Experimental research and modelling of the response of magnetorheological elastomers to cyclic load

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Abstract. Magnetorheological elastomers (MREs) belong to the new group of functional materials called “smart”. The MR effect is increased by choosing the material of the particles with high permeability. The dependence of dynamic moduli and damping of MREs on the external magnetic field and the applied frequency of cyclic shear deformation was studied. Samples of MRE were made of the silicon rubber matrix filled with micron-sized particles. The dynamic stiffness of MRE depends on magnetic flux density and rises with increasing frequency. The loss factor of MRE samples is controlled by the magnetic flux density and it depends also on the testing frequency. The behaviour of viscoelastic materials under uniaxial loading may be represented using rheological models composed of elastic and viscous elements. The dynamic variables storage and loss moduli and loss tangent were determined on the basis of the fitted parameters of the fractional model.

1. Introduction

Magnetorheological elastomers (MREs) are a group of materials with rheological properties very quickly modified by the application of a magnetic field. Magnetorheological elastomers are classified to the group of functional materials called “smart”. Smart materials are intelligent materials that have the ability to change their material properties under the influence of an external impetus such as stress, temperature, moisture, electric or magnetic fields. The rheological properties of MREs (the deformation and flow behaviour under stress) are modified by the application of an external magnetic field. Magnetorheological materials have their viscoelastic and rheological properties altered in the presence of a magnetic field. They demonstrate a quick response to external stimuli [1, 2].

Many factors influence on characteristic response, e.g. size, composition, distribution and volume of the iron particles, the isotropic or anisotropic arrangement of microparticles, elastomeric matrix etc. The MREs consist of magnetically permeable micro particles, dispersed in a non-magnetic matrix.

The unique characteristic of MREs is the shear modulus controlled by the external magnetic field continuously [3]. The MR effect is raised by choosing the material of the particles with high permeability.

The matrix materials of the MREs are silicon rubber, natural rubber, carbon black filled rubber. MRE composites adopt basic properties of the elastomeric matrix such as large deformations, Moulinse effect, frequency and amplitude dependencies, a decrease of stiffness at cyclic loading and viscoelastic time dependent characteristic [4].
1.1 Response of MREs to dynamic loading

The MR effect implies not only an instantaneous and reciprocated increase of the modulus and stiffness of a MR elastomers but also MREs exhibit a field-dependent damping [5]. The loss factor at low frequencies was found to increase by about 30% in the magnetic field [6]. The damping and stiffness of aligned MREs depend on the magnetic field, directions of load and the particle aligning in the elastomer [5, 7].

The response of MREs under dynamic loading has been studied experimentally by many researchers. Jolly et al. [8] presented a model of the magneto-viscoelastic effect of aligned anisotropic MREs in shear taking into account shear stress caused by the magnetic forces of particle dipoles. The results of their experiments showed the material moduli increasing monotonically with the applied field until magnetic saturation of the composite arises between 0.6 T and 0.8 T. The dynamic modulus increases slowly with frequency but the change of the modulus induced by the magnetic field is relatively insensitive to frequency.

All magnetorheological elastomeric composites show Payne effect, i.e. the storage modulus decreases with increasing strain amplitude because of the filler network failure. The Payne effect increases with raising concentration of carbonyl iron microparticles in the composite [5]. The loss factor of the shear modulus also strongly depends on the strain amplitude even for very small amplitudes [9]. The loss factor exhibits the increasing tendency with increasing of the strain amplitude of dynamic loading.

1.2 Modelling of dynamic behaviour of MREs

Main time-dependent features of MREs are derived from the viscoelastic properties of the elastomeric matrix. The dynamic properties depend obviously on the external magnetic field and the content of particles. Several phenomenological models developed as the connections of classical rheological models. The parameters of the model depending on the magnetic flux density are reported Li Y et al and Cantera et al [10, 11].

The four-parameter viscoelastic model based on the classical standard solid model combined with one additional parallel spring which represents the dependence of modulus on the magnetic field was developed by Li W H et al. [9]. Their experimental study contains the harmonic strain-controlled shear loading with different strain amplitudes and frequencies at various magnetic fields. The presented hysteresis loop have elliptical shapes, Authors showed a good agreement between experimental data and the results calculated by the model.

A complex linear viscoelastic model for isotropic MREs was presented by Xin et al. [12]. The viscoelasticity of MREs was divided into a mechanical part and magnetic part. The shear storage and loss moduli are developed using the Kraus model. The magnetic shear storage and loss moduli are derived based on the magneto-elastic theory with consideration of the magnetic saturation. The proposed model was evaluated by the experimental data.

Fractional rheological models are frequently applied recently to describe basic characteristics of different materials, rheological behavior of linear viscoelastic substances. There have been numerous tries to simulate damping properties of MREs using a fractional rheological element. This leads to a significant reduction in the number of parameters that is necessary to determine from experiments.

The model of magneto-sensitive isotropic rubber related to small strain was presented by Blom and Kari [13]. Their constitutive model characterises the amplitude dependence of magnetic sensitivity and viscoelasticity of isotropic MRE. The viscoelastic dependence is described by Abel type integral equation solved via fractional derivative calculus. The model includes the influence of changes in the magnetic field on the parameters.

The constitutive model of isotropic MREs formed on fractional derivatives, which are matrix-particle interaction and the magneto-induced modulus model was developed by Agirre-Olabide et al. [14]. The viscoelastic behavior was modelled using a four-parameter fractional derivative model. The limits of application of the linear viscoelasticity for the magneto-dynamic characterisation of MREs were defined by author Agirre-Olabide et al. [15]. The linear viscoelasticity region limits were examined depending on the particle content, frequency, external magnetic field, the inner structure of the samples and the temperature.
2. Experimental
The magneto-rheological elastomers were produced from room temperature vulcanization silicone rubber as the matrix matter and carbonyl iron microparticles as the filler. Globular carbonyl iron particles of diameters from 2 to 5 µm were supplied by Sigma-Aldrich company. The matrix material of silicone rubber is mixed of ZA 22 base and RZA 22 curative at a ratio of 1:1. Carbonyl iron microparticles of 27 vol% were mixed into the silicone rubber mixture. The polymerization process was performed at room temperature in a vacuum in order to remove bubbles. MRE samples are isotropic.

Dynamic viscoelastic measurements were carried out on an Instron Electropuls 3kN, [1]. The magnetic field induced by an electromagnet was utilized during the cyclic loading on double-shear samples of MRE under controlled shear strain. The detailed information about samples is described in Table 1.

| Sample | Material of matrix | RTV silicone rubber |
|--------|--------------------|---------------------|
|        | Material of filler  | Globular carbonyl iron microparticles |
|        | Sample shape       | Rectangular |
|        | Sample dimensions  | 20x20x5 mm³ |
|        | Glue               | Cyanoacrylate glue with activator |
|        | Volume of carbonyl iron powder | 27% |

The amplitude of the cyclic shear strain was 0.1 and the frequency was changed from 1 to 10 Hz with the step 10 Hz. The samples were cyclically loaded under the magnetic field from 0.1 to 0.8 T. The samples were tested without and with the magnetic field to receive the basic dynamic MRE properties, specifically storage and loss moduli and the loss angle. Dynamic moduli and the loss factor were found out as the function of the magnetic flux density (magnetic field) and the frequency and amplitude of the cyclic deformation in shear. Two methods were used for evaluation of experiments: Phase shift angle determination and evaluation of loss modulus from the area of the hysteresis loop.

2.1 Phase shift angle determination
We suppose that the raw signals, i.e. displacement u(t) and force response F(t), are harmonic functions approximately and we use the Fast Fourier Transform (FFT) in order to determine the phase shift δ at the main excitation frequency [16]. Amplitudes of shear strain and shear stress were determined from raw recorded signals, eq. (1).

\[ \gamma(t) = \gamma_0 + \gamma_a \sin(\omega t) \rightarrow \tau(t) = \tau_0 + \tau_a \sin(\omega t + \delta) \]  

Dynamic modulus \( G^* \) was determined as the ratio between the amplitudes of stress and strain. The storage and loss moduli were calculated as follows

\[ G^* = \frac{\tau_a}{\gamma_a}, \quad G' = G^\ast \cos(\delta), \quad G'' = G^\ast \sin(\delta) \]  

2.2. Loss modulus evaluation from the area of a hysteresis loop
We calculated the loss modulus from the dissipated energy density which corresponds to the area of the hysteresis loop. The dissipation of mechanical energy in the course of one cycle can be expressed as
\[ D = \int_0^T \tau(t) \dot{y}(t) dt = \pi y_0^2 G^* \sin \delta = \pi y_0^2 G'' \] (3)

The density of the dissipated energy was calculated numerically as the mean value of the areas of twenty consecutive steady hysteresis loops in each experiment. The raw signal evaluation can be crucial for successful modelling of the MRE’s response.

3. Modelling
The behavior of viscoelastic properties of MRE samples under uniaxial loading may be described by means of the rheological models containing elastic and viscous elements. The rheological Zener model is a specific case of the generalized Maxwell model assembled of a Maxwell branch in parallel with a spring. Such a model of three parameters is not sufficient for the quantitative representation of the behavior of the MRE. In order to improve the similarity, the number of parameters needs to be increased. This is done by connecting a number of springs and dashpots - to create a so-called ladder model.

The dependency of dynamic moduli can be characterized by a minimum of material constants using the fractional calculus. The fractional Zener model is an adequate uniaxial model for small strains [14, 17]. It corresponds to a linear spring with an elastic modulus in parallel with a fractional Maxwell element. The fractional differential equation is adequately similar to the 1st order differential equation of the classic Zener element.

\[ \sigma(t) + \tau^\alpha \frac{d^{\alpha} \sigma}{dt^\alpha} = (G_m + G_e) \tau^\alpha \frac{d^{\alpha} e}{dt^\alpha} + G_e \varepsilon(t) \] (4)

The model has 4 parameters (elastic part of the viscoelastic member of the model \( G_m \), the stiffness of the spring in elastic branch \( G_m \), the relaxation time \( \tau \) and the fractional parameter \( \alpha \) ) which should be fitted to the experimental data.

The shift from the time domain to the frequency domain is realized by the Fourier Transform. Storage and loss moduli are obtained by separating the real and imaginary part of the dynamic modulus.

\[ G' = G_e + G_m(\tau \omega)^\alpha \frac{\cos(\alpha \pi/2) + (\tau \omega)^\alpha}{1 + 2(\tau \omega)^\alpha \cos(\alpha \pi/2) + (\tau \omega)^{2\alpha}} \] (5)

\[ G'' = G_m(\tau \omega)^\alpha \frac{\sin(\alpha \pi/2)}{1 + 2(\tau \omega)^\alpha \cos(\alpha \pi/2) + (\tau \omega)^{2\alpha}} \tan \delta = \frac{G''}{G'} \] (6)

The loss tangent values evaluated from experiments were fitted to the fractional Zener model.

4. Results and Discussion
4.1. Results of experimental measurements
The dynamic stiffness of MRE depends on magnetic field and rises with increasing testing frequency, see Figure 1 & 2. The loss angle of MRE is controlled by the magnetic field and it also depends on the testing frequency. The dependency of all variables on frequency is slightly increasing in the frequency range of 1 to 10 Hz. The increments of storage and loss moduli between the zero-field and 0.78 Tesla are 32% and 38% resp., the loss angle increase was around 6%.

4.2. Fitted parameters of fractional model
The loss tangent values evaluated from experiments were fitted to the fractional Zener model. We obtained an exceptionally good agreement even for different intensity of the magnetic field (ranging from 0 to 0.6 Tesla) as can be seen in Figure 3. The fitted parameter alpha of the fractional order derivative decreases slightly with the rising magnetic field intensity (Figure 4). This indicates that with the increasing intensity of the magnetic field, the stiffness of the MRE sample increases.
The hysteresis loop record carries all the information we can harvest from a dynamic experiment. It is, therefore, appropriate to fit the model parameters directly to the recorded raw data of strain and stress. The fitting was carried out in Matlab by nonlinear least squares. The dynamic moduli $G'$, $G''$ and loss tangent were determined on the basis of the fitted parameters of the fractional model.

**Figure 1.** The storage modulus dependency on the magnetic field

**Figure 2.** The storage modulus dependency on the frequency

**Figure 3.** The loss angle evaluated from experiments and fitted for different intensity of the magnetic field.

**Figure 4.** The fitted parameter alpha of the fractional order derivative.

5. Conclusion

In this contribution, we presented a study of the mechanical behavior of magneto-sensitive elastomers with the isotropic distribution of the magnetic particles in an external magnetic field and we presented the phenomenological model of the dynamic response based on fractional Zener derivative rheological element. The hysteresis loop record carries all the information we can harvest from a dynamic experiment. Therefore, it is appropriate to fit the model parameters directly to the recorded raw data of strain and stress. The fitted parameter alpha of the fractional order derivative decreases slightly with the rising magnetic field intensity. This indicates that with the increasing intensity of the magnetic field, the stiffness of the MRE sample increases.

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