Proceedings of CONF-FMCE 2026 Symposium: Smart City and Infrastructure Engineering
DOI: 10.54254/2755-2721/2026.CH35293

A Review of Preparation Methods for Magnetorheological Fluids with High Dispersion Stability

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Abstract. Magnetorheological fluid is a kind of intelligent material that is widely used in fields such as damping and vibration reduction, transmission control, sealing and precision machining, and can rapidly undergo rheological transformation under an external magnetic field. However, the large density difference between magnetic particles and carrier fluid easily leads to problems such as particle sedimentation, agglomeration and poor redispersibility, thus limiting the application effect of magnetorheological fluids. This paper reviews the preparation methods of magnetorheological fluids with high dispersion stability, focusing on methods including additive modification, addition of magnetic nanoparticles, surface coating of carbonyl iron powder, introduction of two-dimensional fiber materials and optimization of base fluid. Studies have shown that methods such as using high-viscosity carrier fluid, adding nanomaterials and coating particles to form core-shell structures can effectively improve the dispersion stability, yield stress and redispersibility of magnetorheological fluids. The current development status of the preparation of magnetorheological fluids with high dispersion stability is summarized, and the future directions of multifunctional composite materials, green and environmentally friendly base fluids and intelligent preparation are prospected, so as to provide references for the research and application of high-performance magnetorheological fluids.

Keywords: magnetorheological fluid, dispersion stability, carbonyl iron powder, nanoparticles, additives, two-dimensional materials

1. Introduction

Magnetorheological Fluid (MRF) is an intelligent suspension system prepared by uniformly dispersing micron-sized soft magnetic particles in a carrier fluid with the assistance of an appropriate amount of additives. In the absence of an external magnetic field, MRF behaves as a Newtonian fluid with good fluidity, and magnetic particles are randomly distributed inside the carrier fluid; when subjected to an external magnetic field, magnetic particles will rapidly arrange along the direction of magnetic lines of force and form chain or columnar structures, which significantly increases the internal resistance of the system and sharply raises the viscosity, thus presenting rheological behavior similar to that of a Bingham plastic body [1]. The most remarkable characteristic of MRF is that its rheological properties can be regulated in real time and reversibly

DOI: 10.54254/2755-2721/2026.CH35293

by an external magnetic field. During the transition between liquid and solid-like states, it has the advantages of low energy consumption, fast response speed, continuously adjustable apparent viscosity and high control accuracy, so it has attracted extensive attention. In 1948, American scholar Rabinow first developed a magnetorheological fluid applied to power transmission devices and applied for relevant patents, marking the beginning of research on magnetorheological technology. Since then, researchers at home and abroad have carried out a large number of studies on its composition design, rheological mechanism and engineering applications, promoting the gradual application of MRF in many fields such as vibration damping and isolation systems, transmission and braking devices, sealing technology, precision polishing processing and biomedical equipment [2]. The properties of magnetorheological fluids mainly include dispersion stability, rheological properties, tribological properties, viscosity-temperature characteristics and so on. Among them, the quality of each property will have an important impact on the working efficiency and service life of magnetorheological devices, while dispersion stability and rheological properties are the key factors determining their application effects. When the stability of magnetorheological fluid decreases, its rheological response ability will also be significantly affected, leading to problems such as slower device response speed, fluctuating output performance and shortened service life. Due to the density difference between magnetic particles and carrier fluid, particles are prone to sedimentation under gravity. At the same time, micron-sized magnetic particles have a large specific surface area, and their high surface energy will promote agglomeration between particles, further reducing the dispersion stability of the system [3]. Sedimentation and agglomeration are important factors restricting the long-term stable application of magnetorheological fluids.

To solve the above problems, researchers at home and abroad have proposed a variety of methods to improve the stability and rheological properties of magnetorheological fluids. For example, coating carbonyl iron particles with low-density materials to reduce the density difference between particles and carrier fluid; optimizing particle structure by using non-spherical magnetic particles such as fibrous and flaky ones; compounding magnetic fluid with magnetorheological fluid; introducing functional additives to construct a stable system; using thixotropic network structure to inhibit particle sedimentation; and adopting viscoelastic media to improve suspension capacity. These methods have improved the sedimentation stability and magnetorheological effect of magnetorheological fluids to varying degrees.

In recent years, research hotspots have mainly focused on improving the dispersion stability of magnetorheological fluids by adding functional additives, introducing magnetic nanoparticles, conducting surface coating modification on carbonyl iron powder, adding two-dimensional (2D) materials and optimizing the types of carrier fluids. These strategies can not only effectively inhibit particle sedimentation and agglomeration, but also enhance the field response ability of magnetorheological fluids, providing new research ideas for the development of high-performance magnetorheological materials.

2. Preparation methods of Magnetorheological Fluids

2.1. Improving dispersion stability of Magnetorheological Fluids by additives

Additives are one of the earliest and most widely used methods to improve the dispersion stability of magnetorheological fluids. Their main function is to enhance electrostatic repulsion or steric hindrance between particles, thereby inhibiting sedimentation and agglomeration of magnetic particles. Common additives include polymeric dispersants, surfactants, graphite fibers, silicon carbon fibers, carbon nanotubes, organoclays, silica nanoparticles, etc.

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Domestic and foreign scholars have carried out extensive research on the influence of different types of additives on the properties of magnetorheological fluids. Park et al. [4] investigated the effects of magnetic nanoparticles (chromium dioxide), vacuum grease and viscoelastic media on the sedimentation stability of magnetorheological fluids. The results showed that all the above additives could improve the anti-sedimentation ability of the system to a certain extent. Studies have also found that fibrous particles can provide greater structural support compared with traditional spherical particles, thus effectively enhancing the stability of suspension systems. For example, materials such as organoclays, single-walled carbon nanotubes, mesoporous MCM-41 and nano-graphite fibers all exhibit favorable anti-sedimentation effects [5]. Cho et al. [6] prepared magnetorheological fluids using fumed silica as a functional additive. It was found that silica particles could fill the voids of the body-centered tetragonal structure formed by magnetic particles, further strengthening the chain-like and column-like structures formed under the magnetic field, thereby improving both the rheological properties and dispersion stability of magnetorheological fluids. Premalatha et al. [7] studied the effect of grease dosage on the sedimentation characteristics of magnetorheological fluids. The results indicated that an appropriate amount of grease could effectively improve stability, but the improvement effect tended to saturate when the grease concentration increased to a certain level. Rankin et al. [8] prepared magnetorheological fluids with two different types of grease. Experimental results showed that the addition of grease significantly improved the dispersion stability of the system with basically no negative impact on the magnetorheological effect.

It can be concluded from existing research that magnetic nanoparticles, vacuum grease, viscoelastic media and fibrous nanomaterials can effectively enhance the anti-sedimentation ability and dispersion stability of magnetorheological fluids. Among them, graphite nanofibers and fumed silica can not only strengthen system stability but also improve the magnetorheological effect. An appropriate amount of grease can improve particle suspension on the premise of maintaining basically unchanged magnetorheological properties, but excessive grease concentration has a limited effect on performance improvement. In addition, single-walled carbon nanotubes can promote the formation and repair of particle chain structures under weak magnetic fields, improving the shear yield stress and mechanical properties of the system, which provides a new research direction for the design of high-stability magnetorheological fluids.

2.2. Improving dispersion stability of Magnetorheological Fluids by adding magnetic nanoparticles

Iron oxide nanomaterials have become common functional additives in magnetorheological fluids due to their excellent magnetic response and favorable chemical stability. Among them, iron oxides such as magnetite (Fe_3O_4), hematite (α-Fe_2O_3) and maghemite (γ-Fe_2O_3) are widely used in magnetorheological fluid systems to improve their dispersion stability and magnetorheological properties. In addition, magnetite-based composite nanoparticles prepared by polymerization have also attracted extensive attention from researchers. Such nanoparticles are characterized by low density, fast magnetization speed and large specific surface area, which can effectively enhance the interaction between particles and system stability [9-11]. In recent years, magnetorheological fluids based on ferrofluids have become one of the research hotspots. Compared with traditional magnetorheological fluids, magnetorheological fluids prepared with ferrofluids as the continuous phase possess higher yield stress, which can reach several times that of traditional systems [12]. This is because the Brownian motion network formed by magnetic nanoparticles can effectively coat and support micron-sized magnetic particles, thus inhibiting sedimentation and improving suspension

DOI: 10.54254/2755-2721/2026.CH35293

stability. Therefore, the synergistic dispersion of micron-sized magnetic particles and nano-sized magnetic particles in the system is regarded as an important development direction to improve the comprehensive properties of magnetorheological fluids [13]. Regarding the reinforcing effect of magnetic nanoparticles, Song et al. [14] prepared magnetorheological fluids with carbonyl iron (CI) nanoparticles as additives. It was found that when the addition amount was 1 wt%, both the dispersion stability and yield stress of the system were significantly improved. Chin et al. [15] introduced carbonyl iron microparticles, magnetite nanoparticles (Co-γ-Fe_2O_3 and CrO_2) and surfactants into magnetorheological fluids together. The results showed that the anti-sedimentation performance and magnetorheological effect of the magnetorheological fluids were effectively improved. Oka et al. [16] further studied the influence of volume fractions of magnetic nanoparticles and carbonyl iron microparticles on the properties of magnetorheological fluids. The results indicated that with the increase in the concentration of the two types of particles, both the yield stress and magnetorheological effect of the system were significantly enhanced. Wereley et al. [17] found that there was an optimal range for the addition amount of magnetic nanoparticles. When the volume fraction was 15%–20%, the stability of magnetorheological fluids reached a favorable level; however, when the concentration exceeded 20%, the yield stress showed a downward trend instead. Iglesias et al. [18] prepared magnetorheological fluids by adding 32% iron microparticles and 3% magnetite nanoparticles into ferrofluids without extra stabilizers. The results showed that nanoparticles could effectively inhibit the formation of sediment layers and enhance the magnetorheological effect. Nevertheless, when the nanoparticle content exceeded 7 vol%, a dense coating layer would form on the surface of iron microparticles, weakening the magnetic dipole interaction between particles and thus reducing the magnetorheological effect.

In summary, magnetite-based iron oxides and their composite nanoparticles are a class of high-performance additives for magnetorheological fluids. Nanoparticles can fill the gaps between magnetic particles, strengthen magnetic interactions between particles, and improve the yield stress and dispersion stability of the system. Meanwhile, constructing a micro/nano composite system with ferrofluids as the continuous phase can significantly improve the anti-sedimentation performance and rheological properties of magnetorheological fluids. However, the addition amount of magnetic nanoparticles is not the higher the better. Excessive nanoparticles tend to form a coating layer on the surface of microparticles and weaken the formation of magnetic chain structures. Therefore, the ratio of microparticles to nanoparticles needs to be optimized to achieve the best comprehensive performance.

2.3. Improving dispersion stability of Magnetorheological Fluids by coating carbonyl iron powder

Surface coating modification of carbonyl iron (CI) particles to construct core–shell structured composite particles is regarded as one of the important strategies for improving the dispersion stability of magnetorheological fluids. This method reduces the overall density difference of particles by introducing a low-density functional shell layer on the surface of magnetic particles, thereby slowing down the sedimentation rate of particles and improving the long-term stability of the system [19]. In recent years, polymer coating technology has been extensively studied in the field of magnetorheological fluids. Cho et al. [20] coated the surface of carbonyl iron particles with polymethyl methacrylate (PMMA) to prepare composite particles with a core–shell structure. The results showed that the PMMA coating layer could effectively reduce particle density and thus improve the dispersion stability of magnetorheological fluids. Compared with bare CI particles, the coated particles still maintained an approximately spherical structure but with a smoother surface, and the coating thickness was about 2–10 μm. However, due to the existence of the non-magnetic polymer layer, the saturation magnetization of the particles decreased slightly, leading to a slight reduction in the yield stress of the magnetorheological fluid. Mrlik et al. [21] modified the surface of carbonyl iron particles with cholesteryl chloroformate. The results indicated that the coated particles had better interfacial compatibility with the carrier fluid, which could effectively improve the dispersion stability of magnetorheological fluids. At the same time, the modified particles exhibited excellent thermal stability and redispersibility. At particle concentrations of 20% and 60%, the magnetorheological fluids prepared with coated particles even showed higher yield stress than the uncoated systems. Chuah et al. [22] prepared CI/PS composite particles and dispersed them in the carrier fluid, which also achieved the effects of reducing particle density and improving system stability. In addition, constructing polymer-coated core–shell structures via solvent evaporation has also become a common technique. Researchers have successively developed various composite particle systems such as CI/PVB, CI/polycarbonate (PC), and CI/polystyrene (PS), and optimized their properties by adjusting the coating thickness [22]. Liu et al. [11] formed a SiO_2 coating layer on the surface of carbonyl iron particles using the sol–gel method. The results showed that SiO_2–CI composite particles not only possessed good corrosion resistance but also could increase the viscosity of the carrier fluid, enhancing the shear stress and structural stability of the magnetorheological fluid. Compared with traditional polymer materials, multi-walled carbon nanotubes (MWCNTs) are considered as highly promising coating materials due to their excellent mechanical properties, high specific surface area, and favorable electrical and thermal conductivity. Under an external magnetic field, MWCNT-coated particles can effectively enhance the shear stress and yield stress of magnetorheological fluids while maintaining high magnetic response performance. Fang et al. [23] used 4-aminobenzoic acid (PABA) as a coupling agent to construct an MWCNT coating layer on the surface of carbonyl iron particles and prepared corresponding magnetorheological fluids. Experimental results showed that MWCNT-coated particles could significantly improve the dispersion stability and redispersibility of the system, mainly due to the reduced particle density after coating. It is worth noting that the saturation magnetization of the MWCNT-coated system was 209 $A \cdot m^2/kg$, which was slightly different from the 217 $A \cdot m^2/kg$ of bare CI particles, so its influence on the yield stress of the magnetorheological fluid was limited. Rwei et al. [24] modified the surface of magnetic particles with MWCNTs. It was found that the sedimentation stability and shear stress of the obtained magnetorheological fluid were superior to those of commercial magnetorheological fluid products, while its saturation magnetization (206 emu/g) was close to that of unmodified particles (217 emu/g). Guo et al. [25] immobilized MWCNTs on the surface of magnetic particles using grafting technology. The results showed that with the increase of grafting agent concentration, the dispersion stability of the magnetorheological fluid was further improved. This was mainly attributed to the stronger Brownian motion effect of nanoscale MWCNTs, which could counteract gravitational sedimentation to a certain extent and promote the formation of a stable network structure between particles. In summary, constructing a core–shell structure of carbonyl iron particles is an effective way to improve the dispersion stability of magnetorheological fluids. Polymer coating layers can reduce particle density, improve compatibility between particles and carrier fluid, and enhance thermal stability and redispersibility, thus improving the anti-sedimentation ability and thixotropic properties of the system; however, they may lead to a slight decrease in saturation magnetization and yield stress. In contrast, multi-walled carbon nanotube coating technology can significantly improve the dispersion stability, sedimentation stability, and yield shear stress of magnetorheological fluids while maintaining high magnetic response performance, showing broader application prospects. In the future, the properties of

DOI: 10.54254/2755-2721/2026.CH35293

coating layers can be further optimized through graft modification, composite coating, and multi-scale structural design to achieve the synergistic improvement of stability and rheological properties of magnetorheological fluids.

2.4. Improving dispersion stability of Magnetorheological Fluids by adding 2D fiber materials

In recent years, regulating the morphology of magnetic particles to improve the dispersion stability of magnetorheological fluids has gradually become a research hotspot. Among them, anisotropic materials such as linear particles, two-dimensional lamellar structures and flaky particles, with their unique geometric morphology and large specific surface area, can effectively enhance the structural support between particles, thereby improving the anti-sedimentation performance and magnetorheological effect of magnetorheological fluids [26]. Studies by Kuzhir et al. have shown that when nanorods are used as the dispersed phase to prepare magnetorheological fluids, both the yield stress and dispersion stability of the system can be improved [27]. Bell et al. [28] prepared magnetorheological fluids using iron microwires as the dispersed phase and sodium thiolate solution as the continuous phase, and found that the yield stress of the microwire system was lower than that of traditional spherical particle systems. However, since the contact area between linear particles is small, the irreversible agglomeration between particles is inhibited, thus improving the sedimentation stability. Further studies have found that the composite use of nanowires and spherical magnetic particles can not only improve the system stability, but also enhance the ability to form chain-like structures under the magnetic field, thereby improving the magnetorheological effect. Magnetorheological performance test results show that the magnetorheological effect of magnetorheological fluids using magnetic fibers as the dispersed phase can reach 2 to 3 times that of traditional spherical particle systems. This phenomenon is mainly attributed to the high aspect ratio and anisotropic structure of fibrous particles, which make them easier to form stable and high-strength chain-like structures under the action of a magnetic field. Chiriac et al. [29] further pointed out that particle size and surface oxidation degree also have a significant impact on the magnetorheological effect of magnetorheological fluids. In addition to one-dimensional structural materials, the application of two-dimensional nanomaterials in magnetorheological fluids has also attracted extensive attention. Two-dimensional materials usually possess excellent mechanical, electrical and thermal properties as well as large specific surface area, so they can effectively improve the dispersion stability and rheological properties of magnetorheological fluids [30-32]. Existing studies have shown that composite systems composed of different two-dimensional materials often exhibit more excellent comprehensive properties than single nanoparticle additives [33-35]. For example, reduced graphene oxide/molybdenum disulfide (rGO/MoS_2) composite magnetic particles prepared by hydrothermal method have been used in magnetorheological fluid systems. The results show that the yield stress and shear stress of the rGO/MoS_2 composite magnetorheological fluid are significantly improved. This is mainly attributed to the larger specific surface area provided by the two-dimensional composite structure, and the nanoscale magnetic particles can fill the gaps between microparticles, thereby enhancing the magnetic interaction between particles [36]. Wang et al. [37] prepared magnetorheological fluids using Fe_3O_4/MoS_2 two-dimensional nanocomposites, and the results show that the dispersion stability of the system is significantly improved. It is believed that the unique lamellar structure of MoS_2 nanosheets can form a spatial network similar to "umbrella-like support", effectively hindering the sedimentation of magnetic particles. Upadhyay et al. [38] compared the performance differences between magnetorheological fluids of flaky particles and spherical particles, and found that the sedimentation rate of the flaky particle system was nearly 50% lower than that of spherical particles. Lee et al. [39]

DOI: 10.54254/2755-2721/2026.CH35293

further studied the influence of carbonyl iron particles with different morphologies on the properties of magnetorheological fluids. The results show that magnetorheological fluids prepared with flaky carbonyl iron particles have more excellent dispersion stability. Although their saturation magnetization is slightly lower than that of spherical particle systems, the overall comprehensive performance is more advantageous.

In summary, the anisotropic particle structure is an important development direction for improving the stability of magnetorheological fluids. Linear particles can enhance the formation ability of magnetic chains and improve rheological properties through their high aspect ratio structure; two-dimensional nanosheets and flaky particles can significantly inhibit particle sedimentation relying on their large specific surface area and spatial support effect. Although the preparation processes of nanowires, nanosheets and flaky particles are relatively complex and costly, which limits their large-scale application to a certain extent, they show significant advantages in improving the dispersion stability of magnetorheological fluids, enhancing the magnetorheological effect and optimizing the microstructure, thus possessing broad research and application prospects.

2.5. Improving dispersion stability of Magnetorheological Fluids by optimizing the type and properties of base fluids

Many researchers have proposed using more viscous liquids, such as lubricating oils or gels, as additives or base fluids to improve the dispersion stability of magnetorheological fluids. Polyethylene oxide (PEO) is one of the most commonly used polymers; it is soluble in various solvents and has low toxicity, so it is often used to coat particles. Adding carbon nanotubes to polymer gels can enhance the stability of suspensions [40]. Kim et al. [41] prepared magnetorheological fluids by adding carbon nanotubes to polymer suspensions. The results showed that the rheological properties of PEO-based magnetorheological fluids, including viscosity and field-induced yield stress, were higher than those of conventional magnetorheological fluids, while the sedimentation rate was lower. Wang et al. [42] synthesized magnetorheological gels using PAO base oil and compared them with magnetorheological gels based on magnetic fluids. The results showed that the apparent viscosity of both PAO gels and magnetic fluid gels was higher than that of conventional gels, and the yield stress of magnetic fluid gels was greater than that of PAO-based magnetorheological gels. Kim et al. [43] studied the effect of small-sized soft magnetic CI particles on the properties of polyisobutylene (PIB)/polybutene (PB) fluids. The results showed that the stability and magnetostrictive effect of viscoelastic suspensions based on CI-PIB/PB were significantly improved. However, under zero-field conditions, the use of high-viscosity base fluids adversely affects many properties of magnetorheological fluids. Park et al. [44] prepared magnetorheological gels by dispersing CI particles in water-oil emulsions. The results showed that increasing water content improved the sedimentation stability and magnetostrictive effect of the gels. Mazlam et al. [45] synthesized magnetorheological fluids based on water and hydrocarbons. The results showed that the shear stress of the water-based magnetorheological fluid was higher than that of the hydrocarbon-based one.

Some researchers have used ionic liquids (ILs) as the continuous phase of magnetorheological fluids. Due to their low vapor pressure and extremely low flammability, they are also known as green solvents [46]. Because of their physical adsorption properties, ions on the surface of magnetic particles may increase the repulsion between magnetic particles and reduce particle agglomeration, thereby improving the sedimentation stability of magnetorheological fluids. Sancher et al. [47] prepared magnetorheological fluids using different ionic liquids as base fluids, with magnetite nanoparticles and microparticles as the dispersed phase. The results showed that dispersing 10% by

DOI: 10.54254/2755-2721/2026.CH35293

volume of magnetite microparticles in 1-butyl-3-methylimidazolium hexafluorophosphate improved the dispersion stability of the magnetorheological fluid. Bednarek et al. [48] prepared MRF by adding magnetic silicon steel particles and graphite microparticles to conductive cedar oil. The results showed that the use of graphite particles reduced the resistivity of the magnetorheological fluid by filling the gaps between steel particles and increasing the viscosity of cedar oil, leading to improved stability of the magnetorheological fluid after six weeks.

This indicates that the selection of appropriate surfactants and carrier fluids, as well as the control of interactions between magnetic particles and the carrier fluid, are key to preparing MRF with high dispersion stability.

3. Summary

(1) Fibrous materials, nano-oxides, grease, carbon nanotubes and other additives can synergistically improve the anti-sedimentation performance and magnetorheological effect of magnetorheological fluids. The dosage of additives needs to be reasonably controlled, which serves as a simple and effective method to enhance the stability of magnetorheological fluids.

(2) The introduction of magnetic nanoparticles such as iron oxides and carbonyl iron, or the adoption of ferrofluids as the continuous phase can fill the gaps between particles, strengthen the chain structure, and improve the yield stress and stability of magnetorheological fluids. However, excessive particle concentration will weaken the magnetorheological properties.

(3) The core-shell coating of carbonyl iron powder with polymers, carbon nanotubes and other materials can reduce particle density and improve interfacial compatibility. On the premise of maintaining favorable magnetorheological properties, this modification method can significantly enhance the dispersion stability, thermal stability and redispersibility of magnetorheological fluids.

(4) The application of two-dimensional flaky fiber materials and the optimization of carrier fluids (including high-viscosity oil, ionic liquids and gels) can further inhibit particle sedimentation and broaden the applicable temperature range of magnetorheological fluids. Nevertheless, two-dimensional materials suffer from difficult preparation processes, and high-viscosity base fluids will slightly impair the zero-field rheological properties of magnetorheological fluids.

4. Outlook

Future research will focus on the following aspects to develop high-performance magnetorheological fluids:

(1) Develop novel core-shell structures of carbonyl iron powder compositely coated with carbon nanotubes and two-dimensional materials. This strategy aims to balance high dispersion stability and excellent magnetorheological response, and reduce the negative impact of coating modification on magnetization intensity.

(2) Develop eco-friendly ionic liquids and bio-based carrier fluids to broaden the operating temperature range of magnetorheological fluids, reduce volatilization and corrosion, and adapt to the application requirements of complex working conditions.

(3) Realize the integrated improvement of sedimentation stability, yield stress and redispersibility through multi-component compound modification, by synergistically applying nanoparticles, two-dimensional fibers and functional additives.

(4) Promote the industrial transformation of high-stability magnetorheological fluids from laboratory research to engineering application, develop intelligent and low-cost preparation

processes, and solve the technical bottlenecks in the large-scale preparation of two-dimensional flaky and linear particles.

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