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LINEAR VISCOELASTICITY OF MR FLUIDS: DEPENDENCE ON MAGNETIC FIELDS

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The paper presents investigation of dynamic properties of MR fluids by using a rheometer with parallel-plate geometry. The sample is reduced iron powder based MR suspensions. Linear viscoelastic properties of such sample, which can be variably controlled using a magnetic field, are obtained and summarized based on oscillatory tests. Four field-induced regimes, I, II, III, and IV, are found in the system, which are defined by three critical field strengths: $B_{C1} < B_{C2} < B_{C3}$. MR fluids in regime I through IV experience four typical structural convolutions: coexisting of particles and random chains; coexisting of chains and random clusters; coexisting of clusters and chains; stable clusters. Such results are in good accord with experimental results achieved by Liu's group using light scattering techniques.

1 Introduction

The MR effect is attributed to the field-induced magnetization of the disperse phase relative to the continuous phase [1]. Particle magnetization in an applied magnetic field can be described to leading order as a magnetic dipole. The dipolar particles align head-to-tail in the direction of the magnetic field, resulting in the experimentally observed fibrous structures. The fibrous columns must be broken for the suspension to flow, of which this phenomenon relates to the yield stress. At small field strengths, the field-induced yield stress is predicted to be proportional to the magnetic flux density squared. As the applied field strength increases and the particle magnetization begins to saturate, the yield stress will increase sub-quadratically with flux density, eventually becoming field-independent at large flux densities.

The field-induced structure formation and convolution are directly observed by Liu and her colleagues [2,3] with light scattering technique. Their studies indicate that MR fluids exhibit a complex structural behavior: a gas of Brownian particles changes to columnar solid structures due to induced dipole interactions. Two transition structural regimes are observed: (i) randomly distributed chains and particles and (ii) distinct thin columns and randomly distributed chains and particles. Three structural transition magnetic fields are found, one marking each structural transition, from the initial to final structural regime.

Most of MR devices operate dynamic modes where the fluid behaves as viscoelastic properties [4]. Despite the huge potential commercial importance of MR technology, the current knowledge about viscoelastic behaviors of MR fluids is rather small. Indeed, a good knowledge of MR fluid behaviors is crucial for a better theoretical modeling and understanding of MR technology in general and for developing and optimizing MR fluids for the use in commercial applications [5].

The purpose of this paper is to introduce a new testing method based on oscillatory tests to investigate the field-dependence of MR properties. Viscoelastic properties of MR fluids subjected to a slowly increasing magnetic field are measured using strain amplitude sweep mode. The field dependence of viscoelastic properties will be addressed.

2 Theoretical Background on Linear Viscoelasticity

Sinusoidal oscillations are useful to measure the viscoelastic properties of a material. Among the different ways of measuring the properties, the small-amplitude oscillatory shear test is the most widely used method.

Assuming MR suspension is subjected to an oscillatory strain with sufficiently small amplitude as defined by:

$$\gamma(t) = \gamma_o \sin(\omega t) \tag{1}$$

where γ is the shear strain, t is time, γ_o (<<1) is the shear strain amplitude and ω is the frequency of oscillation in *rad/s*.

The resulting shear stress response, $\tau(t)$, in the linear viscoelastic regime is also a sine wave with the same frequency ω, but shifted by an angle δ with respect to the shear strain wave as written by

$$\tau(t) = \tau_o \sin(\omega t + \delta) \tag{2}$$

where τ_o is the shear stress amplitude. The amplitude ratio is called the dynamic modulus, G_d.

$$G_d = \frac{\tau_0}{\gamma_0} \tag{3}$$

Both G_d and δ depend on only frequency. Linear behavior is generally described in terms of the storage modulus, G', loss modulus, G'', and loss factor $\tan\delta$.

$$G' = G_d \cos(\delta) \tag{4}$$

$$G'' = G_d \sin(\delta) \tag{5}$$

$$\tan\delta = G''/G' \tag{6}$$

The storage modulus, G', also represents the ability of a viscoelastic material to store the energy of deformation, which contributes to the material or device stiffness. The loss modulus, G'', on the other hand, represents the ability of the material to dissipate the energy of deformation as heat. The phase angle, δ, is an indication of whether the viscoelastic material behaves more as solid-like or liquid-like material

3 Experiment

The sample is a suspension of reduced iron powders (BASF AG Co.) in silicone oil with a volume fraction 40%. The SEM image for the reduced iron powders is shown in Fig. 1. As shown in this SEM image, the particles are spherical with average diameter around 4.7 μm and have quite smooth surfaces. The particle density is 7.789 kg/m^3. The silicone oil has a viscosity of 0.1 PaS and density of 963.8 kg/m^3. A small amount of surfactant was added into the suspension so that the particles could suspend in carrier fluid without setting for about 6 hours.

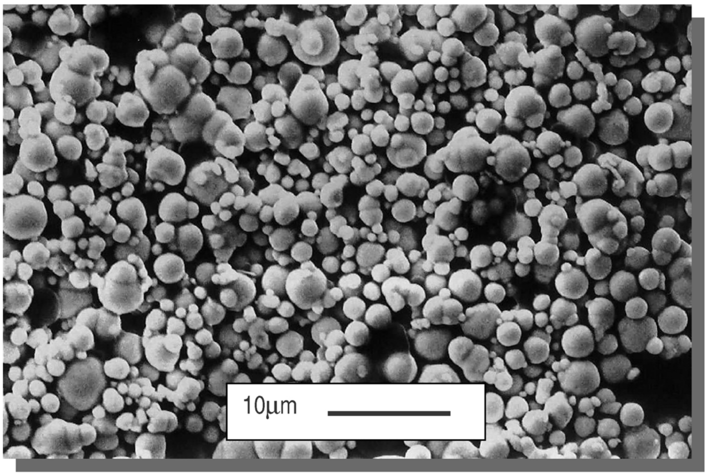

Fig. 1. SEM image of reduced iron powders

The rheology of the sample is measured in a parallel-plate geometry with a Paar Physica rheometer. The system includes a measuring drive system, an electronic control panel, a user-interface software package and a power unit. This rheometer is equipped with an additional MR cell having a parallel plate configuration, as shown in Fig. 2. The MR material is placed in a constant gap of 1.0mm before applying a required magnetic field, which is provided and controlled by a built-in MR cell and a DC power supply, respectively.

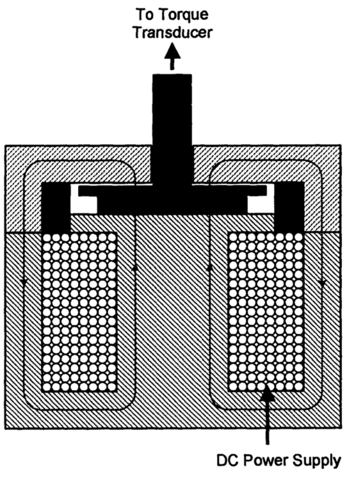

Fig. 2. Schematic diagram of the MR cell

A series of dynamic measurements for successively increasing magnetic field strengths ("*magnetic field sweeps*") is employed to investigate viscoelastic properties of the MR sample. The testing procedure for each measurement is illustrated below. Initially, the sample is sheared at a constant shear rate 100 s^{-1} at a zero field for half a minute to distribute the particles uniformly. Next, the desired magnetic field is applied and maintained for 1 minute without any shear. Then viscoelastic properties of the sample are measured with *strain amplitude sweep mode*: the frequency, ω, is fixed at a given value of 10 rad/s and the strain amplitude, γ_0, is swept from 0.001% to 100% in a logarithmically increasing mode, the relationships for the storage modulus, G', and loss factor, $tan\,\delta$, against strain amplitude can be obtained.

4 Results and discussions

MR fluid performs either *linear viscoelastic* properties or *nonlinear viscoelastic* properties according to the applied strain, γ_0, is below or above a critical strain, γ_c. At large strain shear ($\gamma_0 > \gamma_c$), macrostructures of MR fluids are changed, which result in viscoplastic behaviors [5]. For example, conventional steady-shear tests are widely used to study viscoplastic properties of MR fluids, including dynamic yield stress and apparent viscosity. In this case, MR structures are totally broken because the dynamic yield stress results from the rupture of chains or columns [6]. At small strain shear ($\gamma_0 \leq \gamma_c$), MR fluid behaves as linear viscoelastic properties because such low deformations do not destroy microstructures of MR fluids [5].

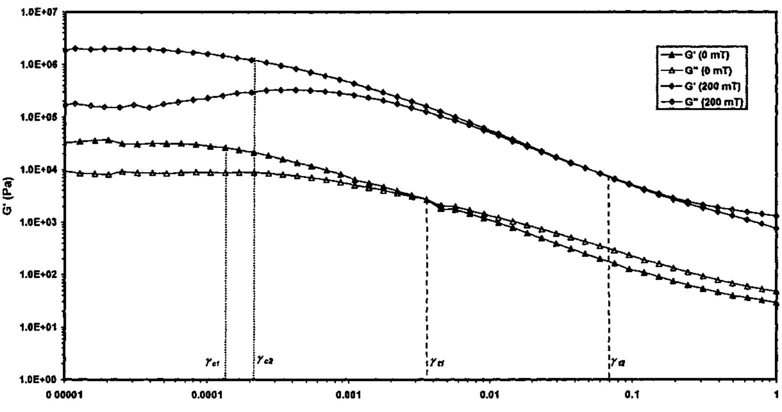

Fig. 3. Plots of G' and G'' versus γ_0 without and with a magnetic applied

In order to determine the strain amplitude rage over which the linear viscoelasticity holds, strain amplitude sweep experiments are carried out at a frequency 10 rad/s over the range of strain amplitude of 0.001% ~ 100%. Fig. 3 shows the graph of storage modulus, G', and loss modulus, G'', versus strain amplitude, γ_0, without and with a magnetic field. Two critical strains, γ_{c1} and γ_{c2}, demarking the end of linear viscoelastic response, are characterized where G' is constant. As shown in this figure, the value of γ_c shows a slightly increasing trend with field on the order of 0.0001 to 0.0003. However, the magnitude of storage modulus increases sharply with increasing magnetic field. The value of G' at the field strength 200 mT is as large as 2.0×10^{6} Pa, which is approximately two orders higher than that without field (~ 3.4×10^{4} Pa). This result demonstrates that MR fluid shows more elastic property with increasing field strength.

In addition, another critical strain, γ_t, is demarked in the figure, which is defined as the crossover of G' and G''. At the ranges of $\gamma<\gamma_t$, $G'>G''$, which implies elasticity dominant; above the value of γ_t, $G''>G'$, MR fluid behaves as more viscous properties. As shown in this figure, the critical value, γ_t, increases steadily with the increment of magnetic field: $\gamma_{t1}(0\,mT)<\gamma_{t2}(200\,mT)$. This result also demonstrates that elastic regime shows a steadily increasing trend with magnetic field.

As shown in Fig. 3 again, the critical strain for distinguishing the linear viscoelastic regime is around 0.01%, i.e., $\gamma_c \approx 0.01\%$. That is, when the strain amplitude is smaller than the critical value, MR fluid behaves as a linear viscoelastic body, and its microstructures is not destroyed. Thus, to avoid rupturing the sensitive structure during the chain formation and convolution, a sufficiently small strain amplitude $\gamma_0 = 0.005\%$ and a constant angular frequency $\omega = 10\,rad/s$ is set and the magnetic field strength is increased continuously from 0.68 mT to 850 mT. Fig. 4 (a) and (b) shows the storage modulus, G', and the loss factor, $tan\,\delta$, as a function of the magnetic field strength, B, respectively. As can been seen from these two figures, there are four significant regimes: I, II, III, and IV. These four regimes are separated by three critical field strengths: $B_{C1} = 18\,mT$, $B_{C2} = 53\,mT$, and $B_{C3} = 470\,mT$.

A. *Regime I*

In this regime, G' is kept a very small constant 6.5×10^{4} Pa. MR fluid behaves as Newtonian fluid. For $B=0$, the suspension is well dispersed without particle aggregate. For small field strengths $B<B_{C1}$, particle aggregate start to form, but gap-spanning chains do not exist. Thus, such conglomerates or structures of magnetic particles become well dissolved and distributed at low magnetic field strength. The suspension in this regime is a mixture of particles and short chains.

B. *Regime II*

Between B_{C1} and B_{C2}, both G' and $tan\,\delta$ show increasing trend with field strength. The increment of G' with field strength indicates that when the field is above B_{C1}, the particle chains are getting longer to span the gap with the increasing field, and meanwhile, few chains interact each other to form into randomly short clusters. The increment of $tan\,\delta$ is not just caused by the increasing length of the chains, but also by the displacement of the chain or cluster fluctuation. Therefore, this suspension in this regime is a mixture of chains and random clusters.

C. Regime III

In contract to regime II, G' in this regime, $B_{C2} < B < B_{C3}$, shows a sharply increasing trend with magnetic field, but $tan\,\delta$ decreases sharply with the increment of field strength. In this regime, the random clusters grow and span the gap as the field strength is increased. The suspension in this regime is a mixture of clusters and chains.

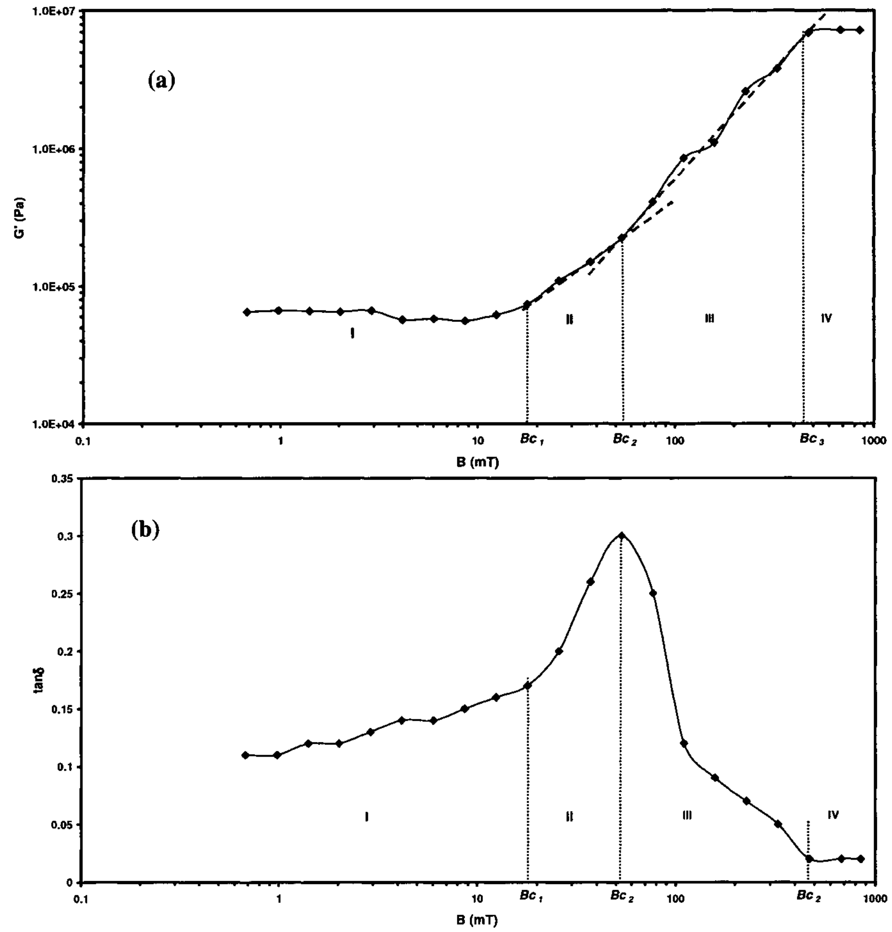

Fig. 4. (a) $G' \sim B$; (b) $tan\delta \sim B$. ($B_{C1} = 18mT, B_{C2} = 53mT, B_{C3} = 470mT$)

D. Regime IV

As the field strength is reached B_{C3}, all the particle chains have formed into stable clusters along the field lines. Such stable clusters have the maximum resistance to prevent flow; consequently, MR fluid has the maximum storage modulus. Further increasing field strength, G' keeps a plateau value because the cluster structures of MR fluid do not change any more.

In general, the field dependence of linear rheology of MR fluid is summarized in Table 1.

Table 1. Field Strength Dependence of MR Rheology

Properties / Field Strength B	Linear Viscoelasticity	Structural Formation & Convolution
$B < B_{C1}$	G'— tanδ—	Coexisting of particles and random chains
$B_{C1} < B < B_{C2}$	$G'\uparrow$ tan$\delta\uparrow$	Coexisting of chains and random clusters
$B_{C2} < B < B_{C3}$	$G'\uparrow$ tan$\delta\downarrow$	Coexisting of clusters and chains
$B > B_{C3}$	G'— tanδ—	Stable clusters

5 Conclusion

In this paper, the field strength dependence of linear viscoelasticity of MR suspension is reported. The experimental study is performed based on oscillatory tests by using a MR rheometer in parallel-plate configuration. The magnetic sweep results indicate that MR fluids exhibit four field-induced regimes, I, II, III, and IV, which are defined by three critical magnetic field strengths: $B_{C1} < B_{C2} < B_{C3}$. Corresponding to these four regimes, micro-structural formation and convolution of MR fluids experiences coexisting of particles and random chains, coexisting of chains and random clusters, coexisting of clusters and chains, and stable chains.

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