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Mesostructural origin of the field-induced pseudo-plasticity effect in a soft magnetic elastomer

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Mesostructural origin of the field-induced pseudo-plasticity effect in a soft magnetic elastomer

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Abstract. A model is presented to studying the pseudo-plasticity effect in a soft magnetic elastomer: a weakly-linked polymer matrix filled with micron-size particles of a ferromagnet with virtually zero coercivity (e.g., iron carbonyl). If the filling fraction of the composite is sufficiently high, such a material displays a remarkable ability to radically change its rheological behavior: being subjected to external magnetic field, the sample loses its elasticity and responds to external mechanical load as if being made of a ductile substance with no tendency to restore its initial shape. In our simulations we show that, considering the system at the mesoscopic scale (as an assembly of particles embedded in an elastic continuum) and taking into account just the magnetostatic and elastic interactions, one is able to qualitatively correct reproduce the field-induced pseudo-plasticity effect.

1. Introduction

Soft magnetic elastomers (SMEs) are functional composite materials obtained by filling soft polymer matrices with ferromagnet microparticles. Under moderate-to-high concentrations of the ferrophase (up to 30 wt.%), SMEs acquire high sensitivity to magnetic fields still retaining a good ability for large deformations. This combination makes these materials quite attractive for applications of which one can name adaptive dampers and vibration isolators, magnetically-controlled manipulators, gripper and other actoric devices of many kinds.

Along with the well-known magnetodeformational [1] and magnetorheological properties [2], e.g., field-induced shape changes and/or modulations of the elasticity moduli and internal friction, SMEs possess some unique and less evident effects. Of those, the most fascinating is the field-induced plasticity also known as magnetic shape memory effect [3, 4, 5, 6]. At the macroscopic scale it manifests itself as a field-induced transition in result of which a SME sample, instead of being elastic (under zero or low field) transforms in a piece of putty (under moderate-to-strong field).

The origin of the effect is doubtless: it is due to the processes which undergo at the mesoscopic scale and originates from the interparticle magnetoelastic interactions. However, mesoscopic models capable of detailing the mechanism of the pseudo-plasticity in a SME are either oversimplified or missing. In this work we present a numerical model that, in our view, on the one hand, is simple enough to make the solution comprehensible but, on the other hand, reveals the physical origin of all the essential effects – magnetic pseudo-plasticity, first place – stemming from mesoscopic structuring induced in a SME by external magnetic field and mechanical load.

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2. Problem statement

Mathematic formulation of the problem splits in two parts: elasticity theory and magnetostatics.

2.1. Elastic problem

Consider a SME sample that occupies a 3D region Ω and consists of an elastic matrix with N embedded solid spherical particles. To be able to subject the sample to mechanical loads, in Ω some auxiliary region Ω_h (not necessarily simply-connected) is included. It is always positioned at the outer border of Ω and does not contain any particles. By its elastic properties, Ω_h is more rigid than the main polymer domain.

The space occupied by i-th particle is denoted as $\Omega_i^{(p)}$, so that the space of the matrix is $\Omega^{(m)} = \Omega \setminus \Omega_h \setminus \Omega_1^{(p)} \setminus \Omega_2^{(p)} \ldots \setminus \Omega_N^{(p)}$. Therefore, each geometrical point inside Ω belongs to one and only one of three elastically different sets: they are either a particle (p), the soft part of the matrix (m), or the rigid part of the matrix (h). Accordingly, we introduce three indicator functions:

$$I_i^{(p)}(\boldsymbol{r}) = \begin{cases} 1, & \boldsymbol{r} \in \Omega_i^{(p)} \\ 0, & \boldsymbol{r} \notin \Omega_i^{(p)} \end{cases}, \quad I^{(m)}(\boldsymbol{r}) = \begin{cases} 1, & \boldsymbol{r} \in \Omega^{(m)} \\ 0, & \boldsymbol{r} \notin \Omega^{(m)} \end{cases}, \quad I^{(h)}(\boldsymbol{r}) = \begin{cases} 1, & \boldsymbol{r} \in \Omega^{(h)} \\ 0, & \boldsymbol{r} \notin \Omega^{(h)} \end{cases}, \tag{1}$$

where $\boldsymbol{r}$ is radius-vector of any inner point of the SME sample.

In a general case, each i-th particle of the considered sample experiences force $\boldsymbol{F}_i^{(p)}$ that is exerted on its center. When subjected to fields and loads, the SME sample is fixed by the part Γ_u of its outer surface whereas another part of the same surface, Γ_f, is subjected to a uniformly distributed mechanical load $\boldsymbol{F}_b$.

Then the set of elasticity theory equations [7] for the problem under study takes the form

$$\nabla \cdot \boldsymbol{\sigma} + \boldsymbol{f} = 0, \quad \boldsymbol{f} = \sum_{i=1}^{N} I_i^{(p)} \boldsymbol{F}_i^{(p)} / V_i,$$

$$\boldsymbol{\sigma} = -p\mathbf{g} + 2G\mathbf{e}, \quad \operatorname{tr} \mathbf{e} = 0, \quad G = G_m I^{(m)} + G_h I^{(h)} + \sum_{i=1}^{N} I_i^{(p)} G_p, \tag{2}$$

$$\mathbf{e} = \frac{1}{2}(\nabla \boldsymbol{u} + \nabla \boldsymbol{u}^{\mathrm{T}}), \quad \boldsymbol{n} \cdot \boldsymbol{\sigma}|_{\Gamma_f} = \boldsymbol{F}_b / S_f, \quad \boldsymbol{u}|_{\Gamma_u} = \boldsymbol{u}^*,$$

where $\mathbf{g}$ is unit tensor, $\boldsymbol{u}$ displacement vector, G_m shear modulus of the matrix, G_h shear modulus of the fixed part of the matrix, G_p shear modulus of the particle (it is much greater than G_m), S_f is the surface area of Γ_f, and λ is Lamé constant.

At the mesoscopic level, forces $\boldsymbol{F}_i^{(p)}$, initiate translational and rotational displacements of i-th particle; at the macroscopic level, the collective effect of those motions results in displacements of the sample border Γ_f. To deal with the situation, it is convenient to introduce the averaged values

$$\boldsymbol{u}_i^{(p)} = \frac{1}{V_i} \int_{\Omega_i^{(p)}} \boldsymbol{u} \, dV, \quad \boldsymbol{u}^{(b)} = \frac{1}{S_f} \int_{\Gamma_i^{(p)}} \boldsymbol{u} \, dS, \tag{3}$$

of the particle and border displacements, respectively; here V_i is the volume of i-th particle and $\Gamma_i^{(p)}$ its surface.

Defining the generalized vectors of forces $\boldsymbol{F}$ and displacements $\boldsymbol{U}$ as

$$\boldsymbol{F} = \{\boldsymbol{F}_1^{(p)}, \boldsymbol{F}_2^{(p)}, \ldots, \boldsymbol{F}_N^{(p)}, \boldsymbol{F}^{(b)}\}, \quad \boldsymbol{U} = \{\boldsymbol{u}_1^{(p)}, \boldsymbol{u}_2^{(p)}, \ldots, \boldsymbol{u}_N^{(p)}, \boldsymbol{u}^{(b)}\}, \tag{4}$$

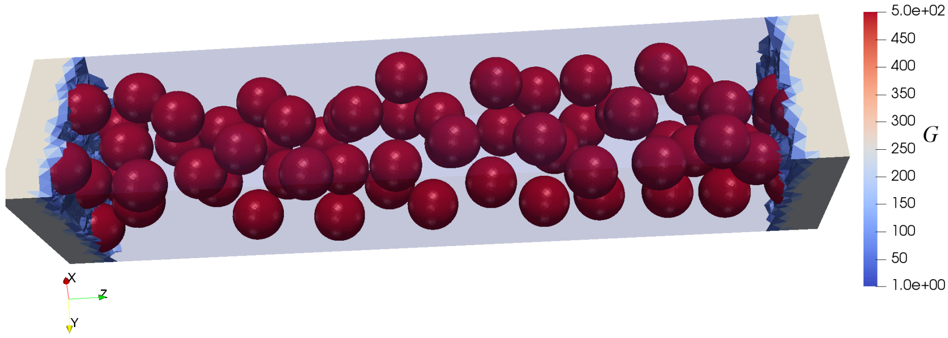

Figure 1. A test SME sample presented as a continuum material with piece-wise shear modulus that assumes the values: $G_m = 1$, $G_p = 500$, $G_h = 250$, see the color scale.

and introducing $\mathbf{C}$, the matrix of linear elastic response of an assembly of solid particles and of the system border Γ_f, we arrive at the relation

$$\boldsymbol{U} = \mathbf{C} \cdot \boldsymbol{F}, \tag{5}$$

that couples the averages $\boldsymbol{u}_i^{(p)}$ and $\boldsymbol{u}^{(b)}$ with the exerted forces $\boldsymbol{F}_i^{(p)}$ $\boldsymbol{F}^{(b)}$:

To find matrix $\mathbf{C}$ one has to $3(N+1)$ times solve the elastic problem (2) using esys-escript (programming tool for implementing finite element method in python [8, 9]). The force vector at i-th step is defined in such a way that $F_k = 0$ if $k \neq i$, and $F_i = 1$. Then vector $\boldsymbol{U}$ is calculated that makes i-th row of matrix $\mathbf{C}$. For further use it is convenient to introduce inverse tensor $\mathbf{L} = \mathbf{C}^{-1}$ so that equation (5) transforms into

$$\boldsymbol{F} = \mathbf{L} \cdot \boldsymbol{U}. \tag{6}$$

The increment of the work generated by forces $\boldsymbol{F}_i^{(p)}$ and $\boldsymbol{F}^{(b)}$ at respective displacements $\boldsymbol{u}_i^{(p)}$ and $\boldsymbol{u}^{(b)}$ is

$$dW = \boldsymbol{F} \cdot d\boldsymbol{U} = \boldsymbol{U} \cdot \mathbf{L} \cdot d\boldsymbol{U}. \tag{7}$$

According to energy conservation law, this work equals the elastic energy stored in the matrix. After integration one gets expression for the total elastic energy in the form

$$W = U_{\text{el}} = \tfrac{1}{2}\boldsymbol{U} \cdot \mathbf{L} \cdot \boldsymbol{U}. \tag{8}$$

2.2. Magnetostatic problem

A point magnetic dipole positioned in the origin of coordinate frame possesses magnetostatic potential [10]

$$\psi = \frac{\boldsymbol{\mu} \cdot \boldsymbol{r}}{r^3}. \tag{9}$$

and induces around it magnetic field

$$\boldsymbol{h}(\boldsymbol{r}, \boldsymbol{\mu}) = -\nabla\psi = -\frac{\boldsymbol{\mu}}{r^3} + 3\frac{\boldsymbol{r}}{r^5}\boldsymbol{\mu} \cdot \boldsymbol{r}. \tag{10}$$

If the dipoles are magnetically soft, i.e., are induced by external field, then the magnetic field created In an arbitrary space point $\boldsymbol{r}$ by an assembly of N such entities positioned at

$\boldsymbol{r}_j = \boldsymbol{r}_j^0 + \boldsymbol{u}_j^p$ and possessing magnetic moments $\boldsymbol{\mu}_j$ due to the presence of external magnetic field $\boldsymbol{H}_0$, writes as

$$\boldsymbol{H}(\boldsymbol{r}) = \boldsymbol{H}_0 + \sum_{j=1}^{N} \boldsymbol{h}(\boldsymbol{r} - \boldsymbol{r}_j, \boldsymbol{\mu}_j). \tag{11}$$

Let us assume that the particles are made of a ferromagnet that magnetizes linearly, i.e., with susceptibility χ independent of the field strength. In that case, the magnetization of a spherical particle is uniform and is given by expression $\boldsymbol{\mu}/V = \boldsymbol{m} = \chi \boldsymbol{H}^{\text{int}}$ with $\boldsymbol{H}^{\text{int}}$ being the field inside the particle. Using relation $\boldsymbol{H}^{\text{int}} = \boldsymbol{H} - 4\pi\boldsymbol{m}/3$ that links vectors of internal and external fields in case of a spherical body with magnetization $\boldsymbol{m}$, one finds the internal field as

$$\boldsymbol{H}^{\text{int}} = \frac{\boldsymbol{H}}{1 + 4\pi\chi/3}, \tag{12}$$

that yields for the magnetic moment of i-th particle:

$$\boldsymbol{\mu}_i = \frac{V_i}{\chi^{-1} + 4\pi/3} \left(\boldsymbol{H}_0(\boldsymbol{r}_i) + \sum_{j=1, j\neq i}^{N} \boldsymbol{h}(\boldsymbol{r}_i - \boldsymbol{r}_j, \boldsymbol{\mu}_j) \right), \quad i = \overline{1, N}. \tag{13}$$

With the use of notations $\boldsymbol{r}_{ij} = \boldsymbol{r}_i^0 - \boldsymbol{r}_j^0 + \boldsymbol{u}_i^{(p)} - \boldsymbol{u}_j^{(p)}$, $r_{ij} = |\boldsymbol{r}_{ij}|$, $\boldsymbol{H}_0(\boldsymbol{r}_i) = \boldsymbol{H}_{0i}$ this equation takes the form

$$\sum_{j=1}^{N} \left[(1 - \delta_{ij}) \left(\frac{\mathbf{g}}{r_{ij}^3} - 3\frac{\boldsymbol{r}_{ij}\boldsymbol{r}_{ij}}{r_{ij}^5} \right) + \delta_{ij}\mathbf{g}\frac{\chi^{-1} + 4\pi/3}{V_i} \right] \cdot \boldsymbol{\mu}_j = \boldsymbol{H}_{0i}, \tag{14}$$

that explicitly presents it as a set of $3N$ linear algebraic equations for the vector components of magnetic moments $\boldsymbol{\mu}_j$ of all N particles contained in the model SME sample.

To make the left-hand side of equation (14) compact, one more notation is introduced:

$$\mathbf{A}^{ij}(\boldsymbol{U}) = \begin{cases} \mathbf{g}/r_{ij}^3 - 3\boldsymbol{r}_{ij}\boldsymbol{r}_{ij}/r_{ij}^5, & i \neq j \\ \mathbf{g}(\chi^{-1} + 4\pi/3)/V_i, & i = j \end{cases}. \tag{15}$$

so that with its aid the set (14) writes as

$$\sum_{j=1}^{N} \mathbf{A}^{ij} \cdot \boldsymbol{\mu}_j = \boldsymbol{H}_{0i}. \tag{16}$$

The last expression could be presented also in a block-matrix form::

$$\begin{pmatrix} [A_{xx}^{ij}] & [A_{xy}^{ij}] & [A_{xz}^{ij}] \\ [A_{yx}^{ij}] & [A_{yy}^{ij}] & [A_{yz}^{ij}] \\ [A_{zx}^{ij}] & [A_{zy}^{ij}] & [A_{zz}^{ij}] \end{pmatrix} \cdot \begin{pmatrix} [(\mu_x)_i] \\ [(\mu_y)_i] \\ [(\mu_z)_i] \end{pmatrix} = \begin{pmatrix} [(H_{0x})_j] \\ [(H_{0y})_j] \\ [(H_{0z})_j] \end{pmatrix} \tag{17}$$

where all the matrices are defined in the same cartesian coordinate frame.

Finally, with appropriately modified notations, the matrix set (17) may be cast in the form

$$\tilde{\mathbf{A}} \cdot \tilde{\boldsymbol{\mu}} = \tilde{\boldsymbol{H}}_0, \tag{18}$$

that enables one to write the magnetostatic the energy of magnetized dipole assembly as

$$U_{mag} = -\tfrac{1}{2} \sum_{j=1}^{N} \boldsymbol{\mu}_j \cdot \boldsymbol{H}_0 = -\tfrac{1}{2} \tilde{\boldsymbol{H}}_0 \cdot \tilde{\mathbf{A}}^{-1}(\boldsymbol{U}) \cdot \tilde{\boldsymbol{H}}_0. \tag{19}$$

2.3. Coupled problem
Uniting the elastic and magnetic terms, i.e., equations (8) and (19), one arrives at the total energy of the system:

$$U_{\text{tot}} = \tfrac{1}{2}\boldsymbol{U}\cdot\mathbf{C}\cdot\boldsymbol{U} - \tfrac{1}{2}\tilde{\boldsymbol{H}}_0\cdot\tilde{\mathbf{A}}^{-1}(\boldsymbol{U})\cdot\tilde{\boldsymbol{H}}_0. \tag{20}$$

Therefore, to find the equilibrium configuration one should minimize function (20) under requirement that the particles do not overlap:

$$\boldsymbol{r}_i^0 + \boldsymbol{u}_i^{(p)} - \boldsymbol{r}_j^0 + \boldsymbol{u}_j^{(p)} > R_i + R_j, \quad i \neq j.$$

To find minimum we use sequential least squares programming algorithm [11].

2.4. Test sample
As a test SME sample we take a prolate rectangular prism pointing along Oz whose dimensions are $l_x = l_y$ and $l_z/l_x = 5$. Its three lateral sides coincide with the coordinate planes xOy, xOz, and yOz. The sample is sketched in Fig. 1, it contains 66 identical spherical particles of a linearly magnetizable ferromagnet. The particles are positioned at random, their volume content is 26%. The distribution of shear modulus in the sample is the same as indicated in at Fig. 1. Boundary conditions on the displacements $\boldsymbol{u}$ are: $u_x|_{x=0} = 0$, $u_y|_{y=0} = 0$, $u_z|_{z=0} = 0$; this excludes Euler instability of the sample under compression

3. Results and discussion
In Figure 2 we present the sequence of snapshots showing configurations of the test sample in the initial state (Fig. 2a), subjected to a longitudinal field (Fig. 2b); under the same field but compressed with force F_b (Fig. 2c) and under the same field but unloaded (Fig. 2d). As seen, the applied field makes the particles to get close and form distinctive chains, see Fig. 2b. The occurring change of longitudinal dimension of the sample is insignificant. This is quite understandable since in a sample where the particle concentration is rather large, the neighboring particles, need to cover but very small distances to pull together. Mechanical pressure exerted on the upper end-wall makes the sample to reduce its longitudinal dimension; the length of chains reduces at the expense of increasing their number, see Fig. 2c. Since all the components of the test sample are assumed to be incompressible, its longitudinal shortening is accompanied by enhancement of the transverse dimensions.

The most interesting effect is evidenced in Fig. 2d. One sees that after switching off the mechanical stress, the sample does not recover the starting configuration of Fig. 2b. Therefore, the presented example demonstrates explicitly that a mechanical load, once exerted and then removed, imparts to a magnetized sample a residual strain that would exist until the field is on. Our simulations show that only after turning off the field, the sample fully recovers its initial configuration, that of Fig. 2a.

The explanation is apparent: the studied system is described by a set of non-dissipative equations. In the initial state it dwells in equilibrium (zero field, zero load), and the internal elastic stresses are completely absent. All the actions on the sample, first, magnetization and then pressure, perturb the initial equilibrium and induce internal (mesoscopic) elastic stresses. As long as the external factors are on, they maintains the elastic stresses in the matrix at a certain level that is higher when both field and load act together, or lower when it is only the field effect. Only after all the perturbing factors are eliminated, the mesoscopic stresses become free to relax and return the system to the initial state.

Therefore, the overall cycle of the sample transformation – passage from stage (a) through (b), (c), (d) and back to (a) in terms of Fig. 2 – is reversible. However, the internal cycle that starts from stage (b) of Fig. 2 through (c) and back to (d) is irreversible. Evidently, the assembly of magnetized particles, when forced to rearrange under pressure, finds a more favorable configuration: under zero mechanical load the energy U_{tot} of pattern (d) of Fig. 2 is lower than that of pattern (b).

The effect of residual strain should strongly depend on the magnitude of applied field $\boldsymbol{H}_0$. Indeed, in the limiting case $H_0 = 0$ the pressure cycle is reversible as there is no magnetostatic term to counteract the elastic one in the energy expression. The pressure cycles of our test sample are presented in Fig. 3 for several values of the applied field. The strain there is defined as $\varepsilon_{zz} = u_z^{\mathrm{top}}/l_z$, where u_z^{top} is the displacement of the sample end-wall which in the initial state is positioned at $z = 0$; the non-dimensional applied stress is characterized by the ratio $F_b/(S_f G_m)$.

As seen, at a non-zero but low field the pressure cycle remains reversible that means that the available magnetic energy gain is yet too small to compete with the elastic stresses. The irreversibility turns up at enhanced fields, where stress–strain curves become loop-shaped that is typical for plastic materials, and the residual strain attains quite high values, up to $\sim 20\%$. Therefore, the magnetic field gives the means to modulate the plasticity of the material.

Notably, the dependence of residual strain on the applied field is not monotonic.

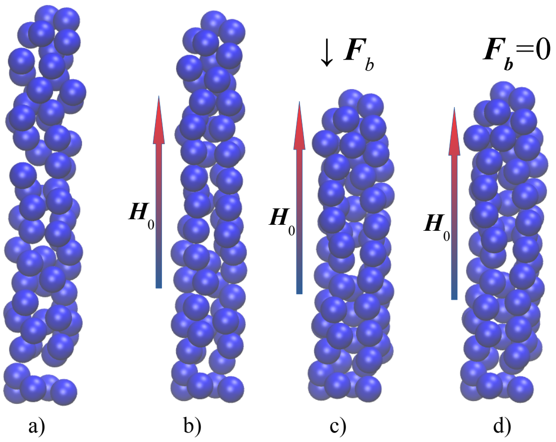

Figure 2. Distribution of the particles in the sample under: (a) – magnetic field $H_0 = 0$ and end-wall force $F_b = 0$; (b) – $H_0 = 10\sqrt{G_m}$ and $F_b = 0$; (c) – $H_0 = 10\sqrt{G_m}$, $F_b = -6\sqrt{G_m}$; (d) – $H_0 = 10\sqrt{G_m}$ and $F_b = 0$.

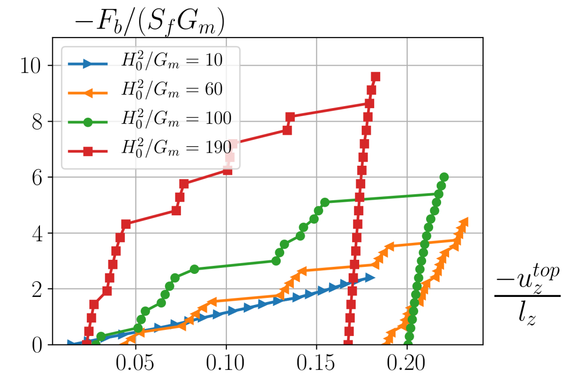

Figure 3. Stress-strain dependencies for the test sample under variation of the applied field.

4. Conclusions

A mesoscopic model of a soft magnetic elastomer that takes into account the magnetic and elastic interactions of the particles is presented. In the performed simulations, a combination of field and pressure cycles exerted on the test sample is studied. It is shown that a material with high concentration of magnetically soft microparticles, if subjected to a constant uniform magnetic field, displays a distinctive pseudo-plasticity effect in a pressure cycle. The magnitude of the effect is strongly depends on the strength of external field in a non-monotonic way. In general, the obtained model dependences agree well with the qualitative experimental evidence reported in literature.

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