JPEDAV (2008) 29:305–311
DOI: 10.1007/s11669-008-9318-8
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A Constitutive Equation for Magnetorheological Fluid Characterization

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(Submitted July 2, 2007)

A microstructural model of the motion of particle pairs in MR fluids is proposed that accounts for both hydrodynamic and magnetic field forces. A fluid constitutive equation is derived, from the model that allows the prediction of velocity and particle structure fields. The analysis is similar to that of bead-spring models of polymeric liquids with replacement of the elastic connector force by a magnetic force. Results for simple shear flow are presented for the case when the two particles remain in close contact so they are hydrodynamically equivalent to an ellipsoid with an aspect ratio of two and only the component of the magnetic force normal to the connecting vector between the centers of the two particles affects motion. The model predicts oscillatory motion of the particle pairs at low magnetic fields. The fluid reaches a steady state at high magnetic fields. The time required to reach the steady state for a given shear rate reduces significantly as the field increases.

Keywords constitutive equation, magnetorheological fluid, particle pair

1. Introduction

Magnetorheological (MR) fluids are two phase suspensions that can reversibly change their state, from a liquid like to a solid/gel like state, upon the application of an external magnetic field. The liquid phase is commonly aqueous or organic,[1] while the solid phase consists of micron size magnetic particles.[2] The change in state, that is accompanied by an increase in viscosity, is due to the alignment of the magnetic particles in chain or column like structures, between the two magnetic poles, upon the application of the magnetic field. This characteristic of MR fluids makes them very suitable for damping related automotive and aerospace applications. Among these applications, the ones receiving most attention are shock absorbers,[3] dampers,[4] brakes[5] and clutches[6] that have already been implemented in luxury automobiles.

While several MR fluid based applications have been developed, our understanding of fluid behavior is incomplete. The literature contains many reports of efforts to experimentally and theoretically characterize MR fluids. However, almost all of these efforts (theoretical approaches, in particular) are limited to dilute suspensions and/or static and quasi-static conditions and therefore not suitable for commercial applications that typically require semi-dilute to concentrated suspensions under dynamic conditions.

Theoretical approaches were initially developed for electrorheological (ER) fluids which are the electric field counterpart of MR fluids. Among the first proposed models was the one by Cutillas et al.[7] Their model developed for ER fluids, predicted particle chain formation, phase separation and a regular pattern of lamellae formation under oscillatory flow, in agreement with the experimental observations reported by Filisko and Henley.[8] Later, Gross et al.[9] assumed that particles aggregate in chains and analyzed the interaction between two finite chains by perturbation theory and numerical analysis for dilute suspensions finding that the range of attraction does not explain chain aggregation. Volkova et al.[10] used a chain model to analyze the experimental shear stress versus shear rate curves of MR fluids. They considered a chain of particles to be stable when the hydrodynamic force at the center of the chain was lower than the attractive force between two adjacent particles. The model is not applicable at high shear rates, when aggregates disintegrate. Pfeil et al.[11] proposed a conservation equation for the evaluation of particle volume fraction when a field is applied to a flowing suspension. For dilute suspensions and quasi-static conditions the equation can predict either the structure evolution in quiescent suspensions (chain formation) or structure evolution in sheared suspensions for low shear rates (lamellae formation). Bechtel et al.[12] developed a set of constitutive equations based on the dumbbell model, assuming that the hydrodynamic and the magnetic forces act only on beads, and that no short or long range hydrodynamic interactions occur. For simple shear flow their constitutive equations reduce to a form similar to the Bingham model.

Several more elaborate models were developed for both electrorheological and magnetorheological fluids by

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Klingenberg et al.,[13] Hass,[14] Mohebi et al.,[15] Volkova et al.,[16] Ly et al.,[17] Sim et al.[18] and Climent et al.[19] While some of these models account for multibody and long range hydrodynamic interactions and may provide more accurate results than the ones mentioned previously, their high computational requirements make them impractical for engineering applications. The models currently used for design purposes are the phenomenological Bingham and Herschel-Bulkley models. While for simple flows these models can predict fluid behavior with sufficient accuracy, for complex applications (e.g. haptic devices) more advanced and accurate models are required.

The microstructural model described in this paper accounts for both the effects of hydrodynamic and magnetic field interactions on velocity and structure in MR fluids and can characterize the fluid rheology for a broad range of particle concentrations in complex flow geometries. A constitutive equation for MR fluids is derived, based on an analysis of the motion of pairs of magnetic particles in the suspending fluid in the presence of a magnetic field. The analysis is similar to that of bead-spring models of polymeric liquids with replacement of the elastic connector force by a magnetic force.

2. Constitutive Equation Development

2.1 The Equations of Motion

The particle pair model as applied to MR fluids consists of two magnetizable particles with the vector connecting the centers of the particles represented by $\bar{Q}$, as shown in Fig. 1. The connecting vector describes the overall orientation and the internal configuration of the two particles. The instantaneous location in space of the center of mass of the two particles relative to the origin of the coordinate system is given by the vector $\bar{R}$.

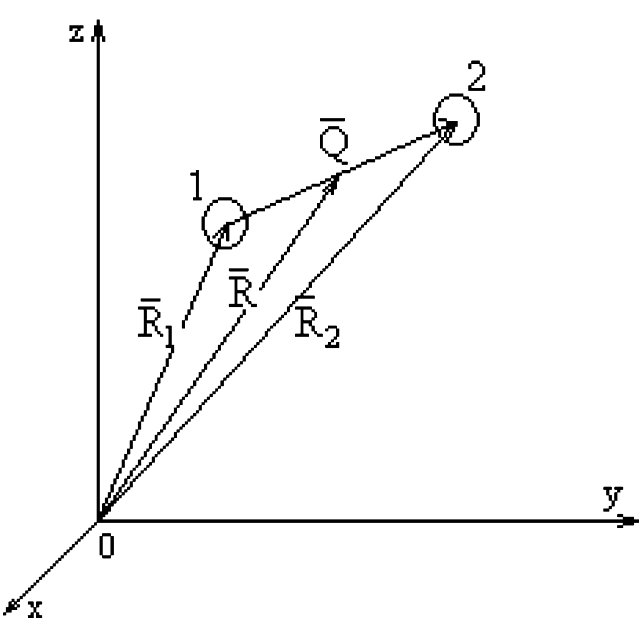

Fig. 1 The geometry of the particle pair model

The kinetic theory for the model requires (a) the establishment of the evolution equation for the particle pair orientation and (b) the development of an expression for the stress tensor.

The evolution equation is obtained by combining the equations of motion of the particles with an equation of continuity describing conservation of system points in configuration space.[20]

In writing the equation of motion for each particle the inertial terms containing the particle masses have been neglected. Each particle was assumed to experience a hydrodynamic drag force (Stokes drag) as it moves through the solution and an external magnetic force. The drag force is given by:

$$F^h = \frac{1}{2}\zeta\left(\dot{\bar{Q}} - \omega\cdot\bar{Q} - \lambda(D - D:\bar{Q}\bar{Q}I)\cdot\bar{Q}\right), \qquad \text{(Eq 1)}$$

where $2\omega = (\nabla v)^T - \nabla v$ and $2D = (\nabla v)^T + \nabla v$ are the vorticity tensor and the rate of deformation tensor, respectively. In the absence of magnetic forces, Eq 1 predicts the particle pair will rotate with the fluid but does not separate; this is identical to the motion of a single ellipsoidal particle in a suspension.[21] The motion is a function of particle aspect ratio through the material constant λ as discussed in the literature. We approximate the friction coefficient ζ with Stokes law as $\zeta = 6\pi\eta_s a$, where η_s represents the solvent viscosity. The difference between the two terms on the right hand side represents the non-affine motion of the particles. If the particles move affinely with the fluid, the difference is zero, and no hydrodynamic force is generated. If the particles move non-affinely, the difference is not zero, and it is proportional to the hydrodynamic force exerted on the particles.

The external magnetic force

$$\bar{F}^m_{ij} = -\frac{\partial U_{m,ij}}{\partial Q_i} = \frac{4\mu_0\pi a^6\chi^2}{3}\left[\frac{H^2}{Q^5}\bar{Q} - \frac{5(\bar{H}\cdot\bar{Q})^2}{Q^7}\bar{Q} + \frac{2(\bar{H}\cdot\bar{Q})}{Q^5}\bar{H}\right] \qquad \text{(Eq 2)}$$

has been derived from the interaction potential between two particles in the point-dipole approximation,[22] i.e.

$$U_{m,ij} = \frac{1}{4\pi\mu_0}\frac{\bar{m}_i\cdot\bar{m}_j - 3(\hat{Q}\cdot\bar{m}_i)(\hat{Q}\cdot\bar{m}_j)}{Q^3}, \qquad \text{(Eq 3)}$$

where $\mu_0 = 4\pi\times10^{-7}$ Tm/A is the permeability of free space and $\hat{Q} \equiv \bar{Q}/|\bar{Q}|$ is a unit vector. The magnetic dipole acquired by the particles in a field $\bar{H}$ (A/m) is $\bar{m}_i = \frac{4}{3}\pi a^3\mu_0\chi\bar{H}$ where a and χ are the particle radius and magnetic susceptibility. In order for the model to account for both far from contact and near contact conditions, the magnetic force has been modified using the Heaviside step function (the unit step function) as follows

$$\bar{F}^m_{ij} = -\frac{\partial U_{m,ij}}{\partial Q_i} = \frac{4\mu_0 \pi a^6 \chi^2}{3}\left[\hbar \frac{H^2}{Q^5}\bar{Q} - \hbar \frac{5(\bar{H}\cdot\bar{Q})^2}{Q^7}\bar{Q} + \frac{2(\bar{H}\cdot\bar{Q})}{Q^5}\bar{H} - (1-\hbar)\frac{2(\bar{H}\cdot\bar{Q})^2}{Q^7}\bar{Q}\right] \quad \text{(Eq 4)}$$

where the Heaviside function $\hbar$ is defined as

$$\hbar\left(\mathrm{tr}\langle\bar{Q}\bar{Q}\rangle - 4a^2\right) = \begin{cases} =1 & \text{for } \mathrm{tr}\langle\bar{Q}\bar{Q}\rangle - 4a^2 > 0 \\ =0 & \text{for } \mathrm{tr}\langle\bar{Q}\bar{Q}\rangle - 4a^2 \le 0 \end{cases}, \quad \text{(Eq 5)}$$

with $\mathrm{tr}\langle QQ\rangle = Q_1Q_1 + Q_2Q_2 + Q_3Q_3$. As introduced in Eq 4 and 5, the Heaviside function prevents particle overlap due to the attractive magnetic force by removing that component of the magnetic force acting in the $\hat{Q}$ direction when the particles are in contact; this is equivalent to introducing an opposing contact force.

The force balance written for the particle pair provides an expression for the change in time of the connecting vector, $\dot{\bar{Q}}$. Substituting the expression for $\dot{\bar{Q}}$ in the equation of change for the distribution function $\psi(\bar{Q}, t)$ (the Smoluchowski equation)

$$\frac{\partial\psi}{\partial t} = -\left(\frac{\partial}{\partial\bar{Q}}\cdot\dot{\bar{Q}}\psi\right), \quad \text{(Eq 6)}$$

multiplying Eq 6 by $\bar{Q}\bar{Q}$ and integrating it over the entire orientation space leads to the time evolution equation for the distribution function

$$\frac{\partial}{\partial t}\langle\bar{Q}\bar{Q}\rangle = (\omega + \lambda\cdot D)\langle\bar{Q}\bar{Q}\rangle - \langle\bar{Q}\bar{Q}\rangle(\omega - \lambda\cdot D) - 2\lambda D : \langle\bar{Q}\bar{Q}\rangle\langle\bar{Q}\bar{Q}\rangle + \frac{4}{\zeta}\frac{\Delta H^2}{Q_0^5}\hbar\langle\bar{Q}\bar{Q}\rangle - \frac{4}{\zeta}\frac{\Delta}{Q_0^7}(2a)^2(3\hbar+2)\bar{H}\bar{H} : \langle\bar{Q}\bar{Q}\rangle\langle\bar{Q}\bar{Q}\rangle + \frac{4}{\zeta}\frac{\Delta}{Q_0^5}\left(\langle\bar{Q}\bar{Q}\rangle\cdot\bar{H}\bar{H} + \bar{H}\bar{H}\cdot\langle\bar{Q}\bar{Q}\rangle\right). \quad \text{(Eq 7)}$$

In Eq 7, $\langle\bar{Q}\bar{Q}\rangle$ is the second order moment of the distribution function defined as[20]

$$\langle\bar{Q}\bar{Q}\rangle = \int \bar{Q}\bar{Q}\psi d\bar{Q} \quad \text{(Eq 8)}$$

$\Delta = \frac{4\mu_0\pi a^6\chi^2}{3}$ and $Q_0 = 2a$. Additionally, note that the fourth order tensor, $\langle\bar{Q}\bar{Q}\bar{Q}\bar{Q}\rangle$, has been approximated as the product of two second order tensors, $\langle\bar{Q}\bar{Q}\rangle\langle\bar{Q}\bar{Q}\rangle$, as suggested in the literature.

The solution of the evolution Eq 7 is required to calculate the particle contribution to the effective fluid stress due to inter-particle magnetic forces from: $\tau^{p,m} = \frac{1}{2}n\left\langle\bar{Q}\bar{F}^m_{ij}\right\rangle$ where $n = 3\phi/4\pi a^3$ is the particle number density and $\phi = 0.32$ is the particle volume fraction. Substituting the magnetic force in this equation results in

$$\tau^{p,m} = \phi\cdot\left[\frac{\alpha\hbar H^2}{Q_0^5}\langle\bar{Q}\bar{Q}\rangle - \frac{\alpha(3\hbar+2)}{Q_0^7}(2a)^2 HH : \langle\bar{Q}\bar{Q}\rangle\langle\bar{Q}\bar{Q}\rangle + \frac{2\alpha}{Q_0^5}HH\cdot\langle\bar{Q}\bar{Q}\rangle\right] \quad \text{(Eq 9)}$$

where $\alpha = \mu_0\ \chi^2 a^5/2$. The particles also give rise to a hydrodynamic drag contribution to the stress given by,

$$\tau^{p,h} = \phi\cdot\left[\mu_1\langle\bar{Q}\bar{Q}\rangle + \mu_2 D : \langle\bar{Q}\bar{Q}\rangle\langle\bar{Q}\bar{Q}\rangle + 2\mu_3\left(D\cdot\langle\bar{Q}\bar{Q}\rangle + \langle\bar{Q}\bar{Q}\rangle\cdot D\right)\right] \quad \text{(Eq 10)}$$

where μ_1, μ_2, and μ_3 are material constants.[22]

2.2 The Stress Tensor

The total stress τ for the two phase MR fluid is calculated as the summation of the solvent-contributed stress and the particle-contributed stresses,

$$\tau = \tau^s + \tau^{p,h} + \tau^{p,m} \quad \text{(Eq 11)}$$

where $\tau^s = 2\eta_s D$ and η_s is the solvent viscosity.

Substituting Eq 9 and 10 in Eq 11 the total stress tensor becomes

$$\tau_{ij} = 2\eta_s D + \phi\cdot\left[\mu_1\langle\bar{Q}\bar{Q}\rangle + \mu_2 D : \langle\bar{Q}\bar{Q}\rangle\langle\bar{Q}\bar{Q}\rangle + 2\mu_3\left(D\cdot\langle\bar{Q}\bar{Q}\rangle + \langle\bar{Q}\bar{Q}\rangle\cdot D\right) + \frac{\alpha\hbar H^2}{Q_0^5}\langle\bar{Q}\bar{Q}\rangle - \frac{\alpha(3\hbar+2)}{Q_0^7}(2a)^2 HH : \langle\bar{Q}\bar{Q}\rangle\langle\bar{Q}\bar{Q}\rangle + \frac{2\alpha}{Q_0^5}HH\cdot\langle\bar{Q}\bar{Q}\rangle\right] \quad \text{(Eq 12)}$$

Equation 12 can be applied to predict the stress tensor for magnetorheological fluids flowing through complex geometries. Stress tensor predictions for simple shear flow of spherical particles in contact are presented in the following paragraph. The magnetic field H was assumed to act perpendicular to the direction of the flow. Therefore, the Heaviside function is zero and the material coefficients are[23] $\mu_1 = 0$, $\mu_2 = 0.3$ and $\mu_3 = 1$. Additionally, Eq 12 simplifies to:

$$\tau_{ij} = 2\eta_s D + \phi\cdot\left[\mu_2 D : \langle\bar{Q}\bar{Q}\rangle\langle\bar{Q}\bar{Q}\rangle + 2\mu_3\left(D\cdot\langle\bar{Q}\bar{Q}\rangle + \langle\bar{Q}\bar{Q}\rangle\cdot D\right) - \frac{2\alpha}{Q_0^7}(2a)^2 HH : \langle\bar{Q}\bar{Q}\rangle\langle\bar{Q}\bar{Q}\rangle + \frac{2\alpha}{Q_0^5}HH\cdot\langle\bar{Q}\bar{Q}\rangle\right] \quad \text{(Eq 13)}$$

For a simple shear flow with $v = \dot{\gamma}x_2$ the equations defining the total stress tensor components are:

$$\begin{aligned}
\tau_{11} &= \phi\cdot\left[\beta_1\langle Q_1Q_1\rangle - \beta_2\langle Q_1Q_1\rangle\langle Q_2Q_2\rangle + 2\mu_2\dot{\gamma}\langle Q_1Q_2\rangle\langle Q_1Q_1\rangle + 2\mu_3\dot{\gamma}\langle Q_1Q_2\rangle\right] \\
\tau_{12} &= \eta_s\dot{\gamma} + \phi\cdot\left[\beta_1\langle Q_1Q_2\rangle - \beta_2\langle Q_1Q_2\rangle\langle Q_2Q_2\rangle + 2\mu_2\dot{\gamma}\langle Q_1Q_2\rangle\langle Q_1Q_2\rangle + \mu_3\dot{\gamma}(\langle Q_1Q_1\rangle + \langle Q_2Q_2\rangle)\right] \\
\tau_{22} &= \phi\cdot\left[\beta_1\langle Q_2Q_2\rangle - \beta_2\langle Q_2Q_2\rangle\langle Q_2Q_2\rangle + 2\mu_2\dot{\gamma}\langle Q_1Q_2\rangle\langle Q_2Q_2\rangle + 2\mu_3\dot{\gamma}\langle Q_1Q_2\rangle\right]
\end{aligned} \quad \text{(Eq 14)}$$

where $\beta_1 = 6a^5\mu_0\chi^2H^2/Q_0^5$ and $\beta_2 = 40a^7\mu_0\chi^2H^2/Q_0^7$. Note that in the following calculations, it is assumed particle pairs always remain in contact and behave hydrodynamically like a single ellipsoidal particle of aspect ratio two.

3. Results

Model predictions of particle orientation and shear stress tensor for different shear rates and magnetic fields are presented here. Results for the imposition of a magnetic field in the absence of flow are illustrated in Fig. 2. Figure 2 shows that when the field is applied the particles will rotate until they reach an equilibrium position in which they are aligned in the field direction. Depending on the applied field strength, the time required for the equilibrium to be reached varies, being smaller at higher magnetic fields.

Under steady shearing conditions, the model predicts oscillatory behavior at low magnetic fields, as shown in Fig. 3. The particle pairs would tend to align with the magnetic field, but the hydrodynamic force arising from fluid shearing is stronger than the magnetic force and particle pairs rotate with the fluid. For sufficiently strong shear flows, the particles will continue to rotate around the direction of the flow as indicated in Fig. 3a and c.

Both, the particle orientation and the 11 and 22 components of the stress show highly oscillatory behavior as opposed to τ_{12} (Fig. 3b and d). This is due to the fact that the primary contribution to the shear stress is that arising from the suspending fluid. At moderate magnetic fields the oscillatory behavior ceases and the fluid reaches a steady state (Fig. 4) in a very short time compared with the period of oscillation recorded at low magnetic fields. If both the shear rate and the field are large (Fig. 4c and d) the steady state establishes soon after the flow begins. The particle-pair equilibrium position is reached somewhere between the direction of the flow and the direction of the field (Fig. 4a and c).

A cumulative graph showing the change of τ_{12} with the magnetic field at a given shear rate is provided in Fig. 5. It can be observed that in the absence of magnetic field the model predicts a shear stress of the suspension larger than that of the fluid itself (denoted as τ^s in Fig. 5). Also, as expected, the shear stress increases as the field increases and after passing through an oscillatory regime reaches a steady state. The magnitude of the field required to achiever the steady state is highly dependent on the shear rate, as indicated in Fig. 6. Identification of the boundary between the oscillatory and steady state regimes, as attempted here, is critical to the design of a MR fluid based device.

Calculations for a broad range of shear rates and field strengths indicate that the viscosity will increase with an

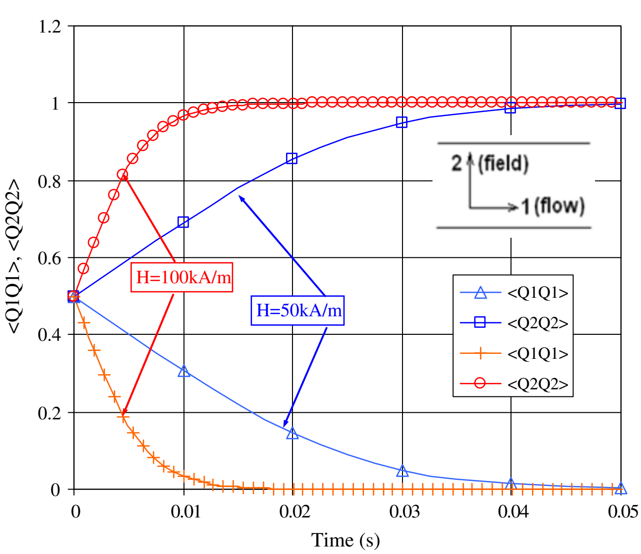

Fig. 2 Particle-pair response to the magnetic field in the absence of flow

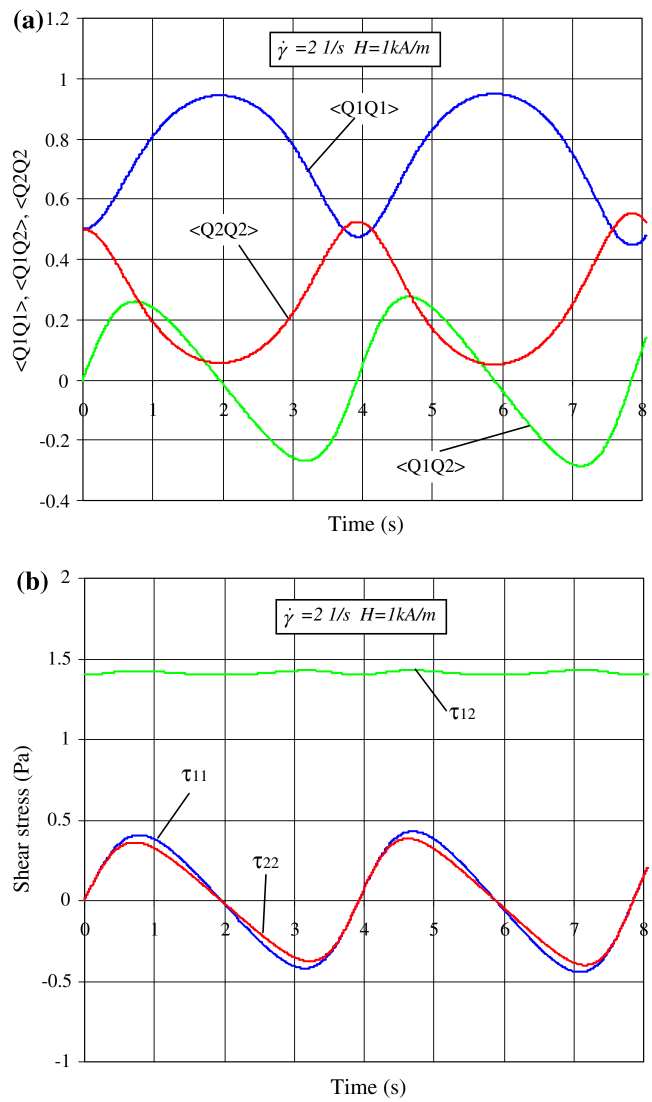

Fig. 3 (a) Particle orientation for magnetic field of 1 kA/m and shear rates of 21/s; (b) Fluid stress for magnetic field of 1 kA/m and shear rates of 21/s

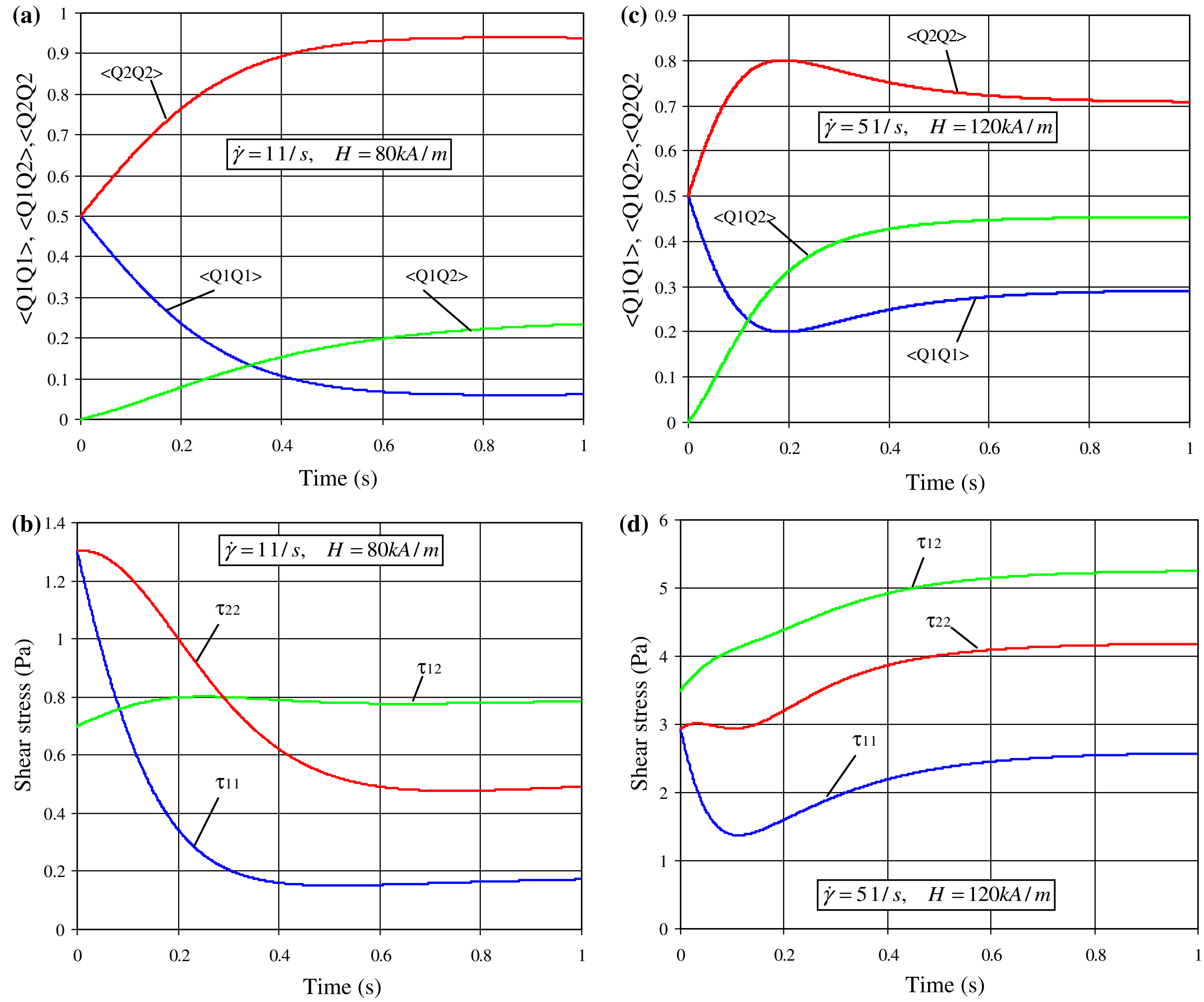

Fig. 4 (a) Particles orientation for magnetic field of 80 kA/m and shear rates of 11/s; (b) Fluid stress for magnetic field of 80 kA/m and shear rates of 11/s; (c) Particle orientation for magnetic field of 120 kA/m and shear rates of 51/s; (d) Fluid stress for magnetic field of 120 kA/m and shear rates of 51/s

increase in the applied field. However, the viscosity reaches a maximum after which it starts to decrease with further increases in field strength. This behavior is due mainly to the particle magnetic contribution to the stress. For a given shear rate, this stress component increases with the magnetic field increase up to a maximum value. Once the maximum has been reached, further increases in magnetic field lead to a decrease in the stress and eventually the stress reaches a lower plateau value as shown in Fig. 7. In all cases, the particle pairs reach a steady configuration at the point of maximum viscosity enhancement—for lower field strengths pairs align more closely with the flow direction and for higher field strengths pairs align more closely with the magnetic field. The particle contribution to the shear stress passes through a maximum since the product $\langle Q_1Q_2\rangle$ $\langle Q_2Q_2\rangle$ in the stress passes through a maximum. This unexpected dependence on orientation arises from the neglect of interactions between particle pairs (only interactions within a pair, not between pairs, are considered here) as they align in the field direction. Such interactions would lead to an increase in the effective aspect ratio of the particles.

The ratio of the effective viscosity to the carrier fluid viscosity is plotted in Fig. 8 as a function of field strength. Figure 8 indicates that the effective fluid viscosity will not increase more than approximately 3.2 times the carrier fluid viscosity. Clearly the field strength required to attain the maximum viscosity enhancement depends on the shear rate. The relationship between shear rate and field strength at the point of maximum enhancement is nearly linear for shear rates greater than 0.1 1/s as illustrated in Fig. 9.

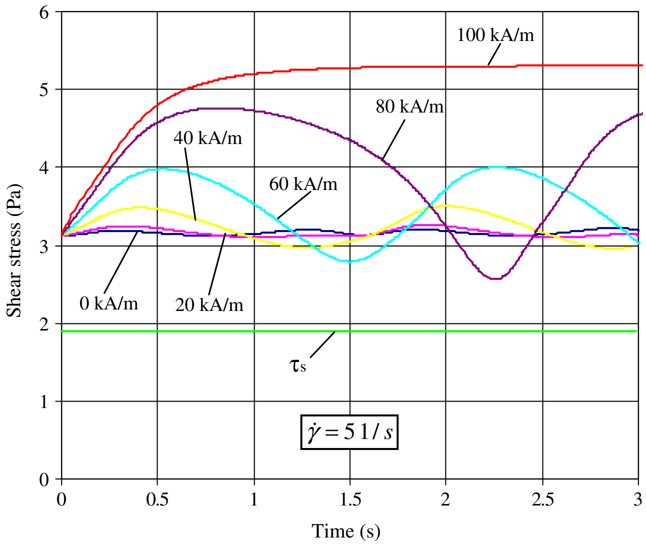

Fig. 5 Fluid stress variation with magnetic field at a shear rate of 51/s

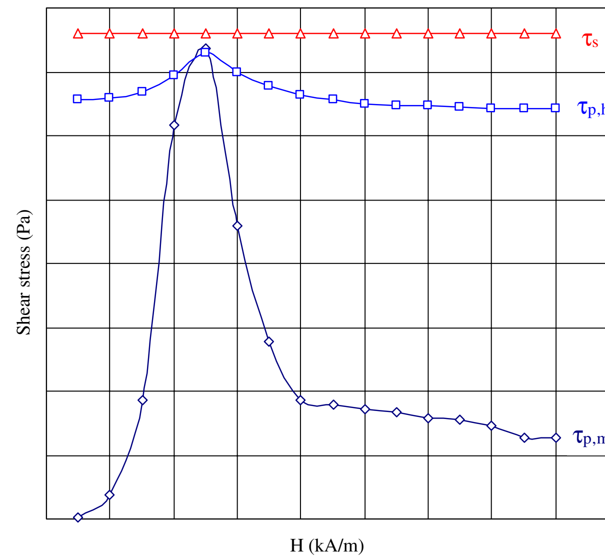

Fig. 7 Field dependence of the three components of the stress for a constant shear rate

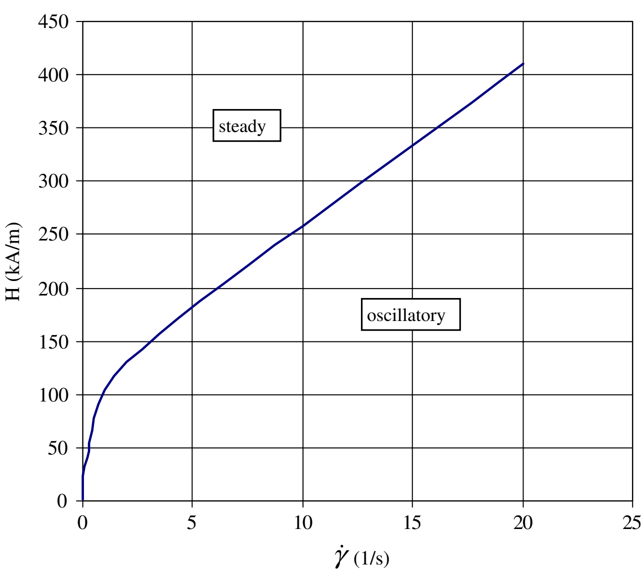

Fig. 6 The oscillatory and steady state regions for the simulated MR fluid

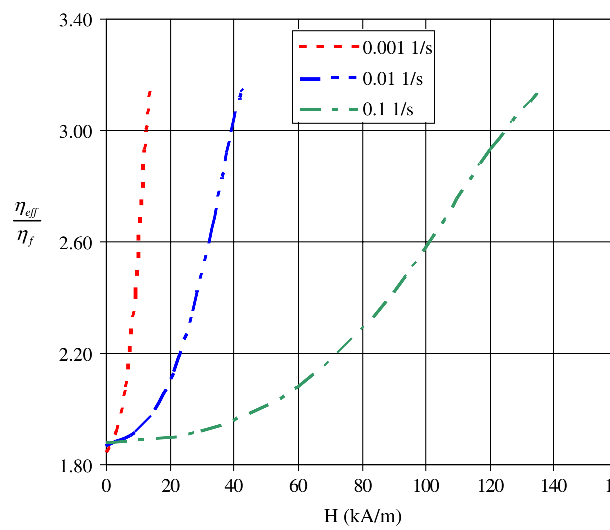

Fig. 8 Variation of equivalent fluid viscosity with magnetic field

4. Conclusions

A stress constitutive equation for magnetorheological fluids has been developed. The equations monitor the evolution of the configuration of particle pairs in the presence of a complex flow and of a magnetic field. The equations account for hydrodynamic drag and magnetic forces. The results obtained for a fluid in simple shear flow with an external magnetic field applied indicate a strong dependence on the applied magnetic field. For low magnetic fields particle pairs oscillate between alignment in the flow direction and in the magnetic field direction. For high magnetic fields particle pairs reach a steady alignment at an intermediate angle between the flow and magnetic field directions. This alignment moves toward the field direction as the field strength increases. The boundary between the oscillatory and steady state regimes as a function of the applied magnetic field and shear rate is identified for design purposes. For the simulated fluid, one can increase the shear viscosity by a factor of ~3.2 by applying a magnetic field; the required field is highly dependent on the shear rate. This increase is highly dependent on the magnetic susceptibility through the stress coefficients (β). These coefficients are proportional to the square of the susceptibility. Unfortunately

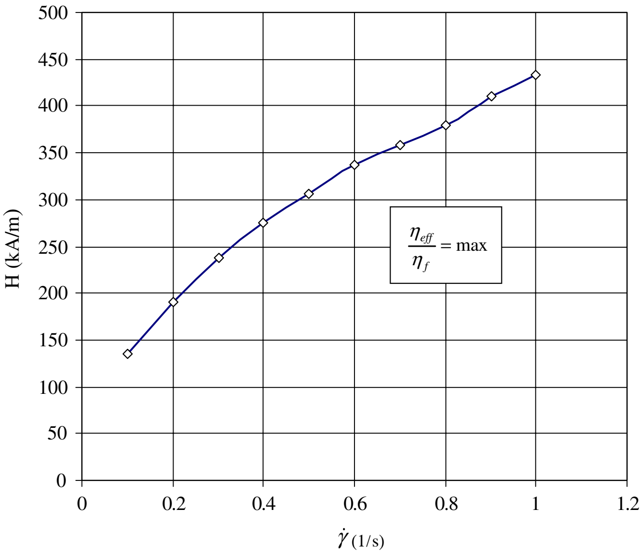

Fig. 9 The maximum equivalent fluid viscosity as a function of magnetic field and shear rate

this is one of the more difficult physical properties to obtain for commercially available fluids.

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