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Elastomer with magneto- and electrorheological properties

Dmitry Yu Borin¹ and Gennady V Stepanov²

Abstract
In this study we introduce an elastic composite, which has been manufactured using a fine carbonyl iron powder coated with a polymeric dielectric shell and dispersed in a silicone elastomer in a way as it is typically done manufacturing magnetorheological elastomers. Due to the used filler such a material possesses the capability of exhibiting magneto- and electrorheological effects. Our experiments have shown that the application of the magnetic field to the composite results in the magnetorheological effect, which becomes stronger in the case of the additional application of an electric field.

Keywords
Magnetorheology, electrorheology, elastomer

Introduction
Most known composites among field-controlled materials are electrorheological (ER) and magnetorheological (MR) fluids and elastomers (Bossis et al., 2002; Jolly et al., 1996; Parthasarathy and Klingenberg, 1996). These are composites of dielectric particles (ER materials) or magnetic particles (MR materials) dispersed in a carrier liquid or an elastic medium. Application of an external electric or magnetic field leads to the particles’ aggregation in structures, which are elongated in the direction of the applied field. The aggregation changes properties of the material. This is widely used for technical applications (Carlson and Jolly, 2000; Kordonsky, 1993; Li and Zhang, 2008). There are different ways to enhance the field response of ER and MR composites.

Among them are targeted selection and modification of the particles and carrier medium, using complex (e.g. bidisperse) compositions, and so on (Akhavan, 2007; Borin and Stepanov, 2013; Böse, 2007; Carlson, 2002). In order to develop an advanced intelligent composite whose properties are actively and reversibly controllable not with just one stimulus, fluids based on dispersed phase that are simultaneously sensitive to electric and magnetic fields (magnetoelectrorheological (MER) fluids) have been introduced in the past (Kordonsky et al., 1994; Korobko et al., 2012).

However, composites based on a liquid matrix have disadvantages such as the gravitational instability of the powder particles and the necessity of using some vessel to contain the fluid, which stimulates further developments. In this article we present for the first time a MER composite based on an elastic matrix. Hereinafter this material will be referred to as ‘MER elastomer’.

The principal aim of the study was to evaluate a synergistic viscoelastic response of a MER elastomer under joint action of the electric and magnetic fields. The article is organized as follows. Firstly, material preparation and used experimental methods are briefly described. In the experimental section separated effects of an applied electric and magnetic field as well as a joint influence of both fields on the viscoelasticity of the MER elastomer are presented. The final section presents conclusions and outlook.

Materials and methods
As a matrix material for MER samples a liquid silicon oligomer (SIEL™ produced by GNICHTEOS) was used. The standard compound SIEL™ is based on a mix of low molecular vinyl-containing rubber and a

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As a filler we used particles of the fine carbonyl iron powder with an average particle size of $d = 4 \, \mu m$. These powder particles were coated with a polymeric dielectric shell. For the coating a deposition method using a water solution of the polyacrylonitrile resin (PAN) and phosphoric acid was utilized. Figure 1 depicts the scheme of a coated particle. The powder particles have a coating with an estimated thickness of 100 nm and a high specific resistance which exceeds 1 \( \Omega \cdot cm \).

MER samples were manufactured dispersing the powder in a silicone elastomer in a way as is usually done manufacturing MR elastomers (Borin and Stepanov, 2013; Böse, 2007; Nikitin et al., 2004; Stepanov et al., 2008). The two components of the matrix, low-molecular vinyl and a hydride-containing ingredient, were first mixed with each other, then with the magnetic filler in a weight concentration of $\sim 70\%$, and were subjected to polymerization at $T = 130 \, ^{\circ}C$.

To evaluate a viscoelastic response of the MER elastomer we used oscillation rheometry. This is a technique commonly utilized to estimate rheological properties of elastomers by measuring their storage ($G'$) and loss modulus ($G''$) (Borin and Stepanov, 2013; Borin et al., 2012; Böse, 2007; Li et al., 2010). For MR and ER materials rotational rheometers occupied with commercially available magnetic or electric cells are used in such a test. However, commercial equipment providing electric and magnetic fields simultaneously does not exist. Therefore, we have combined a rotational rheometer (Anton Paar Physica MCR301) with a custom-made electromagnetic coil and electroductive measuring geometry (plate–plate) using a sliding contact to the rotating upper plate. The used solenoid ensures a homogeneous magnetic field in the area of the measuring gap. We have used a field with a maximal flux density up to 200 mT. It is possible to reach higher fields with the solenoid; however, the field strength was limited in order to avoid an overheating of the coil caused by a high electrical current. The maximum electric field strength used was limited to 2.5 kV/mm due to electric breakdown in the measuring cell.

Response of the composite to an external stimulus was evaluated in several steps. First of all we have analyzed the sample behavior under the separate action of the applied magnetic and electric fields quantifying in this way MR and ER responses correspondingly. Afterwards the sample was subjected to both fields simultaneously and a MER effect was measured. In measurements either the strain at the fixed frequency (amplitude sweep) or the frequency of oscillation at the fixed strain was varied. The wall-sleep problem was taken into account choosing an appropriate surface roughness of the measuring geometries. The normal force during the measurements was kept constant.

### Experimental results

#### MR response

Figure 2 shows the dependence of the storage and loss moduli ($G'$ and $G''$) on the applied strain under the action of an applied magnetic field with various flux densities $B$. The observed behavior with a significant increase of the moduli in the magnetic field is typical for the soft MR elastomers (Böse, 2007; Nikitin et al., 2004; Stepanov et al., 2008). For the small deformations under applied magnetic field with a strength of 200 mT the observed increase of $G'$ and $G''$ was $\sim 480\%$ and $520\%$ respectively. The decrease of the moduli with increased deformation (strain) is referred to as the Payne effect, known from the behavior of filled rubbers, and is correlated with the bond strength between the particles of the filler and the elastic matrix. The frequency dependence of the moduli is shown in Figure 3. An increase of the moduli in the field can be observed. With increasing frequency the increase in the storage modulus by the magnetic field remains fairly constant, while the increase in the loss modulus shows a decreasing trend. This behavior of the material can be attributed to the apparent increase of the cross-linking due to magnetic interactions between the particles.

#### ER response

An application of the electric field leads to more or less similar effects to those measured within MR observations. Results regarding ER effect have shown that the storage and loss moduli $G'$ and $G''$ increase with the increase of an applied electric field magnitude. Under the field action these moduli are increased by about 370\% at the small strain ($< 0.1 \%$). Similar to the MR effect, with increasing amplitude the increase in storage and loss moduli by the electric field becomes smaller (Figure 4). Results of the frequency sweep are presented in Figure 5. An increase of the storage modulus in the field with increasing frequency remains fairly constant, while an increase of the loss modulus becomes smaller.

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Figure 2. Results of the amplitude sweep performed on a MER elastomer obtained at an oscillation frequency of 1 Hz: storage (left) and loss (right) modulus under the action of an applied magnetic field with various flux densities $B$.

Figure 3. Results of the frequency sweep performed on a MER elastomer obtained at a strain of 0.01%: storage (left) and loss (right) modulus under the action of an applied magnetic field with various flux densities $B$.

Figure 4. Results of the amplitude sweep performed on a MER elastomer obtained at an oscillation frequency of 1 Hz: storage (left) and loss (right) modulus under the action of an applied electric field with strength $E$. 
This behavior is as well reasoned with a field influenced interaction between the particles of the filler and the matrix.

**MER response**

A simultaneous application of the magnetic and electric fields results in the further growth of the storage and loss moduli. Results of the amplitude and frequency sweep are given in Figures 6 and 7 respectively. Curves corresponding to the MR effect at 200 mT are given as references. When both fields are applied, moduli exhibit additional increments. Due to the action of both fields, interparticular forces as well as particles–matrix interactions are changed. This is especially reflected in the dependence of the loss modulus on the oscillating frequency. Apparently, the modulus $G''$ became frequency-independent in the applied magnetic and electric fields.

For the better representation of the quantitative fields influences on the viscoelastic response of the composite, relative changes of the storage and loss moduli, as well as a relative change of the loss factor $\tan \delta$, are presented in Figure 8. The loss factor is the ratio between the loss and storage moduli: $\tan \delta = G''/G'$. This parameter is usually used together with $G''$ in order to describe the efficiency of damping caused by the material.

Results presented in Figure 8 clearly demonstrate an enhancing of the MR effect when an electric field is applied (MER effect) in addition to a magnetic field. For the small deformations ($\sim 0.01\%$) and the oscillating frequency of 1 Hz under an applied magnetic field with a flux density of 200 mT the observed increases of $G'$, $G''$, and $\tan \delta$ are $\sim 480\%$, $\sim 520\%$, and $\sim 20\%$ respectively.

When an electric field is additionally applied these moduli exhibited further increments of $\sim 770\%$, $\sim 2110\%$, and $\sim 60\%$ respectively. An observed total effect of
both fields on the moduli is \(-1250, \approx 2630\), and \(\approx 80\%\) respectively. As seen, a simple superposition rule does not work here and a need for adequate and scientifically correct approaches to predict the MER effect is obvious.

A remarkable point, which has not been addressed above, is a dependence of the loss factor on the oscillating frequency. The values of tan\( \delta \) are generally higher at the low frequencies and they decrease with increasing frequency, which results in decreasing of the relative MR and MER effects. This indicates that the loss modulus in the applied field is not significantly influenced by the increasing frequency in the used frequency range as can be seen in Figures 3, 5 and 7.

**Conclusions and outlook**

We have introduced the MER elastomer—an elastic composite with combined MR and ER response. It has been shown that the composite based on a silicon polymeric matrix with embedded magnetic particles coated with a polymeric dielectric shell is highly sensitive to both magnetic and electric fields. Using an oscillatory rheometry it has been found that an application of external fields gives a higher field-strengthening effect than the sum of individual MR and ER effects at the given values of the magnetic and electric fields. In particular the MR effect reaches up to \(\approx 500\%\) \((B = 200 \text{ mT})\) and ER effect reaches up to \(\approx 370\%\)

**Figure 7.** Results of the frequency sweep performed on a MER elastomer obtained at a strain of 0.01%: storage (left) and loss (right) modulus under the action of an applied magnetic field with flux densities \(B\) and electric field with strength \(E\).

**Figure 8.** Relative MER and MR effects in the composite obtained within amplitude sweep at the fixed oscillation frequency of 1 Hz (left) and frequency sweep at the fixed strain of 0.01%. 1: relative change of the storage modulus \(G'\), 2: relative change of the loss modulus \(G''\) and 3: relative change of the loss factor tan\( \delta = G''/G'\) under the action of the applied magnetic field with flux densities \(B = 200\) mT and electric field with strength \(E = 2.5\) kV/mm.
\( E = 2.5 \text{ kV/mm} \), while the observed total MER effect exceeds 1000\% \( (B = 200 \text{ mT}, E = 2.5 \text{ kV/mm}) \).

Obviously, similar to other field-responsive composites, not only the rheological properties of the MER elastomer are changed under external actions. We emphasize that all features typical for conventional soft MR elastomers are expected to be observed for the MER elastomer as well: deformational effect, shape memory effect, field-dependent conductivity and resistivity (Böse, 2007; Nikitin et al., 2004; Stepanov et al., 2008). However, these are topics for future studies.

Furthermore, a theoretical model describing and predicting a response of the composite to the joint action of two independently and simultaneously applied external fields has to be proposed for the correct understanding and interpretation of the MER effect.

**Declaration of conflicting interests**
The authors declared no potential conflicts of interest with respect to the research, authorship, and/or publication of this article.

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