Magnetorheological fluid (MRF) in oscillatory shear and parameterization with regard to MR device properties

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Abstract. A mechanical analogue, consisting of a Bingham element (friction element and damper arranged in parallel) and elastic spring in series, is used to capture the measured amplitude dependences of the storage and loss moduli of an MRF in oscillatory shear. Model predictions for the time-dependent shear stress and resulting moduli at agitation frequency for various amplitudes are discussed. The application to plate-plate geometry, imposing a linear radial shear strain profile, is addressed.

1. Introduction
We present measurements of the storage and loss moduli of a magnetorheological fluid (MRF) versus shear amplitude in a plate-plate magnetorheometer and demonstrate the applicability of a pertinent mechanical analogue to capture the main features. The MRF is a concentrated suspension of spherical micron-sized carbonyl iron particles (CIP) in low viscosity Newtonian base oil. Submitted to a magnetic field, the ferromagnetic particles get magnetized and magnetic particle-particle interaction causes the CIP to arrange in strings parallel to the external field. This structure formation causes an increase of the MRF’s yield stress \( \tau_\gamma \), depending on the magnetic flux density \( B \). The controllability of the flow properties by an external magnetic field is used in MR devices like dashpot dampers (for shock absorbers) and clutches. Here, the performance is mainly controlled by steady shear flow properties of the MRF, approximated by a Bingham model,

\[
\tau = \tau_\gamma(B) + \eta_B(B)\dot{\gamma},
\]

where \( \tau \) is the shear stress, \( \dot{\gamma} \) the shear rate, and \( \eta_B \) the Bingham viscosity. The parameterization is useful to link MRF flow parameters to MR device performance. MR bushings, MR mounts as well as dashpot dampers for controllable damping of oscillatory motions, require additional information about the MRF response in oscillatory shear at various amplitudes. While measurements with imposed sinusoidal shear strain \( \dot{\gamma} = \dot{\gamma}_\text{str} \sin \omega t \) (\( \dot{\gamma}_\text{str} \) shear strain amplitude, \( \omega \) angular frequency) are easily performed by commercial magnetorheometers, a complex MRF response is observed: The resulting oscillatory shear stress strongly depends on the applied shear amplitude, deviates from a sinusoidal wave, and yields a predominantly elastic response at very small shear amplitudes.

2. Experimental
Measurements were performed on a rotational rheometer MRC501 equipped with a MRD180/1T magnetocell (Anton Paar) with modifications to allow online flux density measurements using a Hall probe [1]. The geometry is plate-plate with radius \( R = 10 \) mm and gap \( h = 0.3 \) mm, the flux density...
vector being perpendicular to the plane of shear. For sinusoidal shear, the rheometer software provides a storage and loss modulus $G'(\omega)$ and $G''(\omega)$, respectively, by decomposing the oscillatory shear stress wave into in-phase and off-phase components:

$$\tau = \dot{\gamma} \left[ G'(\omega) \sin \omega t + G''(\omega) \cos \omega t \right]$$  \hspace{1cm} (2)

While Eq. (2) gives an adequate representation of the actual shear stress in the linear viscoelastic regime at small amplitudes, the response becomes nonlinear at high amplitudes and harmonics occur:

$$\tau = \dot{\gamma} \left[ G'(\omega, \dot{\gamma}) \sin \omega t + G''(\omega, \dot{\gamma}) \cos \omega t \right]$$

$$+ \dot{\gamma} \sum_{k=1}^{\infty} \left[ a_k(\omega, \dot{\gamma}) \sin[(2k+1)\omega t] + b_k(\omega, \dot{\gamma}) \cos[(2k+1)\omega t] \right]$$  \hspace{1cm} (3)

The rheometer software, however, filters out the harmonics at $(2k+1)\omega$ and provides the amplitude-dependent moduli $G'(\omega, \dot{\gamma})$, $G''(\omega, \dot{\gamma})$ solely for the agitation frequency $\omega$.

**Figure 1** shows measured moduli versus shear amplitude. A linear viscoelastic behavior - both moduli being amplitude-independent - is found for amplitudes $\dot{\gamma} \leq 0.0005$ (Note: the off-state storage modulus is about four powers of ten smaller). In the small amplitude regime, the MRF responds predominantly elastic, i.e. $G \gg G''$. With increasing amplitude, the loss modulus first increases while the storage modulus remains fairly constant. Finally, both moduli exhibit a strong decrease and $G'' \gg G$ indicates a predominantly viscous response. If plotted versus shear stress amplitude $\dot{\gamma}$ (not shown by a figure), both moduli exhibit a drastic drop at the value of the steady shear yield stress $\tau_y(B)$. **Figure 2** shows the unfiltered shear stress signal versus time for $\dot{\gamma} = 0.01$ (shear stress wave close to sinusoidal), and for $\dot{\gamma} = 1$ (saw tooth shape indicating the generation of many harmonics).

![Figure 1](image1.png)  \hspace{1cm} **Figure 1.** Storage modulus $G'$ and loss modulus $G''$ versus shear amplitude output of the magnetorheometer at $B = 0.39$ T, $\omega = 1.9$ rad/s, and 25 °C.

![Figure 2](image2.png)  \hspace{1cm} **Figure 2.** Unfiltered shear stress signals from the magnetorheometer for two different shear amplitude levels (parameters as in Figure 1).

3. Simple model

**Figure 3** schematically depicts the structure of the MRF under the action of a magnetic field perpendicular to the plane of shear. At rest, the particles form strings and create a static normal stress \( \tau \). Submitted to a small shear strain, the strings experience an essentially affine deformation with the macroscopic shear, i.e. a slight tilt without string rupture. The magnetic particle-particle interaction in the tilted position causes a torque which tries to reorient the strings parallel to the flux density vector, thus causing an elastic reverse force on the moving plate. This is the origin of the elastic response at very small strains. If the acting shear amplitude exceeds the yield stress, however, the strings start to rupture and achieve in part a less tilted arrangement, which means a loss of elastic memory and increased dissipation due to particle movements relative to the base oil. The MRF behavior in large amplitude oscillatory shear thus shifts from predominantly elastic to mainly viscous.

A simple mechanical analogue to capture such a behavior is the model shown in **Figure 4**. It consists of a Bingham element, i.e. a damper of viscosity $\eta_\theta$ parallel to a friction element with yield stress $\tau_y$, arranged in series to an elastic spring of modulus $G_0$ [3]. The Bingham element only
contributes to the deformation $\gamma$ as long as the shear stress $\tau$, acting on both the spring and the Bingham element, exceeds the yield stress. In this case, spring and damper define a relaxation time $\lambda = \eta_B / G_0$.

![Figure 3](image1.png)  
**Figure 3.** Structural model with CIP strings parallel to external field at rest (left), slightly tilted but intact strings at small shear (middle), and tilted but ruptured strings for large shear (right).

As soon as the absolute value of the acting stress $\tau$ becomes smaller than $\tau_\gamma$, the friction element turns rigid again and the deformation of the Bingham element $\gamma_B$, reached up to that instant, remains constant. The following set of equations enables to calculate the time-dependence of the shear stress:

$$\tau = \begin{cases} G_0 (\gamma - \gamma_B) & \text{for } |\gamma| \leq \gamma_\gamma \\ \tau_\gamma \text{sign}(\gamma) + \eta_B \dot{\gamma} - \lambda \dot{\gamma} & \text{for } |\gamma| > \tau_\gamma \end{cases} \quad (4)$$

The differential equation in the second line of Eq. (4) yields for imposed sinusoidal shear

$$\tau(t) = C \exp(-t / \lambda) + \tau_\gamma \text{sign}(\gamma) + \dot{\gamma} \left[ g'' \sin \omega t + g' \cos \omega t \right], \quad (5)$$

where the integration constant $C$ is determined by the value of the shear stress at the change of mode, while the abbreviations $g''$ and $g'$ stand for $g'' = G_0 \omega \lambda / (1 + \omega^2 \lambda^2)$ and $g' = \omega \lambda g''$, respectively.

### 4. Model predictions

**Figure 5** shows examples for the shear stress versus time for $\omega = \pi \text{ rad/s}$, compared to a purely elastic response $\tau_\varepsilon = G_0 \varepsilon$ for $\gamma_B = 0$. In general, the solution to Eq. (4) is composed by subsequent regimes: Once the elastic shear stress exceeds $\tau_\gamma$, the shear stress follows Eq. (5) until $\tau = \tau_\gamma$ is met again. Here, the Bingham element has reached a shear strain $\gamma_B^0$. The latter remains constant in the subsequent elastic phase given by the first line of Eq. (4). From this regime on, the shear stress is periodic in time. Due to the finite $\gamma_B^0$, however, the elastic response is phase shifted compared to $\tau_\varepsilon$.

![Figure 5](image2.png)  
**Figure 5.** Shear stress versus time for various $\tau_\gamma$ values ($G_0 = 3 \times 10^4 \text{ Pa}$, $\eta_B = 10^3 \text{ Pas}$, $\eta_p = 0$).

![Figure 6](image3.png)  
**Figure 6.** Storage and loss modulus versus shear stress amplitude ($\tau_\gamma = 2 \times 10^4 \text{ Pa}$, $\eta_p = 2 \times 10^4 \text{ Pas}$).
the phase shift increasing with the ratio $\eta G_{\phi} / \tau_{\phi}$. When the shear stress becomes smaller than $-\tau_{\phi}$, the friction element opens again and finally reaches a value $-\gamma_{\phi}^0$, which enters in the subsequent elastic response. If an optional damper of viscosity $\eta_{\phi}$ (dotted lines in Figure 4) is arranged in parallel, the response of the Bingham element with spring remains unchanged, but a stress component proportional to the shear rate and $\eta_{\phi}$ is added to the total shear stress.

From the shear stress versus time, the moduli $G'$ and $G''$ for the agitation frequency $\omega$ (base mode) follow as (Note: Integration limits are chosen such that only the periodic shear stress signal is used)

$$G'(\gamma, \omega) = \frac{1}{\gamma} \int_{-\pi/\omega}^{\pi/\omega} \tau(\gamma, \omega, t) \sin(\omega t) dt$$
$$G''(\gamma, \omega) = \frac{1}{\gamma} \int_{-\pi/\omega}^{\pi/\omega} \tau(\gamma, \omega, t) \cos(\omega t) dt.$$  \quad (6)

An example is shown in Figure 6 (model parameters chosen to fit the moduli from Figure 1). Once the shear amplitude exceeds the yield stress, $G'$ monotonously decreases with increasing amplitude, due to the cut-off of the stress amplitude. Below the yield stress, the model yields $G''=0$ for $\eta_{\phi}=0$. Above the yield stress, $G''$ shoots up, passes through a maximum and finally decreases less strongly than $G'$ with increasing amplitude. Noteworthy, the high $G''$ level is mainly caused by the distinct phase shift of shear stress with regard to the imposed shear strain. Large values of the Bingham viscosity $\eta_{\phi}$ also contribute to $G''$, but mainly at high shear amplitudes. The optional viscosity $\eta_{\phi}$ is required to provide a $G''$-plateau in the limit of small amplitudes. From the experimental view, it might be possible that part of the $G''$ plateau at small amplitudes may be due to the limited phase angle resolution of the magnetorheometer for small $G''/G'$. In summary, typical features of the measured moduli are captured by the simple model, while a fit of the measured shape is not achieved. In addition, it is necessary to take into account the radial shear strain profile in the plate-plate measurement (see below).

5. Conversion from physical to engineering parameters

To predict the performance of MR devices, it is necessary to convert the physical model parameters $G_{\phi}, \tau_{\phi}, \eta_{\phi}, \eta_{\phi}$ into the geometry-dependent engineering parameters $\tilde{G}_{\phi}, \tilde{\tau}_{\phi}, \tilde{\eta}_{\phi}, \tilde{\eta}_{\phi}$, which e.g. allow to relate a force to a displacement. A disk clutch of radius $R$ and gap $h$ is chosen as example. This corresponds to the situation in the plate-plate geometry. The Table summarizes relations between torque $M$ and rim shear stress $\tau_{\phi}$ as well as angular displacement $\varphi$ and rim shear strain $\gamma_{\phi}$, respectively, in the linear viscoelastic regime. They yield a conversion factor $f$, applicable to the modulus and optional viscosity.

| $M = \tau_{\phi} \frac{\pi R^2}{2}$ | $M = f \frac{\tau_{\phi}}{\gamma_{\phi}}$ | $\tilde{G}_{\phi} = f G_{\phi}$ | $\tilde{\tau}_{\phi} = f \tau_{\phi}$ |
| $\varphi = \frac{\gamma_{\phi} h}{R}$ | $f = \frac{\pi R^2}{2h}$ | $\tilde{\eta}_{\phi} = f \eta_{\phi}$ | $\tilde{\eta}_{\phi} = f \eta_{\phi}$ |

Table. Relations between physical and engineering model parameters (example plate-plate geometry).

Only approximate values may be given for the parameters of the Bingham element, due to the linear radial shear amplitude profile in the plate-plate gap. The resulting plate-plate moduli $G_{pp}(\gamma_{\phi}, \omega)$ and $G_{pp}(\gamma_{\phi}, \omega)$ for a given rim shear amplitude $\gamma_{\phi}$ may be calculated as

$$G_{pp}(\gamma_{\phi}, \omega) = 4 \int_{0}^{1} x^4 G(\gamma_{\phi} \omega) dx$$
$$G_{pp}(\gamma_{\phi}, \omega) = 4 \int_{0}^{1} x^4 G'(\gamma_{\phi} \omega) dx.$$  \quad (7)

The moduli from Eq. (7) are contained in Figure 6 as dotted lines. Caused by the radial amplitude profile, the moduli are smeared out and somewhat shifted to higher amplitudes. The shape is thus closer to the measurement in Figure 1, obtained by using plate-plate magnetorheometry. This is an example for the complications caused by strain or stress profiles in MR devices. Finally, it should be mentioned that the storage modulus of the MRF may contribute to a shift of the resonance frequency of an MR device. The applicability of a related model to a dashpot damper is demonstrated in [4].

References

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