Chapter

Magneto-Sensitive Smart Materials and Magnetorheological Mechanism

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Abstract

Magneto-sensitive smart materials, also named as magnetorheological (MR) materials, are a class of smart composites prepared by dispersing nanometer- or micrometer-sized ferromagnetic fillers into the different carrier matrix. As the rheological properties can be controlled by an external magnetic field rapidly, reversibly, and continuously, magneto-sensitive smart materials have great application potential in construction, automotive industry, artificial intelligence, etc. In this chapter, a brief history and classification of magneto-sensitive smart materials are firstly summarized. Next, we discuss the state-of-the-art of the magnetorheological mechanism through experimental and theoretical studies, respectively. Finally, the prospect for this material in the future is presented.

Keywords: smart soft material, magnetorheological material, magnetorheology, magnetic dipole theory, viscoelasticity

1. Introduction

Most smart materials imitate natural biological materials, which can respond to the stimuli (like mechanical, thermal, electrical, photic, acoustic, magnetic, chemical, etc.) by changing one or multiple properties to adapt to the changing environment [1]. So far, bioinspired smart materials have become an important research direction in material science. It is difficult for a homogenous material to possess multifunction, so it generally combines the materials with the functions of perception, actuation, control, etc., together in a specific way to design a novel composite with multiple characteristics. The smart material is multilevel with different components; each component has different characteristics and microstructures, and the coupling effect exists between different components, and these components make the smart material show complicated responses to external stimuli. Generally, the adapting ability of smart material to external environment, which is similar to the activated function of biological material, can be dynamically adjusted through the transportation of substance and energy.

Magnetorheological (MR) materials can be regarded as a kind of bioinspired smart materials because their viscoelastic properties can be easily adjusted by an external magnetic field. Magnetorheological materials can be generally classified into MR fluids, MR elastomers, and MR gels according to the type of the carrier matrix and the physical state in the absence of magnetic field [2–4]. The MR

fluid-based practical devices mainly include damper, buffer, clutch, artificial muscle, and so on [5]. Some application examples of MR fluid working under different modes can be found in various dampers, which take MR fluid as working medium. Besides, MR fluid has also found wide application potential in the fields of thermal conduction [6], sound transmission [7], precision machining [8], and biomedicine [9]. The applications of MR elastomer have been widely reported in the fields of adaptive tuned vibration absorber [10], impact absorber [11], active noise abatement barrier system [12], vibration isolator [13], sensor [14–16], and so on. MR elastomer mainly works by changing its modulus through magnetic field during pre-yield stage, which shows different working mechanism with MR fluid (MR fluid mainly works at post-yield stage). The examples for the application of MR gel are relatively less in comparison with MR fluid or MR elastomer, but the unique magneto-induced phenomena have attracted more and more attentions; some work concentrating on the potential applications of MR gel have been reported [17–20].

Here, we will give a brief introduction to different kinds of MR materials, discuss the related experimental and theoretical work when investigating the MR mechanism, and finally propose some future prospects for these magneto-sensitive smart materials aiming at practical applications.

2. The development of magnetorheological materials

2.1 Magnetorheological fluid

Magnetorheological (MR) fluid is the earliest developed magneto-sensitive smart material, which is a particulate suspension by mixing micrometer-sized ferromagnetic fillers, non-magnetic fluid, and some additives together. After applying an external magnetic field, the MR fluid will change from Newtonian-like fluid to semi-solid material quickly (within several microseconds) [21], as shown in **Figure 1a** [22]. The randomly dispersed magnetic fillers are rearranged to form chain-like ordered microstructure through the magnetic interaction. Moreover, the ordered degree of the microstructure is relevant with the magnetic field strength. That is, a stronger magnetic field will induce a more ordered chain-like microstructure parallel with the direction of external magnetic field (**Figure 1b**) [23].

The rheological measurements indicate that MR fluid shows Bingham fluidic behavior under magnetic field, an obvious yield stress exists, and the maximum of yield stress has exceeded 100 kPa [24]. Moreover, the apparent viscosity of MR fluid changes typically 3–4 orders of magnitude by changing the magnetic field strength, presenting a typical MR effect. However, the sedimentation problem due to the density mismatch between carrier matrix and ferromagnetic fillers become one of the bottlenecks to hinder the development of MR fluid. To solve the problems of particle sedimentation and the re-dispersion after the particle aggregation in the carrier matrix, many effective methods have been proposed. Generally, these methods can be classified into two main categories from the aspects of filler and carrier matrix.

Carbonyl iron particle is an ideal candidate to prepare MR fluid due to its low coercive force and high saturation magnetization ($\mu_0 M_s$ = 2.1 T). However, a serious sedimentation problem exists in the MR fluid with carbonyl iron particle. Sometimes, it has to abandon carbonyl iron particle with excellent magnetic property and choose some other magnetic material which could improve the sedimentation of MR fluid for the balance between performance and stability.

Decreasing the size of magnetic fillers is an effective method to improve the stability of MR fluids. Submicron- or nanometer-sized particle shows better

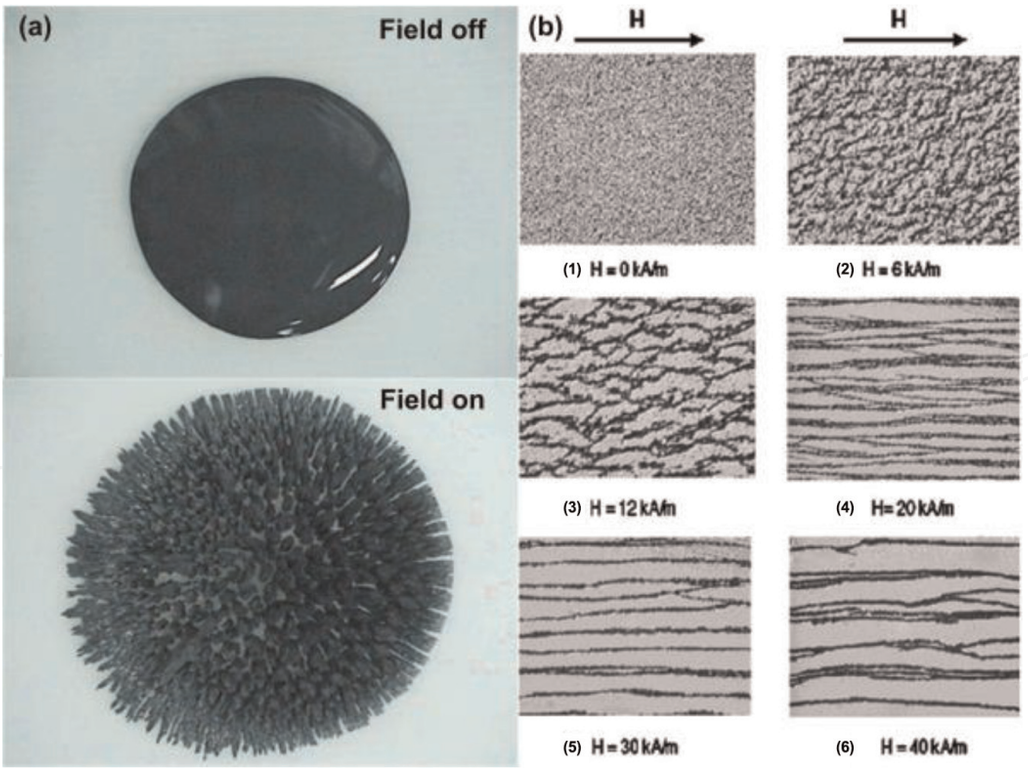

Figure 1.
The images of MR fluid without and with magnetic field [22] (a). The microstructures of MR fluid with different magnetic fields [23] (b).

stability in the non-magnetic carrier matrix in comparison with the micrometer-sized particle because of the Brownian movement and Van der Waals force. Especially, the particulate suspension totally using nanometer-sized ferromagnetic fillers as dispersed phase is named as magnetic fluids or ferrofluids [25–32]. Each ferromagnetic filler in ferrofluids includes single magnetic domain and could disperse in the carrier matrix randomly due to the Brownian movement, so the sedimentation is greatly improved. However, the ferrofluids keep their liquid-like state even under a strong magnetic field, which indicates that it is impossible for ferrofluids to have a high magneto-induced yield stress. In addition, the particle aggregation caused by the particle's nanometer size (or the re-dispersion of particles after removing the magnetic field) is another challenge to promote the further development of ferrofluids [33]. Using the magnetic material with the shape of rod-like or fibroid is regarded as another effective method to improve the stability and MR performance of MR fluids [34].

Except for changing the size and shape of ferromagnetic fillers, the core-shell structured ferromagnetic particle, whose surface is chemically modified by various polymers, is also a good candidate. The core-shell structure not only decreases the density of the particle but also increases the static electrical repulsion between adjacent particles, so the stability and the redispersibility of MR fluid are evidently improved. Moreover, the antioxidation is improved because the surface of the particle is wrapped by polymer [35]. **Figure 2** shows the SEM images of carbonyl irons wrapped by various polymers, which were mainly made by Choi's research group at Inha University.

It is a new approach to balancing the stability and performance of MR fluid by dispersing the submicron- or nanometer-sized ferromagnetic fillers (such as nanotube, submicron- or nanometer-sized carbonyl iron particle, wrought monox, organic clay, clavate ferromagnetic Co-γ-Fe_2O_3, and CrO_2) into conventional MR fluid to change the property of continuous phase (i.e., the matrix) [36–39]. Both of

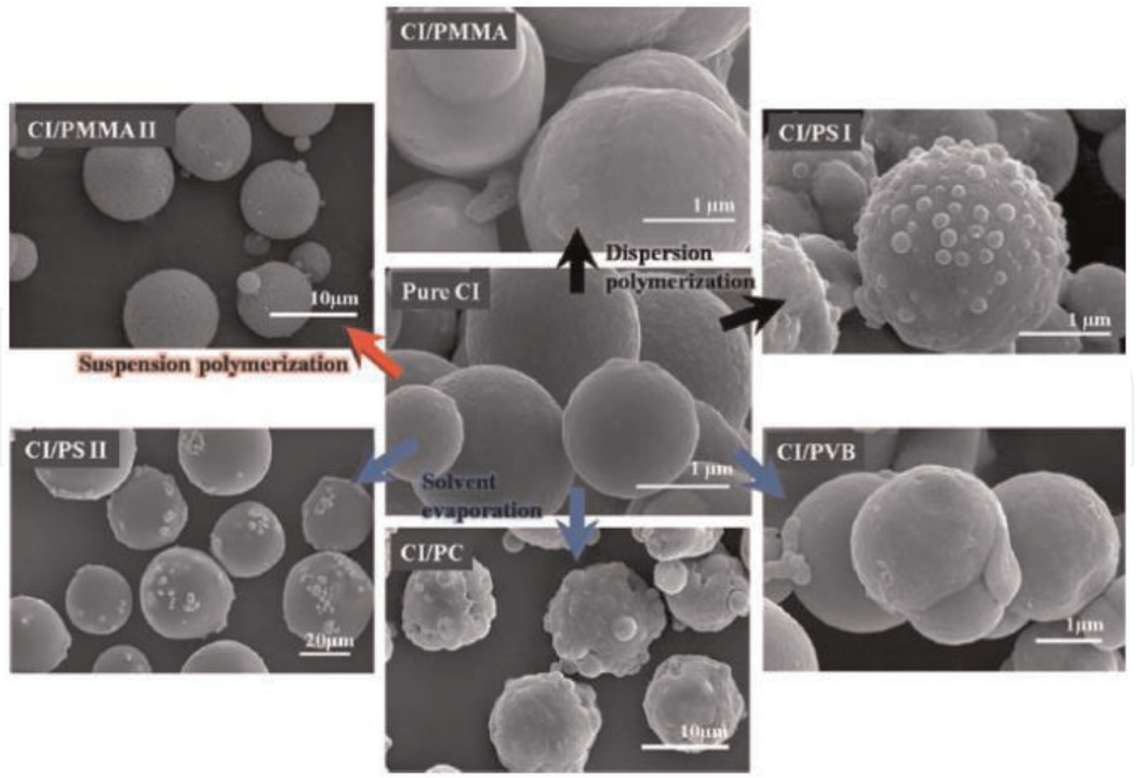

Figure 2.
Pure carbonyl iron and the carbonyl iron wrapped by various polymers [35].

the sedimentation rates of the dispersed phase and the MR effect are improved significantly due to the existence of ion. However, the modification to the carrier matrix, together with the modification to the dispersed particles, could only improve the sedimentation of MR fluid to some extent. To solve the particle sedimentation completely, the non-magnetic liquid matrix can be totally substituted by rubber-like polymer matrix. Thereout, a new magneto-sensitive soft material, that is, MR elastomer, appears later.

2.2 Magnetorheological elastomer

MR elastomer presents solid state even without magnetic field; it inherits the magneto-sensitive feature of MR fluids, but its working principle and application field are quite different from MR fluid. The magnetic fillers are fixed in the polymer matrix after the MR elastomer is prepared, the particles cannot move freely even exposed in a magnetic field, and no "phase transition" appears like happened in MR fluid. MR elastomer carries out the intelligent control mainly through changing its damping and modulus by magnetic field before yield [40]. The ordered chain-like (or column-like) microstructure aligned parallel to the direction of magnetic field generates if an external magnetic field is applied during the vulcanizing process of the polymer matrix. After vulcanization, the ordered microstructure can be solidified in the matrix, and the anisotropic MR elastomer is obtained.

Figure 3 shows the SEM images of MR elastomers pretreated by different magnetic fields. It is clear that chain-like structured anisotropic MR elastomer can be prepared if it is exposed under an external magnetic field during the preconfiguration process. The structure of particle chains can be further adjusted by the magnetic field. The ferromagnetic fillers aggregate more easily under a strong magnetic field, resulting in a higher degree of anisotropy of MR elastomer. Further magnetorheological characterization indicates that the MR elastomer with higher

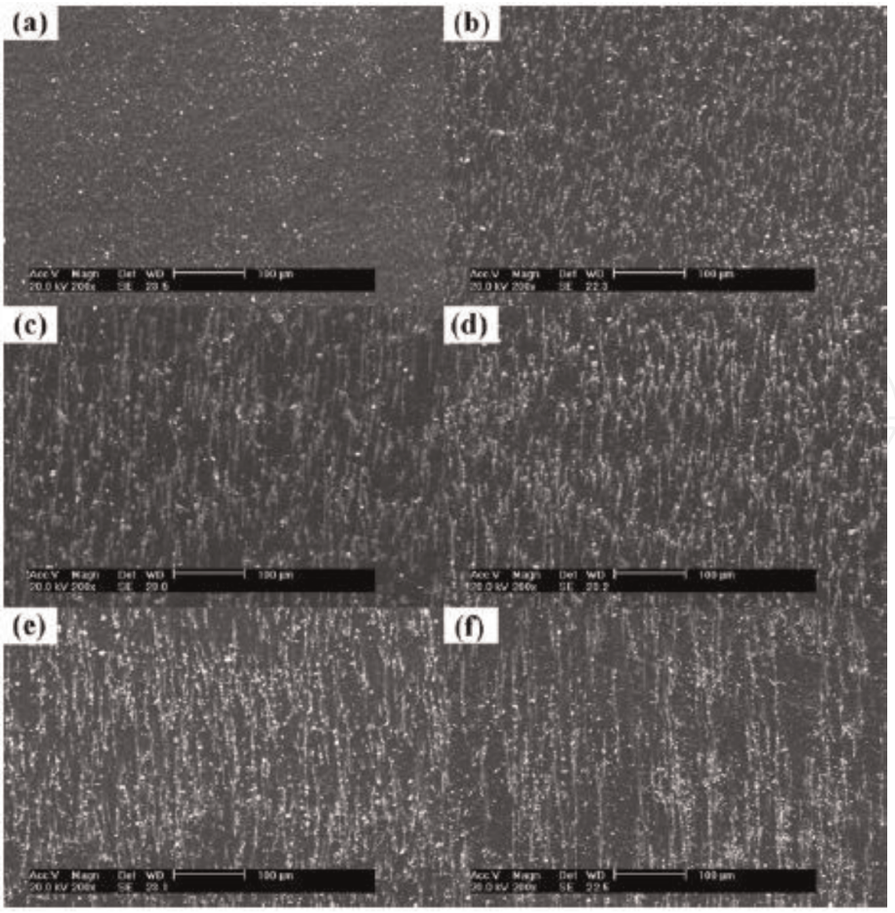

Figure 3.
The SEM images of MR elastomer pretreated by different magnetic fields [41]: (a) 0 mT; (b) 200 mT; (c) 400 mT; (d) 600 mT; (e) 800 mT; and (f) 1000 mT.

degree of anisotropy also possesses high MR effect [41]. So far, many kinds of rubber, such as natural rubber [41], silicon rubber [42], polyurethane rubber [43], cis-polybutadiene rubber [44], nitrile butadiene rubber [45], and thermoplastic elastomer blended by polypropylene and SEBS [46], have been chosen as the candidates for fabricating the MR elastomer.

Although the particle sedimentation is completely solved, the magneto-induced effect and MR effect constrain each other, making the requirement of engineering application not easy to be achieved. Moreover, MR elastomer loses the feature that the particulate microstructure can be easily controlled by magnetic field as like in MR fluid because the ferromagnetic fillers are fixed in the rubber matrix, which indicates that it loses the magneto-controllable flexibility. To pursue a stable MR material with a high MR effect and a strong magneto-induced effect aiming to the engineering application, it needs to abandon the conventional fabricating solutions and redesign a new magneto-sensitive smart material.

2.3 Magnetorheological gel

MR gel is another magneto-sensitive smart material whose continuous phase is viscoelastic. The inelastic matrix makes MR gel present typical viscoelastic characteristics even without external magnetic field. Shiga et al. firstly proposed the concept of MR gel in 1995 [47], and they prepared a series of MR gels with different particle contents by dispersing ferromagnetic fillers into silicone gel and investigated the magneto-dependent viscoelastic behaviors and microstructures of MR gel.

The MR gels can be further classified into liquid-like and solid-like MR gels according to their physical state without magnetic field.

The liquid-like MR gels can be regarded as a kind of special MR fluid with a little mount of polymer solution as additive. The polymer additive can generate a network structure in the matrix to modify the interfacial characteristic of ferromagnetic fillers and their interaction, greatly slowing down the sedimentation velocity of ferromagnetic fillers [48–50]. In comparison with MR fluids, except for improving the sedimentation stability, the off-state viscosity and yield stress of liquid-like MR gels can also be adjusted by adding a certain amount of polymer additives. However, the polymer network increases the moving resistance of ferromagnetic fillers in the matrix, and the response of MR gels to external magnetic field is then decreased accordingly. In addition, the sedimentation problem in the liquid-like MR gels is not completely solved.

Strictly speaking, the magnetic gel reported by Shiga et al. is a kind of solid-like MR gels [47]. The most distinct characteristic of solid-like MR gels is that there is no particle sedimentation problem exists, like MR elastomer. Yet the matrix is not the rubber-like elastic material, the solid-like MR gel cannot be classified into MR elastomer. Recently, these solid-like MR gels have been paid more and more attention [51–57]. A novel solid-like MR gel by mixing micrometer-sized magnetic particles and plasticine-like polyurethane was reported by Xu et al. [58]. As **Figure 4a** shows, this material presents like plasticine without magnetic field, can be changed into any shapes, and remains the status of plastic deformation, so it is named as MR plastomer. MR plastomer deforms along with the direction of the applied external magnetic field (**Figure 4b**). Further microstructure characterization indicates that the randomly dispersed magnetic particles can rearrange to generate chain-like (or column-like) orientated microstructure driven by magnetic force

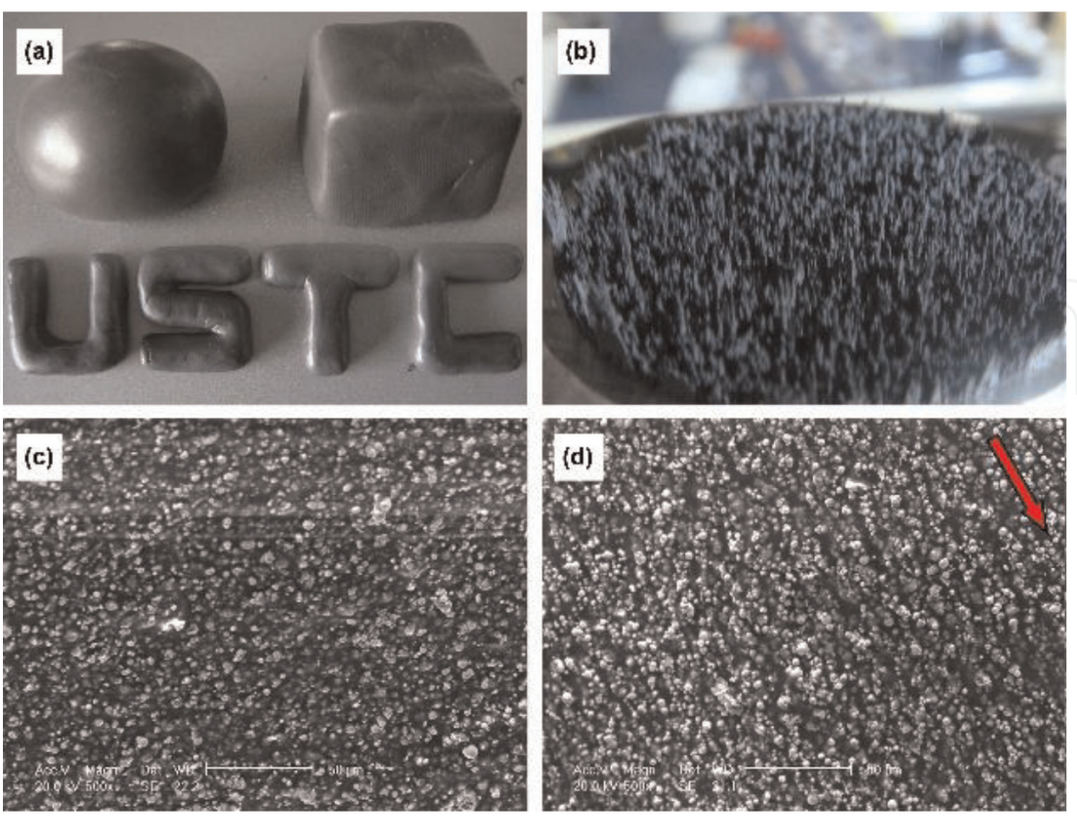

Figure 4.
The images of MR plastomer without (a) and with (b) magnetic field [58]. The SEM images of MR plastomer without (c) and with (d) magnetic field (the direction of magnetic field is marked by the red arrow) [59].

(**Figure 4c** and **d**). In addition, the anisotropic particulate microstructure is kept even the external magnetic field is removed [59].

In one word, an orientated particulate microstructure in the solid-like MR gel can be adjusted through an external magnetic field. Meanwhile, the chain-like (or column-like) microstructure can be fixed in the soft matrix after removing the magnetic field. This unique feature makes solid-like MR gel process the merits existed on MR fluid and MR elastomer (i.e., movability of particles and the "frozen" property of orientated microstructure) at the same time, which is significant to investigate the magneto-mechanical coupling mechanism.

2.4 Other magnetorheological materials

Some other special MR materials, which cannot be simply classified into the most well-known MR materials as mentioned above, were reported in the literature. MR foam is a kind of solid-like polymer composite by pouring MR fluid into porous polymer foam (**Figure 5**); the modulus of the MR foam can be controlled by changing the rheological property of MR fluid through a magnetic field [60–62]. Due to the special porous microstructure, MR foam presents the merits of light-weight, controllable modulus, excellent sound-absorbing property, and so on [63].

To solve the problem of particle sedimentation for MR fluid, Park et al. prepared a kind of novel MR material with excellent sedimentation resistance ability by substituting the fluidic matrix of MR fluid with commercial grease, and they named this MR material as MR grease [64]. MR grease shows typical Bingham fluid behavior; so strictly speaking, MR grease can be regarded as a special MR fluid. Byrom and Biswal reported a colloidal material system by adding micrometer-sized paramagnetic and diamagnetic particles into ferrofluid [65]. Different from the conventional MR fluid, the particles do not generate a chain-like orientated structure parallel with the direction of magnetic field but a fractal net-like microstructure in 2D direction. Further analysis indicated that the fractal net-like microstructure is induced by the magnetic dipole interaction between paramagnetic and diamagnetic particles, and the fractal dimension of the particle aggregates can be controlled by adjusting the concentration of ferrofluid and the ratio of paramagnetic and diamagnetic particles.

A multifunctional magnetic plasticine™ was developed by Xuan et al. [66], and they chose paraffin wax petroleum jelly as the matrix. Except for the high magneto-induced G′ (4.23 MPa) and MR effect (305%), magnetic plasticine™ can be switched between liquid-like state and solid-like state by changing the temperature, which greatly enhances the regulation ability. Shahrivar and de Vicente also reported a thermo-responsive polymer-based magneto-sensitive material [67],

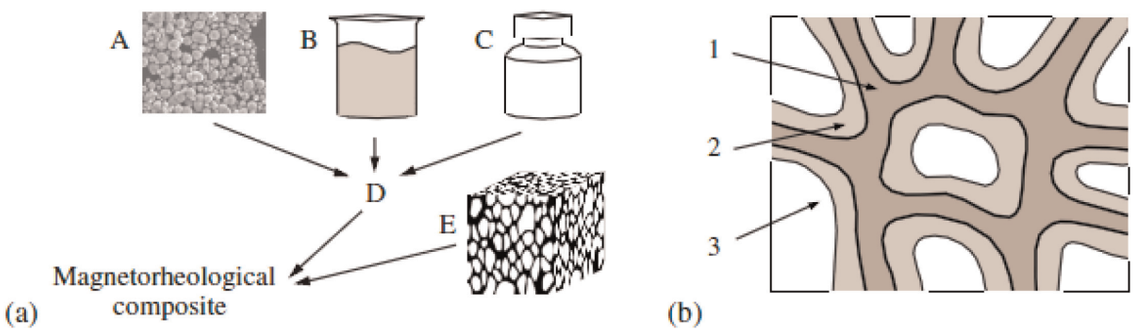

Figure 5.
The fabrication procedure of MR foam (a): pouring the MR fluid D into porous polymer material E. MR fluid D consists of carbonyl iron particles A, carrier fluid B, and additives C; (b) the microstructure inside the foam: microtubule wall 2 is surrounded by MR fluid 1, and the other space is full of air 3 [61].

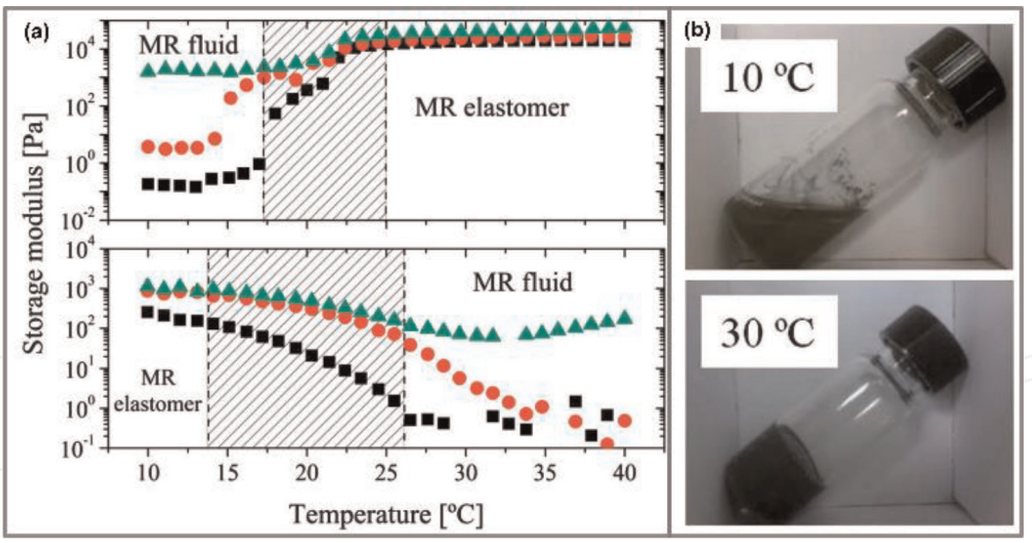

Figure 6.
Temperature dependent G′ of MR composites with PEO-PPO-PEO copolymer solution (upper subfigure) and P-NIPA microgel dispersion (lower subfigure) (a). Photos of the copolymer-based MR material at different temperatures after 1 h at rest (b) [67].

which can easily achieve phase transition by changing temperature. **Figure 6a** depicts that "liquid-to-solid" transition with increasing temperature appears in the MR composite with PEO-PPO-PEO solutions because a repulsive colloidal glass generates, while an inverse temperature-driven phase transition can be achieved for MR composite with P-NIPA microgel dispersions (**Figure 6b**). Besides, the critical temperature of phase transition can be changed by tuning polymer concentration. The multi-responsive MR material is a good effort in the frontier between conventional MR fluid and MR elastomer.

3. Magneto-sensitive properties and MR mechanism

The magnetic field usually leads to a structural rearrangement in soft MR material, and this process has significant influence on the physical properties of soft MR material. Due to the weak restriction of polymer matrix to the ferromagnetic fillers and complexity of polymer matrix, the responses of MR gel to external stimuli are more complicated than those of MR fluid and MR elastomer. For this reason, MR gel presents some unique magneto-electro-thermo-mechanical coupling phenomena. However, the realization on the coupling mechanism of MR gel is far from enough in comparison with those of MR fluid and MR elastomer, and more efforts need to be made through experimental and theoretical studies. Tight correlations exist between these three MR materials; so, there are some similarities in MR mechanism, and we can use the characterization techniques and theoretical models of MR fluid and MR elastomer for reference when studying the MR mechanism of MR gel. Next, we will briefly discuss the characterization methods and theoretical studies for different MR materials.

3.1 Experimental characterization of MR materials

Experimental characterization of MR materials can not only quantitatively evaluate their performance but also provide the necessary parameters for theoretical research or certify the accuracy of the theoretical results, which is the foundation for investigating the MR mechanism. The magneto-induced rheological properties of MR materials under different loading conditions (quasi-static shear, tensile,

compressive loading, dynamic shear, and squeeze loading) are the most important properties, and this is the primary reason for naming this magneto-sensitive smart material as MR material. The influences of relevant factors (i.e., magnetic field, temperature, PH value, particle concentration, shape, size, and so on) on rheological properties were also widely investigated. Besides, the magnetization, electrical conduction, thermal conduction, and magnetostriction of MR materials are studied as well.

The rheological property of MR materials under shear loading is the most used characterization parameters at present. Many famous commercial rheometers, like the Physica MCR rheometer from Anton Paar Company and Discovery hybrid rheometer from TA Company, are designed for the rheological characterization under shear loading.

The magneto-dependent rheological behavior of MR fluid is usually described by Bingham model:

$$\begin{aligned} \tau &= \tau_y + \eta\dot{\gamma} \qquad |\tau| \geq \tau_y \\ \tau &= G_0\gamma \qquad\quad |\tau| < \tau_y, \end{aligned} \tag{1}$$

where τ is shear stress, $\dot{\gamma}$is shear strain rate, τ_y is magneto-dependent shear yield stress, G_0 is the shear modulus before yield, and η is the plastic viscosity. τ_y is defined as the minimum stress of MR fluid to resist the thixotropic effect and start to deform or flow, which can be calculated by fitting the shear stress-strain rate curves of MR fluid by using Bingham model (Eq. (1)). **Figure 7** depicts the typical magneto-sensitive relationship between shear stress and strain rate of MR fluid [68]. Usually, it can be approximately considered the fitting value at the strain rate of zero as τ_y, so it is easily found from **Figure 7** that τ_y significantly increases when the magnetic field strength increases. Therefore, the magneto-dependence of τ_y can be regarded as a characterization parameter to evaluate the magnetorheological effect of MR fluid [69]. This method can also be directly utilized to characterize the magnetorheological property of liquid-like MR gels [70].

Both of the solid-like and liquid-like MR material can be considered as the viscoelastic material, and the dynamic mechanical analysis under oscillatory shear is one of the most important characterization methods for viscoelastic materials. Normally, applying a sinusoidal shear strain to the viscoelastic material, if the amplitude of the strain is small enough, a sinusoidal response stress at the same frequency

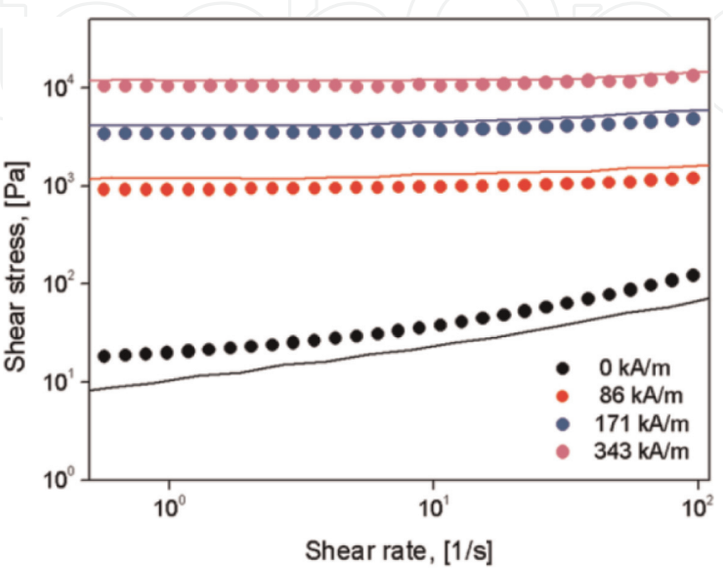

Figure 7.
Shear stress of MR fluid as a function of shear rate under different magnetic fields [68].

but a specified phase shift can be obtained; then, we define the dynamic mechanical properties (i.e., storage modulus G′, loss modulus G″, and loss factor tanδ) of material within the linear viscoelastic (LVE) range as the ratio of response stress to actuation strain in the complex plane. Dynamic mechanical properties are frequently used characterization parameters for investigating the magneto-induced microstructure evolution mechanism of MR materials. For the practical application, most MR materials in the devices are working under the oscillatory shearing mode. More importantly, both solid-like MR elastomer and liquid-like MR fluid can be characterized by dynamic mechanical analyzer (DMA) [71, 72], which indicates that dynamic mechanical analysis can be considered as a universal method to characterize MR materials. Therefore, as the intermediate material system between MR elastomer and MR fluid, the magnetorheological properties of MR gel are mostly studied by DMA [73].

The oscillatory shear mode can be further classified into simple shear and rotating shear according to different measurement principles of commercial devices. **Figure 8a** shows a typical DMA (Tritec 2000, provided by the Triton Technology Co. Ltd., UK). If an external magnetic field generator is added (**Figure 8b**), the magneto-mechanical coupling behaviors of MR elastomer under oscillatory simple shear mode can be investigated [74]. The deformation of the sample under simple shear mode is uniform, which is valuable for theoretical analysis. The rheometer can also be used for dynamic mechanical analysis. **Figure 8c** shows a parallel-plate rheometer (Physica MCR 301, Anton Paar Co., Austria) equipped with a MR accessory (MRD 180), which provides a controllable magnetic field when carrying out a rotating shear experiment (**Figure 8d**). The deformation of the sample under rotating shear mode is inhomogeneous (the shear strain increases linearly in the radial direction of disc-like sample; the shear strain at the center of the sample is zero). Although with different measurement principles, the measurement results obtained from these two kinds of devices show little difference if the amplitude of the actuating strain is small enough. Besides, the magneto-dependent creep and

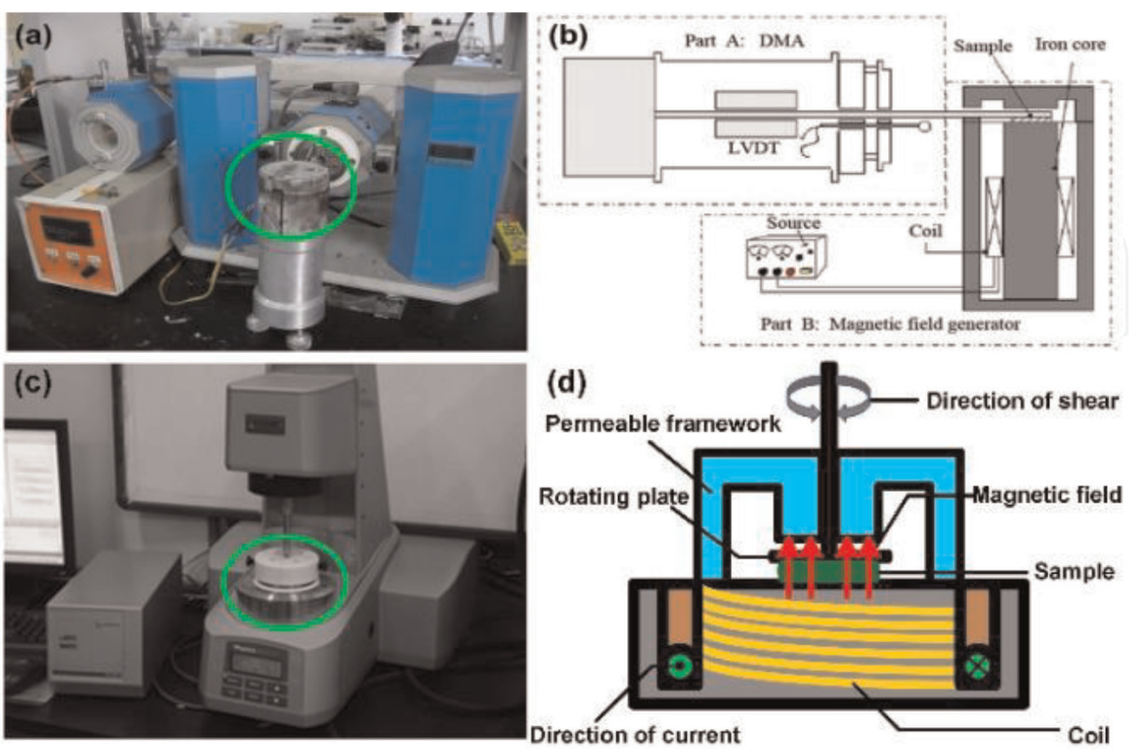

Figure 8.
A modified magneto-mechanical coupled DMA (a) and its measurement schematic diagram (b); a parallel-plate rheometer (c) with a magnetic field generation accessory (d).

recovery [75–77] and stress relaxation behaviors [78] of MR materials under shear loading are very helpful for investigating the magnetorheological mechanism and can be tested with the modified DMA and the rheometer as mentioned above.

The modulus variation of ferromagnetic filler-doped polymer composite can be reflected from the compressive and tensile properties of MR elastomer, which are also frequently used characterization parameters. Bellan and Bossis investigated the influences of magnetic field, particle concentration, and particle distribution on the tensile property of silicon rubber-based MR elastomer [79]. The compressive properties of MR elastomer with different particle distributions (randomly dispersed isotropic and chain-like orientated anisotropic structures) and its magnetic field dependency were investigated by Varga et al., as shown in **Figure 9**. It is found that the compressive modulus of MR elastomer is not only affected by the particle distribution (i.e., the compressive modulus of anisotropic MR elastomer is larger than that of isotropic one under the same loading condition) but also by the magnetic field and loading direction. When the directions of magnetic field, particle chain, and compressive loading are parallel (i.e., the middle test condition in the second row of **Figure 9**), the largest compressive modulus and magneto-induced effect of MR elastomer were observed [80]. In addition, the mechanical properties of MR elastomer under oscillatory squeezing mode were reported by Kallio et al. and Koo et al., respectively [81, 82], which provide valuable experimental data for the application of MR elastomer.

In recent years, many people pay attention to the rheological behaviors of MR fluid under tensile or squeezing loading, and the results suggest that the yield stress of MR fluids is significantly enhanced due to the squeeze-strengthen effect [83–85]. However, the tensile and squeezing behaviors of MR gel are rarely studied to date. Therefore, Xu et al. systematically investigated the squeeze flow behaviors (including quasi-static compressive and tensile behaviors and oscillatory squeeze behaviors) of MR gel [86]. It was concluded that the squeeze flow curve of the solid-like MR gel can be classified into three different deformation regions: elastic deformation, stress relaxation, and plastic flow regions. Yield stresses under both tension and compression are sensitive to the particle distributions, the filler concentration, and the magnetic field. In addition, the magneto-sensitive properties of MR

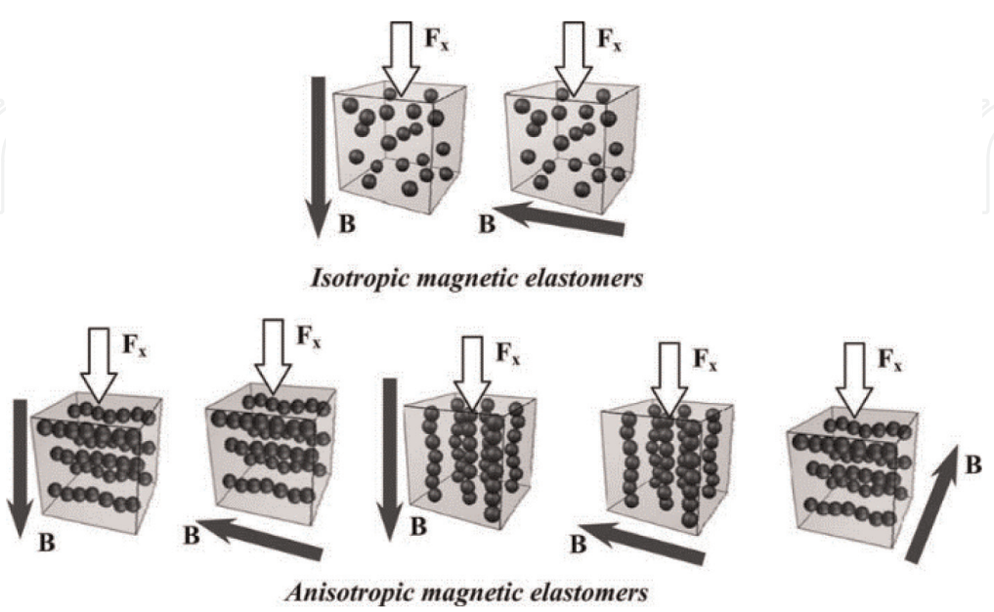

Figure 9.
The magneto-dependent compressive experiments of MR elastomers with different particle distribution states [80]. The white hollow arrow represents the direction of compressive loading, and the solid black arrow represents the direction of magnetic field.

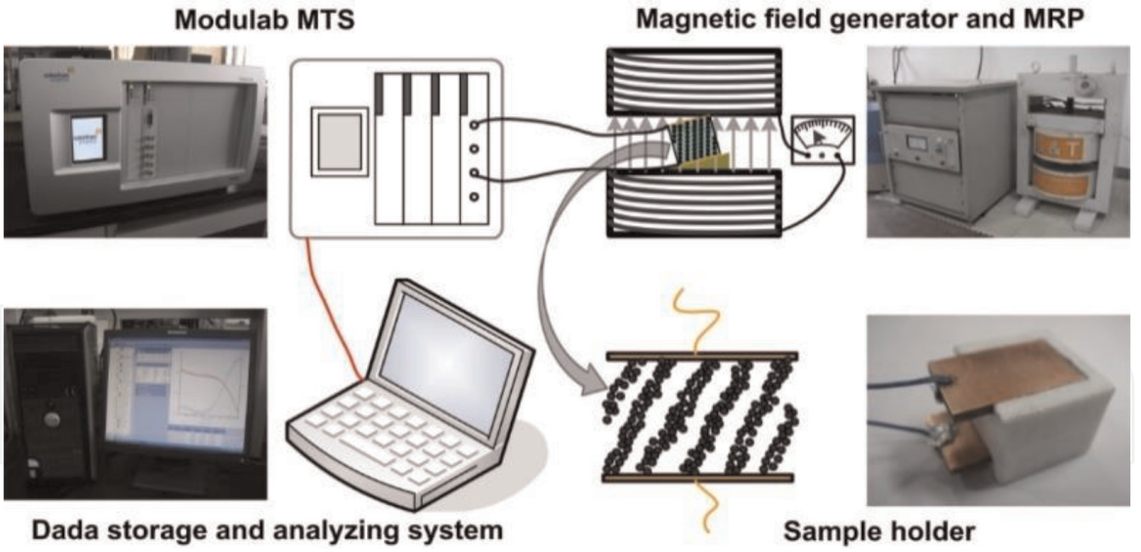

Figure 10.
The magneto-electrical coupling measurement system.

elastomer and MR gel under impact compression were also studied [11, 87]. The compressive modulus can still be strengthened by magnetic field even at high strain rate.

Most magnetic fillers (such as carbonyl iron particles [88], nickel particles [89], and Fe_3O_4 particles covered by silver [90]) are conductive at the same time, so most MR materials also belong to conductive polymer composite. The conductivity of this kind of magneto-sensitive conductive polymer composite is adjusted through external magnetic field except for the particle distribution and particle concentration, presenting a typical magnetoresistance effect [91–93]. Impedance spectroscopy testing is a nondestructive method to quantitatively detect the evolution of the microstructure, which is suitable for analyzing the microstructure evolution mechanism and the interfacial feature of material. **Figure 10** shows a typical experimental setup to test the magneto-sensitive impedance spectroscopy of conductive polymer composite. With this magneto-electrical coupling measurement system, the structure-dependent (**Figure 11a**) and the magneto-induced (**Figure 11b** and **c**) impedance spectroscopy are obtained, and the microstructure-dependent conduction mechanism (**Figure 11d**) can be further analyzed based on the related experimental results [94].

Besides, the investigations on the antioxidation [95], durability [96], and thermal conductivity [6] are helpful to the deep understanding on the magneto-induced mechanism of MR materials as well as some specific practical applications.

3.2 Magnetorheological mechanism

Magnetorheological effect essentially originates from the discrepancy of magnetic permeability between the continuous phase (the carrier matrix) and the dispersed phase (ferromagnetic fillers). Particular magnetization model (i.e., magnetic dipole model) is the most popular microstructural model to explain the magneto-induced effect of MR fluid [21]. If we ignore the multi-body magnetic interaction between particles (i.e., only the magnetic interaction between adjacent particles in a single particle chain is considered) and the multidirectional magnetization in a single magnetic particle (i.e., simplify the micrometer sized ferromagnetic particles as magnetic dipole), the magnetic moment of spherical ferromagnetic particle within the linear magnetization range is:

$$m = 4\pi\mu_0\mu_{cr}\beta a^3 H_0, \tag{2}$$

where a is the radius of magnetic particle, μ_0 is the permeability of vacuum, μ_{cr} is the relative permeability of carrier matrix, H_0 is magnetic field strength, and $\beta = (\mu_{pr} - \mu_{cr})/(\mu_{pr} + 2\mu_{cr})$ is the dimensionless coupling parameter of permeability (μ_{pr} is relative permeability of ferromagnetic particles). As the magnetic field increased, the magnetization of magnetic particle tends to saturation, and the magnetic moment of particle is independent on magnetic field strength, that is

$$m = \frac{4}{3}\pi\mu_0\mu_{cr}a^3 M_s, \tag{3}$$

where M_s represents the saturation magnetization. The parameter λ is usually introduced to represent the ratio of magnetic interaction energy to thermal energy between adjacent ferromagnetic particles [97],

$$\lambda = \frac{1}{4\pi\mu_0\mu_{cr}}\frac{m^2}{r^3}\frac{1}{\kappa T} = \frac{\pi\mu_0\mu_{cr}\beta^2 a^3 H_0^2}{2\kappa T}. \tag{4}$$

If λ is far larger than 1, the magnetic interaction between adjacent particles is far larger than the force induced by Brownian movement, and the particle will generate chain-like (or column-like) oriented microstructure parallel to the direction of magnetic field.

When it flows, the rheological properties of MR fluid are related to λ, the volume fraction of particles ϕ, and Mason number (a dimensionless parameter, Mn).

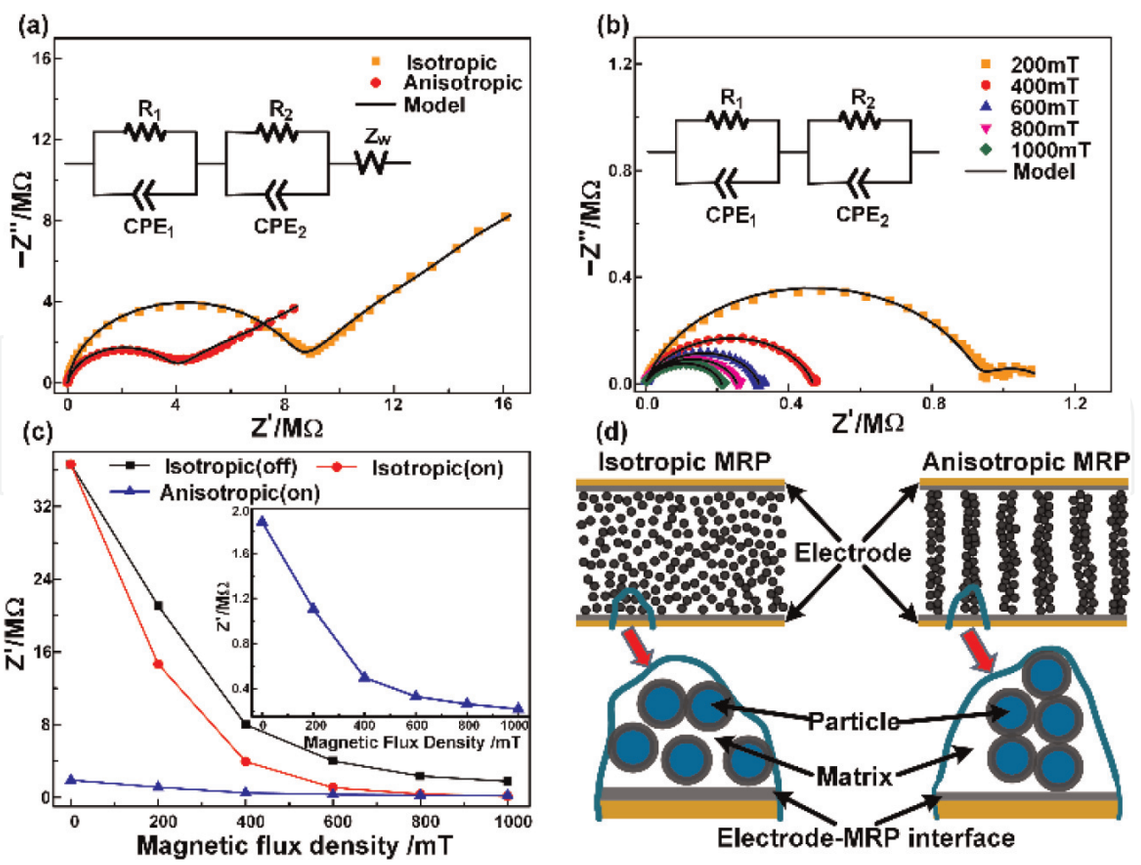

Figure 11.
Nyquist plots of magneto-sensitive conductive polymer composite with different particle distribution states (a) and under different magnetic fields (b); the conductivity of magneto-sensitive conductive polymer composite with different preprocessing methods (c); schematic of magneto-sensitive conductive polymer composites with different particle distributions and corresponding interfaces (d) [94].

In a stable shear flow, *Mn* is defined as the ratio of hydrodynamic drag applied on the ferromagnetic particle to the magneto-static force [98]:

$$Mn = \frac{8\eta_c \dot{\gamma}}{\mu_0 \mu_{cr} \beta^2 H^2}, \tag{5}$$

where η_c is the viscosity of carrier matrix and $\dot{\gamma}$ represents the shear strain rate.

As the most important rheological parameter of MR fluid, the magneto-induced yield stress can generally be explained from macroscopic and microscopic aspects. The macroscopic theoretical model is usually derived according to the minimum principle of energy. It assumes that the ferromagnetic fillers are spherical, cylindrical, or layered and are dispersed evenly in macroscopic theoretical model [99]. These models based on the sub-microstructure only consider the anisotropy of particle aggregation under small strain while the microscopic models consider the interaction between magnetic particles [100]. Most microscopic models ignore the interaction between the structured particle chains and believe that the magnetic interaction between adjacent particles is the main source of yield stress. Therefore, the yield stress of MR fluid can be well predicted when particle content is relatively low, but the assumption is untenable at high particle concentration, which makes a big deviation between theoretical and experimental results.

Figure 12 demonstrates the schematic of classic single-chain magnetic dipole model. When the material deforms by shearing, an affine deformation happens on the particle chain accordingly, which means that the particle moves horizontally along the direction of arrows as shown in **Figure 12a**. For this affine deformation, the distance between adjacent particles is identical before and after deformation. This assumption simplifies the deduction of magneto-induced yield stress. The magneto-induced shear yield stress of MR fluid within the linear magnetization range can be obtained based on the affine deformation assumption:

$$\tau_y = 2.31\phi\mu_0 M_s^{1/2} H_0^{3/2}. \tag{6}$$

In the saturated magnetization range, yield stress can be expressed as:

$$\tau_y^s = 0.086\phi\mu_0 M_s^2 \tag{7}$$

The details for the deduction process of magneto-induced yield stress and the explanation of relevant parameters can be found in the review article about the magnetorheological mechanism written by Bossis et al. [101].

The abovementioned single-chain magnetic dipole model was introduced directly into MR elastomer by Jolly et al. [102]. They predicted the

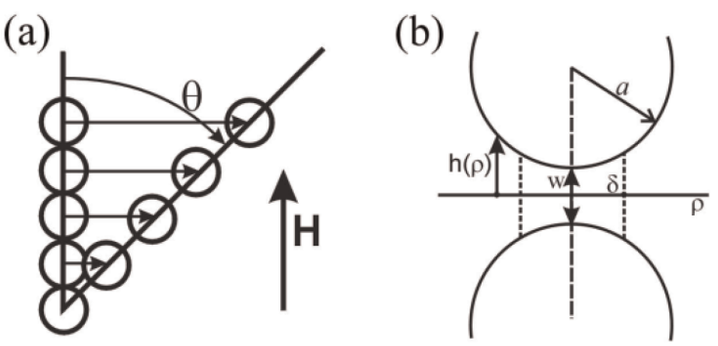

Figure 12.
The schematic of single-chain magnetic dipole model [101]. The affine deformation of particles within a single chain (a) and the geometrical relationship between adjacent particles (b).

magneto-induced elastic modulus by this model, and the theoretical results fit well with the experiment. Davis also calculated the magneto-induced shear modulus by this model, and he found that the magneto-induced shear modulus reaches a maximum if the volume ratio of the ferromagnetic fillers is 27% [103]. However, the influence of magnetized particle on the surrounding particles is not considered by the magnetic dipole model, and the bias of theoretical results from the fact is growing larger with the increasing particle concentration. Shen et al. realized this problem and modified the single-chain magnetic dipole model by considering all of the interaction of the particles within a single chain, which is more in line with the real situation [104]. A finite-column model based on the experimental results was proposed by Chen et al. [41]. However, this model can only predict the magneto-induced modulus of MR elastomer with low particle concentration.

The abovementioned models concentrate on the magnetic interactions between particles, but different from the MR fluid, a strong constraint effect exists between rubber matrix and magnetic particles for MR elastomer. This constraint will influence the magnetic interactions between particles to some extent. Therefore, the theoretical model of MR elastomer considering the coupling effect between rubber matrix and particles is more reasonable. Chen and Jerrams developed a more general magneto-mechanical coupling model of MR elastomer, which includes the magneto-induced mechanical property of particles, the interfacial slipping effect between ferromagnetic particles and rubber matrix, and the viscoelastic properties of rubber matrix [105]. This model could predict the dynamic mechanical properties of MR elastomer with different particle content or different kinds of rubber matrix, revealing the response mechanism of material to external magnetic field. It is a tendency in recent years that constructing the magneto-mechanical coupling model of MR elastomer bases on the theory of continuum medium mechanics [106], more details about the research progress on the theoretical modeling of MR elastomer can be found in the recent review article [107]. Although the numerical results can be obtained after complex mathematical derivation, these theoretical models can deeply reveal the complicated magneto-mechanism coupling mechanism of MR elastomer, which has guiding significance for the optimal design and the practical application of this smart material.

A lot of work has been done on the magnetorheological mechanism of MR fluid and MR elastomer, yet there are little reports on the magnetorheological mechanism of MR gel. On the one hand, MR gel (especially for the solid-like MR gel) has not attracted wide attentions as a new MR material, and the research on it is not enough. On the other hand, the investigation on the magnetorheological mechanism of MR gel is more difficult than those of MR fluid or MR elastomer due to its intrinsic complex MR characteristics. MR gel possesses both the characteristics of mobility of magnetic particles in MR fluid and the stability of oriented microstructure in MR elastomer. These two features are "contradictory" to some extent, but they indeed exist in solid-like MR gel at the same time. Although the magnetic particles are moveable in the carrier matrix of MR gel, the clustering phenomena of particles in MR gel cannot be interpreted by the theory used in MR fluid because the viscous resistance of polymer matrix is far larger than the resistance from the carrier fluid in MR fluid. It is mean that the Mason number of MR gel is far larger than that of MR fluid, the assumption in fluid is invalid in MR gel. In the meantime, the viscous resistance of polymer matrix to the particle is much less than the constraining force of the rubber matrix to the particle in MR elastomer. After applying a magnetic field, the "solidified" ferromagnetic fillers in MR elastomer can only move slightly from the original position, while the ferromagnetic fillers in MR gel can greatly move under a strong magnetic field and a large applied loading; if the direction of magnetic field is changed, the ferromagnetic fillers can even rearrange

to generate chain-like or column-like structure along the new direction of magnetic field. These interesting characteristics that reflect the complexity of MR gel, the rheological behavior of polymer matrix, the magnetic interaction between adjacent fillers, and the interfacial problem due to the relative movement of particle and matrix have to be considered when studying the magnetorheological mechanism of MR gel. In addition, the "huge" change of microstructure after the rearrangement of particles will also make the modeling of the magneto-mechanical coupling behavior of MR gel more difficult.

It is not easy to fully describe the complicated magneto-mechanical coupling behaviors of MR gels. A field theory was developed by Han et al. to describe the magneto-sensitive viscoelasticity of ferrogel based on the principles of non-equilibrium thermodynamics [108]. The responses of ferrogel to different magnetic fields were analyzed by numerical calculation, and the theoretical results consistent with the experimental results under the cyclic magnetic field, which indicates that this theory is reasonable to some extent for the realization to magneto-mechanical coupling mechanism of MR gel. Zubarev evaluated the free energy of ferrogel after tension or compression along with the magnetic field direction by standard methods of statistical physics [109]. The analysis demonstrates that the magnetic field strength, the initial shape of the sample, as well as the particle concentration and the magnetic properties of particles determine the type of magneto-induced deformation (i.e., extension or shrunken). A particle-level molecular dynamics model was employed by Liu et al. to investigate the particle evolution in MR gel under a stable uniform magnetic field [110]. A modified magnetic dipole model is introduced to describe the magnetic interaction between adjacent particles, and this model presents higher precision than classic magnetic dipole model when processing the magnetic interaction of particles close to each other. The rheological behavior of the carrier matrix is described by the Bingham plastic model (Eq. (1)). Some complicated loading methods of magnetic field (such as the rotating magnetic field, as shown in **Figure 13**), which are difficult to be achieved by experiment, can be easily applied through simulation. With this, the 3D evolution of particular microstructure under complicated magnetic field loading conditions can be obtained, which is very important to understand the microstructure evolution mechanism of MR gel.

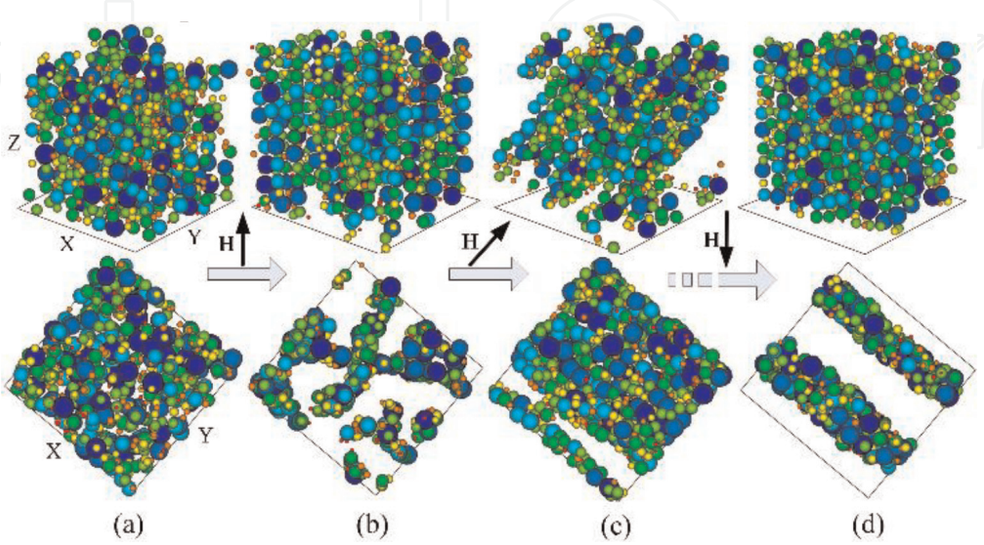

Figure 13.
The evolution process of 3D particular microstructure in MR gel: the initial state before applying magnetic field (a); the magnetic field is parallel with Z axis (b); the magnetic field rotates 45° in the clockwise direction (c); and the magnetic field further rotates 180° in the clockwise direction (d) [110].

The microstructure obtained by the particle-level molecular dynamics model matches well with the results by experimental observation under the same loading condition. However, the coupling effect between ferromagnetic fillers and carrier matrix is not considered in this model, and the Bingham plastic model is also too simple to describe the complicated rheological behavior of the carrier matrix; so, the governing equations which describe the physical behaviors of different components within the MR gel need to be further developed.

4. Conclusions and prospects

As a smart material whose physical properties can be easily controlled, magneto-sensitive composite (i.e., MR material) attracts more and more attentions in recent years. Many works concentrating on the magnetorheological mechanism and application have been reported. Various MR materials aiming at different practical applications were developed, which shows great application potential.

However, the inherent defects existed in conventional MR materials (e.g., the particle sedimentation in MR fluid, the microstructure control of MR elastomer, the bearing capacity of MR gel as structure unit) preclude their wide application. To this end, from the perspective of material preparation, on the one hand, we need to further improve the conventional MR material aiming at the inherent defects; on the other hand, the novel magneto-sensitive material system which meets the requirements of engineering applications should be developed, which means that we could develop multifunctional smart composite that has the magneto-controllable feature (e.g., magneto-sensitive impact-resistant composite, magneto-sensitive conductive composite, magneto-sensitive heat-conducting composite, and so on). From the perspective of mechanism, magneto-sensitive soft material refers to magneto-electro-thermo-mechanical coupling behavior, and it is difficult to describe the response to external stimuli. The difficulties can be summarized as follows: the description of the exact distribution state of dispersed phase before and after exposed under a magnetic field; the description of the discrepancy of size and shape of the dispersed ferromagnetic fillers; the interaction model between the dispersed phase and the continuous phase; the construction of constitutive model of the polymer matrix in the MR elastomer or solid-like MR gel; and the unification of multiscale model from microscale to macroscale. Considering the complexity of true situation, some necessary simplifications have to be made aiming at specific problem; then, the simplified model which could generally reflects the specific mechanism can be developed after ignoring the secondary factors. The numerical simulation is another effective method to investigate the microstructure evolution mechanism of MR material. It is an important research direction to construct the constitutive model of MR material, which could accurately describe the complicated coupling responses to different stimuli (magnetic field, temperature, strain rate, and so on). It is believed that the engineering applications of magneto-sensitive multifunctional material will be more widely concerned with the further realization on the magnetorheological mechanism and the enhancement of the performance of the material.

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