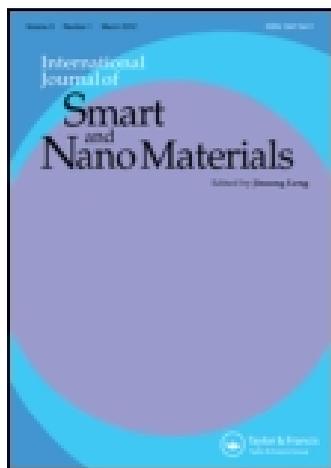


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Recent progress on the magnetorheological plastomers

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Recent progress on the magnetorheological plastomers

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Different from the traditional magnetorheological (MR) fluids and elastomers, the magnetic particles in the plastic MR materials are not ‘deadly’ trapped in the polymer matrix; thus, the MR plastomers exhibit higher MR effects and lower sedimentation. The plastic MR materials have attracted increasing attention, and the relevant fundamental mechanisms and practical applications have been intensively studied due to their unique physical and mechanical properties. In this highlight, we have mainly reviewed the preparation and the rheological properties of the MR plastomers. The formation mechanism of the MR plastomers has also been briefly summarized.

Keywords: magnetorheological; plastomers; MR effects

1. Introduction

As an engineering-applicable smart material, the magnetorheological (MR) material is usually prepared by dispersing magnetic particles into a carrying matrix with high viscosity. Applying under an external magnetic field, the dispersing particles are magnetized and the carrying matrix is strengthened by the as-induced magnetic dipole–dipole interactions (MR effects). These functionalized smart materials are expected to possess both high stability and MR effect to meet the requirements of practical applications. In consideration of the mismatch between the heavy magnetic particles and carrying matrix, an optimum host medium is desirable for developing the high-performance MR materials. The MR fluids (MRFs) have been developed as the first kind of MR materials [1–12] and these have been applied in a wide range of fields such as vibration absorbers, mass dampers, brake, sensors, actuators, base isolator, and mount [13–16]. Unfortunately, the sedimentation of the MRFs is believed to be the most awful point, which limits their practical applications. In the past decades, MR elastomers (MREs) have been developed to defeat the worse stability of the MRFs [17–30]. The entrapment of the magnetic particles within elastic polymer matrixes gives rise to stable mechanical properties of the MREs. Much effort has been made to widen the applications of the MREs [31–35], and several structural models [36–42] have been proposed to investigate the MR mechanism. However, the MREs still face many challenges in the practical applications due to the weak MR effects induced by the hard nature of the polymer matrix.

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Recently, a novel plastic MR material consisted of magnetic particles and soft polymer matrixes has been developed [43–47]. For this type of material, the magnetic particles are highly mobile due to the low cross-linking density of the carrying matrix and the aggregated structures can thus be changed by applying the external magnetic field. Therefore, it exhibits much higher MR effects than the MREs. Similar to the plasticine, this kind of MR material named as MR plastomer (MRP) has the plastic characteristic that enables it to be molded into various shapes [48]. The carrying matrix is critical for the MRP. In the case of highly cross-linking matrix, the magnetic particles were “deadly” anchored in the elastomer (MRE); thus, the high starting shear modulus and low magneto-induced modulus lead to the weak MR effects [49]. If the magnetic particles are highly mobile within the cross-linking polymer matrix, the obtained elastic MR material would exhibit strong MR effects [50]. The MRP is resulted with the further decrease in cross-linking density of polymer matrix. In this case, the magnetic particles could form chain-like structures under the magnetic field, resulting in a very high storage modulus of the material. In comparison with the traditional MR materials, the MRPs are multifunctional. The thermosensitive MRPs can be stored in solid state while working as MRFs under the fluid model [51]. These novel features of the MRPs will find promising applications in vibration dissipation, damping, artificial muscles, drug delivery, valves, and actuators [52–55].

In this highlight, after a brief introduction of the origination of the plastic MR materials and a short summary of the synthetic methods of MRPs, we will demonstrate how the mechanical properties of MRPs are dependent on the detailed structure of polymer matrix and how the strong MR effects are generated by the strong chain-like structures. Finally, we will give discussion on the mechanism of the magnetic field-induced improvement of mechanical properties and the potential application of MRPs in various fields.

2. Magnetorheological plastomers

Since Shiga et al. reported the silicone gels containing dispersed magnetic particles in 1995 [56], the plastic MR materials started to emerge in the scope of relevant research areas. According to the characteristic of the carrying matrix, the plastic MR materials can be classified into three catalogues, i.e., hydrogels, swollen polymer gels, and pure polymer gels (MRP). The magnetic hydrogels have always been prepared by using water-swollen polymer gels as the dispersing phase. Due to the high viscosity of the polymer matrix, the MR hydrogels exhibit higher stability than the MRFs. Similarly, if the polymers are swollen by high boiling point solvent, the as-obtained MR gels (MRGs) are belonged to the swollen polymer gels. Moreover, there are also many kinds of low cross-linking polymers which possess fluidic characteristics. After incorporating the magnetic particles into the above polymer matrixes, the plastic products are obtained and they are defined as the pure polymer gel-based MR materials (MRP).

At the early stage, various water-swollen polymer gels such as the carrageenan [45], gelatin [57], and guar gum [58] are introduced to the MRF to produce the jellylike MR materials. By use of the gel network as support, the settlement problem of the MRFs has been solved. However, a sharp decrease in MR effect is usually observed in these materials because the network can be destroyed by reversibly applying the magnetic field [59]. Although this giant negative MR effect can be conquered by optimizing the experimental parameters, the unexpected degradation of the carbonyl iron (CI) particles is emerged in water [50]. Recently, a kind of novel hydrogel cross-linked by magnetic

particles has been reported [57,60,61]. These magnetic particles are permanently linked to the polymer network; thus, the stability of the hydrogels is substantially improved. The magnetic particles can assemble along the supermolecular hydrogelators to form hybrid nanofibers [57]. Interestingly, as soon as the magnetic field is applied, the viscosity of the magnetic gel is sharply decreased. This work provides a new method to control the mechanical properties of soft materials.

Solvent-swollen polymer matrix has also been considered as an important candidate to improve the MR performances [62–67]. By adjusting the ratio of high boiling point diluents to reactants in the mixing polyurethane-silicone polymer gels, the MR materials can be altered from a liquid state to a solid state [62]. The silicone-based MRGs have a lower viscosity and a higher yield shear stress while polyurethane (PU)-based MRGs exhibit a better sedimentation stability [63]. Moreover, the high mechanical performances are also successfully achieved in the solvent-swollen cross-linked copolyimide (CCPI) [64] and superamolecular polymer systems [65]. Recently, An et al. have reported a new type of physically cross-linked MRGs in which the triblock copolymer polystyrene-block-poly(ethylene-stat-butadiene)-block-polystyrene (SEBS) swollen in paraffin oil was employed as the carrying matrix [46,68–70]. Due to the highly assembled magnetic particles, the as-formed MRGs exhibit a relative MR effect as high as 6000% [46]. Similarly, the MRG has also been prepared by introducing high boiling point solvents to the high cross-linked PU matrix. The dynamic and static yield shear stress is controllable by adjusting the swollen ratio of the PU matrix [66]. Very recently, it has been found that when the swollen solvent exceeds to a critical point (~25 wt%), the final MR gel becomes unstable and the sedimentation appears under a relatively long time (3 months) [67]. Therefore, the swollen ratio of the PU matrix should be controlled within a safe range.

The polymer gels with a low cross-linking density often exhibit a fluidic characteristic. By using such kind of polymers as the carrying matrix, the MR elastomers can be prepared. The first high-performance MRP was prepared by using the low cross-linking PU as the matrix, where the PU matrix was synthesized using toluene diisocyanate (TDI) and poly(propylene glycol) (PPG-1000) as the starting materials (Figure 1(a) and (b)) [48]. The mechanical properties of the PU matrix can be adjusted by varying the molar ratio of hard segment (TDI) to soft segment (PPG-1000); thus, both MRPs and MREs are available in this system. Applying under a magnetic field to the isotropic MRPs (Figure 1(c)), the magnetic forces have driven the CI particles defeat the polymer obstacle to form chain structures, thereby producing the anisotropic MRPs (Figure 1(d)). The mechanical properties of the MR materials are crucially dependent on the inner CI particles assembled structures. The *in situ* observation of the structure evolution opens up a possibility to study the origination of the MR effects. Different from the other two kinds of plastic MR materials, the MRPs have several unique characteristics. First, the matrix of solid-like magnetic gels is not elastic but plastic (Figure 1(a)). Second, the polymer barriers can be easily overcome by the magnetic field-induced torques due to the low cross-linking density of the matrix. Furthermore, a strong MR effect can be achieved since the strong CI chains can be readily formed in the MRP. The last point, the low molecular weight polymer chains can be tangled together and are easily untangled under small disturbance; thus, the self-healing phenomenon is usually observed with the help of magnetic field-induced dipole–dipole interactions [71].

Later, we reported the magnetic plasticine, in which the CI particles were used to substitute the traditional calcium salts dispersed within the paraffin wax/petroleum jelly mixing matrix [51]. Because of the low molecular weight of the paraffin wax and gel-like

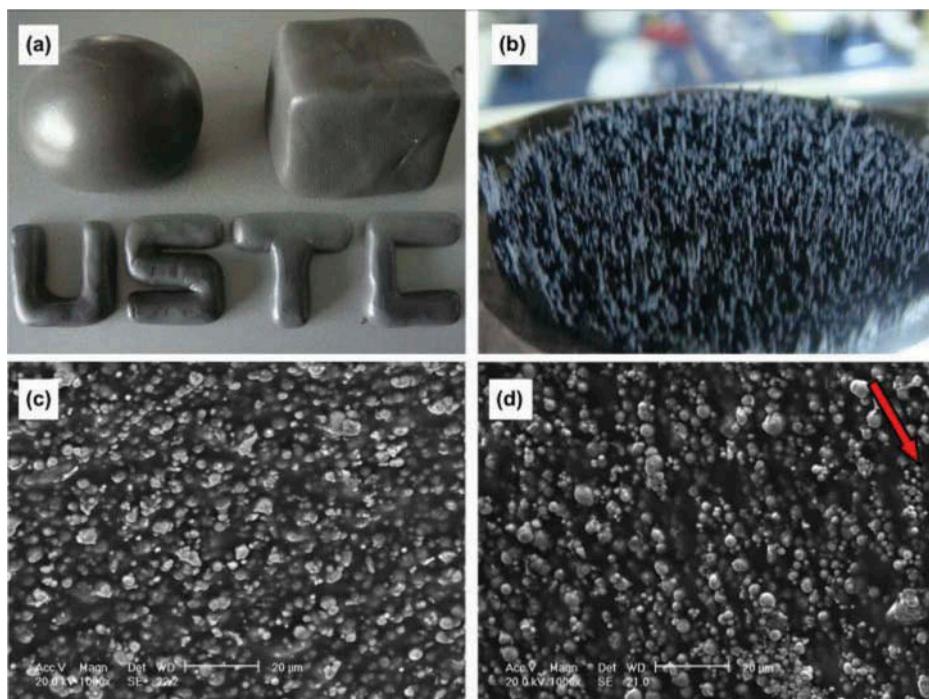


Figure 1. (a) The photograph of MRP with various shapes and (b) the image of the MRP under the magnetic field. The SEM images of (c) isotropic MRP and (d) anisotropic MRP [48]. The scale bar on the SEM images is 20 μm and the red arrow presents the direction of the applied magnetic field.

structure of the petroleum jelly, the as-prepared product possesses all the advantages of plasticine such as the easily shape-molding and good self-healing ability. Similar to the above MRP, the magnetic plasticine exhibits a high MR effect of 430% when the CI content is only 60 wt%. Since the paraffin wax is a phase-changing matrix, this product can be transformed into the MRF when the environmental temperature is higher than 50° C. This feature makes them to be applied reversibly between MRP and MRF by controlling the working temperature. A similar phase-changing MR material based on the thermoresponsive triblock copolymer (Pluronic F127) has also reported [72]. Due to the thermally driven ‘liquid to solid’ transition nature of the carrying matrix, such product behaves like a high-performance MRF when the suspending medium is in the ‘liquid’ phase and then performs as the MRE when the matrix is in the ‘solid’ phase. Clearly, this special kind of thermal sensitive characteristic not only reduces the sentimental defects of the traditional MRFs but also supplies a feasible way for multifunctional MR materials. Actually, the previously reported grease-based MR materials also exhibits such a thermal responsive MR behavior [73,74]. Although the physical state of the MR grease has not been demonstrated in the manuscript, it was estimated to be a kind of MRP with a small starting storage modulus, because the authors have claimed that this product had a strong solid-like structure rather than a fluid-like structure under external magnetic field.

Multifunctional MRP is also achievable by varying the carrying matrix. Shear hardening is a very interesting phenomenon in polymer composite. The storage modulus of the shear hardening polymer increases sharply by increasing the applied shear stress or impact. Introducing the CI particles to the shear hardening polymer makes the novel

MRP possess both high MR effects and excellent shear hardening property [75]. Interestingly, the shock absorbance property of the functional MRP can be tuned by varying the external magnetic field owing to the MR effects. The analysis indicates that the ‘cross bonds’ between the polymer chains and particle–particle forces are attributed to the multifunctional stimulus-responsive properties.

Rheological tests indicate the MRP is different from the MRE or MRF but inherits a part of their advantages. Applying under the magnetic field, the mechanical properties of the polymer matrixes enhanced due to the magnetic dipole–dipole forces. Because the magnetic particles are mobile, they continuously aggregate to form the particle chains until an equilibrium state is reached. The relatively strong chain structure and soft polymer matrix cause the MRPs to have a higher MR effect than the MREs. In comparison with the anisotropic one, the isotropic MRP has a smaller starting storage modulus induced by the randomly dispersed magnetic particles. Different from the MRGs and MRF, the Bingham viscoplastic model is not suitable to describe the mechanical behaviors of the MRPs. Owing to their soft solid-like characteristic, oscillatory shear rheometry has been proven to be a powerful tool to study their mechanical properties [60,65,66,74–77]. In the linear viscoelastic range, the highest magneto-induced storage modulus of the PU-based MRP reaches as high as 6.54 MPa, corresponding to a relative MR effect of 532% (Figure 2(a)). Under applying a constant magnetic field, a noticeable time-dependent increment of the storage modulus is observed for the isotropic MRPs, indicating the

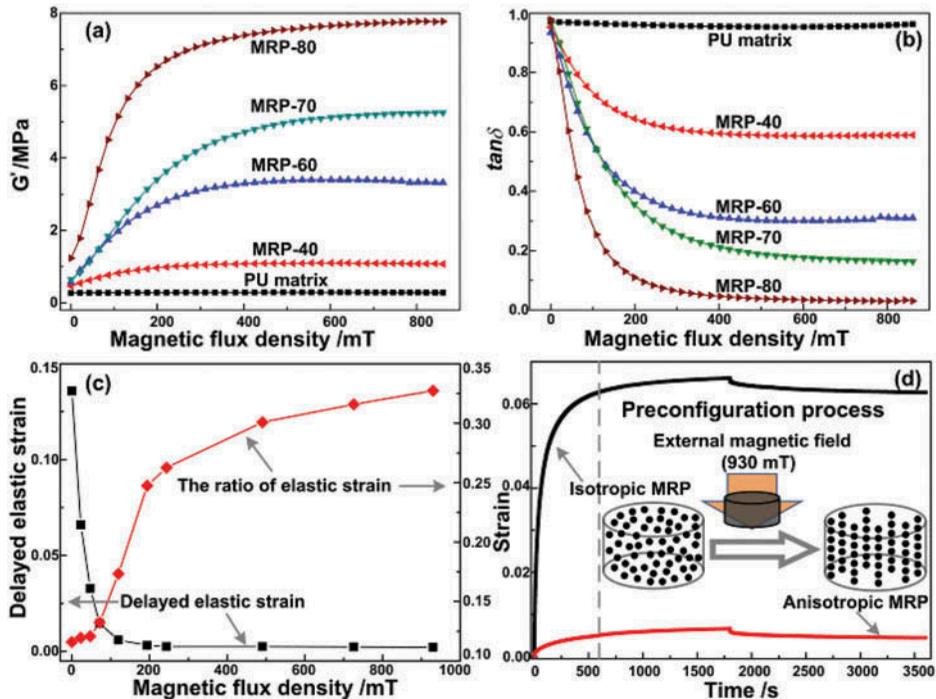


Figure 2. (a) The storage modulus and (b) loss factor of anisotropic MRP under different magnetic fields (The MRP-x denotes the weight percentage of the CI particles) [48]; (c) delayed elastic strain and the ratio of elastic strain to the total strain of the anisotropic MRP under different magnetic fields and (d) the creep and recovery curves of isotropic and anisotropic MRP under a 930 mT magnetic field [81]. (The creep time is 1800 s and the constant stress is set as 2000 Pa.)

structural reorganization processes in these plastic materials [71]. With increase in the magnetic field, the movement of the soft polymer segment decreases as the restriction effect of the CI chains is enhanced, in turn leading to a decrease in the damping factors (Figure 2(b)). As the interfacial slipping between the CI particles and the matrix in the isotropic MRP is larger than the anisotropic MRP, the former exhibits a higher damping factor [48]. Additionally, an obvious magnetostriction effect is observed by monitoring the normal force of MRPs along with the direction of the magnetic field. Therefore, the particle arrangement process and magnetic field strength also generate a great effect on the normal force of MRPs [78,79]. Moreover, the influence of stepwise magnetic field on the storage modulus of MRPs has also been systematically studied. These results have demonstrated that the MR effect and magnetic response velocity are highly dependent on the aggregation structure of the magnetic particles [45,50,67,80].

The MRP exhibits unique nonlinear dynamic properties by loading harmonic strain. The critical points between the linear and nonlinear areas are both found in strain amplitude and frequency dependency testing. Interestingly, although the magnetic field strongly affects the dynamic properties of the MRP, it almost has no influence on the strain- and frequency-induced nonlinearity [71]. By increasing the magnetic field from 0 to 930 mT, the ratio of delayed elastic strain to the total creep strain increases from 11.6% to 32.7% (Figure 2(c)) because of the increment of the restriction effects to the slipping of soft chain segments. Clearly, the MRP transforms from a viscoplastic to a elastoplastic state with increasing magnetic field. In addition, only a small decrease in the modulus is observed in the PU-based MRP with increasing temperature, which is attributed to the low-temperature sensitivity of the PU matrix. The creep strain decreases with increasing temperature under applying a 930 mT magnetic field, while they exhibit a reversible characteristic in the absence of the magnetic field. Although this difference is originated from the magnetic interactions among the CI particles and the coupling effects between the CI particular structures and the polymer matrix, the detailed mechanism of this interesting aspect is still unknown and it is deserved for further investigations. The particle distribution also has a significant influence on the creep behavior of MRP (Figure 2(d)). It is found that the creep strain of isotropic MRP increases more sharply than that of anisotropic MRP at the initial 10 minutes, which reflects that the aggregation process of magnetic particle starts from random dispersion to structured orientation [81].

Besides the normal forces, the squeeze flow behavior of the MRP has also been systematically investigated [82,83]. The elastic deformation region, stress relaxation region, and plastic flow region are presented in the compression and tension testing. It is reported that compressive yield stress and the tensile yield stress are sensitive to the magnetic field, particle concentration, and particle distribution. Because the viscosity of polymer matrix is higher than the silicon oil, the yield stress of the MRP is larger than the relative MRF under the squeeze mode [83]. Moreover, the MRP is a typical soft magnetosensitive polymer composite, so it has a large magneto-deforming effect. The quasi-static shearing testing measurements indicate that the MRP exhibits a high magneto-damping performance, which can also be described by the magneto-enhanced Bingham fluid-like model [84].

The MRP also possesses the typical structural dependent conductivity [85]. As the CI particles are conductive, the electric resistance of the MRP sharply changes once the CI particular structure bridges the two test points. Applying under the magnetic field, both the thickness of the interface layer of electrode-MRP and the gaps between the adjacent particles decrease simultaneously. To this end, the conductivity of the MRP increases since the electron transfer in the particulate structures is improved (Figure 3). Clearly, this

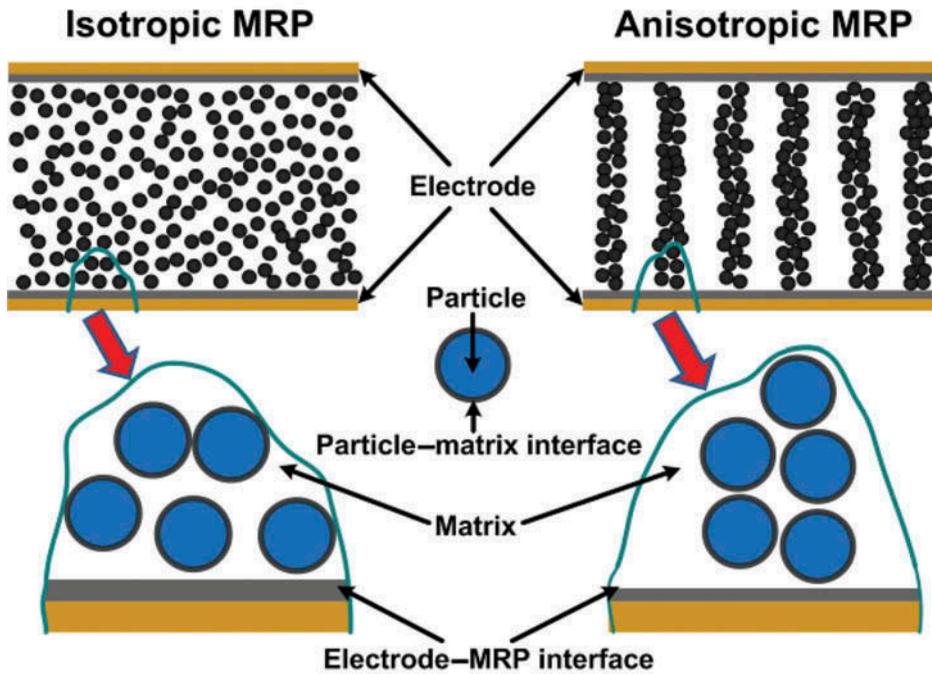


Figure 3. Schematic illustration of the particulate structure of MRP with different particle distribution [85].

work provides a simple method to quantitatively characterize the anisotropy of the MRP, which will help us to understand how the magnetic field affects the interactions between the neighboring CI particles. However, the magnetically dependent conductivity of the MRP has not been carefully explored and more work need to be done to fully understand the exact MR mechanism. Since the MRPs are free-standing and their inner structure is sensitive to the external stimulus, they are expected to be applicable in a number of fields including soft actuator, artificial muscle, and sensors [53–55].

3. Mechanism and simulation

Many interesting/novel properties of the smart MRPs have been reported in recent experimental works. However, the underlying mechanisms of the properties are still mysterious, even though some qualitative mechanisms have already been proposed. Stokesian dynamics theory [86] and computer simulations [87–92] are effective for studying the microstructural formation and evolution processes of MRFs. Therefore, these theories may be useful for describing the unique MR mechanisms of the MRPs due to the similarity of particle movement between the MRPs and MRFs.

With the rapid development of computer science, a large amount of computer-simulation works have been conducted to reveal the magneto-controllable properties of plastic MR materials. Various analytic methods such as the continuous medium theory [93–95], field theory [96], microscopic model [97,98], and Monte Carlo simulations [99] have been developed to describe the magneto-viscoelastic property of ferrogels. Inspired by the aforementioned works, the first simulation work for MRP has already been performed

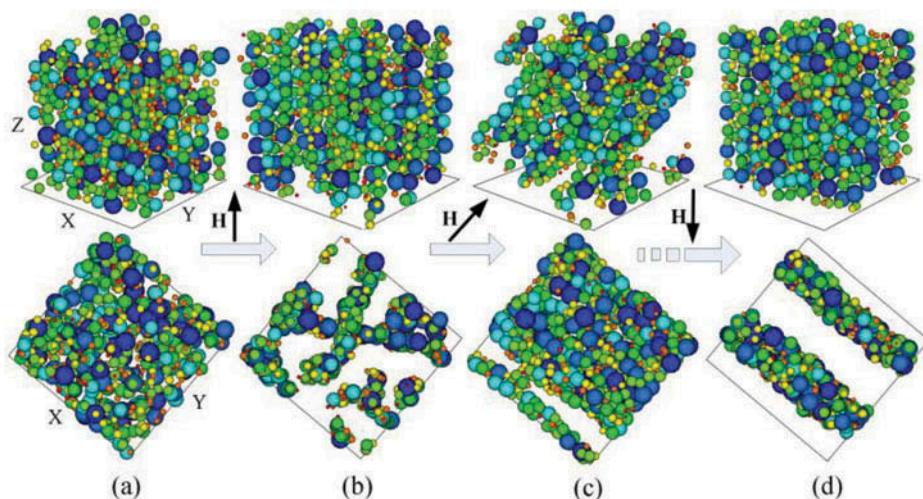


Figure 4. The magneto-induced particulate microstructure of (a) isotropic MRP; (b) anisotropic MRP obtained by applying the magnetic field on the isotropic MRP; (c) and (d) anisotropic MRPs obtained by further rotating the magnetic field. (The upper series shows the axonometric views and the lower series shows the top views. The solid arrows denote the directions of external magnetic field. The particles with the same color indicate the same size) [100].

to demonstrate the magneto-induced rearrangeable characteristics [100]. The size distribution of the CI particles was characterized by a lognormal formula, and a modified dipolar force model was preferably introduced to describe the magnetic interaction of the nearby CI particles. The matrix was treated as a high-viscosity Bingham fluid with a certain yield stress. As an example of the evolution of magneto-induced particulate microstructure in MRPs, Figure 4 illustrates that the initial randomly distributed microstructure (Figure 4(a)) transfers to chain-like/column-like microstructure (Figure 4(b)) after applying an external magnetic field. With the evolution of microstructure, the macroscopic physical or mechanical properties of MRPs vary correspondingly. The chain-like/column-like microstructures can be well remained after removing the external magnetic field, and new microstructures can be formed via the rearrangement of themselves by changing the direction of the magnetic field. Figure 4(c–d) gives an illustration of simulated, magneto-induced rearrangeable microstructure of MRPs from a quasi-steady state to another quasi-steady state in a stepwise rotating magnetic field. Interestingly, the CI particles assemble to planar microstructure in the rotating plane of external magnetic field.

Both the particle–particle model and particle–matrix model are essential for building the entire simulation model. In most of the previous numeric-simulation works, the CI particles were treated as magnetic point dipoles. This treatment leads to some errors in the basic particle model because the CI particles are always aggregated to form large structures in the polymer matrix. Therefore, in the further simulations, proper modification should be conducted on the basic particle model. To this end, an alternative finite-dipole model [101] has been used to solve this problem. Specifically, the magnetization of a particle is represented as a distribution of current density and this method has been proven to be effective for analyzing the dynamic properties of the MR materials. Nevertheless, the current matrix model is mostly based on the model of Newtonian fluid or Bingham fluid, and the motion of magnetic particles in the matrix is treated as Stokesian drag.

These simple models are no longer suitable to deal with the computation of the dynamics properties of MRPs, so these should be improved in future work.

4. Conclusions and perspectives

Plastic MR material has been proven to be a kind of fascinating smart material which possesses both strong MR effects and high stability. The development of novel MRP systems and the optimization of their mechanical properties are considered to be crucial in relevant research areas. In this highlight, we have mainly reviewed the development of the plastic MR materials. The polymer matrix is believed to be the most important parameter for modulating the mechanical properties of the MRPs. Multifunctional MRPs with the combination of MR effects, temperature sensitivity, conductivity sensitivity, and shear thickening/thinning properties certainly deserve future research. The detailed mechanism of the CI aggregation structure transformation, the force transfer during applying the external stimulus, and the formation of the MR effects have remained unclear. Future analysis of the theory model and computer simulation on the structural dependent mechanical properties of the MRP materials will be of high interest.

Due to the unique magnetically dependent mechanical properties, the MRP may find many promising applications in a wealth of areas. Our ongoing project demonstrates that the MRP is effective for the high-performance MR absorber, which should be very useful for intelligent vibration dissipation. The multifunctional MRP and the PU sponge-reinforced MRP can be designed to fabricate the future-generation body armor [75,102]. Furthermore, the magnetic field-dependent conductivity of the MRP also enables them be applied in the field of smart devices such as sensors and on-off switches [103].

Disclosure statement

No potential conflict of interest was reported by the authors.

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