On the Effective Magnetic Properties of Magnetorheological Fluids*

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Abstract

Magnetorheological (MR) fluids represent a class of smart materials whose rheological properties change in response to the application of a magnetic field. These fluids typically consist of small (µm) magnetizable particles dispersed in a non-magnetic carrier fluid that generally contains additives such as surfactants and anti-wear agents [1]. Due to such additives, there is an outer non-magnetic layer on the particles that keeps them from touching. The goal of this paper is to study the effective magnetic behavior of an MR composite as a function of the interparticle distance. To this end, we present and employ a model for the effective magnetic properties of MR fluids with periodic microstructure that is based on the theory of homogenization. Finally, we discuss an interpolating formula for the effective permeability of MR fluids as an extension of the work of Keller [3] and Doyle [4].

1 Introduction

Magnetorheological (MR) fluids are suspensions of micron-sized magnetizable particles (such as iron) in a non-magnetic carrier fluid (such as oil or water). The essential characteristic of these materials is that they can be rapidly and reversibly varied from the state of a Newtonian-like fluid to that of a stiff semi-solid with the application of a moderate magnetic field. This feature, called the MR effect, is a consequence of the fact that, in the presence of a magnetic field, the particles magnetize and form chain-like structures that align in the direction of the applied field as depicted in Figure 1. This columnar microstructure, in turn, dramatically increases its resistance to an applied shear strain (see Figure 2). This feature has inspired the design of new technology and various products such as semi-active dampers, brakes, clutches and numerous other robotic control systems [5] (see also http://www.mrfluid.com).

A typical MR fluid includes special additives such as surfactants and anti-wear agents that enhance its performance in MR fluid based devices [1]. These additives may form a non-magnetic layer on the inclusions that keeps them from actually touching. This minimum interparticle distance affects the overall magnetic properties of the fluids [2]. Understanding the magnetic response of MR fluids and its dependence on microstructure is important to the design of improved...

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MR fluids. So here we study the effective magnetic properties of these fluids as a function of the minimum interparticle distance. Predicting the magnetic properties of these fluids, however, is a challenging task due to the highly nonlinear and oscillatory nature of the magnetization of the constituents. As stated, the characteristic size of the particles is several orders of magnitude smaller than the characteristic size of a sample, rendering standard finite element modeling impractical. Thus, in this paper we present a model for the effective magnetic response of MR fluids that is based on the theory of homogenization. We obtain the constitutive equations that govern the B-H response of MR fluids with periodic microstructure in both the linear and nonlinear regimes. The corresponding numerical results exhibit good agreement with experimental data (from experiments carried out at Lord Corporation). However, it becomes increasingly difficult to obtain robust numerical calculations for very small values of the interparticle distance because the necessary mesh size becomes too small. So we also discuss an interpolating formula for the effective permeability of MR fluids for all (positive) values of the interparticle distance. It extends an interpolating relation due to Doyle from a simple cubic array structure to particle-chains. Doyle’s work is based on that of Keller who analyzed the asymptotic behavior of the effective property as the interparticle distance approaches zero.

This paper is organized as follows. In the next section, we review the mathematical model based on homogenization theory and present analytical formulae for the effective properties of MR fluids with periodic microstructure. In §3, we present experimental data collected for three MR fluids and discuss calculating the permeability of a sample from the experimental B-H curve. Then we discuss the corresponding numerical calculations for the effective properties of MR fluids and compare them with experimental data in §4. Finally in §5, we present an interpolating formula for the effective permeability of MR fluids and investigate its applicability by comparing it with the homogenization calculations.

Figure 1: The MR effect: (a) the particles flow in the absence of magnetic fields; (b) they magnetize and form columns when a magnetic field is applied. (c) Schematic of the microstructure under an applied shear strain.

Figure 2: Shear strain rates versus shear stresses in the post-yield regime, for various values of the magnetic field intensity [6].
2 Mathematical model

2.1 Periodic microstructure

In this section we summarize our model for the effective magnetic properties of MR fluids with periodic microstructure that is based on the mathematical theory of homogenization. The results presented here can be found in the literature (see, e.g., [2, 7]). Let $\Omega \subset \mathbb{R}^n$ (n=2 or 3 depending on the geometry that we consider) be a sample of heterogeneous material that, in the presence of a vertical magnetic field, has periodic microstructure that we now describe. Let $Y$ be the parallelogram defined by

$$Y = \left[ \frac{-c_1}{2}, \frac{c_1}{2} \right] \times \cdots \times \left[ \frac{-c_n}{2}, \frac{c_n}{2} \right]$$

for some constants $c_i$ and assume that the permeability matrix $\mu$ of the sample is $Y$-periodic; that is, $\mu(x) = \mu(x + c_i\epsilon_i)$ for every $x \in \mathbb{R}^n$ and for every $i = 1, \ldots, n$. Here the vectors $\epsilon_1, \ldots, \epsilon_n$ are the unit direction vectors in $\mathbb{R}^n$. Then, as depicted in Figure 3, $Y$ is the basic periodicity cell of $\Omega$. The constituents of our MR fluids are assumed isotropic and therefore, we may write

$$\mu = \mu I$$

where

$$\mu = \begin{cases} 
\mu_p & \text{in the particles}, \\
\mu_c & \text{in the carrier fluid}.
\end{cases}$$

(1)

Now we define $\mu^\epsilon : \mathbb{R}^n \to \mathbb{M}^{n \times n}$ by

$$\mu^\epsilon(x) = \mu \left( \frac{x}{\epsilon} \right),$$

where $\epsilon$ is the ratio of the particle size to the characteristic size of a sample. We note that $\mu^\epsilon$ is $\epsilon Y$-periodic in $\mathbb{R}^n$. Then the magnetic field inside the specimen satisfies the fundamental equation of magnetostatics that, for periodic structures, can be written as

$$-\nabla \cdot \left( \mu \left( \frac{x}{\epsilon} \right) \nabla \Phi^\epsilon \right) = 0$$

(2)

where $\Phi^\epsilon$ is the magnetostatics scalar potential. Since the sample is in the presence of a vertical applied field $H = H_n \epsilon_n$, the boundary conditions that complement (2) are

$$\Phi^\epsilon = 0 \text{ at the bottom, } \Phi^\epsilon = H_n \text{ at the top}$$

(3)

and

$$\frac{\partial \Phi^\epsilon}{\partial n} = 0 \text{ on the sides of the specimen}$$

(4)

where $n$ is the unit normal vector to the surface $\partial \Omega$. Since $\mu^\epsilon$ is highly oscillatory, a direct treatment of the governing equations (2)-(4) is impractical. So, in order to find the macroscopic magnetic properties of MR composites, we employ the theory of homogenization.

Figure 3: Schematic of the periodic microgeometry assumed here.
2.2 Linear homogenization

For low field intensities, MR fluids exhibit linear behavior; that is, $\mu$ is independent of the magnetic field. So we study the asymptotic behavior of (2) as $\epsilon \to 0$. We assume that $\mu(x) \in M_{n \times n}^{\text{sym}}$ is $Y$-periodic, symmetric, uniformly positive definite, and each $\mu_{ij}$ is bounded. Then the theory of homogenization provides the following convergence properties as $\epsilon \to 0$:

$$\nabla \Phi^\epsilon - \nabla \Phi^0 \quad \text{weakly in } L^2(\Omega, \mathbb{R}^n)$$

$$\mu \left( \frac{x}{\epsilon} \right) \nabla \Phi^\epsilon - \mu_{\text{hom}} \nabla \Phi^0 \quad \text{weakly in } L^2(\Omega, \mathbb{R}^n)$$

where $\Phi^0$ satisfies the homogenized problem

$$-\nabla \cdot (\mu_{\text{hom}} \nabla \Phi^0) = 0 \text{ on } \Omega.$$  

The (constant) homogenized matrix $\mu_{\text{hom}}$ is given by

$$(\mu_{\text{hom}})_{ik} = \frac{1}{|Y|} \int_Y \left( \mu_{ik}(y) + \sum_{j=1}^{n} \mu_{ij}(y) \frac{\partial w_k}{\partial y_j} \right) dy.$$  

The function $w_k$ is the unique solution to the auxiliary problem

$$\begin{cases}
\nabla \cdot (\mu(y)(\epsilon_k + \nabla w_k(y))) = 0 \text{ in } Y, \\
w_k \in H^1_{\#}(Y),
\end{cases}$$  

where $H^1_{\#}(Y)$ denotes the set of periodic functions in $H^1(Y)$ with zero mean. Here the convergence properties (5) imply that for sufficiently small $\epsilon > 0$, (7) provides the effective magnetic permeability of the MR composite (i.e., $\tilde{B} = \mu_{\text{hom}} \tilde{H}$). Calculating the homogenized matrix requires solving the boundary value problems (8) for $w_k (k = 1, 2, 3)$ and substituting $w_k$ into equation (7).

2.3 Nonlinear Homogenization

Magnetic materials such as iron possess a magnetic saturation $M_s$; that is, a maximum achievable magnetization that, once reached, does not allow any further magnetization. Thus, for moderate to large valued fields, the permeability is, in fact, a function of the magnetic field $\tilde{H}$. Then we study the asymptotic behavior of

$$-\nabla \cdot (\mu \left( \frac{x}{\epsilon} \right) \nabla \Phi^\epsilon) = 0$$

where the B-H map $(y, \xi) \in \Omega \times \mathbb{R}^n \to \mu(y, \xi) \xi \in \mathbb{R}^n$ is assumed to be $Y$-periodic in $y$. Furthermore, assuming the map to be Lipschitz-continuous, strictly monotone and coercive in $\xi$, then one can prove the following convergence properties of (9):

$$\nabla \Phi^\epsilon - \nabla \Phi^0 \quad \text{weakly in } L^2(\Omega, \mathbb{R}^n)$$

$$\mu \left( \frac{x}{\epsilon} \right) \nabla \Phi^\epsilon - b_{\text{hom}}(\nabla \Phi^0) \quad \text{weakly in } L^2(\Omega, \mathbb{R}^n)$$

where $\Phi^0$ satisfies the homogenized problem

$$-\nabla \cdot (b_{\text{hom}}(\nabla \Phi^0)) = 0.$$  

The map $b_{\text{hom}}(\xi)$ is defined for all $\xi \in \mathbb{R}^n$ by

$$b_{\text{hom}}(\xi) = \frac{1}{|Y|} \int_Y \mu(y, \xi + \nabla w_\xi(y))(\xi + \nabla w_\xi(y)) dy,$$

where $w_\xi$ is the unique solution to the auxiliary problem

$$\begin{cases}
\nabla(\mu(y, \xi + \nabla w_\xi(y))(\xi + \nabla w_\xi(y))) = 0 \text{ in } Y, \\
w_\xi \in H^1_{\#}(Y).
\end{cases}$$
In our calculations, we use an empirical constitutive relation known as the Fröhlich-Kennelly relation given by

\[ \mu(\vec{H}) = 1 + \frac{(\mu_0 - 1) M_s}{(\mu_0 - 1) |\vec{H}| + M_s} \]  

(14)

to determine the permeability of the iron particles at a given field strength \( \vec{H} \). In comparison to the linear case, calculating the effective constitutive law (12) requires solving a continuum of the nonlinear equations (13). In §4 we will present numerical results for both equations (7) and (12).

3 Experimental data

In this section we turn to a discussion of the experimental setup and discuss calculating the permeability of a MR fluid sample from the experimental B-H curve. Experiments were conducted (at Lord Corporation) to measure the magnetic properties of three MR fluids with particle loadings of 10%, 20%, and 30% by volume. The arrangement used to collect the data (KJS model HG-500 magnetic Hysteresis graph) is schematically depicted in Figure 4: a current is passed through an electromagnet and a probe is used to measure the applied field \( \vec{H} \) where \( \vec{H} = H_3 \hat{k} \); the magnetic induction \( \vec{B} = \vec{B}(\vec{H}) \) can then be evaluated from the cylindrical sample with the aid of a search coil. In Figure 5 we display the resulting intrinsic induction \( \vec{B}_i = \vec{B} - \mu_0 \vec{H} \) as

\[ \mu_0 \vec{H} \]

\[ \text{SPECIMEN (MR FLUID)} \]

\[ \text{ELECTROMAGNET} \]

\[ \text{H} \]

\[ \text{B(\text{H})} \]

Figure 4: The experimental setup.

a function of \( \mu_0 \vec{H} \). Since hysteresis is negligible in each case, in our analysis, we only consider data in the first quadrant that correspond to the second part of the hysteresis loop.

\[ \begin{array}{c}
\text{Intrinsic Induction [T]} \\
\text{pH[T]} \\
\end{array} \]

\[ \begin{array}{c}
30\% \\
20\% \\
10\% \\
\end{array} \]

\[ \begin{array}{c}
0.8 \\
0.6 \\
0.4 \\
0.2 \\
0.0 \\
-0.2 \\
-0.4 \\
-0.6 \\
-0.8 \\
\end{array} \]

\[ \begin{array}{c}
-1 \\
-0.5 \\
0.5 \\
1 \\
\end{array} \]

Figure 5: Experimental magnetic induction curves for three MR fluids of different iron volume percents.
For the linear permeability of the sample, we are interested in calculating the permeability as measured at very low fields. Since the magnetic induction is approximately linear for small fields, we take the permeability to be the slope of the B-H curve in this region. We calculate this slope by conducting a linear least squares fit of the data. While it is important to use enough data so that the results are accurate and robust, the fit should be conducted only over the linear regime. To this end, consider the following notation:

\((H_k, B_k) = \) \(k\)th data point of the B-H curve

\(\tilde{H}_k = \) \(\mu_0 H_k\)

\(s_n = \) slope of the fit using the first \(n\) data points

\(b_n = \) y-intercept of the fit using \(n\) data points

\(\tilde{B}_n = s_n \tilde{H}_k + b_n\)

\(r_n = \frac{1}{n-1} \sum_{k=1}^{n} |B_k - \tilde{B}_n|^2\) (residual mean square).

The slope and residual mean square of each fit are shown in Figure 6. We selected the linear regime as the set of \(n\) points whose fit yielded the minimum residual mean square and denote this set by the interval \((0, H_1)\). As a result, the permeabilities and \(H_1\) are summarized Table 1.

<table>
<thead>
<tr>
<th>(%)</th>
<th>(\mu_{\text{eff}})</th>
<th>(\mu_0 H_1) (T)</th>
</tr>
</thead>
<tbody>
<tr>
<td>10%</td>
<td>3.07</td>
<td>0.0142</td>
</tr>
<tr>
<td>20%</td>
<td>5.02</td>
<td>0.0141</td>
</tr>
<tr>
<td>30%</td>
<td>6.97</td>
<td>0.0158</td>
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</tbody>
</table>

Table 1.

4 Numerical experiments

In this section we present the numerical results for the linear and nonlinear effective constitutive laws based on homogenization theory as stated in §2 and compare these with the experimental data that was presented in §3. The specific geometry that we consider here consists of a \(n\)-dimensional (\(n=2\) or 3) static configuration of spherical iron inclusions (such as that in Figure 3). We assume the inclusions form single chains in the direction of the applied field that are periodically dispersed throughout the carrier fluid. We denote by \(a\) the ratio of the vertical interparticle distance, \(2a\), to the particle diameter. We take the (fixed) period along the direction of the field to be unity and the period in the other directions to be \(l\) (for simplicity, it is assumed that all lengths are dimensionless). Then, the periodic microstructure is characterized by the periodicity cell \(Y = [0, l]^{n-1} \times [0, 1]\) as is shown in Figure 7. The physical parameters used here are \(\mu_p = 2000, \mu_e = 1\) and \(\mu_0 M_s = 2.15\) T (tesla) where \(\mu_0 = 4\pi \times 10^{-7} T m/A\) is the permeability of vacuum.
First, we present the numerical results based on our two-dimensional model geometry ($n = 2$) for the linear case. The elliptic equation (8) was solved to compute $(\mu_{\text{hom}})_{22}$ (where $(\mu_{\text{hom}})_{22}$ represents the effective permeability in the direction of the applied field) using a standard finite element method based on piecewise-linear elements. In Figure 8, the experimental data are shown along with the results of our homogenization calculations for values of the ratio $a = 0.008$, $0.012$, $0.016$, $0.02$. We remark that the numerical calculations for $a = 0.016$ agree well with the experimental values for all three fluids.

In Figures 9-12, we depict the effective B-H curves (from linear regime through saturation) based on equations (12)-(13) assuming $a = 0.002$, $0.016$. We constructed the effective B-H curves by interpolating the discretely calculated values of $b_{\text{hom}}(\xi)$ for $\xi = \vec{H} = (0, H_2)$, $\mu_0 H_2 \in (0, 0.8)$. For each fluid, the estimates for $a = 0.016$ provides a good fit not only in the linear region, but through saturation as well.

Figure 9. Effective magnetization for the 10% sample ($a = 0.002, 0.016$) compared to experimental data.
Next we discuss the numerical results for the three-dimensional model geometry \((n = 3)\) and we only consider the linear case. The linear elliptic equations (8) were solved using a commercial finite-element software package [9]. The results for \((\mu_{\text{hom}})_{33}\) are shown in Figure 12 for \(a = .0005, .001, .002, .004\). We find the agreement to be reasonable, given the idealized nature of the assumed microstructure. Indeed, some degree of polydispersity was certainly present within our samples and, of course, the true columnar structures were more complex than simple linear chains. In fact, this complexity is accentuated at larger volume fractions. This may explain the slight increase in the deviation as the particle concentration is increased.

Figure 10: Effective magnetization for the 20\% sample \((a = .002, .016)\) compared to experimental data.

Figure 11: Effective magnetization for the 30\% sample \((a = .002, .016)\) compared to experimental data.

Figure 12. Linear effective permeability computed for the three-dimensional model geometry.
5 Other approximations for $\mu_{\text{eff}}$

In this section we discuss an interpolating formula for $\mu_{\text{eff}}$ of MR composites for $n=3$. We seek an interpolating relation which provides a good approximation for all values of $a$. To this end, we consider linear composites that contain spherical infinitely permeable inclusions (for iron inclusions, $\mu_p = 2000$ so this is a reasonable case to consider). It becomes increasingly difficult to obtain accurate numerical estimates as $a \to 0$. However, J.B. Keller showed that, when the particles are uniformly dispersed (on a cubic lattice), the asymptotic behavior of $\mu_{\text{eff}}$ is logarithmic as $a \to 0$ [3]. Based on this work, Doyle suggested the interpolating formula

$$\mu_{\text{eff}} = \mu_c[A + B \log(\phi - \phi_c)]$$  \hspace{1cm} (15)

where $\phi$ is the volume fraction of the inclusions and $\phi_c = \pi/6$ is the maximum volume fraction attained at $a = 0$ [4]. To find the constants $A$ and $B$, Doyle then considered the following conditions on $\mu_{\text{eff}}$ at $\phi = 0$:

$$\mu_{\text{eff}} = \mu_c$$ \hspace{1cm} (16)

$$\frac{d\mu_{\text{eff}}}{d\phi} = 3\mu_c.$$ \hspace{1cm} (17)

Equation (17) was obtained using the well-known asymptotic estimate attributed to Maxwell [10] given by

$$\mu_{\text{eff}} \approx \mu_c \left( \frac{\mu_p + 2\mu_c + 2\phi(\mu_p - \mu_c)}{\mu_p + 2\mu_c - \phi(\mu_p - \mu_c)} \right).$$ \hspace{1cm} (18)

Maxwell’s estimate is valid for small volume fractions and becomes exact as $\phi \to 0$. From (16)-(17), one can determine $A$ and $B$ and (15) can be written as

$$\mu_{\text{eff}} = \mu_c \left[ 1 - \frac{\pi}{2} \log \left( \frac{1 - 6\phi}{\pi} \right) \right].$$ \hspace{1cm} (19)

Equation (19), known as Doyle’s estimate, is now compared with our homogenization estimates. As is seen in Figure 13, the agreement is good throughout the entire range of volume fractions.

![Figure 13: Homogenization estimates and equation (19) for the effective permeability.](image)

Doyle’s estimate is for cubic structures whereas the microstructure for MR fluids is columnar. In fact, cubic geometry is a special case of the model geometry depicted in Figure 7 with $l = 1$. So it is our intent to seek an extension of Doyle’s estimate for $l \geq 1$. Our preliminary investigations suggest that the $e_3$-component of $\mu_{\text{eff}}$ is logarithmic as $a \to 0$ or, equivalently, as $\phi \to \phi_c$ (the
details are to be reported in future work). Following the procedure described above, we obtain the following extension for \( l \geq 1 \)

\[
\mu_{\text{eff}} = \mu_e \left[ 1 - 3\phi_e \log \left( 1 - \frac{\phi}{\phi_e} \right) \right]
\]

(20)

where \( \phi_e = \pi/6l^2 \) is the maximum volume fraction. Of course, (20) coincides with Doyle’s estimate for \( l = 1 \). The results for the extreme case \( \phi_e \approx \pi/2 \% \) (\( l = 8 \)) are listed in Table 2. Also shown is the relative difference of the interpolation relation from the homogenization estimates. The methods are in reasonable agreement throughout the entire range of possible volume fractions.

<table>
<thead>
<tr>
<th>( \phi ) (%)</th>
<th>Homogenization</th>
<th>Interpolation</th>
<th>Relative Difference</th>
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</thead>
<tbody>
<tr>
<td>0</td>
<td>1.0000</td>
<td>1.0000</td>
<td>-</td>
</tr>
<tr>
<td>0.10</td>
<td>1.0016</td>
<td>1.0032</td>
<td>1.64 \times 10^{-3}</td>
</tr>
<tr>
<td>0.20</td>
<td>1.0037</td>
<td>1.0069</td>
<td>3.12 \times 10^{-3}</td>
</tr>
<tr>
<td>0.50</td>
<td>1.0064</td>
<td>1.0112</td>
<td>4.75 \times 10^{-3}</td>
</tr>
<tr>
<td>0.80</td>
<td>1.0130</td>
<td>1.0165</td>
<td>3.38 \times 10^{-3}</td>
</tr>
<tr>
<td>0.50</td>
<td>1.0212</td>
<td>1.0232</td>
<td>1.91 \times 10^{-3}</td>
</tr>
<tr>
<td>0.60</td>
<td>1.0319</td>
<td>1.0324</td>
<td>4.86 \times 10^{-4}</td>
</tr>
<tr>
<td>0.70</td>
<td>1.0471</td>
<td>1.0475</td>
<td>3.37 \times 10^{-4}</td>
</tr>
<tr>
<td>0.80</td>
<td>1.0926</td>
<td>1.0935</td>
<td>5.56 \times 10^{-4}</td>
</tr>
</tbody>
</table>

Table 2.

In Figures 14-15 we compare the interpolating relation with estimates obtained from homogenization theory (see Figure 12) for \( a = .004, .002, .001, .0005 \). Experimental data are also shown. The interpolating formula is based on infinitely permeable particles, and therefore, yields larger permeabilities than the homogenization model. In comparison to experimental data, the interpolating formula for \( a = .0005 \) exhibits good agreement with experimental data for all three fluids.

Figure 14. Homogenization estimates and equation (20) for the effective permeability compared with experimental data for \( a = .004, .002 \).
6 Conclusions

We presented experimental data that we collected for three MR fluids: 10%, 20% and 30% by volume. We discussed calculating the linear permeabilities using least squares and found them to be approximately in the range from 3 to 7. In order to predict the experimental data, we proposed a model for the effective magnetic properties of MR fluids that is based on the theory of homogenization. Analytical formulas for linear and nonlinear media with periodic microstructure were discussed. For the numerical results, we considered microstructures of monodispersed spherical particles arranged periodically in single particle chains. Interestingly, even with this simplified model, the homogenization results predicted the experimental data with acceptable accuracy. However, the method is not restricted to these specific microstructures or particle shapes. We also derived an interpolating formula for the effective permeability of composites that contain infinitely permeable inclusions arranged in particle-chains. This relation showed good agreement with experimental data. Hence, it may provide a simple yet reasonably accurate method for computing the effective permeability of MR fluids.

7 References


