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Chapter

Magnetorheological Elastomers: Materials and Applications

Taixiang Liu and Yangguang Xu

Abstract

Magnetorheological elastomers (MREs) are a type of soft magneto-active rubber-like material, whose physical or mechanical properties can be altered upon the application of a magnetic field. In general, MREs can be prepared by mixing micron-sized magnetic particles into nonmagnetic rubber-like matrices. In this chapter, the materials, the preparing methods, the analytical models, and the applications of MREs are reviewed. First, different kinds of magnetic particles and rubber-like matrices used to prepare MREs, as well as the preparing methods, will be introduced. Second, some examples of the microstructures, as well as the microstructure-based analytical models, of MREs will be shown. Moreover, the magnetic field-induced changes of the macroscopic physical or mechanical properties of MREs will be experimentally given. Third, the applications of MREs in engineering fields will be introduced and the promising applications of MREs will be forecasted. This chapter aims to bring the reader a first-meeting introduction for quickly knowing about MREs, instead of a very deep understanding of MREs.

Keywords: magnetorheological elastomer, composite material, microstructure, vibration reduction, energy absorption

1. Introduction

Magnetorheological elastomers (MREs) are a type of soft particle-reinforced magneto-active rubber-like composite material, whose physical or mechanical properties can be altered upon the application of a magnetic field [1–5]. MREs can be usually prepared by mixing micron-sized magnetic particles into nonmagnetic rubber-like matrices. In the presence of a magnetic field, MREs exhibit a magnetorheological effect providing a field-dependent physical or mechanical property, for example, a controllable modulus, due to the sensitive response of the magnetic particles to the field. While the field is removed, MREs will reclaim their original, natural property. It is believed that the embryo of MREs is firstly reported by Rigbi and Jilken [1, 6] in 1983, although the discovery of the basic magnetorheological effect can be historically retrospected to the 1940s for magnetic fluid [7]. MREs can be regarded as a solid-state analog to magnetorheological fluids (MRFs) [8–12]. In general, MREs exhibit a unique field-dependent material property when exposed to a magnetic field, and can overcome major issues faced in MRFs, for example, the deposition of iron particles, sealing problems, and environmental contamination. Such advantages offer MREs great potential for designing intelligent devices to be used in various engineering fields, especially in fields that involve vibration.
reduction, isolation, and absorption [1, 13–16]. Recently, the study of the sensing behavior of MREs explosively emerged, for example, for sensing mechanical and magnetic signals [17–22].

From the first report of MREs with soft ferrite particles filling into natural rubber [6], the research of the materials and related preparing methods of MREs develops very quickly. Briefly speaking, MREs consist of three basic components: magnetic particles, nonmagnetic elastic matrices, and additives. For the magnetic particles, higher permeability, higher saturation magnetization, and lower remanent magnetization are highly desirable for obtaining stronger magnetic field-sensitive effect. At present, the micron-sized carbonyl iron powder invented by BASF in 1925 is widely used [23, 24]. For the elastic matrix, there are lots of polymeric rubbers, for example, natural rubber [25, 26], silicone rubber [27–31], poly dimethylsiloxane (PDMS) [32–34], etc., for consideration. According to need, high modulus or low modulus can be chosen, but that the magnetic particles can be locked in the matrix in the absence or presence of a magnetic field is a basic requirement. For the additives, they are determined according to the choice of the particles and the matrix, and silicone oil is usually used as an additive in the fabrication of MREs [1]. Depending on the choice of the matrix, the preparing methods of MREs are many and various, and a high-temperature or room-temperature vulcanization curing method is usually used. Attributed to applying a magnetic field in the curing process, MREs can be prepared with anisotropic particle-formed microstructure. This kind of MREs is called anisotropic MREs. When no field is applied during the curing process, prepared MREs have isotropic particle-formed microstructure and this kind of MREs is called isotropic MREs. It is worth mentioning that the properties of isotropic MREs can differ much from those of anisotropic MREs.

Due to magnetic field-sensitive response of the magnetic particles, the material properties of MREs can be altered by using a magnetic field. For MREs, the magnetorheological effect is defined as the ratio of the value increment of a property at a measured magnetic field to the initial value of that property at zero magnetic field. In most studies, the distinctive change of the storage or loss modulus of MREs is a common concern. The magnetorheological effect is characterized by the ratio of modulus increment $\Delta G$ at a measured magnetic field to the initial modulus $G_0$, i.e., $\Delta G / G_0$. For example, Figure 1 gives the magnetic field-dependent shear storage modulus of MRE. The initial modulus $G_0$ of the MRE is about 0.6 MPa resulting from the initial state of MRE materials. When applying a magnetic field,
the interaction between the magnetic particles of MRE will occur and result in the alteration of the storage modulus. The modulus of this MRE can reach 1.5 MPa in the presence of field and thus the relative change of modulus can be larger than 100%. It should be noticed that, besides the magnetic field, the temperature [36–39], the relative humidity [40], and even the γ radiation [41] can influence the physical or mechanical properties of MREs. Based on the magnetic field-induced change of the physical or mechanical properties of MREs, lots of magnetorheological devices have been designed. In the late 1990s, Ginder et al. [2, 42] and Carlson et al. [3] had done the pioneering works and suggested controllable-stiffness components and electrically-controllable mounts based on MREs. Recently, Li et al. [1] and Ubaidillah et al. [43] have presented state-of-the-art reviews on magnetorheological elastomer devices. From these reviews, one can conclude that MREs can be used in many devices including but not limited to vibration absorbers, vibration isolators, sensors, controllable valves, and adaptive beam structures.

In this chapter, the materials, the preparing methods, the analytical models, and the applications of MREs will be reviewed. In the following section (Section 2), the materials, i.e., the magnetic particles, the nonmagnetic matrix and the additives, as well as the preparing methods of MREs will be introduced. In Section 3, the micro-structures of prepared MREs, the analytical models of MREs, and some typical macroscopic properties of MREs will be presented. The relationship between the microstructures and the macroscopic properties will be qualitatively discussed. Then the applications of MREs will be briefly introduced in Section 4. Finally, Section 5 will give a summary of this chapter.

2. Materials

2.1 Magnetic particles

To the magnetic particles, higher permeability, higher saturation magnetization, and lower remanent magnetization are highly desirable for obtaining stronger magnetic field-sensitive effect. Among a variety of magnetic particle materials, micrometer-sized carbonyl iron (CI) powder is currently widely used as a magnetic particle for preparing MREs. For a quick recognition, Figure 2(a) shows an example of the macroscopic image of CI powder. It shows that the CI powder is a very fine powder material. Figure 2(b) and (c) shows the scanning electron microscopy (SEM) images of CI powder (Type CN, produced by BASF SE Inc.) with different magnifications. The diameter of this kind of CI powder is several micrometers. In general, the size of the magnetic particles can range from several micrometers to hundreds of micrometers [47, 48]. Figure 3 gives the size distribution of CI powder from experimental test. The size distribution can be analytically modeled by a lognormal distribution model as the following equation (Eq. (1)) shows.

\[
P(d) = \frac{1}{d\sigma\sqrt{2\pi}} \exp \left\{ -\frac{(\ln(d) - \mu)^2}{2\sigma^2} \right\},
\]

in which \(P(d)\) is the probability density distribution function of the diameter of CI powder material. \(d\) is the diameter of the CI particle. \(\mu\) and \(\sigma\) are the expectation and variance of \(\ln(d)\). The tap density of CI powder is usually about 3.0 g/cm\(^3\) and the real density of CI powder is about 7.0 g/cm\(^3\).

Beside the size distribution of CI powder, the magnetic property of CI powder draws more attention of researchers. Higher permeability, higher saturation magnetization, and less remanent magnetization of magnetic particles are always highly desirable for
obtaining stronger magnetic field-sensitive effect. As is shown in Figure 4, CI powder shows very high permeability, saturation magnetization, and very little remanent magnetization. The value of saturation magnetization can reach more than 600 kA/m and there is little remanent magnetization when magnetic field is removed. This mainly results from the fact that the content of Fe element in CI powder is usually more than 97.5% in weight fraction. Attributing to the excellent magnetic property, CI powder is widely used for fabricating materials including but not limited to MREs.

Figure 2. Images of carbonyl iron powder. The upper subfigure (a) shows the macroscopic image [44] in daily view and the lower two subfigures (b and c) show the SEM images with different magnifications [45].

Figure 3. The size distribution of carbonyl iron powder (Type CIP-CN) [46].
2.2 Elastic matrices and additives

A basic requirement of elastic matrices for fabricating MREs is that the matrices have soft elastic property, meaning that the matrices can stably hold the magnetic particles under no magnetic field and have a finite deformation under a magnetic field. For the elastic matrices, there are lots of polymeric rubbers that can be considered as candidates, for example, silicone rubber [50], natural rubber [51], butadiene rubber [52], butyl rubber [53], polyurethane [54], polydimethylsiloxane [55], epoxy [56], etc. For instantly having a basic recognition of the matrices, the following Figure 5 gives some examples of image and applications of widely used silicone rubber.

The modulus of these matrices differs much from each other. For example, under normal conditions, the modulus of silicone rubber can be lower than 1.0 MPa [27]. That of natural rubber often reaches several MPa [25]. The shear modulus of PDMS varies with preparation conditions, but is typically in the range of 0.1–3.0 MPa [55]. The modulus of polyurethane can range from 0.01 MPa to several hundred MPa, attributed to its raw materials from fluid-like to solid-like [54]. Among a large amount of rubbers, silicone rubber compounds have characteristics of both inorganic and organic materials, and offer a number of advantages not found in other organic rubbers. From Figure 5, one can know that silicone rubbers have mechanically low modulus and good chemical stability and are nontoxic, nonpolluting, and human-body-friendly in daily use. As is shown in Figure 6, compared to the modulus of other rubbers, the modulus of silicone rubber is much lower within a large range of temperature. Besides, the thermal conductivity of silicone rubber can vary in a wide range. Based on the above-mentioned properties, silicone rubber can be chosen as an ideal soft elastic matrix for preparing MREs and is widely used in fabricating MREs. The other rubber matrices, with some unique mechanical or physical properties for special usage, can also be used in fabricating MREs according to need.

Besides the magnetic particles and the elastic matrices, additives are also key components for preparing MREs. Silicone oil is usually used as an additive in material fabrication of MREs. When the molecules of the silicone oil enter the matrix, the gaps between the matrix molecules are increased, and the conglutination of the molecules is decreased. Apart from increasing the plasticity and fluidity of the matrix, the additives can average the distribution of the internal stress in the materials, which makes a stable material property for MR elastomer materials [1, 58]. The other additives include but are not limited to carbon black [59–61], carbon nanotubes [62–65], silver nanowire [20], Rochelle salt [30], gamma-ferrite additives [66], etc.
2.3 Preparing methods

A simplified illustration of the processing for preparing MREs is shown in Figure 7. Usually, the magnetic particles and the matrix are mechanically mixed with some additives into a mixture. The mixture has a very low yield stress, meaning that the mixture can easily deform and usually creep with itself. Then the mixture vulcanizes at room temperature (called room-temperature vulcanizing, RTV [4]) or high temperature (called high-temperature vulcanizing, HTV [45]) higher than 120°C. During the vulcanizing, in case of applying a magnetic field, the magnetic particles can move in the matrix and gradually aggregate forming chain-like structures along the direction of the field. After the magnetic field-assisted curing,
Magnetorheological Elastomers: Materials and Applications
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anisotropic MREs, meaning that the magnetic particles form chain-like microstructures in MREs, can be prepared. When curing with no magnetic field, the magnetic particles will disperse uniformly in the matrix after vulcanization and thus isotropic MREs are prepared. Figure 8 shows some images of prepared MRE samples.

3. Microstructures and macroscopic properties of MREs

3.1 Microstructures of MREs

As a basic concern in the research of MREs, there are lots of studies focusing on the microstructures of MREs. Almost every report about a newly prepared MRE will show its microstructure. Figure 9 gives some typical SEM images with different times of magnification of the microstructures of carbonyl iron powder-embedded, natural rubber-based MREs samples. The volume fractions of iron particles for all samples are 11%. Figure 9(a) shows the images of a MRE sample cured with no magnetic field. It shows that the carbonyl iron particles randomly and uniformly disperse in the matrix. These two images are typical images showing the microstructures of isotropic MREs. The other images, i.e. Figure 9(b–f), show the microstructures of anisotropic MREs cured with magnetic field. As is shown, the magnetic particles will aggregate forming chain-like microstructures. The stronger the magnetic field intensity is when curing, the longer and thicker the magnetic particle-formed chains, as
the magnetic interaction of neighbor particles is stronger. When the magnetic field is not strong enough, for example, 200 mT, the magnetic particles in the matrix can only move within a small width of range, resulting in that the magnetic particles can only form some short chain-like microstructures. The spaces between these chains are small. With the enhancement of the magnetic field during curing, the spaces will get wider and the anisotropy of MREs will get higher, implying that the properties of MREs will get more anisotropic.

In recent years, the tendency to use quantitative methods instead of a qualitative analysis for structural investigation of the structure of nano- and microstructures has been increasing. Tomographic data open the possibility to achieve detailed quantitative data using techniques of digital image processing [69, 70]. To experimentally study the three-dimensional (3D) microstructures of MREs, Borin's group [71, 72] firstly used the X-ray micro-computed tomography ($X\mu$CT) method to investigate the microstructures of MREs. Balasoiu et al. [70] allowed a detailed structural analysis of both isotropic MREs and anisotropic MREs. Figure 10 shows the exemplary $X\mu$CT images of silicone rubber-based isotropic and anisotropic MRE samples. The iron particles in these MREs have an average particle size of approximately 35 μm. With $X\mu$CT images, single microparticles and aggregates as well as their spatial position can be identified. As can be seen, the particles are distributed homogeneously in the isotropic MRE sample and form chain-like structures in the anisotropic one. Figure 11 shows two extracted particle-formed columns from the reconstructed $X\mu$CT image. In this figure, the direction of the magnetic field was parallel to the longitudinal axis of the cylindrical shape holders and to the gravitational force. From this $X\mu$CT image, one can see how the particle chain forms and can go further to model MREs based on the image. Recently, the motion of particles in MREs was investigated by using $X\mu$CT [73]. It has been shown that $X\mu$CT is a powerful technique to investigate the inner structure of macroscopic samples without destroying the specimen. The $X\mu$CT technique also achieves high spatial resolution and allows the derivation of valuable local and statistical information, such as particle size and position, from the reconstructed 3D images. Furthermore, the nondestructive $X\mu$CT investigations...
of the 3D microstructures of MREs introduce the possibility of investigating the influence of the number and size of columns on the macroscopic mechanical and rheological properties of MREs.

3.2 Analytical models

The study of the microstructure-based analytical model of MREs is always a key work for deeply knowing about MREs. Some pioneering works had been done studying the model of MREs in the late 1990s [74, 75] and a single-chain model was proposed assuming that the MREs are fully filled with single chains (Figure 12(a)). From these studies, it was reported that the optimum particle volume fraction for the largest fractional change in modulus at saturation is predicted to be 27%. Calculations of the zero-field shear modulus perpendicular to the chain axis indicate that it does not exceed the modulus of a filled elastomer with randomly dispersed particles of the same concentration. In 2010, Li and Zhang proposed bimodal particle-based chain-model of MREs (Figure 12(b)) [76]. In their work, theoretical and experimental
studies of the mechanical performance and magnetorheological effects of MREs fabricated with mixtures of large and small particles were performed and an effective permeability model was developed to theoretically analyze the MR effect of bimodal particle-based MR elastomers. Six years later, a composite chain model (Figure 12(c)) was proposed [77, 78]. In this work, a particle chain composed of multiple kinds of particles with different sizes of diameter was introduced and a modeling strategy which accounts for elastic constituents and a nonlinear magnetization behavior of the particles is pursued. Most recently, a 3D multimodal chain model [79] has been proposed. In this model, magnetic particles with log-normal size distribution of diameter were introduced as fillers in soft elastic matrix. At the same time, a finite element model was built according to this model (Figure 12(d)). With the finite element model, one can computationally study the macroscopic physical or mechanical properties of MREs. In addition, as a basic issue, the study of the interaction between two magnetic particles still keeps developing [80–82]. Moreover, besides the microstructure-based analytical model of MREs, the phenomenological continuous medium-based models were also studied [83–93]. These works focus on theoretically and/or experimentally studying the magneto-viscoelastic models for MREs.

3.3 Macroscopic properties

The most important characterization of MREs is that their macroscopic physical or mechanical properties can be altered upon the application of a magnetic field. For a long time, most studies have focused on the magnetic field–induced changes of the modulus or damping of MREs [1]. The shear storage/loss modulus or damping property of MREs can be measured by dynamic mechanical analyzer (DMA) or rheometer. As examples, Figure 13 shows the magnetic field–dependent shear storage modulus and damping of MRE samples. For some natural rubber-based MRE samples with different weight fraction of carbonyl iron particles, one can find that the magnetic field–induced change of their shear storage modulus can reach near or above two times of their initial magnitude. Moreover, as a characterizing of the damping property of MREs, the relationship between shear stress and shear strain, under various magnetic field strengths of silicone rubber-based MRE, is shown in Figure 13 (right). The results show that such MREs have controllable damping properties. The increase of the stress-strain loop area with magnetic field demonstrates that the damping capacity of MREs is a function of applied magnetic field. These field-dependent mechanical properties make MREs much promising in many engineering fields, especially in vibration reduction.
Magnetostriction of MREs is also a key concern when studying MREs [75, 95–97]. Figure 14 shows the simulated magnetostriction in MRE with different volume fractions of structured particle distributions. It shows that the magnetostriction of MREs is magnetic field-dependent. The intenser the field is and the higher the volume fraction of magnetic particle is, the larger the magnetostriction is. Further, full-field magnetostriction/deformation of a silicone rubber-based MRE under uniform magnetic field has been studied [98]. It shows that both isolated particles and grouped particles result in the concave-convex deformation of the MRE sample. Recently, the magnetic field-dependent electrical conductivity of MREs was emergently studied [99–105]. These field-dependent properties make MREs much promising in actuating and sensing.

4. Applications

In 1993, Kordonsky pointed out that magnetorheological effect can be a base of new devices and technologies [106]. Years later, Carlson and Jolly gave an introduction of magnetorheological devices [3]. By possessing variable physical
or mechanical properties when subjected to a magnetic field, MREs are natural candidates to be developed in many applications. In 2014, Li et al. [1] presented a state-of-the-art review on MRE-based devices. From this review, one can find that MREs can be used in many devices including but not limited to vibration absorbers, vibration isolators, sensors, controllable valves, and adaptive beam structures.

4.1 Vibration absorbers

Ginder et al. [107] firstly constructed a simple one-degree-of-freedom mass-spring system—an adaptive tuned vibration absorber—that utilizes MREs as variable-spring-rate elements. After that, the research on vibration MRE-based absorbers developed quickly (e.g., Refs. [13, 108–112]). Figure 15 shows the sketch of a designed MRE-based vibration absorber composed of a semi-active vibration absorption unit and a passive vibration isolation unit. The vibration absorption unit is composed of a magnetic conductor, a shearing sleeve, a bobbin core, an electromagnetic coil winding, and a circular cylindrical MRE vulcanized between the shearing sleeve and the bobbin core. The magnetic conductor, the bobbin core, and the electromagnetic coil are supported on the shearing sleeve through the MRE. The magnetic conductor and the bobbin core are connected by a bolt, and the shearing sleeve is fixed to the lower housing. The outer surface of the shearing sleeve is in clearance fit with the inner surface of the magnetic conductor, and the magnetic conductor can move vertically along the shearing sleeve. The MRE works in pure shear mode, and the magnetic conductor, the bobbin core, and the electromagnetic coil form the dynamic mass of the MRE-based vibration absorber together. The proposed MRE-based vibration absorber can absorb the vibration energy and thus reduce vibration.

4.2 Vibration isolators

Vibration isolators are devices which can isolate an object, such as a piece of equipment, from the source of vibration. Vibration isolators can be categorized into two groups: base isolation and force isolation, and the isolating modes have active and passive vibration isolation [113]. In Ref. [1], Li et al. had given a review on the application of vibration isolators for mechanical engineering and civil engineering. There are many works that focused on the study of MRE-based vibration isolators, for example, Refs. [16, 91, 92, 114–118]. Figure 16 shows an example
of the designed layout and prototype of MRE-based isolator working in squeeze/elongation-shear mode. It shows that the initial vertical stiffness and damping coefficient of the magnetorheological elastomer isolator are $1.14 \times 10^6$ N/m and 495.8 N·s/m, respectively. The relative increase in stiffness and damping is 66.57% and 45.55%, respectively. Due to the properties of controllable stiffness and damping of MRE, the isolation transmissibility and root mean square of acceleration response can be reduced by 41.2% and 65.3%, respectively. The proposed MRE isolator can be used as a controllable stiffness device and has great potential in the field of vibration suppression for heavy equipment. [116].

4.3 Other applications

In addition to the field-sensitive elastic property, MREs possess several functions such as magnetoelasticity, magnetoresistance, magnetostriction, piezoresistance, and thermoresistance [1, 119]. The reasons for these functions are the changes in the spacing between the magnetic particles due to external loadings, which produce variations of the physical or mechanical properties of the MRE materials. Based on their field-sensitive properties, MREs have been developed for use as sensors and actuators, for example, force sensor [120], magnetoresistive sensor [18], magneto-sensitive strain sensor [20], flexible tri-axis tactile sensor [21], self-powered tribo-sensor [22], combined magnetic and mechanical sensor [17, 121], soft actuator [122], actuators for valves [123], MEMS magnetometer [124], etc. Moreover, the microwave response [125, 126] and 3D printing properties of MREs [127–129] have also been recently reported. It is worth being pointed out that the application of MREs is explosively developing.

5. Summary

In this chapter, the materials and applications of MREs are briefly reviewed. Firstly, raw materials, including the magnetic particles, the rubber-like matrices, and the additives, are introduced. As the kind of the raw materials is getting more and more inconstant, there are a variety of raw materials that can be used to prepare MREs.Attributing to the variety of the raw materials, the kinds of prepared MREs are much various and the study on the MRE materials is a long-lasting discovery
or inventive subject. Meanwhile, the XμCT technique can be used to study the microstructures and microstructure-based mechanisms of MREs. With the development of MRE materials, the property of MREs gets more and more various. The application of MREs has quickly developed in the engineering field of vibration reduction and the application range of MREs is getting wider and wider. In addition to the conventional applications in vibration reduction, the sensing and actuating applications of MREs are recently explosively developed. Moreover, the microwave-response and the 3D printing of MREs are newly emerged subjects, which can be much promising in engineering applications in the near future.

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**Conflict of interest**

There is no conflict of interest.

**Author details**

Taixiang Liu¹ and Yangguang Xu²*

¹ Research Center of Laser Fusion, China Academy of Engineering Physics, Mianyang, China

² College of Aerospace Engineering, Chongqing University, Chongqing, China

*Address all correspondence to: xyg@mail.ustc.edu.cn
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Magnetorheological Elastomers: Materials and Applications
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