

# Particle Coatings on Magnetorheological Materials

Subjects: General Materials Science

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## Definition

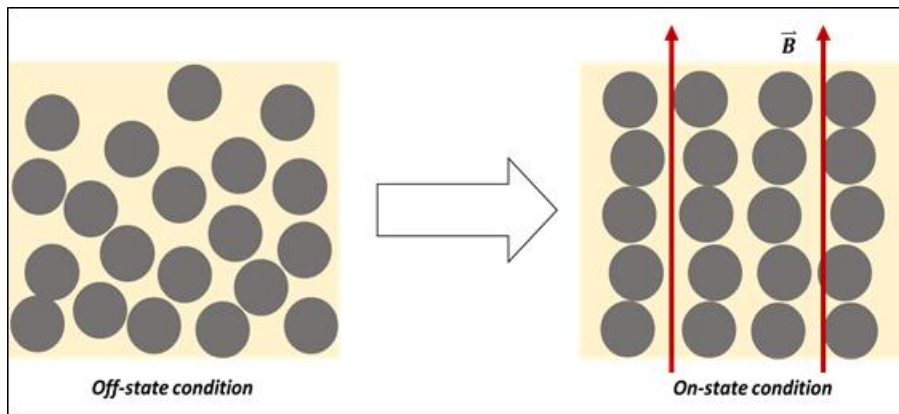
Magnetorheological (MR) material is a type of magneto-sensitive smart materials which consists of magnetizable particles dispersed in a carrier medium. Throughout the years, coating on the surface of the magnetic particles has been developed by researchers to enhance the performance of MR materials, which include the improvement of sedimentation stability, enhancement of the interaction between the particles and matrix mediums, and improving rheological properties as well as providing extra protection against oxidative environments. There are a few coating methods that have been employed to graft the coating layer on the surface of the magnetic particles, such as atomic transfer radical polymerization (ATRP), chemical oxidative polymerization, and dispersion polymerization.

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## 1. Introduction

Magnetorheological (MR) material has captured global interests in the smart materials industry and research, due to its alterable functionalities which correspond with the applied magnetic field. The rheological properties of the materials can be varied accordingly with the application of such external stimuli, therefore MR material is considered to be one of the most promising smart materials which can be applied in engine mounts <sup>[1]</sup>, brake systems <sup>[2][3][4]</sup> and vibration absorbers and dampers <sup>[5][6][7]</sup>, as well as in sensors <sup>[8][9][10]</sup>. These applications have excellent benefits in industries that involve transportation <sup>[11][12]</sup>, seismic prevention <sup>[13]</sup>, soft robotics <sup>[14]</sup>, and prosthetic legs <sup>[15]</sup>.

The first MR material was developed by Rabinow <sup>[16]</sup> in 1948 where a “magnetic fluid clutch” was designed, with a type of magnetizable particles that were mixed in a fluid carrier medium. Under the influence of an external magnetic field, the magnetic particles had been “mutually attracted” and the material “seemingly solidifies”, as mentioned by Rabinow <sup>[16]</sup>. The fluid was then known as magnetorheological fluid, or MRF, with the most commonly used fluid carriers being silicone oil <sup>[17][18][19][20][21]</sup>, synthetic hydrocarbon <sup>[22][23][24][25]</sup>, and mineral oil <sup>[26][27]</sup>. The mechanism of MRF is as illustrated in Figure 1, where during the absence of an external magnetic field (off-state condition), the material behaves like a Newtonian-fluid, however when the external magnetic field is applied upon the material (on-state condition), the micron-sized magnetizable particles in the fluid align to the direction of the applied field. This causes a restriction of the flow of the carrier fluid, thus changing the behavior of the material to become stiffer and more solid-like. This reversible behavior change can occur almost instantaneously. Due to the chain-like structure of the aligned magnetic particles, yield stress is needed to break the particle chains to allow the flow of the fluid, which means that the higher the applied magnetic field to the material, the stronger the particle chain, and thus the higher the force needed to overcome the yield point where the particle chain begins to deform.



**Figure 1.** The mechanism of magnetorheological fluid (MRF) prior and upon the application of external magnetic field, .

MR grease and MR gel were developed later with a much higher viscosity fluid than MRF to minimize the serious sedimentation problem which occurred in MRF, by reducing the density mismatch between the magnetic particles and the carrier medium. MR grease/gel also has a unique property where it possesses solid gel properties at room temperature, but acts like a fluid at a higher temperature.

Meanwhile, magnetorheological elastomer (MRE) is the solid analogue of MRF, where the fluid is replaced by a non-magnetic elastomer matrix as its carrier medium. By using an elastomeric polymer, such as silicone rubber [28][29][30][31], natural rubber [32][33][34][35] and polyurethane [36][37] as its medium, the sedimentation issue in MRF is completely tackled because the magnetic particles are locked within the matrices. It also offers an alternative to the flowy MRF; MRE does not need any vessel to contain it because it is already in a solid form, and therefore there are no leakage or sealing problems in MRE. The particles in MRE are fixed in the network of the elastic polymer, therefore the particles cannot form chain-like structures like MRF when subjected to an external magnetic field, but can only be polarized and mutually attracted to one another, causing a change in the stiffness of the material. The curing process of the material can be done either with or without the presence of an external magnetic field. During the presence of the field, the magnetic particles are aligned in the direction of the magnetic field, causing a columnar particle structure after the sample has cured. This process is called anisotropic curing. Meanwhile, in the isotropic curing method, the magnetic particles are cured and dispersed in no orderly pattern in the MRE due to the absence of a magnetic field. The damping and the storage modulus, as well as loss modulus of these MREs, can be controlled under the influence of various magnitudes of external magnetic fields.

Other less-reviewed MR materials are MR foam and MR plastomer, where the MR foam uses a spongy, absorbent carrier medium such as polyurethane [38][39][40] as its carrier medium. The application of MR foam is also unique; it has been reported that MR foam has a potential application in acoustic absorption [41]. On the other hand, MR plastomer is the latest developed MR material with a carrier medium, such as polypropylene glycol and toluene diisocyanate mixture [42][43], polyvinyl alcohol [44][45], and thermoplastic polyurethane [46], which offers a plasticine-like material that has solid-state between MR gel and MRE. Due to the lack of studies around these forms of MR materials, to the best of our knowledge, there is almost no research involving the incorporation of particle coatings into MR foam and MR plastomer, and therefore in this review, we only focus on the impregnation of coated magnetic particles in the other three MR materials.

## 2. Problems in MR Materials

The advancement of MR materials from one type to another is mainly due to the limitation of one another. For example, because the magnetic particles are much denser than the carrier fluid, sedimentation occurs in MRF during the off-state condition which can lead to another problem—the aggregation of the particles. This aggregation issue may reduce the efficiency of MRF respective to the MR effect that might be reduced over time. Therefore, MR gel/grease was developed to control the

particle-medium density mismatch so that it can have a better redispersibility and thus reduce the sedimentation problem. However, because the matrix-based material is highly viscous, MR gel/grease exhibits a high yield stress at the initial state as well as a lower MR effect compared to MRF.

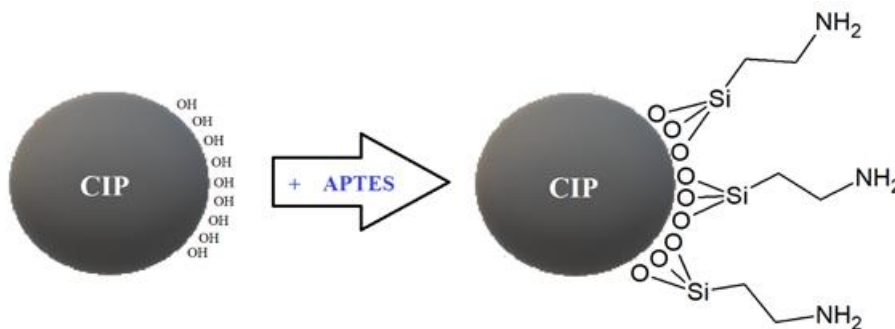
Meanwhile, MRE and MR foam were developed to eliminate the severe sedimentation issue in MRF by embedding the magnetic particles in the elastic and porous matrices, respectively. The leakage and sealant problems in MRF and MR gel/grease has also been overcome by these solid MR materials. However, by locking the magnetic particles in the solid-based matrices, the particles are restricted to move freely and thus inappropriately respond to the applied external magnetic field in the way the particles in MRF do. This affects rheological changes during the off-on state condition in MRE and MR foam, where the MR effect drops several magnitudes in comparison to that of MRF.

Subsequently, the development of MR elastomer may have minimized this MR effect issue, because MR elastomer is reported to have higher MR effect compared to MRE and MR foam, however lower than MRF due to the MR elastomer itself, which exists in the state between both MR materials and possess a high viscosity as well. In addition, the solvent used in MR elastomer may dry up and suffer from a desiccation problem, and therefore it is not suitable for long-term use [44].

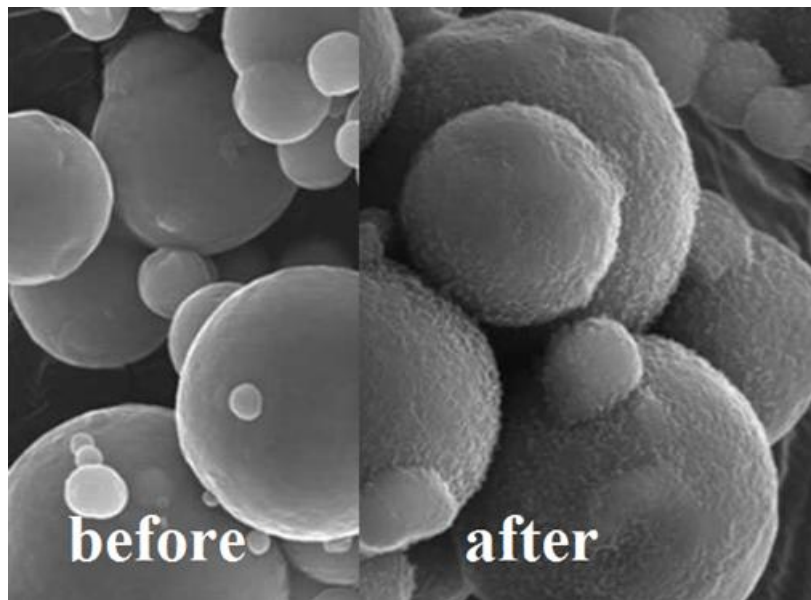
Additionally, because the magnetic particles used in MR materials are made up of metal elements such as pure iron and cobalt, oxidation may also take place, especially for long-term use of applications that could further add problems to MR materials. Due to the huge potential that all respective groups of MR materials could offer, progressive research is continuously being undertaken to improve the internal structure of the materials for enhanced properties. Therefore, a particle coating of the magnetic particles in the MR materials has been proposed as one of the ways to tackle the aforementioned problems.

### 3. Particle Coating in MR Materials

The advantages of coatings in general are known globally; they can provide extra protection for the substrates that is coated as well as enhance the performance of the materials. The coating application on the surface of magnetic particles has emerged in MR studies, to reduce or minimize the issues that have hindered the full capability of MR materials. This is achieved by grafting a layer of polymer onto the surface of the magnetic particles that later will be incorporated in the MR materials. Figure 2 shows an example of a simple coating onto magnetic particles by grafting (3-Aminopropyl) triethoxysilane (APTES) after the surface of the particle has been pre-treated with hydroxyl moieties. On the other hand, Figure 3 shows an example of a micrograph from scanning electron microscopy (SEM) from a successful grafting of coatings onto the magnetic particles as demonstrated by Sutrisno et al. (2013) [47]. It has been proven that particle coating has great benefits to MR materials in terms of improving the sedimentation stability and re-dispersion of the magnetic particles in MRF, protection against wearing and friction, oxidation protection, and enhancement of rheological properties of the MR materials.



**Figure 2.** Grafting of (3-Aminopropyl)triethoxysilane (APTES) onto CIP upon activating the surface of the particle with hydroxyl moieties.



**Figure 3.** Micrograph of before and after a successful grafting of coating on the magnetic particles (Figure has been modified with reference to [\[47\]](#)).

### 3.1. Sedimentation Stability

In a study conducted by Hu et al. (2006) [\[48\]](#), it was found that poly(butyl acrylate)-grafted carbonyl iron particles (CIP) showed a significantly lower settling volume of the magnetic particles in commercial MRF. They also discovered that the sedimentation constant of coated CIP was much lower than that of commercial MRF—the higher percentages of grafted-poly(butyl acrylate) exhibited the lowest sedimentation constant. The team explained that these results are due to the coating layer which acts as a stabilizing layer that can sterically prevent coagulation. Meanwhile, Park et al. (2009) demonstrated that by coating the CIP with poly(methyl methacrylate) (PMMA), the sedimentation problem that occurred in their lubricating oil-based MRF improved due to the lower density of the coated magnetic particles compared to uncoated ones. The re-dispersion of the particles after they were subjected to an external magnetic field was also improved, due to enhanced surface properties such as steric repulsion and electrostatic repulsion of the PMMA coating. A further investigation on the sedimentation stability property of coated magnetic particles in MRF has been conducted by Quan et al. (2014) [\[49\]](#), where a Turbiscan instrument was used to compare the transmission percentage of polystyrene-coated CIP MRF and common silicone oil MRF. At the beginning of the experiment, both MRFs showed zero transmission which represented multiple incident lights scattered through the suspension which could not be transmitted due to many particles that were suspended in both MRFs. However, after 400 min, uncoated CIP MRF exhibited a higher transmission percentage compared to polystyrene-coated CIP MRF, indicating the higher sedimentation stability of the coated sample. The Turbiscan was also used by Tae et al. (2017) [\[50\]](#) to compare the sedimentation ratio of polyaniline (PANI)-coated CIP and pure CIP MRFs by subtracting the transmission ratio from 100% value. In this work, it was found that MRF with PANI-coated CIP exhibited a slower sedimentation speed compared to that of uncoated CIP during the whole 900 min of dispersion period. The team stated that this was due to the thick PANI shell that encapsulated the CIP, causing the density of the particles to reduce significantly. Furthermore, they also claimed that the slightly non-spherical and rough surface of the coated particles also contributed to the improved dispersion stability of the MRF. Moreover, it was also revealed that the time response for coated particles in MRF is faster than that of the uncoated ones, as reported by Nguyen et al. (2014). In this work, the researchers grafted the CIP with silica to be incorporated in MRF for the application of an MR brake, and it was found that the MRF with silica-coated CIP exhibited a higher settling time of 0.450 s compared to 0.342 s for the same MRF with uncoated CIP.

### 3.2. Tribology Properties

Another significant benefit resulting from particle coating in MRF is related to the unique application of

MRF in polishing devices and honing process, as reported by several works [51][52][53][54]. Due to the nature of these works, the magnetic particles used in MRF inevitably experience friction against each other and with the contacting surfaces, which will result in wearing problems of the particles. Furthermore, the use of MRF in dampers and isolators can also cause serious wearing problems to the vessel that contains it [55][58]. Therefore, introducing a coating layer on the surface of the magnetic particles could reduce the fretting problem. It was proven by Bombard and de Vicente (2012) [56], when they compared different grades of commercial CIP in MRF, that it acts as a boundary lubricant between polytetrafluoroethylene (PTFE) and a stainless steel tribopair, which was achieved under a pure sliding contact condition. It was found that OS grade CIP possessed the best friction-reduction behavior compared to the five other grades (OM, OX, HS, HSI and HQ), and even when compared to OM grade that is the same size. This is due to the presence of an amorphous silica coating on the surface of the OS CIP that reduces the friction coefficient of the material and indirectly produces the smallest worn scar diameter in comparison to the other grades. Meanwhile, Zhang et al. (2018) subjected two types of silicone oil-based MRFs to a reciprocating friction and wear test; one was filled with polystyrene-coated CIP and the other one with uncoated CIP. Using a surface profilometer, it was found that the width and the depth of the marks produced by MRF with polystyrene-coated CIP were much narrower and smaller compared to that of uncoated CIP during both off-state and on-state conditions, while the surface of the disc that was coated with CIP MRF was analytically and appeared optically smoother than its counterpart after the wear test. They also discovered that the coefficient of friction of the CIP-coated MRF was lower than that of the uncoated-CIP MRF, especially during the on-state condition. These outcomes undisputedly showed that MRF with polystyrene-coated CIP has better wear and frictional properties than MRF with bare CIP.

In other work conducted by Lee et al. (2015) [57], the CIP was coated with PMMA to be applied in their own MR polishing fluid system. They discovered that by using PMMA-coated CIP in their MRF, the material removal depth of the BK7 glass workpiece was lower while exhibiting lower surface roughness than that of the uncoated one with an optimum rotating wheel speed of 1884 mm/s. This superior surface property of the coated CIP exhibited similar pattern outcomes discovered by the same research team of Lee et al. (2017) [54], but this time, the CIP were coated with a biopolymer, xanthan gum. On the other hand, a study by Hong et al. (2018) [58] also showed the decrease in surface roughness of BK7 glass when the CIP used in their MR polishing fluid was coated with silica, especially when an abrasive was introduced into the MRF. These end results can definitely be utilized in ultra-precision applications in optics and micro parts industries. Another notable outcome achieved by coating the magnetic particles for MR polishing fluid application has been expressed by Salzman et al. (2016) [51], where the CIP was coated with zirconia to be applied as a polishing material on a chemical vapor deposited with zinc sulfide substrate (CVD ZnS). One of the few limitations that hinder MRF from being commercially available for polishing applications is the pebble emergence on the surface of the substrate after the polishing work has been done. The researchers have successfully minimized this problem by using zirconia-coated CIP in their MR polishing fluid in a slightly acidic condition of pH4.

Although there is an increasing number of works which investigate the potential of MRE as a polishing material [59][60], to the best of our knowledge, there is no work that involves coated magnetic particles in MRE for polishing application.

### **3.3. Oxidation**

MR materials contain magnetizable particles that inevitably consist of metal elements such as iron, therefore oxidation of the micron-sized particles undoubtedly can occur; this can happen in wet conditions (with the presence of water molecules) or in dry conditions (without the presence of water molecules, which is commonly known as thermo-oxidation).

It has been proven that oxidation phenomena can present negative impacts on the performance of the MR materials, as stated by Lokander et al. (2004) [61]. In their work, the oxidation of the natural rubber of



MRE increases as the amount of iron particles increases, due to the oxidation of each individual particle. Furthermore, the high concentration of oxygen elements on the surface of the particles, as well as iron elements of the particles, induces iron ions which are some of the root problems of the oxidation issue of MRE, because the particles are directly in contact with the elastomer matrix. In the meantime, Ulicny et al. (2007) [62] validated the adverse effect of oxidation on MRF after the material was tested for durability performance in an MRF fan clutch. It was found that after 540 h of durability tests, the fan clutch torque capacity reduced gradually as the maximum fan speed experienced dropped by 15%. This is said to be due to the increasing oxidation of the iron particles in conjunction with the fact that iron oxides have much lower magnetization compared to elemental iron. The effects of corrosion in MR materials have also been discussed in detail by Plachy et al. (2018) and Burhannuddin et al. (2020) [63] in separate works around MRF and MRE, respectively. Furthermore, rougher surfaces of oxidized particles may lead to irregular polishing performance as well as reducing the lifetime of the vessels that contain the MR materials, because rougher surfaces can cause unnecessary friction between the particles and the wall of the vessels after a long operational period [64]. These problems can be minimized by developing coatings that act as a barrier to the oxidation species. It has been proven by Shafrir et al. (2009) [65] in an accelerated corrosion resistance test to determine the oxidation resistance of their zirconia-coated CIP. This test was done by stirring the coated and uncoated CIP in different batches with pH 4.4 acetic acid aqueous solution. After exposure to the accelerated acidic condition for 22 days, the zirconia-coated CIP did not produce any visible goethite (FeOOH) corrosion product. More evidence to the oxidation resistant coating for magnetic particles in MR materials was demonstrated by Cvek et al. (2015), where the team grafted poly(glycidyl methacrylate) (PGMA) onto the CIP to improve the chemical stability of the particles in acidic condition. After exposing the coated and uncoated CIP to 0.05 M HCl in two different containers, the pH values of PGMA-coated CIP remained almost the same throughout the 90 h of acidic exposure, indicating the CIP corrosion protection provided by the PGMA coating. Meanwhile, the performance of coated and uncoated iron particles in MRE before and after an oxidation test has been displayed by Behrooz et al. (2015) [66]. In their study, the poly(tetrafluoropropyl methacrylate)-coated particles and uncoated particles were embedded into silicone rubber MRE. Both samples underwent an accelerated aging test to induce oxidation of the MREs. It was shown that the MRE with coated particles had the least reduction in effective modulus, and the coated particles were able to preserve the stiffness of the MRE under an oxidative environment.

On the other hand, coated magnetic particles in MR materials also exhibited positive outcomes in terms of thermo-oxidation stability. For instance, a facile coating of silicone oil on the CIP by simply immersing the particles in the dimethyl-silicone oil prior being incorporated into MRE, has been proven to enhance the thermal stability as well as the formation of the cross-linked structure of the MRE [67].

### 3.4. Electromagnetic Shielding

One of the most interesting applications of coated magnetic particles incorporated into MR materials is the ability to shield the materials from electromagnetic waves, which can be used in civil and military fields. This can be achieved when core-shell structure particles, which consist of magnetizable particles coated with electrically conducted shell, are able to absorb unfavorable electromagnetic signals and wave pollution that is subjected to the particles [68]. It has been demonstrated that when tetraethylorthosilicate (TEOS) was grafted onto CIP, the MRE that contained these particles exhibited a higher absorbing ability of ultra-high frequencies of 700 MHz to 1.6 GHz, which is the common operation range of communication and information transmission systems. Through this finding, it was claimed that this was the first time that MRE was reported to have the potential to have sufficient electromagnetic shielding [68].

Furthermore, besides the aforementioned advantages of particle coating in MR materials, there are also other benefits that were obtained indirectly through the coating of the magnetic particles into the MR materials' medium carrier. For instance, in the application of MREs, it was found that the particle's coating has the capability to promote the interfacial interaction between the magnetic particles and elastomer matrices [69], and this would improve inter-bonding between the particles and the matrix. From the

perspective of rheological characterization, it was found that due to the enhancement of the particle-matrix interaction in MRE credited to the grafted coating, the mobility of the particles was enhanced compared to the uncoated particles that were locked within the matrix. This particle mobility reinforcement led to a significant increase in damping property and MR effect of the MRE [70].

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## Keywords

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