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Micromechanics-based hyperelastic constitutive modeling of magnetostrictive particle-filled elastomers

H.M. Yin [a], L.Z. Sun [a,*], J.S. Chen [b]

[a] *Department of Civil and Environmental Engineering and Center for Computer-Aided Design, The University of Iowa, Iowa City, IA 52242-1527, USA*

[b] *Department of Civil and Environmental Engineering, University of California, Los Angeles, CA 90095-1593, USA*

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Abstract

An effective hyperelastic constitutive model is developed for particle-filled elastomer composites based on the microstructural deformation and physical mechanism of the magnetostrictive particles embedded in the hyperelastic elastomer matrix. Two types of loading conditions are considered—magnetic field and mechanical load. Magnetic eigenstrains are prescribed on the particles due to effect of magnetostriction. The effective constitutive relation of the composites during infinitesimal deformation can be established based on Eshelby's micromechanics approach. Since the elastomers normally exhibit finite hyperelastic deformation, the corresponding hyperelastic constitutive law of the composites is constructed in terms of the strain energy densities of the constituents.

Keywords: Elastomer composites; Magnetostriction; Micromechanics; Hyperelasticity; Constitutive relations; Strain energy

1. Introduction

Magnetostriction is a phenomenon that, when a substance is exposed to a magnetic field, its dimensions change (Cullity, 1972; Jiles, 1991; Tremolet de Lacheisserie, 1993). The study of magnetostriction can be traced back to James P. Joule, who first discovered the magnetostriction phenomenon of iron in 1842. Since then, a number of experimental and theoretical works have been done for magnetostrictive homogeneous materials. In recent years, magnetostrictive particle-reinforced heterogeneous composites have attracted much interest in automotive applications such as intelligent devices (Pinkerton et al., 1997; Bednarek, 1999; Carlson and Jolly, 2000) and field-dependent elastomeric components (Ginder et al., 1999, 2000, 2001). Corresponding mathematical models have been developed. For example, Herbst et al. (1997) studied $SmFe_2/Al$ and $SmFe_2/Fe$ composites and proposed a single-sphere model to predict the overall magnetostriction of the composites. Nan (1998) and Nan and Weng (1999) developed an analytical model based on Green's function technique. Davis (1999) used finite element method to analyze the dependence of effective shear modulus of magnetorheological elastomers

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[*] Corresponding author. Tel.: +1-319-384-0830; fax: +1-319-335-5660.

E-mail address: lizhi-sun@uiowa.edu (L.Z. Sun).

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on interparticle magnetic forces. Chen et al. (1999) investigated the effect of the elastic modulus of the matrix on magnetostriction of the composite. Armstrong (2000) presented an analysis of magnetoelastic behavior of dilute magnetostrictive particle composites. It is noted that all these models are limited to infinitesimal strain problems. When the matrix of a magnetostrictive composite is selected from elastic materials such as elastomers or silicones, the deformation of the matrix can be large under loading. Therefore, mechanisms of finite deformation in the matrix should be included in the constitutive relations of magnetostrictive elastomer composites.

Elastomers filled with nonmagnetostrictive particles (such as carbon black) have been studied in terms of their finite hyperelastic behavior. Mullins and Tobin (1965) provided an estimate of effective deformation for carbon-black filled vulcanized rubbers by amplifying the elastic modulus from the composition of matrix and reinforcement. Following Mullins–Tobin's method, Bergstrom and Boyce (1999) used neo-Hookean relation in the rubber to set the ratio of the averaged strain to the measured overall strain, and then obtained a good comparison with experimental data. On the other hand, Govindjee and Simo (1991) investigated the microstructure of the filled elastomers and derived a continuum model that can capture the strain-induced stress softening Mullins effect (cf., Mullins, 1969) during the large deformation. It is noted that these models can only be applied to the particle-filled composites composed of a nonlinear hyperelastic matrix and rigid particles.

In fact, the deformation mechanism of reinforcement has a considerable effect on particle-filled elastomers. Recently, Bednarek (1999) found that magnetostrictive particle-filled elastomers show some unique mechanical properties under the magnetic field. Lanotte et al. (2001) further discussed the deformation mechanism of magnetomechanical coupling for the magnetostrictive composites. In this paper, a micromechanical framework is proposed to investigate the effective mechanical behavior of magnetostrictive particle-filled elastomer composites. Under infinitesimal deformation, the magnetostriction in the particle is treated as a prescribed eigenstrains, and the overall stress–strain relationship is derived by the Eshelby's equivalent inclusion method (Eshelby, 1957). For finite deformation, a strain energy density function of the elastomer composites is derived based on Saint Venant–Kirchhoff assumption (Ciarlet, 1988; Belytschko et al., 2000) and the nonGaussian behavior (Treloar, 1975) of materials. The corresponding hyperelastic constitutive law of the composites is constructed in terms of the invariant-based strain energy which combines the contributions from both the particles and elastomer matrix.

The outline of this paper is as follows. In Section 2, we first established the microstructure of the particle-filled composites. Following Eshelby's equivalent inclusion method, we investigated the eight-particle interaction and obtained the averaged eigenstrain. By using the homogeneous stress boundary condition, we derived the effective elastic constitutive relation of the composites. Considering the magnetostriction as a prescribed eigenstrain, we further obtained the effective magnetostrictive deformation of the composites. Comparisons with the available experimental data and existing models were made. In Section 3, we evaluated the effective free energy in the magnetostrictive composites. We employed the concept of nonGaussian chain in the microstructure so that all particles are connected by the chains. We established the deformation relation between the stretch in elastomer and the total stretch in the particles. We further derived the energy densities in the particles and elastomer, respectively, in order to develop the energy-based hyperelastic constitutive relation of the composite. Finally, in Section 4, we presented numerical results and comparisons based on the proposed model.

2. Linear elastic deformation model

Let us consider a two-phase composite consisting of an elastomer matrix (phase 0) and ferromagnetic spherical particles (phase 1). When the deformation is infinitesimal upon loading, the matrix and the particles can be treated as linearly elastic materials with different elastic constants. For the problem of one particle embedded in an infinite domain, Eshelby (1957) offered an analytical solution through an equivalent inclusion method in which the total domain is assumed to be the same material as the matrix but an eigenstrain is introduced in the particle domain to represent the inhomogeneity. This method has been widely applied in evaluating the effective elastic properties of composites (e.g., Mura, 1987; Nemat-Nasser and Hori, 1999). However, the direct interaction among particles has been ignored since it is impossible to obtain an exact solution.

To account for particle interaction, an approximate treatment is considered here. Let us assume that the matrix and particle have the isotropic elasticity tensors as $\mathbf{C}_0$ and $\mathbf{C}_1$, respectively. Moreover, for any given particle, only the interaction with its eight neighboring particles with the same radius is considered, as shown in Fig. 1.

2.1. Equivalent eigenstrains

Consider the existence of a uniform far-field strain loading $\boldsymbol{\varepsilon}^\infty$. According to Eshelby's equivalent inclusion method, at point $\mathbf{r}$ inside the particle Ω_0 (Fig. 1), the perturbed strain $\boldsymbol{\varepsilon}'(\mathbf{r})$ induced by the inhomogeneity can be related to *equivalent* eigenstrain $\boldsymbol{\varepsilon}^*(\mathbf{r})$ by replacing the particles with the matrix material, following (Ju and Sun, 1999)

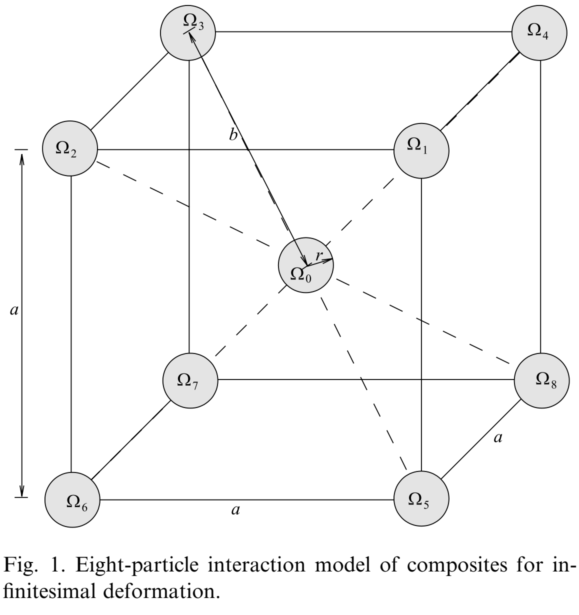


Fig. 1. Eight-particle interaction model of composites for infinitesimal deformation.

$$\mathbf{C}_1 : [\boldsymbol{\varepsilon}^\infty + \boldsymbol{\varepsilon}'(\mathbf{r}) - \boldsymbol{\varepsilon}^{\mathrm{pe}}(\mathbf{r})] = \mathbf{C}_0 : [\boldsymbol{\varepsilon}^\infty + \boldsymbol{\varepsilon}'(\mathbf{r}) - \boldsymbol{\varepsilon}^*(\mathbf{r})], \quad \mathbf{r} \in \Omega_0 \tag{1}$$

where $\boldsymbol{\varepsilon}^{\mathrm{pe}}(\mathbf{r})$ represents the *prescribed* magnetostrictive eigenstrain and Ω_0 designates the domain of particle being considered (see Fig. 1). The symbol ":" denotes the contraction between the fourth-rank tensor and the second-rank tensor. In addition, the perturbed strain $\boldsymbol{\varepsilon}'(\mathbf{r})$ can be derived as

$$\boldsymbol{\varepsilon}'(\mathbf{r}) = \mathbf{S} : \boldsymbol{\varepsilon}^*(\mathbf{r}) + \sum_{i=1}^{n} \int_{\Omega_i} \mathbf{G}(\mathbf{r} - \mathbf{r}') : \boldsymbol{\varepsilon}^*(\mathbf{r}')\,\mathrm{d}\mathbf{r}' \tag{2}$$

where $\mathbf{S}$ is the Eshelby's tensor, and n is the total number of particles surrounded to the considered particle Ω_0. It is noted that the first term of right hand side of the above equation demonstrates the perturbation contribution from the particle itself (Ω_0) while the second term represents the effects from material points $\mathbf{r}'$ in the neighboring particles ($\sum_{i=1}^{n} \Omega_i$). The fourth-rank Green function $\mathbf{G}$ reads

$$\begin{aligned} G_{ijkl}(\mathbf{r} - \mathbf{r}') = \frac{1}{8\pi(1 - \nu_0)\bar{r}^3} [&(1 - 2\nu_0) \\ &\times (\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk} - \delta_{ij}\delta_{kl}) \\ &+ 3\nu_0(\delta_{ik}n_j n_l + \delta_{il}n_j n_k + \delta_{jk}n_i n_l \\ &+ \delta_{jl}n_i n_k) + 3\delta_{ij}n_k n_l \\ &+ 3(1 - 2\nu_0)\delta_{kl}n_i n_j - 15 n_i n_j n_k n_l] \end{aligned} \tag{3}$$

with $\bar{r} = \|\mathbf{r} - \mathbf{r}'\|$, $\mathbf{n} = (\mathbf{r} - \mathbf{r}')/\|\mathbf{r} - \mathbf{r}'\|$, and ν_0 is the Poisson ratio of the matrix.

As an approximation, only the closest eight neighboring particles ($\Omega_i, i = 1, 2, \ldots, 8$) are considered to interact with the particle Ω_0. Substitution of Eq. (2) into Eq. (1) yields

$$\begin{aligned} &-\mathbf{A} : \boldsymbol{\varepsilon}^*(\mathbf{r}) + \mathbf{B} : \boldsymbol{\varepsilon}^{\mathrm{pe}}(\mathbf{r}) \\ &\quad = \boldsymbol{\varepsilon}^\infty + \mathbf{S} : \boldsymbol{\varepsilon}^*(\mathbf{r}) + \sum_{i=1}^{8} \int_{\Omega_i} \mathbf{G}(\mathbf{r} - \mathbf{r}') : \boldsymbol{\varepsilon}^*(\mathbf{r}')\,\mathrm{d}\mathbf{r}' \end{aligned} \tag{4}$$

where the fourth-rank elastic-phase "mismatch tensors" $\mathbf{A} = (\mathbf{C}_1 - \mathbf{C}_0)^{-1} \cdot \mathbf{C}_0$ and $\mathbf{B} = (\mathbf{C}_1 - \mathbf{C}_0)^{-1} \cdot \mathbf{C}_1$.

Performing the volume average on Eq. (4) and considering the same eigenstrains in each particle, we obtain the following equation

$$-\mathbf{A}:\langle\boldsymbol{\varepsilon}^*\rangle_{\Omega_0}+\mathbf{B}:\langle\boldsymbol{\varepsilon}^{\mathrm{pe}}\rangle_{\Omega_0}=\boldsymbol{\varepsilon}^\infty+\mathbf{S}:\langle\boldsymbol{\varepsilon}^*\rangle_{\Omega_0}+\mathbf{g}:\langle\boldsymbol{\varepsilon}^*\rangle_{\Omega_0} \tag{5}$$

where the symbol $\langle\cdot\rangle_{\Omega_0}$ denotes the volume average in Ω_0 and the interaction term

$$\mathbf{g}=\frac{1}{\Omega_0}\sum_{i=1}^{8}\int_{\Omega_0}\int_{\Omega_i}\mathbf{G}(\mathbf{r}-\mathbf{r}')\,\mathrm{d}\mathbf{r}'\,\mathrm{d}\mathbf{r} \tag{6}$$

The above equation can be further simplified as

$$g_{ijkl}=\frac{8\rho^3(14\rho^2-5)}{45(1-\nu_0)}\times\left[(1-5\delta_{IK})\delta_{ij}\delta_{kl}+\delta_{ik}\delta_{jl}+\delta_{il}\delta_{jk}\right] \tag{7}$$

with $\rho=r/b$ where r signifies the radius of particles and b denotes the center-to-center spacing between particle Ω_0 and its neighbor particle Ω_i ($i=1,2,\ldots 8$). It is noted that Mura's (1987) tensorial indicial notation is followed in the above equation; i.e., upper-case indices has the same representation as the corresponding lower-case ones but are not summed (Ju and Sun, 2001; Sun and Ju, 2001).

Through lengthy but straightforward derivation, the averaged eigenstrain can be obtained as

$$\langle\boldsymbol{\varepsilon}^*\rangle_{\Omega_0}=-\Gamma:\left[\boldsymbol{\varepsilon}^\infty-\mathbf{B}:\langle\boldsymbol{\varepsilon}^{\mathrm{pe}}\rangle_{\Omega_0}\right] \tag{8}$$

where

$$\Gamma_{ijkl}=\left(\frac{-m}{(2w-M)(2w+3m-M)}+\frac{M}{2w(2w-M)}\delta_{IK}\right)\delta_{ij}\delta_{kl}+\frac{1}{4w}\left(\delta_{ik}\delta_{jl}+\delta_{il}\delta_{jk}\right) \tag{9}$$

with

$$m=\frac{1}{30(1-\nu_0)}\alpha+\frac{M}{5}$$

$$w=\frac{1}{30(1-\nu_0)}\beta+\frac{M}{5},\quad M=\frac{40(14\rho^2-5)\rho^3}{45(1-\nu_0)}$$

$$\alpha=10(1-\nu_0)\left(\frac{K_0}{K_1-K_0}-\frac{\mu_0}{\mu_1-\mu_0}\right)+2(5\nu_0-1)$$

$$\beta=15(1-\nu_0)\frac{\mu_0}{\mu_1-\mu_0}+2(4-5\nu_0)$$

It is noted that K_0, K_1 are the bulk moduli of the matrix and particle, respectively, and μ_0, μ_1 are their shear moduli.

From the geometrical setting of the composite, the volume fraction of particles ϕ can be calculated as

$$\phi=\frac{\frac{8}{3}\pi r^3}{\left(\frac{2b}{\sqrt{3}}\right)^3}=5.4414\rho^3 \tag{10}$$

Furthermore, since the eigenstrains are the same for all particles, Ω_0 is replaced by Ω in following sections for simplicity.

2.2. *Effective elasticity of composites*

The overall (effective) stress tensor of the composites is defined as

$$\langle\boldsymbol{\sigma}\rangle_V=\frac{1}{V}\int_V\boldsymbol{\sigma}(\mathbf{r})\,\mathrm{d}\mathbf{r} \tag{11}$$

Considering the boundary condition in the far field $\boldsymbol{\varepsilon}^\infty$, the averaged stress can be further expressed as (Mura, 1987)

$$\langle\sigma\rangle_V=\mathbf{C}_0:\boldsymbol{\varepsilon}^\infty \tag{12}$$

On the other hand, the local strain field of composites can be expressed in terms of local stress by using Eshelby's method (Ju and Sun, 1999)

$$\boldsymbol{\varepsilon}(\mathbf{r})=\begin{cases}\mathbf{C}_0^{-1}:\boldsymbol{\sigma}(\mathbf{r})+\boldsymbol{\varepsilon}^*(\mathbf{r}), & \text{in } V_{\mathrm{p}} \text{ (particle domain)}\\ \mathbf{C}_0^{-1}:\boldsymbol{\sigma}(\mathbf{r}), & \text{in } V_{\mathrm{m}} \text{ (matrix domain)}\end{cases} \tag{13}$$

Therefore, the averaged strain tensors over the particle domain Ω and the whole composite domain V can be written, respectively, as

$$\langle\boldsymbol{\varepsilon}\rangle_\Omega=\boldsymbol{\varepsilon}^\infty+(\mathbf{S}+\mathbf{g}):\langle\boldsymbol{\varepsilon}^*\rangle_\Omega \tag{14}$$

and

$$\langle\boldsymbol{\varepsilon}\rangle_V=\boldsymbol{\varepsilon}^\infty+\phi\langle\boldsymbol{\varepsilon}^*\rangle_\Omega \tag{15}$$

Combining Eqs. (15) and (8), we obtain

$$\boldsymbol{\varepsilon}^\infty=(\mathbf{I}-\phi\boldsymbol{\Gamma})^{-1}:\left(\langle\boldsymbol{\varepsilon}\rangle_V-\phi\boldsymbol{\Gamma}:\mathbf{B}:\langle\boldsymbol{\varepsilon}^{\mathrm{pe}}\rangle_\Omega\right) \tag{16}$$

In the absence of prescribed magnetostrictive eigenstrain ($\boldsymbol{\varepsilon}^{\mathrm{pe}}=0$), the effective strain and stress of the composites can be simplified as

$$\begin{aligned}\langle \boldsymbol{\varepsilon}\rangle_V &= (\mathbf{I}-\phi\boldsymbol{\Gamma})\cdot\boldsymbol{\Gamma}^{-1}\cdot\mathbf{A}^{-1} : \langle\boldsymbol{\varepsilon}\rangle_\Omega \\ \langle\sigma\rangle_V &= \mathbf{C}_0 : (\mathbf{I}-\phi\boldsymbol{\Gamma})^{-1} : \langle\boldsymbol{\varepsilon}\rangle_V\end{aligned} \tag{17}$$

The corresponding effective stiffness tensor of the composite reads

$$\overline{\mathbf{C}} = \mathbf{C}_0 : (\mathbf{I}-\phi\boldsymbol{\Gamma})^{-1} \tag{18}$$

It is noted that the above stiffness tensor of the composites is obtained by considering the direct particle interactions. This result recovers Nemat-Nasser and Hori's (1999) formula (Eq. 8.1.8) if the interaction term is ignored.

When the composites are subjected only to magnetostrictive eigenstrain ($\boldsymbol{\varepsilon}^\infty = 0$), the averaged strains $\langle\boldsymbol{\varepsilon}\rangle_\Omega$ and $\langle\boldsymbol{\varepsilon}\rangle_V$ are then shown as

$$\langle\boldsymbol{\varepsilon}\rangle_\Omega = (\mathbf{S}+\mathbf{g}) : \boldsymbol{\Gamma} : \mathbf{B} : \langle\boldsymbol{\varepsilon}^{\mathrm{pe}}\rangle_\Omega \tag{19}$$

and

$$\langle\boldsymbol{\varepsilon}\rangle_V = \phi\boldsymbol{\Gamma} : \mathbf{B} : \langle\boldsymbol{\varepsilon}^{\mathrm{pe}}\rangle_\Omega \tag{20}$$

2.3. *Effective magnetostriction of composites*

Magnetostriction is measured by the fractional change in length $\lambda = \Delta l/l$ (Clark, 1980). The value of λ measured at magnetic saturation is called the saturation magnetostriction λ^{s}. For the isotropic, homogenous magnetostrictive material, the deformation due to saturated magnetostriction has the form of

$$\lambda_{\mathrm{si}}(\theta) = \tfrac{3}{2}\lambda^{\mathrm{s}}(\cos^2\theta - \tfrac{1}{3}) \tag{21}$$

where θ denotes the angle between the magnetization direction and the measurement direction.

For magnetostrictive composites, for simplicity, let us assume that the magnetization direction follows one of the three principal directions of the cube, say x_1-direction. The magnetostrictive eigenstrains are

$$\varepsilon^{\mathrm{pe}}_{11} = \lambda^{\mathrm{s}};\quad \varepsilon^{\mathrm{pe}}_{22} = \varepsilon^{\mathrm{pe}}_{33} = -\tfrac{1}{2}\lambda^{\mathrm{s}};\quad \varepsilon^{\mathrm{pe}}_{ij} = 0 \quad \text{for } i \neq j \tag{22}$$

From Eq. (20), the effective magnetostriction of the composites can be calculated as

$$\langle\varepsilon_{ij}\rangle_V = \frac{\phi}{2w-M}\,\frac{\mu_1}{\mu_1-\mu_0}\,\varepsilon^{\mathrm{pe}}_{II}\delta_{ij} \tag{23}$$

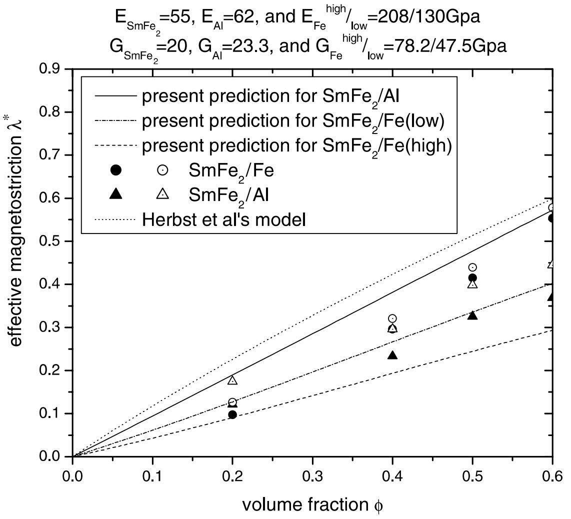


Fig. 2. Comparisons of the effective magnetostriction with the experimental data and Herbst et al.'s model (Herbst et al., 1997).

or the normalized effective magnetostriction of the composite reads

$$\overline{\lambda}^* = \frac{\phi}{2w-M}\,\frac{\mu_1}{\mu_1-\mu_0} \tag{24}$$

Fig. 2 shows the comparison between the above prediction and the reported results of magnetostriction for SmFe$_2$/Al and SmFe$_2$/Fe composites by Herbst et al. (1997). The material constants used are obtained from Herbst et al.'s paper; i.e., elastic Young's moduli for SmFe$_2$, Al and Fe are 55, 62, and 208/130 GPa, respectively. Elastic shear moduli for SmFe$_2$, Al and Fe are 20, 23.3, and 78.2/47.5 GPa, respectively. It is shown that good agreement between the present prediction and experimental data is obtained. It is further illustrated that the present model accounts for both the volume fraction of particles and elastic constants of matrix and reinforcement, whereas Herbst et al.'s analytical model is not sensitive to those material constants.

3. Finite-deformation model

Magnetostrictive particle-reinforced elastomers may display finite, nonlinear deformation under loading. Many hyperelastic constitutive models for homogeneous elastomers are developed in terms of invariant-based strain energy function (see Treloar, 1975), including phenomenological models such as neo-Hookean, Mooney-Rivlin and Ogden. Statistical mechanics offers another way to explain deformation mechanisms of elastomers from the concept of molecular chains. It is shown that the Gaussian chain theory yields the same strain energy density as neo-Hookean model (Treloar, 1975). To study the finite stretch problem, investigators have built networks based on the non-Gaussian chain theory.

For magnetostrictive elastomers, although the elastomer matrix undergoes large deformation, particle reinforcement may still be deformed in the range of infinitesimal strain since the reinforcement is usually much stiffer than the elastomer matrix. In the present model, the nonGaussian chain theory is used to evaluate the energy density in the matrix while infinitesimal deformation theory is adopted to derive the energy density in the particles.

3.1. Geometric analysis

Elastomeric materials are chemically made up of long-chain molecules with a network of freely rotating links that form a network. When particles are embedded in, the chain network will be broken and most of chains will re-connect to particles to form the main network of the composites as microstructurally illustrated in Fig. 3. For simplicity, elastomer chains are assumed to link the central particle to eight neighboring particles. Motivated by Arruda and Boyce's (1993) assertion, the composites considered are always stretched in the principal frame, as described by the three principal stretches λ_1, λ_2 and λ_3.

As shown in Fig. 4, the deformed vector **b** pointing from the center of central particle to its neighboring particle is expressed as

$$\mathbf{b} = \frac{b}{\sqrt{3}}(\lambda_1\mathbf{i} + \lambda_2\mathbf{j} + \lambda_3\mathbf{k}) \tag{25}$$

Deformation vector **r** inside the particle is similarly shown in terms of the three principal stretches $\hat{\lambda}_1$, $\hat{\lambda}_2$, $\hat{\lambda}_3$ of the particle

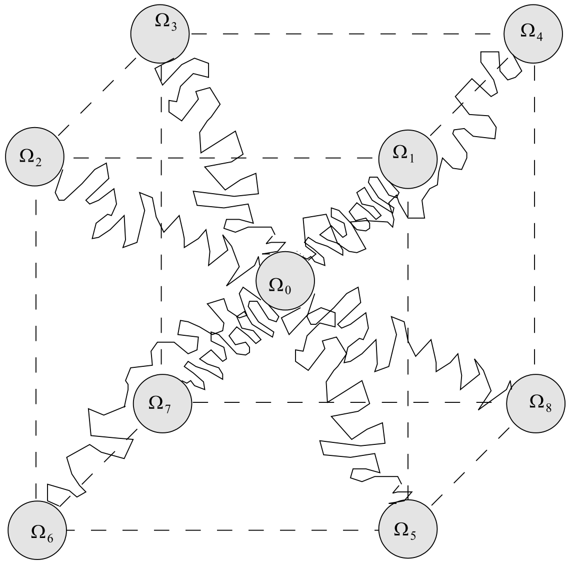


Fig. 3. Chain network between magnetostrictive particles embedded in the elastomer matrix.

$$\mathbf{r} = \frac{r}{\sqrt{3}}(\hat{\lambda}_1\mathbf{i} + \hat{\lambda}_2\mathbf{j} + \hat{\lambda}_3\mathbf{k}) \tag{26}$$

Moreover, the deformed vector **c** along the same direction with the vector **b** has the relationship with the principal stretches $\tilde{\lambda}_1$, $\tilde{\lambda}_2$, $\tilde{\lambda}_3$ of the matrix as

$$\mathbf{c} = \frac{b-2r}{\sqrt{3}}(\tilde{\lambda}_1\mathbf{i} + \tilde{\lambda}_2\mathbf{j} + \tilde{\lambda}_3\mathbf{k}) \tag{27}$$

From the geometric setting $\mathbf{b} = \mathbf{c} + 2\mathbf{r}$, the following relationship among the principal stretches can be derived

$$\tilde{\lambda}_i = \frac{\lambda_i - 2\rho\hat{\lambda}_i}{1-2\rho} \tag{28}$$

3.2. Effective free energy and hyperelastic constitutive law

For magnetostrictive particle-filled elastomers, the effective deformation is induced by magnetostriction in particle and external mechanical loading. To yield the maximum effect of the magnetostriction in the composites, the prescribed magnetic eigenstrains should be consistent with the principal stretch directions of the composites. The

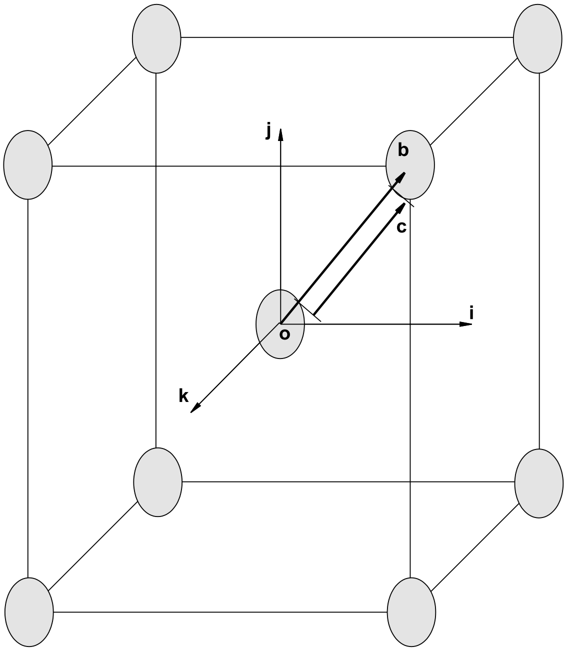


Fig. 4. Eight-particle interaction model of composites for finite deformation.

particle magnetostriction eigenstrain tensor is denoted as

$$\mathbf{E}^{\mathrm{pe}} = \begin{bmatrix} \lambda_1^{\mathrm{s}} - 1 & 0 & 0 \\ 0 & \lambda_2^{\mathrm{s}} - 1 & 0 \\ 0 & 0 & \lambda_3^{\mathrm{s}} - 1 \end{bmatrix} \tag{29}$$

The averaged strain tensor of particles due to magnetostriction is further obtained from Eq. (19) as

$$\widehat{\mathbf{E}}^0 = (\mathbf{S} + \mathbf{g}) \cdot \boldsymbol{\Gamma} \cdot \mathbf{B} : \mathbf{E}^{\mathrm{pe}} \tag{30}$$

and, from Eq. (20), the corresponding effective strain tensor of the composites reads

$$\mathbf{E}^0 = \phi \boldsymbol{\Gamma} \cdot \mathbf{B} : \mathbf{E}^{\mathrm{pe}} \tag{31}$$

Upon external mechanical loading, the total stretches of the composites are considered as finite deformation. The deformation gradient of the composites **F** in the principal frame can be written as

$$F_{ij} = \lambda_{\mathrm{I}} \delta_{ij} \tag{32}$$

Based on Saint Venant–Kirchhoff assumption on hyperelastic materials (see, e.g., Ciarlet, 1988; Belytschko et al., 2000), the relationship between the deformation rate $\widehat{\mathbf{D}}$ of particles and the finite-deformation rate **D** of the composites can be established from infinitesimal theory (Eq. (17)) as

$$\widehat{\mathbf{D}} = \mathbf{A} \cdot \boldsymbol{\Gamma} \cdot (\mathbf{I} - \phi \boldsymbol{\Gamma})^{-1} : \mathbf{D} \tag{33}$$

where

$$D_{ij} = \dot{F}_{ik} F_{kj}^{-1} = \frac{\dot{\lambda}_{\mathrm{I}}}{\lambda_{\mathrm{I}}} \delta_{ij} \tag{34}$$

Through simplification, Eq. (33) can be further expressed as

$$\widehat{D}_{ij} = (\chi_1 D_{II} + \chi_2 D_{mm}) \delta_{ij} \tag{35}$$

where

$$\begin{aligned} \chi_1 &= \frac{1}{2w - M - \phi} \frac{\mu_0}{\mu_1 - \mu_0} \\ \chi_2 &= \frac{1}{3(2w + 3m - M - \phi)} \frac{K_0}{K_1 - K_0} \\ &\quad - \frac{1}{3(2w - M - \phi)} \frac{\mu_0}{\mu_1 - \mu_0} \end{aligned} \tag{36}$$

The Lagrangian strain $\widehat{\mathbf{E}}$ of particles can be derived by integration upon the above equation as

$$\widehat{E}_{ij} = \widehat{E}_{ij}^0 + \left(\chi_1 \ln \frac{\lambda_I}{\lambda_I^0} + \chi_2 \ln \frac{\lambda_1 \lambda_2 \lambda_3}{\lambda_1^0 \lambda_2^0 \lambda_3^0} \right) \delta_{ij} \tag{37}$$

where the stretches with superscripts 0 denote the initial deformation caused by the magnetostriction of particle.

Accordingly, the strain energy of particles can be obtained as

$$\begin{aligned} W_{\mathrm{p}} = {} & \left(\frac{\lambda_{\mathrm{p}}}{2} (\chi_1 + 3\chi_2)^2 + \mu_{\mathrm{p}} (2\chi_1 \chi_2 + 3\chi_2^2) \right) \\ & \times \ln^2 \frac{\lambda_1 \lambda_2 \lambda_3}{\lambda_1^0 \lambda_2^0 \lambda_3^0} + \mu_{\mathrm{p}} \chi_1^2 \left(\ln^2 \frac{\lambda_1}{\lambda_1^0} + \ln^2 \frac{\lambda_2}{\lambda_2^0} + \ln^2 \frac{\lambda_3}{\lambda_3^0} \right) \end{aligned} \tag{38}$$

For the homogeneous elastomer matrix, Arruda and Boyce's (1993) chain model is adopted here to obtain the free energy W_{m} for the matrix, namely

$$W_{\mathrm{m}} = \frac{\mu_0}{6} \int_{\tilde{I}_1^0}^{\tilde{I}_1} \ell^{-1} \left[\frac{\tilde{I}_1^{1/2}}{\sqrt{3N}} \right] \frac{\sqrt{3N}}{\tilde{I}_1^{1/2}} \, \mathrm{d}\tilde{I}_1 \tag{39}$$

where μ_0 is the shear modulus and N is the number of chain segment links of elastomers. The term $\ell^{-1}[\tilde{I}_1^{1/2}/\sqrt{3N}]$ is the inverse Langevin function defined as $\ell(\bullet) = \coth(\bullet) - 1/(\bullet)$. In addition,

$$\widetilde{I}_1 = \widetilde{\lambda}_i \widetilde{\lambda}_i \tag{40}$$

with

$$\tilde{\lambda}_i = \frac{\lambda_i - 2\rho(\widehat{E}_{II} + 1)}{1 - 2\rho} \tag{41}$$

Following Govindjee and Simo (1991), the macroscopic free energy W of the composites is constructed by considering the strain energies from both the particles and the matrix. For the unit cell of composites with total volume V_0 in Fig. 4, the effective strain energy density W can be derived as

$$W = \frac{8(b - 2r)w_{\text{chain}}}{V_0} + \frac{V_{\text{particle}} W_{\text{p}}}{V_0} \tag{42}$$

where w_{chain} is the chain strain energy density per unit length while W_{p} is the strain energy density of the particles. For the elastomer matrix, since the energy density W_{m} per unit volume is related to the chain energy w_{chain} as $W_{\text{m}} = 8bw_{\text{chain}}/V_0$, the total energy of the composites can be further derived as

$$W = (1 - 2\rho)W_{\text{m}} + \phi W_{\text{p}} \tag{43}$$

Once the total energy of the elastomer composites is derived, the hyperelastic stress–strain relations can be developed based on the conventional finite-deformation theory. For example, the effective Cauchy stress σ of the composite has the following dependence on the effective Lagrangian strain $\mathbf{E}$:

$$\sigma_{ij} = \frac{1}{|\mathbf{F}|} F_{ir} F_{js} \frac{\partial W}{\partial E_{rs}} \tag{44}$$

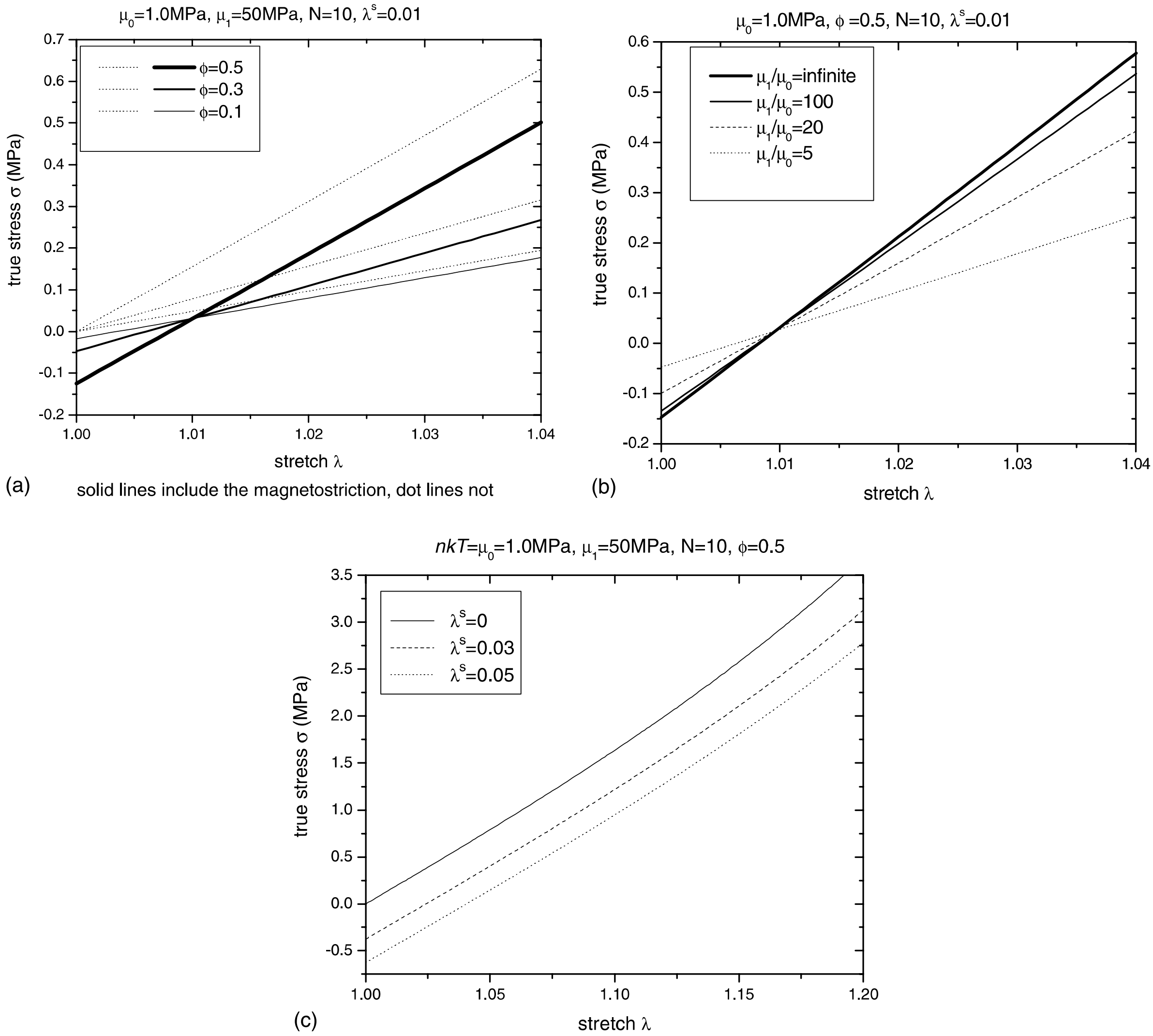
Therefore, a hyperelastic finite-deform constitutive model is developed for magnetostrictive particle-reinforced elastomer composites. This model is formulated in Eqs. (38), (39), (43) and (44) based on a micromechanics approach. The proposed model allows one to estimate the nonlinear hyperelastic stress–strain curves of the magnetostrictive composites.

4. Numerical results and discussion

Stress–stretch curves under uniaxial loading are often referred to as important indicators of the mechanical performance of materials. To illustrate the capability of proposed micromechanics-based model, let us first consider the case of uniaxial stress loading of the composites with a saturated magnetostriction.

Predictions of the proposed method are illustrated in Fig. 5, showing the true stress–stretch curves of magnetostrictive elastomer composites. It is noted that the mechanical uniaxial loading direction is the same as that of the saturated magnetic filed. The selected material constants of the matrix are the shear modulus $\mu_0 = 1$ MPa, the Poisson's ratio $v_0 = 0.5$, and the number of chain segment links $N = 10$. It is shown from Fig. 5(a) that the volume fraction of magnetostrictive particles has a significant effect on the hyperelastic response of the composites. The stiffness of the composites becomes higher with the increase of the particle concentration. Magnetostriction is initiated from the small strain region of the stress–stretch curves. Fig. 5(b) shows that the hyperelasticity of the composites becomes stiffer as the elastic modulus of reinforcement increases. The effect of prescribed magnetostrictive eigenstrains is exemplified in Fig. 5(c). It is shown that the magnetostriction strongly affects the overall tensile hyperelastic deformation. At the same stress level, the composite with magnetostriction exhibits large deformation.

Few experimental data have been published on the magnetostriction and its influence to the finite hyperelastic response of elastomer composites. Mullins and Tobin (1965) and Bergstrom and Boyce (1999) conducted experimental investigations on rigid particle-filled elastomers without magnetostrictions. As a special case, the present model is also able to predict the nonlinear finite-deformation behavior of particle-filled composites by dropping the prescribed eigenstrains. Comparisons are performed between the present prediction and the experimental results from Bergstrom and Boyce (1999) and Mullins and Tobin (1965), as shown in Figs. 6 and 7, respectively. In addition, analytical results from both the Govindjee and



(a) solid lines include the magnetostriction, dot lines not

Fig. 5. Stress–stretch curves of magnetostrictive composites: (a) effect of volume fraction of particles, (b) effect of shear modulus of particles, (c) effect of magnetostrictive eigenstrain.

Simo's (1991) model and the Bergstrom and Boyce's (1999) model are also presented for comparisons. It is noted that the input material constants used in the presented model are consistent with the experimental results and Bergstrom and Boyce's (1999) predictions, as indicated in the figures. Both of Figs. 6 and 7 show that the present model agrees well with those experimental results, especially in the large stretch range. Bergstrom and Boyce's analytical results are similar to the present model, while Govindjee and Simo's predictions underestimate the mechanical response of filled elastomers.

For particle-reinforced elastomer composites, reinforcement particles in general are much stiffer than the matrix. Therefore, many available models ignore the contribution of the strain energy from the particles for simplicity. However, the present model shows that, when the volume fraction of the particles is large or the elastic modulus of particles

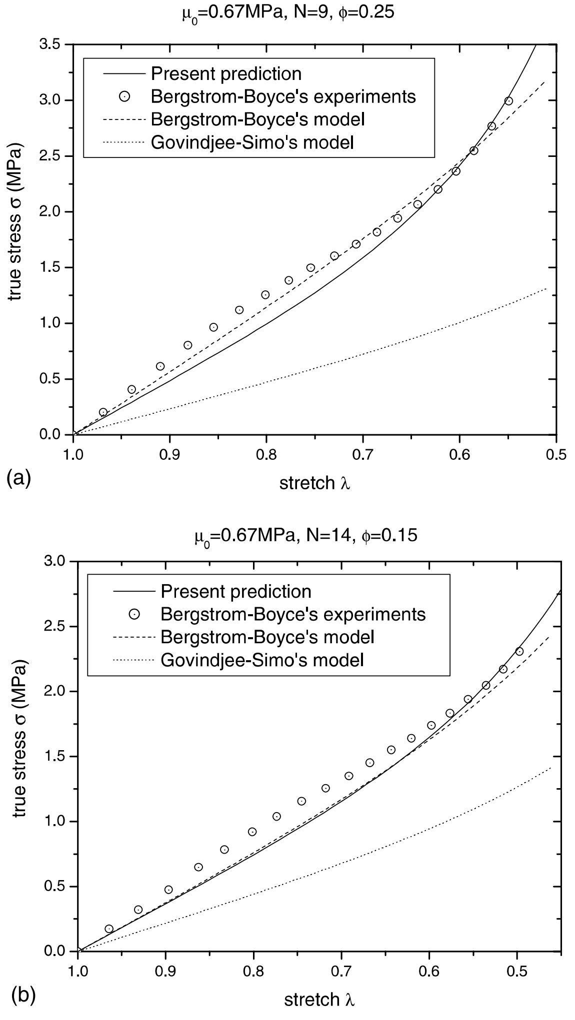


Fig. 6. Comparisons of composite stress–stretch curves with experimental data and other models without magnetostriction: (a) $\phi = 0.25$, (b) $\phi = 0.15$.

is not significantly large compared with that of the matrix, the strain energy in the particles should be considered. Fig. 8 illustrates the effect of shear modulus of reinforcement. It is shown that the modulus of particles has strong effect on the stress–stretch responses of the composites. Bergstrom and Boyce's model with rigid particles would overestimate the mechanical responses of filled elastomers if the material contrast ratio of particles to matrix were less than 20.

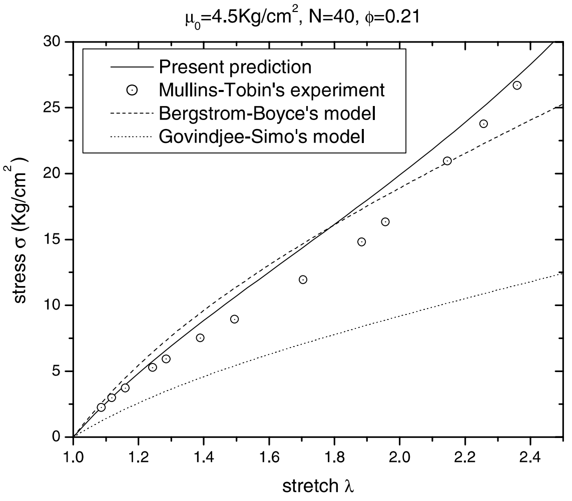


Fig. 7. Comparisons of stress–stretch relation with experimental data and the other model without magnetostriction.

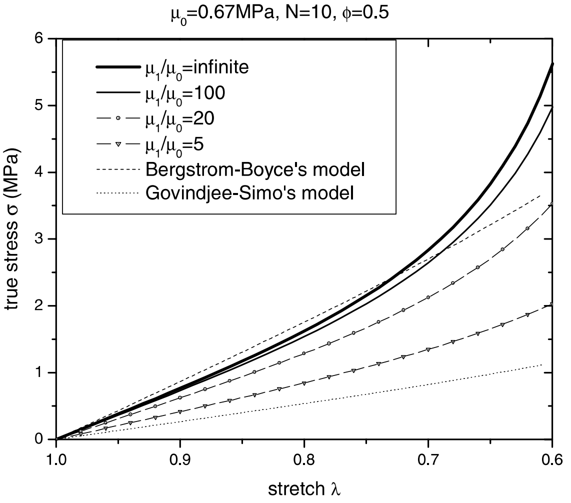
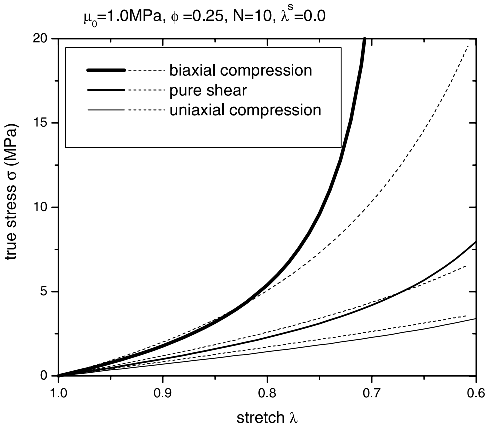


Fig. 8. Comparisons of stress–stretch for different μ_1/μ_0 without magnetostriction.

Mechanical responses under other loading conditions are also calculated. Fig. 9 shows the predictions of nonlinear mechanical behavior of elastomer composites under uniaxial compression, biaxial compression and pure shear from the present model and Bergstrom–Boyce's model. It is found that, for small stretch, the predictions of the



solid lines correspond to present model, dash lines to Bergstrom-Boyce model

Fig. 9. Comparisons of stress–stretch for uniaxial compression, biaxial compression, and pure shear without magnetostriction.

two models are close. For finite deformation, however, the present prediction is different from that of Bergstrom and Boyce's model, since latter is based on neo-Hookean model, which generally yields a smaller stress prediction for the large stretch. The biaxial compression yields the largest difference, while good agreement exists in the uniaxial compression.

5. Conclusions

In this paper, a nonlinear hyperelastic constitutive model is developed for magnetostrictive particle-reinforced elastomer composites, based on the microstructural deformation and physical mechanism of the magnetostrictive particles embedded in the hyperelastic elastomer matrix. Two types of loading conditions are considered—saturated magnetic field and mechanical load. Magnetic eigenstrains are prescribed on the particles due to effect of magnetostriction. Direct particle interaction is taken into account, to accommodate the highly concentrated volume fractions of reinforcement. The proposed hyperelastic model is able to characterize the overall nonlinear elastic stress–stretch relations of the composites under general three-dimensional loading. Model formulations are implemented to investigate the mechanical responses of the composites. Comparisons are conducted among the present results, other existing models, and the experimental data to illustrate the performance of the proposed micromechanics framework.

It is noted that the proposed model is applicable for magnetostrictive particle-filled elastomers under general three-dimensional mechanical loading conditions. However, the applied magnetic field is assumed to be uniaxial. Local deformation mechanisms of interacting particles have been taken into consideration, while the magnetically dipolar interactions between particles are ignored. Future research work on effects of dipolar forces is warranted to improve the magnetomechanical modeling for elastomer composites.

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References

Armstrong, W.D., 2000. The non-linear deformation of magnetically dilute magnetostrictive particulate composites. Mater. Sci. Eng. A A285, 13–17.

Arruda, E.M., Boyce, M.C., 1993. A three-dimensional constitutive model for the large stretch behavior of rubber elastic materials. J. Mech. Phys. Solids 41, 389–412.

Bednarek, S., 1999. The giant magnetostriction in ferromagnetic composites within an elastomer matrix. Appl. Phys. A 68, 63–67.

Belytschko, T., Liu, W.K., Moran, B., 2000. In: Nonlinear Finite Elements for Continua and Structures. John-Wiley, p. 225.

Bergstrom, J.S., Boyce, M.C., 1999. Mechanical behavior of particle filled elastomers. Rubber Chem. Tech. 72, 633–656.

Carlson, J.D., Jolly, M.R., 2000. MR fluid, foam and elastomer devices. Mechatronics 10, 555–569.

Chen, Y., Snyder, J.E., Schwichtenberg, C.R., Dennis, K.W., Falzgraf, D.K., McCallum, R.W., Jiles, D.C., 1999. Effect of the elastic modulus of the matrix on magnetostrictive strain in composites. Appl. Phys. Lett. 74, 1159–1161.

Ciarlet, P.G., 1988. In: Mathematical Elasticity Three-Dimensional Elasticity, vol. 1. North-Holland, p. 130.

Clark, A.E., 1980. Magnetostrictive rare earth-Fe_2 compounds. In: Wohlfarth, E.P. (Ed.), Ferromagnetic Materials, vol. 1. North-Holland, Amsterdam, pp. 531–589.

Cullity, B.D., 1972. Introduction to Magnetic Materials. Addison-Wesley.

Davis, L.C., 1999. Model of magnetorheological elastomers. J. Appl. Phys. 85, 3348–3351.

Eshelby, J.D., 1957. The determination of the elastic field of an ellipsoidal inclusion and related problems. Proc. Roy. Soc. A241, 376–396.

Ginder, J.M., Nichols, M.E., Elie, L.D., Tardiff, J.L., 1999. Magnetorheological elastomers: properties and applications. In: Proceedings Series of SPIE Smart structures and Materials, vol. 3675, pp. 131–138.

Ginder, J.M., Nichols, M.E., Elie, L.D., Clark, S.M., 2000. Controllable-stiffness components based on magnetorheological elastomers. In: Proceedings Series of SPIE Smart structures and Materials 2000, vol. 3985, pp. 418–425.

Ginder, J.M., Schlotter, W.F., Nichols, M.E., 2001. Magnetorheological elastomers in tunable vibration absorbers. In: Proceedings Series of SPIE Smart structures and Materials, vol. 4331, pp. 103–110.

Govindjee, S., Simo, J.C., 1991. A micro-mechanically based continuum damage model for carbon black-filled rubbers incorporating Mullins' effect. J. Mech. Phys. Solids 39, 87–112.

Herbst, J.F., Capehart, T.W., Pinkerton, F.E., 1997. Estimating the effective magnetostriction of a composite: a simple model. Appl. Phys. Lett. 70, 3041–3043.

Jiles, D.C., 1991. Introduction to Magnetism and Magnetic Materials. Chapman and Hall.

Ju, J.W., Sun, L.Z., 1999. A novel formulation for exterior-point Eshelby's tensor of an ellipsoidal inclusion. J. Appl. Mech. 66, 570–574.

Ju, J.W., Sun, L.Z., 2001. Effective elastoplastic behavior of metal matrix composites containing randomly located aligned spheroidal inhomogeneities, Part I: micromechanics-based formulation. Int. J. Solids Struct. 38, 183–201.

Lanotte, L., Ausanio, G., Iannotti, V., Pepe, G., 2001. Magnetic and magnetoelastic effects in a composite material of Ni microparticles in a silicone matrix. Phys. Rev. B 63, 054438,1-6.

Mullins, L., 1969. Softening of rubber by deformation. Rubber Chem. Tech. 42, 339–362.

Mullins, L., Tobin, N.R., 1965. Stress softening in rubber vulcanizates, Part I: use of a strain amplification factor to describe the elastic behavior of filler-reinforced vulcanized rubber. J. Appl. Polym. Sci. 9, 2993–3009.

Mura, T., 1987. Micromechanics of Defects in Solids, Second ed. Kluwer Academic Publishers.

Nan, C.W., 1998. Effective magnetostriction of magnetostrictive composites. Appl. Phys. Lett. 72, 2897–2899.

Nan, C.W., Weng, G.J., 1999. Influence of microstructural features on the effective magnetostriction of composite materials. Phy. Rev. B 60, 6723–6730.

Nemat-Nasser, S., Hori, M., 1999. Micromechanics: Overall Properties of Heterogeneous Materials, Second ed. North-Holland.

Pinkerton, F.E., Capehart, T.W., Herbst, J.F., Brewer, E.G., Murphy, C.B., 1997. Magnetostrictive $SmFe_2$/metal composites. Appl. Phys. Lett. 70, 2601–2603.

Sun, L.Z., Ju, J.W., 2001. Effective elastoplastic behavior of metal matrix composites containing randomly located aligned spheroidal inhomogeneities, Part II: applications. Int. J. Solids Struct. 38, 203–225.

Treloar, L.R.G., 1975. The Physics of Rubber Elasticity. Clarendon Press, Oxford.

Tremolet de Lacheisserie, E., 1993. Magnetostriction: Theory and Applications of Magnetoelasticity. CRC Press.