Coupled magnetic field and viscoelasticity of ferrogel

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Abstract

Composed of a soft polymer matrix and magnetic filler particles, ferrogel is a smart material responsive to magnetic fields. Due to the viscoelasticity of the matrix, the behaviors of ferrogel are usually rate-dependent. Very few models with coupled magnetic field and viscoelasticity exist in the literature, and even fewer are capable of reliable predictions. Based on the principles of non-equilibrium thermodynamics, a field theory is developed to describe the magneto-viscoelastic property of ferrogel. The theory provides a guideline for experimental characterizations and structural designs of ferrogel-based devices. A specific material model is then selected and the theory is implemented in a finite-element code. As numerical examples, the responses of a ferrogel in uniform and non-uniform magnetic fields are analyzed. The dynamic response of a ferrogel to cyclic magnetic fields is also studied, and the prediction agrees with our experimental results. In the reversible limit, our theory recovers existing models for elastic ferrogel, and is capable of capturing some instability phenomena.

Keywords: magnetic field; viscoelasticity; ferrogel; finite element method

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1 Introduction

Although naturally insensitive to magnetic fields, polymers have been made magneto-responsive by embedding iron or magnetite particles. The resulting polymer-based composite materials are often divided into two categories based on the forms of response: ferrogel and magneto rheological elastomer. Characterized by its low mechanical stiffness and usually isotropic filler distribution, ferrogel responds to magnetic stimuli with large deformation. Typical deformation patterns include elongation, rotation and torsion, coiling and bending (Zrínyi et al., 1996, 1998; Snyder et al., 2010; Nguyen and Ramanujan, 2010). Strains of up to 40% when exposed to a non-uniform magnetic field have also been reported (Zrínyi et al., 1997). The deformation of ferrogels in a uniform magnetic field has also been
demonstrated (Raikher et al., 2008; Filipcsei and Zrínyi, 2010). The large deformation capability has made ferrogels a promising material for soft actuators and sensors (Ramanujan et al., 2006; Monz et al., 2008; Qin et al., 2009; Faidley et al., 2010). Magneto-rheological elastomer, on the other hand, responds to magnetic field by changing its mechanical properties, e.g. stiffness (Carlson and Jolly, 2000), and is used in applications like tunable vibration absorbers and damping components (Ginder et al., 2001; Deng et al., 2006; Lerner and Cunefare, 2008). The current paper will focus on the magnetic-field-induced large deformation of ferrogels.

To provide guidance for the design and optimization of ferrogel-based devices, continuous efforts on modeling the coupling behaviors of ferrogels have been made in past decades. Early approaches tend to solve the magnetic field separately and treat the coupling effect by adding field-induced distributed forces and moments (e.g. Zrínyi et al., 1996). More recently, fully coupled nonlinear field theories have been developed (e.g. Dorfmann and Ogden, 2004; Bustamante et al., 2008). The theories consider the coupled elastic deformation and magnetic field, and describe specific material properties by free-energy functions of deformation and magnetic field. The theories have also been implemented numerically to handle boundary-value problems in complex geometries (Dorfmann et al., 2005; Bustamante et al., 2007). However, the elastic theories become deficient when the dynamic response of a viscoelastic ferrogel is of interest. Partly due to the viscoelasticity of the polymer matrices, the responses of ferrogels are often rate-dependent, as demonstrated in various experiments (Zrínyi et al., 1998; Hernández, 2004; Faidley et al. 2010). Very few researches have been carried out on modeling the viscoelastic behaviors of ferrogels. Some models use combinations of linear springs and dashpots to fit the dynamic responses of ferrogels to cyclic magnetic fields (Zrínyi et al., 1998; Rao et al., 2010; Faidley et al., 2010). Without a comprehensive field theory, these models are limited to one dimensional small deformation, and provide little physical insight or guidance for improved designs.

The current paper presents a field theory that fully couples the magnetic field and the large viscoelastic deformation in ferrogels. In Section 2, following the approaches recently used for elastic (Suo et al., 2008) and viscoelastic (Hong, 2010) dielectrics, we define the stress and magnetic fields through an energy approach similar to the principle of virtual work. Such definitions are material-independent and suitable for both equilibrium and non-equilibrium states. Based on the principles of non-equilibrium thermodynamics, Section 3 derives the governing equations for the coupled physics. Under certain physical assumptions, Section 4 proposes a simple material model and Section 5
implements it further into a finite-element method. Finally, numerical simulations of a ferrogel sample in response to uniform and non-uniform magnetic fields are demonstrated as an application of the theory.

2 Stress and magnetic field

To describe the deformation of a ferrogel, we pick the undeformed state to be the reference, in which the material is fully relaxed and no magnetic or mechanical load is present. Following the usual practice in continuum mechanics, we identify a material particle by its position vector in the reference state, \( \mathbf{X} \), and trace the motion by its current position at time \( t \), \( \mathbf{x}(\mathbf{X},t) \). The local deformation and rotation of a material particle is characterized by the deformation gradient \( \mathbf{F}(\mathbf{X},t) = \nabla \mathbf{x} \). In the current paper, the gradient operator \( \nabla \), the divergence operator \( \nabla \cdot \), and the curl operator \( \nabla \times \), indicate differentials with respect to the coordinates in the reference state.

To avoid ambiguity, we define the internal fields as the representatives of external loads, independent of the material properties and the thermodynamic state. Such an approach has been used in the analysis of electro-active polymers (Suo et al., 2008; Hong, 2010). In the reference state, let \( dV(\mathbf{X}) \) be a volume element, and \( dA(\mathbf{X}) \) be the area of a surface element. Correspondingly, we denote the mechanical force in the volume as \( \mathbf{b}(\mathbf{X},t)dV \) and that on a surface as \( \mathbf{t}(\mathbf{X},t)dA \). We define the tensor of nominal stress (i.e. the Piola-Kirchhoff stress of the first kind), \( \mathbf{P}(\mathbf{X},t) \), such that the equation

\[
\int_{\Omega} \mathbf{P} \cdot \nabla \xi dV = \int_{\Omega} \mathbf{b} \cdot \xi dV + \int_{\partial\Omega} \mathbf{t} \cdot \xi dA
\]

holds true for arbitrary test field \( \xi(\mathbf{X}) \) in any domain \( \Omega \) and on its surface \( \partial\Omega \). In the case of a dynamic process, inertial forces are included as the body force. While Eq. (1) serves only as a definition of the stress field, it becomes the principle of virtual work when \( \xi \) is taken to be a virtual displacement field. The stress defined herein recovers the common definition in a state of thermodynamic equilibrium. Applying the divergence theorem to the left-hand side of Eq. (1), via integration by parts, one would easily obtain a mathematically equivalent definition of the nominal stress:

\[
\nabla \cdot \mathbf{P}^T + \mathbf{b} = \mathbf{0}
\]
in the volume of a body and

$$\left( {\mathbf{P}^- - \mathbf{P}^+} \right) \cdot \mathbf{N} = \mathbf{t} \quad (3)$$
onumber

on an interface where the mechanical traction \( \mathbf{t} \) is applied. The labels “+” and “−” differentiate the media on the two sides of the interface, and the unit vector \( \mathbf{N} \) is normal to the interface in the reference state, pointing towards the medium “+”.

In general, a ferrogel may contain conductive parts such as an electromagnetic coil on the surface or in the volume. For simplicity, we take a conceptual idealization and neglect both the mechanical stiffness and the electric resistance of the conductive phases. Upon homogenization, we write the volumetric current density as \( \mathbf{~J}_j \) and the interfacial current density as \( \mathbf{~J}_J \), both measured with respect to the undeformed geometry. Similar as in the definition of nominal stress, we define the nominal magnetic field, \( \mathbf{~H}_H \), such that

$$\int (\nabla \times \mathbf{e}) \cdot \mathbf{~H} dV = \int \mathbf{e} \cdot \mathbf{~J}_j dV + \int \mathbf{e} \cdot \mathbf{~J}_J dA \quad (4)$$

holds true for arbitrary test field \( \mathbf{e} \). Upon application of the divergence theorem, Eq. (4) yields an equivalent definition of the nominal magnetic field in a differential form:

$$\nabla \times \mathbf{~H} = \mathbf{~J} \quad (5)$$

in the volume and

$$\mathbf{N} \times (\mathbf{~H}^- - \mathbf{~H}^+) = \mathbf{~J} \quad (6)$$
onumber

on an interface. Eqs. (5) and (6) are known as Ampère's circuit law (e.g. Guru and Hiziroğlu, 2004).

Since a static magnetic field does no work, here we imagine connecting a ferrogel to a field of current sources, as sketched in Fig. 1. Using a continuum approach, we write the current input by external sources as \( \tilde{I}(\mathbf{X}, t) dV \) in the bulk and \( \tilde{I}(\mathbf{X}, t) dA \) on an interface. The charge conservation dictates that the nominal current densities satisfy
\[ \nabla \cdot \vec{j} = \vec{i} \]  
(7)

in the bulk of a homogeneous material, and

\[ \left( \vec{j} - \vec{j}^+ \right) \cdot \mathbf{N} + \nabla_s \cdot \vec{j} = \vec{I} \]  
(8)

on an interface, where \( \nabla_s \cdot \) is the divergence taken on the interface. Within an interval \( \delta t \), external current sources do work

\[ \int \Phi \vec{I} \delta t dV + \int \Phi \vec{I} \delta t dA, \]  
(9)

where \( \Phi(X,t) \) is the electric potential on a material particle (in its conductive phase). Substituting Eqs. (7) and (8) into (9) and utilizing the divergence theorem, we can write the work done by current sources as

\[ -\int \vec{j} \cdot \nabla \Phi \delta t dV - \int \vec{j} \cdot \nabla_s \Phi \delta t dA. \]  
(10)

While the electric-potential gradient is only defined in the conductive phase, it is possible to continuously extend it into the whole domain and define a vector field \( \vec{A}(X,t) \), such that

\[ \frac{\partial \vec{A}}{\partial t} = -\nabla \Phi \]  

in the conductive phase. It is noteworthy that the choice of \( \vec{A} \) is not unique, and \( \vec{A} \) is not the gradient of a continuous potential. Utilizing the definitions of \( \vec{A} \) and \( \vec{H} \), we may further simplify the work done by the external current sources (10) into a volumetric integral over the whole domain:

\[ \int (\nabla \times \delta \vec{A}) \cdot \vec{H} dV. \]  
(11)

It can be recognized from Eq. (11) that \( \vec{A} \) is the magnetic vector potential in a Lagrange description. One may define the nominal magnetic induction as \( \vec{B} = \nabla \times \vec{A} \).

The soft nature of ferrogels usually results in large deformation, and the geometries in the current and reference states differ significantly. The quantities defined here are nominal fields in a
Lagrange description. When needed, equations in terms of the nominal quantities can be easily rewritten in the current state using the geometric relations between the nominal and true fields, such as

$$\sigma = \frac{P \cdot F^T}{\det F}, \quad j = \frac{F \cdot \tilde{j}}{\det F}, \quad A = \tilde{A}, \quad B = \frac{F \cdot \tilde{B}}{\det F}, \quad \text{and} \quad H = \tilde{H},$$

where $\sigma$, $j$, $A$, $B$, and $H$ are the true stress, true current density, true magnetic potential, true magnetic induction, and the true magnetic field, respectively.

3 Nonequilibrium thermodynamics

Consider a body of ferrogel, loaded mechanically by a field of body force $b$ and surface traction $t$, and electromagnetically by a field of current sources. Associated with a velocity field $\mathbf{x}$ and input current $\tilde{i}$ and $\tilde{I}$, the power of the external work done by the electromagnetic and mechanical loads is

$$\int \int \int \Phi + \Phi + \int b \cdot \mathbf{x} \, dV + \int t \cdot \mathbf{x} \, dA + \int \Phi i \, dV + \int \Phi I \, dA. \quad (13)$$

Let $W$ be the Helmholtz free energy of the material per unit reference volume, and $\dot{W}$ be its material rate of changing. Utilizing the definition of stress and magnetic field, one has the corresponding change rate in the total free energy of the system, $\Pi$, including the potential of the external loads,

$$\dot{\Pi} = \int \left( \dot{W} - P : \dot{F} - H \cdot \dot{H} \right) \, dV. \quad (14)$$

The laws of thermodynamics dictate that the free energy of the system never increases in a physically possible process, $\dot{\Pi} \leq 0$. The inequality must hold true in any volume, and thus

$$\dot{\Pi} = \int \left( \dot{W} - P : \dot{F} - H \cdot \dot{H} \right) \, dV \leq 0 \quad (15)$$

holds true on any material particle for any process, where the equal sign takes place only when the process is reversible, i.e. the system is locally in equilibrium. The thermodynamic equilibrium state of a material particle is fully determined by the deformation gradient and the magnetic induction. For a general inelastic material, the free energy in a non-equilibrium state differs from that in an equilibrium state. To distinguish between them, we introduce the equilibrium Helmholtz free-energy density, and
write it as \( W^{EQ}(F, \Hat{B}) \). From (15) in the case of an equal sign, we obtain the following constitutive relations in an equilibrium state:

\[
\begin{align*}
P^{EQ}(F, \Hat{B}) &= \frac{\partial W^{EQ}(F, \Hat{B})}{\partial F}, \\
\Hat{H}^{EQ}(F, \Hat{B}) &= \frac{\partial W^{EQ}(F, \Hat{B})}{\partial \Hat{B}}.
\end{align*}
\] (16)

in which \( P^{EQ} \) and \( \Hat{H}^{EQ} \) are the nominal stress and the nominal magnetic field in an equilibrium state.

To describe a non-equilibrium state, following the usual approach in finite-deformation viscoelasticity (Lee, 1969), we imagine an intermediate state between the reference state and the current state, which may be achieved by a virtual elastic unloading on the part of the polymer network that is not in equilibrium. The relation between the reference, current, and intermediate states are illustrated in Fig. 2. In order for a full relaxation, the material needs to be divided into infinitesimal particles, which do not necessarily constitute a continuum body in the intermediate state. Denoting the deformation gradient of the intermediate state as \( F^{i}(X, t) \), and that of the current state with respect to the intermediate state as \( F^{c}(X, t) \), we have the multiplicative decomposition of the deformation gradient:

\[
F = F^{c} \cdot F^{i}.
\] (17)

On the other hand, the magnetization of a ferrogel may involve two distinct processes: each filler particle changes its direction of magnetization without rotating its spatial orientation, as shown schematically in Fig. 3b; each particle maintains its magnetization along an easy-magnetization axis, but rotates against the matrix towards the direction of the applied field, as in Fig. 3c. The first process usually dominates in multicrystalline ferromagnetic materials. In general, both processes take place simultaneously in a ferrogel with relatively soft matrix. The two physical processes have distinct characteristic time scales. The magnetic domain switch reaches equilibrium much faster than the rotation of a solid particle in a viscoelastic matrix. Therefore, in the regime where most ferrogels are applied, it may be assumed that the first process is always in equilibrium, while the second process dissipates energy and its rate depends on the viscoelastic property of the matrix. In the current paper, we only consider the case when the first process dominates, so that the magnetic fields are always in equilibrium, \( \Hat{H} \equiv \Hat{H}^{EQ} \). Physically, such an assumption represents ferrogels with relatively large (> 5μm)
filler particles which exhibits less anisotropy, and operated under a field far from saturation. This assumption also implies that the magnetic field under consideration is quasi-static with no propagation of electromagnetic wave, and the energy dissipation due to the induced current in filler particles is neglected.

Following Reese and Govindjee (1998), we assume that the non-equilibrium Helmholtz energy, namely the difference between the Helmholtz free energy of a non-relaxed state and that of an equilibrium state, depends only on the elastic deformation between the relaxed intermediate state and the current state, \( W - W^E \equiv W^{NEQ}(F^e) \). Since the irreversible deformation is more suitable for characterizing a non-equilibrium state, here we take the corresponding deformation gradient \( F^i \) as an internal state variable, and write the total Helmholtz free energy density as

\[
W(F, F^i, \tilde{B}) = W^E(F, \tilde{B}) + W^{NEQ}(F \cdot F^{i-1}).
\]  

To handle more general cases when filler particles exhibit significant magnetic anisotropy and the spatial rotation of particles are important, one may extend the current model by accounting for the dependence on magnetic induction \( \tilde{B} \) in the nonequilibrium free energy density \( W^{NEQ} \).

The total nominal stress is the derivative of the Helmholtz free-energy density with respect to the deformation gradient even in a non-equilibrium state (Coleman and Gurtin, 1967),

\[
P = \frac{\partial W}{\partial F} = P^E + P^{NEQ} \cdot (F^i)^T,
\]

where the inelastic nominal stress tensor \( P^{NEQ} = \frac{\partial W^{NEQ}}{\partial F^e} \). The remainder of inequality (15) which governs the evolution of the inelastic internal variables becomes:

\[
(F^e^T \cdot P^{NEQ} \cdot F^{i-T}) : \dot{F}^i \geq 0,
\]

which physically indicates that the energy of the system only dissipates in inelastic deformation.

In terms of the inelastic true stress \( \sigma^{NEQ} = P^{NEQ} \cdot F^e^T / \text{det} F^e \), inequality (20) can also be written in the current configuration as:
\[ \sigma^{NEQ} : \mathbf{L}' \geq 0, \]  

(21)

where \( \mathbf{L}' \) is the inelastic part of the covariant velocity gradient, \( \mathbf{L}' = \dot{\mathbf{F}} : \mathbf{F}^{-1} - \dot{\mathbf{F}}^e : \mathbf{F}^{-1} = \mathbf{F}^e : \dot{\mathbf{F}}^e : \mathbf{F}^{-1} \).

While inequality (21) is a consequence of the second law of thermodynamics thus must be satisfied by all processes for any material, a kinetic evolution equation in the form

\[ \mathbf{L}' = \mathbf{M} : \sigma^{NEQ} \]

(22)

is often used in practice. The material-dependent fourth-rank mobility tensor \( \mathbf{M} \) needs to be positive-definite to satisfy inequality (20) automatically. By writing the evolution equation in this form, the rigid-body rotation in the inelastic deformation is discarded (Boyce et al., 1989). In general models for the evolution of internal parameters, the mobility tensor may be dependent on various state variables. Equations (16), (19) and (22), together with the definition of state variables in Section 2, form a closed system for the analysis of the coupled magnetomechanical response of viscoelastic ferrogels.

4 A specific constitutive model

To apply the nonlinear field theory developed in the preceding sections to the analysis of ferrogels, one needs to specify the Helmholtz free-energy functions \( W^{EQ}(\mathbf{F}, \vec{\mathbf{B}}) \) and \( W^{NEQ}(\mathbf{F}^e) \), and the mobility tensor \( \mathbf{M} \). In order to characterize the viscoelastic behavior of polymeric materials under various loading conditions, there has been continuous efforts during the past decades on developing equilibrium (e.g. James and Guth, 1943; Treloar, 1975; Flory, 1977; Arruda and Boyce, 1993) and kinetic evolution models (e.g. Lubliner, 1985; Haupt, 1993; Reese and Govindjee, 1998; Bergström and Boyce, 1998). Possible forms of the equilibrium free-energy function \( W^{EQ}(\mathbf{F}, \vec{\mathbf{B}}) \) that couples magnetic field and deformation have also been studied recently (e.g Dorfmann and Brigadnov, 2004; Dorfmann and Ogden, 2004; Kankanala and Triantafyllidis, 2004; Otténio et al., 2008). Here for the purpose of demonstration and qualitative studies, we will construct a simple model.

We assume that the equilibrium free-energy density only consists of the contributions from stretching and magnetization, \( W^{EQ}(\mathbf{F}, \vec{\mathbf{B}}) = W^{EQ}_s(\mathbf{F}) + W^m(\mathbf{B}) \), and the free energy of magnetization only depends on the true magnetic induction \( \mathbf{B} \). The physical decoupling between the deformation and the true magnetic induction field represents a category of materials that have liquid-like magnetization.
behavior independent of the deformation state. For simplicity, we neglect the hysteresis in magnetization and further assume the magnetic property to be linear and the magnetization energy being \( W_m(\mathbf{B}) = \frac{1}{2\mu} \mathbf{B} \cdot \mathbf{B} \), where \( \mu \) is the magnetic permeability. Assuming the free energy of stretching to be purely entropic with Gaussian statistics (Treloar, 1975), \( W_{s\text{EQ}}(\mathbf{F}) = \frac{1}{2} G_{\text{EQ}} \mathbf{F} : \mathbf{F} \), we have the equilibrium free-energy function

\[
W^{\text{EQ}}(\mathbf{F}, \mathbf{B}) = \frac{G_{\text{EQ}}}{2} \mathbf{F} : \mathbf{F} + \frac{1}{2\mu} \left( \mathbf{F} \cdot \mathbf{B} \right) \left( \mathbf{F} \cdot \mathbf{B} \right),
\]

where \( G_{\text{EQ}} \) is the equilibrium or long-term modulus. Similarly, we assume the non-equilibrium free-energy function in the form

\[
W^{\text{NEQ}}(\mathbf{F}^e) = \frac{G^{\text{NEQ}}}{2} \mathbf{F}^e : \mathbf{F}^e = \frac{G^{\text{NEQ}}}{2} \left( \mathbf{F} \cdot \mathbf{F}^{i-1} \right) \left( \mathbf{F} \cdot \mathbf{F}^{i-1} \right)
\]

with non-equilibrium modulus \( G^{\text{NEQ}} \). In the current paper, the material is assumed to be incompressible, with both elastic and inelastic deformations being volume-conservative, \( \det \mathbf{F} = \det \mathbf{F}^i = \det \mathbf{F}^e = 1 \).

Application of free-energy functions (23) and (24) in Eqs. (16) and (19) yields the following constitutive relations:

\[
\mathbf{P} = G^{\text{EQ}} \mathbf{F} + G^{\text{NEQ}} \mathbf{F} \cdot \left( \mathbf{F}^{i-1} \cdot \mathbf{F}^i \right)^{-1} + \frac{1}{\mu} \mathbf{F} \cdot \mathbf{B} \otimes \mathbf{B} - p \mathbf{F}^{-T}, \quad \tilde{\mathbf{H}} = \frac{1}{\mu} \mathbf{F}^T \cdot \mathbf{F} \cdot \mathbf{B},
\]

where \( p \) is an undetermined hydrostatic pressure introduced by the incompressibility constraint. Alternatively, the constitutive relation may also be expressed in terms of true quantities:

\[
\mathbf{\sigma} = G^{\text{EQ}} \mathbf{F} \cdot \mathbf{F}^T + G^{\text{NEQ}} \mathbf{F} \cdot \mathbf{F}^{i-1} \cdot \mathbf{F}^{-T} \cdot \mathbf{F}^T + \frac{1}{\mu} \mathbf{B} \otimes \mathbf{B} - p \mathbf{1}, \quad \mathbf{H} = \frac{\mathbf{B}}{\mu},
\]

with \( \mathbf{1} \) representing the second rank identity tensor. The third term on the right-hand side of Eq. (26), \( \mathbf{B} \otimes \mathbf{B}/\mu \), is usually referred to as the magnetic Maxwell stress. It is noteworthy that the hydrostatic part in the usual form of the Maxwell stress is absorbed in the arbitrary pressure \( p \) introduced by the
incompressibility constraint. The magnetic contribution of stress in this particular form is only a consequence of the specific free-energy function, and should not be generalized to all materials. Under the current assumption that the magnetic field is always in equilibrium, a magnetic contribution does not appear in the inelastic stress $\mathbf{P}^{\text{NEQ}}$ or $\mathbf{\sigma}^{\text{NEQ}}$.

To specify the evolution law for the inelastic deformation, we assume the viscous property of the material to be isotropic in the current state, so that the inverse of the mobility tensor takes the form

$$\mathbf{M}^{-1} = \eta \left( \mathbf{1}^4 - \frac{1}{3} \mathbf{1} \otimes \mathbf{1} \right).$$

(27)

Here $\mathbf{1}^4$ is the fourth rank symmetric identity tensor. When a constant viscosity $\eta$ is used, the inelastic behavior of the material resembles that of Newtonian fluid.

5 Finite-element implementation

To simplify expressions, from now on, we will normalize all stresses and energy densities with the instantaneous modulus, $G = G^{\text{EQ}} + G^{\text{NEQ}}$, magnetic fields with $\sqrt{G/\mu_0}$, magnetic inductions with $\sqrt{\mu_0 G}$, and times with $\eta/G$. $\mu_0$ is the permeability of free space. Without any intrinsic length scale in the model, we will normalize all lengths by an arbitrary length $L$ of the geometry, e.g. the characteristic length of the specimen. The dimensionless fields are noted with an over-bar, e.g. $\overline{p} = p/G$, $\overline{\mathbf{B}} = \mathbf{B}/\sqrt{\mu_0 G}$, and $\overline{\mathbf{x}} = \mathbf{x}/L$.

The material model described in Section 4 has two dimensionless parameters: the relative permeability $\mu_r = \mu/\mu_0$ and the ratio between the equilibrium modulus and instantaneous modulus, $\chi = G^{\text{EQ}}/G$. The parameter $\chi$ characterizes the fraction of the polymer network that has time-independent deformation (Bergström and Boyce, 1998). The viscoelastic material reduces to purely elastic when $\chi = 1$, and becomes a viscous fluid when $\chi = 0$.

Within the current theoretical framework, the mechanical momentum balance and the equilibrium of magnetic field are enforced by the definitions of stress and magnetic field. The definitions are naturally in weak forms as in Eqs. (1) and (4). Applying the specific material model in
Section 4 by substituting Eq. (25) into Eqs. (1) and (4), we arrive at the dimensionless weak forms explicitly as

\[
\int_{\Omega} \left[ (\chi \mathbf{F} - \bar{\rho} \mathbf{F}^{-T}) \cdot \partial \mathbf{F} + (1 - \chi) \left( \mathbf{F}^{T} \cdot \mathbf{F} \right) \right] \cdot \left( \mathbf{B} \otimes \mathbf{B} : \left( \mathbf{F}^{T} \cdot \partial \mathbf{F} \right) \right) dV = \int_{\Omega} \bar{\mathbf{B}} \cdot \bar{\delta} \mathbf{F} dV + \int_{\partial \Omega} \bar{\mathbf{J}} \cdot \bar{\delta} \mathbf{dA} \tag{28}
\]

and

\[
\int_{\Omega} \mathbf{B} \cdot \mathbf{F}^{T} \cdot \mathbf{F} \cdot \partial \mathbf{B} dV = \int_{\Omega} \bar{\mathbf{J}} \cdot \bar{\delta} \mathbf{F} dV + \int_{\partial \Omega} \bar{\mathbf{J}} \cdot \bar{\delta} \mathbf{dA}. \tag{29}
\]

Following the usual approach in finite-element analysis, we add to the weak form

\[
\int_{\Omega} (\text{det} \mathbf{F} - 1) \partial \bar{\rho} dV = 0 \tag{30}
\]

for the volume incompressibility and to determine the field of hydrostatic pressure $\bar{\rho}$.

To evolve the inelastic deformation in the intermediate state, we further write Eq. (22) into the following weak form:

\[
\int_{\Omega} \left[ \mathbf{F} \cdot \mathbf{S}^{\text{NEQ}} \cdot \mathbf{F}^{T} - \mathbf{M}^{-1} \cdot \left( \mathbf{F}^{e} \cdot \mathbf{F}^{i} \right) \right] \cdot \left( \mathbf{F}^{e} \cdot \partial \mathbf{F}^{i} \cdot \mathbf{F}^{-1} \right) dV = 0, \tag{31}
\]

where $\mathbf{S}^{\text{NEQ}} = (1 - \chi) \text{dev} \left( \mathbf{F}^{T^{-1}} \cdot \mathbf{F}^{T} \right)$ is the deviatoric part of the non-equilibrium Piola-Kirchhoff stress of the second kind, and the time derivative $\dot{\mathbf{F}}^{i}$ is taken with respect to the dimensionless time $\tau = tG/\eta$. Considering the symmetric viscous tensor in Eq. (27), we can further simplify Eq. (31) as

\[
\int_{\Omega} \left( \mathbf{F}^{i} \cdot \mathbf{S}^{\text{NEQ}} \cdot \mathbf{F}^{T} \cdot \mathbf{F} - \mathbf{F}^{i} \right) : \partial \mathbf{F}^{i} dV = 0. \tag{32}
\]

Weak forms (28), (29), (30), and (32) constitute a system sufficient to determine the evolution of the dimensionless fields in a ferrogel, $\bar{\mathbf{x}}(\bar{\mathbf{X}}, \tau), \bar{\rho}(\bar{\mathbf{X}}, \tau), \mathbf{F}^{i}(\bar{\mathbf{X}}, \tau)$, and $\bar{\mathbf{A}}(\bar{\mathbf{X}}, \tau)$. However, a ferrogel system may also contain materials that are either very stiff (e.g. a rigid magnet) or extremely compliant (e.g. vacuum or fluid). In vacuum, the deformation fields $\bar{\mathbf{x}}(\bar{\mathbf{X}}, \tau)$ and $\mathbf{F}^{i}(\bar{\mathbf{X}}, \tau)$ are undetermined, while the inclusion of a very stiff body may cause the problem to be numerically ill-conditioned. Instead,
we employ the arbitrary Lagrangian-Eulerian (ALE) method which introduces an artificial deformation field in a vacuum or fluid domain. The artificial deformation, namely the moving mesh, agrees with the actual deformation on the interface between a ferrogel and vacuum, and maximizes the mesh smoothness in vacuum (COMSOL, 2008). On the other hand, the mesh is immobile on a fixed rigid body, \( \mathbf{x} = \mathbf{X} \). To simplify calculation, the weak form of magnetostatics, Eq. (29), is rewritten in the current configuration with the Eulerian magnetic potential \( \Lambda(\mathbf{X}(\mathbf{x}), t) \). We have implemented the formulations in the commercial finite-element package, COMSOL Multiphysics 3.5a, for both 2D and 3D axisymmetric geometries, and used them in the following analyses.

6 Numerical examples

In this section, the responses of viscoelastic ferrogels in magnetic fields will be studied as illustrations of the theoretical framework and numerical method developed. The deformations of a ferrogel in three different magnetic fields will be analyzed. In the first example, the specimen lies inside a solenoid, where the magnetic field is nearly uniform. The second example analyzes the large deformation of a ferrogel induced by a highly non-uniform magnetic field. The third example aims at recovering our recent experimental results of the dynamic response of a ferrogel subject to the cyclic field near an electromagnet.

6.1 A quasi-uniform magnetic field

As a first example, we study the dynamic response of a cylindrical ferrogel inside a solenoid, as shown schematically in Fig. 4a. No mechanical load is present. The computational domain is 3D axisymmetric. A cyclic magnetic field, \( \mathbf{H} = H_0 \sin \omega t \), is applied through an alternating current of dimensionless frequency \( \omega \). In the absence of the ferrogel, the magnetic field inside the solenoid is uniform. However, since the ferrogel has a larger magnetic permeability than vacuum or air, the uniform field is perturbed when the ferrogel is in position. The resulting non-uniform field drives the inhomogeneous deformation of the ferrogel, and the deformed shape further redistributes the magnetic field. The deformed shape and the field lines are plotted in Fig. 4b. In this example, we have taken a relative magnetic permeability \( \mu_r = 2 \) for the ferrogel, and the heterogeneities in the magnetic field and deformation are still relatively small. The resulting behavior of the ferrogel – extension along the field direction – agrees with the experimental observation of a ferrogel in a nearly uniform field.
(Filipcsei and Zrínyi, 2010). The relation between the averaged magnetic field and the averaged magnetic induction is also close to that of a rigid linear magnetic material, as shown in Fig. 4c. Only a very small hysteresis due to the geometric effect is shown on the plot. Taking a representative value for the modulus of the ferrogel, \( G = 10 \text{kPa} \), the dimensionless magnetic field \( \bar{H}_0 = 1 \) approximately corresponds to a dimensional field strength of \( 90 \text{kA/m} \), a value close to the highest achievable uniform field without bringing the material to saturation (Raikher et al., 2008; Snyder et al., 2010).

To show the viscoelasticity effect, we plot the total axial stretch \( \lambda \) and the inelastic stretch \( \lambda^i \) in response to a cyclic field in Fig. 5. When the ferrogel is actuated by a magnetic field at a relatively low frequency, both \( \lambda \) and \( \lambda^i \) are sinusoidal functions of time and are in phase with each other, as shown in Fig. 5a. The stretch is almost fully inelastic, i.e. \( \lambda \approx \lambda^i \), implying that the material is always relaxed. Since the strain only depends on the magnitude and not the sign of the magnetic field, the frequency of deformation doubles that of the applied field. Even though the inelastic response is almost in phase with the applied field, hysteresis loops still appears on the \( \bar{H} - \lambda \) plots in Fig. 5d. This is mainly due to the geometrical nonlinearity introduced by finite deformation. When the magnetic field alternates at an intermediate frequency, the magnitude of stretch is smaller than in the low frequency case, as shown in Fig. 5b. The inelastic stretch \( \lambda^i \) is lower than the total stretch \( \lambda \), and exhibits a finite phase lag due to viscoelasticity. On the trajectory plot of \( \bar{H} - \lambda \) in Fig. 5e, the hysteresis loop stabilizes after a few cycles. When the ferrogel deforms in a magnetic field at a relatively high frequency, the magnitude of the mechanical response is even smaller, as shown in Fig. 5c. The mean value of the inelastic stretch increases gradually and reaches a steady state after many cycles, and the \( \bar{H} - \lambda \) plot in Fig. 5f has negligible hysteresis due to a relatively small deformation. Experiments following the procedure illustrated here may be carried out to identify the viscoelastic properties of ferrogels.

### 6.2 A non-uniform magnetic field

In a magnetic field, each filler particle is magnetized and can be considered as a magnetic dipole. Since the motion of a magnetic dipole is only driven by the gradient of the external field, the deformation of a ferrogel is expected to be much larger if the applied field is non-uniform. In this example, we look at a rectangular ferrogel strip placed in the non-uniform field generated by a pair of magnets, as shown in Fig. 6a. Normalized by the length of the ferrogel strip, \( L \), the geometric
parameters of this example include the distance between the bottom of strip and the axis of magnets $Z = 1$, the size of the magnets $D = 1$, and the distance between the two magnets $D = 2$. While these parameters can all affect the final deformation of the ferrogel (Snyder et al., 2010), as a demonstration, we will only focus on the viscoelastic effect by changing the material parameter $\chi$. The relative permeability of the ferrogel is still taken to be $\mu_r = 2$.

The distribution of the magnetic field is nonuniform: the field maximizes near the axis of two magnets where $H = H_{\text{max}}$, and decays exponentially away from the axis. In the prescribed geometry, $H \approx 0.04H_{\text{max}}$ at the bottom of the undeformed ferrogel. As the ferrogel deforms, the magnetic field near the ferrogel is perturbed, but no noticeable change in $H_{\text{max}}$ is observed near the magnets axis. We will thus use $H_{\text{max}}$ as an indicator for the strength of the applied field.

By symmetry, we establish a 2-D model consisting of half of the ferrogel and one magnet. The deformation pattern of the ferrogel and the distribution of the magnetic field are plotted in Fig. 6b. In addition to the local non-uniform deformation, an overall elongation is induced by the spatial gradient of the magnetic field. The symmetry boundary condition also introduces an artificial constraint to the ferrogel. Due to the lateral non-uniformity of the field, the straight extension of the ferrogel is unstable, and the symmetry may be broken with the ferrogel bending towards one of the magnets. Extra constraints (e.g. by using a glass tube, Nguyen and Ramanujan 2010) are often added to prevent the unwanted lateral bending. On the other hand, when the local field intensity reaches a critical value, a surface instability similar to the Rosensweig instability of ferrofluid will occur (Cowley and Rosensweig, 1967). Without considering surface energy, our model diverges at the onset of this instability. To circumvent this issue, in the following calculations where high field intensity is needed, we add an additional constraint to the edges of the ferrogel by forcing them to be straight, an approach similar to the glass-tube constraint in experiments.

In Fig. 6c, we vary the dimensionless parameter $\chi$ from 0.1 to 0.9, and plot the equilibrium deformation of the ferrogel as a function of the applied field. As expected, when the ferrogel is more liquid-like (smaller $\chi$), its deformation is larger. Compared to the quasi-uniform-field example, the overall deformation caused by the non-uniform field is much larger, even for a solid-like (larger $\chi$)
Fig. 6c also shows that the stretch $\lambda$ increases drastically at higher but finite magnetic field. This trend is related to yet another type of instability observed in the experiments (Zrínyi et al., 1997; Snyder et al., 2010). Such instability is mainly due to the geometry and the spatial distribution of the magnetic field. Multiple equilibrium states can be achieved under the same applied field: the ferrogel can have a smaller elongation and remain in the region of lower magnetic field gradient; alternatively the ferrogel can be highly stretched and reach a position much closer to the axis of the magnets where the gradient of magnetic field is much higher. For simplicity, we neglect the viscoelasticity of the material which plays a minor role in this case, and study the response of a fully elastic ($\chi = 1$) ferrogel. As shown by the solid curves in Fig. 6d, two stable branches of the equilibrium stretch-magnetic-field relation are obtained by prescribing either the undeformed state or a highly stretched state as the initial condition, and gradually increasing or decreasing the applied field. Beyond a certain magnetic field, a ferrogel in the shorter state extends instantaneously to the longer state. Likewise, when the magnetic field decreases below a critical value, the ferrogel originally in the longer state will retract suddenly to the shorter state. In between the two critical values, three equilibrium states are possible. Besides the two stable states, we also obtained the unstable state (shown as the dotted curve on Fig. 6d) by properly constraining the ferrogel. The numerical results agree qualitatively with the existing experimental measurements (Zrínyi et al., 1997; Synder et al., 2010), quantitative predictions may be possible upon calibration of the material behaviors and a more accurate representation of the actual 3-D geometry.

### 6.3 Cyclic response induced by an electromagnet

To further validate the theoretical framework, this example simulates our recent experiment (Faidley et al., 2010). As sketched in Fig. 7a, a ferrogel cylinder is placed on the surface of an electromagnet. We assume the system to be axisymmetric and neglect the friction between the ferrogel and the electromagnet. Fig. 7b shows the deformed shape of the ferrogel and the distribution of the magnetic field. Due to the combined magnetic and gravitational driving forces, the ferrogel reduces in height and increases in diameter. The diameter increase is more significant near the bottom of the ferrogel, where the field gradient is higher. The deformed shape of the ferrogel agrees qualitatively with our observation.
In experiment, it was found that this type of ferrogel (polyvinyl alcohol crosslinked by sodium tetraborate) demonstrates a significant viscoelastic property. Within a relatively short period, the dynamic response to a cyclic field is almost always a sinusoidal oscillation superimposed on a linear creep (Fig. 7c). The magnitude of the sinusoidal oscillation, which is due solely to the magnetic field, depends on the frequency of the field as shown in Fig. 7d. To compare with the experimental results, we plot the numerical solutions of the overall axial strain together with the measured data, both in the time domain (for the response to a magnetic field at 1Hz, Fig. 7c) and in the frequency domain (Fig. 7d). The numerical results agree well with the experiments.

The dimensionless parameters used in calculation are as follows. The relative permeability of the ferrogel, $\mu_r = 2$, is obtained directly from experimental measurements. The ratio between the equilibrium and instantaneous moduli $\chi = 0.09$, the dimensionless specific weight of the ferrogel $\tilde{\rho} = bL/G = 0.017$, and the dimensionless magnitude of the applied magnetic field $\tilde{H}_0 = 0.5$, are determined from fitting the computational results to the experiment. The corresponding dimensional parameters: density of ferrogel at 1700kg/m³, maximum magnetic field at 60kA/m, instantaneous modulus at 20KPa, are all close to measured values. With a viscosity 70kPa∙s, the dynamic response of the material has a characteristic frequency of around 0.3 Hz, and agrees well with that measured as shown in Fig. 7d.

7 Concluding remarks

Based on the principles of nonequilibrium thermodynamics, this paper develops a field theory that couples the large inelastic deformations and magnetic fields in ferrogel. A simple model is specified by assuming a Newtonian-fluid-like kinetic property of the material. Based on the theory and the simple model, a finite element method is further developed so that numerical calculations are possible even in complex geometries. Using the numerical codes, we carry out simulations of a ferrogel in three typical types of magnetic fields. In a quasi-uniform field, the ferrogel extends along the field-direction. In a non-uniform field, the ferrogel moves towards the region of higher magnetic field gradient. In both cases, the responses of a viscoelastic ferrogel are rate-dependent. In a highly non-uniform magnetic field, the instability of a ferrogel caused by geometric nonlinearity is revealed by the model. The dynamic response of a ferrogel driven simultaneously by the constant gravity force and a cyclic non-uniform magnetic field is also studied. The numerical results agree well with our experimental
measurements both in time and frequency domains. When proper material models are adopted and calibrated through experiments, the theory is not only applicable to existing magneto-active materials, but to emerging materials as well.

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References


Figures

**Fig. 1.** Sketch of a ferrogel under combined mechanical and electromagnetic loads. \( P \) indicates the external mechanical load applied by a weight and \( I \) is the current input to the ferrogel by a current source.

**Fig. 2.** A physical deformation is decomposed into two parts through imagining an intermediate state, in which every material particle is elastically relaxed. The fully relaxed material particles in the intermediate state do not need to constitute a continuum body. The inelastic stress in the current state is assumed to be a function of the elastic deformation gradient \( \mathbf{F}^e \) only.
Fig. 3. (a) At a reference state the filler particles are randomly distributed in the polymer matrix. Applying a magnetic field to the ferrogel either changes the magnetization direction of each filler particle without rotating its spatial orientation (b), or rotates particle against the matrix towards the direction of the external field (c). The elliptical shapes are used to show the physical orientations of particles.

Fig. 4. (a) A ferrogel sample is placed inside a solenoid. (b) The magnetic field is slightly perturbed by the ferrogel due to coupling. The color scale indicates the axial stretch $\lambda_3$. (c) The $\overline{H} \sim \overline{B}$ curve is close to linear and demonstrates an insignificant hysteresis (c).
Fig. 5. (a) (b) (c) Total stretch $\lambda(\tau)$ and inelastic stretch $\lambda_i(\tau)$ of a ferrogel in response to a cyclic magnetic field $H(\tau) = H_0 \sin \omega \tau$. (d) (e) (f) Trajectory plots of the stretch $\lambda(\tau)$ with respect to the applied nominal magnetic field $H(\tau)$. Three different dimensionless frequencies are selected for comparison: $\omega = 0.1$ for (a-d), $\omega = 1$ for (b-e), and $\omega = 10$ for (c-f).
Fig. 6. (a) A nonuniform magnetic field is produced by two electromagnets to drive large deformation of a ferrogel. (b) Part of a 2-D model that captures the elongation of the ferrogel. The color scale indicates the longitudinal stretch $\lambda$. (c) The stretch $\lambda$ as a function of the applied field. Several values of the viscoelastic parameters $\chi$ are used. (d) An instability phenomenon as captured by an elastic model.
Fig. 7. (a) Sketch of experimental setup. (b) The ferrogel is shortened longitudinally and expands laterally due to gravity and the magnetic field. The color scale indicates the longitudinal stretch $\lambda$. Numerical results in terms of the average axial strain are compared with experiments in the time domain (c) and in the frequency domain (d).