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Field-induced plasticity of soft magnetic elastomers

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Abstract. A phenomenological model is proposed to describe the plasticity of soft magnetic elastomer composites under an external field. Along with high-elasticity, internal dry friction is introduced whose origin is attributed to the dipole-dipole interaction of the embedded particles. Numerical estimates of the model parameters are obtained from comparison with the measurements performed on iron carbonyl dispersions in soft silicon-rubber matrices.

Introduction

Magnetorheological elastomers are an important class of functional materials. The unique feature ensuring their wide prospects in technical and biomedical applications, is the strong dependence of the mechanical properties on the applied magnetic field [1-9]. Inside this class, soft magnetic elastomers (SMEs) make a separate family distinguished by giant magnetomechanical effects. Typical example is a weakly-linked silicon rubber filled with iron carbonyl grains (2-5 μ m) to the volume fraction $\phi \sim$ 30% [2-4,6,7], its Young modulus being ~100 kPa. SMEs are prepared by polymerization of magnetic dispersions carried out either in the absence or in the presence of external field. Accordingly, the resulting SMEs acquire either a homogenous particle distribution or a chain-like texture [7-9].

The mechanics of SMEs depends crucially on the applied field H. At H = 0 a SME sample responds to elongation up to ~100% in linear elastic (reversible) manner. The same sample under $H \sim 100$ kA/m, being stretched to the same strain, retains a substantial part of it even after the stress is removed. Moreover, this residual strain is the greater the higher H [6]. As soon as the field is switched off, the sample restores its initial shape and properties [4]. The observed state of *field-induced plasticity* and the effect of *magnetic shape memory* that is based on it, are evidently due to re-grouping of the embedded magnetic particles. Hereby we propose a phenomenological model that ties up the field-induced structure changes in SMEs with their above-mentioned macroscopic behavior.

SME in a strain cycle. Discussion of experiment

In Fig. 1 the points present the stress-strain data obtained in loading cycles of an isotropic, i.e., prepared under zero field, silicon rubber SME with $\phi = 30$ vol.% of iron carbonyl [6]. The sample is a cylinder of aspect ratio ≈ 6 magnetized parallel to its axis. At H = const. the sample is stretched slowly along its axis to a certain length, then the load is reduced to zero with the same rate. When, after the cycle, the field is turned off, the sample practically restores its initial size. Thus, the main facts are:

1) deformation is virtually reversible only under H = 0;

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2) with the increase of the applied field the initial elastic modulus grows substantially;

- 3) under deformation in a constant field, the modulus undergoes appreciable changes;
- 4) being subjected to a loading cycle, a magnetized sample gains a notable residual strain which it retains as long as the field is on.

As the magnetic anisotropy of iron carbonyl is very low, at H = 0 the embedded particles are multidomain, and there is no magnetic interaction between them. When the field is turned on, the induced magnetic moments of the particles enter into dipole-dipole interaction that strives to organize chainlike particle aggregates (clusters). In a soft network the dipole forces, which grow rapidly ($\propto 1/r^4$) with the particles closing in, dominate the elastic ones. However, the network, although distorted, does not break, and remains attached to the particles. Under mechanical loading, the joint reaction of the network and the clusters determines the macroscopic response of the material, Fig. 1 is an example.



Figure 1. Loading/unloading deformation processes for a SME cylinder under constant fields 96 kA/m (squares) and 207 kA/m (diamonds); in each test the strain is counted from the size that the sample assumes after magnetization in a stress-free state. Dashed lines present a "uniform" fitting with the model; for the numerical values of the parameters see Table 1.

Let a stress-free SME cylinder be magnetized along its axis by a uniform constant field. The occurring cluster formation results in some macroscopic striction of the sample. When a small load is applied, it is counteracted be the polymer network while the existing clusters behave like solid particles. Further loading increases the local stresses in the matrix. As soon as they reach the "magnetic durability" of the clusters, there begins intense destruction of the existing and formation of the new ones. This endows SME with a mechanism of internal dry friction. Indeed, let the load grow a bit. The network transfers this increment to the clusters, and the latter break in their weakest points. Broken clusters do not resist displacements, and locally the material begins to flow. As the filling is sufficiently high, the moving fragments of a given cluster inevitably approach their analogues emerging from the other ones. From this "debris" the magnetodipole attraction builds up new, more dense, clusters capable to stand the enhanced load; thereby the plasticity flow stops. Macroscopically, stretching of a SME in this regime, being a superposition of ductile and elastic reactions, looks like an elastic response with the effective modulus that is appreciably lower than the initial one.

On halting the deformation, the sample would rest in equilibrium with respect to the actual level of external loading and the internal stress distribution. The reverse process, i.e., quasi-static unloading, induces global shrinking of the network and reduction of internal stresses. At the beginning this does not affect the structure of the clusters; with further decrease of the load, the local stresses on the clusters (now their direction is opposite to that while stretching) induce a rearrangement of the same break/rebuild kind (quasi-plasticity). This regime keeps on until the internal stresses fall below the level at which they are able to rebuild the clusters. In result, under complete unloading the sample

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retains some strain that is maintained by local stresses induced by the set of magnetized clusters whose distribution is different from that existed before the load was applied for the first time.

Turning off the magnetic field radically changes the internal stress balance. As the particles are magnetically soft (no remanence), the magnetodipole forces turn to zero. Accordingly, the particles cease to "sense" each other, and all the clusters fall apart. In result, the local high-elasticity stresses, "frozen" in the presence of the field, get in action. The network (with the particles attached to it) completely restores its configuration, and the SME sample recovers (recalls) its shape.

Model

A simple heuristic scheme (HS) is shown in Fig. 2. Of the two parallel branches, one is a linear elastic spring of rigidity G_1 while another contains a sequence of a spring of rigidity G_2 and a dry-friction element with critical shear S^* . This presents a SME as two interwoven networks which deform affinely. One is purely elastic while another is composed of sequences of elastic and rigid (cluster) segments. The strains of both branches are equal: $\varepsilon = \varepsilon_1 = \varepsilon_2$, the total stress on the HS is the sum of the partial stresses. As follows from Fig. 1, the rheological characteristics of a SME depend on the applied field. Accordingly, we assume that all the parameters of the HS are functions of *H*.

For small loading (the stress in branch 2 is below S^*) only the springs work, so that the net rigidity of the HS is G_1+G_2 . In Fig. 1 this regime corresponds to straight lines departing from zero points. When the stress in branch 2 grows up to S^* (the corresponding strain is $\mathcal{E}_* = S^*/G_2$), the resistance of the dry-friction element falls down to zero, and the stress in branch 2 becomes strain-independent. In this regime the stress induced by the HS is due only to spring 1. Accordingly, the ascend angles of the lines $S(\mathcal{E})$ in Fig. 1 drop down from $\arctan(G_1+G_2)$ to $\arctan(G_1)$. When the strain is halted at some value \mathcal{E}_m , the equilibrium stress equals $S_m = G_1\mathcal{E}_m + S^*$. Here the dry-friction element stops to slide but the stress S^* in branch 2 is maintained by spring 2 stretched to the strain $\mathcal{E}_* < \mathcal{E}_m$, i.e., $S_m = G_1\mathcal{E}_m + G_2\mathcal{E}_*$.





Consider now the diminution of the load. As both springs are stretched, they strive to recover their initial sizes; in the corresponding parts of Fig. 1 the dependencies $S(\varepsilon)$ go down with the tangent $dS/d\varepsilon = -(G_1+G_2)$. When the stain $\varepsilon_m - \varepsilon_*$ is achieved (branch 2 acquires it in result of sliding of the dry-friction element), spring 2 assumes its initial (equilibrium) length while spring 1 is yet stretched. Under further unloading, spring 1 still strives to shrink but to do that it has to compress spring 2. Therefore, the stress in branch 2 becomes opposite to that in the stretching regime. If the elasticity of spring 2 is invariant with respect to elongation/compression, the rigidity of the HS does not change and remains equal G_1+G_2 . When and if the stress in branch 2 achieves $-S^*$, the dry-friction element "takes off". In this situation the only source of shrinking of the HS is the excessive energy of spring 1.

Shrinking stops when the stress on the HS turns to zero, that is the partial stresses compensate each other. The residual strain of the HS in this state is ε_r . The value of ε_r depends on the fact whether or not the dry-friction element worked during the unloading process. This, in turn, is determined by the magnitude of the maximum strain ε_m achieved in the cycle. If ε_m exceeds the critical value

$$\varepsilon_{\max} = \varepsilon_* (2 + G_2 / G_1), \qquad (1)$$

then the residual strain is $\mathcal{E}_r = (G_2/G_1)\mathcal{E}_*$. In the opposite case, it depends linearly on \mathcal{E}_m :

$$\varepsilon_r = G_2 \left(\varepsilon_m - \varepsilon_* \right) / \left(G_1 + G_2 \right). \tag{2}$$

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Numerical values of the three parameters of the HS are obtained from comparison of the model loops with the data of Fig. 1. The spring rigidities are determined from the tangents of the $S(\varepsilon)$ curves. The point ε_* at which the tangent falls down corresponds to the onset of the ductile regime. The value $S(\varepsilon_*) = \varepsilon_*G_1 + S^*$ enables one to estimate the dry friction shear S^* . As the experimental data never give any precise tangent breakup points, these numbers contain some uncertainties. At the first sight, the residual strain ε_r , could be determined more accurately. However, to interpret the obtained value it is necessary to find first, whether the maximal achieved strain ε_m exceeds the value ε_{max} , given by formula (1). For that, one has to employ the estimate of ε_* which accuracy, as mentioned, is not high.

For our HS essential is the quantity ε_* , at which the mechanism of magnetic plasticity (cluster rearrangement) is launched. When choosing the HS parameters to interpret the data of Fig. 1, we try to satisfy two conditions at once: 1) maximal closeness of theoretical and experimental values of the threshold strain ε_* ; and 2) maximal geometric resemblance between the model cycle and the experimental dependence $S(\varepsilon)$. The obtained numerical results are given in Table 1.

Table 1. Estimates for the parameters of the model obtained from comparison with the data
of Fig. 1 (elongation regime).

<i>H</i> , kA/m	G_1 , kPa	<i>G</i> ₂ , kPa	E*	\mathcal{E}_{r}	\mathcal{E}_{m}	S^* , kPa
96	28	164	0.096	0.56	0.9	15.7
207	41	413	0.057	0.57	0.72	23.1

According to Fig. 1, at H = 96 kA/m the observed modulus $dS/d\varepsilon$ at the end of unloading falls down significantly: the curve bulges rightward. On the other hand, under the field 207 kA/m the same effect is virtually absent. A probable cause of such a behavior are slow relaxation processes. If so, on diminution of the unloading rate, the bulging of $S(\varepsilon)$ curves should decease.

Conclusions

A remarkable phenomenon of magnetic shape memory displayed by soft magnetic elastomers is based on the field-induced plasticity effect. Although the details of microscopic behavior of real composites are very complicated, a simple phenomenological model quire reasonably accounts for some essential features of magneto-mechanics of SMEs. The presented analysis shows, in particular, that to explain the mechanical response of the material, one has to assume the presence of internal dry friction. A possible origin for such a mechanism in SMEs based on magnetically soft particles is pointed out.

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