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Material Properties

Study on the damping properties of magnetorheological elastomers based on *cis*-polybutadiene rubber

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Abstract

Magnetorheological elastomers (MREs) are composed of magnetizable particles (iron particles) and a soft rubber-like matrix. Their mechanical properties, including modulus and damping capability, can be controlled by an external magnetic field. The damping properties of MREs, which play an important role in applications, depend mainly on particle content. This paper aims to investigate MRE's damping capabilities by studying two categories of *cis*-polybutadiene rubber-based MREs: isotropic and structured MREs. Both isotropic and structured MRE samples with various iron particle contents (60, 70, 80 and 85 wt%) were fabricated and their damping properties were measured by using a modified dynamic mechanical analyzer (DMA) and a universal testing machine. The results show that the loss factor in the glass transition region decreases with the increment of iron particle content. The loss factors of structured MREs are lower than those of isotropic MREs when the iron particle contents are the same. Furthermore, dynamic testing was conducted to study the effect of strain amplitude, frequency and magnetic field on the loss factor of MREs. In addition, the stress-softening experiments indicate that the ratio of remaining strain energy versus initial strain energy shows a decreasing trend with iron particle content and loading time.

Keywords: Magnetorheological elastomers; *cis*-Polybutadiene rubber; Damping properties; Stress softening

1. Introduction

Magnetorheological (MR) materials have been developing for more than half century since Rabinow first discovered magnetorheological fluids (MRFs) in 1948 [1]. MR materials are a kind of smart materials whose rheological properties can be controlled by an applied magnetic field. Magnetorheological elastomers (MREs) are a newer branch of MR materials. They are solid analogs of MRFs, in which the fluid component is replaced by rubber-like solid materials. Unlike MRFs, MREs have high stability as particles inside are not prone to settlement. The matrix includes silicon rubber, natural rubber, polyurethane sealant, acrylonitrile, and polybutadiene as well as their blends [2–6].

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MREs can be fabricated either without or with a magnetic field. The former results in isotropic MREs [6,7], while the latter results in anisotropic or structured MREs [3]. The major difference between the two types of MREs is the formation of chain-like and columnar structures in the matrix along the direction of the magnetic field before curing for the structured MREs. The modulus of MREs can be continuously and reversely changed within milliseconds by applying and removing the magnetic field [8].

Such properties indicate that MREs are promising for many applications, such as adaptive tuned vibration absorbers, stiffness tunable mounts and suspensions, and variable impedance surfaces [9–12]. It is noted that the performances of MRE devices are very dependent on the damping capabilities of MRE materials, so it is crucial that these properties are studied. However, there is little work to address this important matter. To this end, this paper aims to study the influence of the iron particle content as well as the microstructure (isotropic or structured) on the damping properties of MREs. Because *cis*-polybutadiene has relatively low damping, it is chosen as the MRE matrix here. The stress softening of MREs and the MR effect are also experimentally investigated.

2. Experimental

2.1. Preparation of materials

Two types of spherical magnetic filler were used to fabricate MREs. They were purchased from BASF, Germany (model CN and SU). The first has carbonyl iron particles with size distribution: $d10 = 3.5\,\mu m$, $d50 = 6\,\mu m$ and $d90 = 21\,\mu m$. The second has the distribution: $d10 = 0.7\,\mu m$, $d50 = 1.7\,\mu m$, $d90 = 3.4\,\mu m$. The matrix material was *cis*-polybutadiene rubber, manufactured by ShangHai GaoQiao Petrochemical Corporation. The vulcanization system was a conventional sulfur curing system. The recipe is: 100 phr rubber, 5 phr ZnO, 1 phr stearin, 2 phr MDA, 3 phr sulfur, 1 phr cz and 45 phr plasticizer. The mass fractions of the carbonyl iron particles in the matrix were 60%, 70%, 80% and 85%. The contents of model CN and SU particles were the same. The iron particles and other additives were first mixed into the rubber through conventional rubber mixing techniques on a two-roll mill. Then, the mixture was put into a mold. Two types of MREs, isotropic and structured, were prepared. The isotropic MREs were cured at 160 °C for 24 min under a pressure of approximately 10 MPa. The structured MREs were subjected to an external homogenous field of 1300 mT at 160 °C for 8 min, and then the samples were vulcanized at 160 °C for 16 min under a pressure of approximately 10 MPa. The magnetic field for the pre-structured process was provided by a self-developed magnet-heat couple device, whose working principle and schematic structure were detailed in our previous work [4].

2.2. MRE property testing

The damping properties of MRE samples were evaluated at low strain (including dynamic tension under temperature scanning and dynamic shear under strain amplitude, frequency sweep and magnetic flux density scanning) and at high strain (quasi-static extension at room temperature).

2.2.1. Dynamic tension

The dynamic mechanical performances of the MREs were obtained by using a dynamic mechanical analyzer (DMA). The samples were analyzed in tensile mode at a frequency of 1 Hz, with a strain of 0.5% and a temperature range from −105 to 30 °C. The samples were about $10 \times 10 \times 1\,mm^3$.

2.2.2. Strain and frequency sweep testing

Both strain amplitude sweep and frequency sweep modes were employed to measure dynamic performance of MRE samples. With these measurements, MR effect and loss factor ($\tan\delta$) were obtained under various magnetic fields. The frequency ranged from 0.1 to 40 Hz, and the strain amplitude was from 0.1% to 1.3%. The magnetic flux density ranged from 0 to 800 mT. The samples were about $10 \times 10 \times 3\,mm^3$.

2.2.3. Quasi-static testing

A Universal Testing Machine (JPL-2500) at a crosshead speed of 500 mm/min was used for quasi-static testing. The dumbbell specimens were about 6 mm wide and 2 mm thick in the central section. The samples were subjected to four cycles in tension from 0% to 100% and back to 0% strain. After the samples were rested for 24 h at room temperature, a fifth extension was carried out.

3. Results and discussion

3.1. The glass transition temperature and tan δ

The relationships between the loss factor (tan δ) and the temperature for both isotropic and structured MREs with various iron particle contents are shown in Figs. 1 and 2, respectively. As observed from these figures, the peak loss factor values of the MREs decrease from 1.908 to 0.998 for isotropic samples and from 1.644 to 0.994 for structured ones with the addition of the iron particles. The temperature corresponding to tanδ can be approximately regarded as the glass transition temperature, Tg [13]. From these two figures, Tg increases from −89.5 to −82.2 °C for isotropic MREs and from −88.5 to −78.8 °C for structured MREs.

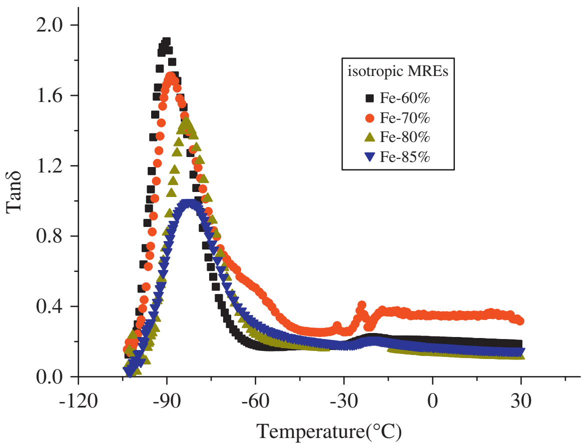

Fig. 1. Relationship between tan δ and temperature for isotropic MREs with different content of iron particles.

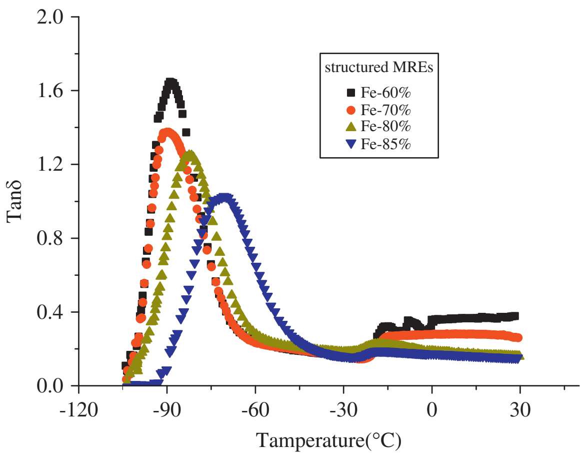

Fig. 2. Relationship between tan δ and temperature for structured MREs with different content of iron particles.

These results can be explained through the free volume theory. Transition from glassy to rubbery phase contributes to mobility of the polymer segments. The distance between particles is much smaller with the increment of particle content from 60% to 85%, which can be observed in Fig. 3(a, b and a′, b′). The free volume that offers the space for mobility of the segments will decrease. Therefore, the temperature at which the rubber segments can mobilize will increase. At the same time, the free volume change is relative to the damping properties of MREs. The loss factor decreases when the free volume becomes smaller. The properties are in agreement with the results for particle-reinforced EPDM composite [14].

Moreover, the tanδ values for isotropic MREs are larger than those of structured MREs at the same iron particle content. As a chain of particles exists in structured MREs, the distance between the particles is much closer than that of isotropic MREs at the same particle content, which is seen in Fig. 3(a, a′ and b, b′).

It should be noted that the tanδ alone cannot be used to evaluate damping properties of MREs [15]. Both the width and the height of loss factor curves should be carefully considered. The loss factor of isotropic MREs can reach a maximum value when the iron particle content is 70%, as observed in Fig. 1.

3.2. Dynamic performances

Relative MR effect is a key parameter for evaluating MRE performance. The relative MR effect is defined as the difference between the magneto-induced shear modulus and the zero-field shear modulus. The results of relative MR effect for isotropic and structured elastomers with different iron particle content are shown in Fig. 4. As observed from this figure, relative MR effect for both isotropic and structured MREs increases steadily up to a maximum value of 80% when the addition of iron particles is up to 70%. After this, the relative MR effect shows a decreasing trend, because the zero-field modulus of the elastomers is very large. In addition, the relative MR effect for structured MREs is larger than that for isotropic MREs at the same particle content.

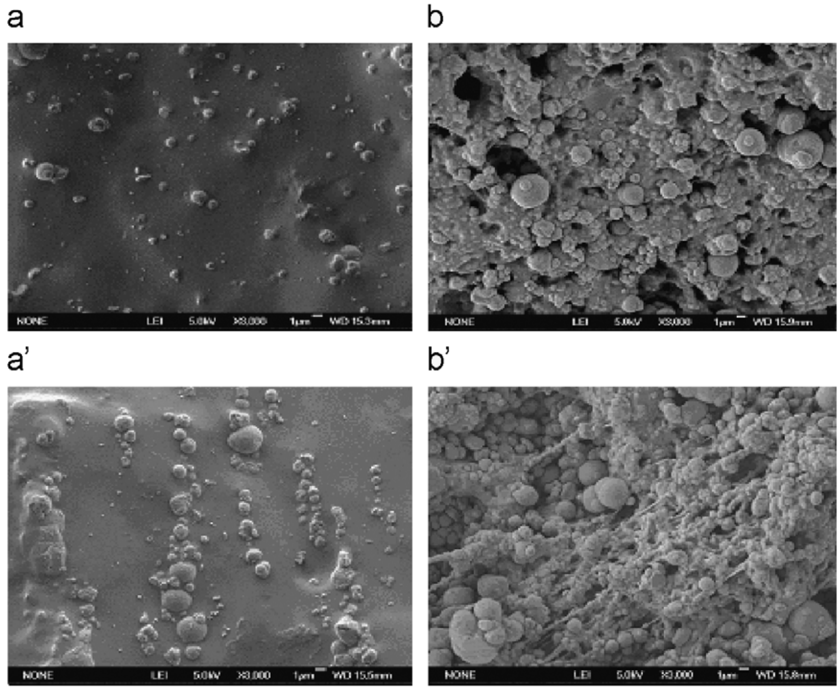

Fig. 3. The distribution of particles for isotropic (a and b) and structured (a′ and b′) MREs with the iron particle contents of 60% and 80% viewed by SEM.

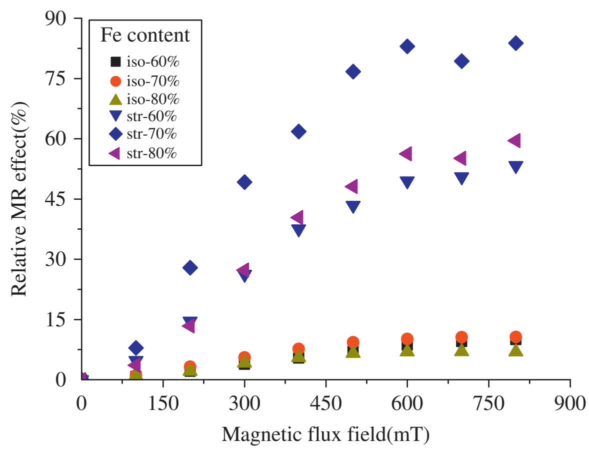

Fig. 4. Relationship between relative MR effect and magnetic flux field for isotropic (iso-) and structured (str-) MREs with different iron particle contents in the matrix at the frequency of 10 Hz.

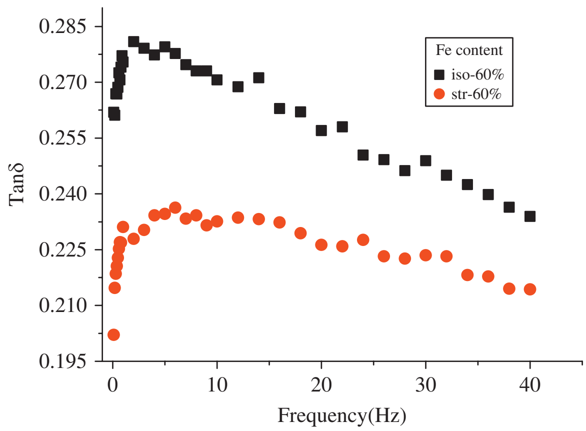

Fig. 5. Relationship between $\tan\delta$ and frequency for isotropic (iso-) and structured (str-) MREs with the iron particle contents of 60%.

The frequency dependence of the loss factor is shown in Fig. 5 for isotropic and structured MREs with the same iron particle content of 60%. The results indicate that the loss factor firstly increases sharply with the increment of frequency and then the values decrease gradually with further increasing frequency. The maximum loss factor is 0.279 and 0.236 for isotropic and structured MREs, respectively, when the frequency is 5 Hz. The loss factor of isotropic MREs is larger than that of structured MREs for the whole frequency range.

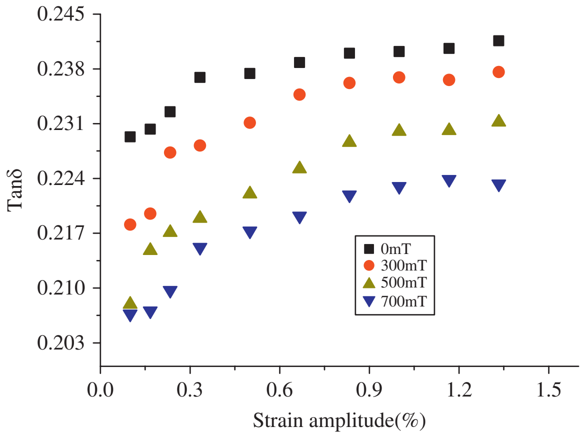

Fig. 6. Relationship between $\tan\delta$ and strain at the different magnetic flux field for structured MREs with the iron particle contents of 70%.

The strain amplitude dependency of the loss factor is shown in Fig. 6 for structured MREs with an iron particle content of 70% at various magnetic flux densities. The results indicate that the loss factor increases steadily with the increment of strain, but it reaches a plateau when the amplitude is above 1%. The loss factor shows a decreasing trend with magnetic field ranging from 0 to 700 mT. It is noted that the maximum change of the loss factor is very small, ranging from 0.02 to 0.015.

3.3. *Stress softening*

For the uniaxial tension experiments with filled composite, the stress-softening effect should be taken into account through repeated extensions. The phenomenon is known as the Mullins effect, which can also be used to evaluate energy loss. This effect is represented by the following equation $R_i' = R_i/R_1 \times 100\%$, where R_i' is the percentage of remaining strain energy, R_i is the strain energy at the ith extension and R_1 is the strain energy at the first extension.

Fig. 7 shows the stress–strain relations of both isotropic and structured MREs with the iron particle content of 60% (a, a′), 70% (b, b′) and 80% (c, c′), respectively. The specific data are listed in Table 1. The results indicate that all samples tend to soften further, with the remaining strain energy decreasing with increasing number of cycles. The remaining strain energy decreases (energy loss increases) with the addition of iron particles. When the samples are put aside for 24 h at room temperature, the stress recovers to almost the original values of the first extension, except for the material with iron particle content of 80% (the remaining strain energy appears to exceed 100% due to measurement errors). When the iron particle content is 80%, the composite is damaged at high strain because of debonding of the elastomers from the particles; hence the stress cannot recover to the original value. The comparison of these data listed in Table 1 implies that the remaining strain energy for isotropic MREs is lower than that of structured MREs at the same particle content. The mechanism of stress softening is consistent with carbon-black-filled rubber, which can be explained by Bueche's model [16]. He claimed that the stress softening was due to the shorter chains between the particles becoming taut and tearing loose from the particles when the samples were tensioned. The formation of particle chains exists in the structured MREs, the chains between particles are smaller and the stress softening and energy loss will become more intense.

These results are different from the variation of damping properties from dynamic tension measurement. This is because the stress-softening results were obtained under high strain, while the damping properties were obtained through dynamic testing under low strain. The difference results from the mechanism of energy loss.

4. Conclusions

The influence of iron particle content on damping properties as well as MR effects of *cis*-polybutadiene rubber-based MREs was studied. The results demonstrate that the peak loss factor values decrease from 1.908 to 0.998 for isotropic samples and from 1.644 to 0.994 for structured ones with the addition of iron particles. Also, the $\tan\delta$ values for structured MREs are lower than those of isotropic MREs at the same iron particle content. Tg increases from −89.5 to −82.2 °C for isotropic MREs and from −88.5 to −78.8 °C for structured MREs with the addition of iron particles. However, the variation of damping properties is contrary to the results from a stress-softening experiment. The remaining strain energy decreases with the increase of iron particle contents in the matrix, and the remaining strain energy for isotropic MREs is lower than that of structured MREs at the same particle content. The difference results from the mechanism of energy loss. The relative MR effect can be up to a maximum value of 80% when the iron particle

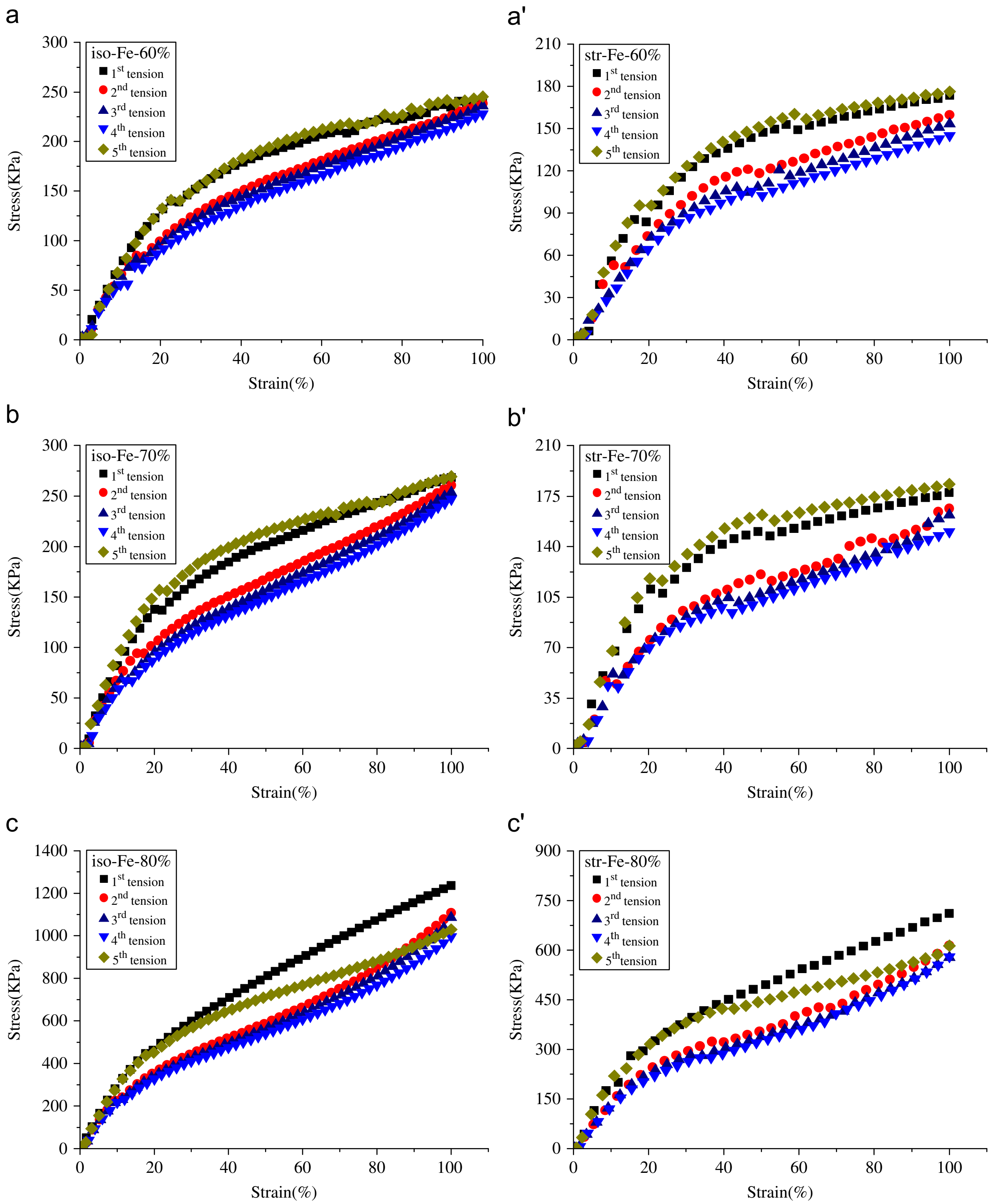

Fig. 7. Stress-softening effect of isotropic (iso-) and structured (str-) MREs with the iron particle contents of 60% (a, a′), 70% (b, b′) and 80% (c, c′), respectively.

Table 1
Rate of remaining strain energy for isotropic and structured MREs with different contents of iron particles

Iron particles content	R_i'			
	R_2' (%)	R_3' (%)	R_4' (%)	R_5' (%)
Isotropic (60%)	88.7	86.1	81.6	100.1
Structured (60%)	86.1	79.9	75.2	101.5
Isotropic (70%)	86.7	81.4	77.7	100.8
Structured (70%)	81.1	76.9	73.4	101.8
Isotropic (80%)	77.7	74.2	70.5	86.7
Structured (80%)	76.8	72.2	70.1	89.9

content is 70%. The influences of strain amplitude, frequency and magnetic flux field on the damping capacities of MREs are small, in the range of 0.02–0.015.

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