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INVESTIGATION ON THE NONLINEAR MECHANICAL PERFORMANCE OF NOVEL HYBRID MAGNETORHEOLOGICAL MATERIALS**

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ABSTRACT To improve mechanical performance of magnetorheological elastomers (MRE), two novel hybrid magnetorheological elastomers which were embed with magnetorheological fluids (MRF) and magnetorheological gels (MRG) were fabricated. In this work MRF and MRG were respectively injected into different numbers of holes which were punched on MREs specimens regularly. The nonlinear mechanical properties of magnetorheological fluid-elastomers (MRFE) and magnetorheological gels-elastomers (MRGE) have been evaluated by a modified dynamic mechanical analyzer (DMA) and researched respectively. As experimental results shown, not only the foundation modulus of two novel hybrid magnetorheological materials were higher? than both MRF's and MRG's but also their magnetorheological effects (MR) were better than MRE's. The mechanical properties of the two new hybrids were different with traditional MRE's. Besides, their dynamic properties changed according to the different volume ratio of MRF and MRG which were injected into MRE specimens. A new model based on theory of composite materials was used to explain the nonlinear mechanical properties of the hybrids, and it fit the experimental data well.

KEY WORDS nonlinear mechanical performance, magnetorheological fluid-elastomers, magnetorheological gels-elastomers, magnetorheological effect

I. INTRODUCTION

Magnetorheological (MR) materials are a class of smart materials whose mechanical and magnetic properties can be varied by applying of an external magnetic field[1]. Since the MR effect was discovered by Rabinow in 1948[2,3], MR materials have become a large family with MR fluids, MR foams, MR gel and MR elastomers (MREs) etc.[4]. The most common MR materials are MR fluids (MRFs) which yield stress and apparent viscosity can be changed from Newtonian liquid to non-Newtonian liquid when a magnetic field is applied[5]. MR elastomers (MREs) are a new branch of MR materials. Their mechanical properties, including modulus and damping capability, can be controlled by an external magnetic field. MREs are composed of magnetizable particles (iron particles) and soft rubber-like matrix[6-10]. MREs are divided to as isotropic MREs and anisotropic (chain-like structure) MREs based on different curing conditions. The MR effect of chain-like structure MREs is larger than that of isotropic MREs under the same strength of magnetic field[11-13]. Isotropic magnetic gels, that are gels containing randomly dispersed magnetic particles, are able to react to magnetic field gradients as introduced by Zrinyi's pioneer work[14-15] in the 90's. It was just in 2003 that ferrofluid-based anisotropic magnetic gels with frozen-in magnetic order and able to respond to homogeneous fields was reported[16].

Magnetorheological gels (MRGs), which is a composite fluid of magnetic particles suspended in polymer gels, is known as a new generation of MRFs and used in vibration control and damping devices[19]. These fluids have the advantages of being able to control off-state viscosity and to reduce the settling rate of the magnetic particles in the fluid[20]. MREs are promising for many

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applications, including adaptive tuned vibration absorbers[17], stiffness tunable mounts and suspension[18], and variable impedance surfaces[19-20]. It is important to improve mechanical performance of magnetorheological elastomers, because MREs devices often work in dynamic modes. Here, we report that an improved mechanical performance of MREs can be successfully obtained by injecting MRFs and MRGs into different numbers of holes which were punched on MREs specimens regularly. The nonlinear mechanical properties of magnetorheological fluid-elastomers (MRFE) and magnetorheological gels-elastomers (MRGE) including modulus and damping capability have been evaluated by a modified dynamic mechanical analyzer (DMA), respectively.

II. EXPERIMENTAL

2.1 Sample Preparation

The MRE components include natural rubber (NR), carbonyl iron particles (the average radius is 7 μm) and other additives containing carbon black, ZnO, stearin, MDA (4,4´-Methylenedianiline), sulphur, cz (N-cyclohexyl-2-benzothiazole sulphenamide), plasticizer and carbon black[21]. In samples the mass fraction of the carbonyl iron particles was 60%. The fabrication procedure of MREs was as follows: the iron particles and other additives were firstly mixed into the rubber in a double-roll mill. The resulting material is then compression-molded into a mold in the self-developed Magnet-Heat Coupled Device, which is composed of a magnetic field, a mold, and a controllable heating system, as shown in Fig. 1.

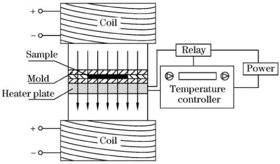


Fig. 1. Principle diagram of PS device.

The mixture is then exposed to the magnetic field and tightly fixed with heat plate. The magnetic field is capable of applying the external magnetic flux density of 0 to 1T over the samples. The heat plate is conterminal with a temperature controller whose temperature can be set in the range from 50 ℃ to 200 ℃. In our experiments, the mixture was exposed to a magnetic field of 800 mT at 160 ℃ for 30 min, after which sample was cured at 160 ℃ for 20 min under a pressure of 1.5 atm. MREs specimens were prepared which was 5 mm thick. Then the beam-like samples (10 mm (L) × 10 mm (W) × 5 mm (T)) which were cut from the MREs specimens, were used to punched holes on. Different numbers of holes which were punched on MRE specimens regularly. And the holes were parallel to the direction of chain-like structure in MREs. Then MRFs and MRGs, the mass fraction of the carbonyl iron particles were both 60%, were respectively injected into holes which were 1.5mm diameter. At last holes were sealed with MREs. Then MRFEs and MRGEs samples were prepared for testing, as shown in Fig. 2.

2.2 Testing

Samples were prepared according to the procedure described above. Then the mechanical properties of samples such as modulus and damping factor were evaluated by using our modified dynamic mechanic analyzer (DMA)[8,9]. The principle diagram of the modified DMA is shown in Fig. 3. This system is composed of two parts: Part A and Part B. Part A is a common DMA (Triton Technology Ltd. UK, Model Tritec 2000B), Part B is a self-developed electromagnet which can generate a variable magnetic flux density up to 1 T. The specimen was set between the driving draft

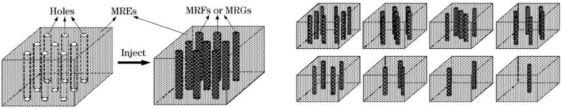


Fig. 2. Schematic diagram of sample geometries and injecting process.

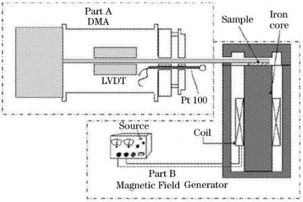


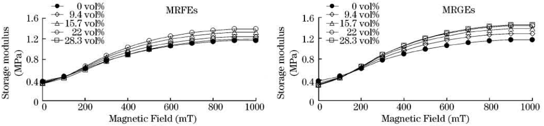
Fig. 3. Sketch and principle diagram of the modified DMA.

of Part A and the magnetic core of Part B. When the draft moves, the specimen was deformed in a shear mode. This system applies a fixed oscillatory strain to the specimen and measures the amplitude and phase of the output force, from which the shear storage modulus G' can be calculated.

III. RESULTS AND DISCUSSION

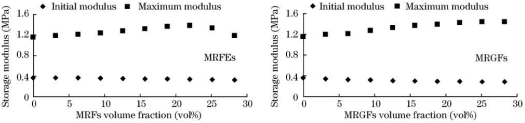
The effect of applied magnetic field on storage modulus of MRFEs and MRGEs were investigated using our modified dynamic mechanic analyzer (DMA). The results of the storage modulus versus different applied magnetic flux densities with different volume ratio of MRFs and MRGs are shown in Fig. 4. The volume fractions of MRFs and MRGs were varied from 0vol% to 28.3vol%. As can be seen from these experimental data, the field-induced modulus G' and G'' of each samples exhibits an increasing trend with increasing magnetic flux density. This is because that with the increase the magnetic field intensity, the average particle-particle interaction increases. which lead to an increase in mechanical stiffness due to hardening of the filler strings with respect to the polymer matrix. However, for the samples which synthesized by injecting MRFs, the modulus firstly increase with increasing MRFs volume fraction before 22vol%, and then decreases with increase of MRFs volume fraction (Fig. 4 (a)). For example, the magneto-induced modulus of MRFEs, $G'_{B=1000\ \mathrm{mT}}$, is 1.16 MPa when the MRFs content is 0vol% compared to 1.39 MPa the MRFs content is 22vol%, and 1. 19 MPa the MRFs content is 28. 3vol%. It is clear that the magneto-induced storage modulus reaches a maximum 1. 39 MPa when the MRFs content is 22vol%. At the same time, modulus of MRGs samples always increase with increasing MRFs volume fraction and magnetic flux density, as show in Fig. 4 (b). For example, the magneto-induced storage modulus of MRGEs, $G'_{B=1000\ \mathrm{mT}}$, is 1.16 MPa when the content is 0vol% compared to 1. 45 MPa the MRGs content is 28. 3vol%.

In order to clearly demonstrate that dependence, the initial modules G_0 (when magnetic flux density is 0), the maximum modules G_{Max} which is the storage modulus when the particles is at magnetic saturation and absolute modulus difference $\Delta G = G - G_0$ are displayed in Fig. 5. The volume fractions of MRFs and MRGs were varied from 0vol% to 28. 3vol%. As shown in Fig. 5 (a)



(a) (b)

Fig. 4. Dependence of storage modulus G' on applied magnetic field for different MRFs (a) and MRGs (b) volume fraction.



(a) (b)

Fig. 5. Increment of initial G' and maximum G' for different MRFs (a) and MRGs (b) volume fraction.

and (b), the initial modules G_0 of both MRFEs and MRGEs samples slowly decrease with increasing the MRFs and MRGs volume fractions. For example, the initial modules G_0 of MRFEs or MRGEs are both 0.37 MPa when the MRFs or MRGs content is 0vol% compared to 0.33 MPa and 0.29 MPa respectively when the MRFs or MRGs content is 28.3vol%. Besides, maximum modules G_{Max} of MRFEs firstly increases until MRFs volume fraction is 22vol%, and then trend of G_{Max} is decreasing with continually increased MRFs volume fraction. But G_{Max} of MRGEs increases with increasing MRGs volume fraction. So it is demonstrated that mechanical properties of MREs can be improved by injecting MRFs or MRGs into it.

The absolute MR effect is a key parameter for evaluating MRE performance. The MR effect is commonly used to characterize the material. MR effect is defined as MReffect $= \Delta G_{\mathrm{Max}}/G_0$, in which G_0 is the initial modulus and $\Delta G_{\mathrm{Max}} = G_{\mathrm{Max}} - G_0$. G_{Max} is the storage modulus when the material is at magnetic saturation (900 mT). In this study, the applied magnetic field is from 0 to 1000 mT. The relation between MR effect and magnetic field for different MRFs and MRGs volume fraction was shown in Fig. 6. Obviously, the MR effects of all kinds of samples show a remarkable change with increasing magnetic flux density and the maximum MR effects of samples without MRFs or MRGs is 216% (1000mT), as shown in Fig. 6 (a) and (b). But trends of absolute maximum MR effects difference ($\Delta\mathrm{MReffect}_{\mathrm{Max}} = \mathrm{MReffect}_{\mathrm{Max}} - \mathrm{MReffect}_{0\mathrm{vol}\%}$) with different MRFs and MRGs volume fraction are dissimilar. As can be seen from Fig. 6 (a), MR effects of MRFEs samples reaches a maximum when MRFs volume fraction of MRFEs samples is 22vol%, and the maximum MR effects is 314%. In the meantime, MR effects of MRGEs samples always increases with increasing MRFs volume fraction and magnetic flux density as shown in Fig. 6 (b), and the maximum MR effects is 399%. It is obviously that MR effects can be remarkably improved by injecting MRFs or MRGs into MREs, and MRGEs samples has higher MR effects than MRFEs when they are in the same volume fraction.

The considered composite is supposed to consist of three transversely anisotropic phases. V_E, V_F and V_G are the volume fractions of the matrix (MREs) and the inclusion phases (MRFs and MRGs), respectively. According to the theory of elastic mechanics, elastic analysis of transversely anisotropic composite can be written as

$$\sigma_{xx}^{i} = \sigma_{xx}^{E} = 0, \quad \sigma_{yy}^{i} = \sigma_{yy}^{E} = 0, \quad \sigma_{xy}^{i} = \sigma_{xy}^{E} \neq 0$$

The stress-strain relationship can be expressed as

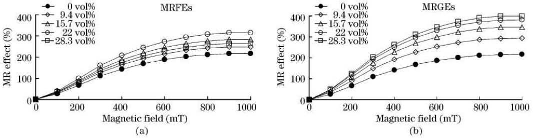


Fig. 6. Dependence of the MR effect on applied magnetic field for different MRFs (a) and MRGs (b) volume fraction.

$$\varepsilon_{xy} = \frac{\sigma_{xy}}{E_{xy}} = V_i\varepsilon^i_{xy} + V_E\varepsilon^E_{xy} = V_i\frac{\sigma^i_{xy}}{E^i_{xy}} + V_E\frac{\sigma^E_{xy}}{E^E_{xy}} = \left(\frac{V_i}{E^i_{xy}} + \frac{V_E}{E^E_{xy}}\right)\sigma_{xy} \tag{1}$$

Thus, the shear modulus of the hybrid magnetorheological elastomers can be get

$$G = \frac{E^E_{xy}}{1 - V_i(1 - E^E_{xy}/E^i_{xy})} \tag{2}$$

$$E^E_{xy} = G^E, \quad E^i_{xy} = G^i$$

G is the effective shear modulus of the hybrid MREs. V_i the volume fractions of the inclusion. G^E and G^i are the shear modulus of MREs and the inclusion (MRFs or MRGs).

$$\gamma = \frac{\tau}{G_s} - \lambda_s \tanh\left(\frac{\tau}{\tau_s}\right) - \left[1 - \tanh\left(\frac{\tau}{\tau_s}\right)\right]\frac{\lambda_s}{M_s}M^2 \tag{3}$$

The model is suitable for MREs based on different rubber matrix. Additionally, this model only depends on three material parameters, initial shear modulus G_0, saturation shear modulus G_s and saturation magnetorheological shear strain λ_s. All of them are easily measured in experiments.

So the effective shear modulus (G^E) of MREs can be got

$$G^E = \frac{\tau}{\gamma} \tag{4}$$

In the preyield region, MR suspension behaves as a viscoelastic solid and therefore the shear below yield stress (at stains on the order of 10^{-3}) is given by[22]

$$\tau = G\gamma \tag{5}$$

According to the research of Ginder[23], the shear modulus of MRFs or MRGs can be written

$$G' \approx 3\phi\mu_0 M_s H \tag{6}$$

ϕ, μ_0, M_s and H are the volume fraction of carbonyl iron particles, the magnetic conductivity, the local saturation of magnetization and Magnetic field intensity, respectively.

Substituting Eq. (4) and Eq. (6) into Eq. (2), a constitutive model for MRFEs or MRGEs can be got. In order to obtain a convenient constitutive model for engineering applications, those constants can be easily determined by the experimental data. And the model results are shown in Fig. 7. It can be seen that the model can accurately predict the initial shear modulus (B = 0) and maximum shear modulus (B = 1000 mT) of MRFEs and MRGEs with different volume ratio of MRFs and MRGs. However, the maximum shear modulus of MRFEs when the MRFs content is more than 22vol%, predicted by the model, do not fit the experimental data very well. It means that the model

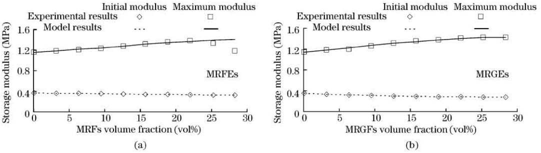


Fig. 7. Increment of initial G' and maximum G' for different MRFs (a) and MRGs (b) volume fraction.

fails to simulate the experimental phenomenon of MRFEs when the MRFs content is more than 22vol% ,which is because MRFs is considered as solid and solid-liquid coupling is not considered. The issue will be researched in our future work.

Ⅳ. CONCLUSIONS

Injecting MRFs or MRGs into different numbers of holes which were punched on MRE specimens regularly provide an easy way to improve the mechanical properties of the magnetorheological materials under external magnetic fields. The mechanical properties such as storage modulus G′ and MR effect for all MRFEs and MRGEs samples exhibit an increasing trend with increasing magnetic flux density. The magneto-induced modulus and MR effect of MRGs samples always increase with increasing MRFs volume fraction, but modulus and MR effect of samples which was injected with MRFs firstly increases with increasing MRFs volume fraction before 22vol% ,and then modulus decreases with continuing increase of MRFs volume fraction. All their mechanical properties, especially MR effect, exhibit significant improvement compared with traditional MREs. A new model based on theory of composite materials was used to explain the nonlinear mechanical properties of the hybrids, and it fit the experimental data well.

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