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Multiplicative electro-elasticity of electroactive polymers accounting for micromechanically-based network models

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Abstract

Electro-active polymers are materials which exhibit coupled electro-mechanical behavior at large strains. They respond by a deformation to an applied electrical field and are applied in advanced industrial environments as sensors and actuators, for example in robotics, biomimetics and smart structures.

A predictive modeling of these materials must account for characteristic features of their microstructure, consisting of an amorphous matrix of cross-linked polymer chain matrix and possibly a fraction of crystalline particles. The polymer network has an important effect on the electro-mechanical overall response, in particular in the large strain regime. The key intention of this work is to outline a general modeling structure for electroactive polymers on the macroscopic continuum level, that incorporates existing micromechanically-based network models for cross-linked polymers in a modular format. In order to account for electric dipoles associated with crystalline particle aggregates embedded into the polymer network, we consider a link to the network model based on multiplicative decompositions of the deformation gradient into electrically-induced and stress-producing parts. This includes a separate constitutive modeling of an electrically-induced stretch driven by the particle dipoles. Here, we incorporate Lee- and Clifton-type right and left decompositions, where the latter seems more appropriate for the modeling of electroactive polymers due to its dependence on the true electric field. Furthermore, micromechanical structures for a deformation-dependent permittivity of the polymer chain matrix are taken into account. We develop details of a unified modeling structure and its numerical implementation for those alternative kinematic assumptions, and combine it with the so-called microsphere network model of rubber elasticity, that exploits a homogenization over a chain orientation space. This provides an advanced model problem for the application of the proposed constitutive framework for EAPs.

Keywords: Electro-mechanics; Electroactive polymers; Micromechanics; Finite strains; Variational principles; Coupled problems

1. Introduction

1.1. Definition of EAPs and existing modeling frameworks

Electro-active polymers (EAPs) are materials with a polymer chain network microstructure, which exhibit coupled electro-mechanical behavior. They respond by a deformation to an applied electrical stimulus, an effect first observed

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by Röntgen in 1880 in an experiment on a rubber strip subjected to an electric field. EAP materials are divided into two main groups: field-activated and ionic materials. In *field-activated* EAPs, the activation is driven by Coulomb-type *electrostatic forces* between opposite charges, i.e. Maxwell stresses, which are created inside the dielectric as a result of an applied electric field. These materials allow fast response speeds, however, at high voltage. *Ionic* EAPs including gels and conductive polymers are actuated by electric-field-induced *diffusion of mobile ions* that causes a deformation. They operate at low voltage, however, perform at a low speed. Advanced EAP materials may achieve large electrically induced strains, making them the closest man-made materials to biological muscles. As a consequence, they are applied in many technological environments, e.g. in robotics, biomimetic and smart structures as sensors and actuators, see Bar-Cohen [1,2], Bar-Cohen and Zhang [3] and Smith [4]. Recent investigation shows that enhanced electro-mechanical properties can be achieved in certain types of composite materials, see Tian et al. [5] and Ponte Castañeda and Siboni [6].

The theoretical foundations for the *continuum modeling of electro-mechanical interactions* in solids were developed in the 1950s and 1960s, see Toupin [7], Truesdell and Toupin [8], Eringen [9], Landau and Lifshitz [10], Pao [11], Hutter et al. [12], Maugin [13], Eringen and Maugin [14,15] and Kovetz [16]. The recent developments with a particular focus on the constitutive modeling of field-activated EAPs Dorfmann and Ogden [17,18], McMeeking and Landis [19], Vu and Steinmann [20], Zhao et al. [21], Suo et al. [22], Ponte Castañeda and Siboni [6], Thylander et al. [23], Jiménez and McMeeeking [24], and Rosato and Miehe [25] will be particularly useful for our subsequent investigations. Recent numerical implementations of finite electro-mechanics combined with variational principles are proposed by Vu et al. [26], and Vu and Steinmann [20] for non-dissipative and by Miehe and Rosato [27], Rosato and Miehe [25], and Miehe et al. [28] for dissipative response.

1.2. Micromechanical ingredients to include in the modeling

This work focuses on the micromechanically-based continuum modeling of field-activated EAPs. These materials can further be subdivided into *dielectric elastomers*, which are activated by Coulomb forces, *ferroelectric polymers*, which exhibit spontaneous polarization and show piezoelectricity when poled and electrostriction in a non-poled phase, and *electrostrictive graft elastomers*, where the activation is caused by molecular alignment. All these materials have as a basic ingredient of their microstructure at least a fraction of an amorphous network, consisting of cross-linked polymer chains similar to natural or synthetic rubbers. This network has an important effect on the electro-mechanical overall response, in particular, when the material undergoes large strains. There is a substantial body of literature for the construction of highly predictive micromechanically motivated models in the pure mechanical context. The key intention of this work is to outline alternative model structures of finite electro-mechanics, which incorporate these network models in a modular format.

The elastic response of cross-linked polymers is dominated by an extreme reformability and can be well explained by statistical micromechanics, see for example Treloar [29] for an introduction. Elasticity of polymers above the glass transition temperature is achieved by a molecular microstructure consisting of very flexible and mobile long chain molecules and a three-dimensional network that is formed by occasional cross-links between molecules. The dominant contribution to the elastic response of rubber-like materials is due to changes in conformations of network constituents, yielding the so-called entropy elasticity theory. Entropic elasticity of chain molecules is well established in the context of statistical mechanics, see Kuhn [30,31], Kuhn and Grün [32], Treloar [29], Flory [33] and references cited therein. In the literature, many constitutive models for the macroscopic elastic response of rubbery polymers have been developed, see Boyce and Arruda [34], and Miehe et al. [35] for an overview. *Purely phenomenological macro-models* involve invariant or principal stretch based isotropic free energy functions, often having polynomial structures. The most advanced formulations are those of Ogden [36,37]. However, these approaches lack relations to the molecular structure of the material. This is achieved by *micromechanically-based network models*, such as the three chain model proposed by James and Guth [38], the eight chain model suggested by Arruda and Boyce [39] and the affine full network models considered in Treloar [40], Treloar and Riding [41], and Wu and van der Giessen [42]. It is well-known that the affinity assumption between microscopic and macroscopic deformation is not in agreement with experimental observations, in particular in the range of large deformations. Consequently, Boyce and Arruda [34] argued that the eight chain model yields more realistic results than the seemingly more precise affine full network models. A further improvement provides the non-affine microsphere model proposed in Miehe et al. [35], which allows a flexible modeling of the locking stretches in multi-dimensional deformations.