Constitutive model for shear yield stress of magnetorheological fluid based on the concept of state transition

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Abstract
Magnetorheological fluid (MRF) is a smart material whose rheological properties can be varied by a magnetic field; it has been applied in the development of semiactive dampers for a variety of applications. The material essentially consists of a suspension of magnetic particles in a nonmagnetic carrier fluid. It is important to understand the magnetic response of MRF and its dependence on several parameters for improving and designing MRF devices. The purpose of this work is to develop a constitutive model that describes the behavior of the shear yield stress of the material as function of the magnetic field and composition. Taking into account that the material changes its rheology and apparent viscosity according to magnetic field, a magnetically induced state transition is proposed; by the use of a state transition equation, a constitutive model for shear yield stress is defined, consisting of an expression that relates composition of the material and the stimulus applied, it also associates the volume fraction of particles, magnetic field and the material that composes the particles.

Keywords: magnetorheological fluid, smart material, model, constitutive equation

(Some figures may appear in colour only in the online journal)

1. Introduction
Magnetorheological fluid (MRF) is a smart material whose rheological properties can be varied by an external magnetic field (Susan-Resiga 2009, Bompos and Nikolakopoulos 2011), changing from a liquid Newtonian fluid state to that of a stiff semisolid (Simon et al 2001).

MRFs were first developed in 1940s by Winslow and Rabinow (Ginder et al 1996) and, because of their mechanical properties can be controlled, there have been triggered applications of different industries like automotive dampers, prosthetic legs, clutches, among others (Koo et al 2006, Çeşmeci & Engin 2010, Shah et al 2014). The material essentially consists of a suspension of micron-sized magnetically permeable particles dispersed in a nonmagnetic carrier fluid (Simon et al 2001, Sedlacik et al 2013), which may include surfactants, antiwear, thermal-stability, and other stability agents (Phulé and Ginder 1998). The mechanism responsible of this behavior is the induced magnetic interaction of particles within the matrix (Jolly et al 1996). Such particles, in the presence of a magnetic field, get magnetize and move from a random position into anisotropic chain-like structures parallel to the field streamlines (Sedlacik et al 2013). Consequently, the mechanical energy needed to yield chain-like structures increases according to the magnetic field (Carlson and Jolly 2000). The configuration and rigidity of the chain structure will depend upon several factors including the strength and distribution of the applied field (Jolly et al 1996, Laun et al 2008). Macroscopically, it is observed that when exposed to a magnetic field MRF does not flow unless acted by at least some critical shear stress.
called shear yield stress $\tau_y$ (Molyet et al 2006), such behavior is represented at figure 1.

The change in state is accompanied by an increase in viscosity (Ciocanel et al 2006). However, the property that is observed to change upon an increase in applied field is not the viscosity, but rather the shear yield stress, defining the flow regime (Weiss and Duclos 1994). Thus, a MRF could be defined as a material whose flow ability is reversibly and controllable by an external magnetic field (Laun et al 2008) and, because of it, shows a magnetically variable shear yield stress (Nakano et al 2005) which produces a change in its apparent viscosity (An and Kwon 2003).

Understanding the response of MRF and its dependence on microstructure is important for the design of the material. Predicting the magnetic properties of these fluids, however, is a challenging task due not only to the size and density of the particles, but also to their nonlinear magnetization (Simon et al 2001). In addition, the material, shape and coating of the suspended particles have shown to importantly affect the MRF performance (Sedlacik et al 2011, Sedlacik et al 2013, Shah et al 2014). Several authors as Carlson and Ginder have tried to explain its behavior in form of a constitutive equation but due to its nonlinear magnetization there is not a model which relates the maximum shear yield stress, material composition and magnetic field with the shear yield stress in the same expression. For instance, the purpose of this work is to develop a semi-empirical constitutive model that describes the behavior of the shear yield stress of the material as function of the magnetic field, the particle’s material and amount.

2. Proposition of state transition

The alignment of particles can be considered as a magnetically induced state transition. Based on this it is assumed, as depicted at figure 2, that there is a quasi-liquid state (hereafter liquid), when the particles are in random position, and a quasi-solid state (hereafter solid) when the particles are fully aligned. State transition is shown at figure 3; without magnetic field, volume fraction of the liquid state is $\sim 1$, as magnetic field increases towards infinity, volume fraction of the solid state increases to $\sim 1$.

Macroscopically, transition to solid state can be identified by the increase of the shear yield stress as function of the magnetic field, then, shear yield stress is the key parameter to describe the state transition. According to literature, it has the behavior illustrated at figure 4 and depends of the amount and composition of particles on the carrier fluid. At some value of magnetic field shear yield stress becomes constant due to alignment of particles, such value is known as the saturation shear yield stress (Ginder and Davis 1994). At that point it is considered that liquid–solid state transition is finished.
3. Formulation of model for state transition and yield stress of MRF

A constitutive model for state transition of MRF is based on the work of Cortes et al for stainless steels (Cortes Ramirez et al 1992, Tsuta and Cortes Ramirez 1993). In the case of MRF, the liquid and solid states are the aggregates and the microstructural transition between them is the basis of the model; this way, it is in terms of the magnetic field, saturation stress of each state and its volume fraction.

3.1. Constitutive model for shear yield stress

Volume fraction \( V_l \) of each state is defined as:

\[
V_{fil} = \frac{V_l}{V_i}, \quad V_{fs} = \frac{V_s}{V_i},
\]

where subscripts \( l, s \) and \( t \) indicate liquid, solid and total, respectively. Cortes et al constitutive model of flow stress for multi-phases aggregate adapted to MRF is:

\[
\tau_y = V_{fi} \cdot \tau_{y}^{sat} + V_{fs} \cdot \tau_{ys}^{sat},
\]

where \( \tau_y \) is the shear yield stress, \( \tau_{y}^{sat} \) and \( \tau_{ys}^{sat} \) are the saturation shear yield stress of each state.

3.2. Kinetics of magnetic field induced liquid–solid state transition

Based on the state transition at figure 3, \( V_l \) varies by:

\[
V_0 + V_{fs} = 1.
\]

Magnetic field induced solid state is described by state transition equation (STE):

\[
V_{fs} = \left[ 1 + \left( \frac{H}{H_c} \right)^{-B_{MRF}} \right]^{-1},
\]

where \( H_c \) is the characteristic magnetic field at which 50% of the state transition occurs and \( B_{MRF} \) is a constant fitted based on the experimental data represented as in figure 7.

Due that solid state grows from liquid state:

\[
V_{fs} = 1 - V_{fi}.
\]

4. Experimental quantification of state transition

4.1. Material

Three Lord MRFs are used for this work: MRF-122EG, MRF-132DG and MRF-140CG. Details of these materials are listed at table 1.

4.2. Methodology

4.2.1. Shear yield stress–magnetic field characterization.

The constitutive model is fitted by considering the experimental \( \tau_y-H \) data of figure 5 provided by the MRF supplier (LORD Corporation 2010).

4.2.2. State transition estimation. From the \( \tau_y-H \) data of figure 5, liquid–solid state transition is estimated by considering the \( \tau_y \) axis a \( V_l \) axis, with values between 0 and 1. It is assumed that at maximum magnetic field (when shear stress tends to be constant) \( V_{fs} \) is 1, at 50% of the state transition an inflexion point of \( \tau_y-H \) curve exists and \( V_l \) of both states is 0.5, such point is the characteristic magnetic field \( H_c \). This transition for every MRF is plotted at figure 6.

4.2.3. Estimation of saturation yield stress. By equation (6) proposed by Ginder et al \( \tau_y^{sat} \) is:

\[
\tau_y^{sat} = \frac{4}{5\sqrt{2}} \cdot \xi (3) \cdot \phi \cdot \mu_0 \cdot M_s^2,
\]

where \( \xi(3) \) is a constant determined by Ginder et al \( \mu_0 \) is the permeability of vacuum and \( M_s \) is the saturation magnetic field of the material used as particles.

4.3. Results

4.3.1. Yield stress–magnetic field characterization. Figure 5 shows the plot of \( \tau_y-H \) for MRF-122EG, MRF-132DG and MRF-140CG.

4.3.2. State transition estimation. Based on the experimental data from figure 5, the estimated saturation values on table 1 and the assumption that \( V_{fs} \) is ~1 at saturation, figure 6 shows the estimated liquid–solid state transition based on the variation of \( V_l \) of each state as function of \( H \) for each MRF.

From figure 6, \( H_c \) can be expressed as a function amount \( \phi \) of particles:

\[
H_c = \alpha \phi^2 + \beta \phi + \chi,
\]

where \( \alpha, \beta \) and \( \chi \) are constants which depend of the material of the particles.

4.3.3. Estimation of saturation yield stress. Resulting \( \tau_y^{sat} \) as function of \( M_s \) by using equation (6) and the same values of
constants than Ginder et al (listed at table 3) for each MRF is listed at table 2.

5. Fitting of constitutive model

5.1. Kinetics of state transition

Based on the experimental $V_y-H$ data of figure 5, STE is fitted. The values of $\alpha$, $\beta$ and $\chi$ model the dependency of $H_c$ to the volume fraction of particles $\phi$, and they are estimated based on the experimental values. In order to determine the role of $B_{MRF}$, $V_{fs}$ values are plotted versus $H/H_c$ values of the three MRFs. Per least squares method, it is shown that $B_{MRF}$ can be defined by a specific value, which is independent of $H$ as shown at figure 7. Substituting equation (7) into (4):

$$V_{fs} = \left[ 1 + \left( \frac{H}{\alpha \phi^2 + \beta \phi + \chi} \right)^{-B_{MRF}} \right]^{-1}.$$  (8)

The values of $\alpha$, $\beta$ and $\chi$ for the MRFs used in this work are listed at table 3.

5.2. Saturation shear yield stress

Assuming that the carrier fluid has a $M_s$ of 0, due that such material does not respond to magnetic fields:

$$\tau_{ys}^{sat} = 0.$$  (9)

| Parameter | Value | Units       |
|-----------|-------|-------------|
| $\alpha$  | -1547.2 | kA m$^{-1}$ |
| $\beta$   |     844  | kA m$^{-1}$ |
| $\chi$    |   -30.544 | kA m$^{-1}$ |
| $B_{MRF}$ |      2.41 | —           |
| $M_s$     |   831.23   | kA m$^{-1}$ |
| $\mu_0$   |   4$\pi \times 10^{-7}$ | H m$^{-1}$ |

Table 2. $\tau_{ys}^{sat}$ of MRF-122EG, MRF-132DG and MRF-140CG fluids.

| Material       | $\tau_{ys}^{sat}$ |
|----------------|-------------------|
| MRF-122EG      | $2.38 \times 10^{-8}M_s^2$ |
| MRF-132DG      | $3.46 \times 10^{-8}M_s^2$ |
| MRF-140CG      | $4.32 \times 10^{-8}M_s^2$ |

Table 3. Material constants determined from experimental data for MRF-122EG, MrF-132DG and MRF-140CG fluids.
6. Validation of constitutive model

Substituting equations (6), (8) and (9) into (2), a constitutive model for shear yield stress of MRF is obtained in terms of the magnetic field, amount and material of particles:

\[ \tau_y = \left[ 1 + \left( \frac{H}{\alpha \mu + \beta \rho + \chi} \right)^{-\beta \text{sat}} \right]^{-1} \left[ \frac{4}{5^{3/2}} \cdot \xi(3) \cdot \rho \cdot \mu_0 \cdot M_s^2 \right] \]

(10)

where \( H \) is in A m\(^{-1} \), and \( \tau_y \) results in Pascals (Pa). From figure 5, \( \tau_{y \text{ sat}} \) values are used for fitting \( M_s \) and \( B_{\text{MRF}} \) using least squares method. The values of the constants of equation (10) are listed at table 3.

Constitutive model represented by equation (10) is applied and verified by comparison with experimental \( \tau_y - H \). Comparison plots are shown at figure 8.

6.1. Comparison with other constitutive models

The accuracy of the proposed constitutive model is evaluated by comparing it with the equations proposed by Dave and Ginder.

6.1.1. Dave equation. In his work MR fluid and devices in the real world, Carlson cites an empirical equation proposed by Dave to provide a practical and convenient description of virtually any MR fluid, which is:

\[ \tau_y = C \cdot 271700 \cdot \rho^{1.5239} \cdot \tan h \left( 6.33 \times 10^{-6} \cdot H \right), \]

(11)

where \( \rho \) is the volume fraction of iron particles, \( \tau_y \) is in Pa, \( H \) is the magnetic field intensity in A m\(^{-1} \), and the constant \( C \) equals 1.0, 1.16 or 0.95 depending on whether the carrier fluid is hydrocarbon oil, water or silicone oil. This equation generates a curve that describes the nonlinear behavior of \( \tau_y \) as function of \( H \). Due that analyzed MRFs in this work have a hydrocarbon oil \( C \) is 1. Comparison with constitutive model based on STE (assuming carrier fluid is oil thus \( C \) is 1) is shown at figures 9–11.

6.1.2. Ginder equations. Ginder proposes a series of equations to predict \( \tau_y \) and \( \tau_{y \text{ sat}} \) of MRF, such equations are obtained as an approach from Finite Element Analysis of interaction between magnetic particles in a carrier fluid.

Equation for saturation shear yield stress is equation (6), already applied in this work. For application, equation (12) describes the increase of \( \tau_y \), its limit is obtained by equation (6) and constants listed at table 3. Comparison with constitutive model based on STE is shown in figures 9–11.

\[ \tau_y = \sqrt{6} \cdot \rho \cdot \mu_0 \cdot M_s^{1/2} \cdot H^{3/2}. \]

(12)

6.1.3. Bingham model. Equation (10) can be integrated into Bingham model (Nishiyama et al 2002), this way shear stress can be determined for MRFs as function of viscosity \( \eta \) and shear rate \( \gamma \):

\[ \tau = \tau_y(H) + \eta \gamma. \]

(13)
140CG by STE model, Ginder and Dave equations.

Accuracy of the model is quantified through experiments an own mathematical expression already applied in other kind of materials and the stimulus applied, in this case a magnetic field. The equation associates the volume fraction of particles, magnetic field and the material that composes the particles. The model can be substituted in Bingham equation and applied to describe shear stress of MRF as function of shear rate and viscosity.

The model allows designing MRF by selecting the degree of sensitivity to magnetic field and the saturation yield stress. From the behavior observed, it can be considered that a good MRF should have a high saturation yield stress and low magnetic sensitivity, this way there is more range of applicability of the material in reconfigurable devices such as dampers or prosthetic legs.

From experimental liquid–solid state transition estimation is observed that MRF with larger amount of particles saturates at a lower magnetic field that those with less particles. It is considered that this occurs due that a larger amount of particles makes the material more sensitive to the magnetic field. An estimation of this can be done by assuming that as lower results the characteristic magnetic field of MRF the material is more sensitive to magnetism.

STE is used for describing state transition, even if it is not clearly sigmoidal, the equation results flexible and can get a good fitting with experimental data.

A constitutive model for yield stress of MRF is proposed consisting of an expression that relates composition of the material and the stimulus applied, in this case a magnetic field. The equation associates the volume fraction of particles, magnetic field and the material that composes the particles. The model can be improved by preparing an own model that relates the amount of particles with the saturation yield stress, this way the robustness of the model would be improved by not considering constants that depend of Ginder’s model (ξ(3)) or were adjusted specifically for Lord Corporation MRFs (α, β, χ).

Due to that this constitutive model achieved a good fitting with experimental data it is demonstrated that effect of magnetic field on rheology of MRF can be described based on a mathematical expression already applied in other kind of smart materials such as NiTi alloys (Cortes Ramirez et al 2012), electroactive polymers and SMP (Varela-Jimenez et al 2010; Varela-Jimenez 2011).

### 7. Discussion

A liquid–solid state transition in MRF was proposed having a sigmoidal behavior, considering that nucleation is started by the magnetic field and is applied and as it is increased the volume fraction of the solid state also becomes higher.

Experimental yield stress–magnetic field data for commercial MRF allowed to estimate the state transition, demonstrating that magnetic field has an immediate effect on the material by observing that even if the magnetic field is low it produces an increase on the yield stress. Then, it can be considered that solid state growth stage starts almost immediately with a very short nucleation stage, in contrast with the phase transformation of austenite in NiTi alloys or active state of shape memory polymers (SMP) on which nucleation occurs and a specific amount of heat must be applied to activate the growth of the transition (Varela-Jimenez et al 2010, Cortes Ramirez et al 2012).

From experimental liquid–solid state transition estimation is observed that MRF with larger amount of particles saturates at a lower magnetic field that those with less particles. It is considered that this occurs due that a larger amount of particles makes the material more sensitive to the magnetic field. An estimation of this can be done by assuming that as lower results the characteristic magnetic field of MRF the material is more sensitive to magnetism.

### 8. Conclusion

A constitutive model for shear yield stress of MRF is proposed consisting of an expression that relates composition of the material and the stimulus applied, in this case a magnetic field. The equation associates the volume fraction of particles, magnetic field and the material that composes the particles...
represented by a series of constants mathematically fitted. The final model has better performance than Dave and Ginder models for all the MRF, reducing the MSE at least 3.9 times for Ginder model and 1.9 for Davis. The structure of the proposed model allows that it can be simply introduced into a spreadsheet or finite element analysis software for preliminary design of MRF devices.

The model is successfully fitted for the MRF-122EG, MRF-132DG and MRF-140CG (LORD Corporation 2010) to describe shear stress as function of magnetic field, material and amount of particles. The constitutive model is based on equation which has been applied to describe behavior of other materials such as SMP or stainless steels, demonstrating that smart materials can be modeled by a common equation based on a state transition.

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