MAGNETORHEOLOGICAL ELASTOMERS: EXPERIMENTS AND MODELING

N. Triantafyllidis, **K. Danas** *Laboratoire de Mécanique des Solides, C.N.R.S. UMR7649, Ecole Polytechnique, ParisTech, 91128 Palaiseau, France. Téléphone : 0196335798; 0169335786, Adresse(s) électronique(s) : nick@lms.polytechnique.fr, kdanas@lms.polytechnique.fr*

S.V. Kankanala *BD Medical – Medical Surgical Systems, Sandy UT 84070, USA Adresse(s) électronique(s) : sundeep_kankanala@bd.com*

Mots clés : Magnetoelasticity; Magnetomechanical processes; Finite strain; Energy methods; Nonlinear elasticity

1 INTRODUCTION

Magnetorheological elastomers (MREs) are a class of solids that consist of a rubber matrix filled with magnetizable particles, typically sub-micron sized iron particles (see Rigbi and Jilkén (1983) and Ginder et al. (1999)). The interest in these materials stems from their strong magnetoelastic coupling properties. The application of an external magnetic field **h**, tends to align the initially random magnetization vectors of the particles with the applied external field. As a result the interparticle magnetic forces result in the macroscopic magnetostriction of the MRE.

Although theoretical research in general coupled field theories in mechanics was actively conducted from the early fifties to the early seventies, the attention that this area received in the next twenty years was considerably diminished, perhaps due to the absence of technologically relevant applications. One might also speculate as an additional factor the lack, at the time, of the appropriate computer hardware that is required to handle the complicated numerical calculations necessary for the solution of the resulting boundary value problems (geometric nonlinearities, coupling between mechanical and magnetic fields). The situation changed in the late nineties, due to an application-driven strong interest in MR devices (see for example Carlson and Jolly (2000)) and recent experimental investigations of Ginder et al. (1999) on MREs and their applications as well as the more recent studies of Hubert et al. (2003), Coquelle and Bossis (2006), Diguet et al (2009,2010).

From a theoretical viewpoint, the excellent monograph by Brown (1966), which was published in the mid-sixties, presents a direct (using conservation laws) and a variational (based on a potential energy) approach, both resulting in the same governing equations and boundary conditions for the finite strain magnetoelasticity of non-conducting solids. Further refinements and complications in finite strain magnetoelasticity were proposed in the early seventies by Maugin and Eringen (1972), which included magnetic couple stresses and spin interactions stresses as a result of the dependence of the solid's free energy not only on the gradient of deformation and magnetization but on the spatial gradient of the magnetization as well. On the numerical side, most of the literature on the subject (e.g. Hirsinger and Billardon (1995), Huang et al. (1999)) solve the mechanical and magnetic problems independently and then iterate until the problem converges. Other researchers solve the coupled problem directly (Ren et al. (1995)), or using fully-coupled FFT algorithms (Brenner, 2010), but in most of these cases a small strain formulation is adopted and different expressions are used for the calculation of the body forces in each one of these works.

An intriguing feature in the continuum modeling of MREs are the different expressions for the magnetic (Maxwell) part of the total stress and the body forces, depending on adopted hypotheses. In an effort to reconcile theses differences Dorfmann and Ogden (2003) and Kankanala and Triantafyllidis (2004) have proposed fully coupled magneto-mechanical formulations for these materials. In particular, Kankanala and Triantafyllidis (2004) have derived a consistent, fully Lagrangian variational formulation with a local energy minimum at equilibrium (instead of the saddle points in the principles proposed by Brown (1966)), which is appropriate for the finite element implementation of coupled magneto-mechanical problems. In particular, this unified variational approach has already been applied successfully in the simpler context of electromagnetic forming (where an eddy current approximation is used and there is no magnetization present in the solid) (Thomas and Triantafyllidis, (2009)). However, the numerical implementation of these coupled variational principles remains an open question.

All the above-mentioned work on MREs is phenomenological and based on continuum models. However, due to the recent interest in particle-filled MREs, there are recent efforts to use micromechanical models and mean-field homogenization techniques to predict their overall properties based on particle volume fraction and particle shape and distribution information (e.g. Daniel et al. (2008), Corcolle et al. (2009), Diguet et al. (2009, 2010), Ponte Castañeda and Galipeau (2010), Galipeau and Ponte Castañeda (2012)). One should also mention here other recent studies involving magnetoelastic interactions, such as magnetostriction and the development of fine microstructures in thin films (e.g. James and Kinderlehrer (1993)). However, in our opinion, there are still several open questions that need to be answered. For instance, the use of appropriate independent variables in the homogenization problem (i.e., magnetization M or magnetic field **H** or magnetic field B as in Ponte Castañeda and Galipeau (2010)) is of crucial importance, especially for numerics. Finally we should also mention the very recent work of Kuo, Slinger and Bhattacharya (2010) that addresses the issue of optimizing magnetoelastic coupling by microstructure modification.

The present study is divided into two parts and makes use of the works of Kankanala and Triantafyllidis (2004) and Danas et al. (2012). In the first part a general presentation of the governing equations for MREs is given. Moreover, using the energy approach we propose a minimum, fully-coupled variational principle which is appropriate for finite element implementation. In the second part of this work, we present recent experimental results for MREs with particle-chain microstructures subjected to prestressing and arbitrary magnetic fields. Then, we propose a transversely isotropic energy density function that is able to reproduce these experimental measurements. In order to explain (i) the counterintuitive effect of dilation under zero or compressive applied mechanical loads for the magnetostriction experiments and (ii) the importance of a finite strain constitutive formulation even at small magnetostrictive strains, we propose microscopic mechanisms of deformation which are further supported by full-field coupled FEM microstructural calculations.

2 THEORETICAL ANALYSIS

In this section, we present briefly the theoretical background for magnetoelastic solids. More specifically, the starting point for the exact or approximate solution of all boundary value problems is based on the construction of appropriate variational principles. Thus, following Kankanala and Triantafyllidis (2004), we define the potential energy $\mathcal{E}(\mathbf{u},\mathbf{M},\mathbf{A})$ of the magnetoelastic solid as

$$\mathcal{E}(\mathbf{u},\mathbf{M},\mathbf{A}) = \int_V \rho_0(\psi - \mu_0\,\mathbf{h}_0\cdot\mathbf{M} - \mathbf{f}\cdot\mathbf{u})\mathrm{d}v + \frac{1}{2\mu_0}\int_{\mathbf{R}^3} \|\nabla\times\mathbf{A} - \mu_0\rho_0\mathbf{M}\|^2\,\mathrm{d}v - \int_{\partial V}\mathbf{t}\cdot\mathbf{u}\mathrm{d}a, \quad (1)$$

where $\psi(\mathbf{C},\mathbf{M})$ is the free energy plus magnetic dipole energy (termed also “anisotropy energy”), and is a function of $\mathbf{C} = \mathbf{F}^T\cdot\mathbf{F}$ (right Cauchy-Green tensor), $\mathbf{F}$ is the deformation gradient, $\mathbf{M}$ is the magnetization per unit mass, $\mathbf{h}_0$ is the externally applied magnetic field, $\mathbf{f}$ is applied non-magnetic body force (e.g., gravity) per unit mass, $\mathbf{A}$ is the vector potential for the magnetic field perturbation, ρ_0 is the reference mass density, μ_0 is the magnetic permeability of free space and $\mathbf{t}$ is the externally applied mechanical traction. Notice that the magnetic energy appears in the second integral over the entire space $\mathbf{R}^3$ since the magnetic field also exists outside the solid under consideration, which occupies a volume V with boundary ∂V. The above energy is a functional of the independent variables $\mathbf{u}$, $\mathbf{M}$, and $\mathbf{A}$.

The Euler-Lagrange equations with respect to $\mathbf{A}$ are Ampere’s equations and interface conditions (across an interface with reference normal $\mathbf{n}$), namely:

$$\mathcal{E}_{,\mathbf{A}}\,\delta\mathbf{A} = 0 \;\Rightarrow\; \nabla\times\mathbf{H} = \mathbf{0} \quad \text{in} \quad \mathbf{R}^3, \quad \mathbf{n}\times[\![\mathbf{H}]\!] \quad \text{on} \quad \partial V, \quad \mathbf{H} = \mathbf{h}\cdot\mathbf{F}, \quad (2)$$

where $\mathbf{H}$ and $\mathbf{h}$ are the h-fields in the reference and current configurations, respectively. Variation with respect to specific magnetization $\mathbf{M}$ gives the magnetic part of the constitutive response:

$$\mathcal{E}_{,\mathbf{M}} = 0, \quad \Rightarrow \quad \frac{\partial\psi}{\partial\mathbf{M}} = \mu_0\,\mathbf{h}. \quad (3)$$

Finally variation with respect to displacement $\mathbf{u}$ gives the equations of mechanical equilibrium and corresponding interface conditions:

$$\mathcal{E}_{,\mathbf{u}} = 0, \quad \Rightarrow \quad \nabla\cdot\mathbf{\Pi} + \rho_0\mathbf{f} = 0 \quad \text{in} \quad V, \quad \mathbf{n}\cdot[\![\mathbf{\Pi}]\!] = \mathbf{T}. \quad (4)$$

Here, $\mathbf{T}$ is the mechanical traction in the reference configuration and the first Piola-Kirchhoff stress, $\mathbf{\Pi}$, is related to the total Cauchy stress $\boldsymbol{\sigma}$ by

$$\mathbf{\Pi} = J\,\mathbf{F}^{-1}\cdot\boldsymbol{\sigma},\quad \boldsymbol{\sigma} = \rho\left[2\,\mathbf{F}\cdot\frac{\partial\psi}{\partial\mathbf{C}}\cdot\mathbf{F}^T + \mu_0\,(\mathbf{M}\,\mathbf{h} + \mathbf{h}\,\mathbf{M})\right] + \mu_0\left[\mathbf{h}\,\mathbf{h} - \frac{1}{2}\,(\mathbf{h}\cdot\mathbf{h})\;\mathbf{I}\right], \tag{5}$$

where $J = \det\mathbf{F}$ and $\rho = \rho_0/J$ is the current mass density.

Notice from (5) that the total Cauchy stress $\boldsymbol{\sigma}$ has a mechanical part (the term in ψ, recognized from finite elasticity) plus a magnetic part (termed Maxwell stress) and is obviously symmetric. It is worth noticing that even in the absence of material ($\rho_0 = 0$) the Maxwell stress component is nonzero, a concept which is a bit strange for solid mechanics where the concept of stress is associated with the presence of a solid.

The advantage of using a unified variational principle is evident from the absence of need to consider separately electromagnetic body forces and interface conditions (the body forces are simply found as the divergence of the Maxwell stress in (5)). It should also be pointed out here that, as seen from (5), the Maxwell stress is nonlinear in terms of the magnetic field quantities, thus explaining the need for a full Lagrangian formulation of the variational principle.

3 EXPERIMENTS AND MODELING

Experiments are carried out (Danas et al., 2012) for MREs comprising 25% of iron particles of sizes ranging from $0.5\mu m$ to $5\mu m$ cured in a 0.8T magnetic field. The application of a magnetic field during the curing process leads to formation of particle chains aligned with the curing field direction. The experiments involve three different setups; (a) uniaxial stress tests in the direction of a magnetic field which is aligned with the particle chains, (b) uniaxial stress tests in the direction of a magnetic field which is perpendicular to the particle chains and (c) simple shear tests where the particle chains are initially aligned with the applied magnetic field, as shown in Fig. 1.

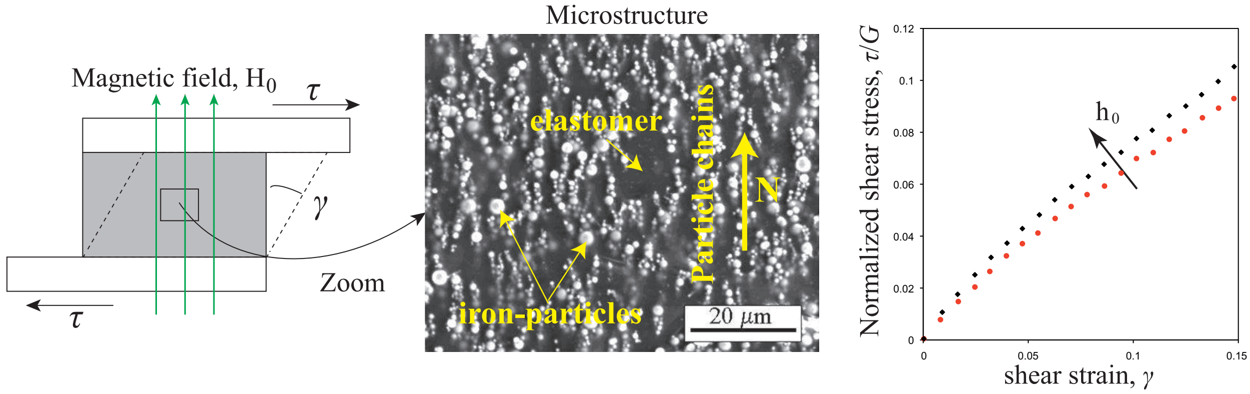

FIG. 1 – *An illustration of the stiffening effect of a magnetic field on the shear stress-strain behavior of a MRE (left) with particle-chain microstructure, initially aligned with the applied magnetic field (center). The stiffening in the mechanical response when a magnetic field* $\mathbf{h}_0$ *is applied is due to inter-particle magnetic forces (right).*

The other part of this work pertains in finding an energy density function ψ that best fits the experiments reported above (Danas et al., 2012). The material under investigation is a transversely isotropic composite since the iron particles form chains along a certain direction. This implies that the free energy density ψ should also depend on the unit vector $\mathbf{N}$, (see Fig. 1), which defines the initial orientation of the particle chains. Thus, one has

$$\psi = \psi(\mathbf{C},\mathbf{N},\mathbf{M}),\quad \mathbf{C} = \mathbf{F}^T\bullet\mathbf{F},\quad \mathbf{N}\bullet\mathbf{N} = 1. \tag{6}$$

The reader is referred to the work of Danas et al. (2012) for the derivation and detailed expressions of the energy function ψ.

Fig. 2 shows experimentally measured magnetostriction strain $\Delta\varepsilon$ versus the applied nondimensional magnetic field $h/\rho_0 M_s$ for uniaxial stress tests (with ρ_0 denoting the initial material density). In Fig. 2a, the magnetostriction is plotted for different preloads σ/G, which are aligned with the applied magnetic field and the particle chain ($\mathbf{h} \parallel \mathbf{N}$). The magnitude of magnetostriction increases in

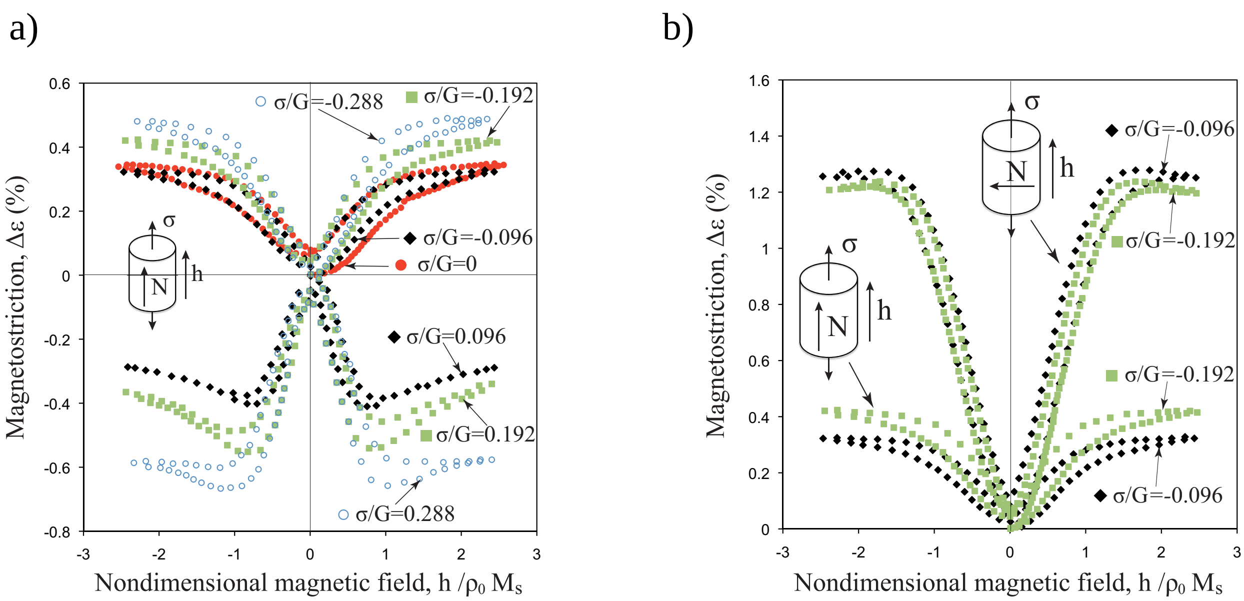

FIG. 2 – *Magnetostriction $\Delta\varepsilon$ versus the applied nondimensional magnetic field $h/\rho_0 M_s$ for various prestresses, σ/G, aligned with the applied magnetic field. Part (a) and (b) correspond particle chains parallel (**h** $\parallel$ **N**) and perpendicular (**h** $\perp$ **N**), respectively, to the applied magnetic field.*

absolute value with the magnitude of the nondimensional preloads. Interestingly, the magnetostriction response is not symmetric with respect to the sign of the prestress in Fig. 2a. Notice that the sample expands ($\Delta\varepsilon > 0$) for zero or negative prestresses and contracts for adequately large tensile prestresses. This asymmetry implies a strong nonlinear effect of the applied prestress on the resulting magnetostriction. In turn, Fig. 2b, shows the strong influence of the particle-chain orientation upon the overall magnetization and magnetostriction of the MRE specimens. In order to unfold the corresponding underlying microstructural deformation mechanisms leading to the pronounced prestress and particle-chain orientation effects, FEM full-field calculations are also carried out (not shown here for brevity).

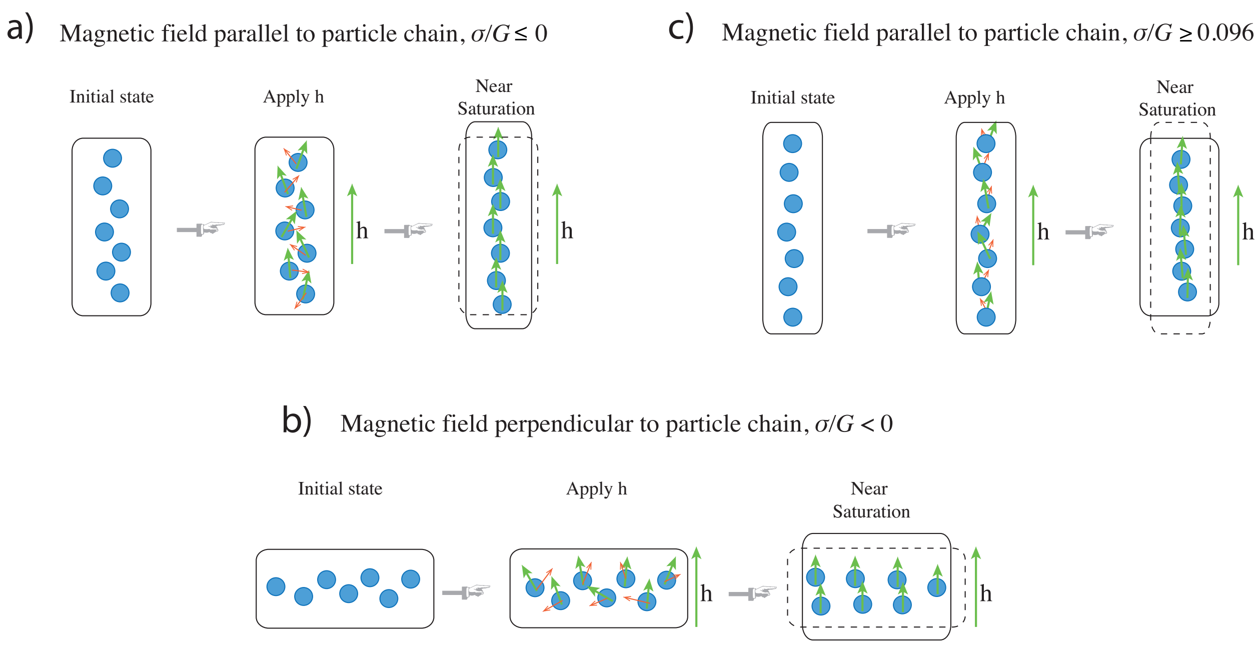

FIG. 3 – *Deformation micromechanism explaining the influence on magnetostriction of particle chain orientation and prestresses σ/G (with G denoting the shear modulus of the MRE). The direction of the large (green-color) arrows indicates the direction of effective magnetic dipoles, i.e., from south to north pole. The small (red-color) arrows indicate the direction of motion of the particle due to the magnetic forces.*

In an attempt to explain the magnetostrictive behavior depicted in Fig. 2 and particularly the counterintuitive results for the magnetostriction (e.g., $\Delta\varepsilon > 0$ for no prestress), Danas et al. (2012) have proposed a mechanism, sketched in Fig. 3, which could explain the previously observed responses. The proposed local deformation mechanism to be detailed in the following has its roots in the original work of Klingenberg and Zukoski (1990) (in the context of electrorheological suspensions) and Lemaire and Bossis (1991) (in the context of magnetic suspensions), who found that between a pair of particles subjected to a magnetic (or electric) field, there exists a restoring force which is, in general, non-aligned to the applied magnetic (or electric) field and tends to align the particles with the applied magnetic (or electric) field so that they form magnetic (or electric) dipoles. Similar observations have also been made in the more recent work of Borcea and Bruno (2001) in the context of two-particle magnetostatic systems at small strain as well as in Diguet et al. (2009) in the context of isotropic systems (who used a dipole-to-dipole particle interaction instead of a continuum model for the finite-sized particles to model extension of the MRE during application of magnetic fields).

As sketched in Fig. 3a for $\sigma/G \leq 0$, the particles are taken to be somewhat aligned in a staggered configuration. However, it is important that we do not allow for a perfect alignment of the particles in accord with the electron micrograph shown in Fig. 1. Then, by application of the field **h** parallel to the particle chain, the particles become magnetic dipoles with effective magnetization direction indicated by the large (green-color) arrows that tend to align themselves with the externally applied magnetic field. The optimal configuration would be the one that the south magnetic pole of a particle on top approaches the north magnetic pole of the particle below. In order to achieve such a configuration, the particles must move in a direction almost perpendicular to **h**, as indicated by the small (red-color) arrows in Fig. 3a.

This interparticle motion leads to a contraction in the direction normal to **h** and consequently due to matrix incompressibility to an overall extension of the MRE along **h**. Similarly in Fig. 3b, when the particle chain is perpendicular to the applied magnetic field **h** , the repulsive forces between the neighboring particles are even stronger than in the parallel case and hence lead to an even higher overall magnetostriction. In contrast, in Fig. 3c, due to the adequately large positive prestress, the interparticle distance increases and attractive forces between particles appear now in the direction of the applied magnetic field, leading to an overall compressive magnetostriction.

Theoretical predictions, based on the energy density of equation (6) are compared to experimental results in the case of simple shear loading and a uniaxial stress test in the direction of a magnetic field which is (i) aligned with the particle chains and (ii) perpendicular to the particle chains (not shown here for brevity). In all three cases, The model gives an excellent agreement with experiments for relatively moderate magnetic fields but has also been satisfactorily extended to include magnetic fields near saturation (see detailed discussion in Danas et al. (2012)).

4 CONCLUSIONS AND FUTURE WORK

The present combined experimental and theoretical investigation of MREs subjected to coupled mechanical and magnetic loading reveals the many challenging features of these materials. This study shows the adequacy of the anisotropic, finite strain continuum formulation for the description of these materials, while at the same time it demonstrates the importance of microgeometry in the macroscopic magnetoelastic coupling response of the composite. Given the need in applications to produce MREs with strong magnetoelastic coupling, it is desirable to build a) microscopic models to study these coupling mechanisms in detail and b) mean-field (i.e., homogenization) models to investigate more efficiently the influence of matrix properties, particle distribution and shape on the macroscopic magnetomechanical response of these composites. On the practical side, mean field theories are a valuable tool to optimize coupling properties (e.g., Galipeau and Ponte Castañeda (2012); Kuo et al. (2010)) in these materials. Studies in these directions are currently under way by the authors.

REFERENCES

Borcea, L. and Bruno, O., 2001. On the Magneto-elastic Properties of Elastomer-ferromagnetic Composites. *J. Mechanics Physics Solids*, 49, 2877-2919.

Brenner R., 2010. Computational approach for composite materials with coupled constitutive laws. *Z. Angew. Math. Phys.*, 61, 919–927.

Brown, W. F., 1966. Magnetoelastic Interactions. Springer-Verlag, New York.

Carlson, J. D., Jolly, M. R., 2000. MR Fluid, Foam and Elastomer Devices. *Mechatronics*, 10, 555–569.

Coquelle, E., Bossis, G., 2006. Mullins effect in elastomers filled with particles aligned by a magnetic field. *Int. J. Solids Struc. 43*, 7659 – 7672.

Corcolle, R., Daniel, L., Bouillault, F., 2009. Intraphase fluctuations in heterogeneous magnetic materials, *Journal of Applied Physics* 105(12), 123913.

Danas, K., Kankanala, S.V., Triantafyllidis, N., 2012, Experiments and modeling of iron-particle-filled magnetorheological elastomers, *J. Mech. Phys. Solids* 60, 120 – 138.

Daniel, L., Hubert, O., Buiron, N., Billardon R., 2008. Reversible magneto-elastic behavior: a multiscale approach, *J. Mech. Phys. Solids* 56(3), 1018–1042.

Diguet, G., Beaugnon, E., Cavaillé, J., 2009. From dipolar interactions of a random distribution of ferromagnetic particles to magnetostriction. *J. Magn. Magn. Mater.* 321, 396–401.

Diguet, G., Beaugnon, E., Cavaillé, J., 2010. Shape effect in the magnetostriction of ferromagnetic composite. *J. Magn. Magn. Mater.* 322, 3337–3341.

Dorfmann, A., Ogden, R.W., 2003, Magnetoelastic modelling of elastomers, *Eur. J. Mech. A/Solids 22*, 497–507.

Galipeau, E., Ponte Castañeda, P., 2012, The effect of particle shape and distribution on the macroscopic behavior of magnetoelastic composites, *Int. J. Solids Struct.* 49, 1 – 17.

Ginder, J., Nichols, M., Elie, L., Tardiff, J., 1999. Magnetorheological elastomers: Magnetorheological elastomers: properties and applications, *Smart Structures and Materials 1999: Smart Materials Technologies Ed. by M. Wuttig, Proc. of SPIE*,3675, 131–138.
Hirsinger, L., Billardon, R., 1995. Magneto-elastic Finite Element Analysis Including Magnetic Forces and Magnetostriction Effects. IEEE Transactions on Magnetics, 31, 1877–1880.

Hubert, O., Daniel, L., Billardon R., 2003. Experimental analysis of the magnetoelastic anisotropy of a non-oriented silicon iron alloy, *Journal of Magnetism and Magnetic Materials* 254, 352–354.

Huang, L., Mandeville, R. E. and Rolph III, W. D., 1999. Magnetostatics and Coupled Structural Finite Element Analysis. *Computers and Structures*, 72, 199–207.

James, R., Kinderlehrer, D., 1993. Theory of magnetostriction with applications to tbxdy1xfe2. *Phil. Mag. B* 68, 237–274.

Kankanala, S.V., Triantafyllidis, N., 2004, On finitely strained magnetorheological elastomers, *J. Mech. Phys. Solids* 52, 2869 – 2908.

Klingenberg, D.J., Zukoski, C.F., 1990. Studies on the steady-shear behavior of electrorheological suspensions. *Langmuir* 6, 15Ð24.

Kuo, H.Y., Slinger, A., Bhattacharya, K., 2010. Optimization of magnetoelectricity in piezoelectric–magnetostrictive bilayers. *Smart Materials and Structures* **19**, pp. 125010.

Lemaire, E., Bossis, G., 1991. Yield stress and wall effects in magnetic colloidal suspensions. *J. Phys. D: App. Phys.* 24, 1473.

Maugin, G.A., Eringen, A.C., 1972. Deformable magnetically saturated media. i. field equations. *J. Math. Phys.* 13, 143–155.

Moffett, M.B., Clark, A.E., Wun-Fogle, M., Linberg, J., Teter, J. P., McLaughlin, E.A., 1991, Characterization of Terfenol-D for magnetostrictive transducers, *J. Acoust. Soc. America* 89, 1448 – 1455.

Ponte Castañeda, P., Galipeau, E., 2011. Homogenization-based constitutive models for magnetorheological elastomers at finite strain. *J. Mech. Phys. Solids*, 59, 194–215.

Ren, Z., Ionescu, B., Besbes, M., and Razek, A., 1995. Calculation of Mechanical Deformation of Magnetic Materials in Electromagnetic Devices. *IEEE Transactions on Magnetics*, 31, 1873–876.

Rigbi, Z., Jilkén, L., 1983. The response of an elastomer filled with soft ferrite to mechanical and magnetic influences. *J. Magn. Magn. Mater.* 37, 267–276.

Thomas, J.D., Triantafyllidis, N., 2009. On electromagnetic forming processes in finitely strained solids: Theory and examples. *J. Mech. Phys. Solids* 55, 2863–2873.