Two-particle element of a magnetorheological elastomer under a cyclic magnetic field

A M Biller, O V Stolbov, Yu L Raikher

Institute of Continuous Media Mechanics – the Division of Perm Federal Research Center, Russian Academy of Sciences, Ural Branch, Perm, 614013, Russia

E-mail: kam@icmm.ru

Abstract. The results of modelling the behaviour of a pair of magnetisable spherical particles embedded in a viscoelastic elastomer with Kelvin rheology are presented for the case, where an ac magnetic field is exerted along the intercenter line of the particles. This system is considered as a small-scale (mesoscopic) structure element of a magnetorheological elastomer. The system in question is known to be able for hysteretic behaviour under the quasistatic cycle of the field. The particles initially positioned well apart, at some finite field strength, fall onto one another (cluster) and dwell in this state until the field decreases well below the value at which the cluster has been formed. Under dynamic cycling of the field, the viscous friction interferes the particle displacement process and impedes occurrence of the magnetodeformational hysteresis of the element. Starting from small-amplitude overdamped oscillations and enhancing the role of magnetic forces over viscous ones, we show how the anharmonicity of the system grows while it approaches the transition threshold, above which the dynamics of the element includes the cluster state. The dynamic hysteresis of magnetisation that accompanies the mechanical oscillations of the particles is presented as well.

1. Introduction

Among the many newly developed materials, the so-called “smart” ones attract special interest because of their properties which can be controlled and adjusted to variable external conditions. Such materials include magnetorheological elastomers (MRE), the composites consisting of an elastomer matrix filled with micron-sized magnetisable particles. A distinctive feature of these materials is their magnetosensitivity: when an external magnetic field is applied, the particles inside the composite magnetise and interact with each other changing their spatial distribution. This causes modification of a number of the MRE macroscopic properties: elasticity and viscosity, magnetic and electric permeability, conductivity, etc. Usage of MRE as essential working components in adaptive dampers, actuators, valves, medical appliances, etc. is an actively developing applicational trend. In such devices, the main exploited effect is the change of rheological characteristics and shape of MRE elements in response to the applied magnetic field or mechanical load in the real-time regime. Therefore, the dependence of the dynamic characteristics of MREs on the applied field and load is an important issue; this is demonstrated by a rather large number of published experimental works on this subject [1, 2, 3, 4, 5, 6].
2. Two-particle element

2.1. Quasistatic situation

To understand the mechanisms of inner interactions in MREs, which govern rearrangement of their internal structure, one should study them at the mesoscopic level, where the matrix and particles are considered separately and each one is treated as a continuous medium with its own set of properties to which scope the hypotheses on the interaction between the phases are added. In line with this viewpoint, the present paper considers a set of two magnetic particles embedded in an elastic matrix as a basic element of an MRE capable of illustrating the magnetomechanical response of the material. This model element consists of a cylindrical sample whose outer dimensions are chosen under the requirement that a pair of spherical particles occupies about 30% of its geometric volume. The particles have identical radii $a$, and are positioned at the cylinder axis in such a way that their intercenter vector $l$ lies along this axis. The latter is taken as the $Oz$ axis of the cylindrical coordinate frame whose origin is placed at the centre of the sample.

![Figure 1. Model two-particle element of an MRE.](image)

As well known, the most strong shape and magnetic effects are displayed by MREs based on soft polymer matrices with Young moduli $E \sim 5 - 30$ kPa, see [4], for example. The internal viscosity of those compounds estimated from their relaxation times which are of the order of a few seconds [7]. This time scale is inherent to MREs filled with microparticles (2–5 µm) of carbonyl iron that has the initial magnetic susceptibility is of the order of $10^4$ and saturation magnetisation of 1500 kA/m. In view of what follows, we note that under not too strong fields and strains, the nonlinearity of both the particle magnetisation and the matrix elasticity might be neglected, at least in the first approximation.

When the element sketched in Fig. 1 is subjected to an external uniform magnetic field $H_0 = (0, 0, H_0)$, the magnetic polarisation of the particles induces their mutual attraction striving to move them closer. However, as soon as the particles yield to this tendency, the deformed matrix generates restoring elastic forces, so that a new equilibrium state of the pair is defined by the balance of those opposing effects.

The magnetomechanical response of the above-described two-particle element under quasistatic conditions was studied in detail in Refs. [8, 9]. In that modelling, nonuniform deformation of a nonlinearly elastic elastomer matrix and nonuniform and nonlinear magnetisation of the particles were taken into account as well. It turned out that within a certain domain of material parameters of the particles and matrix, the system displays bistable behaviour. Namely, when the MRE element is subjected to a quasistatic cycle of the magnetic field, the magnetomechanical hysteresis occurs: the path that the particles pass under the increase of the field does not coincide with that undergone during the field decrease. The main parameter governing the effect is $\beta = \mu_0 M_s^2 / c_1$, where $\mu_0$ is the magnetic constant, $M_s$ saturation
magnetisation of the ferromagnetic material, and $c_1$ elastic constant of the Mooney-Rivlin model assumed for the polymer.

Further on, to characterise the configuration of the two particle element, we use nondimensional units, scaling all the distances with the particle radius. Therefore, the current intercenter distance of the pair and the relative change of the interparticle gap (the closest distance between the surfaces of the particles) are defined as

$$q = l/a, \quad \text{and} \quad \epsilon = (q - 2)/(q_0 - 2),$$

respectively; here $q_0$ is the initial interparticle distance and $q = 2$ its limiting value attained when the particles come into tight contact. As seen, the gap parameter $\epsilon$ varies from unity in the initial state of the element to zero when the particles collapse in a pair cluster.

Figure 2. Quasistatic magnetomechanical hysteresis loops for a pair of particles with intercenter distance $q_0 = 3$; solid lines correspond to the case of particles made of a linearly magnetisable ferromagnet, dashed lines are for the case of a ferromagnet that magnetises according to the Frölich-Kennelly law [10] with the parameters given in the text. Note virtual coincidence of the curves of both types, the difference is visible only in the vertical lines denoting the jumps to and from the cluster state.

In Fig. 2 an example of quasistatic bistable behaviour is presented for the element with initial intercenter distance $q_0 = l_0/a = 3$ and magnetomechanical parameter $\beta = 2500$, that corresponds to a MRE with $M_s = 1500 \text{kA/m}$ and Young modulus of the matrix $8 \text{kPa}$. Here and below we express the external field strength in nondimensional units as $h_0 = H_0/M_s$.

The adopted set of material parameters implies that the polymer is rather soft so that the collapsing field is low: $h_0 \approx 0.04$ that corresponds to $H_0 \approx 60 \text{kA/m}$ (or 750 Oe) that is very far from the saturation level. That is why the magnetomechanical hysteresis loops of the linearly and nonlinearly magnetising particles do not differ much. Further on, being after qualitatively understanding of the dynamic processes, we restrict the consideration by linear magnetisation approximation using it for evaluating the interparticle magnetic interaction via the linear multipole solution model. This treatment is not at all unrealistic since iron carbonyl microparticles magnetise virtually linearly in the fields up to few kOe. Although a linearly magnetising substance does not have saturation, in below we keep the adopted value of $M_s$ as the unit of the field scale; this should be recalled when transforming the results back to the dimensional form.

2.2. Time-dependent formulation

In the case of a time-dependent field, the motion of the particles, besides the magnetic and elastic forces, which are potential, is affected by viscous friction which has two evident sources. One stems from the interaction of eddy currents induced inside the metal grains by the applied field.
However, in the low-frequency (up to few kHz) range that we aim at, this effect is negligible. The dissipation mechanism that is relevant in the considered situation results from the particle movement relative to the matrix. Provided the motion is not too fast, the arising viscous force acting on a particle is Stokes-like, i.e., proportional to its velocity.

We begin with the case of small particle displacements, i.e., small matrix strains. In that case, one may assume that the particles move only along the upper branch of the magnetomechanical hysteresis loop, see Fig. 2. In linear-strain approximation, the stress $\sigma$ arising in the matrix can be represented as a sum of elastic and viscous parts as

$$\sigma = \sigma_{el} + \sigma_{visc} = \lambda I_1(\varepsilon) I + 2G\varepsilon + \eta \dot{\varepsilon}. \quad (2)$$

Here $\varepsilon$ is the strain tensor, $\lambda$ is the Lamé’s first parameter, shear modulus $G$ and viscosity $\eta$ of the material. The adopted representation corresponds to the Kelvin-Voigt model of a viscoelastic medium, where the elastic and viscous elements are connected in parallel.

Under linear magnetisation law, the interparticle magnetic force may be described with the aid of analytical expressions derived in Ref. [8]. In the here adopted units the result of [8] for the magnetic attraction force per particle writes as

$$F_m = \mu_0 H_0^2 a^2 \tilde{F}_m(q), \quad (3)$$

where nondimensional function $\tilde{F}_m$ in a complicated way depends on the interparticle distance $q$ but is independent of the applied field strength. Introducing volume density $f_m$ of this force in such a way that it is nonzero only inside the particles and is always directed along the local magnetic field, one arrives at the time-dependent force balance equation

$$\nabla \cdot \sigma + f_m = 0. \quad (4)$$

Here the inertia terms are omitted as being negligible, see Section 6 for estimates.

Finally, we introduce the dimensionless time scale $\tilde{t} = t/\tau_R$ basing it on the stress relaxation time $\tau_R = \eta/G$ of the Kelvin-Voigt model. Transformation of the time derivatives $\dot{\varepsilon} = \tau \ddot{\varepsilon}$ yields the stress tensor in the form

$$\sigma = G\tilde{\sigma} = G \left[ \frac{\lambda}{G} I_1(\varepsilon) I + 2\varepsilon + \dot{\varepsilon} \right], \quad (5)$$

so that the equation of motion (force balance) writes as

$$\tilde{\nabla} \cdot \tilde{\sigma} + \kappa h_0^2 \tilde{f}_m = 0, \quad (6)$$

where $\tilde{\nabla} = a\nabla$ and the coefficient alongside the magnetic force is

$$\kappa = \mu_0 M_s^2 a^3 / G. \quad (7)$$

The magnetoelastic problem under study is completed by adding to it the kinematic relations

$$\varepsilon = \frac{1}{2} \left[ (\nabla \mathbf{u})^T + (\nabla \mathbf{u}) \right] \quad (8)$$

(where the superscript denotes matrix transposition) and stress-free conditions for all boundaries.
3. Oscillations of the interparticle distance

Let an ac uniform external magnetic field

$$h_0 = h_A \sin(\omega t) = h_A \sin(\tilde{\omega} \tilde{t}), \quad \tilde{\omega} = \omega \tau_R$$

(9)

of amplitude $h_A$ be applied along the axis of the MRE element; here $\omega$ is dimensional frequency. For the steady field oscillations (9), the magnetomechanical problem (5)–(8) was solved numerically in axisymmetric formulation by the finite element method with the aid of library fenics for the python language.

As the magnetic particles are fully polarisable and do not possess any residual magnetisation, the magnetic force between them does not depend on the direction of the applied field and, thus, would oscillate with doubled frequency. Accordingly, the same would be the time dependence of the forced changes of the interparticle gap $\epsilon$.

3.1. Low-field amplitudes

Let us compare solution of the dynamic problem with the results of quasistatics. For that, we consider the field-induced oscillatory solutions of the set (5)–(8) and vary the frequency. The corresponding plots of the interparticle distance $\epsilon(h_0)$ are presented in Fig. 3.

![Figure 3](image)

Figure 3. Interparticle distance as a function of the applied field strength in the quasistatic case (solid line), and in steady oscillatory regimes induced by ac field of amplitude $h_A = 0.035$ and frequencies $\tilde{\omega} = 0.03$ (dashed), 0.15 (dash-dotted), 0.75 (dotted), and 3.75 (long dashed).

For comparison, in Fig. 3 the quasistatic loop same as in Fig. 2 is presented having been expanded with allowance for the fact that the field that may assume the values of both signs, as it happens for the ac field. However, the dynamic cycles in Fig. 3 are calculated for the field whose amplitude does not exceed the quasistatic threshold value, and, thus, no jumps of the interparticle distance occur. This means that the shown loops are entirely due to the dynamic hysteresis – the viscosity-induced lag between the field (the cause) and the displacement (the response) – because under this condition the system performs cycles near the upper branch of the quasistatic magnetodeformational curve and never fall in the cluster state. Indeed, at the lowest frequency, the loop is narrow (almost adiabatic) and closely follows the quasistatic one, see the dashed line. With the increase of frequency, the loop becomes wider which effect is maximal when the dispersion condition $\tilde{\omega} \sim 1$ is attained, see the dotted line in Fig. 3. Upon further growth of frequency, the response goes down as the exciting field oscillates too fast to entrain the particles to motion; accordingly, the loops shrink and become more “horizontal”. They also become more “shallow”: the minimal values, which they attain, progressively increase, less and less differing from unity that means that the span of the particle displacements diminishes.
For a given frequency $\omega$, the greater viscosity, i.e., $\tau_R$, the lower parameter $\tilde{\omega}$, the smaller number of cycles the system has to undergo between the initial state and attaining a stable limiting cycle. This is illustrated by Fig. 4.

Note that the specific “baglike” shape of the loop at the lowest frequency. For that curve, the displacement swing is maximal, and for that regime the nonlinear dependence of the magnetic force, see (3), manifests itself. Indeed, the magnetic attraction force even in the simplest (dipole) approximation grows as $1/q^3$ when the particles approach. In the multipole solution, which takes into account mutual polarisation of the particles, this dependence enhances substantially (the power exponent increases) at closer distances, see [8]. That is why the same increment of $h_0^2$ – we remind that the external driving force is proportional to the square of the field – induces larger displacements. Due to that, the right part of the loop in Fig. 4a is wider that its left part. As Figs. 4b and c show, this effect fades out fast as soon as the particles displacement amplitude becomes smaller.

Figure 4. Establishing the steady dynamic magnetodeformational hysteresis cycle of the pair for the field amplitude $h_A = 0.035$ under frequencies $\tilde{\omega} = 0.15$ (a), 0.75(b) and 3.75(b)

3.2. Oscillations in pretransitional region

Evidently, the most interesting case of the problem under study are the oscillations of the particle pair, when the ac field is strong enough to provoke the particle collapse. During some part of the field period, where the field strength is the highest, despite the viscous impediment, the particles would dwell in the cluster state falling on each other at the beginning of this time interval and falling apart at its end. The chances to obtain this regime analytically, due to its strong nonlinearity, are very poor, and numerical modelling seems the only possible way to describe it. Unfortunately, the numerical tools used here are inept for an adequate description of this field-frequency domain: the employed calculation procedure becomes unstable there. Nevertheless, even with the present technique, we are able to get some notion of the occurring transition.

The results of those tests are shown in Fig. 5. Unlike Fig. 4, here the amplitude of the applied field exceeds the quasistatic threshold $h_*$ of the particle collapse, cf. Fig. 2. In this situation, the transitions of a separated pair into cluster and back become possible. However, the fact whether or not such a transition would really occur depends on the frequency of the field. Indeed, in the presence of viscous friction, the transition to the cluster state, i.e., a substantial change of the interparticle distance, needs a finite time $\tau_*$. If at a given frequency in each half-period of the ac field, the condition $h_0(t) > h_*$ holds for a time interval longer than $\tau_*$, then the particle clusterisation should take place. If, to the contrary, this interval is shorter that $\tau_*$, clusterisation is impossible. Although the time parameter $\tau_*$ is not calculable analytically, the numerical plots of Fig. 5 for the certain amplitude imply that $0.5 < \tau_* < 0.6$. This follows from the difference of
the cycle shapes; the one at higher frequency is closed and located at $\epsilon \geq 0.5$, while the one at lower frequency – within the framework of present work – falls down to $\epsilon \ll 1$ and never returns.

In our viewpoint, the above-presented findings imply that the forced oscillations of the pair where cluster formation takes place are an interesting issue, whose solution would be useful for shading light on the internal dynamics of MREs. Because of that, this work deserves further investigation, both analytical and numerical.

![Graphs showing magnetization curves](image)

**Figure 5.** Steady magnetization curves of the model element under ac field of amplitude $h_A = 0.045$ and frequencies $\omega = 1.2$ (a), 0.6 (b), 0.5 (c), and 0.4 (d)

4. **Magnetisation oscillations**

As the MRE element under study is made of the magnetically polarisable particles of a finite size, the changes of interparticle distance modulate their mutual magnetisation. In result, the magnetic moment of the element undergoes cyclic oscillations. This effect could be evaluated from the carried out calculations and enables one to obtain the dynamic hysteresis loops of the model system under study.

In Fig. 6 plots of the magnetisation of the particle are presented for a constant subcritical ($H_0 < h_c$) amplitude of the ac field and different frequencies. Note that, due to the fact that in an MRE containing only magnetically soft particles, the shape of the dynamic magnetisation curves $m(h_0)$ look different in comparison with customary ones. The loops display two symmetrical
Figure 6. Steady magnetization curves of the model element under ac field of amplitude $h_A = 0.035$ and frequencies $\tilde{\omega} = 3.75$ (a), 0.75 (b), 0.15 (c), and 0.03 (d).

“inflation” regions at the ends, which are the result of the viscous friction. As it should be, the area of the loops has the maximum at $\tilde{\omega} = \omega R\tau \sim 1$, where the energy dissipation due to the particle mechanical relaxation is the highest.

5. Conclusion
Modelling of the behaviour of a pair of magnetisable spherical particles embedded in a viscoelastic elastomer with Kelvin rheology is presented for the case, where an ac magnetic field is exerted along the intercenter line of the particles. This system might serve as a small-scale (mesoscopic) structure element of a magnetorheological polymer. A fundamental fact is that the system in question displays hysteretic behaviour under the quasistatic cycle of the field provided the latter is strong enough. The magnetic particles, initially positioned well apart, at some finite field strength fall onto one another (cluster) and dwell in this state until the field decreases well below the value at which the cluster has been formed. Under dynamic cycling of the field, the viscous friction interferes the particle displacement process and, provided the frequency of the field is high enough, impedes occurrence of the magnetodeformational hysteresis of the element. Starting from small-amplitude overdamp ed oscillations and enhancing the role of magnetic forces over viscous ones, we show how the anharmonicity of the system grows while it approaches the transition threshold, above which the dynamics of the element includes the
cluster state. The dynamic hysteresis of magnetisation that accompanies mechanical oscillations of the particles is presented as well.

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6. Appendix:
Let us give a simple estimate for the reference times of the examined system. For this purpose, consider a solid spherical particle that moves slowly (in the Stokes regime) inside a viscoelastic medium. The resistance force acting on a particle of radius \(a\) in such a situation comprises the elastic and viscous parts:

\[
f_{el} = -\gamma x = -6\pi a G x, \quad \text{and} \quad f_{visc} = -\zeta \dot{x} = -6\pi a \eta \dot{x},
\]

where \(G\) is the shear modulus and \(\eta\) is viscosity. Then the equation of motion of the particle of mass \(m\) writes as

\[
m \ddot{x} = -\zeta \dot{x} - \gamma x.
\]

As follows from (11), the time during which the particle velocity assumes its steady value (the inertial time) by the order of magnitude equals the ratio of the particle mass to the coefficient of viscous resistance of the medium, while the stress relaxation time is defined by the ratio of the viscous and elastic coefficients of the medium:

\[
\tau_I \sim m/\zeta, \quad \tau_R = \zeta/\gamma.
\]

A typical stress relaxation time \(\tau_R\) of a soft elastomer (weakly-linked silicone rubber) that makes the base of MRE is of the order of few seconds. With the assumed in above value of the shear modulus, this yields the elastomer viscosity to be in the interval from 1 and 10 kPa·s. Meanwhile, the inertial time \(\tau_I\) for a particle of a few microns in diameter made of iron (specific density \(\rho \sim 8 \times 10^3\) kg/m\(^3\) is

\[
\tau_I \simeq \rho V/(6\pi a \eta) = 2pa^2/9\eta \sim 10^{-12}\ s;
\]

Here, to be particular, we set \(a \simeq 5\) \(\mu\)m and \(\eta \simeq 10\) kPa·s. Therefore the inertial time \(\tau_I\) comes out to be more than ten orders of magnitude shorter than the stress relaxation time \(\tau_R\). This implies that when considering the processes, which the particle undergoes with the reference times about \(\tau_R\), the inertial forces may be safely neglected.

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