

# Magnetorheological Elastomers Based on Isobutylene–Isoprene Rubber

**Yinling Wang, Yuan Hu**

*State Key Laboratory of Fire Science, University of Science and Technology of China, Hefei 230026, People's Republic of China*

**Yinling Wang, Huaxia Deng, Xinglong Gong, Peiqiang Zhang**

*CAS Key Laboratory of Mechanical Behavior and Design of Materials, Department of Mechanics and Mechanical Engineering, University of Science and Technology of China, Hefei 230027, People's Republic of China*

**Wanquan Jiang, Zuyao Chen**

*Department of Chemistry, University of Science and Technology of China, Hefei 230026, People's Republic of China*

**Magnetorheological (MR) elastomers are a group of smart materials whose modulus can be controlled by the application of an external magnetic field. In this paper, MR elastomers based on isobutylene–isoprene rubber were prepared by the common manufacturing procedure of rubber and the corresponding MR effect, mechanical properties, and thermal stability were investigated. The results showed that MR effect varied with the volume content of iron particles and a maximum of 20% in MR effect was obtained at 15 vol% of iron particles. The relationship between MR effect and microstructure was discussed in detail. Mechanical tests showed that iron particles could improve the tensile strength and hardness. However, compared with carbon black with the same volume content, the reinforcing effect was far worse. TG analysis showed the thermal stability of isobutylene–isoprene rubber was improved by incorporation of iron particles. POLYM. ENG. SCI., 46:264–268, 2006. © 2006 Society of Plastics Engineers**

## INTRODUCTION

Magnetorheological (MR) materials are a group of smart materials whose rheological properties can be controlled by the application of an external magnetic field. Up to now two main kinds of MR materials have been reported, MR fluids

and MR elastomers. MR fluids are well-known smart materials, which exhibit Newtonian-like behavior in the absence of a magnetic field and become a weak viscoelastic solid with a certain yield stress when applying a magnetic field [1]. However, the settlement of magnetic particles has been a serious problem that affects the performance of MR fluids.

MR elastomers can be thought of as a new generation of MR materials that has distinct properties from those of MR fluids. They have controllable field-dependent modulus while MR fluids have a field-dependent yield stress. The obvious advantage of MR elastomers is that the particles are not able to settle with time and then have stable MR performance. MR elastomers also have a sensitive response to magnetic fields and the time of response of MR elastomers is less than 10 ms [2]. MR elastomers hold promise in adaptive tuned vibration absorbers, stiffness tunable mounts, automobile suspensions [2], and artificial muscles [3].

MR elastomers are mainly composed of magnetic particles and elastic polymer matrix. The magnetic particles used are usually carbonyl iron particles and the reported polymer matrix included soft silicone elastomers [4–6], poly (vinyl alcohol) [7], gelatin [8], hard natural rubber [9], and RTV polyurethane sealant [10]. In general, magnetic particles are first embedded in the uncured polymer and then the mixture is cured under a strong magnetic field. Thus, the magnetic particles form special chainlike structure in the direction of the magnetic field, which results in the controllable shear modulus with the magnetic fields. This kind of MR elastomers can be called anisotropic MR elastomers. Up to now, the reported best relative effect for MR elastomers based on silicon rubber can be up to 60% [11]; however, their me-

*Correspondence to:* Y. Hu; e-mail: yuanhu@ustc.edu.cn; X. L. Gong; e-mail: gongxl@ustc.edu.cn

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chanical properties are poor. And that of the MR elastomers based on natural rubber is about 30% and the absolute MR effect is  $\sim 0.7$  MPa at a magnetic field of 0.8 T [9]. But the typical needed magnetic field is very strong at about  $8 \times 10^6$  A/m [5], which makes its manufacture complicated and difficult to be broadly applied.

Recently, Lokander and Stenberg [12, 13] studied the isotropic magnetorheological rubbers that are prepared without applying magnetic fields. They found that isotropic magnetorheological elastomers using large irregularly shaped iron particles had a large absolute MR effect that is about 0.4 MPa at 0.24 T. In the previous work, the new MR elastomers based on PU/Si-rubber hybrid were studied [14]. The maximum increase in shear modulus of this kind of MR elastomers can be up to 0.5 MPa at 0.2 T and the relative MR effect is higher than that reported in the literature.

Isobutylene–isoprene rubber (IIR) has good thermal and oxidative stability, chemical resistance, and high damping and has been applied broadly, especially in vibration absorbers. To make practical MR elastomers useable for adaptive tuned vibration absorbers, in this paper, the isotropic MR elastomers are prepared using IIR as the matrix. The MR effect, mechanical properties, and thermal properties of the MR elastomers are studied here in detail.

## EXPERIMENTAL

### Materials

The used spherical carbonyl iron particles are commercial FTF-4 type with the size range of 3–5  $\mu\text{m}$  bought from Hebao Nanomaterial. IIR, carbon black (N660) and the other corresponding additives were provided by Grandtour Tire (Anhui).

### Sample Preparation

Magnetorheological (MR) elastomers based on isobutylene–isoprene rubber (IIR) with different volume contents of carbonyl iron particles were prepared by the common manufacturing procedure of rubber. Carbonyl iron particles were mixed into the rubber together with vulcanization system in a two-roll mixer. Then the mixtures were vulcanized at 175°C for 10 min. To compare the mechanical properties, a series of samples containing carbon black instead of iron particles with the same volume content were also prepared under the same conditions.

### Mechanical Measurements

Tensile tests were performed on a Universal testing machine (WD-5D, Changchun, China) with a crosshead speed of 50 mm/min at 25°C. The average of five tests was reported here.

The magnetorheological effect was evaluated by measuring the dynamic shear modulus with and without an applied magnetic field. The schematic equipment set-up and the principle of the tests can be seen in [15].

The magnetic field was made by electromagnet, and the magnetic induction through samples was kept at 0.24 T. The samples tested were  $30 \times 10 \times 2$  mm<sup>3</sup> and sandwiched between a brass plate and an aluminum plate.

The morphologies of MR elastomers with different volume contents of carbonyl iron particles were observed using an XL30 ESEM at an accelerating voltage of 10 kV. All the samples were coated with a thin layer of gold before SEM observations.

The electrical resistance of MR elastomers was measured using plate electrode of ZC-36-type high resistance meter and the corresponding resistivity was calculated according to the instructions. For resistance measurements, the quadrate samples with dimensions of  $10 \times 10 \times 2$  mm<sup>3</sup> were prepared.

The samples were analyzed by thermogravimetric analysis (TGA), using a Netzsch STA-409c thermal analyzer under air flow from 25 to 600°C at the rate of 10°C/min.

## RESULTS AND DISCUSSION

### Magnetorheological Properties and Microstructure

Figure 1 shows the relative MR effect of MR elastomers with different volume contents of iron particles. Here, the relative MR effect meant the relative change of shear modulus of MR elastomers. It can be seen that the MR effect increases in the beginning and decreases when the content of iron particles is 15%.

It was considered that for isotropic MR elastomers, the change of shear modulus was due to the deformation of MR elastomers under magnetic fields. And the deformation was the accumulative results of that of every part of matrix resulted from the dragging of iron particles to the direction of magnetic fields. When the content of iron particles was less, the accumulative results were less which resulted in the small MR effect. With the increase of iron particles, the MR effect would improve accordingly. However, up to a certain content of iron particles a network of iron particles formed in the matrix. The modulus mainly depended on the network and the potential of deformation under magnetic fields was little with little content of IIR matrix and contributed little to the change of modulus. So, the MR effect would decrease at a certain content of iron particles.

Figure 2 shows the microstructure of MR elastomers based on IIR with different contents of iron particles. It can be seen that when the content of iron particles surpassed 15%, a network of iron particles had formed in the matrix. To further prove the existence of the network of iron particles, the volume resistivity of MR elastomers with different contents of iron particles was tested in Fig. 3. After 15 vol%, there was a rapid decrease of volume resistivity that indicated the formation of a connected network throughout the bulk of the rubber matrix. So, according to the assumption mentioned earlier, the trend of MR effect changing with the content of iron particles was understandable.

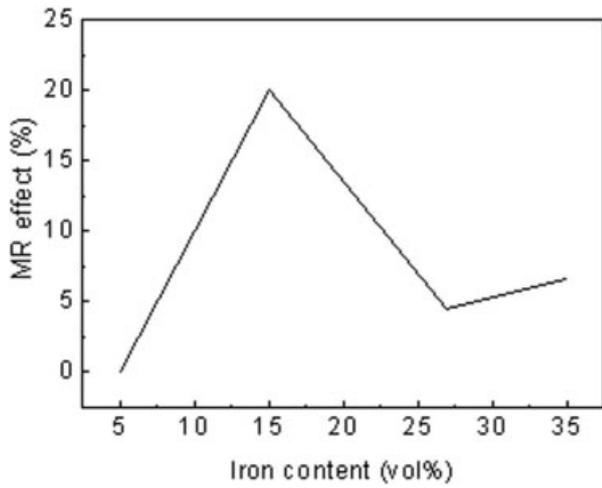


FIG. 1. MR effect of MR elastomers with different volume contents of iron particles.

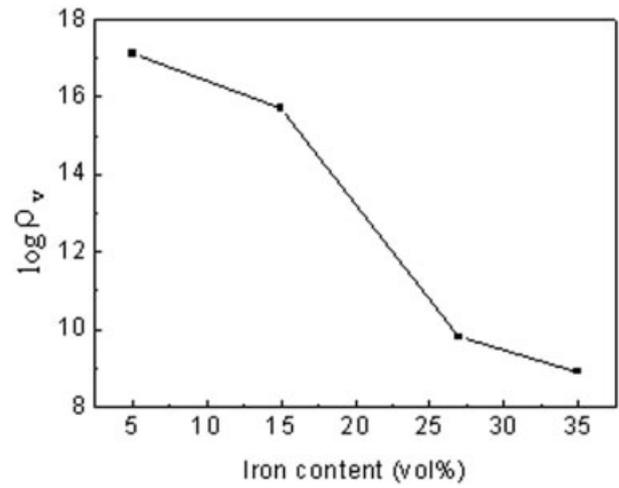


FIG. 3. Volume resistivity as a function of the volume content of iron particles.

### Mechanical Properties

The mechanical properties including tensile strength, hardness, and elasticity of MR elastomers with different

contents of iron particles were studied and compared with the same content of carbon black. Figure 4 shows the variation of tensile strength with different contents of fillers.

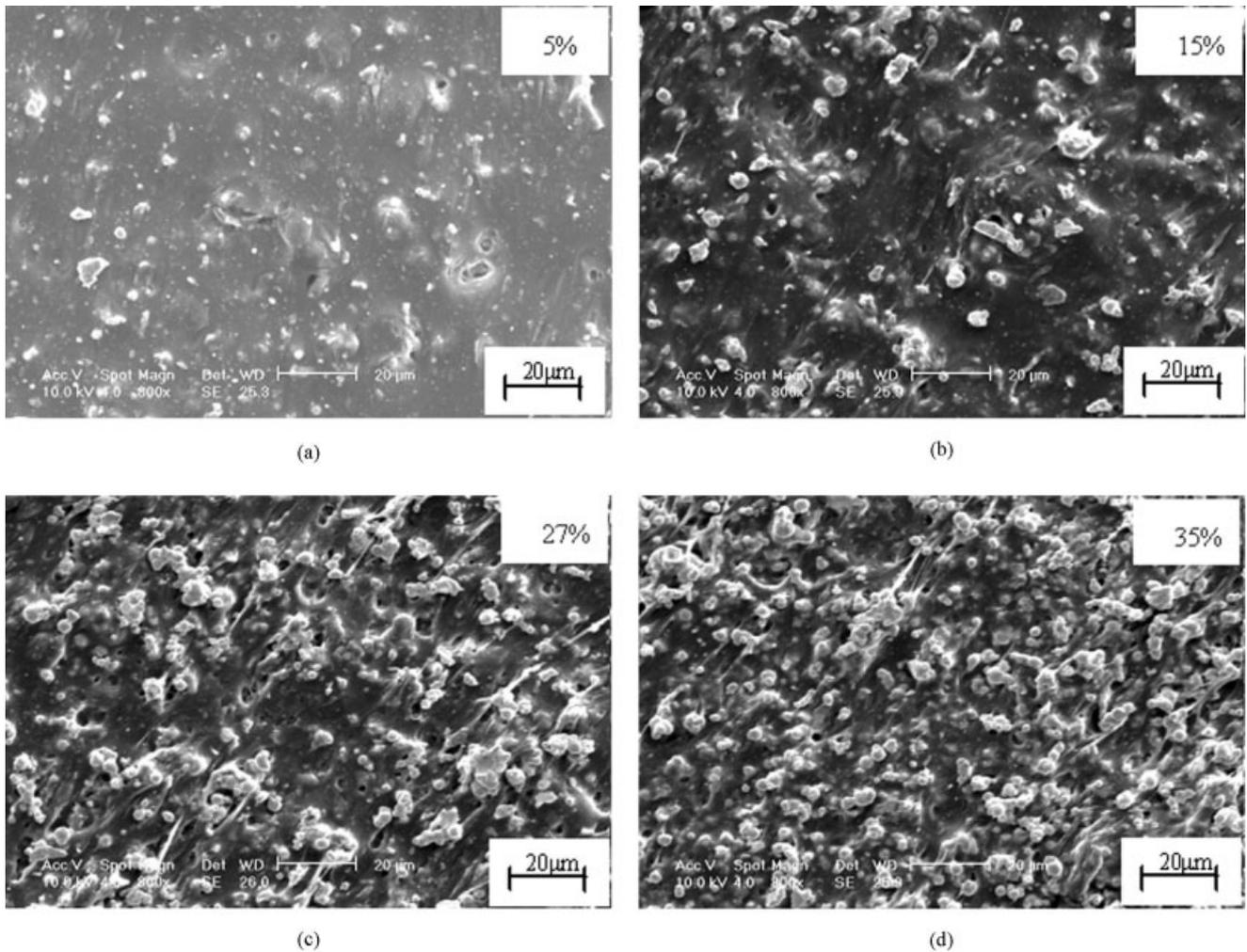


FIG. 2. Microstructure of MR elastomers with different contents of iron particles.

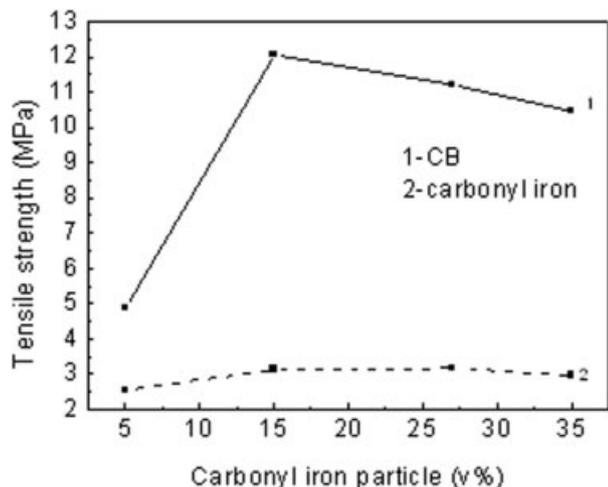


FIG. 4. Tensile strength as a function of the content of CB (carbon black) (1) and iron particles (2).

It can be seen that the tensile strength of MR elastomers increased with the increase of iron particles until the content of 27%. However, their tensile strength was far smaller compared with those with the same content of carbon black. At 15 vol% of fillers, the tensile strength of IIR containing carbon black was about four times of that of MR elastomers. This might be due to the larger size of carbonyl iron particles and the weak interaction with the matrix for their polar surface properties. The weak interaction with the matrix can be seen from the microstructure in Fig. 2. The gaps between iron particles and matrix can be seen clearly. So, it can be concluded that if MR elastomers were to be put into application, the reinforcement of the material should be considered.

Figure 5 shows the variation of elongation at break with the content of filler. It can be seen that the elongation at break of IIR filled with carbon black had a maximum at 15 vol%, but that of MR elastomers increased always with the

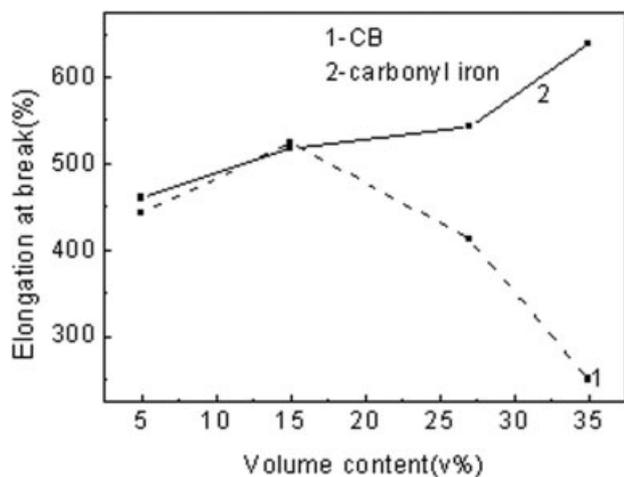


FIG. 5. Elongation at break as a function of the content of CB (1) and iron particles (2).

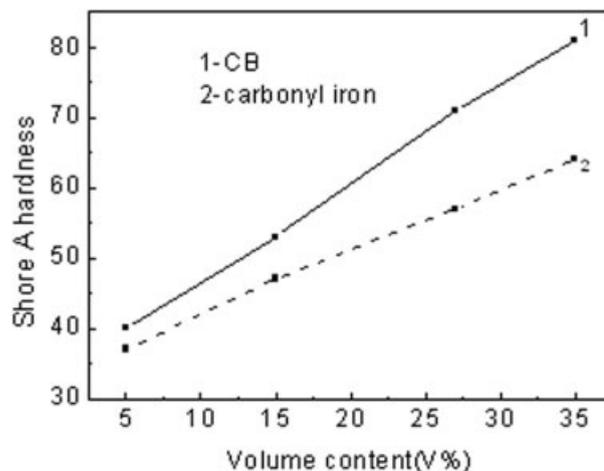


FIG. 6. Shore A hardness as a function of the content of CB (1) and iron particles (2).

content of iron particles and the elongation was larger at the same volume content. This further proved that the interaction between iron particles and IIR matrix was weak and could not prevent the movement of molecular chains.

Figure 6 shows the dependence of shore hardness on the filler content. The curve reveals that the hardness increases almost linearly with the content of fillers and the degree of increase of hardness of MR elastomers is still smaller than that of IIR containing carbon black.

Figure 7 shows the variation of elasticity with the content of fillers. It can be seen that the elasticity of IIR is low and it decreases with the content of fillers. And at the same volume content, the value of elasticity of the MR elastomers was almost the same with that of IIR elastomers containing carbon black. It can be deduced that the elasticity mainly depended on the content of IIR matrix and had less relationship with the size and properties of fillers.

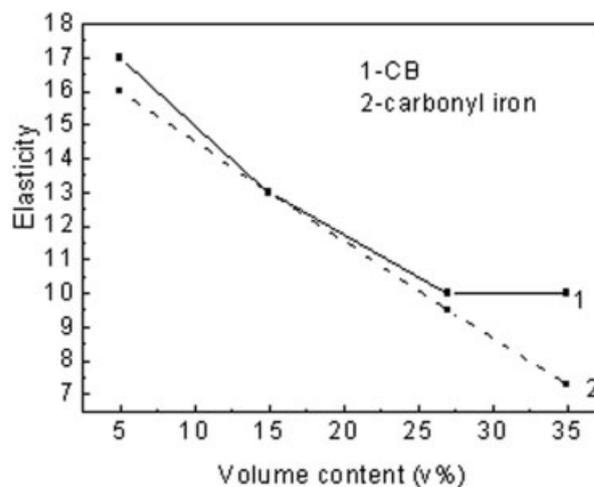


FIG. 7. Elasticity as a function of content of CB (1) and iron particles (2).

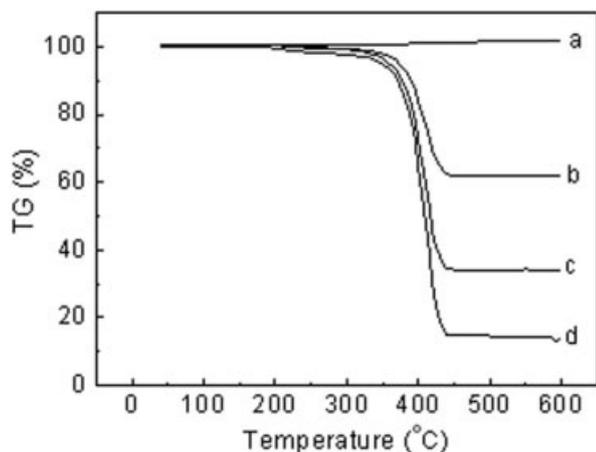


FIG. 8. TG analysis of (a) pure Fe (b) MR elastomers with 15% vol Fe (c) MR elastomers with 5% vol Fe (d) IIR filled with 5% vol CB.

### Thermal Properties

Thermal stability is an important factor which affects the service life of rubber. The effect of iron particles on the thermal properties of IIR was investigated by TG analysis and the result can be seen in Fig. 8.

Figure 8 shows that the mass of pure iron particles was kept constant within tested temperature in  $N_2$  atmosphere, and so the mass loss of MR elastomers was entirely due to the thermal degradation of IIR. The temperature at 5% mass loss of IIR of MR elastomers with different contents of iron particles and that of samples for comparison are listed in Table 1. It can be seen that the temperature increases with the sequence 5% CB < 5% Fe < 15% Fe, which indicates that IIR containing iron particles had better thermal properties compared with IIR filled with carbon black and the thermal stability was improved by increase of iron particles.

### CONCLUSIONS

MR elastomers based on IIR rubber were prepared by the common manufacturing procedure of rubber. The MR effect varied with the content of iron particles and a maximum of 20% in MR effect at 15 vol% was obtained. The relationship between MR effect and microstructure was discussed in detail.

The mechanical properties including tensile strength,

TABLE 1. Temperature at 5% mass loss ( $^{\circ}C$ ).

Sample	Temperature at 5% mass loss
5%CB	347.4
5%Fe	359.7
15% Fe	375.4

hardness, and elasticity were also studied. The results showed that tensile strength increased with the content of iron particles; however, the tensile strength was far lower than that of IIR filled with the same content of carbon black due to their larger size and weak interaction with the matrix. The hardness increased with the content of iron particles and the elasticity decreased with the content of iron particles.

TGA measurements suggested that the thermal stability of MR elastomers based on IIR can be improved by increasing the content of iron particles.

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