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Abstract

A systematic development of a continuum model is presented, which is capable of describing the magneto-mechanical behavior of magnetic polymer gels commonly referred to as “ferrogels”. In the present research, ferrogels are treated as multicomponent, multiphase materials. They consist of a polymer network (P), fixed magnetic particles (f), mobile magnetic particles (m), and liquid (L). By considering ferrogels as multicomponent materials, interaction among constituents of ferrogels can be captured. This helps in understanding the process occurring inside ferrogels under the influence of external stimuli, such as magnetic fields. In our modeling approach, the field equations of ferrogels are derived within the framework of the theory of mixtures. The basic equations include Maxwell's equations, balance of mass, linear momentum, angular momentum, energy, and entropy. In the framework of the theory of mixtures, balance relations are first presented at the constituent level also referred to as partial balance relations. By summing partial balance relations over all constituents and imposing the restrictions of theory of mixtures, balance relations of mixture (for the ferrogel) are obtained. In the current work the specific magnetization (magnetization per density) is considered as an evolving variable. It is demonstrated that balance of angular momentum is satisfied using the evolution equation of specific magnetization and constitutive laws. In the process of modeling, a suitable free energy function is introduced and thermodynamically consistent constitutive laws are formulated. Introducing certain assumptions, a reduced model of the ferrogel, a coupled magneto-mechanical formulation, is subsequently presented. The reduced model consists only of a polymer network (P) and fixed magnetic particles (f). It is concluded that the reduced model compares well to the existing ones in the literature. The magneto-mechanical problem based on the reduced model is solved in 2D using the finite element method. The only unknowns for the finite element method implementation are mechanical displacement and magnetic potential. Deformation of a ferrogel in a magnetic field is subsequently investigated. Elongation and contraction of a ferrogel are observed when a magnetic field is applied in the x- and y-directions, respectively. The numerical results were compared with existing experimental work in the literature. A good qualitative agreement was found between numerical and experimental results.

Keywords

Ferrogels, modeling, theory of mixtures, magneto-mechanical formulation

Introduction

Ferrogels are magneto-sensitive materials, the fabrication of which was first reported in the late 90s (Zrínyi et al., 1996, 1997). They primarily consist of a polymer gel and magnetic filler nanoparticles, for example, magnetite Fe_3O_4 (Gollwitzer et al., 2008). Polymer gels are usually synthesized by chemically cross-linking an array of polymer networks. By application of an external magnetic field, magnetic particles orient themselves towards the applied field. Since some of the particles adhere to the polymer network, ferrogels deform in the direction of the applied magnetic field, see Figure 1.

Depending on their preparation method, ferrogels can turn into isotropic or anisotropic gels (Bohlius et al., 2004). In the case of an isotropic ferrogel, the magnetic

particles are randomly dispersed throughout the polymer matrix, see Figure 2(a). Isotropic ferrogels are only sensitive to the gradient of a magnetic field showing no remnant magnetization (Jarkova et al., 2003). The magnetic particles in the anisotropic ferrogels, on the other hand, form chain-like structures as a consequence of placing the ferrogel in a magnetic field in the course of preparation (Bohlius et al., 2004; Collin et al., 2003;

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Figure 1. Ferrogel before application of the magnetic field (left). Deformation of a ferrogel in the magnetic field (H) (right).

Figure 2. Isotropic (a) versus anisotropic (b) ferrogel.

Varga et al., 2003), see Figure 2(b). Anisotropic ferrogels are therefore not only responsive to a homogeneous magnetic field, but also show magnetization due to shear deformation (Bohlius et al., 2004).

Ferrogels are often characterized by their low stiffness (below 10 kPa), large deformation of up to 40%, and high water absorption (Faidley et al., 2010; Han et al., 2011; Park et al., 2008; Raikher and Stolbov, 2003, 2005; Zrínyi et al., 1997). Owing to their unique properties, ferrogels are suitable for a variety of mechanical (An and Shaw, 2003; Kato et al., 1997; Meng and Hu, 2010; Mitwalli et al., 1997; Monz et al., 2008; Zimmermann et al., 2011, 2006; Zrínyi et al., 1998) and medical (Chan et al., 2013; Lao and Ramanujan, 2004; Liu et al., 2008, 2006; Qin et al., 2009; Satarkar and Hilt, 2008; Thévenot et al., 2013) applications.

Shortly after the introduction of ferrogels in the late 90s, the first theoretical models were suggested at macroscopic level for both isotropic and anisotropic ferrogels. Raikher and Rusakov (2001) were among the first to introduce a magnetomechanical model for ferrogels with a focus on magnetodynamics of viscoelastic ferrogels. Ever since, several other models have also been proposed at both small and finite strains (Allahyarov et al., 2014; Diguët et al., 2009; Filipcsei and Zrínyi, 2010; Morozov et al., 2009; Raikher and Stolbov, 2003, 2005, 2008; Voltairas et al., 2003; Zubarev, 2012; Zubarev and Elkady, 2014). Considering the viscoelastic response of ferrogels, a few efforts have also been devoted to developing magneto-viscoelastic models (Faidley et al., 2010; Han et al., 2011).

Several authors made use of a macroscopic hydrodynamic-like model to describe the behavior of

isotropic ferrogels (Jarkova et al., 2003), uniaxial ferrogels (Bohlius et al., 2004), and recently ferronematic gels (Brand and Pleiner, 2014). In this modeling approach, magnetization is considered as an additional degree of freedom.

Some researchers, on the other hand, proposed ferrogel models at microscopic and mesoscopic scales. In these models, the magnetic particles were modeled as hard spheres along with harmonic springs, resembling the elastic matrix (Annunziata et al., 2013; Sánchez et al., 2013; Tarama et al., 2014; Weeber et al., 2012; Wood and Camp, 2011). Some authors used numerical tools, for example, Monte Carlo simulations (Wood and Camp, 2011) or Molecular Dynamics (Weeber et al., 2012) to simulate the magnetoelastic behavior of ferrogels. Recently a scale-bridging model has also been suggested by Menzel (2014) to bridge between the microscopic level and the macroscopic scale.

In the majority of the existing models (Jarkova et al., 2003; Raikher and Stolbov, 2003; Voltairas et al., 2003) only the polymer gel as a whole and one species of magnetic particle is considered. In the present study, ferrogels are considered as multicomponent media consisting of a polymer network (P), fixed magnetic particles (f), mobile magnetic particles (m), and liquid (L). By considering ferrogels as a multicomponent media, interaction among constituents of ferrogels can be captured. This may help with the understanding of the process occurring inside ferrogels under the influence of external stimuli, such as magnetic fields.

The motivation behind this research stems from the fact that despite recent developments in modeling the magneto-mechanical behavior of ferrogels, there have hardly been any attempts to propose a macroscopic model based on the multiphase, multicomponent nature of ferrogels. A similar modeling approach has recently been reported for *electrically* stimulated gels (Attaran et al., 2015). The current work intends to propose a model for *magnetically* stimulated gels (i.e. ferrogels) in a similar fashion.

Our ultimate goal is to obtain a magneto-mechanical formulation capable of describing mechanical deformation of the ferrogel placed in a magnetic field. In

addition, the influence of the magnetic field on the material consisting of magnetic particles is sought.

By imposing appropriate assumptions, a reduced model for ferrogels is obtained. The reduced model consists only of a polymer network (P) and fixed magnetic particles (f). It will be demonstrated that the reduced model is comparable to the existing ones in the literature, for example, Jarkova et al. (2003) and Eringen and Maugin (1990). This reduced model is further solved using the finite element method (FEM) in 2D where the only degrees of freedom are mechanical displacement and magnetic potential.

This article is structured as follows: In section ‘Theory’ the systematic development of a continuum model for ferrogels is presented. This includes a brief description of the kinematics of multicomponent systems and postulation of the respective balance equations. Thermodynamically consistent constitutive laws are also derived in this section. A reduced model for ferrogels is introduced in section ‘Reduced model for ferrogels’ where a suitable free energy function is given and the respective constitutive laws are subsequently derived. Using the constitutive laws and balance equations presented in section ‘Theory’, a coupled magneto-mechanical formulation for ferrogels is presented. The basic settings for FEM implementation and the numerical simulation results are given in section ‘Deformation of a ferrogel in a magnetic field’. This work is concluded in the final section with a short summary and highlights of the current article.

Theory

General overview

It is the purpose of this section to present a systematic development of a continuum model for ferrogels within the framework of the theory of mixtures (Ehlers, 2002) and the electromagnetism of continua (de Groot and Suttorp, 1972; Eringen and Maugin, 1990). This model is used to understand the process occurring inside the gel under the influence of an externally applied magnetic field. Owing to its multiphase, multicomponent nature, it is assumed that our system—here referred to as domain $\mathcal{B}$ —consists of (i) a gel phase ($\mathcal{G}$) and (ii) a surrounding fluid ($\mathcal{F}^{ex}$), see Figure 3, such that

$$\mathcal{B} = \mathcal{G} \cup \mathcal{F}^{ex} \quad \text{and} \quad \mathcal{G} \cap \mathcal{F}^{ex} = \emptyset$$

The gel phase ($\mathcal{G}$) and the surrounding fluid ($\mathcal{F}^{ex}$) are enclosed by the surfaces $\partial\mathcal{G}$, and $\partial\mathcal{F}^{ex}$, respectively. The boundary between the gel phase and the surrounding fluid moves at a velocity of w_i . The boundary of the whole domain, $\partial\mathcal{B}$, is defined as

$$\partial\mathcal{B} = \partial\mathcal{F}^{ex} \setminus \partial\mathcal{G}$$

Figure 3. Decomposition of the system ($\mathcal{B}$) into a gel phase ($\mathcal{G}$) and the surrounding fluid ($\mathcal{F}^{ex}$). The boundary between the gel and the surrounding fluid moves at velocity w_i .

Figure 4. Microstructure of the gel phase; it is composed of a polymer network (P), fixed magnetic particles (f) attached to the polymer network, collectively representing the solid phase ($\mathcal{S}$), and an interstitial fluid phase ($\mathcal{F}^{int}$) containing a liquid (L) which is a carrier of the mobile magnetic particles (m).

In the above definitions $\cup$ indicates the union of two sets, $\cap$ the intersection of two sets, $\emptyset$ the empty set and $\setminus$ indicates the remainder of two sets.

In this research the gel phase ($\mathcal{G}$) and the jump condition to the surrounding external fluid ($\mathcal{F}^{ex}$) will only be given. The gel phase is a composition of a solid phase ($\mathcal{S}$) and an interstitial fluid phase ($\mathcal{F}^{int}$), see Figure 4. The constituents of the gel phase are summarized in Table 1. In the rest of the text the gel phase is referred to as ferrogel.

Basic settings

The problem is formulated in a Cartesian coordinate system. For this, index notation is used. A general vector $\mathbf{u}$ is defined by

$$\mathbf{u} = \sum_{i=1}^3 u_i \mathbf{e}_i = u_i \mathbf{e}_i \quad (1)$$

where $\mathbf{e}_i$ are the unit vectors of the coordinate system and u_i are the coordinates of the vector $\mathbf{u}$ in the x_i -direction. The Einstein summation convention, that is, the summation over identical indices in a term is performed.

The differential operator $(\cdot)_{,i}$ is accordingly defined using index notation as follows

Table 1. Constituents of solid phase and fluid phase.

Solid phase ($\mathcal{S}$)	Interstitial fluid ($\mathcal{F}^{\text{int}}$)
Polymer network (P)	Liquid (L)
Fixed magnetic particles (f)	Mobile magnetic particles (m)

$$\frac{\partial(\cdot)}{\partial x_i} = \partial_i(\cdot) = (\cdot)_{,i} \quad (2)$$

Throughout the text α refers to components of the ferrogel ($\alpha = \text{P, f, m, L}$), β indicates fixed and mobile magnetic particles ($\beta = \text{f, m}$) and γ points to fixed and mobile magnetic particles as well as the Liquid ($\gamma = \text{f, m, L}$).

Kinematics

To describe the motion of ferrogels, the kinematics of multicomponent systems are used. The kinematics are based on the concept of superimposed and interacting continua (Ehlers, 2002). In this concept it is assumed that there exists a motion function κ_i^α for each component α inside the ferrogel. This motion function maps each material point Q^α (at $t_0 = 0$) onto a common spatial point in the current configuration (at $t > 0$), that is

$$x_i = \kappa_i^\alpha(X_I^\alpha, t) \quad ; \quad \alpha = 1, \dots, N \quad (3)$$

where X_I^α is the position vector of the material point Q^α at $t_0 = 0$, x_i the position vector of all components in the current configuration at $t > 0$, N is the total number of components in the medium and t is the time. Conversely, we define the inverse of the motion mapping as

$$X_I^\alpha = \kappa_I^{\alpha-1}(x_i, t) \quad ; \quad \alpha = 1, \dots, N \quad (4)$$

As an example, the motion of a binary mixture is illustrated in Figure 5.

It follows from equations (3) and (4), and Figure 5 that each component of the ferrogel has a unique reference point at $t_0 = 0$. At $t > 0$, however, one spatial point represents the position of all the components (Ehlers, 2002).

Following this assumption, the displacement vector u_i^α of the α th component is given as the difference between the current and the initial position

$$u_i^\alpha = x_i - X_I^\alpha \delta_{ji} \quad (5)$$

where δ_{ji} is the Kronecker delta defined as

$$\delta_{ji} = \begin{cases} 1 & ; i = j \\ 0 & ; i \neq j \end{cases}$$

We can now define the deformation gradient tensor F_{iJ}^α of the α th component and its inverse, $F_{iJ}^{\alpha-1}$ as

Figure 5. Motion of a binary system.

$$F_{iJ}^\alpha = \frac{\partial x_i}{\partial X_J^\alpha} \quad \text{and} \quad F_{iJ}^{\alpha-1} = \frac{\partial X_J^\alpha}{\partial x_i} \quad (6)$$

The determinant, J^α , of F_{iJ}^α can accordingly be defined as

$$J^\alpha = |F_{iJ}^\alpha| = \left| \frac{\partial x_i}{\partial X_J^\alpha} \right| > 0 \quad (7)$$

where $| \cdot |$ is the determinant of a second order tensor. The Green–Lagrange strain tensor of the α th component, E_{IJ}^α , that is, the strain tensor in the reference configuration, is defined as

$$E_{IJ}^\alpha = \frac{1}{2} (F_{kI}^\alpha F_{kJ}^\alpha - \delta_{IJ}) \quad (8)$$

For the infinitesimal strain assumption (i.e. $\|u_{i,j}\| \ll 1$), the distinction between the current and the reference configurations is negligible and the infinitesimal strain tensor ε_{ij}^α is defined as

$$\varepsilon_{ij}^\alpha = \frac{1}{2} (u_{i,j}^\alpha + u_{j,i}^\alpha) \quad (9)$$

The infinitesimal strain assumption further leads to the linearized determinant of F_{iJ}^α being

$$J^\alpha = |F_{iJ}^\alpha| \approx 1 + u_{i,i}^\alpha = 1 + \varepsilon_{ii}^\alpha \quad (10)$$

The velocity of the α th component v_i^α is defined as

$$v_i^\alpha = \frac{\partial \kappa_i^\alpha(X_I^\alpha, t)}{\partial t} = \frac{\partial x_i}{\partial t}(X_I^\alpha, t) \quad (11)$$

In the present work, the polymer network (P) and the fixed magnetic particles (f) have velocities equal to that of the solid phase ($\mathcal{S}$), that is, $v_i^P = v_i^f = v_i^S$. This consideration becomes essentially important since fixed magnetic particles are attached to the polymer network and are not just trapped within the networks. That means both the polymer network and fixed magnetic particles have the same velocity.

In addition, the velocity of the interstitial fluid $v_i^{\mathcal{F}^{\text{int}}}$ is defined as $v_i^{\mathcal{F}^{\text{int}}} = \frac{1}{\rho^{\mathcal{F}^{\text{int}}}} (\rho^L v_i^L + \rho^m v_i^m)$, where $\rho^{\mathcal{F}^{\text{int}}}$ is the density of the interstitial fluid, ρ^L is the density of

the liquid, v_i^L is the velocity of the liquid, ρ^m is the density of the mobile particles, and v_i^m is the velocity of the mobile particles. In this context, $v_i^{\mathcal{F}^{\text{int}}}$ is regarded as the barycentric velocity of the interstitial fluid.

Basic equations

In this section, basic equations for a magneto-mechanical problem are described. The formulations are presented in Lorentz–Heaviside units.

To postulate balance relations for the mixture, we first express partial balance relations. Imposing restrictions and assumptions of the theory of mixtures, balance relations of the whole mixture (ferrogel) are derived.

Maxwell's equations. For the considered ferrogel we assume that there exists no electric charge and/or electric flux, and the ferrogel is non-polarizable. These assumptions lead to the expression of Maxwell's equations of magnetostatics as

$$\mathbb{B}_{i,i} = 0 \text{ in } \mathcal{G} \quad (12a)$$

$$e_{ijk}\mathbb{H}_{k,j} = 0 \text{ in } \mathcal{G} \quad (12b)$$

with the relationship

$$\mathbb{H}_i = \mathbb{B}_i - \mathbb{M}_i \text{ in } \mathcal{G} \quad (13)$$

Here, $\mathbb{B}_i$ is the magnetic induction vector, $\mathbb{H}_i$ the magnetic field strength vector and $\mathbb{M}_i$ the magnetization vector.

The jump conditions are given by

$$[\mathbb{B}_i]n_i^{\mathcal{G}} = 0 \text{ on } \partial\mathcal{G} \cap \partial\mathcal{F}^{\text{ex}} \quad (14a)$$

$$e_{ijk}[\mathbb{H}_k]n_j^{\mathcal{G}} = 0 \text{ on } \partial\mathcal{G} \cap \partial\mathcal{F}^{\text{ex}} \quad (14b)$$

where, e_{ijk} is the Levi–Civita symbol defined by $e_{ijk} = -e_{jik} = -e_{ikj} = -e_{kji}$, and $e_{123} = 1$, $[\] = \text{variable}^{\mathcal{G}} - \text{variable}^{\mathcal{F}^{\text{ex}}}$, and $n^{\mathcal{G}}$ is the unit normal vector defined positive in the outwards direction at the ferrogel boundary.

For the cases where no electric current is present, the magnetostatic problem can be described using a scalar magnetic potential Ψ^{mag} such that

$$\mathbb{H}_i = -\Psi_{,i}^{\text{mag}} \text{ in } \mathcal{G} \quad (15)$$

with the jump condition given by

$$[\Psi^{\text{mag}}] = 0 \text{ on } \partial\mathcal{G} \cap \partial\mathcal{F}^{\text{ex}} \quad (16)$$

Using equation (15), equation (12b) is identically satisfied.

Balance of mass. The partial balance of mass for the α th component ($\alpha = \text{P, f, m, L}$) is expressed as

$$\frac{\partial \rho^\alpha}{\partial t} + (\rho^\alpha v_i^\alpha)_{,i} = \hat{\rho}^\alpha \text{ in } \mathcal{G} \quad (17)$$

with the jump condition expressed as

$$[\rho^\alpha(v_i^\alpha - w_i)]n_i^{\mathcal{G}} = \tilde{\rho}^\alpha \text{ on } \partial\mathcal{G} \cap \partial\mathcal{F}^{\text{ex}} \quad (18)$$

where ρ^α is the mass density, $\hat{\rho}^\alpha$ is the mass production density, and $\tilde{\rho}^\alpha$ is the mass production per area of the α th component.

In the theory of mixtures the sum of partial balance relations over all constituents has to be equal to the single-phase balance (Ehlers, 2002). Using

$$\sum_\alpha \rho^\alpha = \rho \quad (19a)$$

$$\sum_\alpha \rho^\alpha v_i^\alpha = \rho v_i \quad (19b)$$

$$\sum_\alpha \hat{\rho}^\alpha = 0 \quad (19c)$$

$$\sum_\alpha \tilde{\rho}^\alpha = 0 \quad (19d)$$

one obtains the balance of mass for the ferrogel as

$$\frac{\partial \rho}{\partial t} + (\rho v_i)_{,i} = 0 \text{ in } \mathcal{G} \quad (20)$$

with the jump condition expressed as

$$[\rho(v_i - w_i)]n_i^{\mathcal{G}} = 0 \text{ on } \partial\mathcal{G} \cap \partial\mathcal{F}^{\text{ex}} \quad (21)$$

where ρ is the mass density of the ferrogel, v_i the barycentric velocity of the ferrogel, and w_i is the velocity of the moving boundary.

Equations (19c) and (19d) indicate that there is neither a mass production nor a mass sink in the ferrogel as a whole. The mass production density of component α , $\hat{\rho}^\alpha$, is included in order to later be able to model, for example, a transition of fixed magnetic particles to mobile magnetic particles, and vice versa.

It is often of a greater interest to restate the balance of mass in terms of specific density of the individual components, $\tilde{c}^\alpha$ defined as

$$\tilde{c}^\alpha = \frac{\rho^\alpha}{\rho} \quad (22)$$

where $\sum_\alpha \tilde{c}^\alpha = 1$, indicating there are no voids in the ferrogel and it is completely saturated.

Defining the material time derivative with respect to the barycentric velocity, v_i as

$$(\dot{\ }) = \frac{\partial(\)}{\partial t} + (\)_{,i} v_i$$

equation (17) is now rewritten as

$$\rho \dot{\tilde{c}}^\alpha + j_{i,i}^\alpha = \hat{\rho}^\alpha \text{ in } \mathcal{G} \quad (23)$$

with the jump condition given as

$$[[j_i^\alpha + \rho \tilde{c}^\alpha(v_i - w_i)]] n_i^G = \tilde{\rho}^\alpha \quad \text{on } \partial\mathcal{G} \cap \partial\mathcal{F}^{ex} \quad (24)$$

where j_i^α is the mass flux vector of the α th component defined as

$$j_i^\alpha = \rho^\alpha(v_i^\alpha - v_i) \quad \text{in } \mathcal{G} \quad (25)$$

and $\sum_\alpha j_i^\alpha = 0$.

Balance of linear momentum. Imposing the following restrictions—according to the theory of mixtures (Ehlers, 2002)

$$\rho b_j = \sum_\alpha (\rho^\alpha b_j^\alpha + \hat{\rho}^\alpha v_j^\alpha) \quad (26a)$$

$$\sigma_{ij} = \sum_\alpha (\sigma_{ij}^\alpha - \rho^\alpha(v_i^\alpha - v_i)(v_j^\alpha - v_j)) \quad (26b)$$

the balance of linear momentum for the ferrogel is given as

$$\rho \dot{v}_i - \sigma_{ji,j} = \rho b_i + b_i^E \quad \text{in } \mathcal{G} \quad (27)$$

Here, b_j is the body force vector of the ferrogel, b_j^α the body force vector of the α th component, σ_{ij} the mechanical stress tensor of the ferrogel, σ_{ij}^α the mechanical stress tensor of the α th component, σ_{ji}^E the Maxwell's stress tensor expressed in our context as (de Groot and Suttrop, 1972; Eringen and Maugin, 1990)

$$\sigma_{ji}^E = \mathbb{B}_j \mathbb{H}_i - \frac{\delta_{ij}}{2} (\mathbb{B}^2 - 2\mathbb{M}_q \mathbb{B}_q) \quad \text{in } \mathcal{G} \quad (28)$$

and b_i^E is the magnetic body force vector given by

$$b_i^E = \sigma_{ji,j}^E = \mathbb{M}_j \mathbb{B}_{j,i} \quad \text{in } \mathcal{G} \quad (29)$$

Please note that the second term on the right hand side of equation (26a) stems from mass production of different components α multiplied with respective velocities v_i^α .

The jump condition for the balance of linear momentum is now stated as

$$[[\rho v_j(w_i - v_i) + \sigma_{ij} + \sigma_{ij}^E]] n_i^G = -\tilde{b}_j \quad \text{on } \partial\mathcal{G} \cap \partial\mathcal{F}^{ex} \quad (30)$$

where $\tilde{b}_j$ is the prescribed body force per area of the ferrogel.

Balance of energy. The balance of internal energy for the ferrogel is expressed as

$$\rho \dot{u} + q_{i,i} - \sigma_{ij} v_{j,i} = -\mathbb{M}_i \dot{\mathbb{B}}_i + \rho \bar{r} + \sum_\alpha j_i^\alpha b_i^\alpha \quad \text{in } \mathcal{G} \quad (31)$$

with

$$\rho \bar{u} = \rho u - \mathbb{M}_i \mathbb{B}_i \quad \text{and}$$

$$\rho \bar{r} = \rho r - \sum_\alpha [j_i^\alpha b_i^\alpha + \hat{\rho}^\alpha(u^\alpha + \frac{1}{2}(v_i^\alpha - v_i)(v_i^\alpha - v_i))]$$

where, u is the specific internal energy of the ferrogel, q_i is the heat flux vector of the ferrogel, and r is the specific external heat supply.

In postulation of equation (31), the relations

$$\rho u = \sum_\alpha [\rho^\alpha u^\alpha + \frac{\rho^\alpha}{2}(v_i^\alpha - v_i)(v_i^\alpha - v_i)] \quad (32a)$$

$$\rho r = \sum_\alpha [\hat{u}^\alpha + \rho^\alpha b_i^\alpha(v_i^\alpha - v_i) + \hat{\rho}^\alpha(u^\alpha + \frac{1}{2}(v_i^\alpha - v_i)(v_i^\alpha - v_i))] \quad (32b)$$

$$q_i = \sum_\alpha [q_i^\alpha - \sigma_{ji}^\alpha(v_j^\alpha - v_j) + \rho^\alpha u^\alpha(v_i^\alpha - v_i) + \frac{\rho^\alpha}{2}(v_i^\alpha - v_i)(v_j^\alpha - v_j)(v_j^\alpha - v_j)] \quad (32c)$$

are used in the framework of the theory of mixtures (Ehlers, 2002), where u^α is the specific internal energy, $\hat{u}^\alpha$ the specific internal energy production, and q_i^α is the heat flux vector of the α th component. The corresponding jump condition is then expressed as

$$\left[\left[\rho(u + \frac{1}{2}v^2)(w_i - v_i) + w_i \frac{1}{2}\mathbb{B}^2 - q_i + \sigma_{ij}v_j \right] n_i^G \right] = -\tilde{b}_i w_i - \tilde{r} \quad \text{on } \partial\mathcal{G} \cap \partial\mathcal{F}^{ex} \quad (33)$$

where $\tilde{r}$ is the specific external heat supply per area of ferrogel.

Balance of internal angular momentum. The balance of internal angular momentum for the ferrogel is expressed as (de Groot and Suttrop, 1972)

$$\rho \left(\frac{\dot{S}_i}{\rho} \right) + J_{pi,p}^S - e_{ijk}(\sigma_{jk} + \sigma_{jk}^E) = m_i - e_{ipq}x_p \rho b_q \quad \text{in } \mathcal{G} \quad (34)$$

where S_i is the internal angular momentum density, J_{ji}^S the flux of the internal angular momentum, $m_i = \sum_\alpha m_i^\alpha$ the external angular momentum acting on the ferrogel, and m_i^α is the angular momentum acting on the α th component. The corresponding jump condition is given by

$$[[[S_i(v_j - w_j) + J_{ji}^S]] n_j^G] = \tilde{m}_i - e_{ipq}x_p \tilde{b}_q \quad \text{on } \partial\mathcal{G} \cap \partial\mathcal{F}^{ex} \quad (35)$$

where $\tilde{m}_i$ is the external angular momentum acting on the boundary of the ferrogel. Please note that by setting $m_i = e_{ipq}x_p \rho b_q$ and $\tilde{m}_i = e_{ipq}x_p \tilde{b}_q$ the right-hand sides of equations (34) and (35) vanish. That means no external angular momentum acts in the volume and on the boundary of the ferrogel. Additional discussion on

balance of internal angular momentum with respect to $\frac{S_i}{\rho}$ as an internally varying variable is elaborated on in section 'Reformulation of balance of internal angular momentum' and Appendix 1.

Balance of entropy. The balance of entropy for the ferrogel is expressed as

$$\rho \dot{\eta} + \eta_{i,i} = \hat{\eta}^{in} + \hat{\eta}^s \quad \text{in } \mathcal{G} \quad (36)$$

with the jump condition formulated as

$$[\rho \eta (v_i - w_i) + \eta_i] n_i^{\mathcal{G}} = \tilde{\eta}^{in} + \tilde{\eta}^s \quad \text{on } \partial \mathcal{G} \cap \partial \mathcal{F}^{ex} \quad (37)$$

where η is the specific entropy of the ferrogel, η_i is the conductive entropy density flux vector, $\hat{\eta}^{in}$ is the induced part of the entropy production density, and $\hat{\eta}^s$ is the spontaneous entropy production density of the system. The $(\sim)$ in equation (37) represents the corresponding entropy production term per area of the ferrogel.

Constitutive relations

In this section thermodynamically consistent constitutive laws are introduced. By incorporation of constitutive laws into basic equations, the governing equations of the ferrogel are obtained.

Second law of thermodynamics. Thermodynamically consistent constitutive relations must satisfy the second law of thermodynamics. This law is fulfilled if the spontaneous entropy production density $\hat{\eta}^s$ is positive, that is

$$\hat{\eta}^s \geq 0 \quad \text{in } \mathcal{G} \quad (38)$$

The specific free energy f of the ferrogel is linked to the internal energy density $\bar{u}$ by the following Legendre-Transformation

$$f = \bar{u} - T\eta + \mathbb{B}_i m_i \quad (39)$$

Equation (31) can be written as

$$\begin{aligned} \rho T \dot{\eta} = & \rho \bar{r} - q_{i,i} + \sigma_{ij} v_{j,i} + \sum_{\alpha} j_i^{\alpha} b_i^{\alpha} \\ & + \rho \mathbb{B}_i \dot{m}_i - \rho \dot{f} - \rho \eta \dot{T} \end{aligned} \quad (40)$$

where T is temperature and m_i is the specific magnetization defined as

$$m_i = \frac{\mathbb{M}_i}{\rho} \quad \text{in } \mathcal{G} \quad (41)$$

As $T > 0$, the second law of thermodynamics, inequality (38), with the help of equations (36) and (40) is now expressed as

$$\begin{aligned} T \dot{\eta}^s = & T \eta_{i,i} - T \rho \dot{\eta}^{in} + \rho \bar{r} - q_{i,i} + \sigma_{ij} v_{j,i} + \rho \mathbb{B}_i \dot{m}_i \\ & + \sum_{\alpha} j_i^{\alpha} b_i^{\alpha} - \rho \eta \dot{T} - \rho \dot{f} \geq 0 \quad \text{in } \mathcal{G} \end{aligned} \quad (42)$$

For the considered ferrogel, the specific free energy $f = f(F_{iJ}^P, \tilde{c}^{\gamma}, m_i^{\beta}, m_{i,j}^{\beta}, T)$ is now assumed to be a function of the following independent variables

F_{iJ}^P : deformation gradient tensor of the polymer,
 $\tilde{c}^{\gamma}$: specific density of the component γ ,
 m_i^{β} : specific magnetization of the component β ,
 $m_{i,j}^{\beta}$: gradient of the specific magnetization of the component β ,
 T : temperature,

where $\gamma = f, m, L$ and $\beta = f, m$. A motivation for the choice of F_{iJ}^P and $\tilde{c}^{\gamma}$ as independent variables is outlined in Appendix 2.

Now to fulfill the second law of thermodynamics inequality (42), it is sufficient to (i) introduce the constitutive equations as

$$\sigma_{ij} = \overset{R}{\sigma}_{ij} + \overset{D}{\sigma}_{ij} = \rho \frac{\partial f}{\partial F_{jI}^P} F_{iI}^P - \sum_{\beta} \overset{R}{\mathcal{B}}_{iq}^{\beta} m_{q,j}^{\beta} + \overset{D}{\sigma}_{ij} \quad \text{in } \mathcal{G} \quad (43a)$$

$$\mu^{\gamma} = \frac{\partial f}{\partial \tilde{c}^{\gamma}} \quad \text{in } \mathcal{G} \quad (43b)$$

$$\mathbf{b}_i^{\beta} = \overset{R}{\mathbf{b}}_i^{\beta} + \overset{D}{\mathbf{b}}_i^{\beta} = - \frac{\partial f}{\partial m_i^{\beta}} + \overset{D}{\mathbf{b}}_i^{\beta} \quad \text{in } \mathcal{G} \quad (43c)$$

$$\mathcal{B}_{ji}^{\beta} = \overset{R}{\mathcal{B}}_{ji}^{\beta} + \overset{D}{\mathcal{B}}_{ji}^{\beta} = \rho \frac{\partial f}{\partial m_{i,j}^{\beta}} + \overset{D}{\mathcal{B}}_{ji}^{\beta} \quad \text{in } \mathcal{G} \quad (43d)$$

$$\eta = - \frac{\partial f}{\partial T} \quad \text{in } \mathcal{G} \quad (43e)$$

where μ^{γ} is the chemical potential of the γ th component, $\mathbf{b}_i^{\beta}$ is the local magnetic induction vector of the β th component, and $\mathcal{B}_{ji}^{\beta}$ is the exchange force tensor (a second order tensor related to the gradient of the specific magnetization (Eringen and Maugin, 1990)) of the β th component, and (ii) satisfy the following inequality

$$\begin{aligned} & \overset{D}{\sigma}_{ij} v_{j,i} - \sum_{\beta} \dot{m}_i^{\beta} \rho \overset{D}{\mathbf{b}}_i^{\beta} + \sum_{\beta} (\dot{m}_i^{\beta})_{,j} \overset{D}{\mathcal{B}}_{ji}^{\beta} \\ & - \frac{T_{,j}}{T} \bar{q}_j + j_q^P [b_q^P + \frac{1}{\rho^P} h_{iJ}^P F_{iJ,q}^P + \frac{1}{\rho^P} (h_{qJ}^P F_{jJ}^P)_{,j}] \\ & + \sum_{\alpha} j_i^{\alpha} (b_i^{\alpha} - \mu_{,i}^{\alpha}) \\ & + \rho \sum_{\beta} \dot{m}_i^{\beta} [\mathbb{B}_i + \mathbf{b}_i^{\beta} + \frac{1}{\rho} \mathcal{B}_{ji}^{\beta}] \geq 0 \quad \text{in } \mathcal{G} \end{aligned} \quad (44)$$

where

$$h_{iJ}^P = \rho \frac{\partial f}{\partial F_{iJ}^P} \quad (45a)$$

$$\bar{q}_j = q_j + h_{ij}^p F_{ij}^p \frac{j_j^p}{\rho^p} - \sum_{\gamma} \mu^{\gamma} j_j^{\gamma} + \sum_{\beta} \mathcal{B}_{ji}^{\beta} \dot{m}_i^{\beta} \quad (45b)$$

The derivation of this inequality is outlined in Appendix 3. In the definitions given above, the stress tensor σ_{ij} , the local magnetic induction vector $\mathbf{b}_i^{\beta}$, and the exchange force tensor $\mathcal{B}_{ji}^{\beta}$ contain (i) a reversible potential contribution ($\overset{R}{}$) and (ii) a remaining part ($\overset{D}{}$). Please note that the reversible potential part results from differentiation of the specific free energy function. The remaining part contains a reversible, non-potential contribution and a dissipative part (compare this with Jarkova et al., 2003, Bohlius et al., 2004, and Brand and Pleiner, 2014).

The dissipative part of the local magnetic induction vector of magnetic particles, $\mathbf{b}_i^{\beta}$, pertains to the “so-called spin-lattice relaxation phenomenon of the magnetic particles” (Eringen and Maugin, 1990).

In our approach we restrict the formulation to reversible terms $\overset{R}{\sigma}_{ij}$, $\overset{R}{\mathbf{b}}_i^{\beta}$, $\overset{R}{\mathcal{B}}_{ij}^{\beta}$ —resulting from differentiation of the specific free energy function—by explicitly setting reversible non-potential and dissipative parts equal to zero, that is, $\overset{D}{\sigma}_{ij} = 0$, $\overset{D}{\mathbf{b}}_i^{\beta} = 0$, $\overset{D}{\mathcal{B}}_{ji}^{\beta} = 0$. We also assume zero flux of the polymer network, that is, $j_q^p = 0$, and zero body force of the γ th components, that is, $b_i^{\gamma} = 0$.

We further assume that the process is isothermal and therefore $\dot{T} = 0$ and $T_{,j} = 0$. Inequality (44) reduces now to

$$-\sum_{\gamma} j_i^{\gamma} \mu_{,i}^{\gamma} + \rho \sum_{\beta} \dot{m}_i^{\beta} [\mathbb{B}_i + \mathbf{b}_i^{\beta} + \frac{1}{\rho} \mathcal{B}_{ji,j}^{\beta}] \geq 0 \text{ in } \mathcal{G} \quad (46)$$

By introducing

$$j_i^{\gamma} = -D^{\gamma} \mu_{,i}^{\gamma} \text{ in } \mathcal{G} \quad (47)$$

where D^{γ} is the diffusion coefficient of the γ th component, the first term in (46) is always positive. The discussion on the rest of the inequality is given in Appendix 1.

Reformulation of balance of internal angular momentum. Additional discussion on balance of internal angular momentum with respect to $\frac{\mathbb{B}_i}{\rho}$ as an internally varying variable is presented in this section. To fulfill inequality (46) and since $\mathbf{b}_i^{\beta}$ and $\mathcal{B}_{ji}^{\beta}$ are defined using constitutive equations

$$\sum_{\beta} \dot{m}_i^{\beta} [\mathbb{B}_i + \mathbf{b}_i^{\beta} + \frac{1}{\rho} \mathcal{B}_{ji,j}^{\beta}] = 0 \text{ in } \mathcal{G} \quad (48)$$

is prescribed a priori.

In equation (48), sum of the magnetic induction, the local magnetic induction and the divergence of the exchange force tensor is referred to as the effective magnetic field (Eringen and Maugin, 1990). Assuming

equation (47) is fulfilled, equation (48) indicates that in order to satisfy inequality (46), the evolution of the specific magnetization must be perpendicular to the effective magnetic field.

It has to be noted that provided the temperature is well below the Curie temperature of the material under consideration (e.g. iron oxide has a Curie temperature of around 500°C (Thévenot et al., 2013)), the specific magnetization may be regarded as saturated with time-independent magnitude (Eringen and Maugin, 1990). Therefore

$$|\mathbf{m}_i^{\beta}| = \text{const.} \text{ in } \mathcal{G} \quad (49)$$

and consequently

$$\dot{\mathbf{m}}_i^{\beta} \mathbf{m}_i^{\beta} = 0 \text{ in } \mathcal{G} \quad (50)$$

as well as

$$\mathbf{m}_{j,i}^{\beta} \mathbf{m}_j^{\beta} = 0 \text{ in } \mathcal{G} \quad (51)$$

Using equations (48) and (50), the evolution of specific magnetization

$$\dot{\mathbf{m}}_i^{\beta} = \gamma^{\beta} e_{ijk} \mathbf{m}_j^{\beta} [\mathbb{B}_k + \mathbf{b}_k^{\beta} + \frac{1}{\rho} \mathcal{B}_{pk,p}^{\beta}] \text{ in } \mathcal{G} \quad (52)$$

is obtained, where γ^{β} is the gyromagnetic coefficient of the β th components ($\beta = \text{f, m}$). It will be explained in Appendix 1 that the balance of angular momentum equation (34) is satisfied using equation (52) if

$$\mathcal{B}_{pk}^{\beta} \mathbf{m}_k^{\beta} = 0 \text{ in } \mathcal{G} \quad (53a)$$

$$e_{ijk} [\sigma_{jk} + \sum_{\beta} (\mathbf{m}_{j,p}^{\beta} \mathcal{B}_{pk}^{\beta} - \mathbf{m}_j^{\beta} \rho \mathbf{b}_k^{\beta})] = 0 \text{ in } \mathcal{G} \quad (53b)$$

are satisfied using the constitutive laws.

Reformulation of the constitutive relations. To fulfill equations (73) and (74), and to ensure the invariance of the free energy function $\mathfrak{f}$ against orthogonal transformations, the independent variables for the isothermal process are now chosen as (Eringen and Maugin, 1990)

$$E_{IJ}^p = \frac{1}{2} (F_{kl}^p F_{kl}^p - \delta_{IJ}) \quad (54a)$$

$$\mathcal{M}_I^{\beta} = \mathbf{m}_j^{\beta} F_{jI}^p \quad (54b)$$

$$\mathcal{M}_{IJ}^{\beta} = \mathbf{m}_{p,r}^{\beta} \mathbf{m}_{p,s}^{\beta} F_{rl}^p F_{sl}^p \quad (54c)$$

so that $\mathfrak{f}$ can be written as

$$\mathfrak{f} = \mathfrak{f}(E_{IJ}^p, \tilde{c}^{\gamma}, \mathcal{M}_I^{\beta}, \mathcal{M}_{IJ}^{\beta}) \quad (55)$$

Constitutive relations in (43) as well as equation (47) are now stated as

$$\sigma_{ij} = \rho \frac{\partial \mathfrak{f}}{\partial E_{IJ}^p} F_{il}^p F_{jl}^p - \rho \sum_{\beta} \mathbf{b}_i^{\beta} \mathbf{m}_j^{\beta} \text{ in } \mathcal{G} \quad (56a)$$

$$\mu^\gamma = \frac{\partial f}{\partial \tilde{c}^\gamma} \quad \text{in } \mathcal{G} \quad (56b)$$

$$\mathbf{b}_i^\beta = -\frac{\partial f}{\partial \mathcal{M}_i^\beta} F_{il}^P \quad \text{in } \mathcal{G} \quad (56c)$$

$$\mathcal{B}_{ij}^\beta = 2\rho \frac{\partial f}{\partial \mathcal{M}_{ij}^\beta} \mathbf{m}_{i,r}^\beta F_{rj}^P F_{jl}^P \quad \text{in } \mathcal{G} \quad (56d)$$

$$j_i^\gamma = -D^\gamma \mu_{,i}^\gamma \quad \text{in } \mathcal{G} \quad (56e)$$

For small strain problems, the variables $F_{ij}^P, E_{IJ}^P, \mathcal{M}_I^\beta, \mathcal{M}_{IJ}^\beta$ are approximated as

$$F_{ij}^P \rightarrow \delta_{ij} + u_{i,j} \quad (57a)$$

$$E_{IJ}^P \rightarrow \varepsilon_{ij}^P \quad (57b)$$

$$\mathcal{M}_I^\beta \rightarrow \mathbf{m}_i^\beta \quad (57c)$$

$$\mathcal{M}_{IJ}^\beta \rightarrow \mathcal{M}_{ij}^\beta = \mathbf{m}_{p,i}^\beta \mathbf{m}_{p,j}^\beta \quad (57d)$$

and the set of constitutive relations may be given as

$$\sigma_{ij} = \rho \frac{\partial f}{\partial \varepsilon_{ij}^P} - \rho \sum_\beta \mathbf{b}_i^\beta \mathbf{m}_j^\beta \quad \text{in } \mathcal{G} \quad (58a)$$

$$\mu^\gamma = \frac{\partial f}{\partial \tilde{c}^\gamma} \quad \text{in } \mathcal{G} \quad (58b)$$

$$\mathbf{b}_i^\beta = -\frac{\partial f}{\partial \mathbf{m}_i^\beta} \quad \text{in } \mathcal{G} \quad (58c)$$

$$\mathcal{B}_{ji}^\beta = 2\rho \frac{\partial f}{\partial \mathcal{M}_{jr}^\beta} \mathbf{m}_{i,r} \quad \text{in } \mathcal{G} \quad (58d)$$

$$j_i^\gamma = -D^\gamma \mu_{,i}^\gamma \quad \text{in } \mathcal{G} \quad (58e)$$

where $f = f(\varepsilon_{ij}^P, \tilde{c}^\gamma, \mathbf{m}_i^\beta, \mathcal{M}_{ij}^\beta)$.

Reduced model for ferrogels

The model presented in the previous section is intended to be simulated numerically in later sections. The extension of this model, however, makes the numerical simulation a tedious task. Owing to these limitations, a reduced model is introduced that is suitable as a starting point for a numerical simulation.

Basic assumptions

Considering the following assumptions, a reduced model for ferrogels is presented in this section that consists only of the solid phase (S), see Figure 6.

- (1) Only isothermal and reversible processes will be considered.
- (2) Mobile magnetic particles are not taken into account.
- (3) It is assumed that no liquid is present.

Figure 6. Microstructure of a reduced model for ferrogels; it is composed of a polymer network (P) and fixed magnetic particles (f) attached to the polymer network.

- (4) Small strain theory is used to describe the deformation of ferrogels.
- (5) Mass density of ferrogels remains constant.

In this section, the primary intention is to demonstrate the functionality of our modeling approach under the assumption of small strain. This assumption, only valid for small deformation and small magnetic field strength, gives an overview of the qualitative performance of the model. It also reduces the calculation time using FEM.

Due to assumptions 3 and 4, balance of mass for fixed magnetic particles (f) needs special treatment as follows. It is shown in Appendix 2 that

$$\rho = \frac{\dot{\rho}^P}{|F_{jj}^P| \tilde{c}^P} \quad (59)$$

where $\dot{\rho}^P$ is the initial mass density of the polymer network (P). In the small strain regime, equation (59) can be written as

$$\rho = \frac{\dot{\rho}^P}{(1 + \varepsilon_{ii}^P) \tilde{c}^P} \quad (60)$$

Since $\tilde{c}^f + \tilde{c}^P = 1$, balance of fixed magnetic particles is now stated as

$$\tilde{c}^f = 1 - \frac{\dot{\rho}^P}{\rho} \frac{1}{1 + \varepsilon_{ii}^P} \quad \text{in } \mathcal{G} \quad (61)$$

The constituents of such ferrogels are summarized in Table 2.

Figure 6 and Table 2 imply that the reduced model contains only the solid phase of the ferrogel.

Superparamagnetic behavior of ferrogels

Despite the ferromagnetic ordering of magnetic particles, the magnetic behavior of a system of nanoparticles exhibits a transition to a quasi-superparamagnetic state. Ferrogels and ferrofluids are good examples of such systems. This transition usually takes place well below the Curie temperature of the material, for example, around room temperature (Veverka et al., 2007).

The critical size of magnetic nanoparticles in such systems, at which this transition occurs, is material

Table 2. Constituents of reduced model for ferrogels.

Solid phase ($\mathcal{S}$)	Interstitial fluid ($\mathcal{F}^{\text{int}}$)
Polymer network (P)	–
Fixed magnetic particles (f)	–

dependent. For example, for magnetite Fe_3O_4 and maghemite $\gamma\text{-Fe}_2\text{O}_3$ the critical size has been reported to be slightly below 20 nm (Veverka et al., 2007). A typical consequence of such a transition is that in the absence of an applied magnetic field, magnetic moments of the particles are randomly oriented, creating zero net magnetization (Eringen and Maugin, 1990).

With no ferromagnetic ordering, exchange interaction between magnetic moments of neighboring particles can be neglected (Eringen and Maugin, 1990). This leads to setting the gradient of the specific magnetization to zero, that is, $\mathbf{m}_{i,j}^\beta = 0$ and hence $\mathcal{B}_{ij}^\beta = 0$.

Should $\frac{1}{\gamma^\beta}$ be extremely small (i.e. large γ^β), $\dot{\mathbf{m}}_i^\beta$ can be neglected [Eringen and Maugin, 1990]. Equation (52) therefore reduces to

$$e_{ijk} \mathbf{m}_j^\beta (\mathbb{B}_k + \mathbf{b}_k^\beta) = 0 \quad (62)$$

In the following a suitable free energy function is formulated and the respective constitutive laws are introduced. The outcome of this section is a coupled magneto-mechanical model to describe the behavior of ferrogels.

Free energy function and constitutive equations

Based on the superparamagnetic behavior of ferrogels and the assumptions of section ‘Basic assumptions’, a polynomial approximation for the specific free energy is presented in this section. Further by assuming a constant density of fixed magnetic particles, this free energy contains all terms of linear and quadratic order in ε_{ij}^P and $\mathbf{m}_i^f$. This free energy represents isotropic material behavior and satisfies parity and time-reversal conditions. The linear contribution containing only ε_{ii}^P is omitted as no eigenstress should occur. Since $\mathbf{m}_i^f \mathbf{m}_i^f = \text{const.}$, this contribution also does not appear in the free energy function.

This approximation also contains cubic terms - with at least one contribution of ε_{ij}^P and one of $\mathbf{m}_i^f$. Setting $\varepsilon_{ij}^P = \varepsilon_{ij}$ and $\mathbf{m}_i^f = \mathbf{m}_i$ such a free energy function is formulated as

$$\rho_0 \mathbf{f} = \frac{1}{2} \mathbb{C}_{ijkl} \varepsilon_{ij} \varepsilon_{kl} + \frac{1}{2} \lambda_{ijkl} \varepsilon_{ij} \mathbf{m}_k \mathbf{m}_l \text{ in } \mathcal{G} \quad (63)$$

where, ρ_0 is the reference mass density, $\mathbb{C}_{ijkl}$ is the fourth order elasticity tensor, and λ_{ijkl} is the magneto-strictive tensor (Eringen and Maugin, 1990). The elasticity tensor $\mathbb{C}_{ijkl}$ and the magneto-strictive tensor λ_{ijkl} have the following structures (Jarkova et al., 2003)

$$\mathbb{C}_{ijkl} = K \delta_{ij} \delta_{kl} + G (\delta_{ik} \delta_{jl} + \delta_{il} \delta_{jk} - \frac{2}{3} \delta_{ij} \delta_{kl}) \quad (64a)$$

$$\lambda_{ijkl} = B_0 \delta_{ij} \delta_{kl} + B_1 (\delta_{ik} \delta_{jl} + \delta_{il} \delta_{jk} - \frac{2}{3} \delta_{ij} \delta_{kl}) \quad (64b)$$

where K is the bulk modulus, G is the second Lamé constant also known as shear modulus, and B_0 and B_1 are the magneto-strictive coefficients.

Assuming $\rho \approx \rho_0$, constitutive equations are now presented for σ_{ij} , and $\mathbf{b}_i$ as

$$\sigma_{ij} = \rho \left[\frac{\partial \mathbf{f}}{\partial \varepsilon_{ij}} - \mathbf{b}_i \mathbf{m}_j \right] \quad (65)$$

$$= \mathbb{C}_{ijkl} \varepsilon_{kl} + \frac{1}{2} \lambda_{ijkl} \mathbf{m}_k \mathbf{m}_l + \lambda_{ijkl} \varepsilon_{kl} \mathbf{m}_q \mathbf{m}_l \text{ in } \mathcal{G}$$

$$\mathbf{b}_i = - \frac{\partial \mathbf{f}}{\partial \mathbf{m}_i} = - \frac{1}{\rho_0} \lambda_{ijkl} \varepsilon_{jk} \mathbf{m}_l \text{ in } \mathcal{G} \quad (66)$$

Please note that constitutive relations defined above contain reversible potential parts only, and therefore the overhead script $\overset{R}{()}$ has been dropped for simplicity.

Coupled magnetomechanical problem for ferrogels

Utilizing Maxwell’s equations and the balance laws presented in section ‘Basic equations’ as well as the constitutive equations introduced in section ‘Free energy function and constitutive equations’, a coupled magneto-mechanical problem is obtained. The summary of field equations, number of equations and number of unknowns of this magneto-mechanical problem is presented in Tables 3 and 4. The corresponding boundary, initial, and jump conditions are summarized in Boxes 1 and 2.

Table 3. Summary of field equations in the three dimensional setting for a coupled magneto-mechanical problem.

Equation	Nr. of equations
Primary equations	
$\mathbb{H}_i = - \Psi_{,i}^{\text{mag}}$	3
$\Psi_{,ii}^{\text{mag}} = \mathbb{M}_{i,i}$	1
$\sigma_{ji,j} + \mathbb{M}_{j,i} \mathbb{B}_{j,i} = 0$	3
Auxiliary equations	
$\varepsilon_{ij} = \frac{1}{2} (u_{i,j} + u_{j,i})$	6
$\mathbb{M}_i = \rho \mathbf{m}_i$	3
$\mathbb{H}_i = \mathbb{B}_i - \mathbb{M}_i$	3
Constitutive equations	
$e_{ijk} \mathbf{m}_j (\mathbb{B}_k + \mathbf{b}_k) = 0$	3
$\sigma_{ij} = \mathbb{C}_{ijkl} \varepsilon_{kl} + \frac{1}{2} \lambda_{ijkl} \mathbf{m}_k \mathbf{m}_l + \lambda_{ijkl} \varepsilon_{kl} \mathbf{m}_q \mathbf{m}_l$	9
$\mathbf{b}_i = - \frac{1}{\rho_0} \lambda_{ijkl} \varepsilon_{jk} \mathbf{m}_l$	3
Total number of equations	34

Box 1: Boundary and initial conditions of a magneto-mechanical problem

- Boundary conditions for magnetic variables

$$\begin{aligned}\Psi^{\text{mag}} &= \bar{\Psi}^{\text{mag}} & \text{on } \partial\mathcal{G}_{\Psi^{\text{mag}}} \\ \mathbb{B}_i n_i &= \bar{\mathbb{B}} & \text{on } \partial\mathcal{G}_{\mathbb{B}}\end{aligned}$$

- Boundary conditions for mechanical variables

$$\begin{aligned}u_i &= \bar{u}_i & \text{on } \partial\mathcal{G}_{u_i} \\ (\sigma_{ji} + \sigma_{ji}^E) n_j &= \bar{t}_i & \text{on } \partial\mathcal{G}_{t_i}\end{aligned}$$

with

$$\sigma_{ji}^E = \mathbb{B}_j \mathbb{H}_i - \frac{\delta_{ij}}{2} (\mathbb{B}^2 - 2\mathbb{M}_q \mathbb{B}_q)$$

Box 2: Jump conditions for a magneto-mechanical problem.

- Jump conditions for magnetic variables

$$\begin{aligned}[\![\mathbb{B}_i]\!] n_i^{\mathcal{G}} &= 0 & \text{on } \partial\mathcal{G} \cap \partial\mathcal{F}^{\text{ex}} \\ [\![\Psi^{\text{mag}}]\!] &= 0 & \text{on } \partial\mathcal{G} \cap \partial\mathcal{F}^{\text{ex}}\end{aligned}$$

- Jump conditions for magnetization

$$\left[\left[\frac{\rho}{\gamma} \mathbf{m}_i (v_j - w_j) \right] \right] n_j^{\mathcal{G}} = 0 \quad \text{on } \partial\mathcal{G} \cap \partial\mathcal{F}^{\text{ex}}$$

- Jump conditions corresponding to balance of linear momentum

$$\left[\left[\rho v_j (w_i - v_i) + \sigma_{ij} + \sigma_{ij}^E \right] \right] n_i^{\mathcal{G}} = -\tilde{b}_j \quad \text{on } \partial\mathcal{G} \cap \partial\mathcal{F}^{\text{ex}}$$

Deformation of a ferrogel in a magnetic field

Deformation of a ferrogel is numerically investigated based on the reduced model for ferrogels and the coupled magneto-mechanical formulation presented in the previous section. Therefore we assume that (i) the ferrogel is in equilibrium, and quasi-superparamagnetic behavior of the ferrogel prevails, and (ii) the mass density of the magnetic particles and that of the polymer network remain constant.

Finite element discretization

For numerical treatment of the coupled magneto-mechanical problem for ferrogels, FEM is employed. The resulting nonlinear system of equations is solved using the Newton–Raphson (NR) method. For a 2D case in this work, degrees of freedom are identified as the scalar magnetic potential (Ψ^{mag}) and the displacement ($u = u_x$) in the x - and the displacement ($v = u_y$) in the y -directions. The generalized displacement vector then reads

$$\hat{u}_i = [u \ v \ \Psi^{\text{mag}}]^T \quad (67)$$

In the framework of the NR method, the system of equations to be solved is usually expressed as (Attaran et al., 2015)

$$\mathbf{K}(\hat{u}_i^n) \Delta \hat{u}_i^n = -\mathbf{R}(\hat{u}_i^n) \quad (68)$$

where $\mathbf{K}$ is the Jacobian matrix, $\Delta \hat{u}_i^n$ is the increment of the generalized displacement vector defined as $\Delta \hat{u}_i^n = \hat{u}_i^{n+1} - \hat{u}_i^n$, and $\mathbf{R}$ is the residuum vector containing residuals of the system of differential equations. The Jacobian matrix of the resulting system of equations reads

$$\mathbf{K}(\hat{u}_i^n) = \begin{bmatrix} \mathbf{K}_{uu} & \mathbf{K}_{uv} & \mathbf{K}_{u\Psi^{\text{mag}}} \\ \mathbf{K}_{vu} & \mathbf{K}_{vv} & \mathbf{K}_{v\Psi^{\text{mag}}} \\ \mathbf{K}_{\Psi^{\text{mag}}u} & \mathbf{K}_{\Psi^{\text{mag}}v} & \mathbf{K}_{\Psi^{\text{mag}}\Psi^{\text{mag}}} \end{bmatrix} \quad (69)$$

where $\mathbf{K}_{uu}$, $\mathbf{K}_{vv}$, $\mathbf{K}_{uv}$, and $\mathbf{K}_{vu}$ are the contributions of the displacement field to the Jacobian matrix. $\mathbf{K}_{\Psi^{\text{mag}}\Psi^{\text{mag}}}$ is the contribution of the magnetic field and $\mathbf{K}_{u\Psi^{\text{mag}}}$, $\mathbf{K}_{v\Psi^{\text{mag}}}$, $\mathbf{K}_{\Psi^{\text{mag}}u}$, and $\mathbf{K}_{\Psi^{\text{mag}}v}$ are the contributions to the Jacobian matrix due to the coupling between the magnetic and displacement fields.

Numerical test set-up

In this section the necessary set-up for the numerical simulation is outlined. The simulation domain (15 mm $\times$ 11 mm) consists of a ferrogel strip (5 mm $\times$ 1 mm), which is assumed to be surrounded by a vacuum, see Figure 7. Boundaries of the simulation domain are fixed. The ferrogel is also constrained in the middle to avoid any rigid body rotations. Two test cases will be investigated. In the first test case, an external magnetic field is placed in the x -direction. In the second test case a magnetic field is applied in the y -direction. To apply a magnetic field in the x -direction a magnetic potential is prescribed on the left ($\Psi^{\text{mag}}|_{\text{left}} = 1.5\text{A}$) and right ($\Psi^{\text{mag}}|_{\text{right}} = -1.5\text{A}$) boundaries of the simulation domain. To apply a magnetic field in the y -direction a magnetic potential is prescribed on the top ($\Psi^{\text{mag}}|_{\text{top}} = -1.5\text{A}$) and bottom ($\Psi^{\text{mag}}|_{\text{bottom}} = 1.5\text{A}$) of the simulation domain.

The material parameters used for the numerical simulation are listed in Table 5. Please note that for the numerical simulation SI units are used. The elastic and magnetic material parameters are determined based on the experimental work of Gollwitzer et al. (2008). They studied the deformation of a spherical ferrogel in a homogeneous magnetic field. The elastic parameters, that is, the Young's modulus (E) and Poisson's ratio (ν) are the same as given in Gollwitzer et al. (2008). Young's modulus and Poisson's ratio of Gollwitzer et al. (2008) are also used to calculate the bulk modulus (K) and the shear modulus (G). The magnetostriction

Table 4. Summary of the number of unknowns and their mathematical symbols in the three dimensional setting for a coupled magneto-mechanical problem.

Field variable	Symbol	Nr. of variables
Scalar magnetic potential	Ψ^{mag}	1
Magnetization vector	$\mathbb{M}_i$	3
Magnetic induction vector	$\mathbb{B}_i$	3
Magnetic field vector	$\mathbb{H}_i$	3
Specific magnetization of the fixed magnetic particles	$\mathbf{m}_i$	3
Local magnetic induction vector of the fixed magnetic particles	$\mathbf{b}_i$	3
Displacement vector	$\mathbf{u}_i$	3
Mechanical stress tensor	σ_{ij}	9
Strain tensor	ε_{ij}	6
Total number of unknowns		34

Figure 7. Simulation domain consisting of a ferrogel strip and the surrounding vacuum. **Figure 8.** Deformation of a ferrogel in a magnetic field. The magnetic field is placed in the x-direction. For the purpose of visualization the deformed ferrogel has been magnified.

coefficients (B_0 and B_1) are determined according to the procedure described in Appendix 4.

Numerical results

The resulting deformation of a ferrogel is presented in this section for two test cases. For each test case the results are presented with and without the magnetostriction effect.

Test case 1. In the first test case, a magnetic field is applied in the x-direction. The resulting deformation of a ferrogel is presented with and without magnetostriction effect. The results indicate that the ferrogel elongates when the magnetic field is applied in the x-direction, see Figure 8.

Test case 2. In the second test case, the magnetic field is applied in the y-direction. As before, the resulting deformation of a ferrogel is presented with and without the magnetostriction effect. The obtained results show the contraction of the ferrogel as the magnetic field is applied in the y-direction, see Figure 9.

Figure 9. Deformation of a ferrogel in a magnetic field. The magnetic field is placed in the y-direction. For the purpose of visualization the deformed ferrogel has been magnified.

Comparison with experimental work. The results of both test cases are compared with the experimental work of Filipcsei and Zrínyi (2010) and Gollwitzer et al. (2008). Both test cases are qualitatively in-line with these works. The above results also show that magnetostriction influences the deformation of a ferrogel. As evident in Figures 8 and 9, upon deformation, the ferrogel becomes slightly curved on the boundaries. This is

Table 5. Material parameters.

Name	Symbol	Value	Dimension
Bulk modulus	K	14.21×10^3	Pa
Shear modulus	G	0.87×10^3	Pa
Mass density	ρ	1085	kg m^{-3}
Magneto-strictive coefficient	B_0	-0.30	$\text{kg}^3 \text{A}^{-2} \text{s}^{-2} \text{m}^{-5}$
Magneto-strictive coefficient	B_1	-0.46	$\text{kg}^3 \text{A}^{-2} \text{s}^{-2} \text{m}^{-5}$
Permeability of the vacuum	μ_0	1.25663×10^{-6}	N A^{-2}

believed to be due to the singularities of the magnetic field on the corners of the ferrogel. To fully understand the cause of this problem, more simulations are required using various geometrical entities, material parameters, and field intensities.

Evaluation of the model

Imposing certain assumptions, a reduced model for ferrogels was introduced in this work. This model agrees with the model proposed by Eringen and Maugin (1990) and Jarkova et al. (2003) under the equilibrium conditions. These assumptions specifically are: (i) setting evolution of the specific magnetization to zero ($\dot{m}_i = 0$), (ii) ignoring exchange forces among magnetic particles ($\mathcal{B}_{ij} = 0$), and (iii) assuming a constant density of the polymer network and magnetic particles ($\rho^p = \rho^f = \text{const.}$).

Summary and conclusion

A coupled magneto-mechanical problem for ferrogels—consisting of a polymer network (P), fixed (f) and mobile (m) magnetic particles, and liquid (L)—was presented in this work. The derivation of the coupled problem was carried out in the framework of the continuum mechanics of multicomponent systems and the electro-magnetism of continua. Thermodynamically consistent constitutive relations were also formulated.

In many of the existing models in the literature (Jarkova et al., 2003; Raikher and Stolbov, 2003; Voltairas et al., 2003), only a polymer gel as a whole and one species of magnetic particles are considered. Regarding the ferrogel as a multicomponent material, interaction among constituents of the ferrogels can be captured. This helps to understand the process occurring inside ferrogels under the influence of external stimuli, such as magnetic fields.

A reduced model for ferrogels—consisting of a polymer network (P) and fixed (f) magnetic particles—was later presented. To obtain a reduced model for ferrogels, we assumed that all processes were isothermal and reversible, and deformation of ferrogels was described in the small strain regime. The mass density of ferrogels

was set to be constant. A superparamagnetic behavior of magnetic particles was assumed.

The magneto-mechanical problem based on the reduced model was solved in 2D using FEM. The only unknowns for the FEM implementation were mechanical displacement and magnetic potential. Deformation of a ferrogel in a magnetic field was subsequently studied. Two test cases were considered. In the first test case a magnetic field was applied in the x -direction. Elongation of a ferrogel was observed in this case with and without the magnetostriction effect.

Conversely, contraction of a ferrogel was observed in the second test case when a magnetic field was applied in the y -direction with and without the magnetostriction effect. The numerical results were also compared with experimental investigations taken from the literature. A good qualitative agreement was found between the numerical results and experimental work.

With our modeling approach, we were able to investigate (i) the influence of the magnetic field on the polymer gel containing magnetic particles (ferrogel) and (ii) the resulting mechanical deformation of a ferrogel. Thus, depending on the applied magnetic field, the resulting deformation of a ferrogel, and the attainable restoring forces, for example, of a ferrogel actuator could be determined.

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Notes

1. Mobile magnetic particles can either diffuse within the ferrogel or become trapped inside the polymer networks. The mobility of magnetic particles in the gel depends on the degree of cross-linking of the polymer network. Ferrogels containing both fixed and mobile magnetic particles (in the form of trapped particles) have already been produced in the laboratory (Frickel et al., 2011).
2. This holds true only for isotropic ferrogels where the particles are randomly distributed throughout the gel. A remnant magnetization is observed in anisotropic ferrogels where they have been produced in the presence of a magnetic field (Bohlius et al., 2004).

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Appendix I Discussion on balance of internal angular momentum

With the help of equations (28) and (41), it can be shown from equation (52) that

$$\begin{aligned} \sum_{\beta} \rho \frac{\dot{\mathbf{m}}_i^{\beta}}{\gamma^{\beta}} &= \rho \sum_{\beta} e_{ijk} \mathbf{m}_j^{\beta} [\mathbb{B}_k + \mathbf{b}_k^{\beta} + \frac{1}{\rho} \mathcal{B}_{pk,p}^{\beta}] \\ &= e_{ijk} \mathbb{M}_j \mathbb{B}_k + \sum_{\beta} e_{ijk} \mathbf{m}_j^{\beta} [\rho \mathbf{b}_k^{\beta} + \mathcal{B}_{pk,p}^{\beta}] \quad (70) \\ &= e_{ijk} \sigma_{jk}^E + \sum_{\beta} e_{ijk} \mathbf{m}_j^{\beta} [\rho \mathbf{b}_k^{\beta} + \mathcal{B}_{pk,p}^{\beta}] \end{aligned}$$

From equations (70) and (34) we have

$$\begin{aligned} \rho \left(\frac{\dot{S}_i}{\rho} \right) - \sum_{\beta} \frac{\dot{\mathbf{m}}_i^{\beta}}{\gamma^{\beta}} &= -J_{pi,p}^S \\ &+ e_{ijk} \sigma_{jk} - \sum_{\beta} e_{ijk} \mathbf{m}_j^{\beta} [\rho \mathbf{b}_k^{\beta} + \mathcal{B}_{pk,p}^{\beta}] \quad (71) \end{aligned}$$

Assuming

$$\frac{S_i}{\rho} = \sum_{\beta} \frac{\mathbf{m}_i^{\beta}}{\gamma^{\beta}} \quad \text{in } \mathcal{G} \quad (72a)$$

$$J_{pi}^S = \sum_{\beta} (J_{pi}^S)^{\beta} = - \sum_{\beta} e_{ijk} \mathbf{m}_j^{\beta} \mathcal{B}_{pk}^{\beta} \quad \text{in } \mathcal{G} \quad (72b)$$

and

$$\mathcal{B}_{pk}^{\beta} \mathbf{m}_k^{\beta} = 0 \quad \text{in } \mathcal{G} \quad (73)$$

Equation (71) as well as equation (34) can be restated as

$$e_{ijk} [\sigma_{jk} + \sum_{\beta} (\mathbf{m}_{j,p}^{\beta} \mathcal{B}_{pk}^{\beta} - \mathbf{m}_j^{\beta} \rho \mathbf{b}_k^{\beta})] = 0 \quad \text{in } \mathcal{G} \quad (74)$$

where equations (73) and (74) have to be identically satisfied by the constitutive relations. For more details concerning the motivation of equations (72b) and (73), please refer to Appendix 5.

In summary, by fulfillment of equations (47), (52), (73) and (74), the inequality (46) is satisfied.

The jump condition equation (35) is now expressed as

$$\begin{aligned} &\left[\left[\sum_{\beta} \frac{\rho}{\gamma^{\beta}} \mathbf{m}_i^{\beta} (v_j - w_j) - \sum_{\beta} e_{ipk} \mathbf{m}_p^{\beta} \mathcal{B}_{jk}^{\beta} \right] \right] \mathbf{n}_j^{\mathcal{G}} \\ &= 0 \quad \text{on } \partial \mathcal{G} \cap \partial \mathcal{F}^{ex} \quad (75) \end{aligned}$$

Introducing equations (56), it can be shown that using equation (56d) and equation (51), equation (73) is fulfilled. Accordingly, using equations (56a), (56c), and (56d), equation (74) is satisfied.

Appendix 2 Identification of independent variables

In this section the independent variables are identified for the purpose of constitutive modeling. The considered ferrogel consists of:

- polymer (P);
- fixed magnetic particles (f);
- liquid (L);
- mobile magnetic particles (m).

We assign symbol γ to fixed magnetic particles (f), liquid (L), and mobile magnetic particles (m) such that

$$\{\gamma\} = \{f, L, m\}$$

and symbol β to fixed magnetic particles (f) and mobile magnetic particles (m) such that

$$\{\beta\} = \{f, m\}$$

In the following we verify whether the following variables can be considered as independent variables.

The first set of variables to verify is the specific density of the γ th component $\tilde{c}^\gamma$. Since

$$\sum_{\gamma} \tilde{c}^\gamma + \tilde{c}^P = 1 \Rightarrow \tilde{c}^P = 1 - \sum_{\gamma} \tilde{c}^\gamma. \quad (76)$$

equation (76) shows $\tilde{c}^P$ depends on $\tilde{c}^\gamma$ and therefore is not an independent variable.

To verify if the deformation gradient of polymer F_{iJ}^P is an independent variable we first recall equation (6)

$$F_{iJ}^P = \frac{\partial x_i(X_Q^P, t)}{\partial X_J^P} \quad (77)$$

By defining the material time derivative with respect to “P” as

$$(\cdot)^P = \frac{\partial(\cdot)^P}{\partial t} + (\cdot)_{,i}^P v_i^P \quad (78)$$

we can express the material time derivative of the deformation gradient tensor of the polymer network $\dot{F}_{iJ}^P$ as

$$\dot{F}_{iJ}^P = \frac{\partial^2 x_i(X_Q^P, t)}{\partial X_J^P \partial t} = v_{i,j}^P = v_{i,j}^P F_{jJ}^P \quad (79)$$

By assigning $\alpha = P$ and $\dot{\rho}^P = 0$, equation (17) is rewritten as

$$\begin{aligned} \frac{\partial \rho^P}{\partial t} + (\rho^P v_i^P)_{,i} &= \dot{\rho}^P + \rho^P v_{i,i}^P = \dot{\rho}^P + \rho^P \frac{\dot{V}_{F_{iJ}^P}}{|F_{iJ}^P|} \stackrel{!}{=} 0 \\ \Rightarrow (\rho^P |F_{iJ}^P|)^{\times} &= 0 \Rightarrow \rho^P |F_{iJ}^P| = \dot{\rho}^P |F_{iJ}^P| = \dot{\rho}^P \text{ with } |F_{iJ}^P| = 1 \\ \Rightarrow \rho &= \frac{\dot{\rho}^P}{|F_{iJ}^P|^{\frac{1}{\alpha^P}}}. \end{aligned} \quad (80)$$

From equation (80) it is concluded that either ρ or $|F_{iJ}^P|$ can be chosen as independent variables.

The independent variables are now chosen as:

- specific density $\tilde{c}^\gamma$ for $\gamma = f, L, m$;
- deformation gradient tensor of the polymer F_{iJ}^P ;
- the specific magnetization vector and its derivative $\mathbf{m}_i^\beta, \dot{\mathbf{m}}_{i,j}^\beta$, provided that they fulfill the condition $\mathbf{m}_i^\beta \mathbf{m}_i^\beta = \text{const.}$ and $\mathbf{m}_{i,j}^\beta \mathbf{m}_i^\beta = 0$;
- temperature T .

Appendix 3 Derivation of inequality (44)

From the free energy density $\mathfrak{f} = \mathfrak{f}(F_{iJ}^P, \tilde{c}^\gamma, \mathbf{m}_i^\beta, \dot{\mathbf{m}}_{i,j}^\beta, T)$, equations (43) and (45) we obtain

$$\begin{aligned} \rho \dot{\mathfrak{f}} &= h_{iJ}^P \dot{F}_{iJ}^P + \sum_{\alpha} \rho \mu^{\gamma} \dot{\tilde{c}}^{\gamma} - \sum_{\beta} \rho \mathbf{b}_i^{\beta} \dot{\mathbf{m}}_i^{\beta} \\ &+ \sum_{\beta} \mathcal{B}_{ji}^{\beta} \dot{\mathbf{m}}_{i,j}^{\beta} + \rho \frac{\partial \mathfrak{f}}{\partial T} \dot{T} \end{aligned} \quad (81)$$

Using the following auxiliary calculations:

- $\dot{F}_{iJ}^P = \dot{F}_{iJ}^P - F_{iJ,q}^P (v_q^P - v_q)$
 $= v_{i,j}^P F_{jJ}^P + v_{i,j} F_{jJ}^P - v_{i,j} F_{jJ}^P - F_{iJ,q}^P (v_q^P - v_q)$
 $= v_{i,j} F_{jJ}^P + (v_i^P - v_i)_{,j} F_{jJ}^P - F_{iJ,q}^P (v_q^P - v_q)$
 $= v_{i,j} F_{jJ}^P + \left(\frac{j_i^P}{\rho^P} \right)_{,j} F_{jJ}^P - F_{iJ,q}^P \left(\frac{j_q^P}{\rho^P} \right)$
- $(\mathbf{m}_{i,j}^\beta)^{\cdot} = \frac{\partial \mathbf{m}_{i,j}^\beta}{\partial t} + \mathbf{m}_{i,k}^\beta v_k = (\dot{\mathbf{m}}_i^\beta)_{,j} - \mathbf{m}_{i,k}^\beta v_{k,j}$
- $h_{iJ}^P \dot{F}_{iJ}^P = h_{iJ}^P \left(v_{i,j} F_{jJ}^P + \left(\frac{j_i^P}{\rho^P} \right)_{,j} F_{jJ}^P \right) - F_{iJ,q}^P \frac{j_q^P}{\rho^P}$
- $h_{iJ}^P \left(\frac{j_i^P}{\rho^P} \right)_{,j} F_{jJ}^P = T \left[\frac{h_{iJ}^P F_{jJ}^P}{T} \frac{j_i^P}{\rho^P} \right]_{,j}$
 $- (h_{iJ}^P F_{jJ}^P)_{,j} \frac{j_i^P}{\rho^P} + \frac{T_{,j}}{T} h_{iJ}^P F_{jJ}^P \frac{j_i^P}{\rho^P}$
- $\rho \mu^{\gamma} \dot{\tilde{c}}^{\gamma} = \mu^{\gamma} (\dot{\rho}^{\gamma} - j_{i,i}^{\gamma}) \stackrel{!}{=} -\mu^{\gamma} j_{i,i}^{\gamma}$
 $= -T \left[\frac{\mu^{\gamma}}{T} j_i^{\gamma} \right]_{,i} + \mu_{,i}^{\gamma} j_i^{\gamma} - \frac{T_{,i}}{T} \mu^{\gamma} j_i^{\gamma}$
- $\mathcal{B}_{ji}^{\beta} (\mathbf{m}_{i,j}^\beta)^{\cdot} = \mathcal{B}_{ji}^{\beta} \left[(\dot{\mathbf{m}}_i^\beta)_{,j} - \mathbf{m}_{i,k}^\beta v_{k,j} \right]$

$$\begin{aligned}
\bullet \quad {}^R\mathcal{B}_{ji}^\beta(\dot{\mathbf{m}}_i^\beta)_{,j} &= ({}^R\mathcal{B}_{ji}^\beta - {}^D\mathcal{B}_{ji}^\beta)(\dot{\mathbf{m}}_i^\beta)_{,j} \\
&= -{}^D\mathcal{B}_{ji}^\beta(\dot{\mathbf{m}}_i^\beta)_{,j} + T \left[\mathcal{B}_{ji}^\beta \frac{\dot{\mathbf{m}}_i^\beta}{T} \right]_{,j} \\
&\quad - \mathcal{B}_{ji,j}^\beta \dot{\mathbf{m}}_i^\beta + \frac{T_{,j}}{T} \mathcal{B}_{ji}^\beta \dot{\mathbf{m}}_i^\beta
\end{aligned}$$

equation (81) is then rewritten as

$$\begin{aligned}
\rho \dot{\mathbf{f}} &= v_{i,j} \left[h_{ij}^P F_{jj}^P - \sum_\beta {}^R\mathcal{B}_{jq}^\beta \mathbf{m}_{q,i}^\beta \right] + \rho \frac{\partial \mathbf{f}}{\partial T} \dot{T} \\
&\quad + T \left[\frac{h_{ij}^P F_{jj}^P}{T} \frac{j_j^P}{\rho^P} - \sum_\alpha \frac{\mu_\alpha}{T} j_j^\alpha + \sum_\beta \mathcal{B}_{ji}^\beta \frac{\dot{\mathbf{m}}_i^\beta}{T} \right]_{,j} \\
&\quad - \frac{j_q^P}{\rho^P} \left[(h_{qj}^P F_{jj}^P)_{,j} + F_{ij,q}^P h_{ij}^P \right] + \sum_\alpha \mu_{,i}^\alpha j_j^\alpha \\
&\quad + \frac{T_{,j}}{T} \left[h_{ij}^P F_{jj}^P \frac{j_j^P}{\rho^P} - \sum_\gamma \mu_{,j}^\gamma j_j^\gamma + \sum_\beta \mathcal{B}_{ji}^\beta \dot{\mathbf{m}}_i^\beta \right] \\
&\quad - \sum_\beta \dot{\mathbf{m}}_i^\beta \left[\rho \mathbf{b}_i^\beta - \rho \mathbf{b}_i^\beta + \mathcal{B}_{ji,j}^\beta \right]
\end{aligned} \quad (82)$$

The second law of thermodynamics, inequality (42), can now be expressed as

$$\begin{aligned}
T \hat{\eta}^s &= \rho \bar{r} - T \rho \hat{\eta}^i - \rho \dot{T} (\eta + \frac{\partial \mathbf{f}}{\partial T}) \\
&\quad + v_{i,j} \left[\sigma_{ji} - h_{ij}^P F_{jj}^P + \sum_\beta {}^R\mathcal{B}_{jq}^\beta \mathbf{m}_{q,i}^\beta \right] \\
&\quad + T \left[\eta_j - \frac{q_j}{T} + \sum_\gamma \frac{\mu_\gamma}{T} j_j^\gamma - h_{ij}^P F_{jj}^P \frac{j_j^P}{(T \rho^P)} - \sum_\beta \mathcal{B}_{ji}^\beta \frac{\dot{\mathbf{m}}_i^\beta}{T} \right]_{,j} \\
&\quad - \frac{T_{,j}}{T} \left[q_j + h_{ij}^P F_{jj}^P \frac{j_j^P}{\rho^P} - \sum_\gamma \mu_{,j}^\gamma j_j^\gamma + \sum_\beta \mathcal{B}_{ji}^\beta \dot{\mathbf{m}}_i^\beta \right] \\
&\quad + \frac{j_q^P}{\rho^P} \left[\rho^P b_q^P + h_{ij}^P F_{ij,q}^P + (h_{qj}^P F_{jj}^P)_{,j} \right] \\
&\quad + \sum_\alpha j_i^\alpha (b_i^\alpha - \mu_{,i}^\alpha) \\
&\quad + \sum_\beta \dot{\mathbf{m}}_i^\beta \left[\rho \mathbb{B}_i + \rho \mathbf{b}_i^\beta + \mathcal{B}_{ji,j}^\beta \right] \\
&\quad - \sum_\beta \dot{\mathbf{m}}_i^\beta \rho \mathbf{b}_i^\beta + \sum_\beta (\dot{\mathbf{m}}_i^\beta)_{,j} \mathcal{B}_{ji}^\beta \geq 0
\end{aligned} \quad (83)$$

Defining

$$\bar{q}_j = q_j + h_{ij}^P F_{jj}^P \frac{j_j^P}{\rho^P} - \sum_\gamma \mu_{,j}^\gamma j_j^\gamma + \sum_\beta \mathcal{B}_{ji}^\beta \dot{\mathbf{m}}_i^\beta \quad (84a)$$

$$\eta_j = \frac{\bar{q}_j}{T} \quad (84b)$$

$${}^R\sigma_{ij} = \rho \frac{\partial \mathbf{f}}{\partial F_{ij}^P} F_{ij}^P - \sum_\beta {}^R\mathcal{B}_{iq}^\beta \mathbf{m}_{q,i}^\beta \quad (84c)$$

$$T \hat{\eta}^i = \bar{r} \quad (84d)$$

Figure 10. A ferrogel strip, fixed at one end and free to move at the other end. n_i^I and n_i^{II} are the normal unit vectors at the boundary of the ferrogel.

$$\eta = - \frac{\partial \mathbf{f}}{\partial T} \quad (84e)$$

inequality (83) is rewritten as

$$\begin{aligned}
v_{i,j} \frac{D}{T} \sigma_{ji} - \frac{T_{,j}}{T} \bar{q}_j + \frac{j_q^P}{\rho^P} \left[\rho^P b_q^P + h_{ij}^P F_{ij,q}^P + (h_{qj}^P F_{jj}^P)_{,j} \right] \\
+ \sum_\alpha j_i^\alpha (b_i^\alpha - \mu_{,i}^\alpha) - \sum_\beta \dot{\mathbf{m}}_i^\beta \rho \mathbf{b}_i^\beta + \sum_\beta (\dot{\mathbf{m}}_i^\beta)_{,j} \mathcal{B}_{ji}^\beta \\
+ \sum_\beta \dot{\mathbf{m}}_i^\beta \left[\rho \mathbb{B}_i + \rho \mathbf{b}_i^\beta + \mathcal{B}_{ji,j}^\beta \right] \geq 0
\end{aligned} \quad (85)$$

Appendix 4 Determination of the magnetostriction coefficients

Gollwitzer et al. (2008) suggested a linear relation between the elongation of a ferrogel sample and the magnetic field expressed as

$$\varepsilon_i = c_i \mathbb{M}^2 \quad (86)$$

where, $i = x$ and z , with x perpendicular and z parallel to the magnetic field. c_x and c_z were determined as $c_x = -6.1 \times 10^{-5} (\text{m/kA})^2$ and $c_z = 13.4 \times 10^{-5} (\text{m/kA})^2$ (Gollwitzer et al., 2008).

To find the magnetostriction coefficients (B_0 and B_1), a relation between the strain (elongation) and the square of the magnetization is sought using the magneto-mechanical problem in section ‘Reduced model for ferrogels’. This relation is then compared to equation (86) to find B_0 and B_1 .

For this purpose a ferrogel strip is considered which is fixed at one end and is free to move at the other ends, see Figure 10. An external magnetic field $\mathbb{H}_0$ is then placed in the z -direction resulting in magnetization $\mathbb{M}_0$ in the same direction. The following equilibrium conditions are defined on the boundaries of the ferrogel

$$[[\mathbb{B}_i]] n_i = 0 \quad (87a)$$

$$[[\mathbb{H}_i]] t_i = 0 \quad (87b)$$

$$(\sigma_{ij} + \sigma_{ij}^E) n_j|_{(In)} = (\sigma_{ij}^E) n_j|_{(Out)} \quad (87c)$$

where (In) indicates inside the gel and (Out) indicates outside of the gel. In order to obtain the results in SI units, the Maxwell’s stress tensor σ_{ij}^E is rewritten as

$$\sigma_{ji}^E = \mathbb{B}_j \mathbb{H}_i - \frac{\delta_{ij}}{2} \left(\frac{1}{\mu_0} \mathbb{B}_k \mathbb{B}_k - 2 \mathbb{M}_q \mathbb{B}_q \right) \quad (88)$$

where $\mu_0 = 1.25663 \times 10^{-6} \text{ N A}^{-2}$ is the magnetic permeability of the vacuum. Solving equations (87) for σ_{xx} and σ_{zz} leads to

$$\sigma_{xx} = -\frac{1}{2} \mu_0 (\mathbb{M}_y^2 + \mathbb{M}_z^2) \quad (89a)$$

$$\sigma_{zz} = -\frac{1}{2} \mu_0 (\mathbb{M}_x^2 + \mathbb{M}_y^2) \quad (89b)$$

$$\sigma_{xz} = \sigma_{xy} = 0 \quad (89c)$$

Since we know that $\mathbb{M}_x = \mathbb{M}_y = 0$ and $\mathbb{M}_z = \mathbb{M}_0$, equations (89) reduces to

$$\sigma_{xx} = -\frac{1}{2} \mu_0 \mathbb{M}_0^2 \quad (90a)$$

$$\sigma_{zz} = 0 \quad (90b)$$

$$\sigma_{xz} = \sigma_{xy} = 0 \quad (90c)$$

Using the constitutive relations equations (65) and (66) presented in section ‘Reduced model for ferrogels’ and assuming $\varepsilon_{xx} = \varepsilon_{yy}$, a system of equations is obtained where B_0 and B_1 are the only unknowns.

Solving this system of equations, the magneto-strictive coefficients are determined.

Appendix 5 Reformulation of the flux of internal angular momentum

Using

$$J_{ml}^S = a_m \mathbf{m}_l - e_{lqr} \mathbf{m}_q \mathcal{B}_{mr} \quad (91)$$

with the restriction

$$\mathcal{B}_{mr} \mathbf{m}_r = 0 \quad (92)$$

the variable J_{ml}^S may be expressed in terms of the variables a_m and $\mathcal{B}_{mr}$, bearing in mind that $\mathbf{m}_r$ are known variables. The replacement of the variables is unique as is shown by the explicit solution of equation (91) using (92) resulting in

$$\begin{aligned} a_m &= J_{ml}^S \frac{\mathbf{m}_l}{\mathbf{m}_q \mathbf{m}_q} \text{ and} \\ \mathcal{B}_{mr} &= J_{ml}^S e_{lrp} \frac{\mathbf{m}_p}{\mathbf{m}_q \mathbf{m}_q} \end{aligned} \quad (93)$$

In our case, motivated by equation (71), a_m equals zero.