On the Yield Stress of Magnetorheological Fluids

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HIGHLIGHTS

• Effect of magnetic field and particle volume fraction are investigated experimentally.
• MRFs with different particle concentration are tested through steady stress sweep test.
• A nonlinear model is proposed for the yield stress of MRFs.
• Model includes two parameters, covers a wide field range and captures magnetic saturation.
• A modified form of the magnetic dipole model is also proposed.

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ABSTRACT

Magnetorheological fluids (MRFs) are a category of functional materials that exhibit magneto-mechanical coupling. These materials exhibit a reversible and instantaneous change from a free-flowing Newtonian fluid to a semi-solid state upon application of a magnetic field. In contrast to ordinary fluids, MRFs can tolerate shear stresses up to a yield value in the presence of a magnetic field. The yield stress strongly depends on intensity of the applied magnetic field and volume fraction of magnetic particles. As the yield stress is the most important parameter of an MRF and must be considered in the design of MR devices, in this work, effects of magnetic field and volume fraction of particles are investigated both experimentally and theoretically. MRF samples with the same carrier fluid but different particle concentrations are analyzed, and an empirical model is proposed for the yield stress of MRFs that covers a wide field strength range and also captures magnetic saturation of the MR fluids. Though the model is mathematically simple, it also includes the effect of particle concentration such that once calibrated, it can be utilized for different particle concentrations as well. Moreover, a modified form of the magnetic dipole model is proposed to model the yield stress of MRFs where an exponential distribution function is utilized to describe the arrangement of particle chains in the presence of a magnetic field. It is shown that, though the model has a simple mathematical formulation, it leads to a reasonable distribution of chains compared to previous similar models.

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1. Introduction

Magnetorheological fluids (MRFs) are a category of functional materials that exhibit magneto-mechanical coupling. This means that, a magnetic stimulation can change their mechanical properties. The inverse action may also be possible however, it is not as interesting as the direct coupling (Bastola and Hossain, 2020; Akbari and Khajehsaeid, 2021). An MRF is a suspension of ferromagnetic microparticles in a nonmagnetic carrier fluid. Most MRFs include some additives as well mainly to prevent agglomeration and sedimentation of particles. These materials exhibit a reversible and instantaneous change from a free-flowing Newtonian fluid to a semi-solid state upon application of a magnetic field where the randomly dispersed magnetic particles get mutually attracted and rearranged to form chains with the tendency to be aligned in direction of the field as shown in Fig. 1.

In the presence of a magnetic field, due to the chains' resistance against flow, MRFs can tolerate shear stress until a threshold value. This threshold is called the yield stress of MR fluid. Yield stress strongly depends on strength of the applied magnetic field. Beyond the yield, the chains collapse however, the mutual attraction of particles can still significantly increase the viscosity compared with the zero-field (off-state) condition (Guerrero-Sanchez et al.,...
The off-state viscosity and yield stress are the most important characteristics of an MRF. Changes in viscosity are reversible for MRFs and occur in a few milliseconds (Chioriero and Science, 2017; Ghaffari et al., 2015). Consequently, the intensity of the applied field can be assumed as the controlling parameter to adjust both yield stress and viscosity of MRFs. There are a few parameters which play role in these properties; type, size, geometry and volume/weight fraction of magnetic particles, viscosity of carrier liquid as well as additives (Chiriac and Stoian, 2010; Leung et al., 2007). The off-state viscosity and yield stress are the most important parameters in the literature as well. For example, Li and Peng (Li et al., 2005) employed the normal probability distribution function to describe the orientation of particle chains with respect to the magnetic field. They also proposed a micro–macro model for the magneto-mechanical behavior of MRFs (Peng and Li, 2007). They used a statistical approach to analyze aggregation of particles as magnetic dipoles forming chains and its contribution to resistance against flow. They assumed normal PDF to describe the orientation of particle chains and investigated the effect of important parameters on the behavior of MRFs.

Yi et al. (Yi et al., 2010) investigated the same problem dropping out the assumption of small particles compared with the inter-particle distances. However, they assumed that the chains are far enough from each other so that the interaction between particles from different chains is negligible. They called it the exact dipole model (EDM). Contribution of a single chain into the shear resistance of an MRF is shown schematically in Fig. 3 both for SDM and EDM. At a small $\theta$, the shear component of the magnetic force increases as well until it reaches a maximum. However, for a large $\theta$ the shear component decreases sharply as the inter-particle distances have increased due to the deformation.

Zhao et al. considered the supplementary magnetization of dipoles induced by the neighboring ones (which is often neglected), in addition to the magnetization by the applied magnetic field (Zhao et al., 2012). They showed that the results of their Enhanced Model (EM) are close to the SDM rather than the EDM. They justified that, as the approximation of particles as magnetic dipoles may enhance the effect of magnetization, the inherent assumption of the SDM partly reduces this enhancement. However, as in the EDM this assumption has been removed, the enhancement of the dipole model is not compensated.

In this study, MRF samples with a range of particle volume fraction are tested through flow ramp analysis to evaluate the effects of

![Fig. 1. Schematic of an MRF in off-state (left), and on-state (right).](image1)

![Fig. 2. Pre-yield chains deformation under an external magnetic field and a shear stress.](image2)

![Fig. 3. Schematic of a single chain's resistance against flow as a function of its orientation.](image3)
of both magnetic field strength and volume fraction of particles on their yield stress. A nonlinear empirical model is proposed to describe the yield stress in terms of the above parameters. Mathematical simplicity, coverage of the whole magnetic field range as well as magnetic saturation of MRFs are sought as the important parameters for the model. The results are compared with the linear and power laws which are widely used in the literature. Moreover, a modified magnetic dipole model is proposed in conjunction with an exponential probability distribution function to describe the particle chains arrangement under the effect of magnetic field and shear stress.

2. Mechanical modeling of the yield stress

2.1. Background

Several researchers have attempted to determine dependency of MRFs’ yield stress on the effective parameters. A number of models have also been proposed however, there is no accurate and reliable correlation that can be used to predict the yield stress of a wide range of MRFs. The effect of magnetic field has received more interest in comparison with other governing parameters. Though a quadratic relation with the magnetic field is proposed in a majority of studies, some experimental observations do not approve this. For example, Claracq et al. investigated the mechanical properties of MRFs at a wide range of magnetic field strength [Claracq et al., 2004]. Their experimental work was conducted at low shear rates but included a 5%-30% volume fraction of particles. They came up with a power law, with exponent 1.5:

$$\tau_y \propto \sqrt{H^3}$$  \hspace{1cm} (1)

Where $\tau_y$ is the yield shear stress and $H$ is the strength of the applied magnetic field. Vereda et al. followed the concept of particle magnetization $M_p$. Their measurements on magnetization of suspensions showed that, $M_p$ can be used to scale the yield stress at low particle concentrations [Vereda et al., 2011]. They proposed the below relation:

$$\tau_y = 2.19 \times 10^{-1} \phi (M_p)^2$$  \hspace{1cm} (2)

where $\phi$ is the volume fraction of particles. Chin et al. also came up with a linear relationship between volume fraction of the carbonyl-iron particles (CIP) and yield stress for dilute MRFs ($\phi < 20\%$). However, they emphasized that the yield stress grows faster at higher volume fractions of CIP [Chin et al., 2001]. Similar to [Claracq et al., 2004], their experimental observations in terms of the field dependency resulted in the below relation:

$$\tau_y \propto \phi M_p^2 B^2$$  \hspace{1cm} (3)

where the magnetic induction $B$ is defined as $B = \mu_0 (H + M_s)$. In general, most investigations have resulted in power law relations. One can refer to [Bossis et al., 2002; de Vicente et al., 2011] among others:

$$\tau_y \propto \phi^\beta H^\gamma$$  \hspace{1cm} (4)

Most studies propose a linear relationship with the particle volume fraction $n = 1$. However, as the magnetic particles may have a nonlinear magnetization, different exponents ($m$) are proposed for low, intermediate, and high magnetic fields. $m = 2$ is used for a linear magnetic material at low magnetic fields while for intermediate fields $m = 3/2$ has been shown to work better in several works. The smaller value of $m$ may be due to saturation of particles at intermediate fields. When the saturation magnetization is achieved, the yield stress no longer varies with the applied field ($m = 0$).

A reliable relation for the yield stress can be used along with a constitutive relation to model the mechanical behavior of MRFs. There are a few constitutive models in the literature, among them the Bingham model is the most well-known and widely used one [Bingham, 1922]:

$$\begin{align*}
\tau > \tau_y : & \quad \tau = \tau_y + \eta \gamma \\
\tau < \tau_y : & \quad \gamma = 0
\end{align*}$$  \hspace{1cm} (5)

In Eq. (5), $\tau$ is the shear stress, $\eta$ is viscosity of the fluid and $\gamma$ is the shear rate. According to (5), shear stress $\tau$ splits into a yield stress $\tau_y$ and a Newtonian term; multiplication of the high shear rate viscosity $\eta$, and the shear rate $\gamma$. The fundamental assumption of the Bingham model is that the flow curve is simply shifted by $\tau_y$ due to the presence of a magnetic field. In other words, when the applied shear stress is larger than the yield stress ($\tau > \tau_y$), the material flows with a post-yield viscosity, $\eta$. When the shear stress is equal to or less than the yield stress ($\tau \leq \tau_y$), the material behaves like a solid with no remarkable flow. This model assumes that the shear rate does not have any effect on the post-yield viscosity. Other models like Casson [Casson, 1959] and Herschel-Bulkley [Herschel and Bulkley, 1926] have also been proposed as shown in (6) and (7), respectively. All these models preserve the above fundamental assumption and mainly try to replace the Newtonian term by a nonlinear term to get better fits of experimental data.

$$\begin{align*}
\tau > \tau_y : & \quad \sqrt{\tau} = \sqrt{\tau_y} + \sqrt{\eta \gamma} \\
\tau < \tau_y : & \quad \gamma = 0
\end{align*}$$  \hspace{1cm} (6)

$$\begin{align*}
\tau > \tau_y : & \quad \tau = \tau_H + k \gamma^n \tau < \tau_y : & \quad \gamma = 0
\end{align*}$$  \hspace{1cm} (7)

The coefficients $\tau_y$, $\eta$, $\tau_H$, $k$ and $n$ vary with magnetic field intensity so the above models need to be calibrated at the particular field intensity that they are aimed to be utilized [Gabriel and Laun, 2009]. Another nonlinear constitutive model has recently been proposed which possesses remarkable features in predicting the mechanical behavior of MRFs [Asiaban et al., 2020]. It has been shown that the predictions of this model are in better correlation with the experimental data compared with the other above-mentioned models. The model is represented as follows:

$$\tau = a_1 \gamma + a_2 B + a_3 B \ln(1 + \gamma)$$  \hspace{1cm} (8)

Where $a_1$, $a_2$ and $a_3$ are the model parameters. $a_1$ is the off-state viscosity of the MRF (viscosity of the fluid in absence of magnetic field) which can be determined from the off-state flow curve (shear stress vs. shear rate at zero field). It is assumed that, MRFs behave almost like Newtonian fluids in the off-state. The second term indicates the dynamic yield stress and the third term corresponds to the shear thinning of MRFs at high shear rates under external magnetic field, thus $a_2$ and $a_3$ can be determined simultaneously from an arbitrary on-state flow curve [Asiaban et al., 2020]. It is observed that, in contrast to [Vereda et al., 2011; Chin et al., 2001], the above model suggests a linear relationship between the yield stress and magnetic field.

In the following, the yield stress of MRFs will be investigated thoroughly to derive an accurate relation between the yield stress and the main influencing parameters, i.e., magnetic field intensity and concentration of particles. Such a relation is required to be used in conjunction with an appropriate constitutive model to provide a reliable mechanical model for MRFs.
2.2. Experimental investigations and mathematical modelling

In the present study, to investigate the yield stress of CIP-based MRFs more precisely, 4 MRF samples have been thoroughly tested in the shear-rate range of 0.001–1000 1/s over a wide range of magnetic field intensity. The combinations were originally developed by Asibian et al. (Asibian et al., 2020). They studied magneto-rheology of MRF samples made of synthetic engine oil as the carrier liquid and carbonyl iron powder as the magnetic particle. In this work, 4 samples called D, E, F and G (according to (Asibian et al., 2020) among the others are examined in terms of the yield stress dependency on the magnetic field and the volume fraction of particles. The samples are made with low (D), moderate (E and F), and high (G) particle weight fractions to cover a rather wide range of CIP content. Regarding the range of viscosity of the samples, the flow ramp test has been employed here to determine yield stresses of the samples. The tests were carried out by means of a parallel plate Anton Paar MCR302 rheometer equipped with MRD180 magnetic accessory. The particle weight and volume fractions of the studied samples are shown in Table 1 where the relation between volume and weight fraction is obtained as:

\[
\phi = \frac{a}{1 - a + b} 
\]

Where \(a\), \(b\), and \(c\) are the carrier fluid’s density, particles’ density, and the weight fraction of particles, respectively.

The calculated yield stresses are shown in Fig. 4 in comparison with the linear and power laws mentioned in (8) and (1), respectively. The test results follow the same trend as reported in (Yang et al., 2019). The figure shows that, the yield stresses increase by both the flux density and the CIP weight/volume fraction. It is also observed that, the linear approximation is more accurate than the power law for these samples. Only for the sample G, the power law is comparable with the linear relation in terms of accuracy. This implies that, the utilized power law is only suitable for MRF samples with high CIP volume fraction. For lower CIP contents, the linear approximation works better thought like the power law it cannot capture magnetic saturation of the MRFs. It is concluded that, the dependency on the magnetic field cannot be described by a single power law as the rate increases first and then decreases as the material approaches its magnetic saturation level. This means that, different power laws will be required for low, moderate, and high field strengths. As mentioned earlier, power laws cannot describe the magnetic saturation, in particular that the saturation level is higher for samples with higher particle content. Therefore, in the present work, a new empirical model is proposed for the yield stress of MRFs that covers a wide field range and also reflects saturation of the MR fluids at high field intensities. Such a model must include both the effect of particle concentration and the field strength:

\[
\tau_s = A \sinh^{-1} \left( \frac{B}{B_{sat}} \right)^2 
\]

Where \(A\) and \(B_{sat}\) are model parameters that depend on the volume fraction of particles. The model predictions are shown in Fig. 5 in comparison to the experimental results. It is observed that the proposed relation well captures variation of the yield stresses with the magnetic flux density. The model is also capable to reflect the magnetic saturation of the samples though it occurs at different field intensities for samples with different particle volume fraction.

The obtained parameters are shown in Fig. 6 in terms of the volume fraction of CIP. It is observed that, variation of \(A\) can be approximated by a quadratic function with respect to \(\phi\). To be more precise, \(A\) is exhibiting a saturating nature with respect to \(\phi\), too however, in the present work for the sake of simplicity a quadratic form has been used which works satisfactorily in the studied range. On the other hand, \(B_{sat}\) exhibits an increasing but saturating variation by increase of \(\phi\) meaning that, the MRF is approaching its saturation level of CIP content. The below relations are proposed for the parameters introduced in (10):

\[
A(\phi) = a\phi^2, \quad B_{sat}(\phi) = \sinh^{-1} \left( \frac{2\phi}{\phi_{sat}} \right) 
\]

Where \(a\) and \(\phi_{sat}\) are material parameters. \(\phi_{sat}\) is the saturation volume fraction of CIP which theoretically refers to 100% weight and volume fraction. However, in the reality due to the large difference in densities of magnetic particles and carrier fluids, as the weight fraction increases, the volume fraction is still far lower. For example, in the case of the samples studied here, as the weight fraction goes beyond 85%, the volume fraction is still around 30%. Thus, \(\phi_{sat}\) depends on the relative density of the particles and the carrier fluid. The model parameters are reported in Table 2.

In the following, the obtained results will be examined by means of micromechanical approaches to obtain a reasonable description for the arrangement of the particle chains in the presence of an external magnetic field.

2.3. Micromechanical approach: A modified magnetic dipole model

As discussed in Section 1, several researchers have employed the magnetic dipole model to determine the yield stress of MRFs. In these models, a variety of probability distribution functions have also been utilized to describe the arrangement of particle chains with respect to the direction of the field. Some works have utilized the simple dipole model (SDM) which assumes that the particles are very smaller than the distance between particles however, this may not be true for intense MRFs. On the other hand, Yi et al. proposed a model called exact dipole model (EDM) which does not need this assumption (Yi et al., 2010). Such micromechanical models investigate the mutual attractions between the magnetized particles in an MRF and first try to formulate the contribution of a single chain to the resistance of MRF against shear flow. The simple dipole model results in:

\[
T_{ch} = \sum_{k=1}^{n-1} \frac{4\mu_0 \pi \mu^2 H^2}{3\Delta^3} (\cos^2 \theta - 1) \cos^2 \theta \sin \theta 
\]

Where \(T_{ch}\) is the contribution of a single chain into MRF’s yield stress. In (12), \(\mu_0\), \(\tau\), \(H\), \(r\), \(\theta\) and \(\Delta\) are the vacuum permeability, particle’s magnetic susceptibility, field strength, radius of particles, orientation of the chain with respect to the magnetic field, and particle’s center-to-center distance, respectively. \(n\) shows the number of particles on a single chain. However, according to the exact dipole model, the contribution of a single chain has been formulated as:

\[
T_{ch} = \sum_{k=1}^{n-1} \frac{2E_k \mu_0 \pi r^2 H^2}{3} \cos^2 \theta \sin \theta 
\]

Where

\[
E_k = \sum_{i=0}^{\infty} \frac{k^2 (2r + 2t + d)^i (-1)^i k^r (2r + 2t + d)}{\sqrt{(k^2 (2r + 2t + d)^i - r^2 \cos^2 \theta)^3}} 
\]
In (14), $t$ and $d$ correspond to thickness of the non-magnetic coating and inter-particle distance, respectively. As discussed in section 1, the enhanced finite element simulations show that the SDM results in more accurate predictions rather than the EDM despite its inherent simplifying assumption (Zhao et al., 2012). Therefore, one may be able to employ an appropriate PDF along with (12) to predict the shear yield stress of MRFs by a rather simple micromechanical model. In this work, an exponential PDF is employed to describe the distribution of particle chains with respect to the direction of applied field:

$$P(\theta) = \lambda e^{-\lambda\theta}$$

where $\lambda = \lambda(\phi, B)$

(15)

Where $\lambda$ is the rate parameter of the exponential PDF. Some distributions according to the exponential PDF are shown in Fig. 7 for different values of $\lambda$. It is observed that, a small $\lambda$ corresponds to rather random distribution of chains while a higher $\lambda$ shows that, most chains are aligned with the applied magnetic field.

The overall yield shear stress can be formulated as $\tau_y = \tau_0 + \tau_y$ in which $\tau_0$ is the yield stress of the MRF in the absence of external magnetic field and $\tau_y$ is the overall contribution of the particle chains. Substituting $A = k\phi$ and $B = \mu_0(1 + \gamma)H$ in (12), we have:

$$T_{oh} = \sum_{k=1}^{n-1} \frac{4\pi^2\chi^2B^2}{3\mu_kk^2(1 + \gamma)^3\phi^3} (5\cos^2\theta - 1) \cos^4\theta \sin\theta$$

(16)

Fig. 4. Linear and power laws in comparison with the experimental yield stresses for: left) samples D and E, right) samples F and G.

Fig. 5. Yield stresses of the MRF samples; comparison of the model results with the experimental data.

In (14), $t$ and $d$ correspond to thickness of the non-magnetic coating and inter-particle distance, respectively. As discussed in section 1, the enhanced finite element simulations show that the
According to Fig. 2, the number of particle chains in the control volume \(\Delta V = \delta \times 1 \times 1\) should be determined in order to calculate the overall chains contribution to the MRF yield stress. The number of chains \(N\) can be calculated by means of the volume fraction of particles:

\[
N = \frac{3\delta \phi}{4\pi r^3}
\]  

(17)

Substituting (17) into (16), assuming \(\delta \approx 2r\) and using the probability distribution function (15) for the particle chains, we have:

\[
\tau_p^{\prime} = \sum_{k=1}^{N-1} \frac{\phi r^3 \lambda^2}{8 \mu_h \lambda^4 (1 + \lambda)^2} \int_0^{\pi/2} (5\cos^3 \theta - 1) \cos^3 \theta \sin \theta \lambda e^{-\lambda r} d\theta
\]  

(18)

Eq. (18) can be used to determine the distribution of particle chains at different magnetic flux densities. Yield stress values obtained from experimental investigations have been used here to determine the rate parameter in (18). The results are shown in Fig. 8 where the values of \(\lambda\) show that, the chains are almost randomly oriented in the off-state (see Fig. 7 as well) as well as in a weak magnetic field however, they become more and more aligned with the applied magnetic field as the field intensity increases. Moreover, MRFs with higher particle content need higher field intensity to get the chains aligned with the field. The results contrast with those obtained in (Guo et al., 2014) where \(\lambda\) was determined in the range of a few thousands. Such high values of \(\lambda\) imply that almost all the chains are aligned with the magnetic field even in the off-state as well as for very weak fields which is obviously not true.

According to Fig. 8, it can be concluded that the modified magnetic dipole model combined with the exponential PDF is a useful method to determine the chains distribution in an MRF as well as the yield stress of MRFs.

### 3. Summary and conclusions

In this work, effects of magnetic field and volume fraction of particles on the yield stress of MRFs were investigated both experimentally and theoretically. This was due to the crucial importance of the yield stress in evaluation of MRFs as well as design of MR devices. MRF samples with the same carrier fluid but different CIP volume fractions were analyzed through flow ramp tests to determine their yield stresses. A nonlinear model was proposed for the yield stress of MRFs where an exponential distribution function was utilized to describe the arrangement of particle chains in the presence of magnetic field. As the model leads to a reasonable distribution of chains, it was concluded that it can be a useful tool in predicting MRFs yield stress or chains arrangement at the presence of external magnetic field.

Yield stress is one of the important characteristics which play role in selection/design of MRFs for a variety of applications. Thus, it is very useful to know how the yield stress changes with respect to the applied field and the magnetic particle content. CIP is the most widely used magnetic particle in MRFs, meaning that the results of the present work would be applicable to a wide range of applications that use CIP-based MRFs. On the other hand, for the control purposes of MR devices (like clutches and dampers), a relation which predicts the MRF’s yield stress is crucial for computation of the output parameters like torque, force, velocity, etc.

**CRediT authorship contribution statement**

**H. Khajehsaeid:** Supervision, Formal analysis, Conceptualization, Methodology. **N. Alaghehband:** Writing – original draft, Data curation. **P.K. Bavil:** Writing – review & editing, Validation, Visualization.

**Declaration of Competing Interest**

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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