Magneto-rheological Suspensions  
— Physical Mechanisms and Modeling —

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A magneto-rheological suspension (MRS) is a particulate suspension which shows a dramatic increase in flow resistance upon application of an external magnetic field. The fundamental physical process is believed to be that the field induces polarization of each particle with respect to the carrier material, and the resulting interparticle forces cause aggregates of particles to form in the field direction. While recent years have witnessed the appearance of several applications using these tunable flow properties, optimal use of MRS technology is still hindered by our incomplete understanding of the underlying mechanisms. This paper surveys recent developments which have improved our understanding of several of the key issues governing the rheological behavior of MRS. In particular, experiments using small strain rheometry and oscillatory shear flow have given insights into the viscoelastic nature of the aggregates before they yield. A recent advance in modeling has been the two fluid continuum approach, which may help the establishment of a more general constitutive framework for MRS.

Key Words: Magneto-rheological suspensions / Polarization / Aggregates / Field-responsive fluids

1. INTRODUCTION

Recent years have witnessed continued strong interest in magneto-rheological suspensions (MRSs). These are “smart fluids” which show a dramatic but reversible increase in flow resistance when placed under an externally applied magnetic field. There has been considerable research activity worldwide in both industry and academe with the aim of improving our understanding of the fundamental mechanisms behind the behavior of MRS. This paper sets out to present a snapshot of some of the recent developments and key unresolved issues related to the mechanisms and modeling of the rheological behavior of MRS.

The tunable flow resistance displayed by these particulate suspensions offers many potential applications, such as adjustable vibration damping devices, clutches and control valves. This wide range of potential engineering uses is a major reason for the ongoing research effort into MRS technology. It is worth noting that some MRS devices are just beginning to appear on the market, such as adjustable automobile shock absorbers (Delphi Automotive Systems LLC, USA) and active vibration dampers for civil engineering structures (e.g. used on the recently constructed Dong Ting Bridge in China\(^1\)). Indeed, MRS technology was the subject of a recent article in the widely read *Popular Science* magazine (“Best of What’s New 2002” feature\(^2\)). Historically, the study of MRS began with the pioneering report by Rabinow in 1948\(^3\), and from the mid-1990s there was an explosion of interest in the field due to advances in manufacturing the particulate materials and compact electromagnetic systems. The focus of this paper will be on the physical mechanisms behind the rheological behavior - readers interested in applications are encouraged to look at a recent review article on that aspect of MRS technology.\(^4\) In general, it should be kept in mind that for most engineering applications, an “ideal” MRS should display a large field-induced change in flow resistance, and have good long term stability against sedimentation and irreversible flocculation.

Despite the ongoing efforts towards industrial application of MRS described above, there still remain many gaps in our knowledge concerning the microstructural mechanisms behind the field-induced rheological behavior. This review paper will highlight some recent advances made in this area, as well as describe key issues which still require investigation. It should be pointed out that review articles on MRS have appeared in the literature over the years.\(^5\)-\(^8\) There are also available...
proceedings of recent international conferences dealing with MRS, the most recent being held in 2001.9,10) This paper will therefore primarily focus on developments since 2001.

This review will be structured as follows. In Section 2, to establish the background, as well as provide a useful guide to the reader new to the field of MRS, a brief review of the main points of the mechanisms behind the field-induced response will be presented. In Section 3, we present a summary of some of the recent advances made in our understanding of the governing mechanisms behind these materials, from both experimental (sub-section 3.1) and theoretical/modeling perspectives (sub-section 3.2). In Section 4, we present some concluding remarks and highlight the issues which still seem to require more investigation.

2. OVERVIEW OF MRS BEHAVIOUR

In this section we will briefly review the main features of MRS, including the basic modeling approaches which have been developed up to now. This section will conclude with a short discussion of some of the shortcomings of the currently available approaches, together with an introduction to a related system, the electro-rheological fluid.

Typically, an MRS consists of a suspension of micron-sized, magnetizable particles (e.g. carbonyl iron) dispersed in an appropriate carrier fluid. The fluid shows a rapid and reversible increase in flow resistance when an external magnetic field is applied. To date, most rheological tests of these systems have been carried out under steady shearing deformation. An example is shown in Fig.1 for a suspension of carbonyl iron particles dispersed in silicone oil. It is apparent that the rheological behavior under steady shearing (with the field applied normally to the shearing planes) can represented by the Bingham fluid model, with the shear stress \( \tau \) given by

\[
\tau = \tau_y + \eta \dot{\gamma}.
\]

Here \( \dot{\gamma} \) is the shear rate and \( \eta \) is the plastic viscosity. \( \tau_y \) is the yield stress induced by the magnetic flux density (B), and depends strongly on B. Typically, MRS can show quite large yield stresses : for example, a 100kPa yield stress under a 1T magnetic flux density has been reported.5,11) In addition to the shear stress, these fluids are known to show positive normal forces under steady shear flow and strong magnetic fields. 12, 13)

The essential mechanism behind the field-induced flow resistance of MRS is as follows: The external field induces polarization within each particle relative to the carrier fluid, and the resulting magnetoostatic interaction forces between the particles lead to the formation of aggregates aligned in the direction of the field. If the sample is deformed, the presence of these elongated aggregates causes the viscosity to increase dramatically (Fig.2). The simplest model of the interparticle forces is based on the “point dipole” approach, which assumes that the particles are spheres which interact through dipoles of fixed strength located at the center of each sphere (the strength of the dipole is taken to be the magnitude of a dipole induced in an isolated sphere under the external field). Based on this simplified interaction force, there have been many studies using particle-level computer modeling to study the aggregate deformation, as well as theoretical calculations of the mechanical strength of each aggregate assuming an idealized arrangement of the particles (e.g. single-width chain of particles, or particles arranged in a lattice). This body of work

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig1.png}
\caption{An example of the field-induced change in rheological properties. Shear stress is plotted against shear rate for an MRS consisting of carbonyl iron particles in silicone oil (volume fraction 0.35) under various magnetic flux densities B, as indicated (units: teslas).}
\end{figure}

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig2.png}
\caption{A schematic diagram of the microstructure in an MRS under an external magnetic flux density (B) and shear flow. The induced dipoles in each particle are represented by the arrows. The dipole-dipole interactions lead to the formation of elongated aggregates which cause the increase in shear stress. Note that particle size has been exaggerated for clarity - typically the particles are micron-sized and the gap is ~ 1mm.}
\end{figure}
has been well summarized in review articles.\textsuperscript{6-7)}

The requirement that the particulate material in MRS be magnetisable means that there is a limited range of possible materials that can be used. Micron-sized carbonyl iron particles are commonly used, since they show low coercivity (low remnant magnetisation) and high magnetic permeability.\textsuperscript{14)} The carrier fluid is often silicone oil or some light mineral oil, and particle volume fractions are typically of the order of 0.3. In addition, newer materials continue to be developed, such as aqueous suspensions of monodisperse colloidal polystyrene spheres which contain magnetic Fe\textsubscript{2}O\textsubscript{3} grains.\textsuperscript{15)} Another recent development is the Magnetic Compound Fluid by Shimada and co-workers - here nm-sized magnetite particles are added to the MRS and the performance of the material as a polishing fluid is significantly enhanced.\textsuperscript{16)} A useful review of the material aspects of MRS technology has been compiled by Phule,\textsuperscript{17)} to which the interested reader is referred.

Despite all this research activity, there still remain shortcomings in our understanding of the mechanisms behind MRS behavior, and here we briefly outline some of the major unresolved issues—recent efforts at addressing these issues will be described in Sections 3 and 4 (to follow). Firstly, most of the characterization and modeling of these materials have been carried out under steady shear flow, and there is a need to extend this to a general constitutive framework relating the deformation geometry and mechanical response. Towards this end, careful rheometric measurement of these systems under small strain flows (e.g. small amplitude oscillations, shear start-up or creep response) or non-shearing flow geometries (e.g. squeeze flow) will improve our understanding of the relationship between aggregate microstructure and the rheological response. The effects of particle size is an important issue which requires clarification. On the modeling side, it would be fair to say that there is a need to move away from calculations of interparticle forces in neatly arranged aggregate structures: there still seems to be a considerable gap between these idealized situations and the complex flow and field geometries encountered in many of the proposed engineering devices.

To conclude this section, it should be pointed out that there is an analogous system to MRS based on electrical effects: the so-called electro-rheological fluid (ERF). An ERF is a field-responsive particulate suspension whereby the mismatch in dielectric properties between particles and carrier liquid leads to particle polarization and aggregation. Under weak fields, there are many similarities in the physical mechanisms behind the response of MRS and ERF: both involve linear polarization of the particles and subsequent formation of aggregates. Under high fields, however, differences between the electric and magnetic systems emerge—e.g., ERF begin to display non-ohmic conduction effects, whereas magnetic saturation effects become important for MRS. Extensive review articles on the mechanisms behind ERF behavior are available in the literature.\textsuperscript{18-21)
to scale as the magnetic flux density \((B)\) as \(B^{1.4}\) (Fig.3), which is in close agreement with the \(B^{1.5}\) dependence predicted for the field-induced interparticle force.\(^{25,26}\) On the other hand, the timescale for growth in \(G'\), which is expected to be related to the timescale of aggregate formation, was found to vary as \(B^{-0.4}\) ie the stronger the field, the faster the growth in \(G'\). At present, there do not appear to be any theoretical models which can explain this field dependence of the timescale. The typical magnitudes measured, under a magnetic flux density of 0.54T, were \(G'\) increasing from 2kPa (before the application of the field) to 5MPa, with an initial jump in \(G'\) after the application of the field (over less than 0.1s), followed by a much more gradual increase of \(G'\) to the final value over 1 s.

Another study using small amplitude oscillatory shear measurements has been carried out on a system under constant magnetic field, and numerical inversion of the linear viscoelastic data to find the relaxation time spectrum has been attempted.\(^{27}\) The results indicate that the MRS under a moderate field of 0.5T has a rather broad distribution of relaxation times, perhaps the variety of rearrangement processes of particle configurations which can be realized in an aggregate under a strong field. In addition, Li et al.\(^{28}\) report an examination of the onset of non-linear viscoelasticity as strain amplitude is increased: it was found that the viscosity component of the material’s response increased with increasing strain amplitude, reflecting the break up or yielding of the aggregate structures. Lastly, although not experimental, it is appropriate to mention here the recent contribution from Sim et al.\(^{29}\), who carried out a particle-level computer simulation of the oscillatory response of a suspension of linearly polarisable spheres (actually an ERF system, which is identical to MRS under weak fields). These workers determined the Lissajous plots (stress vs strain) for large amplitude strains, and found a novel strain-overshoot phenomenon which they explained in terms of the cyclical reformation process of the aggregates.

Another experimental technique which has succeeded in illuminating key aspects of the field-induced response of MRS is based on a physically related system, the “inverse fluid”. This consists of magnetically inert particles (e.g. polystyrene or silica particles) dispersed in a magnetizable ferrofluid. Interestingly, application of an external magnetic field to this system will induce magnetostatic interactions between the particles analogous to those in MRS, and the particles will form elongated aggregates in the field direction, with an accompanying change in rheological properties.\(^{30,31}\) Figure 4 gives an example of the flow curves obtained with an inverse fluid under different magnetic field strengths. It will be seen that the stress levels are somewhat lower than conventional MRS (Fig.1) - indeed it is not proposed to use the inverse fluid as an alternative to MRS in engineering applications, but rather that it should serve as a useful “model” system. The advantage of the inverse system is that it enables us to explore the effects of particle size and shape on magneto-rheological phenomena - such effects are difficult to explore with conventional MRS since the particles must be made from magnetizable materials. For example, experiments by de Gans and co-workers\(^{30}\) used silica particles somewhat smaller (53nm to 189nm) than those typically used in MRS and examined the effects of particle size. They found that there was a tendency for the magneto-rheological performance to improve as larger sized particles were used, although it needs to be kept in mind that the particles in this study were subject to considerable thermal agitation (an effect not usually
encountered in conventional MRS which contain micron-sized particles). It is expected that this inverse fluid approach will find wide application in fundamental studies, enabling us to examine for example the effect of particle size and shape on magneto-rheological behavior. For completeness, it should be noted that while this inverse fluid technique can give us useful insights into the role of the particulate geometry, issues such as the field dependence of the response are not expected to be the same as conventional MRS, since the magnetic response of magnetizable particles (e.g., carbonyl iron) has a field dependence quite different to that of ferrofluids, which consist of nm-sized magnetite particles strongly affected by thermal motion.

Finally, it should be emphasized that most fundamental studies of MRS have focused on the response under simple, uniform shear flow with the magnetic field applied perpendicularly to the shearing planes. In many engineering applications, however, flow geometries more complex than simple shearing are often encountered: for example, a flow field close to squeeze flow (Fig.5) is encountered in optical polishing devices based on MRS.\(^{32}\) To fill this gap in our knowledge, recently there have been experimental studies of the behavior of MRS in deformation geometries more complex than uniform shear flow. For example, Yamamoto and Nakano\(^{33}\) have examined the response of an MRS in a specially developed oscillatory pressure flow rheometer, and have proposed a non-linear dynamic mechanical model to explain the complex waveforms observed. In addition, compressional or squeeze flow (Fig.5), whereby the aggregates are deformed in the same direction as the applied field, has been the object of recent investigations. A comparison of the field \((B)\) dependence of the response under start-up of shear flow and start-up of compressional flow has been reported.\(^{34}\) It was found that the shear stress vs strain curves during shear start-up (constant shear rate) scaled as \(B^{1.4}\), in agreement with theoretical predictions for the mechanical response under shearing (exponent of 1.5).\(^{25,26}\) On the other hand, the curves of resistance force vs strain when constant velocity squeeze flow was applied to the sample scaled as \(B^{0.91}\). Clearly, this difference in field dependence reveals that there are fundamental differences between the mechanical behaviors under shear and squeeze flows, and presently there are no theories available which predict the behavior of MRS under compression. Another interesting perspective on MRS in compressional mode has been provided by Tao,\(^{35}\) who suggested that “superstrong” field responsive fluids could be achieved by first slightly compressing the aggregates normally before applying a shearing deformation. This was confirmed by (i) microscopic studies on suspensions of carbonyl iron particles, where it was directly observed that thick columns of aggregates tended to form after the compression step, and (ii) rheological measurements where the field-induced yield stress was found to increase up to 800 kPa (under a field of 458 kA/m). Thus, although still preliminary, it would appear that these studies may have marked the beginning of the characterization of MRS behavior in flow geometries more complex than uniform, simple shear flow.

### 3.2 Theory and Modeling

Parallel with the experimental studies described above, there has been some progress in recent years towards the development of theory and modeling strategies for MRS. As described in Section 2, the bulk of the modeling work carried out up to now has assumed spherical, magnetizable particles dispersed in a Newtonian carrier liquid. Within this framework, there have been particle-level computer simulations carried out and theories developed, and much of this work has been reviewed thoroughly elsewhere.\(^{6-8}\)

A recently developed modeling approach, which provides a new way of looking at MRS, is worth reviewing. This takes a continuum perspective, and uses a “two fluid” approach to model the mass transport (i.e., particle flux) in MRS.\(^{36}\) The starting point is the two fluid model originally developed for non-colloidal (i.e., hard sphere, non-Brownian) suspensions in shear flow.\(^{37}\) In the MRS we have two phases: the particles and the carrier liquid. A mass conservation equation for the particulate phase is introduced which involves the particle migration flux - the latter in turn can be expressed in terms of the divergence of \(\sigma_p\), the particle-contributed stress tensor. A momentum balance equation for the particle phase is also

![Fig.5 Schematic of the squeeze flow geometry.](image)
introduced which involves the average hydrodynamic force per particle and, again, the divergence of $\sigma_p$. The key issue is thus to find a suitable expression for $\sigma_p$, and here the results of calculations by Shkel and Klingenberg[38] are used to write down an expression for the field-induced particle-contributed stress in terms of the magnetostrictive coefficients. Hydrodynamic interactions between the particles are also accounted for, using the approach of Morris and Boulay.37 The resulting set of equations can be analyzed or solved numerically to predict (i) the changes in the microstructure after application of a magnetic field to a quiescent, dispersed suspension, or (ii) the structures formed during steady shear flow. For quiescent systems, the model predicts the formation of particle-rich columnar structures aligned in the field direction, as observed experimentally. For the steady shear flow, the model predicts the formation of particle-rich “stripes”, which lie in planes containing the flow velocity vector and the magnetic field vector. There have in fact been reports of such layered structures in ERF.39 It should be pointed out that, at present, this two fluid approach is based on the assumption that the initial suspension is isotropic, and further that the polarization is linear (i.e., weak fields). It therefore provides a model for calculating the changes in the particle distribution through the system, but is not able to reproduce the field-induced rheological behavior. Nevertheless, it is felt that this continuum modeling approach is worth taking note of, since it represents a departure from the traditional way of modeling MRS i.e., based on “interacting particles in aggregates”. Indeed, continuum approaches such as this two fluid model may in the future provide a path for developing a general constitutive framework for MRS, which would be able to deal with, for example, the effects of non-uniform magnetic fields.

4. CONCLUDING REMARKS

A brief survey has been presented of some of the recent developments in our understanding of the mechanisms behind the response of magneto-rheological suspensions. It was seen that many insights into the strength of the aggregates can be gained through small strain rheometry and the use of physically analogous systems such as the inverse fluid. This improved understanding of the mechanical behaviour of the aggregates will eventually enable us to find directions for optimizing the performance of MRS. In addition, measurements of the response of this material in deformations other than simple, uniform shear flow showed that the mechanical behavior (indeed the field dependence) is sensitive to the geometry of the flow field. The incorporation of these effects presents a major challenge to developers of theoretical models of MRS behaviour.

As a final note, it is pointed out that there still remains much work to be done to tackle the issue of particle sedimentation as well as explore the possibility of improving performance with suitable wall conditions. Sedimentation is a major issue in MRS due to the high metallic content of the particles, and one promising strategy is to employ a rheologically complex carrier matrix e.g. a strongly shear thinning material. With this, the particle sedimentation under gravity would be hindered, but particle aggregation would still be able to occur rapidly under the stronger magnetostatic forces. A preliminary study using grease as the matrix has shown promising results.40 Regarding the wall conditions, there is the possibility of enhancing the performance of the MRS through the use of suitably profiled wall surfaces (e.g. to provide anchor points for the aggregates). However, very little work appears to have been done to systematically study the effect of wall conditions on MRS performance. There thus remain many challenges to be met before the full potential of MRS technology on engineering can be realized - it is hoped that vigorous research efforts will continue to uncover the mechanisms responsible for the behavior of these fascinating materials.

REFERENCES