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Effect of local elasticity of the matrix on magnetization loops of hybrid magnetic elastomers

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ABSTRACT

To model magnetization loops of magnetorheological elastomers (MREs) with magnetically hard filler, we consider an assembly of single-domain particles possessing inversion-symmetrical shape and embedded in a soft polymer matrix. To describe the intrinsic behavior of the particle magnetic moments under an applied field, the Stoner-Wohlfarth approach is employed. Unlike the case of solid matrix, the particle in a MRE is able to rotate relative to its elastic environment, so that its equilibrium orientation results from the balance between the magnetic torque (exerted by the applied field at any magnetic moment that does not point along the field) and the elastic torque generated by the matrix. We assume that elastic resistance to the field-induced particle rotation could be presented as comprising of two contributions. The first one is valid for any not perfectly spherical particle and is independent of the particle–matrix adhesion. It reflects the fact that, when trying to rotate, the particle has to "shoulder its way" by deforming the adjoining regions of the matrix. In that case, for angular deviations up to 90° from the initial position, the resistance torque increases. However, as soon as the rotation angle grows up to the value but infinitesimally exceeding 90°, the elastic torque changes its sign and from now on forces the particle to rotate to 180°, where it attains the geometrical position that coincides with the initial one. Evidently, this process is of the barrier type: both orientations of the particle are equal in elastic energy. The second mechanism stems from the "memory" that a given particle has of its initial state and may be caused, for example, by some macromolecules grafted to its surface while curing the matrix of the MRE. This restoring force always tends to drag the particle to its 0° (initial) position. Therefore, the observed magnetization of a MRE sample comes out as a result of joint interplay of the Stoner-Wohlfarth and the two elastic mechanisms. In our simulations, we show that the proposed model in a natural way accounts for the two essential features observed (solely or together) in experiment, namely: (*i*) weak net coercivity of the MREs, whose filler particles as themselves are highly coercive, and (*ii*) asymmetric positions of the magnetization loops with respect to $H = 0$ point at the $(M - H)$ plane.

1. Introduction

Composite materials consisting of a soft polymer matrix and embedded into it magnetic particles, can change their physical properties in response to the applied magnetic field. Due to this specific ability, such smart materials are called magnetorheological, magnetoactive or magnetosensitive elastomers. Being synonyms, these three terms are used interchangeably in literature [1–3]. The interest to these field-responsive materials is based on wide prospects of their technical and high-tech [4–6], and biomedical [7,8] applications. In below, we discuss some aspects of internal magnetomechanics of these composites under the action of external fields and hereafter use MRE as a unique acronym designating these composites.

Hybrid magnetic elastomers containing a mixture of two distinctly different magnetic fillers – magnetically hard (MH) and magnetically soft (MS) microparticles – make a special class of MREs. As shown in Refs. [9–11], the magnetic properties of hybrid MREs differ significantly both from those of customary MREs with just magnetically soft filler and from those of solid magnetics. In this work we investigate theoretically a model hybrid MRE which contains only MH particles. To justify this choice, we remark that, according to the available experimental data, the MS component, if added, apart from increasing the saturation magnetization of the material, does not bring any new specifics in magnetic behavior

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of a sample. Experimental investigation of the MH particles magnetization has been presented in Refs. [1,9–11].

In what follows, we, first, develop a model describing the response of a magnetically hard particle embedded in a soft elastomer matrix. We then use that model to simulate magnetization loops of the samples and, finally, give an example of application of the theory taking for that the data obtained on MREs filled with NdFeB microparticles.

2. Model

When constructing the model, we aim at the experimentally available MREs where the MQP-S-11-9-20001-070 powder (Magnequench, Inc.) was used as a filler. This powder consists of initially isotropic roundish NdFeB-alloy particles with mean linear size of 46.8 μm, according to the manufacturer. Due to production imperfections, a considerable fraction of the particles come out resembling prolate or oblate ellipsoidal shapes than are perfectly spherical.

According to the manufacturer, the particles are produced during centrifugal or spinning cup atomization, which is a type of rapid solidification processes [12]. A NdFeB microparticle of that kind consists of metallurgical grains with the size about 20 nm [13] that is below the critical size of single-domainness for NdFeB (0.21 μm [14]). Since in the melt-spun materials the domain walls coincide with the grain borders [15], then during magnetization these single-domain grains should fairly accurately obey the Stoner-Wohlfarth (SW) model [16]. The same should apply to the materials produced by the HDDR (hydrogenation, disproportionation, desorption, and recombination) method, which yield the grains about 0.3 μm [17].

When subjected to a strong field (first magnetization), each grain (domain) of the NdFeB microparticle switches in the direction of the field. On turning off the field, the elementary magnetic moment falls to the closest (in orientational sense) potential well. The resulting net magnetic moment of the particle, in accordance with the SW model, is a half of the maximal value [10,16] Due to that, under a cycled field (its negative values included) the microparticle displays a magnetization loop very similar to the hysteresis obtained with the SW model for an assembly of grains with random distribution of uniaxial anisotropy axes [10]. Hereby, we neglect this detail and approximate magnetization of a NdFeB particle, once subjected to a strong field, with a single SW cycle. As it will be demonstrated in Section 4, the discrepancies resulting from that deliberate simplification are of some quantitative nature but are not relevant for the main line of the work: introducing an extended concept of magnetization for the MREs filled with MH particles.

Bender et al. [18,19] modified the conventional SW energy expression to allow for the linear elastic response (Hook resistance) of the matrix to mechanical rotations of the particles. It has been done in order to study polymer-based dispersions with acicular magnetically hard filler. Hereby we modify this magnetoelastic approach in the following way. Consider a typical MRE based on weakly linked silicone rubber plasticized with silicone oil. In such a matrix, an MH particle sits inside a cavity formed around it during the polymerization process. It is tightly enveloped by the environment but is not too strongly "glued" to it. Because of that, in response to an exerted mechanical torque, the particle would strive to rotate. Moreover, in this motion the plasticizer distributed all over the matrix would serve as a lubricant.

If the particle in question is to some extent anisometric, then for its rotation it ought to move apart the surrounding matrix because, when turned from the equilibrium position, the particle is not any longer congruent with the cavity. Apparently, this inference equally applies to prolate and oblate objects. Let $\boldsymbol{n}$ be the unit vector of major axis of an axially symmetrical particle, and $\boldsymbol{v}$ the unit vector characterizing the enveloping cavity (Fig. 1) that has the same shape. Those vectors coincide ($\boldsymbol{n} = \boldsymbol{v}$) in equilibrium, but turn from one another under particle rotation. Evidently, the resistance torque grows with the deviation angle ω until the particle attains position $\boldsymbol{n} \perp \boldsymbol{v}$, i.e., $\omega = 90^\circ$. As soon as this orientation is traversed, the torque changes its sign: in order to restore its shape, the elastic cavity now generates the stress that urges the particle towards position $\boldsymbol{n} = -\boldsymbol{v}$, i.e., to $\omega = 180^\circ$. Whatever the exact form of angular dependence of the torque, one concludes that this function is even with respect to the direction $\omega = 90^\circ$, i.e., orthogonal configuration $\boldsymbol{n} \perp \boldsymbol{v}$.

This implies that the corresponding term to be added to the particle energy is of the barrier type. With regard to that, we approximate the in-cavity energy barrier term as $-QV(\boldsymbol{nv})^2 = -\boldsymbol{QV}(\boldsymbol{nv})^2 = -\cos^2\omega$, where Q is the effective elastic modulus and V the particle volume.

To account for a general case, the Hook-like term should be retained to the particle energy expression as well. The basis for that is at least twofold. First, the point-to-point adhesion at the particle/matrix interface is not total zero, so that, when rotating, the particle has to pull and unwind some macromolecules attached to its surface. Second, even if the barrier-type energy prevails, a real particle hardly has perfect inversion-symmetrical shape; then, upon turning it by 180°, its congruence with the cavity would be incomplete. For such a particle the energy wells (minima) positioned at $\boldsymbol{n} = \pm\boldsymbol{v}$ are not equally deep: the one at $\boldsymbol{n} = \boldsymbol{v}$ is preferable. Therefore, to allow for possible coupling of the particle with its initial position, we, as in Ref. [19], add to the energy expression a quasi-Hookean term $GV\arccos^2(\boldsymbol{nv})$.

Let the magnetic moment $\boldsymbol{m}$ of the considered MH particle at the initial state be directed along $\boldsymbol{n}$. Then the above-described in-cavity 180°-rotation causes mechanical inversion of the particle magnetic moment without changing its position inside the particle: $\boldsymbol{m}$ rotates together with $\boldsymbol{n}$. Meanwhile, the SW process renders a different (non-mechanical) type of re-orientation of $\boldsymbol{m}$ that does not require any mobility of the particle axis $\boldsymbol{n}$. This means that in reality the vectors $\boldsymbol{m}$ and $\boldsymbol{n}$ are independent orientational variables.

With allowance for the above-given explanations we present the orientation-dependent energy of an MH particle embedded in an elastic matrix and subjected to a quasistatic external field $\boldsymbol{H} = \boldsymbol{Hh}$ in the form

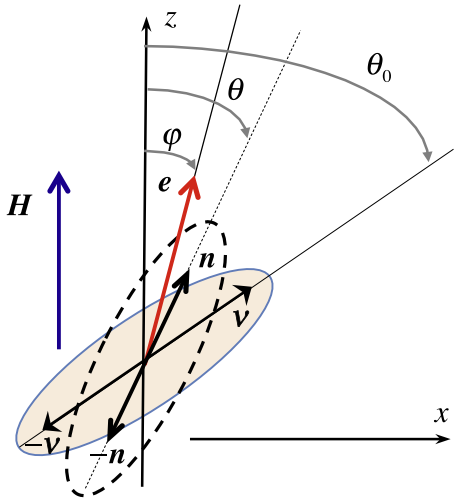

Fig. 1. MH particle rotation in an elastic matrix.