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# Multiplicative magneto-elasticity of magnetosensitive polymers incorporating micromechanically-based network kernels

G. Ethiraj, C. Miehe*

*Institute of Applied Mechanics (CE), Chair 1 University of Stuttgart, Pfaffenwaldring 7, Stuttgart 70569, Germany*

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ABSTRACT

Magnetosensitive elastomers are a class of composite materials whose mechanical response may be altered by application of magnetic fields. Such materials have tunable mechanical properties and find use in controllable stiffness devices and applications for active control of structural components. In this work we present a novel approach to the macroscopic magneto–elastic modeling of magnetorheological elastomers (MREs) at finite strains, whose structure accounts in a modular format for micromechanically-based ingredients. Keeping in mind the composite nature at the microscale, we develop a constitutive formulation that provides *two microstuctural-based kernels* for (i) the energy of the magnetized rigid iron particles and (ii) the elastic energy of the deformable polymer network. The latter is achieved by a *multiplicative elasto-magnetic split* of the deformation gradient. Here, a left decomposition is proposed that includes a magnetostrictive deformation in terms of the spatial (true) magnetization vector of the composite material. This approach induces an *anisotropic Eulerian metric* depending on the magnetization, which is used to map standard isotropic chain statistics into anisotropic ones. As a consequnce, the approach allows to make use of *micromechnically-based isotropic network models for polymers* in a modular format and extends their application to the anisotropic coupled magnetomechanical response. In particular, such a formulation allows the inclusion of the *microsphere model* for network-based elasticity, that has been successfully applied to the modeling of rubber-like polymers. We discuss details of a unified modeling structure within a variational formulation of finite magneto-elasticity, and outline ingredients of the finite element implementation of the coupled problem. The modeling capabilities are demonstrated by solving application-oriented boundary value problems.

## 1. Introduction

### *1.1. Definition of MREs and existing modeling frameworks*

Magnetorheological elastomers (MREs) are a class of solids that consist of rubber matrix embedded with micro- or nano-sized magnetizable particles such as iron. As a result of this specific composite nature, the mechanical properties of these materials can be varied by the application of magnetic fields. This fact gives a wide scope for the exploitation of such materials in industry. It is therefore of great interest to be able to reliably model or predict the properties of such materials by constitutive equations

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* Corresponding author. Tel.: +49 711 66379; fax: +49 711 66347.
*E-mail address:* cm@mechbau.uni-stuttgart.de (C. Miehe).

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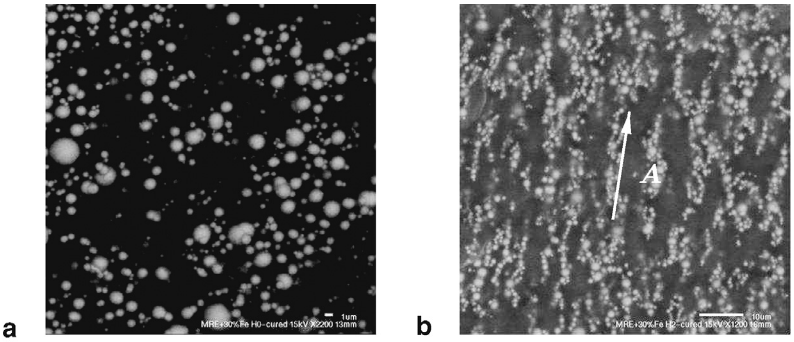

**Fig. 1.** Scanning electron microscope (SEM) pictures of microstructure of MRE-iron particles in silicone M4601 matrix, Kallio (2005). (a) Isotropic or random distribution of iron particles in polymer matrix and (b) iron particles aligned in a preferred direction (transversally isotropic) indicated by the Lagrangian structural vector ***A***.

and present the solution and analysis of representative boundary value problems. Also, since deformations of elastomers are typically quite large, there is an added need to develop the constitutive theory in a finite strain context.

MREs may be classified into two categories on the basis of the arrangement of the magnetizable particles in the polymeric matrix: (i) isotropic and (ii) anisotropic. Isotropic MREs have the magnetizable particles uniformly distributed throughout the matrix while anisotropic MREs have these particles aligned along a particular direction, see Fig. 1. Such an arrangement is brought about by applying a magnetic field during the curing process such that the particles form chain like structures in that direction. In this paper, we focus on the *isotropic category* of MREs.

Experimental work and simple one dimensional models for isotropic and anisotropic MREs have been presented by Jolly, Carlson, Muñoz, and Bullions (1996), Davis (1999), Bellan and Bossis (2002) and Varga, Filipcsei, and Zrínyi (2005, 2006). These contributions focus on estimating the change in shear modulus of the material motivated by ab initio calculations for magnetic dipole–dipole interaction and experiment. The dissertation by Kallio (2005) covers preparation and characterization of MREs with data on static and dynamic properties. The articles by Bednarek (1999) and Ginder, Clark, Schlotter, and Nichols (2002) are also valuable contributions in this context. The work by Diguet (2010) addresses, additionally, micromechanically-based modeling aspects and considers the shape effect in some detail among other aspects. All the above works provide valuable data and hints for continuum approaches of the type presented here. We recall the classical works of Toupin (1956), Brown (1951a, 1951b,1966); Maugin (1988), Maugin and Eringen (1977) and Tiersten (1964, 1987, 1990) which have laid the foundations of finite strain thermo-electro-magneto-mechanics. These works form the background for more advanced models presented more recently, that are thermodynamically consistent and account for magnetomechanically coupled finite deformation. We refer to the works Dorfmann and Ogden (2003, 2004a, 2004b) and Bustamante, Dorfmann, and Ogden (2007a) which have been extended to include viscoelastic response by Saxena, Hossain, and Steinmann (2013). Kankanala and Triantafyllidis (2004, 2008) further consider stability issues of the coupled problem, while Danas, Kankanala, and Triantafyllidis (2012) present experimental results on anisotropic MRE and a generalized anisotropic material model that is fitted to the obtained experimental data.

Computational models at the microscale level have furthered the understanding of the micromechanics of MREs. While Borcea and Bruno (2001) consider pair-wise interaction of magnetic dipoles to obtain constitutive relations that are exact to second-order in the volume-fraction of the particles, Ponte Castañeda and Galipeau (2011) and Galipeau and Ponte Castañeda (2012) use a homogenization approach to obtain the constitutive response for magnetorheological elastomers at finite strain. We also make note of the work Weeber, Kantorovich, and Holm (2012) that presents microscopic mechanisms for deformations of ferrogels. The particular form of the multiplicative elasto-magnetic split of the deformation gradient chosen for our continuum model is consistent with the atomistic simulations presented therein.

### *1.2. Micromechanically-based modular approach for modeling of MREs*

With the above mentioned literature as background, we aim to fill in what is missing in literature, namely a fully coupled finite element implementation of a thermodynamically consistent material model for isotropic magneto-elastic elastomers, that is *motivated from micromechanics* and is consistent with experimental observations. The modeling approach is directly motivated by considering magneto–mechanical interactions on the microscale of such composite materials. Furthermore, a knowledge of the composition of these materials down to the microscale, i.e. iron particles embedded in a rubber matrix, compels us to look at the finite strain modeling of rubberlike polymers: a field with a long and rich history. A detailed review of the literature in modeling of rubber is skipped here for the sake of brevity and we recall only works on *micro-mechanically-based network models*, such as the three chain model proposed by James and Guth (1943), the eight chain model suggested by Arruda and Boyce (1993) and the affine full network models considered by Treloar (1954), Treloar and Riding (1979) and Wu and van der Giessen (1993). A further improvement is provided by the non-affine micro-sphere model proposed in Miehe, Göktepe, and Lulei (2004), which allows a flexible modeling of the locking stretches in multi-dimensional deformations. In Miehe et al. (2004), the microsphere model was equipped with a characteristic micro-tube deformation that is linked to the macroscopic deformation.