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2007-01-01

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Chen, L., Gong, X.L., Jiang, W.Q., Yao, J.J., Deng, H. & Li, W.H. (2007) Investigation on Magnetorheological Elastomers Based on Natural Rubber. *Journal of Materials Science*, Volume 42, Number 14, 5483-5489, doi:10.1007/s10853-006-0975-x

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3 Investigation on magnetorheological elastomers based on natural 4 rubber

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7 Received: 12 April 2006 / Accepted: 19 September 2006 / Published online: ■
8 © Springer Science+Business Media, LLC 2007

9 **Abstract** Magnetorheological Elastomers (MR Elastomers or MREs) are a kind of novel smart material, whose
10 mechanical, electrical, magnetic properties are controllable
11 under applied magnetic fields. They have attracted
12 increasing attentions and broad application prospects. But
13 conventional MREs are limited to wide applications
14 because their MR effects and mechanical performances are
15 not high enough. This paper aims to optimize the fabrication
16 method and to fabricate good natural rubber based
17 MREs with high modulus by investigating the influences of
18 a variety of fabrication conditions on the MREs performances,
19 such as matrix type, external magnetic flux density,
20 and temperature, plasticizer and iron particles. Among
21 these factors, the content of iron particles plays a most
22 important contribution in shear modulus. When the iron
23 particle weight fraction is 80% and the external magnetic
24 flux density is 1 T, the field-induced increment of shear
25 modulus reaches 3.6 MPa, and the relative MR effect is
26 133%. If the iron weight fraction increases to 90%, the
27 field-induced increment of shear modulus is 4.5 MPa. This
28 result has exceeded the best report in the literatures
29 researching the MREs on the same kind of matrix. The
30 dynamic performances of MREs were also experimentally
31 characterized by using a modified Dynamic Mechanical
32 Analyzer (DMA) system. The effects of strain amplitude
33 and driving frequency on viscoelastic properties of MREs
34 were analyzed.

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Introduction

Magnetorheological (MR) materials belong to a class of function materials and smart materials, due to their rheological properties can be changed continuously, rapidly and reversibly by applied magnetic fields. Recently, MR materials play important roles in the domain of the automotive vehicles, architecture, and vibration controls, etc. [1].

The most common MR materials are MR fluid (MRF) [2], comprising micro-sized or sub-micro-sized magnetizable particles dispersed in liquid-state materials. Two or three orders of magnitude may happen on the yield stress and apparent viscosity as well as the suspension system changes from Newtonian liquid to non-Newtonian liquid when a magnetic field applied on MRF [3–5].

MRE is the solid-state analogue of MRF, and a new branch of MR materials. The problems existing in MRF such as particle sediment are well overcome via replacing the fluid matrix by solid matrix, such as rubber. MRE is the resulting composites made up by soft magnetic particles embedded in a polymer. The interactions between magnetic particles under a magnetic field result in field-dependent mechanical performances [6, 7]. Having both excellences of MRF and elastomers, MRE has attracted considerable interest recently [8–10]. Some MRE based devices have been reported. For example, a proof-of-concept

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63 variable-rate automotive suspension bushing consisting of
64 concentric outer and inner sleeves based on MRE was
65 designed by Ford Research Laboratory [10].

66 A few research groups used silicone rubber, gels and
67 resin as soft matrix [6, 8, 11, 12], which can be easily
68 processed from liquid precursors; several kinds of MREs
69 are prepared on polymers which are in possession of
70 excellent mechanical properties such as natural rubber and
71 nitrile rubber [10, 13]. However, MREs fabricated with
72 such methods are limited for wide applications because
73 they are difficult to own good bearing capacity and good
74 MR effects at the same time.

75 This work aims to fabricate high-efficiency natural
76 rubber based MREs. The effects of fabrication conditions
77 (matrix type, external magnetic flux density, and tempera-
78 ture), and materials (plasticizers and iron particles) on
79 MRE performances were experimentally investigated.
80 Dynamics properties of the fabricated MREs were also
81 characterized and analyzed (Fig. 1).

82 Experimental

83 Preparation of MRE materials

84 The fabrication of MREs consists of three major steps:
85 mixing, forming pre-configuration and sulfuration. The
86 mixture is processed with conventional rubber-mixing
87 techniques. A Double-Roll Mill (Taihu Rubber Machinery
88 Inc. China, Model XK-160), is used to fabricate rubber.
89 When the machine is running, two rolls are rotating on
90 opposite directions with different speeds whilst the roll gap
91 can be set in a very small scale. The massive natural rubber
92 on the rolls is subjected to strong extrusion pressure and
93 shear force. Through the rolls uninterrupted rotating, the
94 molecular chains in natural rubber are breakdown, and the

natural rubber losses its elastic and becomes viscous body gradually. So crosslinkers and processing aids, the carbonyl iron particle and plasticizers can be easily added into the natural rubber. The resulting material is then compression-molded into a mold in the self-developed Magnet-Heat Coupled Device, as shown in Fig. 2. It is composed of a magnetic field, a mold, and a controllable heating system. The MRE sample is in the mold which is exposure to the magnetic field and tightly fixed with heat plate. The magnetic field is generated by a magnetic coil, which is capable of applying the external magnetic flux density of 0 to 1 T over the samples. The heat plate is conterminal with a temperature controller whose temperature can set in the range from 50 °C to 200 °C. During the pre-configuration stage, the heating system and the magnetic field are both turned on so that both the temperature and the external magnetic flux density can be set properly. The particles are magnetized and then form chains aligned along the field direction. 30 min later, the procedures of forming pre-configuration is finished. After shutting down the magnetic field, the temperature is raised to 153 °C. At this condition, the sample is on sulfuration for 15 min. Then the MRE based on natural rubber is prepared. The used carbonyl iron particles are supplied by BASF, Germany, model CD , the particle size distribution: $d_{10} = 3 \mu\text{m}$, $d_{50} = 6 \mu\text{m}$, $d_{90} = 11 \mu\text{m}$. The natural rubber, plasticizers and other additives are provided by Hefei Wangyou Rubber Company, China. The main ingredients in plasticizers are vaseline and paraffine.

For the purpose of comparison, other MRE samples based on silicone rubber matrix are also prepared. The iron particle, Dimethyl-silicon oil (Shanghai Resin Factory, China, with the viscosity of 300cp) and RTV silicone rubber (Xida Adhesives Factory, China, Model 704) are mixed together, then the hybrid is put into the mold under the magnetic flux density of 1 T, for curing up to 24 h at room temperature.

In the experiment, a Tesla gauge (Shanghai Hengtong MagnetoElectricity Co. Ltd, China) is used to test the magnetic flux density outside the MRE.

Dynamic testing system of MRE performance

A Dynamic Mechanical Analyzer (DMA) is the common equipment for dynamic testing on viscoelastic material. In this work, the DMA (Triton Technology Ltd. UK, Model Tritec 2000B) system, is modified to characterize MRE performances by introducing a self-made electromagnet which can generate a variable magnetic flux density up to 1 T (sketch map shown in Fig. 2). This system applies a fixed oscillatory strain to the specimen and measures the amplitude and phase of the output force, from which stress, modulus (shear storage modulus G' and loss modulus G'')

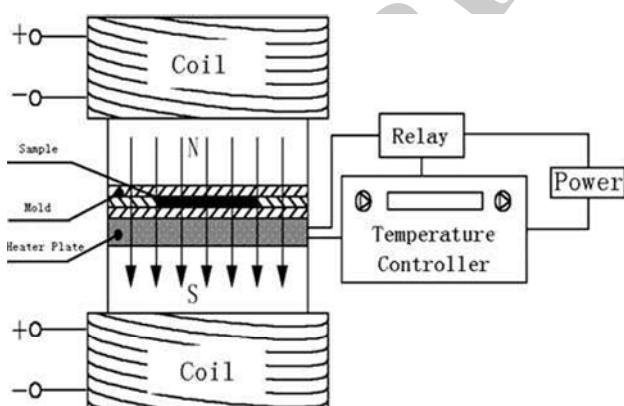
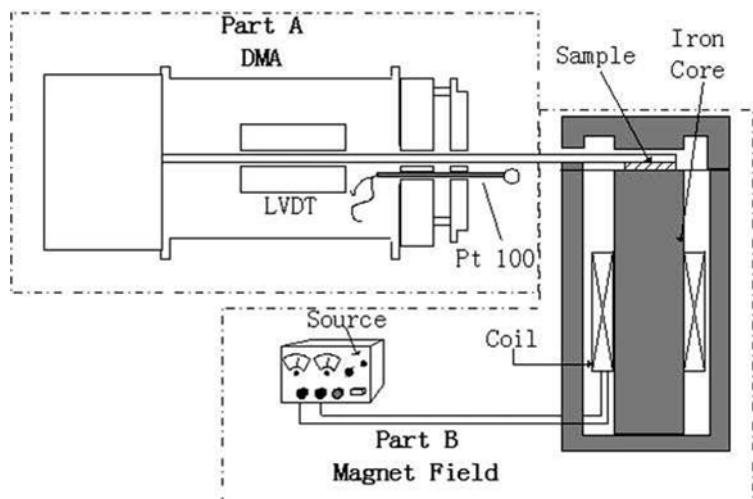


Fig. 1 The sketch map of self-assembled magnet-heat coupled device. The dimensions of sample in the mold are 80 mm × 80 mm × 3 mm (length, width and thickness)

Fig. 2 A sketch map of magnet-mechanics coupled DMA. The dimensions of testing sample are $10 \text{ mm} \times 10 \text{ mm} \times 3 \text{ mm}$ (length, width and thickness). The direction of the external magnetic flux density is perpendicular to surface of the testing sample



146 included) and the loss tangent ($\tan \delta = G''/G'$) can be
 147 calculated. Testing involved recording the modulus and the
 148 loss tangent of various specimens at various frequencies,
 149 strains and applied magnetic fields. In the context, the shear
 150 storage modulus is studied and the phrase "modulus"
 151 refers to the shear storage modulus.

152 The experiment is started in the room temperature, and
 153 the increment of temperature of the electromagnet is less
 154 than 3°C during the stage of the whole experiment.

155 Mechanical measurements

156 In order to supply reference for properties of materials and
 157 applications, the basic mechanical properties of MRE are
 158 also measured. Tensile strength, angle tear strength, resili-
 159 ence factor and hardness are the most basic and important
 160 factors of mechanical performances in rubber industry [14],
 161 and are tested on JPL mechanical test machine, JC-1007
 162 elasticity test machine, LX-A hardness gauge, respectively.
 163 These apparatus are all manufactured by Jiangdu Jingcheng
 164 Test Instruments Factory, China.

165 Results and discussion

166 Influence of fabrication conditions on the MRE 167 performances

168 The influences of matrix type, external magnetic flux
 169 density (B), and temperature in the stage of forming pre-
 170 configuration, content of plasticizers and iron particles on
 171 the MRE performances are experimentally investigated,
 172 and discussed below. It is noted that all percentages used in
 173 the context refer to weight percentages.

174 When the dynamic testing done in paragraph 3.1, the
 175 exciting frequency is fixed as 5 Hz and the dynamic stain
 176 amplitude is set at 0.3%.

Matrix type

177 Two kinds of MREs based on silicone rubber and natural
 178 rubber are prepared. The two kinds of MREs have the same
 179 ingredient proportions (60% of iron particle, 10% of
 180 plasticizers, and 30% of matrix). The only difference be-
 181 between them is the matrix used: one is natural rubber and the
 182 other is silicone rubber. Mechanical performances in terms
 183 of tensile/tear strength, resilience factor and harness of the
 184 two kinds of MREs were measured and compared. As can
 185 be seen from Table 1, MRE based on natural rubber gen-
 186 erally have better performances than that based on silicon
 187 rubber. For example, both the tensile strength and the tear
 188 strength of nature rubber based MREs are almost 10 times
 189 as that of silicone rubber based MREs. Therefore, MREs
 190 whose matrix is well mechanical performance polymers
 191 such as natural rubber instead of soft materials, would gain
 192 wide applications (Fig. 3).

External Magnetic flux density in forming pre-configuration

194 In this group, four natural rubber based MRE samples with
 195 the same compositions (60% of iron particles, 20% of natural
 196 rubber, and 20% of plasticizers) were pre-configu-
 197 198

Table 1 Comparison of mechanical performance of MRE based on natural rubber and silicon rubber

Test samples	Tensile strength/ MPa	Angle tear strength N/mm	Resilience factor	Hardness
Silicone rubber MRE	0.7	1.7	28%	33
Natural rubber MRE	6.5	16.3	52%	45



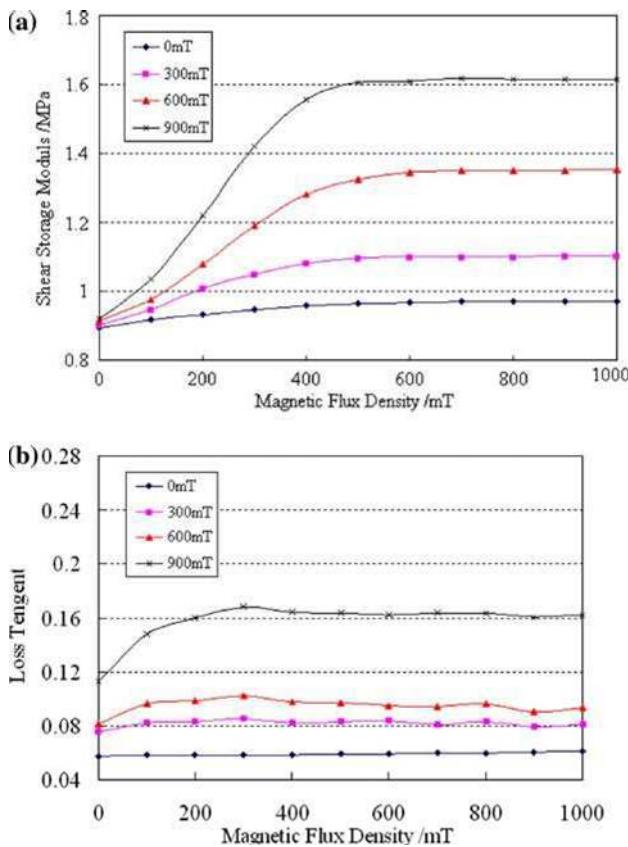


Fig. 3 Increment of the magneto-induced modulus (a) and loss tangent (b) with applied magnetic strength for MREs pre-configured under $B = 0, 300, 600$ and 900 mT, respectively

199 rated at the temperature of 80°C , but fabricated at four
200 external magnetic fields with $B = 0, 300, 600, 900$ mT,
201 respectively. The field dependence of shear modulus for
202 these four samples is shown in Fig. 4(a). As can be seen
203 from this figure, shear modulus of each sample shows an
204 increasing trend with the external magnetic flux density
205 prior to the iron particle saturation. This is because the
206 shear modulus comes from the actions of magnetizable
207 particles. When iron particles reach saturation magnetiza-
208 tion, the actions between magnetizable particles can't vary
209 with the external magnetic flux density, thus, the magneto-
210 induced modulus reach the maximum. By comparing these
211 four samples, it is found that strong external magnetic flux
212 density applied in forming pre-configuration leads to the
213 high magneto-induced modulus. For example, the maxi-
214 mum modulus of MRE pre-configured in the magnetic
215 flux density of 900 mT is above 1.6 MPa while the one
216 pre-configured without field is 0.9 MPa. This is obvious
217 because stronger external magnetic flux density helps to
218 form more stable chain or column structures and conse-
219 quently induce higher magneto-induced modulus. It is also
220 indicated in Fig. 4(a) that external magnetic flux density in
221 forming pre-configuration has no influence on the MRE

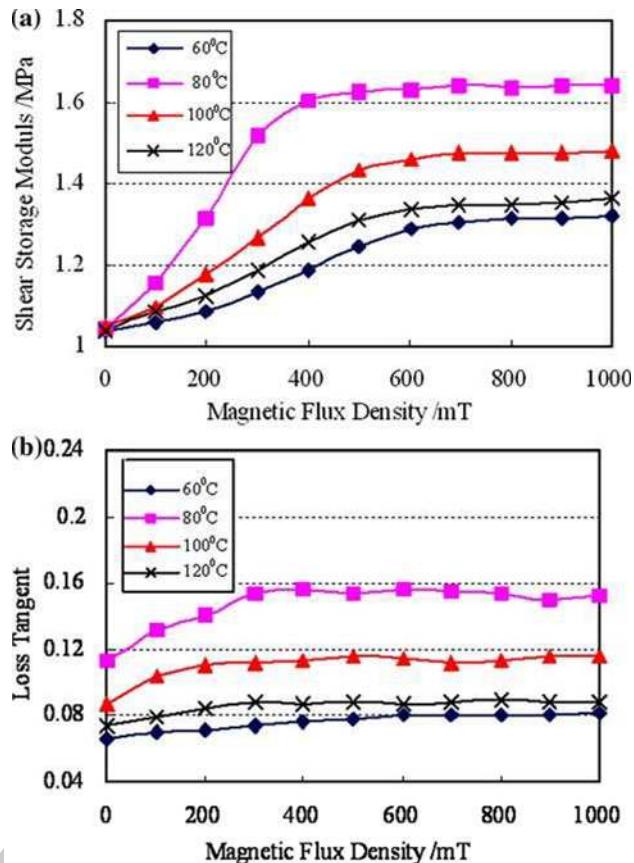


Fig. 4 Increment of the magneto-induced modulus (a) and loss tangent (b) with applied magnetic strength for MR elastomers pre-configured at the temperature of 60°C , 80°C , 100°C and 120°C , respectively

zero-field modulus. It is because there is no magnetic interaction between the iron particles in MRE when no magnetic field is applied. On the other hand, the field dependence of loss tangent for these four samples is shown in Fig. 4(b). It can be seen from this figure that the loss tangent follows the same rule to the external magnetic flux density during the stage of forming pre-configuration.

Temperature in forming pre-configuration

In this group, four natural rubber based MRE samples with the same compositions (60% of iron particles, 20% of natural rubber, and 20% of plasticizers) were pre-configured in the applied external magnetic field with $B = 1$ T, but at four different temperatures of 60°C , 80°C , 100°C , 120°C , respectively. The field dependence of modulus for these four samples is shown in Fig. 5. It can be seen from this figure that the MRE pre-configured in 80°C has the best MR effects in this group. This result is probably due to temperature effect on the natural rubber matrix. It is known that the natural rubber is a sort of temperature dependence

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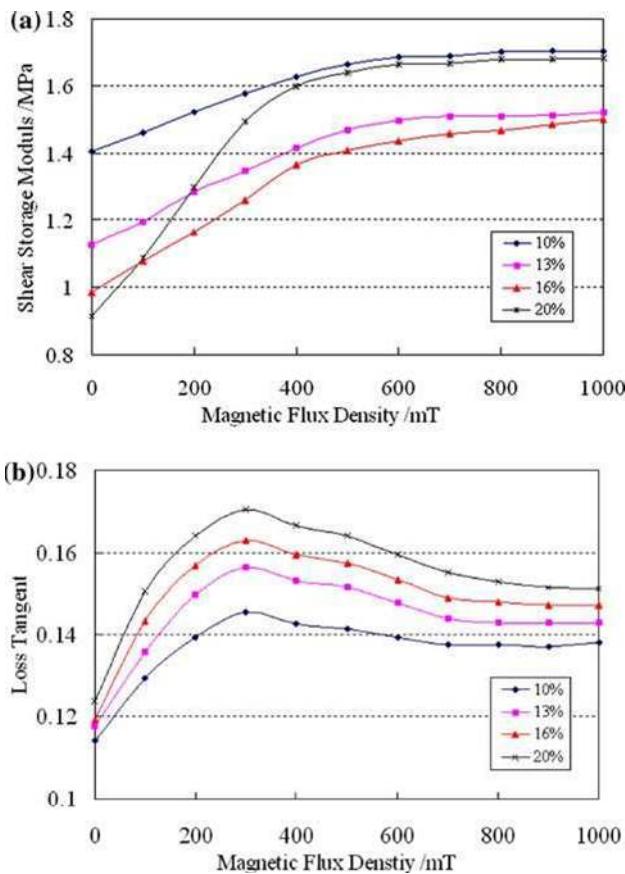


Fig. 5 Influence of content of plasticizers on the MR effects of variation of magneto-induced modulus and loss tangent

241 of viscoelastic material. Such material behaves as elastomer at room temperature but turns into a soft viscoplastic
242 or fluid substance when it is heated. Further increasing
243 temperature, the matrix will become hard again because
244 chemical crosslinking plays an increasing role with
245 increasing temperature. As shown in Fig. 5, 80 °C is the
246 ideal temperature for the particles to move and form ordered
247 pre-configuration. Same as the sample group in 3.1.3,
248 the zero-field moduli are not changed by the results of pre-
249 configuration.
250

251 Content of plasticizers

252 In this group, four MRE samples based on natural rubber
253 are fabricated with 60% of iron particles, pre-configured
254 in the applied external magnetic field with $B = 1$ T and at
255 the temperature of 80 °C. But the contents of plasticizers
256 are 10%, 13%, 16% and 20%, respectively. Plasticizer is a
257 kind of additions in the rubber technology; it can be dissolved
258 in rubber after mixing them together. Plasticizers act as the lubricant and let the molecular chains of rubber
259 glide easily, and then the rubber matrix shows a low
260 adhesiveness. So adding the plasticizers in the MREs is
261

expected to improve MRE performances, because plasticizers can not only change the rubber mechanisms but also modify particle properties. Figure 6 shows the influence of plasticizers on MRE performances. From this figure, the zero-field moduli (G_0 , the shear storage modulus of MRE when the external magnetic flux density $B = 0$) of the samples with 10% and 20% weight fraction of plasticizer are 1.4 MPa and 0.9 MPa, respectively. Also, their corresponding saturation magneto-induced moduli(ΔG , the change of the shear storage modulus when saturation magnetization) are 0.2 MPa and 0.7 MPa. So the relative MR effects $\Delta G/G_0$ are 14% and 78%, respectively. Therefore, the amount of plasticizers in the matrix plays an important role in improving MR effects, especially the relative MR effects. The field dependence of loss tangent is shown in Fig. 6(b), where the loss tangent firstly increases steadily with the increment of external magnetic flux density up to a maximum value at 300 mT. Above $B = 300$ mT, the loss tangent shows a decreasing trend with flux density. This may be due to the temperature effect in the testing system. The testing sample is attached to the

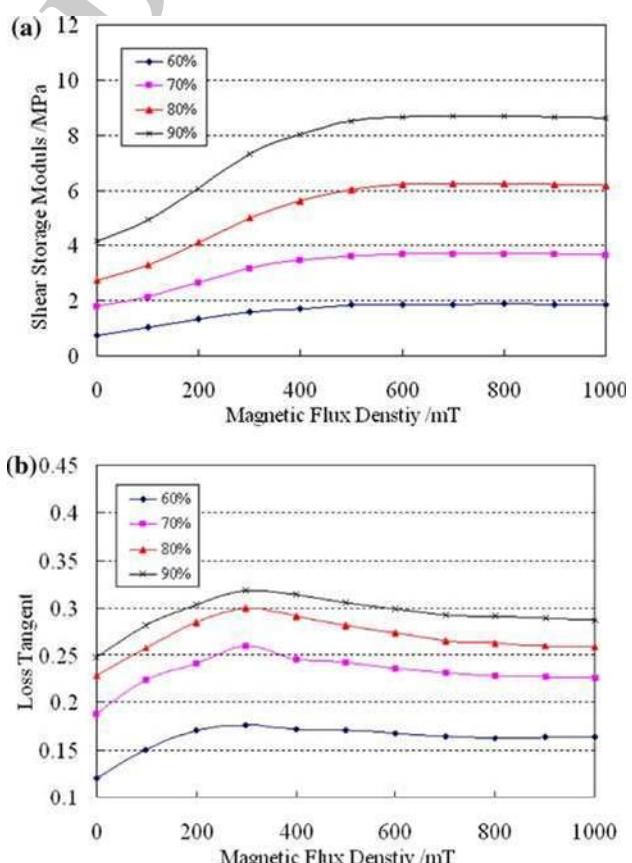


Fig. 6 Dependence of MR effects of variation of magneto-induced modulus and loss tangent on content of iron particles embodied in MREs



283 electromagnet whose temperature rises steadily when the
284 magnetic flux density increases.

285 Content of iron particles

286 In this group, four MRE samples based on natural rubber
287 are pre-configured in the applied external magnetic field
288 with $B = 1$ T and at the temperature of 80 °C. According to
289 the results in 3.1.5 and in order to get the best MR effect,
290 the ratio of plasticizers to natural rubber is set at 1. But
291 their iron particles contents are 60%, 70%, 80% and 90%,
292 respectively. The influence of content of iron particles on
293 MR effects are shown in Fig. 7 and summarized in
294 Table 2. The results show that the magneto-induced
295 modulus increases dramatically with the particle content
296 increases. For example, the magneto-induced modulus is as
297 high as 4.5 MPa when the content is 90%. This is because
298 the modulus is induced by interactional force between the
299 iron particles. So, more the particles are, higher the mag-
300 neto-induced modulus is. However, the increment of iron
301 particles also enhances the zero-field modulus, which may
302 decrease the relative MR effect. For example, the relative
303 MR effect is reduced from 133% to 107% when particles
304 content changed from 80% to 90%. It is also shown in
305 Table 3 that the mechanical performances of MRE filled
306 with different content of iron particles are quite different.
307 Increment of content of iron leads to the decrement of
308 tensile strength and angle tear strength of MRE. Thus, it is
309 not applicable to fabrication practical MRE by solely
310 increasing particle contents.

311 Dynamic properties of MREs

312 MRE based device often operates in dynamic mode. So the
313 study of dynamic properties of MREs will provide an
314 important reference to their practical applications.

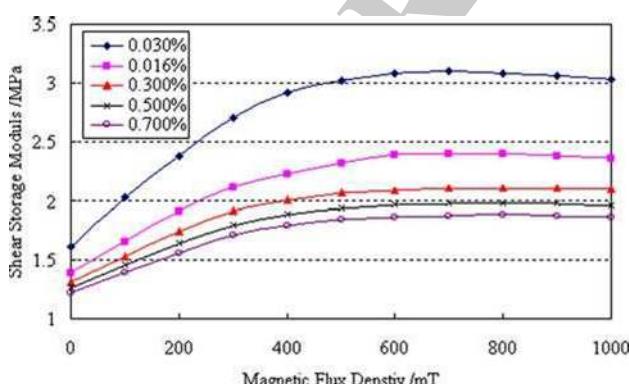


Fig. 7 Shear storage modulus as a function of magnetic strength measured at different strain amplitudes

Table 2 MR effect of four MRE samples filled with different percentage of the iron particles

Content of Fe	G_0	ΔG	$\Delta G/G_0$	$\text{Tan}\delta_0$	$\text{Tan}\delta_{\max}$
60%	0.9	0.7	78%	0.12	0.17
70%	1.8	1.9	110%	0.18	0.24
80%	2.7	3.6	133%	0.20	0.27
90%	4.2	4.5	107%	0.25	0.31

Zero-field moduli G_0 , is the shear storage modulus of MRE when the external magnetic flux density $B = 0$ and saturation magneto-induced moduli ΔG is the change of the shear storage modulus when saturation magnetization, $\Delta G/G_0$ is the relative MR effect, while $\text{Tan}\delta_0$ is the zero-field of the loss tangent and $\text{Tan}\delta_{\max}$ is the maximum of the loss tangent in the range of applied field from 0 to 1 T

Table 3 Mechanical performances of four MRE samples filled with different percentage of the iron particles

Content of Fe	Tensile strength / MPa	Angle tear strength N/mm	Resilience factor	Hardness
60%	3.25	11.4	28%	35
70%	2.27	10.7	21%	46
80%	1.29	7.6	14%	67
90%	0.32	3.7	5%	85

The dynamic testing is carried out using the modified DMA system. The sample measured is fabricated with the contents of iron particles of 60%, and plasticizers of 16%, at the applied external magnetic field with $B = 1$ T, and at the temperature of 80 °C in the stage of forming pre-configuration. Figure 8 shows the field dependence of modulus at various strain amplitudes, where the driving frequency is fixed as 5 Hz. The experimental results demonstrate the MREs behave as classical viscoelastic materials. In other words, the modulus of MREs shows a decreasing trend with applied strain amplitude. When the applied strain amplitude increases, the distance between particles within MRE will increase. This will induce the decrease of interactive forces between particles which result in the decrease of the magneto-induced modulus.

Under the same dynamic stain amplitude of 0.7%, the frequency dependence of MRE shear modulus is measured and the result shows that the exciting frequency has little influence on the magneto-induced modulus.

Conclusions

The effects of both fabrication and working conditions on MRE performances were experimentally explored in this paper. Main finding are summarized below.

- 338 • Replacing the silicone rubber with natural rubber as the
 339 matrix can get good MREs with improved mechanical
 340 performances. This could be the first step for MREs to
 341 walk out from laboratory and walk into practical
 342 applications.
- 343 • The optimal pre-configuration conditions are: augmenting
 344 the magnetic fields, setting the temperature at
 345 80 °C, and adding more plasticizers to matrix.
- 346 • The content of iron particles plays a significant role in
 347 improving MRE performances. When the iron particle
 348 weight fraction is 80%, the MRE shear modulus at the
 349 applied external magnetic field with $B = 1$ T reaches
 350 3.6 MPa, and the relative effects is 133%. When 90%
 351 iron particles are embedded in MRE, the magneto-
 352 induced modulus reaches as high as 4.5 MPa. This
 353 result has exceeded the best report in the literatures
 354 regarding MREs based on the same kind of matrix. But
 355 increment of content of iron leads to the decrement of
 356 tensile strength and angle tear strength of MRE. Thus, it
 357 is not applicable to fabrication practical MRE by solely
 358 increasing particle contents.
- 359 • MREs behave as viscoelastic materials and their
 360 dynamic properties were measured using the modified
 361 DMA. The shear modulus decreases with the increment
 362 of strain but is almost independent of driving fre-
 363 quency.

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