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A rheological model of the dynamic behaviour of magnetorheological elastomers

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A rheological model is described that was developed to simulate the dynamic behaviour of magnetorheological elastomers (MREs). The viscoelasticity of the polymer composite, magnetic field induced properties and interfacial slippage between the matrix and particles were modelled by analogy with a standard linear solid model, a stiffness variable spring and a spring-Coulomb friction slider respectively. The loading history and rate dependent constitutive relationships for MREs were derived from the rheological model. The hysteresis loop from shear strain - shear stress plots, which determines the shear modulus and loss factor, were obtained from substituting cyclic loading into these constitutive relationships. The dynamic behaviours of MREs were simulated by changing parameters in the rheological model to influence MREs' performances. The simulation results verified the effectiveness of the model.

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I. INTRODUCTION

Magnetorheological elastomers (MREs) are 'intelligent' materials having physical properties that can be reversibly and instantaneously controlled by an external magnetic field [1]. they consist of micron sized Typically, magnetically permeable particles dispersed in a polymer matrix. Upon the application of a magnetic field, the particles form ordered structures that result in field-induced performance improvements. Over the past few years, MREs have attracted increasing attention and have been considered for applications as adaptive stiffness elements in vibration absorbers [2], automotive bushing [3] and piezoelectric power actuators [4].

Recently, many methods have been proposed to examine the behaviour of magnetorheological (MR) materials. For example, Ginder et al of the Ford Research Laboratory analysed average magnetic induction using finite element analysis and computed the shear stresses from the field using Maxwell's stress tensor [5]. Jolly et al [6] extended a simple dipole model, based on the magnetic interactions between two adjacent particles, to approximate MRE performances. Davis [7] calculated the saturated field-induced shear modulus by considering the interactions in a single particle chain. Chen at al [8] created a finite length model based on the microstructural observation of the particles forming the discontinuous and finite length column structures in polymer matrices. Most of these models exhibit good accuracy in predicting MR effects caused by embedded ferromagnetic particles. However, the properties of the MRE can't be comprehensively characterized by only considering the MR effect which is the merely the performance induced by the ferromagnetic particles. The effects of the polymer matrix and its interaction with embedded particles also play an important role in establishing MRE performance. Moreover, an MRE component is commonly subjected to dynamic loading in engineering applications. Its mechanical properties, including shear modulus and damping ratio, are greatly dependent on the amplitude and frequency of the dynamic loading [2, 9]. This results in uncertainty in characterising MRE performance and brings difficulties in system control when using MREs in vibration absorbing technology. Therefore, this work aims to develop a theoretical model by considering the effect of the matrix, embedded particles and the interaction between them, for the prediction of mechanical behaviour of MREs subjected to dynamic loading and external magnetic fields.

This paper is organized as follows: Section 2 described the methodology of the rheological model; the simulation results of the model are reported in Section 3 along with an investigation of the influence of parameters in the model. The reliability of the model was also discussed in this section; Conclusions are given in Section 4.

II. METHODOLOGIES

The present model is based on rheology methods allowing examination of the dynamic behaviour of MREs. The contribution to the overall performance of an MRE of each of the following influences is taken into account:

i) the viscoelasticity of the polymer composite (the matrix material filled with ferromagnetic particles),

ii) the magnetic-field-induced mechanical properties and

iii) the interfacial slippage between the matrix and particles. They are modelled by analogy with a standard linear solid model (combination of two spring elements and a dashpot element), a stiffness variable spring and a spring-Coulomb friction slider respectively, shown in Fig.1. The constitutive equations for each component are given in the text following the figure.



Fig.1 A rheological model for examining dynamic properties of MREs comprising three components to simulate (a) the viscoelasticity of the polymer composite, (b) the magnetic-field-induced mechanical properties and (c) interfacial slippage between the matrix and the particles.

a. Viscoelasticity of the polymer composite

The matrix of an MRE is a rubber, which is a typical viscoelastic material. So the mechanical properties of the polymer composite without the application of a magnetic field can be approximately simulated using a standard linear solid model shown as component I in Fig.1. When the component is subjected to external loading causing shear strain γ for small strains, the shear stress is proportional to the shear strain in the spring elements and has a linear relationship with the shear strain rate in the dashpot element. So it can be assumed that

$$\tau_1 = G_1 \gamma \,, \tag{1}$$

$$\tau_2 = G_2 \gamma_2, \tag{2}$$

$$\tau_2 = \eta \,\gamma_1 \,, \tag{3}$$

$$\gamma_2 + \gamma_2 = \gamma , \qquad (4)$$

where G_n (n=1 and 2) is the shear modulus of the spring elements (see Fig.1), τ_n and γ_n are the corresponding shear stress and shear strain and η is the dynamic viscosity of the dashpot element.

b. Magnetic-field-induced mechanical properties

When an MRE is exposed to an applied magnetic field, the embedded ferromagnetic particles are magnetized. The magnetic forces between the particles result in a field-induced modulus. As the modulus is field dependent, a stiffness variable spring (shown in Fig.1) is used to simulate the magnetic-field-induced mechanical properties.

The field-induced stress can be calculated by taking the derivative of the dipole energy density (U) with respect to the shear strain [6]

$$\tau_3 = \frac{\partial U}{\partial \gamma},\tag{5}$$

Considering the dipole interaction between adjacent ferromagnetic particles within the chain, the magnetic energy density of an MRE can be expressed as

$$U = \frac{\phi^2 (\gamma^2 - 2) J_p^2}{8\pi \mu_1 \mu_0 \pi (1 + \gamma^2)^{5/2}},$$
 (6)

where ϕ is the magnetic particle volume fraction and this value is taken as 33% for an optimized MRE with a maximum MR effect [10], μ_1 is the relative permeability of the medium, approximated to 1 in a vacuum. $\mu_0 = 4\pi \times 10^{-7} N/A^2$ and $J_p = \mu_0 M_p$ is the dipole moment magnitude per unit particle volume.

The nonlinearity of the magnetic field strength versus internal magnetisation is described by the Frohlich-Kennely law [11]:

$$M_{p} = \frac{(\mu_{p} - 1)M_{s}H}{M_{s} + (\mu_{p} - 1)H},$$
(7)

where M_s is the saturated magnetisation and it is assumed that $\mu_0 M_s = 2.1T$ for ferromagnetic materials, μ_p is the permeability of ferromagnetic materials in low fields, $\mu_p = 1000$, and *H* is the external magnetic field strength.

Form equation (5)-(7), the field-induced stress can be expressed as a function of γ and *H*:

$$\tau_{3} = \frac{27\phi^{2}\mu_{0}(\mu_{p}-1)^{2}M_{s}^{2}H^{2}\gamma(4-\gamma^{2})}{4\pi\mu_{1}[M_{s}+(\mu_{p}-1)H]^{2}(1+\gamma^{2})^{7/2}},$$
(8)

c. Interfacial slippage

Previous MRE experimental investigations indicated that the interfacial bond conditions between embedded particles and the surrounding matrix have great influence on MRE properties [12]. For example, the shear modulus is enhanced and the damping ratio of MRE is decreased with the incremental increase of interfacial bond strength. In the present model, the interfacial slippage is simulated using a component of a spring-Coulomb friction slider as shown in Fig.1.

When the component is subjected to a small loading, the slider is fixed and the length of the spring extends proportionally to the loading. When the loading reaches a critical value τ_c , the slider begins to slip and the spring retains a constant length. Thereafter, if a load is applied in the opposite direction with increasing magnitude, the spring firstly becomes compressed and finally the slider will once more slip. The slippage of the friction slider is used to model the interfacial behaviour between the particles and the matrix under external loading. The critical value τ_c denotes the interfacial bond strength.

Their constitutive relationship can be written as:

$$\overset{\bullet}{\tau_4} = G_4 \gamma \qquad \text{when } -\tau_c < \tau_4 < \tau_c \,, \qquad (9)$$

$$\tau_4 = \operatorname{sgn}(\gamma)\tau_c$$
 outside this range, (10)

where τ_4 is the shear stress in the spring and friction slider element and G_4 is the shear modulus of the spring element. Both of these parameters can be obtained from macroscopic slippage experimentation, where the critical force and displacement for slippage determines τ_c and τ_c/G_4 respectively. In the present model, τ_c is a variable and its influence on the MRE behaviour is investigated. The effect of G_4 is not considered and assumed to be equal to 1 MPa.

Stress in each component makes a contribution to the external loading, so it is assumed that

$$\tau = \sum_{i=1}^{4} \tau_i \tag{11}$$

From above Eqns. (1)-(11), the constitutive relationships of MREs can be solved using a Laplace transform and numerical methods.

In this research, a differential equation solution toolbox in Mathcad was used to obtain the numerical solutions. After substituting a harmonic strain $\gamma(t) = \gamma_0 \cos(2\pi f t)$ into the equations set, a time dependent stress was accordingly derived. There was a phase difference between the strain and stress due to the energy dissipation caused by the viscoelasticity of the matrix and interfacial slippage. Consequently, the stress and strain formed a hysteresis loop from which the dynamic properties, including shear modulus and loss factor, can be calculated.

The value of the complex shear modulus is equal to the ratio of the amplitudes of the shear stress and shear strain:

$$G = \frac{\tau_0}{\gamma_0},\tag{12}$$

The area enclosed by the hysteresis loop represents the energy dissipated internally during one cycle. The loss factor is proportional to the ratio of the dissipated energy (ΔW) during one complete cycle to the maximum stored energy (W) from the inception of the loading until it reaches a maximum, represented by

$$\tan \delta = \frac{\Delta W}{W} / 2\pi \tag{13}$$

where
$$\Delta W = \oint \tau d\gamma$$
, (14)
 $W = \int_{\gamma_0}^{\gamma_0} \tau d\gamma$ (15)

$$W = \int_0^{\gamma_0} \tau d\gamma , \qquad (15)$$

III. SIMULATION RESULTS AND DISCUSSIONS

The dynamic behaviour of MREs was investigated by changing parameters in the rheological model to influence the performance of the composite. These parameters were i) intrinsic factors in MREs, modulus (E_0) and dynamic viscosity (η) of polymer composite and interfacial bond strength (τ_s) and ii) external factors including loading amplitudes (γ_0), frequency (f) and magnetic field strength (H_0). All of the parameters were obtained from the related experiments. They were ascribed a reasonable range of values for the simulations described in this work.

3.1 Influence of interfacial bond strength (τ_s) and strain amplitudes (γ_0)

Simulations were carried out for interfacial bond strengths ranging from 0.001 to 10 MPa at several strain amplitudes of 0.01, 0.1, 0.5 and 1. Other parameters were set as constants:- $E_0 = 1MPa$,

 $\eta = 8500 \text{Pa} \cdot \text{s}, f = 1Hz \text{ and } H_0 = 50k \text{ A/m}.$

Fig.2 depicts the simulation results of the variations of shear modulus in MREs with interfacial bond strength for four different strain amplitudes. It can be seen that each MRE shear modulus firstly rises with an increase in interfacial bond strength. This is so because for a structured system, a stronger internal connection results in less internal freedom and a higher stiffness for the whole system. However, when bond strength is raised to a critical value, no interfacial slippage takes place. So the stiffness and shear modulus is unaffected by the increase in bond strength and becomes constant. This result is also shown in Fig.2.

In addition, comparing each curve in Fig.2 for an MRE having applied shear strains with different amplitudes, it can be seen that, when an MRE is subjected to a large load, the shear modulus is lowered. This simulation result is consistent with the Payne effect [13]. It is worth noting that when the interfacial bond strength is sufficiently strong, the Payne effect is not present in a conventional filled rubber. In this situation, the shear modulus of the filled rubber is not influenced by strain

amplitudes. However, Fig.2 shows that the result is different for common filled rubbers and MREs. This is due to a reduction in the magnetic interaction between the embedded ferromagnetic particles with increased strain amplitude [6]. Hence, the magnetic-field-induced shear modulus is accordingly affected by the strain amplitude.



Fig.2 Dependence of the shear modulus of an MRE on interfacial bond strength at several values of strain amplitude



Fig.3 Dependence of the loss factor of an MRE on interfacial bond strength at several values of strain amplitude

Fig.3 shows the simulation results of the variations of loss factor of MREs with interfacial bond strength for four different strain amplitudes. It can be seen that interfacial bond strength firstly progressively increases the MREs' loss factor up to a maximum and then sharply decreases it to a constant. These results can be explained using energy dissipation methods. The previous research indicated that energy dissipation in interfacial slippage was the predominant factor in determining the loss factor of an MRE [12]. In a Coulomb friction system, the slippage friction

force rises with the increase in bond strength and obviously, the bond strength decreases the interfacial slippage displacement to zero. Therefore, the interfacial energy dissipation, approximately equal to the value of friction multiplied by the slippage displacement, should increase to a maximum before decreasing to zero. Accordingly, the loss factor of MREs follows the same trend as shown in Fig.3. These results indicate that the interfacial bond strength has significant effects on the properties of MREs. Conversely, these effects can be used in the design of MREs. For example, coating the ferromagnetic particle is a method of controlling interfacial layers leading to the development of MREs possessing the desired properties.

3.2 Influence of frequency of loading (f) and dynamic viscosity of the polymer composite (η)

Simulations were carried out for loadings with frequency ranging from 1 to 20 Hz at several values of dynamic viscosity of the polymer composite of 4000, 8000, 12000, 16000 and 20000 Pa · s . Other parameters were set at constants:- $E_0 = 1M$ Pa , $\gamma_0 = 0.1$, $\tau_c = 0.01MPa$ and $H_0 = 50k A/m$.

Fig.4 delineates the simulation results of the variations of shear modulus of MREs with frequency of loading for five different values of dynamic viscosity of the polymer composite. It can be seen that the shear modulus of an MRE rises with the increase in frequency of loading. Fig.4 also shows that the shear modulus of an MRE rises with the increase in the dynamic viscosity of the matrix material, which is a parameter that can be obtained from stress relaxation experimentation on MREs. Fig.5 presents the simulation results of the variation of loss factor of MREs with frequency of loading for five different values of dynamic viscosity of polymer composite. It can be seen that increasing the frequency of loading first increases the loss factor of MREs up to a maximum, before the loss factor progressively declines. The simulation results indicated that the influence of the frequency of loading and the dynamic viscosity of polymer composites on the dynamic behaviour of an MRE obeys the same rules as those for the performance of conventional viscoelastic materials.

Figs.2-5 illustrate that the proposed rheological model is capable of simulating the effect of intrinsic characteristics of MRE and the

dependency of external stimuli. It is thus a useful tool in the determination of the real-time mechanical behaviour of an MRE used in a vibration control system.



Fig.4 Dependence of the shear modulus of an MRE on frequency of loading at several values of dynamic viscosity of polymer composite



Fig.5 Dependence of the loss factor of an MRE on frequency of loading at several values of dynamic viscosity of polymer composite

3.3 Influence of magnetic field strength (H) and the modulus of the polymer composite (E_0)

Simulations were carried out for magnetic field strengths ranging from 0 to50 kA/m at values of modulus for the polymer composite of 0.1, 0.5, 1, 2 and 5 MPa. Other parameters were set at constants:- $\gamma_0 = 0.1$, $\tau_c = 0.01MPa$, $\eta = 8500Pa \cdot s$ and f = 1Hz.

Fig.6 shows the simulation results of the variations of loss factor of MREs with magnetic field strength for five different values of modulus of the polymer composite. It can be seen that the loss factor of the MRE decreases with increases in

magnetic field strength, particularly for MREs with low moduli. This result conforms well to the observed behaviour of these materials. This is so because the magnetic field induces a magnetic attraction between the embedded ferromagnetic particles inside the MRE, which determines the interaction between particles and the surrounding matrix. Therefore, strong magnetic field strength results in a high interfacial pressure. In a strong magnetic field, the high pressure prevents interfacial slippage. Consequently, the loss factor of the MRE is reduced in this situation. Fig.6 also shows that MREs based on matrix materials with small moduli possess large loss factors. The can be explained by the large difference in modulus between the soft polymer matrix and hard iron particles leading to a stress concentration in the interfacial layer allowing interfacial slippage. The softer the matrix is, the larger the difference in modulus and more interfacial slippage can take place. Therefore the loss factor of an MRE is enhanced with a decrease in modulus of the matrix material.



Fig.6 Dependence of the loss factor of an MRE on magnetic field strength at several values of modulus of polymer composite

Fig.7 demonstrates the influence of magnetic field strength on the shear modulus of MREs based on matrices with different values of modulus. This is commonly termed the MR effect and has been extensively researched in the previous decade. To show this MR effect more clearly, some transformation was implemented. The shear modulus without a magnetic field is denoted as G_0 , and the field-induced shear modulus is expressed as ΔG . The relative change in shear modulus of an MRE, calculated by $\Delta G/G_0$, is shown in Fig.8.



Fig.7 Dependence of the shear modulus of an MRE on magnetic field strength at several values of modulus of polymer composite



Fig.8 Dependence of the relative MR effect of an MRE on magnetic field strength at several values of modulus of polymer composite

It can be seen that increasing the magnetic field strength progressively raises the shear modulus of an MRE up to a maximum. This is so because the incremental increase in shear modulus is due to the attraction of magnetized particles. The higher the applied magnetic field, the stronger the attraction and hence the greater the stiffness of an MRE is. When the magnetic field increases to a critical value, the magnetization of particles is saturated and the attraction between magnetisable particles cannot vary with field strength. The field-induced modulus thus reaches a maximum. The results confirmed that the shear modulus of an MRE can be controlled by an external magnetic field which is why an MRE is regarded as a smart material. Also, when comparing different curves in Fig.8, it is found that an MRE with a softer matrix material has a much higher MR effect. This result indicates that to get a high MR effect, it is preferable to use a softer matrix material, such as a room temperature vulcanized silicon rubber or a plasticized natural rubber.

IV. CONCLUSIONS

A rheological model that examines the dynamic mechanical behaviour of MREs has been presented. It modelled the viscoelasticity of the polymer composite, magnetic-field-induced properties and interfacial slippage between the matrix and particles by analogy with a standard linear solid model, a stiffness variable spring and a spring-Coulomb friction slider respectively. The constitutive relationship of the rheological model was derived allowing modelling of hysteresis loops for MREs subjected to a harmonic excitation and the determination of shear modulus and loss factor.

The effectiveness of the model was verified by simulating the properties of MREs possessing a range of intrinsic characteristics and subjected to various external stimuli. The simulation results were consistent with the theories for mechanisms of polymer composites and electromagnetism. The proposed rheological model is capable of predicting the dynamic behaviour of MREs possessing any volume fraction of iron particle and properties of the matrix material. It offers a practical tool in the design and development of MREs. In particular it can be employed as an effective input parameter model in the system control of MRE devices.

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