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STUDY OF MECHANICAL BEHAVIOR AND MICROSTRUCTURE OF MAGNETORHEOLOGICAL ELASTOMERS

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Magnetorheological elastomers' mechanical property is greatly influenced by the microstructure of magnetizable particles embedded in the rubber matrix. When synthesizing magnetorheological elastomers, usually the mixture is cured in the presence of a magnetic field and particles are arranged in chainlike or columnar structure after the crosslink, in order to get remarkable effect controlled by the magnetic field. However curing under the magnetic field will face some problems, *e.g.* the preparation will become not convenient.

In this paper magnetorheological elastomers are prepared in various volume fractions in the absence of a magnetic field. Their dynamic viscoelastic properties are tested by a system designed by ourselves. Their microstructures are observed by scan electronic microscope. Finally the inherent relation is revealed between the magnetoviscoelasticity and the distribution of particles in the matrix as well as the components of the mixture and the chemical technique. A kind of typical microstructure is found to be relative to the magnetoviscoelasticity of magnetorheological elastomers prepared in the absence of a magnetic field. This study also provides a guide when designing and preparing this kind of smart materials.

1 Introduction

Since magnetorheological (MR) effect was discovered in 1948 [1], after a transitory interest, MR materials have nearly been forgotten because of the lack of preparation technology. Recently, with the development of preparation technology, their quick response, good reversibility and controllable performance make them again to be a kind of fascinating smart materials.

MR fluids (MRF) are a kind of colloidal suspension which can change its phase between liquid and solid under the control of magnetic field. It is composed of oil with low permeability and ferrous particles with micrometer size. When MR fluids are exposed to a magnetic field, the ferrous particles are magnetized and attracted by each others to form chains and columns furthermore in the direction of external magnetic field which changes its phase to solid. When it is sheared under a stress above its yield stress, it will show characteristics of liquid phase again and the yield stress is determined by the external magnetic field. When the external field is withdrawn, it will recover its phase from solid to liquid [2]. MRF's unique characteristics make them to be widely used in various devices, such as dampers, clutches, and brakes [3-5].

To make the materials to be more stable, MR elastomers (MRE) want to fix these chainlike and columnar structures in the matrix. They contain micrometer size ferrous particles, which are mixed into polymer media. Then the mixture is cured in the presence of a magnetic field to remain the chainlike and columnar structures in the matrix. When such kind of materials is exposed to an external magnetic field, the elastic modulus will change with the intensity of applied field. These field-dependent characteristics make MRE have promising applications, such as components having changeable modulus [6-8].

However, to fabricate MRE under an external magnetic field limits greatly its application in industry. Although some attempts have been tried to prepare MRE without external field [9], the shear modulus only change about 10%, and the mechanism is also not analyzed. Obviously the chainlike or columnar structures will not exist in the MRE prepared without external magnetic field. So, what makes such kind of MRE still have the field-dependent performance? And how can make their performance reach best state? This paper tries to prepare MRE under nature condition without external magnetic field. Their dynamic performances are measured by a system developed by ourselves. These performances are also compared with their microstructures. The mechanism is found to determine the performance of MRE prepared without external magnetic field. This mechanism is relative to the composing of materials, and gives a guide to fabricate MRE without external magnetic field.

2 Preparation of MR Elastomers and Experimental Setup

To prepare the MR elastomers, carbonyl iron particles with average diameter of about $3\mu m$, 704 RTV silicon rubber and silicone oil are determined. After carbonyl iron particles have been immersed in silicone oil, they are mixed with 704 RTV silicone rubbers. Then the mixture is put into a vacuum case to remove the air bubbles inside it. After cured about 24 hour in room temperature without magnetic field, MR elastomers are prepared.

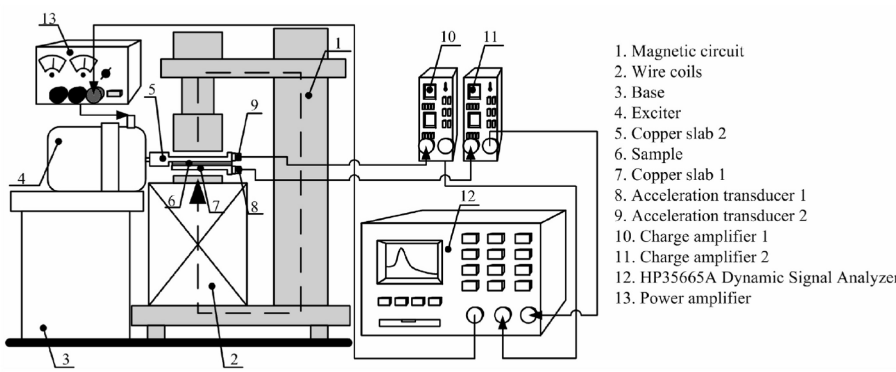


Fig.1 Schematic of the performance testing setup

The setup to study MR elastomers' mechanical performance is shown in Figure (Fig.) 1. The dashed line shows the magnetic route. The magnetic intensity is controlled by the electrical current intensity in the coil. The induced magnetic field is imposed in the direction of sample's thickness. The used sample is about 30mm in length, 10mm in width and 6mm in thickness. Its upper and lower surfaces are adhered to two copper slabs respectively. Then they are inserted into the gap of the magnetic route. Slab 2 is forced to vibrate by an exciter, which is driven by a random signal source from a power amplifier whose signal is provided by a HP35665A Dynamic Signal Analyzer. The frequency of the source ranges from 50Hz to 1000Hz. The vibrations of the two slabs are monitored by two same type of piezoelectric acceleration transducers. The two signals from the transducers are input into two same types of charge amplifiers. Then they are sent to the HP35665A Dynamic Signal Analyzer for processing.

MR elastomers with the lower slab are regarded as a system with a single degree of freedom whose complex stiffness is $k_\tau(1+i\eta)$, where k_τ, η are stiffness and loss factor respectively. And it can be stated that $k_\tau = GA/h$, where A is the area of the sample's shear surface, h is the thickness and G is the shear modulus of MR elastomer. The mechanical parameters of MR elastomers can be obtained from the ratio (*i.e.* transfer function) of the response signal (*i.e.* the lower transducer) to the excitation one (*i.e.* the upper transducer) in the frequency domain. The transfer function can be expressed as

$$T(w) = \frac{K_\tau(1-i\eta)}{-Mw^2 + K_\tau(1+i\eta)} \qquad (1)$$

where M is the effective mass and ω is the angle frequency. Based on equation (1), the mechanical parameters of MR elastomers can be calculated by the data obtained from HP35665A.

To observe the microstructure, a slice of the sample is placed into an environmental scan electronic microscope (SEM). The microscope is XT30 ESEM-TMP, which is made by Philip of Holland. It can observe and analyze samples with their primitive state even containing water or oil. It provides us the intuitionistic images of the samples that illustrate the microstructure of particles in the MR elastomers.

3 Experimental Results

To study the mechanism of MR effect and the best composition of MR elastomers, many kinds of MR elastomers with different percentages of carbonyl iron particles, silicone oil and silicone rubber are prepared. Some of them are shown in table (Tab.) 1. Among them, No.7 and No.3 are the same sample. They are list on both of series 1 (sample 1-4) and series 2 (sample 5-8) in order to compare the samples conveniently.

Table 1 The composing of the samples (weight percentage)

Sample No.	Percentage of carbonyl iron particles	Percentage of silicone oil	Percentage of silicone rubber	Sample No.	Percentage of carbonyl iron particles	Percentage of silicone oil	Percentage of silicone rubber
1	60%	0%	40%	5	20%	20%	60%
2	60%	10%	30%	6	40%	20%	40%
3	60%	20%	20%	7	60%	20%	20%
4	60%	30%	10%	8	70%	20%	10%

Firstly, the sample series 1 is investigated. Their weight percentages of carbonyl iron particles are fixed as 60% and weight percentages of silicone oil are 0%, 10%, 20% and 30%, respectively, the residual percentage is silicone rubber. Their shear moduli (*versus* frequency) when they are at nature state and are exposed to a magnetic field of 200 mT are shown in Fig. 2. The absolute variations of moduli between the two states do not change with frequency. The original and the variation of moduli nearby the frequency of formant are list in Tab. 2.

From Fig. 2 and Tab. 2, it can be found that for samples with fixed percentage of carbonyl iron particles, the shear modulus of MR elastomers exposed to no magnetic field decreases when the percentage of silicone oil increases. When the percentage of silicone oil reaches 20% the modulus has maximum variation of about 26%.

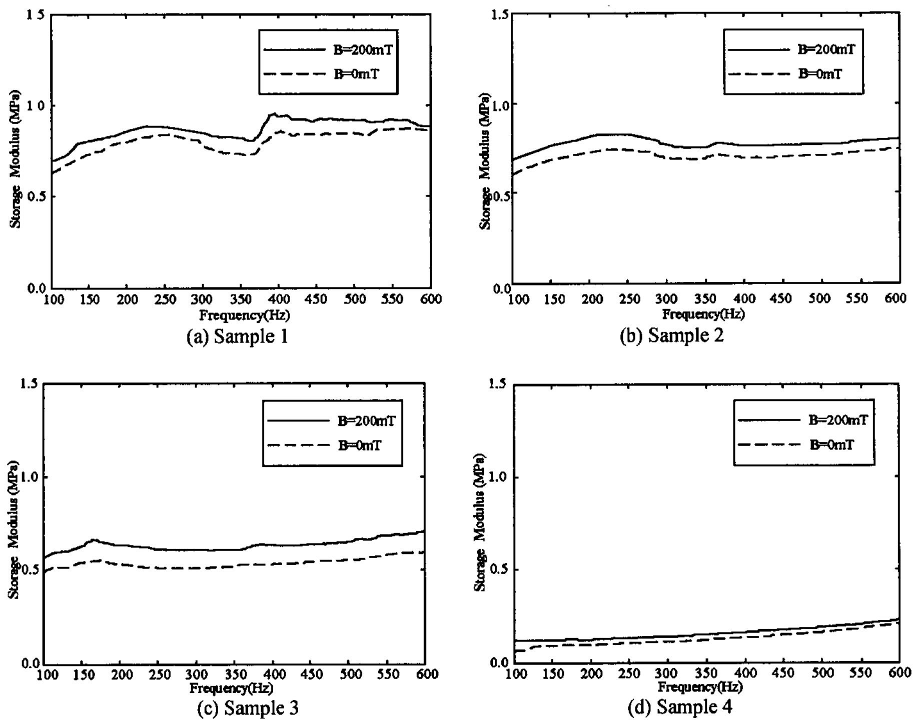
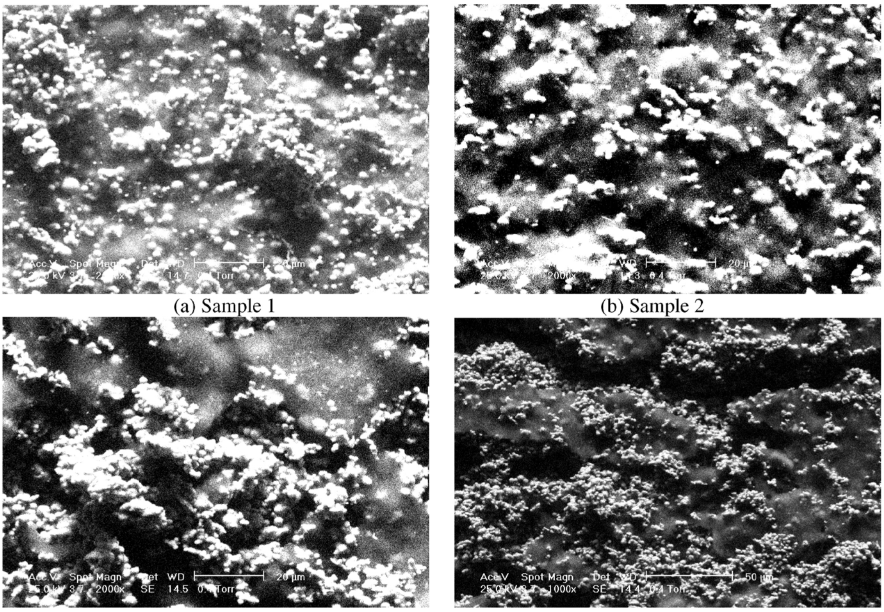


Fig.2 Variation of shear modulus with frequency (B=0 and B=200mT) ---sample series 1

Table 2 Variation of shear modulus of sample series 1

Sample No.	1	2	3	4
G_0	0.85MPa	0.75MPa	0.53MPa	0.15MPa
ΔG	0.06MPa	0.08MPa	0.14MPa	0.03MPa
$\Delta G/G_0$	7%	11%	26%	20%

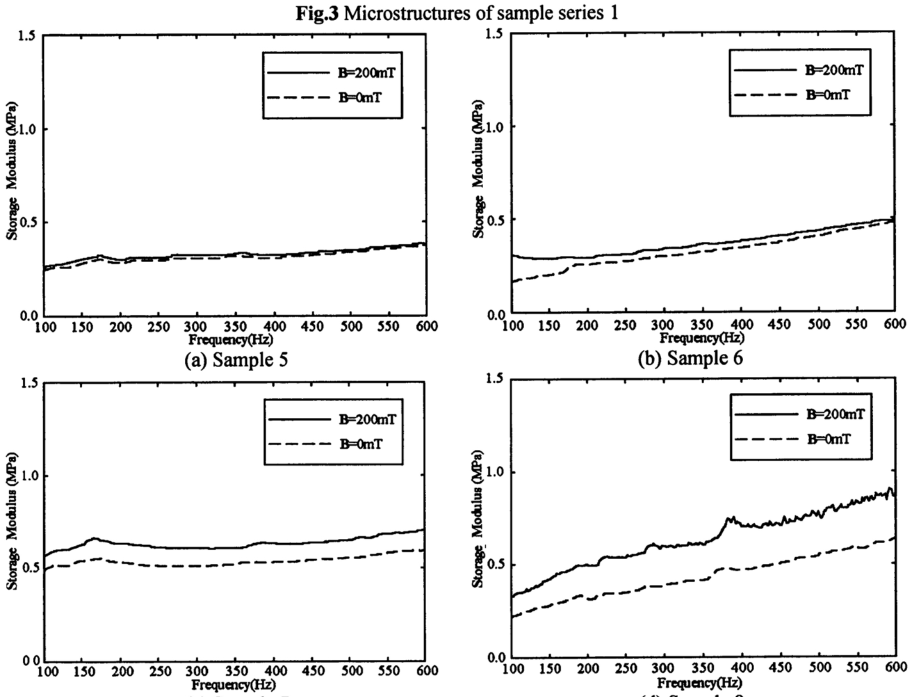
Their microstructure images obtained by SEM are shown in Fig. 3. When there is no silicone oil (sample 1), the particles disperse randomly in the matrix. When there is 20% percentage of silicone oil (sample 3), the particles in the sample can attach each other and self-assemble a partial microstructure like "grape" because of the help of the additive (silicone oil). This kind of microstructure is mainly composed of carbonyl iron particles with silicone oil between their gaps. They are dispersed in the silicone rubber. When the sample is exposed to a magnetic field, the particles in the microstructure are magnetized and move slightly to form more regular structure in the help of the lubrication of silicone oil, which results in high MR effect. When the percentage of silicone oil is larger or smaller than 20%, the samples have smaller MR effect than the sample with 20% silicone oil. This accords to the results of Fig. 3. Fig. 3(b) shows that the microstructures in sample 2 are not as good as those in sample 3. Fig. 3(d) shows that the matrix of sample 4 no longer has rubber's fine construction because it contains too much silicone oil. Therefore, 20% silicone oil gives the best condition to form the self-assembled microstructure and to move slightly when they are exposed to a magnetic field.



(a) Sample 1 (b) Sample 2

(c) Sample 3 (d) Sample 4

Fig.3 Microstructures of sample series 1



(a) Sample 5 (b) Sample 6

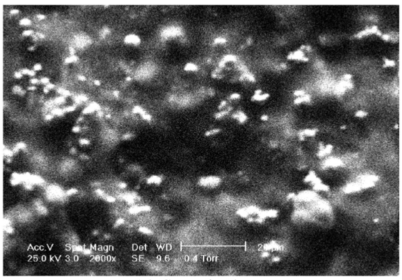
(c) Sample 7 (d) Sample 8

Fig.4 Variation of shear modulus with frequency （B=0 and B=200mT） ---sample series 2

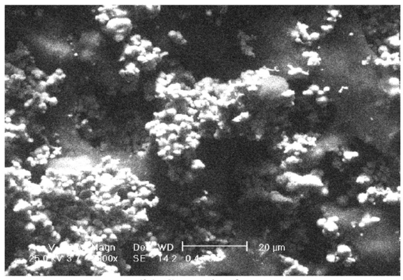
Then, let's investigate the sample series 2. Their weight percentages of silicone oil are fixed as 20% and weight percentages of carbonyl iron particles are 20%, 40%, 60% and 70%, respectively, the residual percentage is silicone rubber. Their shear moduli (*versus* frequency) when they are at nature state and are exposed to a magnetic field of 200 mT are shown in Fig. 4. The variations of moduli between the two states are list in Tab. 3. Their microstructures are shown in Fig. 5.

Table 3 Variation of shear modulus of sample series 1

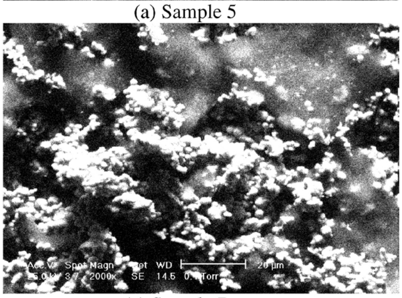
Sample No.	5	6	7	8
G_0	0.29MPa	0.3MPa	0.53MPa	0.33MPa
ΔG	0.01MPa	0.03MPa	0.14MPa	0.17MPa
$\Delta G/G_0$	3%	10%	26%	51%



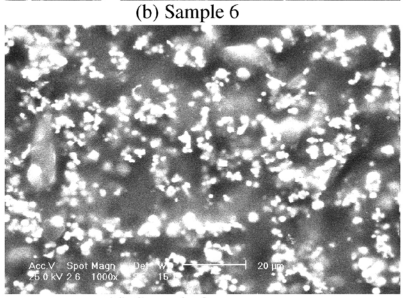
(a) Sample 5



(b) Sample 6



(c) Sample 7



(d) Sample 8

Fig.5 Microstructures of sample series 2

It is similar to the sample series 1 that the absolute variations of moduli do not change with frequency. MR effect increases while the percentage of carbonyl iron particles increases, except that the variation of modulus of sample 4 changes differently with different frequency. When the percentage of carbonyl iron particles is 20% (sample 5), it is difficult to find the self-assembled microstructure in the sample. When the percentage of carbonyl iron particles reaches 40% (sample 6) the self-assembled microstructure can be seen clearly. When the percentage of carbonyl iron particles is 60% (sample 7) particles form the most beautiful self-assembled microstructures. In sample 8, the percentage of silicone rubber is lack because there is too much carbonyl iron particles. The lack of rubber makes the sample become too floppy to keep the toughness of rubber. So the best percentage of carbonyl iron particle is about 60%.

4 Conclusions

MR elastomers are fabricated under nature condition without external magnetic field. Their performances and the microstructures are also studied. It has been found that the construction in such MR elastomers is no longer chains or columns of particles. By the help of additive, such as silicone oil, the particles form a kind of self-assembled microstructure in MR elastomers. This kind of microstructures is composed by particles and additive, they are dispersed randomly in the matrix. When this kind of MR elastomers is exposed to a magnetic field, the particles in the microstructure are magnetized and move slightly by the lubrication of additive to form regular construction, which results in great MR effect. The key technique to improve the performance of MR elastomers prepared without magnetic field is how to form the partial self-assembled microstructure. Experiments show that when preparing such MR elastomers by using carbonyl iron particles, silicone rubber and silicone oil, the MR elastomers have best MR effect when their percentages are 60%, 20% and 20%, respectively.

Further work will be done to validate this kind of microstructure, to observe the formation process of the microstructure and to observe the shift of particles in the structures when they are exposed to a magnet field.

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