Study of the Physical Properties of Carbonyl Iron Particles-Oriented Magneto-Rheological Elastomer†

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The physical properties of an anisotropic magneto-rheological elastomer were examined. The magneto-responsive particles were incorporated into a natural rubber matrix at a volume fraction of 0, 10, 20, 30, 40 and 50 vol. %. The magneto-rheological elastomers were prepared using a newly proposed neodymium magnet inserted into the mold to align the particles efficiently. The tensile strength of the anisotropic magneto-rheological elastomer decreased abruptly, whereas the hardness was higher than that of isotropic magneto-rheological elastomer. At an input current of 3A, magneto-rheological elastomer containing 30 vol. % magneto-responsive particle showed the maximum magneto-rheological effect, which was explained by the ratio of the shear modulus of anisotropic magneto-rheological elastomer with that of isotropic magneto-rheological elastomer and the ratio of the shear modulus shift was 59 %. Based on these results, the orientation of magneto-responsive particle can affect the damping properties of magneto-rheological elastomer.

Key Words: Magneto-rheological elastomer, Natural rubber, Anisotropic magneto-rheological elastomer.

INTRODUCTION

Rubber vibration isolators are used for a range of purposes and play a key role in the automotive industry. Because these conventional isolators only absorb a restricted range of frequencies due to their inherent modulus, efficient control of wide-ranging vibrations is difficult. For this reason, a number of studies have been performed and a magneto-rheological elastomer (MRE) was suggested to be one of the best solutions for this problem¹.

Magneto-rheological elastomer is a composite, in which the rubber matrix is filled with magneto-responsive particles (MRPs). Therefore, the modulus of the composite can be varied according to the applied magnetic field. These unique phenomena can help reveal the chain like formation of filled magneto-responsive particle along with the direction of the applied magnetic field²⁻⁴. If the magnetic field is applied to a composite before curing the matrix, randomly dispersed magneto-responsive particle in the matrix can be arranged along the direction of the magnetic field. This can result in a broader absorbable frequency range. The magneto-rheological fluid, whose matrix is viscous fluid, is another modulus variable material similar to magneto-rheological elastomer. On the other hand, the relatively lower modulus of the fluid matrix, the settlement of fine magneto-responsive particle and the need for a fluid

container can be problematic. These problems can be solved by replacing the viscous fluid matrix with an elastic solid matrix. The matrix material, magneto-responsive particle concentration and magnetic flux density are factors that affect the modulus in magneto-rheological elastomer. Most studies on magneto-rheological elastomer, however, are restricted to isotropic magneto-rheological elastomer (i-MRE) cured without pre-orienting the particles after compounding²⁻⁷.

This paper proposes a newly designed mold to induce the pre-orientation of magneto-responsive particle. Anisotropic magneto-rheological elastomer (a-MRE) containing pre-oriented magneto-responsive particle could be produced by exposure to magnetic fields prior to curing. Natural rubber was chosen as the elastomeric matrix and micro sized magneto-responsive particle was used as the filler with various loads to examine the magneto-rheological effect and mechanical properties of the magneto-rheological elastomer. The magneto-rheological properties of the magneto-rheological elastomer were examined by applying a range of external magnetic fields to the magneto-rheological elastomers.

EXPERIMENTAL

Natural rubber (NR, RSS#1, Malaysia) was used as the matrix of the magneto-rheological elastomer. The structure of the carbonyl iron particles (s-1641, international specialty

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Products, US), which were used as magneto-responsive particles (MRPs), was spherical and the mean diameter of the particles was 3–5 μm . The loading was 0, 10, 20, 30, 40 and 50 vol %. For the vulcanization of the natural rubber matrix, 2.5 phr of sulfur, 5 phr of ZnO, 2 phr of stearic acid and 0.8 phr of sulfenamide were used.

Compounding was processed using an open-roll mill at room temperature. After mastication of natural rubber, ZnO and stearic acid were added to the natural rubber matrix followed by the addition of magneto-responsive particle. The sulfur was added after mixing and dispersing the magneto-responsive particle to prevent scorching of the natural rubber matrix during the compounding process. The final magneto-rheological elastomer compound was stored at room temperature for 24 h for stabilization prior to use.

Preparation of magneto-rheological elastomer specimen: Previous studies to produce magneto-responsive magneto-rheological elastomer reported several problems⁸. First, the orientation of magneto-responsive particle disappeared when vulcanization was performed in a hydraulic hot press. The high pressure of the hydraulic hot press broke down and rearranged the magneto-responsive particle, which had been oriented in a rubber compound between neodymium (Nd) magnets. Second, many of the bubbles rose from the magneto-rheological elastomer specimen when the bumping process was skipped to reduce the re-orientation of the magneto-responsive particle. A newly designed mold with a Nd magnet inserted was used to improve these phenomena, as shown in Fig. 1. The new mold consisted of an aluminum body to protect the Nd magnet from the high pressure of the hydraulic hot press and the Nd magnet was used to induce the orientation of magneto-rheological elastomer during the curing process. The dimensions of a 3 mm thick aluminum-coated Nd magnet were 600 mm $\times$ 120 mm $\times$ 350 mm. The surface magnet density was 0.6 Tesla.

In this study, two types of magneto-rheological elastomer were prepared for comparison. The isotropic magneto-rheological elastomer (i-MRE) was prepared by the homogenous distribution of magneto-responsive particle in the matrix. The curing characteristics of the compound were measured using a rubber rheometer (DRM-100, Daekyung engineering, Korea). Vulcanization was carried out using a hydraulic hot press at 160 $^{\circ}\text{C}$ and 15 kPa for 90 % of the measured cure time (t_{90}) of the compound. Fig. 1 shows the anisotropic magneto-rheological elastomer (a-MRE) prepared with a Nd magnet inserted mold. The magneto-rheological elastomer sheet and mold were pretreated at 80 $^{\circ}\text{C}$ for 0.5 h. The orientation of magneto-responsive particle was expected to take place during pretreatment. Finally, the magneto-rheological elastomer was cured at 160 $^{\circ}\text{C}$ for 0.5 h to modify the orientation of the magneto-responsive particle.

Mechanical properties and morphology of the magneto-rheological elastomer: The tensile properties of the magneto-rheological elastomer were examined at room temperature without an external magnetic field using a universal testing machine (DUT-501, Daekyung engineering, Korea). The gauge length and crosshead speed was 20 mm and 500 mm/min, respectively. The hardness of the magneto-rheological

elastomer with different compositions was measured using a Shore A durometer. The morphology of i-MRE and a-MRE was examined by scanning electron microscopy (JSM-5200, JEOL, Japan).

Fig. 1. Neodymium magnet inserted mold : (a) design drawing, (b) inside of real mold

Measurements of the magneto-rheological effect: The magneto-rheological (MR) effect was examined using a self-modified fast Fourier transform analyzer. **Scheme-I** presents the structure of the fast Fourier transform analyzer. The electro-magnet applied fast Fourier transform analyzer was equipped with a fixed end beam for stable measurements. The lower accelerator measured the vibration generated from the base. The upper accelerator measured the vibration transferred

from the modulus variable magneto-rheological elastomer using a magnetic flux generator. The oscillator gave a shear force on the magneto-rheological elastomer as created by inertia.

Scheme-I: Self-modified fast Fourier transform analyzer for measurement of shear modulus of magneto-rheological elastomer specimen

To examine the shear modulus of the magneto-rheological elastomer due to the induced current, white noise excitation applied to the entire system was generated by a shaker beneath the fixed-fixed end beam. When the entire system was excited, the magneto-rheological elastomer connecting the magnetic flux generator and oscillator was deformed in the shear direction. The shift in the natural frequency of the oscillator occurred by changing the shear stiffness of the magneto-rheological elastomer. The measured signals were sent to the fast Fourier transform analyzer. The transfer function could be obtained using a fast Fourier transform analyzer. After identifying the natural frequency of the oscillator in the transfer function, the frequency was substituted into the equation of the relationship between the shear modulus and frequency. The shear modulus of the magneto-rheological elastomer due to the induced current could be obtained using this process.

RESULTS AND DISCUSSION

Morphology of the magneto-rheological elastomer: The morphology of the magneto-rheological elastomers was examined by SEM (Fig. 2). Fig. 2a shows the morphology of i-MRE containing 30 vol % magneto-responsive particle dispersed randomly in the natural rubber matrix. i-MRE showed no deformation of magneto-responsive particle, whereas a-MRE presented an apparent orientation (Fig. 2b). The orientation of the magneto-responsive particle was maintained during the curing process of the magneto-rheological elastomer, even though the bumping process was used to remove the bubbles generated. The reason was that the magnetic field was applied continuously during the bumping process. Therefore, it was expected that the a-MRE would show the best magneto-rheological effect.

Mechanical properties of the magneto-rheological elastomer: Fig. 3 and Table-1 present the tensile properties of i-MRE and a-MRE at various loadings. In case of the i-MRE, the tensile strength and elongation at break decreased dramatically when more than 30 vol. % magneto-responsive particle was added, whereas they were slightly lower when up to

Fig. 2. Morphology of magneto-rheological elastomers with the type of manufacturing process: (a) i-MRE, (b) a-MRE

30 vol. % magneto-responsive particle was added. Both i-MRE and a-MRE showed a much greater decrease in tensile properties when more than 40 vol % magneto-responsive particle was added. This suggests that the mechanical properties of magneto-rheological elastomer decreased with increasing amounts of magneto-responsive particle due to a decrease in miscibility between the organic matrix and inorganic magneto-responsive particle. In the case of a-MRE, the tensile strength of a-MRE decreased with increasing magneto-responsive particle content. This was attributed to the orientation of magneto-responsive particle. **Scheme-II** shows the direction of the orientation of magneto-responsive particle on a-MRE specimen. In contrast to i-MRE, the orientation of magneto-responsive particle in a-MRE was perpendicular to the direction of the tensile direction. Therefore, it is believed that the lower tensile strength of a-MRE would result in a higher orientation of magneto-responsive particle in a-MRE. A comparison of the data in Fig. 3 and Table-1 showed that the tensile strength of a-MRE was lower than that of i-MRE and the two materials showed a different trend with the magneto-responsive particle loading.

Fig. 3. Tensile properties of MRE with the amounts of magneto-responsive particles : (a) Tensile strength, (b) Elongation at break

Type of MRE	Vol. % of particles	0	10	20	30	40	50
i-MRE	Tensile strength (MPa)	25.9	25.4	23.4	23.8	16.0	10.8
	Elongation at break (%)	685	695	673	643	597	540
a-MRE	Tensile strength (MPa)	25.9	8.7	5.2	5.9	5.4	3.8
	Elongation at break (%)	685	513	477	461	418	294

Scheme-II: Schematic diagram for the direction of particle orientation and test

Table-2 (Fig. 4) lists the change in the magneto-rheological elastomer hardness with the addition of magneto-responsive particle. The hardness of magneto-rheological elastomer linearly increased with increasing magneto-responsive particle content regardless of the type of magneto-rheological elastomer. The hardness of a-MRE was higher than that of i-MRE when both magneto-rheological elastomers containing the same amounts of magneto-rheological elastomer were compared. The higher hardness of a-MRE than i-MRE was attributed to the measured direction of hardness. The hardness was measured along the orientation direction of magneto-responsive particle. In addition to the results of the tensile strength, the hardness of the magneto-rheological elastomer is an important estimating the orientation of the magneto-responsive particle.

Magneto-rheological effect of the magneto-rheological elastomer: When the external magnetic field applies to magneto-rheological elastomer, the orientation of the magneto-responsive particle affects the modulus of the magneto-rheological elastomer. The change in modulus is the so called magneto-rheological (MR) effect and can be explained by eq. 1.

$$\text{MR effect (\%)} = \frac{\Delta G}{G_i} \times 100 \quad (1)$$

where G_i indicates the initial shear modulus without a magnetic field and ΔG means the change in initial modulus when the magnetic field is applied to the magneto-rheological elastomer. Therefore, a higher magneto-rheological effect means a higher performance of magneto-rheological elastomer. This study introduced a new method, as explained by eq. 2, to compare a-MRE with i-MRE.

$$\text{Modified MR effect (\%)} = \frac{(G_{a\text{-MRE}} - G_{i\text{-MRE}})}{G_{i\text{-MRE}}} \times 100 \quad (2)$$

where $G_{i\text{-MRE}}$ means the shear modulus of i-MRE in a magnetic field and $G_{a\text{-MRE}}$ means that of a-MRE in the field at a certain input current.

Vol. % of particles	0	10	20	30	40	50
i-MRE	36	38	41	45	47	53
a-MRE	36	40	45	53	60	60

Fig. 4. Hardness of magneto-rheological elastomer with the amounts of magneto-responsive particles

The magneto-rheological effect was measured using a fast Fourier transform analyzer described in **Scheme-I** and the input current was 0~3 A. Fig. 5 shows the effect of the input current on the change in the shear modulus of i-MRE and a-MRE containing 0~50 vol % of magneto-responsible particle. Most specimens showed that magnetic saturation occurs after an input current of 2A. The shear moduli of both i-MRE and a-MRE at a certain input current increased with increasing amount of magneto-responsible particle. Magneto-responsible particle has a unique character to orient to the direction of the magnetic field generated by an input current. The number of particles that can be oriented to the direction of the magnetic field should increase with increasing amount of magneto-responsible particle in the natural rubber matrix. Therefore, more particles in the composite lead to growth in the shear modulus of the magneto-rheological elastomer. The shear moduli of both i-MRE and a-MRE with a certain amount of magneto-responsible particle also increased with increasing input current. This phenomenon is more remarkable for a-MRE than i-MRE. As mentioned, a-MRE is a composite, in which pre-orientation was induced by a magnet prior to the curing process, whereas i-MRE underwent a curing process without a pre-orientation of the particles after compounding. Consequently, when a magnetic field was applied to the magneto-rheological elastomer specimens, the particles in a-MRE changed their orientation to the direction of the magnetic field, whereas those in i-MRE did not. Therefore, a-MRE showed a higher magneto-rheological effect than i-MRE.

Fig. 5. Change of shear moduli of MREs according to input current : (a) i-MRE, (b) a-MRE

Fig. 6 shows the modified magneto-rheological effect of a-MRE compared to i-MRE at an input current of 3A, which was calculated using eq. 2. The maximum variance in shear modulus was 59 % at 30 vol % magneto-responsible particle. The variance in shear modulus decreased with increasing magneto-responsible particle content over 30 vol %. This means that the magneto-rheological effect on the particle orientation was optimized at 30 vol%, which corresponds to the mechanical properties of magneto-rheological elastomer.

Fig. 6. Modified magneto-rheological effect of a-MRE compared with i-MRE

Conclusion

This study examined the effects of the orientation of particles on the mechanical properties, hardness and magneto-rheological properties. Two different types of magneto-rheological elastomer were manufactured using a specially designed magnetic-inserted mold. The tensile strength and elongation at break decreased and the hardness increased with increasing amount of magneto-responsible particle. At the same vol % of magneto-responsible particle, a-MRE showed lower tensile strength and higher hardness than those of i-MRE due to a difference in the orientation direction and testing direction. An external magnetic field was applied to the magneto-rheological elastomer. Most of specimens showed magnetic saturation at a current of 2A regardless of the isotropy of the particles. At an input current of 3A, the variance in shear modulus between a-MRE and i-MRE showed a maximum of 59 % at 30 vol % magneto-responsible particle. Based on these results, the chain-like formation of magneto-responsible particle in the natural rubber matrix played a key role in controlling the change in magneto-rheological elastomer modulus.

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