Study of magnetorheology and sensing capabilities of MR elastomers

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Abstract. This study focuses on the magnetorheology and sensing capability of graphite based Magnetorheological Elastomers (Gr MREs). By introducing graphite (Gr) to conventional MREs, the Gr MREs are derived. The anisotropic sample with 20% graphite weight fraction was selected to be compared with anisotropic conventional MREs. The microstructures of anisotropic Gr MREs and conventional MREs were observed. Both steady state tests and dynamic tests were conducted to study rheological properties of the samples. For dynamic tests, the effects of strain amplitude, and frequency on both storage modulus and loss modulus were measured. For sensing capability, the resistance of selected Gr MREs under different magnetic fields and external loadings is measured with a multi-meter. Either higher magnetic field or more external loading results in the resistance increment. Based on an ideal assumption of perfect chain structure, a mathematical model was proposed to investigate the relationship between the MRE resistance with the external loadings. In this model, the current flowing through the chain structure consists of both tunnel current and conductivity current, both of which depends on external loadings. The modelling parameters were identified and reconstructed from comparison with experimental results. The comparison indicates that both experimental results and modelling prediction agree favourably well.

1. Introduction

Magnetorheological elastomers (MREs) are smart materials where polarized particles are suspended in a non-magnetically solid or gel-like matrix [1]. These materials exhibit characteristics that their moduli can be reversely controlled by an external magnetic field. MREs have recently found a variety of applications, such as adaptive tuned vibration absorbers, dampers, sensors, and so on [2-4]. There are two kinds of MREs which are anisotropic and isotropic [5,6]. In anisotropic samples, polarised particles can be arranged as chains in polymer media such as silicon rubbers and natural rubbers.

MREs generally consist of three major components: magnetizable particles, matrix, and additives. Iron particles are generally used as the filler material to fabricate MREs, this is because iron has one of the highest saturation magnetisation values of metallic elements as well as high permeability, low remnant magnetisation and high saturation magnetisation. High permeability and saturation magnetisation are thought to provide high inter-particle attraction and thereby a high MR effect. The effect of particle shape, size and volume fraction on the overall MRE performances have been intensively investigated. Lokander and Stenberg [7] measured the MR effect for isotropic nitrile rubber MRE with varying sizes and content of iron particles. The MR effect was larger for materials

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with ASC300 iron (particle size < 60 μm) than for materials with carbonyl iron (particle size 3.9-5.0 μm). Shiga et al. [8] measured the increase in shear modulus as a function of the particle volume fraction. For aligned MREs the change in shear modulus increases with an increasing particle volume fraction. When the concentration of filler is higher than 30 vol.%, the mechanical properties of the composite deteriorate rapidly and the stiffening of the material is larger than the increase of the MR effect. Both natural rubber and silicone rubber are used as typical matrixes [9]. Natural rubber is an elastomer. The purified form of natural rubber is the chemical polyisoprene which can also be produced synthetically. Heat is normally required to vulcanise silicone rubber. The silicone rubber and a vulcanising silicon sealant (at room temperature), is mixed with silicon oil to changing its ductility. The silicon oil is selected on the basis of preliminary studies with different elastomers. Additives are used to adjust the mechanism properties or electrical performance of MREs. Silicone oil is an additive to increase the gaps between the matrix molecules and to decrease the gaps between the conglutination of molecules. Apart from increasing the plasticity and fluidity of the matrix, the additives can average the distribution of internal stress in the materials, which makes them ideal for fabricating MRE materials [10]. Graphite powder is also a kind of additive which can affect the magnetorheology and electrical conductivity of MREs [11,12].

By introducing graphite microparticles into the elastic matrix, the Gr MREs is derived that behave a lower electrical conductive characteristic, which has a potential to work as a sensing material for development of force and magnetic field sensors [11]. The Gr MREs show resistance changing when an external load is applied to MRE samples. By this feature of MREs, the external stress signal can be converted to a resistance signal, which can be used potentially in a force sensor.

Kchit and Bossis [13] found that the initial resistivity of metal powder at zero pressure is about 108 Ωcm for pure nickel powder and 106 Ωcm for silver coated nickel particles. The change in resistance with pressure was found to be an order of magnitude larger for a MRE composite than for the same volume fraction of fillers dispersed randomly in the polymer. Wang et al. [14] proposed a phenomenological model to understand the impedance response of MREs under mechanical loads and magnetic fields. Their results showed that MRE samples exhibit significant changes in measured values of impedance and resistance in response to compressive deformation, as well as applied magnetic field. Bica [12] found that MRE with graphite micro particles (~14%) is electroconductive. The magnetoresistance has an electric resistance whose value diminishes with both the increase of the intensity of the magnetic field and with the compression force. The variation of resistance with magnetic field intensity is due to the compression of MRE with graphite microparticles. In the approximation of the perfect elastic body, the sums of the main deformations and the compressibility module of MRE with graphite microparticles, depend on the magnetic field intensity. Li et al. [11] introduced graphite into conventional MREs and found a MRE sample with 55% carbonyl iron, 20% silicon rubber and 25% graphite powder has the best performance. The test result showed that at a normal force of 5 N, the resistance decreases from 4.62 kΩ without a magnetic field to 1.78 kΩ at a magnetic field of 600 mT. The decreasing rate is more than 60%. This result also demonstrated the possibility of using MREs to develop a sensor for measuring magnetic fields. This result indicates that the detection is very sensitive to the normal force. When the normal force is 15N, the field-induced resistance only has less than 28% change from 0.65 kΩ at 0 mT to 0.47 kΩ at 600 mT. Tian et al. [15] proposed a representative volume unit (RVU) to analyze the resistance of Gr MREs theoretically. Many Gr MREs samples with various graphite weight fractions were fabricated and measured for the resistance. They found that the samples’ resistance decreases with increasing either the external force or the intensity of applied magnetic field. Meanwhile, the sample with higher graphite concentration shows a lower resistance. Both Bica’s [12] and our previous work [11,15] experimentally demonstrated the graphite based MREs exhibit conductive properties; however, the investigation of the mechanism lacks in-depth discussion. For example, very few reports in literature have been developed to investigate the effect of external loading on the electric properties. This is the major motivation of this research.
2. Experimental

2.1. MRE fabrication and microstructure observation
The materials used for the Graphite MR Elastomers are: silicone rubber (Selleys Pty. LTD); silicone oil, type 378364 (Sigma-Aldrich Pty. LTD); carbonyl iron particles, type C3518 (Sigma-Aldrich Pty. LTD); and graphite powder, type 282863 (Sigma-Aldrich Pty. LTD). The particle sizes of graphite powder are about 20 μm, while the iron particles’ diameter is between 3 μm and 5 μm.

In this study, a graphite based anisotropic MRE sample (graphite weight fractions Gr = 20% wt) was fabricated to be compared with the conventional anisotropic MRE sample. Table 1 shows the compositions of the two anisotropic MRE samples. All the samples contain the same compositions of 10 g carbonyl iron particles, 3 g silicone rubber and 3 g silicone oil. The only difference is that the graphite based MRE has 20% graphite weight fraction.

| Sample          | Carbonyl iron | Silicone oil | Silicone rubber | Graphite | Graphite weight fraction (Gr %) |
|-----------------|---------------|--------------|-----------------|----------|--------------------------------|
| Conventional MRE | 10 g          | 3 g          | 3 g             | 0 g      | 0%                             |
| Graphite based MREs | 10 g    | 3 g          | 3 g             | 4 g      | 20%                            |

After the samples were fabricated, LV-SEM (JSM 6490LV SEM) was used to observe microstructures of MRE samples. Figures 1&2 show the surface imaging for MRE microstructures.

![Figure 1. Microstructure of anisotropic conventional MRE (Gr 0%).](image1)

![Figure 2. Microstructure of anisotropic Gr MRE (Gr 20%).](image2)

In figure 1 that shows the microstructure of anisotropic conventional MRE, the carbonyl iron particles array in chains in the matrix. However, figure 2 demonstrates that in anisotropic Gr MREs, when the carbonyl iron particles array in chains, the graphite powders disperse in the matrix randomly. The reason of this phenomenon is that the magnetic field can not affect on the graphite powders but only on the carbonyl iron particles. So by the magnetism, the carbonyl iron particles move to chains along the same direction as the magnetic field in the matrix.

By comparing figure 1 & 2, we can see that the carbonyl iron chains in the sample without graphite have a better lines performance. The reason is that when the mixed paste of carbonyl iron, silicone rubber, silicone oil and graphite is curing under the magnetic field, the graphite powders in graphite MREs affect the carbonyl iron particles’ movement. The more graphite in the mixture, the more effects are applied on to the carbonyl iron chains, which influence the magnetorheology of MREs.

2.2. Rheological experimental device
A rotational Rheometer (MCR 301, Anton Paar Companies, Germany) and a Magneto Rheolgical Device (MRD 180, Anton Paar Companies, Germany) were used to measure the Gr MREs’
mechanical properties. The Magneto Rheological Device is equipped with an electromagnetic kit which can generate a magnetic field perpendicular to the direction of the shear flow. A 20 mm diameter parallel-plate measuring system with 1 mm gap was used. The samples were sandwiched between a rotary disk and a base parallely. In this study a steady-state rotary shear and oscillatory shear were both used for the experiments.

In this experiment the magnetic flux density of the sample of MRE ($B_{\text{MRE}}$) in the measuring gap depends not only on the current ($I$) applied to the samples, but also on the magnetic properties of MRE materials. The relationship between $B_{\text{MRE}}$ versus $I$ is found to be: $B_{\text{MRE}} = 220 I$, where the units of $B_{\text{MRE}}$ and $I$ are in mT and amp (A), respectively.

In the following test, the test current varies from 0 A to 2 A with the increment 0.5 A, whose intensity of magnetic field is 0 mT to 440 mT with the increment 110 mT.

2.3. Sensing capability experimental measurement

Figure 3 shows a schematic of the experimental device used. In this setup a long plastic plate is used to hold the weight to apply the load to the Gr MRE samples. A Gaussmeter (HT201, Hengtong magnetoelectricity CO., LTD) is used to test the intensity of the magnetic field. A multimeter (Finest 183, Fine Instruments Corporation) measures the resistance in the Gr MRE samples.

![Figure 3. sketch of the experimental device](image)

3. Results and discussion

3.1. Steady state rheological test results

Under rotary shear the shear stress and shear strain of MREs under fields varying from 0–440 mT were measured. The MR effect was evaluated by measuring the shear strain-stress curve of the sample with and without a magnetic field applied.

Figure 4 (a) & (b) show the strain-stress curve of the two samples at 5 different magnetic field intensities ranging from 0 to 440 mT. The slope of the strain-stress curve is the shear modulus of the material. As can be seen in the figures, both the samples’ shear modulus show an increasing trend with increasing magnetic field intensity before they reach magnetic saturation at high field strength, which proves that both the MRE samples exhibit obvious MR effects. Also from Figure 4, the shear stress shows a linear relationship with the shear strain when the strain is within a range. This means the MRE acts with linear viscoelastic properties when the strain is below a limitation. For conventional MREs, the limitation is around 50% shear strain, which was previously reported [22]. When the graphite weight fraction increases from 0 to 20%, the range of linearity decreases from 50% to around 7%. For the strain over the limitation, the shear modulus reaches a saturation (maximum value) and
decrease steadily. This could be due to sliding effect. The higher magnetic field intensity leads to higher steady shear stress.

Figure 4. Strain-stress curve versus magnetic field (anisotropic MRE) (a) Gr 0% and (b) 20%.

For each curve, the slope equals to the ratio of peak shear stress to the relevant shear strain, which is the shear modulus. By analyzing the slopes of the curves, it is easy to see that the more graphite in the material, the less growth of slopes when the magnetic field increase from 0 to 440mT. This is because of the contributions of graphite powders to the stiffness of the samples and to pull down the volume fraction of iron particles in the samples. The graphite increases the initial stiffness of graphite MREs, thus the stiffness changing from the MR effect can not be as the same as the conventional MREs. Meanwhile, the graphite added to the sample downgrades the volume fraction of iron particles in the sample, which plays crucial role in determining the MRE effect of the MR materials. Therefore, the MR effect of Gr MREs is lower than the conventional MREs.

3.2. Dynamic rheological tests results
In order to obtain the dynamic mechanical behavior of MRE, both angular frequency sweep tests and strain amplitude sweep tests were used. Five sets of data were collected for different amplitudes of oscillation, according to the various magnetic fields input to the samples of MR elastomers. Same as the steady state tests, five different magnetic field intensities, 0, 110, 220, 330 and 440 mT, were used in this experiment. The amplitude of shear strain in angular frequency sweep tests is set at 1% and the input frequency was 5 Hz in the strain amplitude sweep tests.

3.2.1. Strain amplitude sweep. In the strain sweep test, the storage and loss moduli were tested by varying strain from 0.01% to 100% at different magnetic fields. Figure 5 & 6 show the changing of storage modulus at the strain amplitude sweep.

Figure 5. Storage Modulus versus strain amplitude sweep (anisotropic MRE Gr 0%).

Figure 6. Storage Modulus versus strain amplitude sweep (anisotropic MRE Gr 20%).
In figures 5 & 6, the overall trend of storage modulus is to decrease with the strain amplitude. It goes down smoothly within 10% shear strain and begins to drop significantly over 10% shear strain, which means at the high shear strain, the storage modulus are much smaller than that at low shear strain.

Figures 7 & 8 show the storage modulus versus magnetic field at 0.1% and 10% shear strain, respectively. The two shear strains are the beginning and end of linear range. In these two figures we can see that the storage modulus shows an increasing trend with the intensity of magnetic field. The ratio of storage modulus at 440 mT to that at 0 mT is the MR effect. The MR effect of anisotropic conventional MRE is around 6.3 and 5.4 at 0.1% shear and 10% shear, respectively; when the graphite weight fraction increases to 20%, the MR effect decreases to around 2.5 at 0.1% shear and to 2.38 at 10% shear. This proves that with the growth of graphite, the MR effect decreases.

3.2.2. Angular frequency sweep. In this test, the strain is set at 1%. According to the experimental equipments, the angular frequency was varied from 6 to 100 1/s at different magnetic fields as 0, 110, 220, 330 and 440 mT. The figures 9 & 10 show the storage modulus curves of the MRE samples at frequency sweep.

From the figures above, we can see that in the log-log scale, the storage moduli of the two samples are both increasing linearly with the growth of angular frequency. This means with a higher angular frequency, the samples have bigger storage modulus. This logarithmically linear relationship of the storage modulus to the angular frequency can be used to predict the storage modulus at a certain frequency. The effect of the graphite weight fraction on the MR effect was shown in figures 11 & 12, from which it can be seen that with higher graphite weight faction, the samples have a bigger storage modulus. This also proves that the graphite powders contribute to the initial stiffness of MRE samples.
Figure 11. Storage modulus versus angular frequency sweep (without magnetic field).

Figure 12. Storage modulus versus angular frequency sweep (440mT magnetic field).

The ratio of storage modulus at 440 mT to the storage modulus at 0mT is the MR effect. The ratio of anisotropic conventional MREs is around 5.2, when the graphite weight fraction increases to 20%, the MR effect is only around 2.4. This again means with the growth of graphite, the MR effect decreases. This phenomenon is because graphite powders contribute to increase the samples’ stiffness and to lower iron particles volume fraction in the sample, therefore the MR effect can only have less values on the Gr MRE samples than conventional MREs.

3.3. Sensing capability measurement results and discussion

After the test, the resistance data of anisotropic Gr MRE with 20% graphite weight fraction is shown in figure 13.

Figure 13. Resistance versus load (anisotropic MRE Gr 20%).

As can be seen from figure 13, in a fixed magnetic field when the external load increases from 0 to 10 N the resistance of Gr MREs reduces at all different magnetic field intensities. With smaller load the resistance changes significantly but it decreases slowly when the load is more than 5 N. Meanwhile it is clear that under a fixed external load, the higher magnetic field intensity leads to a lower resistance value. For instance, at 5 N external force, the resistance decreases from 1086.3 kΩ (without the magnetic field) to 918 kΩ at a 440 mT magnetic field. This trend can be seen in Figure
13. Therefore, the total trend is the decline in resistance with raising the magnetic field or increasing the external load.

To explain how the microstructure affects the resistance of Gr MRE, from the Dipole Model we announced a representative volume unit (RVU) that consists of two neighbouring hemispheres and the surrounding polymer matrix, which can be regarded as the minimum volume element in the conventional MRE. Figure 14 shows the longitudinal section of the unit. [15]

![Figure 14. The longitudinal section of RVU.](image)

In the theoretical approach, with the help of Fowler-Nordheim equation, Hertz theory and theory of magnetism [16-21], we got a final equation showing the resistance $R_g$ of Gr MREs.

$$
R_g = \frac{\lambda_e \lambda_i l}{3A \phi \left[ \frac{2\alpha E \exp \left( -\frac{h\beta}{2r_p E} \right) + \sigma_f}{r_p h} \right] \left( r_{i0} + r_p \left( \sigma_0 + \sigma_i + \sigma_2 \right)^{1/3} - \sigma_0^{1/3} \right) \left( \frac{3\pi \left( 1 - \nu^2 \right)}{2E_p} \right)^{1/2} (1)
$$

By substituting the known and constant coefficients into Equation 1, a simplified equation is derived.

$$
R_g = \frac{1.78 \times 10^{-2}}{10^{-8} + 0.000002 \left( \left( 2000 + \sigma_1 + \sigma_2 \right)^{1/3} - 2000^{1/3} \right) \times 0.001166} (2)
$$

where $\sigma_f$ is the compressive stress from the external force and $\sigma_2$ is the compressive stress from the external magnetic field.

To show the comparison between the experimental result and the theoretical prediction (anisotropic MRE with graphite weight fraction 20%) clearly, four series of both the experimental result and the theoretical result are selected, as shown in Figure 15. Figure 15 shows that the experimental result and theoretical result couldn’t match each other perfectly, but that the trends of both the experimental result and the theoretical result are the same.

Similarly some other comparison can be made from the data. At the fixed external force (which is expressed as load) when the magnetic field intensity increases the resistance of the sample decreases. The data under three external loads such as 1 N, 5 N and 10 N were chosen to compare the experimental and theoretical results in figure 16. With an increase in the magnetic field intensity, the sample’s resistance decreases. A higher external applied load leads to lower resistance of Gr MREs.
4. Conclusion
An anisotropic Gr MREs with 20% graphite weight fractions was selected to compare with anisotropic conventional MREs about rheology and to be studied its sensing capability in this study. LA SEM was used to observe their microstructures. This observation shows that the graphite powders effect the forming of carbonyl iron chains by connecting two iron chains in parallel and connecting the disconnected iron chains the graphite contributes to the conductivity of MREs.

The steady state and dynamic tests such as strain amplitude sweep and angular frequency sweep were used to test the magnetorheology of Gr MREs. With the help of graphite in MREs, the Storage and Loss Moduli are both changed. The steady state tests showed that the graphite decreases or even diminish the viscoelastic linear range of MREs. The dynamic test proved that the samples with higher graphite weight fraction show higher initial storage and loss moduli and lower MR effects.

Based on a Dipole model, a representative volume unit was presented to show the resistance of ideal anisotropic MREs. The current flowing through the ideal chain structure were derived by taking into account both the tunnel current and conductivity current. The exponential parameters were identified and then used to reconstruct modelling predictions. The comparison between experimental results with modelling predictions indicates that the proposed mathematical model performs well in investigating the sensing capabilities of the graphite based MRE elastomers.

Totally compared by the microstructures and experimental data from this study, we got the major effects of graphite in Gr MREs: (a) Graphite powders affect the forming of iron lines when the mixture of iron particles, graphite powders, silicone rubber and silicone oil is curing. The graphite particles lead to an increment of initial mechanical properties and a decrement of the MR effect because of the volume fraction of iron particles was lowered by adding the graphite powders to the sample, (b) For a Gr MREs sample, the graphite powder can connect the off-state lines and link the adjacent lines parallelly, which significantly increases the conductivity of the Gr MREs sample.

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