the strong correlation between the macroscopic dissipative behavior of the electroactive membrane and the distribution properties at the level of the micro scale. Specifically, the adopted damage model considers the complex mechanism of links breaking and recrosslinking at the network polymer scale.

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Schematically, the model describes the amorphous polymeric material as a mixture of elastic material (here modeled by an Ogden energy function) and damageable material. To take care of the amorphous character of the polymer, the damageable fraction is assigned as a distribution of materials with variable activation and breaking thresholds. Moreover, to describe the healing effect (recrosslinking) at the base of the cyclic hysteretic behavior, we assume that upon unloading a fraction of previously broken material can reform (D'Ambrosio et al., 2008; De Tommasi et al., 2009). This approach has shown to be efficient in the description of the Mullins effect for rubberlike material (De Tommasi and Puglisi, 2007) and biomaterials (De Tommasi et al., 2010).

After introducing the three-dimensional variational approach for the description of the hysteretic behavior of EAP bodies (Sect. 2), we focus on the important case of equibiaxial strains in membranes (Sect. 3), representing an optimal deformation class to increase the performance of energy harvesting devices (Huang et al., 2013) and a diffuse class of deformations also in EAP actuators (Plante and Dubowsky, 2006). We obtain explicit relations between the force, the voltage and the stretch, depending from the history of deformation through the only maximum previously attained value of the first invariant of the Cauchy–Green strain tensor (Sect. 3). As a result we show that the hysteresis model is computationally efficient and thus we believe that the suggested approach can be important in the development of optimal algorithm for the control of EAP devices.

As we show in Sect. 4 by considering stress-stretch cycles for fixed values of the voltage and voltage-stretch cycles at fixed values of the prestress, the model captures many important effects observed in the hysteresis behavior of EAPs (York et al., 2010): the stress decreases with increasing voltages; the activation is improved by increasing the level of prestress; Return Point Memory property of the cycles; reproducibility of internal cycles.

Finally, by an experimental comparison with spherical EAP actuators in (Gooyers, September 2009-Febuary 2012; Ahmadi et al., 2013), we show the possibility of a quantitative prediction of the hysteretic behavior in EAPs. Indeed after deducing the constitutive properties of the material by analyzing the zero voltage pressure-stretch cycle, we deduce the behavior at non-zero voltage, obtaining an accurate quantitative prediction of the hysteresis cycles of EAP ballon.

2. Electromechanical variational approach for EAP bodies

In order to study the electromechanical behavior of the dielectric polymer when dissipative damage and hysteresis are considered, in this section we extend to electroactive polymers the variational formulation proposed in De Tommasi et al. (2013b), which is based on the minimization of a Griffith-type energy to account for the continuous process of damage at the microscopic scale of the material.

Let $\boldsymbol{f}_t : \mathscr{B}_0 \to \mathscr{B}_t$ be the deformation of an EAP body $\mathscr{B}_0$ in the current configuration $\mathscr{B}_t$ at time $t \in (0,T)$. Let then $\mathbf{F}_t = \nabla \boldsymbol{f}_t$ be the deformation gradient with respect to points $\boldsymbol{X} \in \mathscr{B}_0$ of the reference configuration. We assume that two fully compliant electrodes under a potential difference V_t are attached on the boundary of the EAP body. Following a typical constitutive assumption for EAP dielectrics (ideal fluid dielectric), we consider a deformation independent dielectric permittivity ε. More general constitutive assumption can be considered (Dorfmann and Ogden, 2005), taking care of possible constitutive electromechanical couplings; however, we point out that the assumption of ideal dielectric material adequately describes the experimental behavior observed in several EAP devices (Suo, 2010).

Under this assumption it is possible to additively decompose the total potential energy of the system as follows:

$$G_t = \Psi_t + \frac{1}{2} Q(f_t, \varphi_t) V_t - \mathscr{W}^M(f_t) - \mathscr{W}^E(f_t, \varphi_t) \quad t \in (0,T), \tag{2.1}$$

where Ψ_t is the total internal (purely mechanical) energy at time t (specified in the following), $\varphi_t : \mathscr{B}_0 \to IR$ is the material description of the electrostatic potential $\overline{\varphi}_t : \mathscr{B}_t \to IR$ (i.e. $\varphi_t = \overline{\varphi}_t \circ \boldsymbol{f}$), $Q = Q(\boldsymbol{f}_t, \varphi_t)$ is the electrostatic charge on the electrodes, $\mathscr{W}^M$ and $\mathscr{W}^E$ are the mechanical and electrostatic work terms, respectively (in the following, when unnecessary, we suppress for simplicity of notation the explicit dependence on the time t). Since $\mathscr{W}^E = QV$ we have

$$G = \Psi - \frac{1}{2} QV - \mathscr{W}^M. \tag{2.2}$$

The effectiveness of this approach relies on the possibility of solving the electrostatic problem for an arbitrary state of deformation; this circumstance was shown possible in some important cases of technological interest (see Puglisi and Zurlo, 2012 and references therein), like the one considered in this work.

As anticipated above, to describe the complex damage and healing effects characterizing the polymer, we here consider the microstructure based phenomenological model for amorphous materials proposed in De Tommasi et al. (2006); De Tommasi et al. (2008). Roughly speaking (we refer the reader to these papers for a detailed discussion on the physical interpretation and mechanical assumptions of the model) we describe the EAP as an amorphous material, constituted at each point by a fraction α of elastic material with energy density ψ_e and a fraction $(1-\alpha)$ of damageable material with energy density ψ_d, that takes care of the scission and recrosslinking effects at the scale of the polymeric network.

The fraction α represents an elastic foundation which ensures non-vanishing strength even in the case of damage saturation. For this fraction we consider an Ogden energy function (Ogden, 1972)

$$\psi_e = \sum_{m=1}^{N} \frac{\mu_m}{a_m} \left(\lambda_1^{a_m} + \lambda_2^{a_m} + \lambda_3^{a_m} - 3 \right) \tag{2.3}$$

where $\lambda_i \, (i = 1,2,3)$ are the eigenvalues of $\mathbf{V} := \sqrt{\mathbf{B}}$, with $\mathbf{B} := \mathbf{F}\mathbf{F}^T$ the left Cauchy–Green tensor, satisfying the incompressibility assumption $\lambda_1\lambda_2\lambda_3 = 1$, and where μ_m and a_m are material constants.

Regarding the damageable fraction $(1-\alpha)$, following (De Tommasi et al., 2006; De Tommasi et al., 2008), in order to describe the activation and breaking phenomena of the network chains, we consider at each point a distribution of materials that activate and break at different levels of strain. Specifically, we introduce a Neo-Hookean type energy for the damageable fraction, and correspondingly we assume that activation and breaking effects depend only on the first invariant of deformation $I = \mathrm{tr}\mathbf{B}$. Thus there exist two material parameters $I_a < I_b$ such that when $I = I_a$ the material activates and when $I = I_b$ the material breaks. For $I_a \le I \le I_b$ the energy density depends on the relative deformation tensor $\widehat{\mathbf{F}}$ measured from the activation configuration $\mathbf{F} = \mathbf{F}_a$ (delivering $I = I_a$), with $\widehat{\mathbf{F}} = \mathbf{F}\mathbf{F}_a^{-1}$. To this scope we refer to the 'relative' strain measures $\widehat{\mathbf{B}} = \widehat{\mathbf{F}}\widehat{\mathbf{F}}^T$ and $\widehat{I} = \mathrm{tr}\widehat{\mathbf{B}}$. As a result for the damageable fraction the energy density function is

$$\psi_d = \begin{cases} 0 & \text{if} \quad I < I_a \\ \dfrac{\mu_d}{2}\left(\widehat{I} - 3\right) & \text{if} \quad I_a < I < I_b \\ \dfrac{\mu_d}{2}\left(\widehat{I}_b - 3\right) & \text{if} \quad I > I_b \end{cases}, \tag{2.4}$$

where μ_d is the shear modulus. In order to describe material disorder, it is further assumed that the fraction of damageable material

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