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Non-linear viscoelastic response of magnetic fiber suspensions in oscillatory shear

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ABSTRACT

This paper reports the first study on the large amplitude oscillatory shear flow for magnetic fiber suspensions subject to a magnetic field perpendicular to the flow. The suspensions used in our experiments consisted of cobalt microfibers of the average length of 37 μm and diameter of 4.9 μm, dispersed in a silicon oil. Rheological measurements have been carried out at imposed stress using a controlled stress magnetorheometer. The stress dependence of the shear moduli presented a staircase-like decrease with, at least, two viscoelastic quasi-plateaus corresponding to the onset of microscopic and macroscopic scale rearrangement of the suspension structure, respectively. The frequency behavior of the shear moduli followed a power-law trend at low frequencies and the storage modulus showed a high-frequency plateau, typical for Maxwell behavior. Our simple single relaxation time model fitted reasonably well the rheological data. To explain a relatively high viscous response of the fiber suspension, we supposed a coexistence of percolating and pivoting aggregates. Our simulations revealed that the former became unstable beyond some critical stress and broke in their middle part. At high stresses, the free aggregates were progressively destroyed by shear forces that contributed to a drastic decrease of the moduli. We have also measured and predicted the output strain waveforms and stress–strain hysteresis loops. With the growing stress, the shape of the stress–strain loops changed progressively from near-ellipsoidal one to the rounded-end rectangular one due to a progressive transition from a linear viscoelastic to a viscoplastic Bingham-like behavior.

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1. Introduction

Magnetorheological (MR) fluids are suspensions of magnetic micron-sized particles in a carrier liquid. The MR fluid particles are subject to a reversible aggregation and the MR fluid develops a large yield stress upon application of a magnetic field. This phenomenon is known as the magnetorheological effect [1,2]. So, the magnetic and mechanical properties of these fluids can be efficiently controlled by external magnetic fields and this makes them attractive for several industrial applications. Controlled stiffness dampers and magnetorheological finishing of optical surfaces have been successfully commercialized during the last decade and the other magnetorheological smart devices, such as controlled hydrodynamic bearings, are being developed [3]. The improvement of the real time control of these devices and the increase of the controllability range of their control parameters (flow rate, stiffness) are still important issues. In general, there are two ways of solving these problems: either by optimization of the fluidic and the magnetic designs of the considered device or by improving the properties of the MR fluid.

One way of improvement of the MR fluid stability and mechanical properties consists of using magnetic micro- or nanofibers instead of conventional spherical particles. The magnetic fiber suspensions have shown better sedimentation stability [4] and developed a yield stress a few times larger than that of the magnetic suspensions of spherical particles at the same magnetic field intensities and the same particle volume fraction [5–10]. Such enhanced magnetorheological effect in fiber suspensions has been explained in terms of the interfiber solid friction [11] and by enhanced magnetic permeability of these suspensions as compared to the permeability of conventional MR fluids [9,12]. Note that the similar particle shape effect has been observed in electrorheological (ER) fluids [13–16] and was attributed to both the physical overlapping of the elongated particles and to their strong dielectric properties [17,18]. Kor and See [19] have recently carried out particle level simulations on ER fluids and

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also found the stress enhancement in suspensions of needle-like particles.

Almost all the studies on elongated particle-based MR or ER fluids were focused on the steady shear or the small amplitude oscillatory shear flows. These studies give an important insight into the understanding of the general rheological behavior of the suspensions but have limited applications in ER and MR smart devices. For instance, in MR dampers, the MR fluid is subject to a reciprocal large amplitude straining motion. Thus, investigation of the large amplitude oscillatory shear (LAOS) flow of magnetic fiber suspensions would be very helpful for this application. Apart from its engineering importance, the LAOS response of the magnetic fiber suspensions could bring valuable information on their nonlinear viscoelasticity in the wide range of the excitation frequencies.

In general, the LAOS tests have been successfully used to characterize the rheological properties of various kinds of soft materials, such as polymers, wormlike micelles, colloidal gels, biopolymer networks, see for instance [20,21]. These tests consist of imposing a sinusoidal strain (or stress) to the material and recording its stress (or, respectively, strain) response. In the nonlinear viscoelastic regime, i.e. at large strains or stresses applied, the material response is non-sinusoidal with higher harmonics appearing. Fourier analysis of the measured response signal allows one to learn about a rheological behavior exhibited by the material (shear thinning/thickening, strain hardering/softenning). The general framework of treatment and interpretation of the LAOS data has been developed by Wilhelm [22] and Ewoldt et al. [23].

A number of works has been devoted to the LAOS response of conventional ER fluids and MR fluids, both composed of spherical particles [24–32]. The electrorheological and magnetorheological effects are essentially similar in nature, so both ER and MR fluids show similar behavior in oscillatory shear. In most of the works, the first harmonic (or fundamental) shear moduli were used as the principle measure of the viscoelastic response of these fluids. In experiments, both storage and loss moduli have been found to increase with the magnetic (or electric) field strength because of the field-induced aggregation of the MR (or ER) fluids. At small strains, the aggregates spanned the gap between rheometer plates and the MR and ER fluids showed an elastic behavior with the storage modulus substantially larger than the loss modulus. Nevertheless, the loss modulus was non-negligible and even a few orders of magnitude larger than the one of the dispersing liquid of the suspension. Such viscous response of the suspensions was interpreted in the two following ways. First, McLeish et al. [33] supposed a co-existence in the ER fluid of the gap-spanning particle chains with the free chains attached to the wall by one of the ends. The gap-spanning chains were supposed to move affinelly with the suspending liquid and contributed to the suspension storage modulus, while the free chains moved out of phase with the rheometer walls and bended under the hydrodynamic forces exerted by the suspending liquid. Thus, a small fraction of the free chains contributed to the suspension loss modulus. Moreover, different chain bending modes gave an infinite series of relaxation times. To fit their theory to experimental data on the loss modulus, the authors adjusted the volume fraction of the free chains. On the other hand, Klingenberg [34] has carried out particle level simulations and shown that the non-negligible viscous response of the ER fluids could arise from non-affine motion of the particles inside the gap-spanning aggregates even at strains as small as 10^{-4}.

With increasing strain amplitude, the storage modulus experiences a slow decrease until some critical strain followed by an abrupt decrease above this strain. The loss modulus also decreases with the strain but in a less extent, so that, at large strains, both moduli are usually of the same order of magnitude signifying the transition from solid to liquid like behavior [35]. Parthasarathy and Klingenberg [26,27] explained the smooth initial decrease of the storage modulus by a short-scale rearrangement of particles in aggregates and they interpreted the further abrupt drop of the modulus by the large-scale structure rearrangement and by rupture and reformation of gap-spanning structures.

In most cases, the frequency dependence of the shear moduli of ER and MR fluids followed a Maxwell-like behavior with the storage modulus increasing monotonically and attaining a high frequency plateau and the loss modulus having a maximum at intermediate frequency. Such dependence supports, at least qualitatively, both McLeish's "free chain hypothesis" [33] and Parthasarathy and Klingenberg's [27] observations of small and large scale cluster rearrangements.

A deeper understanding of the nonlinear viscoelastic response of the ER and MR fluid could be achieved by analyzing the stress–strain hysteresis loops, called Lissajous plots. When increasing the amplitude of the applied strain, the suspension stress response becomes non-linear and the Lissajous plots change their shape from the ellipsoidal shape at low strains to the parallelogram-like one at high strains [24]. Martin and Odinek [25] have attributed this particular shape of the Lissajous plots to rapid fragmentation and aggregation of particle chains in oscillatory shear. They have proposed a phenomenological equation describing kinetics of chain aggregation/fragmentation and developed a theoretical model, which reproduced the experimental hysteresis loops reasonably well with a single fit parameter. Parthasarathy and Klingenberg [27] have also well reproduced the parallelogram-like hysteresis loops in their simulations and explained them in terms of plastic Bingham-like behavior. Namely, as the direction of shear is reversed, the stress varies rapidly until the structure yields, and the stress becomes independent of strain.

Tracking back to the ER and MR fluids composed of elongated particles, only the paper of Tsuda et al. [15] reports on the nonlinear viscoelastic response of such fluids – the ER whisker suspension. In their paper, the authors measured the complex shear modulus as function of the applied stress and have found a similar abrupt decrease of the modulus with increasing stress as in the case of suspensions of spherical particles. However, the solid-to-liquid transition in whisker suspensions (corresponding to the drop of the modulus) occurred at higher critical stresses. This was explained by the formation of branched intricate structures in whisker suspensions that were mechanically stiffer than the column structures in suspensions of spheres. No results have been presented on the stress and strain waveforms. Theoretical models have neither been reported to describe the LAOS response of the ER or MR suspensions of elongated particles. It is worth to mention the dipole interaction models of Kanu and Shaw [17] and de Vicente et al. [9] that predicted the elastic modulus of, respectively, ER and MR suspensions of needle-like particles in the linear viscoelastic regime at small applied stresses or strains.

In this paper we have carried out a detailed investigation of the large amplitude oscillatory shear flow of the magnetic fiber suspensions in the presence of an external magnetic field. In experiments, we apply a sinusoidal shear stress and measure the output strain waveform. We examine effects of the magnetic field strength, stress amplitude and excitation frequency on the strain signal as well as on the shear moduli of the fiber suspension. Particular attention is focused on the stress dependence of the loss modulus which, to our opinion, has never been clearly explained even for conventional suspensions of spherical particles. Finally, we develop a microstructural model that allows describing, at least semi-quantitatively, all observed rheological behaviors in the non-linear viscoelastic regime. In particular, to explain relatively high values of the loss modulus, we assume a specific structure of the fiber suspension composed of both percolating aggregates and the free branches attached by one extremity to either the walls of a flow cell or to the percolating aggregates. These free branches move out of phase with

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