

NONHOMOGENEOUS, NON-LINEAR DEFORMATION OF POLYMER GELS SWOLLEN WITH MAGNETO-RHEOLOGICAL SUSPENSIONS

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Polymer gels swollen with a magneto-rheological suspension are highly elastic materials with considerable magnetic susceptibility. In this work the magnetic field induced deformation and motion of these magnetic polymer systems is discussed. We present a continuum material model by introducing magnetic equations into non-linear elasticity theory. The material properties of these magnetic rubber-like substances are characterized with a Langevin type magnetization and a neo-Hookean strain energy function. The non-linear character of the equations that describe the material properties and the nonhomogeneity of the deformation lead to a unique deformation mechanism. In order to demonstrate the characteristics of the magnetic field induced deformations we present numerical and finite element calculations and compare them with experimental results.

1 Introduction

Novel magnetic- and electric field sensitive polymeric materials have been developed recently. [1-3] In these polymer gels fine nanoparticles are embedded into a polymer network resulting in a highly elastic, field sensitive material. In this paper we deal with polymers containing magnetic particles. These so-called ferrogels are superparamagnetic with a considerable magnetic susceptibility of the order 0.01-0.1. Ferrogels are capable of undergo giant deformations (the strain can reach 1.5) in a magnetic field with ordinary strength.

Our goal is to understand and describe the deformation of ferrogels in magnetic field. To accomplish this we need to introduce magnetic equations into the non-linear elasticity theory. Magnetic interactions are not usual in elasticity problems. Since the field, and the force alike, might have a spatial distribution different material points experience force of different strength and direction, which leads to a complex, nonhomogeneous deformation. Not only the nature of the resulting deformation, but also the treatment of body forces is different from that of surface tractions.

In case of a highly elastic magnetic medium the non-linear character of both elastic and magnetic interactions lead to some very novel features of the deformation process. The non-linearity of the governing interactions and the spatial distribution of the force density make the treatment fairly complicated and result in deformations with some unique features as well as with special patterns never seen in deformations induced by surface tractions. In the following chapter we discuss the mechanism of magnetic field induced deformations. Then, we briefly outline the continuum material model for ferrogel, a rubber elastic, superparamagnetic medium. In order to compare the model with real experiments we show the solution of the equations of motion in a simple, one-dimensional situation and compare it with experimental results. Finally, one- and three-dimensional finite element calculations are shown in order to illustrate the real deformation of a ferrogel sample.

2 Deformation induced by magnetic field

The ferrogel is a polymer network swollen with a MR suspension. Most of the magnetic properties of ferrogels are similar to that of ferrofluids. They are superparamagnetic with a fast magnetic relaxation. The main difference is that the particles cannot move. Their motion is restricted by the polymer matrix. In uniform magnetic field, the moments of the particles rotate towards the direction of the field, but cannot form chains or alike. The ferrogel gets magnetised and its elastic modulus changes depending on the strength of the magnetising field. [4] It experiences no net force and does not get deformed. However, when it is placed into an inhomogeneous magnetic field, the particles experience forces. These magnetic forces acting on the individual particles generate a stress distribution, which becomes balanced by the network elasticity, owing to the strong adhesive interactions between chains and particles. Due to the cross-linking bridges and entanglements in the network, changes in molecular conformation can accumulate and lead to macroscopic deformation or – if the external field changes in time - motion. The magnetic field drives and controls the deformation or motion, and the momentary equilibrium shape is determined by the balance between magnetic and elastic interactions.

As an example to the nonhomogeneous deformation of ferrogels in magnetic field we present the deformation of a magnetic gel sheet in Fig 1. Picture ‘A’ and ‘B’ show the gel in undeformed and deformed states, respectively. The complexity and nonhomogeneity of the deformation is well seen on the transformation of the grid painted on the surface of the gel.

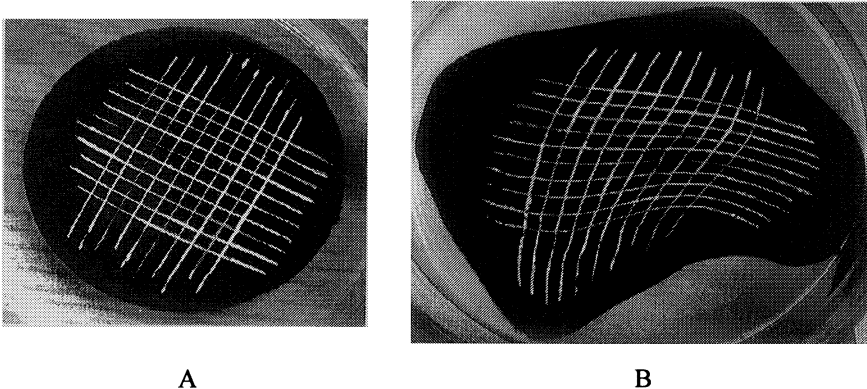


Fig.1: Illustration of the complex nonhomogeneous deformation of a ferrogel sheet. Undeformed (A) and deformed (B) states. The magnetic field was induced by permanent magnets placed around the gel.

3 Mechanical model for ferrogels

The mechanical behaviour of the gel is assumed to be hyperelastic, thus exists a strain-energy function W for which the neo-Hooke law is a good approximation:

$$W = \frac{G}{2} (\lambda_1^2 + \lambda_2^2 + \lambda_3^2 - 3) \quad (1)$$

where the material parameter G is the initial shear modulus. For incompressible materials, such as ferrogels, the equation of the constraint $\lambda_1 \lambda_2 \lambda_3 = 1$ is also needed.

The mechanical balance equations written with the nominal stress are

$$\text{Div } \mathbf{t}(\mathbf{X}) + \mathbf{b}_o(\mathbf{X}, t) = \mathbf{0}, \tag{2}$$

The vector \mathbf{b}_o is the body-force written with the terms of the initial configuration. On a continuous magnetic material due to the applied magnetic field it can be written as

$$\mathbf{b}(\mathbf{x}, t) = (\text{grad } \mathbf{M}(\mathbf{x}, t))\mathbf{H}(\mathbf{x}, t). \tag{3}$$

The vector $\mathbf{H}(\mathbf{x}, t)$ denotes the magnetic field strength and $\mathbf{M}(\mathbf{x}, t)$ represents the magnetization of the material. The ferrogel shows superparamagnetic material behavior. Its magnetization follows the one-valued equation [5]

$$\mathbf{M}(\mathbf{x}, t) = \Phi_m M_o \alpha(\xi) \frac{\mathbf{H}}{H}, \tag{4}$$

with the Langevin function $\alpha(\xi)$. The parameter Φ_m stands for the volume fraction of the magnetic particles in the gel, M_o represents the saturation magnetization of the material of the particles, m is the magnetic moment of an individual particle and equals to $M_o V_m$ in the case of a mono-domain particle, where V_m is the volume of the particle. At moderate magnetic fields linearized relation can be assumed between the magnetization and magnetic field

$$\mathbf{M} = \mu_o \chi \mathbf{H}, \tag{5}$$

where μ_o represents the permeability of the vacuum. This simplification results in a simplified expression for the magnetic body-force, namely

$$\mathbf{b}(\mathbf{x}, t) = \mu_o \chi (\text{grad } \mathbf{H})\mathbf{H}. \tag{6}$$

Substitution the constitutive relation into the equilibrium equation (eq.2), we obtain a set of three partial differential equations of the second order for the coordinates of the material points. If the material is incompressible, the hydrostatic pressure field p must be also calculated from the incompressibility constraint. For time-dependent problems, boundary conditions and initial conditions for the displacement and velocity fields are necessary. For quasi-static analysis only boundary conditions should be defined.

4 Uniaxial deformation

Because of the relatively complex form of the Langevin-type magnetization and the magnetic force density it is not possible to achieve analytical solution for Eq.2 even in simple magnetic field distributions. In order to reveal the characteristics of the magnetic field induced deformations of a magnetic continuum it is worth to examine simple one-dimensional situations.

Let us consider a very simple physical situation Long and thin ferrogel cylinder is suspended in water vertically. The axis of the gel cylinder (z) is parallel with the magnetic field and its gradient. In this case, the deformation of the gel is uniaxial and can be considered as one-dimensional. In this situation eq.2 results in the following second-order, non-linear ordinary differential equation:

$$G \left(\frac{d^2 u_z(Z)}{dZ^2} + \frac{2}{(du_z(Z)/dZ)^3} \frac{d^2 u_z(Z)}{dZ^2} \right) + M(u_z(Z)) \frac{dH(u_z(Z))}{dZ} = 0 \tag{7}$$

Here $u_z(z)$ means the displacement of the material point Z along the z axis. The relevant boundary conditions are: $u_z(0)=0$, and $t(Z_m)=0$. Here 0 and Z_m represents the position of the top and bottom end of the gel, respectively.

In our one-dimensional experiments the ferrogel cylinder was elongated by an inhomogeneous magnetic field induced by a solenoid-based electromagnet that was placed under the gel. The distribution of the field was determined along the axis of the gel by a teslameter.

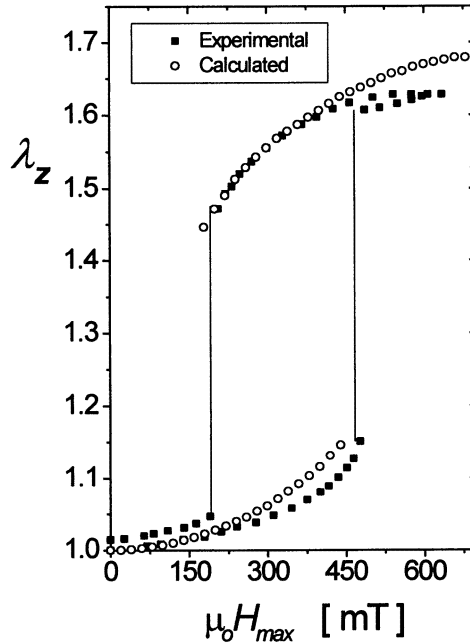


Fig.2: Uniaxial elongation of a ferrogel cylinder. The points represent the displacement of the bottom end of the gel. The blank points were calculated on the basis of Eq.7.

A typical result of the uniaxial experiments can be seen in Fig.2, where the displacement of the bottom end of the gel cylinder is plotted against the current intensity. It may be seen that a non-continuous change in the length appears within an infinitesimal change in the current intensity. While decreasing the current, another non-continuous size transformation occurs. The unique property of ferrogel's magnetoelasticity is the non-continuity, which is due to the non-linearity of the magnetic interactions. In Fig.2 we have also plotted the numerical solution of eq.7. The points fit quite well to the measured ones indicating the continuum model holds for ferrogels.

5 Finite element calculation

As mentioned previously, the analytical determination of the displacement field is generally extremely complicated; therefore a numerical procedure is necessary to calculate the deformation. Nowadays the finite element method (FEM) is prevalent in

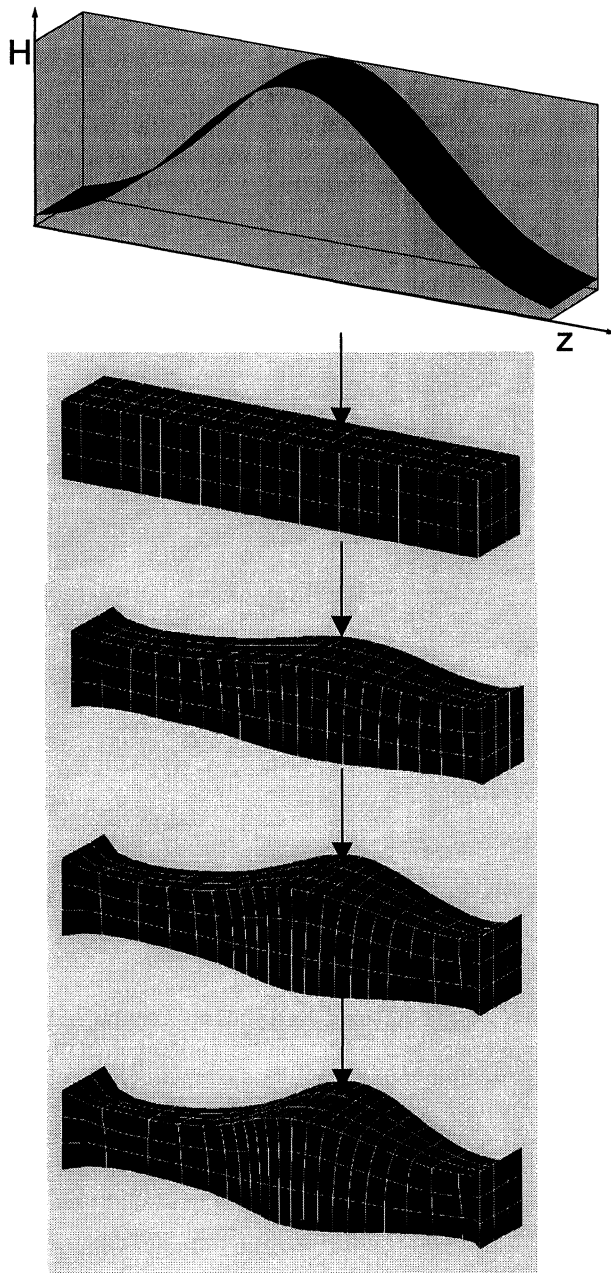


Fig.3: FEM calculation of the deformation of a ferrogel block in a uniaxial magnetic field. The field distribution is shown at the top. The arrows indicate the maximum field strength.

solid mechanics and provides a useful tool to study non-linear deformations in magnetic field. In what follows, we expose the results of our FEM procedure on ferrogels. With FEM fully 3D analysis can also be performed. In theory any arbitrary experimental situation can be modelled and analysed. As an illustrative example, in Fig.3 we present

the deformation of a ferrogel block in a uniaxial field parallel with the gel axis. Looking at the set of pictures one might associate with the motion of a living worm. And indeed, the deformation and motion of ferrogels has a close relation with the one of simple living organisms. This is due to the nonhomogeneity of the magnetic field induced deformations.

6 Conclusion

To summarize our results, we can conclude that the above-presented mechanical model for magnetic continuums can successfully describe the deformation in static magnetic fields. With the aid of numerical or FEM calculations based on the model, it is possible to reveal the basic characteristics of the magnetic field induced deformations (i.e. non-continuity, nonhomogeneity) or predict the deformation of a highly elastic magnetic substance in an arbitrary magnetic field distribution. However, the effect of the field distribution on the resulting deformation, which is probably the most interesting point, has not analyzed yet. We leave this for future works. The model can also serve as a basis for designing ferrogel-based actuators or artificial muscles whose importance does not need to emphasize.

References:

1. M. Zrínyi, *Trends in Polymer Science* **5**, 277 (1997)
2. M Zrínyi, L. Barsi, D. Szabó, H.-G. Kilian, *J. Chem. Phys.* **106**, 5685 (1997)
3. K. Bohon, S. Krause, *J.Polym.Sci.:Polym.Phys.* **36**, 1091 (1998)
4. T. Mitsumata, K. Ikeda, J. P. Gong, Y. Osada, D. Szabó, M. Zrínyi, *J. Appl. Phys.* **85**, 1 (1999)
5. D.Szabó, I.Czakó-Nagy, M.Zrínyi,A.Vértes, *Journal of Colloid and Interface Science* **221**, 166 (2000)