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Research articles

Negative coercivity of magnetic elastomers filled with magnetically hard particles

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ABSTRACT

This research is dedicated to the specifics of the behavior exhibited by elastomer filled with magnetically hard (high-coercivity) particles influenced by magnetic fields. Composite materials of this type are also mentioned as magnetorheological, magnetoactive, and hybrid. There have been done studies on the magnetization processes taking place in ME's containing magnetically hard spherical NdFeB-alloy particles. As was found out, the coercivity exhibited by a sample can be significantly weaker in comparison to that demonstrated by the filler in pure form. It was also discovered that the coercivity may have negative values in positive magnetic fields. These effects are determined by the magnitude relation of the remnant magnetization of the sample, elasticity of the polymer matrix, strength of the external magnetic field, and the capability of magnetic particles to turn inside the polymer matrix as the magnetic field changes polarity to opposite.

1. Introduction

Investigations described in this article were inspired by the studies launched several years ago on magnetic elastomers (ME) simultaneously containing both magnetically soft and magnetically hard particles. Noticed to exhibit specific magnetic and rheological properties [1–6], such materials have been designated as 'hybrid', which emphasizes the differences they demonstrate in comparison to the classic magnetorheological elastomer.

For example, introduction of magnetically hard particles into the composition results in a higher loss tangent shown by the material after being magnetized [4]. It may be considered a promising feature for its applicability in primitive damping devices not supplied with permanent or electro-magnets. In particular, hybrid magnetic elastomers are offered for use in damping devices and acceleration sensors [7–10].

Investigation of the rheological properties of hybrid magnetic elastomer (HME) unveiled the asymmetric behavior of its elasticity modulus measured as a function of magnetic field direction [1,5,6]. Studies of the magnetic properties of HME's carried out at previous stages demonstrated that they exhibited coercivities significantly weaker than that of the pure filler used for their fabrication. In order to give these materials a more profound investigation, there were prepared specimens filled with magnetically hard particles only. Such materials demonstrate low coercivities, which was explained by rotation of the particles inside the polymer after subjecting the samples to magnetizing [11–13]. A number of publications have been dedicated to the mathematical description and simulation of this rotation phenomenon taking place during hysteresis measurements [14,15]. The issue of the mechanical properties of these composites was considered in recent studies [16,17].

Whereas in previous investigations there was used a low-coercivity YMM-Q-grade NdFeB-alloy powder characterized by a value of 2900 Oe [11–13], in the present study a powder with a coercivity of 9000 Oe was introduced into the specimens. In addition, priority was given to the filler with spherical-type particles. Owing to the fact that in previous studies flake-like high-coercivity particles were determined to be the cause of quick degradation of matrices subjected to deformations, this choice seems quite logical making materials filled with spherical particles most perspective for possible applications.

2. Samples and methods

Specimens of hybrid magnetic elastomer (HME) were fabricated using the two-component silicone elastomer compound SIEL-254, a product of State Scientific Research Institute for Chemical Technologies of Organoelement Compounds. Its two parts, a vinyl- and a hydride-containing agent, are initially liquid and polymerize into a 3-D cross-linked net when mixed together and treated thermally. For the purpose

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Table 1
Sample compositions and elastic modulus magnitudes

Sample #	Concentration of filler (wt.%)	Concentration of oil M100 in liquid matrix-to-be (wt.%)	G′ (kPa)
1	75	65	50
2	75	40	100
3	75	25	400
4	75	0	1000

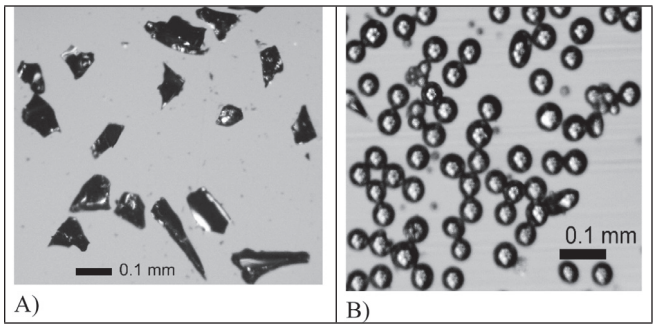

Fig. 1. Morphologies of NdFeB-alloy particles: A) YMM-Q-grade with planar geometrically irregular shapes, B) MQP-S-11-9-grade with spherical shape and an average diameter 50 μm.

of fabrication samples with desired rigidity, we also added silicone oil M100 at concentrations shown in Table 1. At the same time, the oil served as a modifying agent used to make the filler hydrophobic and prevent aggregation of its particles. As has been mentioned above, the filler taken for preliminary experiments (Sample 0) was a YMM-Q-grade NdFeB-alloy powder with particles having irregular geometries and exhibiting Hc = 2900 Oe, whereas the rest of specimens were fabricated on the basis of an MQP-S-11–9-grade powder with spherical particles and Hc_1 (descending) = Hc_2 (ascending) = 9000 Oe (Fig. 1).

According to the detailed description provided in [4], the fillers were added to make a concentration of ~77 wt% and thoroughly mixed with the liquid matrix-to-be. The degassing procedure was followed by distributing the resulting suspension among moulds and curing at a temperature of ~110 °C. Polymerized and removed, cylindrical samples with a diameter and thickness of 14 and 2 mm, respectively, were tested for rheological properties. Their shear modulus was measured on an Anton Paar Physica MCR301 rheometer with amplitude and frequency set to be 0.01% and 1 Hz, respectively, as a result of which the magnitudes listed in Table 1 were obtained.

In order to determine and characterize the specific features of processes taking place inside the hybrid elastomer, a series of minor magnetization curves were recorded for samples with different rigidities. The measurements were performed on cylindrical-shaped specimens having 2 mm in thickness and diameter using a Lake Shore 7407 vibrating-sample magnetometer, USA. Based on recording minor magnetization curves resulting from external magnetic field oscillations predetermined to be symmetric with respect to zero-field and at a certain amplitude, they were carried out according to the scheme shown in Figs. 2–4. The specimen forced into vibration was exposed to the field growing from zero to a maximum corresponding to point 1, also referred to as the initial magnetizing field of a hysteresis loop. After that, the field decreased to zero to begin to grow in the opposite direction and stop at point 2 corresponding to the magnitude of the initial magnetizing field with the opposite sign. The magnetization of the sample thus subjected to a polarity reversal (re-magnetizing procedure) followed the field change along the descending branch of the hysteresis loop. On the way back, the field decreased to zero again and grew up to the initial maximum, thus making a complete cycle at point 1. Suffering another re-magnetizing, the sample changed its condition along the

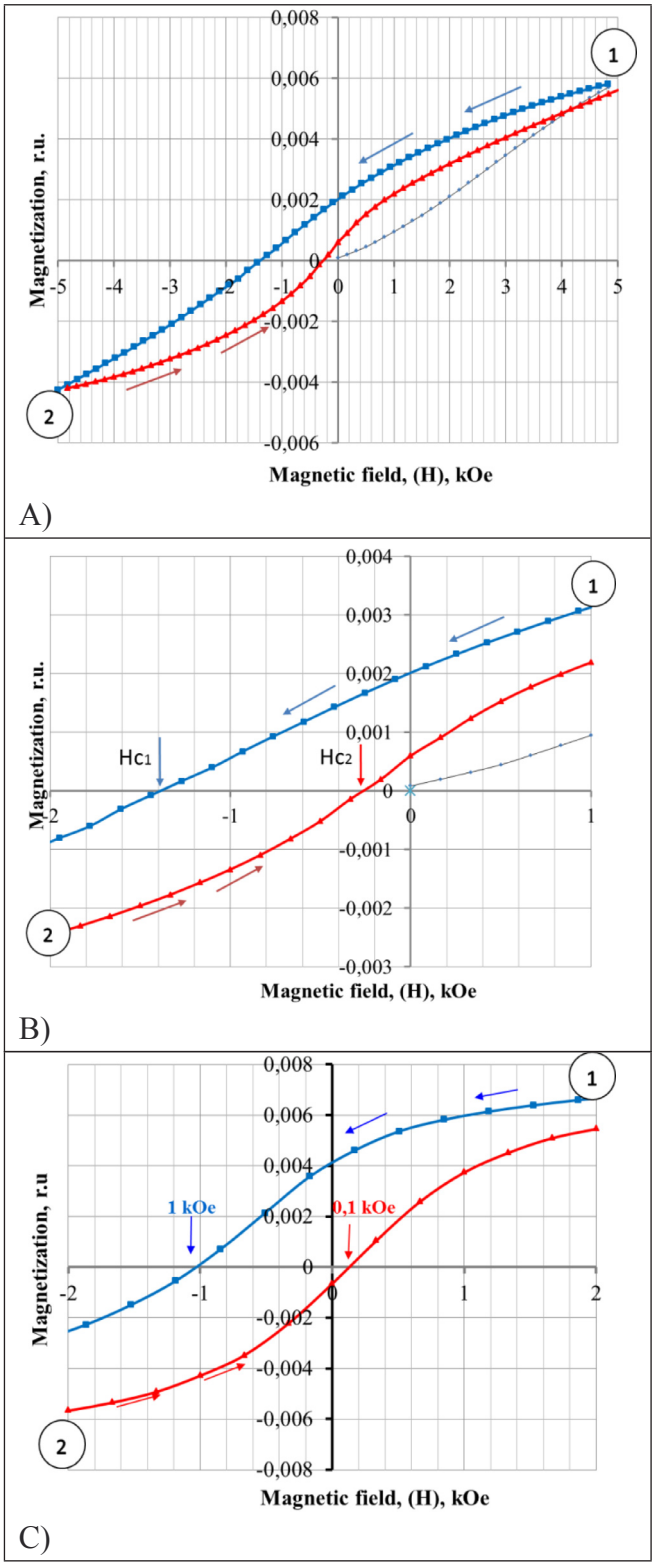

Fig. 2. Magnetization hysteresis loops recorded for Sample 0 initially magnetized at A) 5 kOe, B) 5 kOe, scaled-up version of A, and C) 10 kOe.

ascending curve. Finally, the external magnetic field decreased from point 1 to zero to start another cycle at a higher amplitude. As a result, a series of minor hysteresis loops with amplitudes successively increasing in the sequence 3, 6, 9, 12, 15 and 20 kOe were recorded for every specimen. The field change step was 100 with an interval of 2 s in all cycles. Magnetizing of the samples was carried out stepwise.

3.7. Description of the negative coercivity phenomenon

The results obtained indicate that negative coercivity emerges when the ascending curve of the hysteresis loop is measured. The specifics of this phenomenon are determined by the direction of the initial magnetizing, which is also responsible for the asymmetry of the loop. This relationship appears owing to the elastic forces occurring inside the polymer matrix, which tend to return the magnetized particle into the position, in which it was before the initial magnetizing. The set of forces influencing the magnetized particles inside the polymer placed in a magnetic field are schematically shown in Fig. 13.

Influenced by a magnetic field (1), the particle gets magnetized along the axis of easy magnetization (3) resulting in an overall magnetization vector (2) co-linear with the external field. This magnetization vector is responsible for the force causing rotation of the particle to line its moment up with the external field. When the magnetic moment corresponding to this vector exceeds the influence of elastic forces, the particle starts turning after the external magnetic field until the driving factor is balanced by the growing resistance (4) of the polymer tending to return the particle to the initial position. The elastic force of the matrix is proportional to the elasticity modulus and degree of rotational deviation from equilibrium. Fig. 14 shows the possible stages of magnetization and rotation of the particle inside an ME sample during recording of a complete hysteresis loop.

Fig. 14 presents a descriptive model of processes occurring inside the magnetic elastomer. The filler particles are distributed randomly and may be both plate-like and spherical as the shape factor does not make critical difference. The particles illustrated on the drawing are elongated for a better presentation and understanding of processes taking place inside the polymer during its magnetizing and re-magnetizing. An average particle selected initially (point 0) is positioned at an angle of 45^0 with respect to the direction of the external field. Influenced by the field, it becomes magnetized and starts participating in the interactions shown in Fig. 14. Leaving the initial position it turns after the magnetic field (points 1, 2, 3), after which the external field decreases to zero making it possible for the particle to return to the initial position (points 4, 5). (At this point, we purposely simplify the overall picture by not taking into account interactions among magnetically hard magnetized particles.) At point 5, the external magnetic field is zero, but the particle possesses a remnant magnetization directed along the initial magnetic field.

As the reverse field becomes stronger, the particle (point 6) either gets re-magnetized changing own polarity if the polymer matrix is rigid, or turns after the field if the matrix is soft. In stronger reverse fields the particle tends to turn more intensively overcoming the elastic forces of the matrix. At the same time, owing to the fact that the real system contains a big number of particles, they may behave in different ways partly rotating or partly re-magnetizing, which might be determined by their concrete positions and coercivities, matrix elasticity, and magnetic field intensity.

As the reverse magnetic field decreases, the particle, influenced by the elastic forces of the polymer matrix, returns to the initial state, as a result of which its magnetic moment once becomes co-linear with the direction of initial magnetizing (points 9, 10, 11). At point 11, even before the moment when the reverse magnetic field has diminished to zero, the particle practically completely turns back and creates a positive magnetic moment in a negative field.

Presenting an understanding of processes taking place inside magnetic elastomer, for simplicity the proposed model takes into consideration no structuring effects among magnetized particles.

On the strength of all the forces influencing the magnetized particle, its magnetic moment may have positive values in a range of negative magnetic fields.

4. Conclusions

1. Particles inside the polymer matrix are capable of rotating under the influence of external magnetic field. At certain magnitudes of magnetic field, remnant magnetization, and elasticity of the polymer matrix the magnetic susceptibility curve may reveal both re-magnetizing and rotation of particles.
2. The hysteresis loop is asymmetric, which is determined by the direction of the initial magnetizing of the ME sample.
3. At a certain relation between the magnitudes of the remnant magnetization of the filler, the initial magnetic field, and elasticity of the polymer matrix the conditional coercivity exhibited by the sample on the ascending branch of the hysteresis loop may be negative, which means that the magnetization of the material has a positive value at a negative magnetic field. This effect is determined by the rotation of the magnetized particles caused by the elastic forces occurring inside the polymer and tending to return the particles into their initial states.
4. The negative coercivity effect reveals itself in the magnetic elastomer filled with both spherical and flake-like particles.
5. In the case of elastic matrices, the coercivity exhibited by ME samples magnetized in strong magnetic fields is determined by the elasticity of the polymer, being proportional to it.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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