European Journal of Mechanics A/Solids xxx (2014) 1–12

Contents lists available at ScienceDirect

European Journal of Mechanics A/Solids

journal homepage: www.elsevier.com/locate/ejmsol

Multiscale analysis of the electromechanical coupling in dielectric elastomers

Noy Cohen [a], Gal deBotton [a,b,*]

[a] Dept. of Mechanical Engineering, Ben-Gurion University, Beer-Sheva 84105, Israel
[b] Dept. of Biomedical Engineering, Ben-Gurion University, Beer-Sheva 84105, Israel

ARTICLE INFO

Article history:
Received 8 October 2013
Accepted 26 February 2014
Available online xxx

Keywords:
Dielectric elastomers
Variational principles
Multi-scale analysis

ABSTRACT

The coupled electromechanical response of dielectrics is analyzed in terms of the local response and the distribution of its microstructural unit elements. A comparison between two variational statements for a continuous and a discrete systems results in an estimate for the macroscopic coupled electromechanical energy-density function. Next, a few types of unit elements are considered and the corresponding behaviors of dielectrics with a random distribution of these elements are calculated. Additionally, to highlight the possible effect of local constraints among the unit elements, a deterministic model for a network of polymer chains is proposed and its response to electric excitation is determined. The practically common case of a uniaxial stretch aligned with the electric field is examined in detail and the results are compared with widely accepted macroscopic models and available experimental results.

1. Introduction

Dielectric elastomers are materials that deform under electrostatic excitation. These light weight, flexible and available actuators can be used in a wide variety of applications such as artificial muscles (Bar-Cohen, 2001), energy-harvesting devices (McKay et al., 2010), micropumps (Rudykh et al., 2012), and tunable wave guides (Gei et al., 2011; Shmuel et al., 2012), among others. Roughly, the principle of the actuation is the attraction between two oppositely charged electrodes attached to the sides of a thin soft elastomer sheet. Due to Poisson's effect, the sheet expands in the transverse direction. This electromechanical coupling is characterized by a quadratic dependence on the applied electric field as theoretically described in Toupin (1956) and experimentally verified (*e.g.*, Kofod et al., 2003). However, the feasibility of these materials is limited due to the high electric fields required for meaningful actuation (~ 100 MV/m) (Pelrine et al., 2000; Stoyanov et al., 2010). This is because the relatively low ratio between the dielectric and the elastic moduli. Thus, common flexible polymers have low dielectric moduli while polymers with high dielectric moduli are usually stiff. Nonetheless, a few recent works demonstrated that this ratio may be improved. Experiments carried out by Huang et al. (2004) discuss organic composite EAPs attaining more than 8% actuation strain in response to an activation field of 20 MV/m. Stoyanov et al. (2010) show that by embedding conducting particles in a soft polymer the actuation can be dramatically improved. In parallel, theoretical works dealing with enhancement of coupling in composites also hint at the possibility of improved actuation with an appropriate adjustment of their microstructure (Tian et al., 2012; Galipeau and Ponte Castañeda, 2013; Rudykh et al., 2013; Gei et al., 2013).

The above recent findings motivate an in depth study of the electromechanical coupling in elastic dielectrics and its relation to their underlined microstructure. A *macroscopic level* investigation of this coupling has been performed by Toupin (1956). Dorfmann and Ogden (2005) developed an invariant-based representation for the constitutive behavior of EAPs. The possible influence of the deformation and its rate on the electromechanical coupling was considered too (*e.g.*, Ask et al., 2012; Gei et al., 2013). Moreover, application of the micro-sphere approach (*e.g.*, Miehe et al., 2004) to this class of materials was considered by Thylander et al. (2012). However, to the best of the authors' knowledge, a *microscopic level* analysis that relates the mechanical response of dielectric elastomers to electrostatic excitation in terms of their molecular microstructure is not yet available. Such an analysis will shed light on the origins of the electromechanical coupling in elastic dielectrics and may assist in the design of new and improved EAPs.

* Corresponding author. Dept. of Mechanical Engineering, Ben-Gurion University, Beer-Sheva 84105, Israel. Tel.: +972 8 647 7105.
E-mail address: debotton@bgu.ac.il (G. deBotton).

http://dx.doi.org/10.1016/j.euromechsol.2014.02.022
0997-7538/

Due to the wide usage of polymers in the last century, the *uncoupled* relations between their molecular microstructure and electrostatic and mechanical behaviors have been intensively investigated. The resulting models provide accurate descriptions for the overall response of the polymers and are broadly used by the scientific and the engineering communities. Microscopic analyses of the electrostatic behaviors of dielectrics can be found in Onsager (1936); Fröhlich (1986) and Pao and Hutter (1975), among others. Commonly, the starting point is the behavior of a dipole in an electric field. One such example is the Lorentz model that enables to express the relative permittivity and polarization of the material in terms of the number of mols per unit volume (*e.g.,* Blythe and Bloor, 2008). From a mechanical point of view, the pioneering works of Treloar (1943); Flory and Rehner (1943) relate the deformation of a polymer due to a mechanical loading to its microscopic properties, such as the number of chains per unit volume, by using statistical considerations. Wang and Guth (1952) and Arruda and Boyce (1993) among others, followed these works and considered models with different distributions of the chains in the polymer. These works stress the need for an analogous analysis concerning the *coupled* electromechanical behavior.

The goal of the present work is to perform a multiscale analysis that takes into account the distribution of the underlined microstructure and its local response to the applied electric field. We begin with variational statements for the internal energy and the electrical enthalpy (*e.g.,* Ymeri, 1997; McMeeking and Landis, 2005; deBotton et al., 2007; McMeeking et al., 2007) and consider two electrostatic equivalent systems. One is described in terms of macroscopic field variables while the other is written in terms of the corresponding discrete microscopic quantities. Leaning on the assumption that the electric field induced on a dipole by its neighbors is smaller than the applied electric field, a comparison between the two statements leads to the desired connection between the overall behavior and the molecular structure of the dielectric. Next, a few possible microscopic constitutive models are assumed and the overall coupled behaviors of dielectrics with different spatial distributions of the microstructure are determined. Lastly, in a way of an example, a deterministic polymer chain model which assumes specific geometries of the chains before and after the deformation is analyzed. This model assumes, in accordance with Treloar (1943), that all chains undergo the same deformation and that at the reference configuration the chains are uniformly distributed throughout the material. We conclude with a numerical example concerning the practically common case of an electric excitation aligned with the mechanical stretch. The predictions of the different models are compared and the influence of the microstructure and its evolution on the overall behavior is analyzed. The results are also compared with common phenomenological models (Tiersten, 1990) and with invariant based models of Dorfmann and Ogden (2005). The trends of the predicted coupled behavior agree with recent experimental results (Pelrine et al., 1998; Kofod et al., 2003; Wissler and Mazza, 2007; McKay et al., 2009; Tagarielli et al., 2012) as well as with the Gaussian chain model of Cohen and deBotton (2014).

2. Theoretical background

Consider the deformation of a body subjected to electromechanical loading from a referential configuration to a current one. 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. The material point locations are $\mathbf{X}$ and $\mathbf{x}$ before and after the deformation, respectively. The deformation is characterized by the mapping $\mathbf{x} = \boldsymbol{\chi}(\mathbf{X})$, and its gradient is

$$\mathbf{F} \equiv \nabla_{\mathbf{X}} \boldsymbol{\chi}, \tag{1}$$

where the operation $\nabla_{\mathbf{x}}$ denotes a derivative with respect to the referential coordinate system. The ratio between the volume of an element in the current and the reference configurations $J \equiv \det \mathbf{F}$ is strictly positive. The velocity of a material point is $\mathbf{v}(\mathbf{x})$, and the spatial velocity gradient is

$$\mathbf{L} = \nabla_{\mathbf{x}} \mathbf{v}, \tag{2}$$

where $\nabla_{\mathbf{x}}$ denotes a derivative with respect to the current coordinate system. For later use we recall that $\dot{\mathbf{F}} = \mathbf{L}\mathbf{F}$.

The induced electric field $\mathbf{E}$ on the body satisfies the governing equation

$$\nabla_{\mathbf{x}} \times \mathbf{E} = 0, \tag{3}$$

in the entire space. Accordingly, a scalar quantity, the electric potential ϕ, may be defined such that $\mathbf{E} = -\nabla_{\mathbf{x}} \phi$. The electric displacement field $\mathbf{D}$ in the vacuum surrounding the body is given via $\mathbf{D}_v = \varepsilon_0 \mathbf{E}$, where ε_0 is the permittivity in vacuum. Within the dielectric body $\mathbf{D} = \varepsilon_0 \mathbf{E} + \mathbf{P}$, where $\mathbf{P}$ is the electric dipole-density, also referred to as the polarization. The electric displacement is governed by the equation

$$\nabla_{\mathbf{x}} \cdot \mathbf{D} = \rho_f, \tag{4}$$

where ρ_f is the density of the free charge per unit volume. In this work we limit our treatment to the case of dielectrics with no free charge density.

The referential counterparts of the electric field and the electric displacement are (Dorfmann and Ogden, 2005),

$$\mathbf{E}^{(0)} = \mathbf{F}^T \mathbf{E}, \tag{5}$$

$$\mathbf{D}^{(0)} = J \mathbf{F}^{-1} \mathbf{D}. \tag{6}$$

These referential quantities abide the governing equations $\nabla_{\mathbf{x}} \times \mathbf{E}^{(0)} = 0$ and $\nabla_{\mathbf{x}} \cdot \mathbf{D}^{(0)} = 0$. Unlike $\mathbf{E}$ and $\mathbf{D}$, the referential polarization is not uniquely defined, and here we adopt the definition

$$\mathbf{P}^{(0)} = J \mathbf{F}^{-1} \mathbf{P}. \tag{7}$$

This ensures that the referential polarization is an energy conjugate to the electric field such that $1/J \mathbf{E}^{(0)} \cdot \mathbf{P}^{(0)} = \mathbf{E} \cdot \mathbf{P}$.

Assuming no body forces, the internal stress $\boldsymbol{\sigma}$ that develops in the dielectric due to electromechanical loading satisfies the equilibrium equation

$$\nabla_{\mathbf{x}} \cdot \boldsymbol{\sigma} = \mathbf{0}. \tag{8}$$

The pull back of the stress to the reference configuration $\boldsymbol{\sigma}^{(0)} = J \boldsymbol{\sigma} \mathbf{F}^{-T}$ is denoted the nominal stress. The equilibrium equation in terms of this stress measure is $\nabla_{\mathbf{X}} \cdot \boldsymbol{\sigma}^{(0)} = \mathbf{0}$.

The electrical boundary conditions on the dielectric boundary are given in terms of either the electric potential or the charge per unit area ρ_a. In the former case at the boundary ϕ is continuous and in the latter $(\mathbf{D} - \mathbf{D}_v) \cdot \hat{\mathbf{n}} = -\rho_a$, where $\hat{\mathbf{n}}$ is the outward pointing unit normal. Practically, in EAPs, ρ_a is the charge on the electrodes. The mechanical boundary conditions are given in terms of the displacement or the mechanical traction $\mathbf{t}$. The electric field induces the Maxwell stress

$$\boldsymbol{\sigma}^M \equiv \varepsilon_0 \mathbf{E} \otimes \mathbf{E} - \frac{\varepsilon_0}{2} \mathbf{E} \cdot \mathbf{E} \mathbf{I}, \tag{9}$$

Please cite this article in press as: Cohen, N., deBotton, G., Multiscale analysis of the electromechanical coupling in dielectric elastomers, European Journal of Mechanics A/Solids (2014), http://dx.doi.org/10.1016/j.euromechsol.2014.02.022