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A generalized electro-elastic theory of polymer networks

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ABSTRACT

A rigorous multi-scale analysis of the electromechanical coupling in dielectric polymers is conducted. The body couples stemming from a misalignment between the electric field and the electric-dipole density vector are studied and the conservation laws for polymer networks are derived. Using variational principles, expressions for the polarization and the stress are determined. Interestingly, it is found that the stress tensor resulting from coupled loadings in which the electric field is misaligned with the principal stretch directions is not symmetric and the asymmetry arises from the body couples. Next, the electromechanical response of a chain is analyzed. The deformations of the individual polymer chains are related to the macroscopic deformation via two highly non-linear constraints - the first pertaining to the compatibility of the local deformations with the imposed macroscopic one and the second stems from the symmetric part of the stress at equilibrium. In accord with the proposed framework, an amended three-chains model is introduced. The predictions of this model are found to be in excellent agreement with experimental findings. Lastly, the behavior of a polymer subjected to a simple shear and an electric field is studied. The offset between the electric field and the principal directions gives rise to body couples, a polarization that is not aligned with the electric field, and an asymmetric stress tensor.

1. Introduction

Electro-active polymers (EAPs) are dielectrics that deform in response to an electric stimulus. Thanks to their light weight, flexibility, availability and ability to undergo large deformations, EAPs can be used in a wide variety of applications such as artificial muscles (Bar-Cohen, 2001), energy-harvesting devices (McKay et al., 2010; Springhetti et al., 2014; Tutcuoglu and Majidi, 2014), micro-pumps (Rudykh et al., 2012) and tunable wave guides (Galich and Rudykh, 2017; Shmuel, 2015), among others. The work of Goshkoderia and Rudykh (2017) also revealed that the instability phenomenon can be taken advantage of in EAPs with a periodic structure. The actuation of EAPs is enabled due to the attraction between two oppositely charged electrodes attached to the faces of a soft thin polymer film. As a result of the Poisson's effect, the film expands in the transverse direction. The pioneering analysis of Toupin (1956) revealed that the electro-mechanical coupling is characterized by a quadratic dependence on the electric field. This result was later verified experimentally by Kofod et al. (2003),Wissler and Mazza (2007) and McKay et al. (2009), among others. Currently, the feasibility of EAPs is hindered due to the high electric fields (~ 100 MV/m) required for a meaningful actuation. This shortcoming is the result

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of the relatively low ratio between the electric and the elastic moduli (Pelrine et al., 2000a). Specifically, common flexible polymers have low dielectric moduli while polymers with high dielectric moduli are usually stiff.

Recent efforts to enhance the actuation suggested that this ratio may be improved. For example, the experimental work of Huang et al. (2004) demonstrated that organic composite EAPs experience more than 8% actuation strain in response to an electric field 20 MV/m. Embedding conducting particles in a soft polymer also proved to dramatically enhance the actuation (Stoyanov et al., 2010). In parallel, many theoretical works suggested that the actuation of dielectric composites can be improved with appropriate micro-structural adjustments (Tian et al., 2012; Galipeau and Castañeda, 2012; Rudykh et al., 2013; Lopez-Pamies, 2014; Spinelli et al., 2015, among others). These findings motivate a multi-scale analysis that accounts for the microstructural changes resulting from the electro-mechanical loading.

In the past, the response of polymers subjected to a purely mechanical loading has been extensively investigated at all length scales. Macroscopically, the behavior of polymers undergoing large deformations was modeled by Rivlin (1948), Gent (1996), Ogden (1997), and Lopez-Pamies (2010), and among others. The microscopically based analysis of Kuhn and Grün (1942) resulted in a Langevin based constitutive relation that led to various multi-scale models such as the 3-chain model (Wang and Guth, 1952), the tetrahedral model (Flory and Rehner, 1943; Treloar, 1946) and the 8-chain model (Arruda and Boyce, 1993). A macroscopic and a microscopic analyses of the electric response of dielectrics were carried out by Tiersten (1990) and Hutter et al. (2006), among others.

Examination of the coupled electro-mechanical response began with the macroscopic analysis of Toupin (1956). This work inspired Dorfmann and Ogden (2005) to derive an invariant-based constitutive relation characterizing the coupled behavior. In addition, the electrostrictive properties of EAPs were phenomenologically modeled by Zhao and Suo (2008), Ask et al. (2012b), Ask et al. (2012a), Jimenez and McMeeking (2013) and Gei et al. (2014), among others. Cohen et al. (2016) carried out a multi-scale entropy-based analysis of the coupled response of polymers subjected to an electric field that is aligned with the direction of a principal stretch. This microscopically motivated analysis resulted in expressions for the polarization and the stress that account for the evolution of the microstructure and provided an insight into the microscopic mechanisms that control the actuation. Furthermore, explicit approximations for the polarization and the stress in polymers composed of long-chain molecules were computed. The predictions according to these approximations were found to be in good agreement with experimental findings (Cohen and deBotton, 2016) and suggested that the electro-mechanical coupling can be enhanced by microscopic design (Cohen, 2017).

The current work expands the theory developed by Cohen et al. (2016) to the general case in which there is no restriction on the orientation of the electric field with respect to the deformation. To this end, a rigorous multi-scale analysis, starting from the monomer level, is conducted to determine the electro-mechanical response. It is shown that the application of an electric field at an offset to the principal stretches gives rise to body couples and, as a consequence, a non-symmetric stress tensor and a polarization that is not aligned with the electric field.

The paper is organized as follows. Section 2 provides a theoretical background. In Section 3, the electric body couples stemming from the dipolar monomers composing the polymer chains are examined and the conservation laws for polymer networks are derived. By employing variational principles, we obtain expressions for the polarization and the stress. In Section 4, the coupled response of a polymer chain comprising dipolar monomers is analyzed. The macroscopic response of the polymer is determined by an integration process from the chain to the macroscopic level. The multi-scale analysis reveals that the stress tensor is not necessarily symmetric and that the local deformations of the chains must satisfy two constraints - one pertaining to the compatibility with the macroscopic deformation and the other stemming from the symmetric part of the stress at equilibrium. In Section 5, the three-chains model of Wang and Guth (1952) is amended to comply with the constraints of the proposed framework. The merit of this model is attested through three sets of experimental findings in Section 6. Additionally, the three-chains model is used to predict the behavior of an EAP subjected to a simple shear and an electric field. The conclusions are summarized in Section 7.

2. Theoretical background

Consider the deformation of a dielectric body from a referential configuration to a current one as a result of an electro-mechanical loading. The body occupies a region $V_0 \in \mathbb{R}^3$ with a boundary ∂V_0 before the deformation and a region $V \in \mathbb{R}^3$ with a boundary ∂V at the current configuration. $\mathbf{X}$ and $\mathbf{x}$ denote the locations of the material points before and after the deformation, respectively. The mapping of the material points from the reference to the current configuration is $\mathbf{x} = \boldsymbol{\varphi}(\mathbf{X})$ and the corresponding deformation gradient is

$$\mathbf{F} = \nabla_{\mathbf{X}} \boldsymbol{\varphi}, \tag{1}$$

where the notation $\nabla_{\mathbf{X}}$ denotes the gradient operation with respect to the referential coordinate system. The determinant of the deformation gradient, $J = \det(\mathbf{F}) > 0$, is the ratio between the volumes of an infinitesimal element in the current and the reference configurations.

The velocity of the material points is $\mathbf{v}(\mathbf{x}, t)$ and the corresponding spatial velocity gradient is

$$\mathbf{L} = \nabla_{\mathbf{x}} \mathbf{v} = \dot{\mathbf{F}} \mathbf{F}^{-1}, \tag{2}$$