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**13– 2002**

**Preprint, no 13 – 02 / 2002**

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# Modelling the Hyperelasticity of Magnetic Field Sensitive Gels

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The continuum theory, previously developed to quantitatively account for the large deformations observed in gels endowed with electric properties, is extended to magnetic field sensitive gels (ferromagnetic or diamagnetic in origin). The derived analytical formula for the dependence of the gel displacement on the magnetic field, can be applied, either to control recently developed biomimetic valves and possible artificial muscles constructions, or to interpret similar phenomena in biophysics.

PACS numbers: 46.25.Hf, 75.50.Mm, 75.80.+q, 82.70.Gg, 83.80.Gv, 85.70.Ec, 87.68.+z

Piezoelectric, magnetostrictive and shape memory alloys have for long been used in industrial and medical applications. There is an increasing need nowadays for new materials, with biomimetic functionalities, that combine low cost and high efficiency. Hydrogels with thermoelectromagnetic or chemical properties are good candidates, since they combine more efficient actuation or sensory mechanism (large deformations) along with minimum investment on expensive rough materials. Phenomenological models have been proposed by the authors, to control medical applications of magnetic fluids in and drug delivery and eye surgery [1, 2]. Experiments on: *pH* [3], thermo- [4] and electromagnetic [5–8] sensitive gels have been performed recently, confirming their capability to mimic muscle contraction (artificial muscles). The challenge for theory is to express the observed nonlinear deformations as a function of the applied fields. Attempts in that direction are the models developed in [9–11] for magnetic gels in nonuniform and uniform magnetic fields, when hysteretic effects are present in the magnetoelastic constitutive laws, and in [12] by us, for electrogels in uniform electric fields. Our work on electrogels succeeded in determining both the initial slope of the strain-electric field relation, as well as the saturation effects at high electric fields. It is the aim of the present work to extend the results of our previous effort [12] to magnetic gels, either ferromagnetic (*ferrogels*) or diamagnetic in origin. We discuss also the possibility of the present approach to control the operation of biomimetic valves [3] and explain similar diamagnetic deformations in biophysics [13].

We begin by summarizing the general continuum theory of magnetoelasticity. The formulation is analogous to our previous work [12] and is based on that of Toupin [14]. Hereafter, bold and double bold characters will denote vector and tensor fields, respectively. We consider that the ferrogel deforms as a continuous body, which in the reference (undeformed) configuration occupies a region  $\Omega \subset \mathcal{S}$ , of the whole space  $\mathcal{S}$ , inside the closed surface  $\partial\Omega$ .

The material points are identified by their position vectors  $\mathbf{X}$  in  $\Omega$ , with Cartesian coordinates  $X_A$  ( $A = 1, 2, 3$ ). After the deformation, the ferrogel occupies the region  $\Omega_d$  and a point originally denoted by  $\mathbf{X}$  is deformed to the position  $\mathbf{x}$ , with coordinates  $x_i$  ( $i = 1, 2, 3$ ) and deformation gradient  $\mathbb{F} \equiv \nabla_{\mathbf{X}} \otimes \mathbf{x}(\mathbf{X})$ . For a uniform static applied magnetic field  $\mathbf{H}_0$ , the equilibrium problem is described by the partial differential equations:

$$\nabla_{\mathbf{X}} \cdot \mathbb{S} + \mathbf{f}_m = \mathbf{0}, \quad \text{in } \Omega \quad (1)$$

$$\nabla \cdot \mathbf{B} = 0, \quad \text{in } \mathcal{S} \quad (2)$$

$$\nabla \times \mathbf{H} = 0, \quad \text{in } \mathcal{S} \quad (3)$$

and the jump conditions

$$[\mathbb{S}^T + J (\mathbb{F}^{-1} \mathbb{T}_m)^T] \mathbf{N} = \mathbf{0}, \quad \text{on } \partial\Omega \quad (4)$$

$$[\mathbf{B}] \cdot \mathbf{n} = 0, \quad \text{on } \partial\Omega_d \quad (5)$$

$$\mathbf{t} \cdot [\mathbf{H}] = 0, \quad \text{on } \partial\Omega_d. \quad (6)$$

in the reference configuration. Here

$$\mathbf{f}_m = \nabla_{\mathbf{X}} \cdot (J \mathbb{F}^{-1} \mathbb{T}_m) = J \mu_0 (\mathbf{M} \cdot \nabla) \mathbf{H}, \quad (7)$$

is the magnetic body force with

$$\mathbb{T}_m = \mathbf{B} \otimes \mathbf{H} - \mu_0 H^2 \mathbb{I}/2, \quad (8)$$

the Maxwell stress tensor, due to Einstein and Laub [14],

$$\mathbb{S} = J \mathbb{F}^{-1} \mathbb{T}, \quad (9)$$

is the nominal stress tensor [17],

$$\mathbf{B} = \mu_0 (\mathbf{H} + \mathbf{M}), \quad (10)$$

is the magnetic induction,  $\delta(\mathbf{x}) = 1$  for  $\mathbf{x} \in \Omega_d$  and  $\delta(\mathbf{x}) = 0$  for  $\mathbf{x} \in \mathcal{S} - \Omega_d$ ,  $[\mathbf{A}] \equiv A_{\text{out}} - A_{\text{in}}$ ,  $\mathbf{H}$  is the total magnetic field,  $\mathbf{H} = \mathbf{H}_0 + \mathbf{H}_D$ , with  $\mathbf{H}_D$  the demagnetizing field,  $\mathbf{M}$  is the magnetization vector per unit volume,  $J = \det \mathbb{F}$ ,  $\mathbb{T}$  is the Cauchy stress tensor,  $\nabla_{\mathbf{X}}$  and  $\nabla$  are the gradient vector operators in the reference and present configurations, respectively,  $\mu_0$  is the magnetic permeability of vacuum,  $\mathbb{I}$  is the identity tensor,  $\otimes$  and  $\cdot$  denote tensor and inner product, respectively,  $\mathbf{t}$  is

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the unit tangential vector on  $\partial\Omega_d$  and  $\mathbf{N}$  and  $\mathbf{n}$  are the outward unit vectors on  $\partial\Omega$  and  $\partial\Omega_d$ , respectively, with

$$\mathbf{n} = J(\mathbb{F}^{-1})^T \mathbf{N}. \quad (11)$$

If we decompose the magnetic field  $\mathbf{H}$  in tangential and normal components on  $\partial\Omega$ , neglect mechanical surface tractions and make use of the definitions (7-11) and the jump conditions (5-6), the balance of magnetomechanical surface tractions (4) reduces to:

$$\mathbb{S}^T \mathbf{N} = \mu_0 M_n^2 \mathbf{n}/2, \quad M_n \equiv \mathbf{M} \cdot \mathbf{n}, \quad \text{on } \partial\Omega. \quad (12)$$

The BVP (boundary value problem) (1-6) is derived from an energy variational principle [14]. The derivation is not unique and depends on the form of the magnetostatic energy. Thus, many equivalent, but not identical, formulations are used in the literature. The interested reader should consult the footnotes in [15] for this controversial issue, in the analogous electromechanical problem. The variational principle imposes constraints on the form of the constitutive relations, which in our case read:  $\mathbb{S}(\mathbb{F}, \boldsymbol{\mu}) \equiv \partial W / \partial \mathbb{F}$  and  $\mathbf{H}(\boldsymbol{\mu}, \mathbb{F}) \equiv \partial W / \partial \boldsymbol{\mu}$ . The free energy depends both on the strain and the magnetization,  $W = W(\mathbb{E}, \boldsymbol{\mu})$ , where  $\mathbb{E} = (\mathbb{F}^T \mathbb{F} - \mathbb{I}) / 2$  is the Green finite strain tensor and  $\boldsymbol{\mu} = \mathbf{M} / \rho$  is the magnetization vector per unit mass.  $\rho$  is the density in  $\Omega_d$ , which is related to the density  $\rho_0$  in  $\Omega$  through the equation  $\rho_0 = J\rho$ . The exact forms of the above constitutive laws are also determined by the material symmetry and the second law of thermodynamics [16]. For small concentration of the magnetic micro- or nanoparticles (*diluted ferrogel*), the magnetization contribution to the free energy density  $W$  is negligible and the constitutive equations become:

$$\mathbb{S} \simeq \mathbb{S}(\mathbb{F}) = \frac{\partial W}{\partial \mathbb{F}}, \quad (13)$$

$$\mathbf{M} \simeq \mathbf{M}(\mathbf{H}). \quad (14)$$

Henceforth, we will restrict our attention to diluted ferrogels, with constitutive equations of the form (13-14) and vanishing mechanical surface traction. If we introduce the magnetostatic potential  $\Phi$  in (2-3) with  $\mathbf{H} \equiv -\nabla\Phi$ , and express the nominal stress tensor  $\mathbb{S}$  in terms of the Biot stresses  $t_1^{(1)}, t_2^{(1)}, t_3^{(1)}$  (the principal values of the Biot stress tensor,  $\mathbb{T}^{(1)} = (\mathbb{S}\mathbb{R} + \mathbb{R}^T \mathbb{S}^T) / 2$ , [17]), where  $\mathbb{R}$  is the finite rotation tensor, we obtain a complicated but well posed problem. Even for relatively simple prescribed deformation modes, that satisfy (1), the nonlinearities involved in (13-14), make the resultant potential problem a formidable task. Before any effort to solve the BVP (1-6), the difficult to prove issues of existence and uniqueness of solutions, should be addressed. Nevertheless, we will construct our model upon the general theory, without complete rigour on the satisfaction of BVP (1-6), by introducing simplifications, guided by the observed geometry of deformation, as well as from physical considerations.

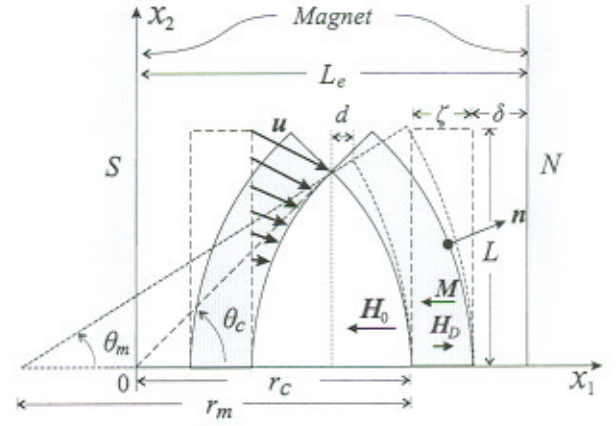


FIG. 1: Problem geometry.

The following model can be applied, either to determine the magnetic field dependence of the deformation of a single ferrogel, or of a pair of diluted ferrogels, placed symmetrically between the poles of a magnet, in such a distance apart that do not interact with each other in the absence of the external magnetic field. In the presence of a uniform applied magnetic field  $\mathbf{H}_0$ , the pair of ferrogels admit symmetrical deformations, with respect to the middle parallel plane to the poles of the magnet, that are  $L_e$  distance apart (see figure 1). We consider that the ferrogels are homogeneous, isotropic and diluted, in the form of rectangular blocks of cubic cross-section, of width  $\zeta$  and height  $L$ . Though the further analysis is performed for paramagnetic gels or ferrogels (positive magnetic susceptibility), it can be applied also to diamagnetic gels, just by changing the sign of the magnetic susceptibility and considering deformations in the opposite direction from the one shown in figure 1. Experiments of this kind with plants, have been announced recently [13]. Due to the prescribed symmetry of the deformation for the pair of ferrogels, we restrict our discussion to a single ferrogel (the right ferrogel of figure 1) and we will recall the presence of the second ferrogel only when the condition for valve operation will be derived. We assume that the ferrogel admits plane deformations of the form

$$r = f(X_1), \quad \theta = g(X_2), \quad x_3 = X_3, \quad (15)$$

shown in figure 1. Then the displacement field

$$\mathbf{u} = \mathbf{X} - \mathbf{x}(\mathbf{X}), \quad (16)$$

corresponds to the bending of the dashed rectangle, of figure 1, into a section of a circular disc, with radius difference  $\Delta r = \zeta$ . The ferrogel is kept fixed at points  $\mathbf{X} = (r_m, 0)$  and  $(r_m + \zeta, 0)$ , that is:

$$\mathbf{u}(r_m, 0) = \mathbf{u}(r_m + \zeta, 0) = 0. \quad (17)$$

Due to the incompressibility constrain, ferrogel's volume  $v$  remains invariant, during the magnetically driven deformation,  $v(\Omega) = v(\Omega_d)$ . This condition determines the

maximum deflection angle  $\theta_m$  in terms of geometrical parameters:

$$\theta_m = 2L/(2r_m + \zeta). \quad (18)$$

The principal stretches  $\lambda_i$ ,  $i = 1, 2, 3$  are given, due to (15) by the relations:

$$\lambda_1 = f'(X_1), \quad \lambda_2 = f(X_1)g'(X_2), \quad \lambda_3 = 1, \quad (19)$$

where the prime denotes differentiation with respect to the argument. The incompressibility constraint  $\lambda_1 \lambda_2 \lambda_3 = 1$  results, due to (19), after separation of variables, to the solution:

$$r = \sqrt{2A_1 X_1 + A_2}, \quad \theta = X_2/A_1 + A_3. \quad (20)$$

The unknown constants  $A_i$ ,  $i = 1, 2, 3$  are determined from the conditions (17)

$$\begin{aligned} A_1 &= r_m + \zeta/2 = L/\theta_m, \\ A_2 &= -r_m(r_m + \zeta) = (\zeta/2)^2 - (L/\theta_m)^2, \\ A_3 &= 0. \end{aligned} \quad (21)$$

In order to simplify the magnetostatic mathematical analysis, without sacrificing the physics of the problem, we assume that the magnetization vector  $\mathbf{M}$  and the magnetic field  $\mathbf{H}$ , either inside or outside the ferrogel, are constant and collinear to the external uniform applied field  $\mathbf{H}_0$ :

$$\mathbf{H}_{\text{in}} = \chi_H \mathbf{H}_0, \quad (22)$$

$$\mathbf{H}_{\text{out}} \simeq \mathbf{H}_0, \quad (23)$$

$$\mathbf{M} = \chi_m \mathbf{H}_{\text{in}} = \chi_m \chi_H \mathbf{H}_0, \quad (24)$$

$$\mathbf{H}_0 = -H \hat{\mathbf{E}}_1, \quad H = \text{const.} > 0, \quad (25)$$

where  $\chi_H$  and  $\chi_m$  are dimensionless functions of the magnetic field  $H$  and the shape of the ferrogel  $L/\zeta$ , and  $\hat{\mathbf{E}}_i$  and  $\hat{\mathbf{e}}_i$ , ( $i = 1, 2$ ) are the unit vectors in the reference and present configuration, respectively. The above assumptions are valid for the bulk of the ferrogel but not close to the boundary  $\partial\Omega$ . The solution (22-25) satisfies the magnetostatic problem (2-3), while the jump condition (5) reduces to

$$(1 - \chi) \mathbf{H}_0 \cdot \mathbf{n} = 0, \quad \text{on } \partial\Omega_d, \quad (26)$$

with  $\chi = \chi(H, L/\zeta) = \chi_H(1 + \chi_m)$  the magnetic susceptibility. In general,  $\mathbf{H}_{\text{in}} < \mathbf{H}_{\text{out}}$ , due to the presence of demagnetizing effects, so  $0 \leq \chi_H \leq 1$  and  $\chi_m > 0$ . Due to the assumptions (22-25),  $\mathbf{f}_m = \mathbf{0}$  from equation (7). Then, due to the constitutive laws (13-14), the equilibrium problem (1) reduces to the pure mechanical one:

$$\nabla_{\mathbf{X}} \cdot \mathbb{S} = 0. \quad (27)$$

By following an analysis identical with the one presented in [12] we can solve (27) in terms of the Biot stresses  $t_1^{(1)} = \tau(\lambda) = C_1 \lambda$  and  $t_2^{(1)} = -\lambda^2 \tau(\lambda)$  with

$$\lambda = B_1/r = [2\theta_m X_1/L + \zeta^2 \theta_m^2 / (4L^2) - 1]^{-1/2}, \quad (28)$$

and  $\lambda_1 = 1/\lambda_2 = \lambda$ . In accordance with our assumption for a diluted ferrogel, the balance of the magnetomechanical surface tractions (12) results, due to the solutions (22-25) to  $\tau(\lambda) = \mu_0 M_n^2/2$ , or equivalently to:

$$2\lambda - h^2(1 - \chi_H)^2 \cos^2 \theta = 0, \quad (29)$$

with  $h \equiv H/H_p$ ,  $H_p \equiv (2C_1/\mu_0)^{1/2}$ , on the boundary  $X_1 = r_m + \zeta$ . Since  $\mathbf{H}_0 \cdot \mathbf{n} \neq 0$  on  $\partial\Omega_d$  the condition (26) implies that  $\mathbf{H}_{\text{in}} = \mathbf{H}_{\text{out}}$  or  $\chi = 1$ . In order to preserve the positive definite character of the strain-energy function  $W(\lambda) = C_1 \lambda^2/2$  we must have  $C_1 > 0$ . Notice that equation (29) does not hold for every  $\theta$  on  $\partial\Omega$ , but since our primary concern is to model available experimental data, we just have to satisfy (29) only for the maximum deflection angle  $\theta_m$ , since what is measured in the experiments is the displacement of the upper free part of the ferrogel, for given  $H$  and  $L/\zeta$ . Then, from (28) and (29) we obtain, for  $X_1 = r_m + \zeta$  and  $\theta = \theta_m$ :

$$h^2(1 - \chi_H)^2 [1 + \zeta\theta_m/(2L)] \cos^2 \theta_m = 2. \quad (30)$$

If we solve (30) for  $\theta_m$  and substitute the result in

$$u \equiv u_x/(r_m + \zeta) = 1 - \cos \theta_m, \quad (31)$$

we derive the displacement  $u$ , as a function of the applied field  $h$  and its shape  $L/\zeta$ , provided that the function  $\chi_H = \chi_H(h, L/\zeta)$  will be specified. Unfortunately, the solution  $\theta_m$  of (30) and thus  $u$  of (31) are singular at  $h = 0$ , for a diluted ferrogel,  $\chi_H(0, L/\zeta) = 0$ , and thus they do not correspond to the expected vanishing of the deformation in the absence of applied magnetic field  $u(h = 0) = 0$ . This singularity is a consequence of isotropy, collinearity and especially homogeneity introduced in (22-25). Since the main physical mechanisms, observed in the experiments, are present in our model, we can overthrow the singularity at  $h = 0$ , by neglecting (18) and replacing the geometrical definition of  $\lambda$  (28), with a suitable function of  $h$  and  $L/\zeta$ . Thus, if we substitute,

$$\lambda = \lambda_0(1 + \chi_H)/(1 + \gamma \chi_H), \quad (32)$$

with  $\gamma \geq 1$  and  $\lambda_0 \equiv h^2(1 - \chi_H)^2/2$ , in (29) and solve for  $\cos \theta_m$ , the displacement (31) reduces to

$$u \simeq u_x/(r_m + \zeta) = 1 - [(1 + \chi_H)/(1 + \gamma \chi_H)]^{1/2}. \quad (33)$$

The longer is the surface of the ferrogel, that is exposed to the uniform external field, the larger the demagnetization effects induced on it and as a consequence the smaller the total magnetic field inside. In order to take into account this shape dependence of the constitutive law (24) we admit for  $\chi_H$  the simple power law:

$$\chi_H = \alpha (L/\zeta)^{2\beta} h^\beta, \quad (34)$$

where all  $\alpha$ ,  $\beta$  and  $\gamma$  are dimensionless constants. Although the form (32) has the drawback that  $\lambda(h = 0) =$

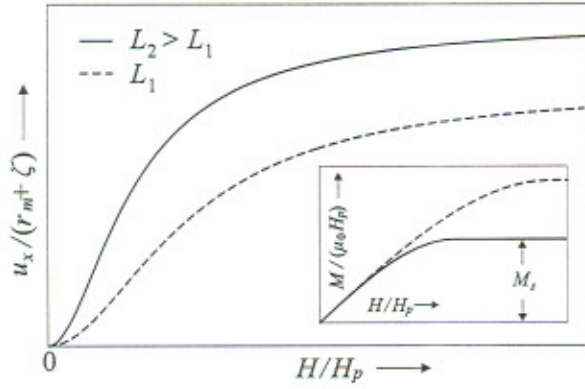


FIG. 2: The normalized displacement  $u_x/(r_m + \zeta)$  and magnetization  $M/(\mu_0 H_p)$ , as functions of the normalized applied magnetic field  $H/H_p$ , for varying length  $L$  of the ferrogel.

0, compared to the expected  $\lambda(h=0) = 1$ , it recovers the observed  $u(h=0) = 0$ , due to (34). Due to (24) and (26) the magnetic constitutive relation reads

$$m(h) = \begin{cases} h(1 - \chi H) & \text{for } h < h_{sat} \\ m_{sat} & \text{for } h \geq h_{sat} \end{cases} \quad (35)$$

with  $m \equiv M/H_p$  and

$$m_{sat} = \beta h_{sat}/(1 + \beta), \quad (36)$$

$$h_{sat} = (\zeta/L)^2 [\alpha(1 + \beta)]^{-1/\beta}. \quad (37)$$

The response (displacement) of the ferrogel to the applied magnetic field, is depicted in figure 2, according to the model (33-34), for  $\alpha > 0$ ,  $\beta = 2$ ,  $\gamma > 1$ ,  $r_m + \zeta \simeq L_e$  and varying  $L$ , such that  $0 < \chi_H < 1$ . The magnetic constitutive law (35-37) is also enclosed in the same figure. Due to the flexibility on the selection of the dimensionless parameters  $\alpha$ ,  $\beta$ ,  $\gamma$ , our model can estimate correctly both the initial slope, as well as saturation effects of the displacement-magnetic field behavior. Notice that increase of the length  $L$  of the ferrogel leads, as expected from demagnetizing effects, to higher displacement and smaller magnetization for the same applied magnetic field. Unfortunately, though equation (35) is nonlinear for  $\beta \neq 0$ , it corresponds to unit initial susceptibility,  $\chi_{ini} \equiv m'(0) = 1$ , since  $\chi = 1$ . This drawback can be restored, in a particular experiment, by replacing  $(1 - \chi_H)$  in (35) with  $(\chi - \chi_H)$  and assuming that  $\chi > 1$ .

Returning to the case of a pair of ferrogels, that behave as a biomimetic valve, and due to the symmetries considered, we can determine the critical magnetic field for valve operation (shaded deformed state of ferrogels in figure 1) as the one that corresponds to  $d = 0$ , or equivalently to  $r_m = r_c$ ,  $\theta_m = \theta_c$ , where

$$d = r_m(1 - \cos \theta_m) - r_c(1 - \cos \theta_c). \quad (38)$$

Notice that in general  $r_m = r_m(H, L)$ , but we have to be cautious about the form that we will assign to this

function, in order to preserve the monotonicity of the displacement function, with respect to  $H$  and  $L$ .

In summary, we have developed the theoretical framework for studying large deformations in ferrogels, when hysteretic effects are not present in the constitutive laws. Our model includes all the information for quantitative interpretation of magnetic field dependent deformations and valve operations, since it comprises the main physical mechanisms and the geometrical attributes of the deformation (magnetic microparticle concentration, nonlinearities on constitutive laws, maximum deflection angle, aspect ratio). The effect of an inhomogeneous applied magnetic field can also be studied, with the cost of complicating the solution procedure. Due to its generality, the present analysis easily conforms with similar observed mechanisms in biophysics (paramagnetic,  $0 < \chi < 1$ , or diamagnetic,  $\chi < 0$ , elastic responses of plants and biological tissues in magnetic fields [13]), as well as with prototypes in the rapidly developing field of microelectromechanical systems (MEMS), and their medical counterparts, biomedical microdevices (Bio-MEMS). Extension of the approach to pure elongation of the ferrogel, in suitably applied magnetic fields is straight-forward, with direct application to artificial muscle modelling.

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