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Modelling of iron-filled magneto-active polymers with a dispersed chain-like microstructure

Prashant Saxena [a, b], Jean-Paul Pelteret [a], Paul Steinmann [a, *]

[a] Chair of Applied Mechanics, University of Erlangen–Nuremberg, Paul-Gordan Straße 3, Erlangen 91052, Germany
[b] Center for Integrative Genomics, University of Lausanne, Génopode Building, 1015 Lausanne, Switzerland

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Magneto-active polymers are a class of smart materials commonly manufactured by mixing micron-sized iron particles in a rubber-like matrix. When cured in the presence of an externally applied magnetic field, the iron particles arrange themselves into chain-like structures that lend an overall anisotropy to the material. It has been observed through electron micrographs and X-ray tomographs that these chains are not always perfect in structure, and may have dispersion due to the conditions present during manufacturing or some undesirable material properties. We model the response of these materials to coupled magneto-mechanical loading in this paper using a probability based structure tensor that accounts for this imperfect anisotropy. The response of the matrix material is decoupled from the chain phase, though still being connected through kinematic constraints. The latter is based on the definition of a ‘chain deformation gradient’ and a ‘chain magnetic field’. We conclude with numerical examples that demonstrate the effect of chain dispersion on the response of the material to magnetoelastic loading.

1. Introduction

Magneto-active polymers (MAPs) are smart materials in which the mechanical and the magnetic properties are coupled with each other. Typically these elastomers are composed of a rubber matrix filled with magnetisable iron particles. The magnetisable particles are usually between 1 and 5 μm in diameter and kept between 0 and 30% by volume of the entire mixture (Boczkowska and Awietjan, 2009; Ginder et al., 2002; Jolly et al., 1996; Co Ting Keh et al., 2013). The application of an external magnetic field causes the magnetisation of iron particles and the resulting particle–particle and particle–matrix interactions lead to phenomena such as magnetostriction and a change in the overall material stiffness (Danas et al., 2012; Varga et al., 2006). These elastomers have received considerable attention in recent times due to their potential uses in a variety of engineering applications, such as variable stiffness actuators (Böse et al., 2012) and vibration suppression by energy absorption (Co Ting Keh et al., 2013; Yalcintas and Dai, 2004).

Mathematical modelling of the coupling of electromagnetic fields in deformable continua has been an area of active research in the past. In particular, we note the contributions of Landau and Lifshitz (1960), Livens (1962), Tiersten (1964), Brown, (1966), Pao and Hutter (1975), Maugin and Eringen (1977), Maugin (1988), and Eringen and Maugin (1990). The advancement of MAP (along with electro-active polymer) fabrication in the laboratory setting, and hence their wider availability in recent decades, has led to another surge in research in this area. Furthermore, as opposed to metallic alloys and ceramics, newly developed polymer based materials can undergo very large deformations. This has resulted in focused explorations in the nonlinear regime of their response.

Recent developments in this field, based on the classical works mentioned above, are largely due to Brigadnov and Dorfmann (2003), Dorfmann and Ogden (2004, 2014), and Kankanala and Triantafyllidis (2004). The former's (Dorfmann and coworkers) work is based on the definition of a ‘total’ energy density function that implicitly accounts for magnetic and coupled energy stored in the polymer; while the latter's approach is to minimise a generalised potential energy with respect to internal variables, thereby yielding the relevant governing equations and boundary conditions. It is shown that any one of the magnetic induction, magnetic field, or magnetisation vectors can be used as an independent input variable and the other two obtained through constitutive relations. Based on these formulations, several nonlinear deformation problems have been studied by, for example, Dorfmann and Ogden (2005), Otténio et al. (2008), Bustamante et al. (2011a), and Danas

* Corresponding author. Tel.: +49 9131 8528501; fax: +49 9131 8528503.
E-mail address: paul.steinmann@ltm.uni-erlangen.de (P. Steinmann).

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et al. (2012). Steigmann (2009) and Maugin (2009) have discussed several important issues concerning the modelling of coupled magneto-electro-elasticity using continuum approaches. Further newer developments pertain to using implicit theories (Bustamante and Rajagopal, 2013) and rate-dependent theories (Saxena et al., 2013, 2014) for modelling more general effects, but are beyond the scope of this work.

MAPs can exhibit isotropic or anisotropic properties depending on the kind of fabrication process used. If the elastomers are cured in the presence of an external magnetic field, the magnetisable particles tend to form chain-like arrangements lending an overall directional anisotropy to the material. Experiments on such materials (Varga et al., 2006) have shown that anisotropic MAPs tend to have stronger coupling with the external magnetic field and are therefore more likely to be used in engineering applications.

The modelling of soft elastomers with a directional anisotropy is a subject of research in its own right. For example, averaging approaches have been adopted by Galipeau and Ponte Castãneda (2012) and Yin et al. (2006) to capture the microscopic behaviour of aligned magnetisable particles in soft carrier. In contrast, Rudykh and Bertoldi (2013) directly represented the chain-type microstructure using a laminate structure. Another common method of incorporating anisotropy, and that adopted within this work, is by using structural tensors. As described by Spencer (1971) and Zheng (1994), these can be coupled with the right Cauchy–Green deformation tensor to obtain scalar invariants through symmetry arguments. The invariants are then used as an independent input in the energy density function defining the material properties. This method has been used by, among others, Shams et al. (2011) for modelling pre-stressed elastic solids, Holzapfel and Gasser (2001) for modelling fibre-reinforced composites, and Bustamante (2010) and Danas et al. (2012) for modelling MAPs with a directional anisotropy. One needs to choose at least a minimum number of invariants for completeness (Destrade et al., 2013) and take care while performing energy decomposition for numerical implementation of incompressible materials (Sansour, 2008). Another approach towards this problem is by decoupling the response of the matrix and the anisotropic part, thereby considering different kinematic variables and energies for each. This has been used by Klinkel et al. (2004) in the case of anisotropic elasto-plasticity and by Nedjar (2007) for modelling anisotropic visco-elasticity. Based on this latter approach, Saxena et al. (2014) recently presented a model for nonlinear magneto-viscoelasticity of anisotropic MAPs.

Recent experiments have shown another rather important feature in the microstructure of anisotropic iron-filled MAPs, namely that the particle chains formed due to the curing of an MAP under an external magnetic field are not all aligned in the same direction. The chains combined together have an average alignment in the direction of the magnetic field applied during curing, but individual chains do have an observable dispersion that may possibly influence the macroscopic response of the material. Modelling of this phenomenon and demonstration of the effect of chain dispersion on the overall macroscopic response of an MAP are the main contributions of this paper. We note that, mathematically, this phenomenon is similar to the dispersion of fibres in biological tissues as is discussed in the papers by Gasser et al. (2006), Holzapfel and Ogden (2010) on the modelling of blood vessels, and Federico and Herzog (2008) on articular cartilage. In these works, the authors considered a generalised structure tensor based on a probability density function that accounts for the dispersion of embedded fibres.

Numerical methods, and in particular finite element analysis, have been widely used in the study of magneto-sensitive materials in order to understand and predict both their micro- and macroscopic behaviour. The formation of particle chains in magneto-rheological fluids, effectively characterising the pre-cured state of an MAP, has been investigated by Ly et al. (1999) and Simon et al. (2001). For the case of solid carriers, Boczkowska et al. (2010), Chen et al. (2013) and Vogel et al. (2014) have studied the movement of magnetic particles in elastomers. A coupled scalar magnetic potential formulation has been utilised to predict both the magnetic and deformation fields at the macroscopic level, where consideration of the surrounding free space is necessary. For example, Kannan and Dasgupta (1997) adopted this approach to study the magnetostrictive behaviour of MAPs and their application in mini-actuators, Zheng and Wang (2001) investigated the magnetisation of a ferromagnetic plate and Bermúdez et al. (2008) demonstrated its application to electromagnets. Furthermore, the shear behaviour of a magnetised block in free space has been considered by Marvalova (2008) and Bustamante et al. (2011a), the latter of whom also investigated its contractile behaviour.

The remainder of this paper is arranged in the following manner: In Section 2 we outline the fundamental aspects of continuum mechanics pertaining to magnetoelasticity. Following this, in Section 3 we provide a motivation and the mathematical formulation of the dispersed magnetisable particle chains that comprise the MAP microstructure. We then detail a decoupled energy model for quasi-incompressible media in Section 4, and the associated energy model for the free space. In Section 5, we briefly present the finite element formulation used for performing the numerical computations. Analytical and finite element examples, used to demonstrate the behaviour captured by the constitutive model, are presented in Sections 6 and 7 respectively. Lastly, some concluding remarks are presented in Section 8.

2. Kinematics, balance laws and boundary conditions

We consider a body composed of a quasi-incompressible magnetoelastic material which, in a state of no stress and no deformation, occupies the reference configuration $\mathscr{B}_0$ with a boundary $\partial\mathscr{B}_0$. In this state, the free space surrounding the body is denoted by $\mathscr{S}_0$ and the entire domain by $\mathscr{D}_0 = \mathscr{B}_0 \cup \mathscr{S}_0$. On a combined mechanical and magnetic static loading, the body occupies the spatial configuration $\mathscr{B}_t$ at time t with the boundary $\partial\mathscr{B}_t$. The corresponding configurations for the free space and entire domain are denoted by $\partial\mathscr{B}_t$ and $\mathscr{D}_t = \mathscr{B}_t \cup \mathscr{S}_t$, respectively. A deformation function $\boldsymbol{\varphi}$ maps the points $\mathbf{X} \in \mathscr{D}_0$ to the points $\mathbf{x} \in \mathscr{D}_t$ by the relation $\mathbf{x} = \boldsymbol{\varphi}(\mathbf{X})$. The deformation gradient tensor is given by a two-point tensor $\mathbf{F} = \nabla_0 \boldsymbol{\varphi}$, ∇_0 being the differential operator with respect to $\mathbf{X}$. The determinant of $\mathbf{F}$ is given by $J = \det \mathbf{F}$ such that the condition $J > 0$ is always satisfied. For the case of an incompressible material, as presented in Section 6, the constraint $J \equiv 1$ is enforced.

It is assumed that the material is electrically non-conducting and that there are no electric fields. Let $\boldsymbol{\sigma}$ be the symmetric total Cauchy stress tensor (Dorfmann and Ogden, 2004) that takes into account magnetic body forces, ρ be the mass density, $\mathbf{f}_m$ be the mechanical body force per unit deformed volume, $\mathbf{a}$ be the acceleration, $\mathbb{b}$ be the magnetic induction vector in $\mathscr{D}_t$, and $\mathbb{h}$ be the magnetic field vector in $\mathscr{D}_t$. The balance laws are expressed as (Brown, 1966; Maugin and Eringen, 1977)

$$\nabla\cdot\boldsymbol{\sigma} + \mathbf{f}_m = \rho\mathbf{a}, \quad \boldsymbol{\sigma}^t = \boldsymbol{\sigma} \quad \text{in } \mathscr{B}_t; \quad \nabla\times\mathbb{h} = \mathbf{0}, \quad \nabla\cdot\mathbb{b} = 0 \quad \text{in } \mathscr{D}_t. \tag{1}$$

Here ∇ denotes the differential operator with respect to $\mathbf{x}$ in $\mathscr{D}_t$. Equation $(1)_1$ is the statement of balance of linear momentum, equation $(1)_2$ is the statement of balance of angular momentum, equation $(1)_3$ is a specialisation of the Ampère's law, and equation $(1)_4$ is the statement of impossibility of the existence of magnetic