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Using oscillatory shear to probe the effects of bidispersity in inverse ferrofluids

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Abstract

The effects of particle size distribution on the magnetorheological response of inverse ferrofluids was investigated using controlled mixtures of two monodisperse non-magnetisable powders of sizes 4.6 μm and 80 μm at constant volume fraction of 30%, subjected to large amplitude oscillatory shear flow. In the linear viscoelastic regime (pre-yield region), it was found that the storage and loss moduli were dependent on the particle size as well as the proportion of small particles, with the highest storage modulus occurring for the monodisperse small particles. In the nonlinear regime (post yield region), Fourier analysis was used to compare the behaviour of the 1st and 3rd harmonics (I_1 and I_3 respectively) as well as the fundamental phase angle as functions of the applied strain amplitude. The ratio of I_3/I_1 was found to become more pronounced with decreasing particle size as well as with increasing proportion of small particles in the bidisperse mixtures. Furthermore, the phase angle was able to clearly show the transition from solid-like to viscous behaviour. The results suggested that the nonlinear response of a bidisperse IFF is dependent on particle size as well as the proportion of small particles in the system.

Keywords : inverse ferrofluid, large amplitude oscillatory shear flow, particle size distribution, bidisperse, linear/nonlinear viscoelastic, Fourier analysis

1. Introduction

Magnetorheological and electrorheological fluids (MRF and ERF respectively) are field responsive materials which are generally composed of micron-sized particles, and they show dramatic increases in their flow resistances upon application of external magnetic or electrical fields. For both systems, this field responsive behaviour is attributed to the formation of field-induced dipoles within each particle. Dipole-dipole interactions between the particles lead to the formation of chains or aggregates in the direction of the applied field, and this in turn leads to the increase in the flow resistance of the material. More details on these physical mechanisms and materials are provided in a number of review articles (Block and Kelly, 1988; Parthasarathy and Klingenberg, 1996; See, 1999). Furthermore, due to their tunable flow properties, these fluids are used in a variety of engineering applications such as damping and high precision polishing (Kordonski and Golini, 2000; Stanway *et al.*, 1996; Klingenberg, 2001; Bombard *et al.*, 2002; Magnac *et al.*, 2006).

A third type of field responsive suspension, the inverse ferrofluid, has been receiving increased attention in recent years. Inverse ferrofluids (abbreviated as IFF) may be used to model the behaviour of MRF and ERF, and they are comprised of a magnetisable carrier liquid (which is a ferrofluid consisting of nano-sized magnetite particles) into which is dispersed micron-sized non-magnetic solid particles. Due to the difference in particle size between the larger non-magnetic particles and the nano-sized particles in the carrier liquid, the larger particles would behave as if they were in a continuum. Similar to the behaviour of a conventional MRF, in the presence of a magnetic field dipolar interactions are induced in the IFF between the micron-sized particles and aggregates are formed (Skjeltorp, 1983). Consequently, IFF also show field induced increases in their flow resistances which are analogous to MRF and ERF, although with a smaller magnitude (Bossis and Lemaire, 1991; Popplewell *et al.*, 1995; de Gans *et al.*, 1999; 2000; Volkova *et al.*, 2000; Saldivar-Guerrero *et al.*, 2005). The existing studies of IFF have made use of micron-sized non-magnetic particles (Lemaire *et al.*, 1995; Popplewell and Rosenweig, 1995; 1996; Saldivar *et al.*, 2005; 2006), and nano-sized particles (de Gans *et al.*, 1999; 2000).

A significant advantage of IFF over MRF is that a wide range of non-magnetic particles are currently available in different sizes which may be used to probe the effects of particle size distribution (de Gans *et al.*, 2000). Already, similar studies of particle size and shape have been done for ERF (Ota and Miyamoto, 1994; Shih and Conrad,

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1994; Wu and Conrad, 1998; Parthasarathy and Klingenberg, 1999; See *et al.*, 2002), and for MRF (Trendler and Böse, 2004; Kittipoomwong and Klingenberg, 2005; Wereley *et al.*, 2006). The simulations carried out by Kittipoomwong and Klingenberg (2005) showed that yield stress was higher for bidisperse MRF than for monodisperse MRF. They attributed this to the fact that the smaller particles had the effect of inducing the larger ones to form chainlike structures. These results were similar to the experimental findings on ERF by See *et al.* (2002), but different from the experimental findings of Wu and Conrad (1998). Wereley *et al.* (2006) investigated the effects of bidispersity on a MRF composed of micron-sized and nano-sized particles. They found that the yield stress increased with an increase in nano-particle concentration of up to 20%. In excess of 20% nano-particle concentrations, the yield stress decreased to a value lower than the initial value observed for the monodisperse micro-particle case.

Attempts to experimentally investigate the rheology of IFF with micron-sized particles have mainly focused on either monodisperse systems (Popplewell and Rosenweig, 1995; 1996; Saldivar *et al.*, 2005) or polydisperse systems with a broad size distribution (Lemaire *et al.*, 1995; Saldivar *et al.*, 2006). This paper will focus on the behaviour of bidisperse systems with two distinct particle sizes. As such, we will now briefly review the literature for systems where the effects of particle size polydispersity have been explicitly examined. Lemaire *et al.* (1995) compared the flow curves for an IFF composed of monodisperse silica particles of average size 45 μm with a polydisperse system with particle sizes in the range 5-50 μm. They found that there was no difference in the flow curves for these two systems. Saldivar *et al.* (2006) investigated the viscoelastic behaviour of three IFF systems made up of two monodisperse particles (11 μm and 3 μm respectively) and a polydisperse particulate system (mean diameter of 1.08 μm). They found that the storage modulus for the monodisperse particles increased with particle volume fraction. The storage modulus was also found to be independent of particle size for the monodisperse particles, and this agreed with a theoretical prediction by de Gans *et al.* (1999) for the case of nano-sized dispersions. In comparing the polydisperse system with the monodisperse systems, they found that the value of the storage modulus for polydisperse systems increased more slowly than those for the monodisperse systems as the magnetic field was increased. They suggested that this was due to the effects of non-magnetic interactions. All these papers have considered the case of broad size distribution. An alternative approach to considering the effects of particle size distribution would be to investigate the simplest case of a polydisperse system i.e. a bimodal or bidisperse system. This will be considered in this paper.

The usual method of characterizing ERF, MRF or IFF is steady shear flow, where the flow curves of these materials are measured. As reported elsewhere, some issues arise with the use of flow curves for characterizing IFF (Ekwebelam and See, 2006). On the other hand, another useful alternative approach would be to make use of oscillatory flow to investigate the onset of non-linear viscoelastic behaviour of the material as the strain amplitude is increased. Furthermore, a number of MRF and ERF devices operate under dynamic mode where they are subjected to oscillatory flow, so it is important to understand their behaviour under these flow conditions (Li *et al.*, 2003). The oscillatory shear approach has already been used by a number of workers for ERF (Weiss *et al.*, 1994; Parthasarathy and Klingenberg, 1995; Parthasarathy and Klingenberg, 1999; Otsubo *et al.*, 1992; Gamota and Filisco, 1991a; 1991b; 1991c; Gamota *et al.*, 1993) and MRF (Weiss *et al.*, 1994; Li *et al.*, 2003; Claracq *et al.*, 2004; Wereley *et al.*, 2006). Of more relevance, this technique has been used for monodisperse and polydisperse micron-sized IFF systems (Saldivar *et al.*, 2006) as well as monodisperse nano-sized systems of IFF (de Gans *et al.*, 1999; 2000) as discussed above. Thus far, we have not found any published work which shows that this technique has been applied to a bidisperse micron-sized IFF system, and this will be the focus of this work.

In this paper, we will look at the effect of mixing two particles of distinctly different sizes (keeping volume fraction constant) in an IFF. Furthermore, we will use oscillatory shear tests to characterize the system, and in doing so we will follow two main approaches. In the first place, we will consider the linear viscoelastic moduli (which are used to characterize the "stiffness" of the sample). In addition, as the materials show yielding behaviour at large strains, Fourier analysis of the stress waveforms will be used (here, in particular, we will utilize the concept of increased third harmonics). Fourier analysis has been shown in general to be a useful tool for characterizing complex materials (Wilhelm *et al.*, 1995; Sim *et al.*, 2003), and this technique will be applied to the analysis of IFF in this paper.

The structure of this paper is as follows: the materials, equipment and experimental method are described next; the test carried out was an oscillatory shear flow test, and we include a description of the Fourier technique here. In the following section we display the results from this test, and then we discuss the implications of these results in the context of both the material's stiffness, as well as Fourier analysis. The paper ends with a conclusion section.

2. Method

2.1. Raw Materials

The ferrofluid used was supplied by Ferrotec Inc. USA

Table 1. The composition of particles used for the bidisperse IFF system

Sample name	4.6 μm particles: proportion in mixed suspension (vol. %)	80 μm particles: proportion in mixed suspension (vol. %)
100:0	100	0
75:25	75	25
50:50	50	50
25:75	25	75
0:100	0	100

(type EFH1) and consisted of a mineral based fluid into which were dispersed nano-sized magnetite particles. The fluid had a viscosity of 6×10^{-3} Pa.s at 27°C and a density of 1.21 g/ml. The disperse phase consisted of spherical particles of polymethylmethacrylate and polyethylene, both of which had average particle sizes of 4.6 μm and 80 μm respectively. The bidisperse IFF was made up by dispersing mixtures of varying ratios of these powders into the ferrofluid, and each mixture was dispersed to an overall volume fraction of 30%. Microscopic observation of the IFF showed that there were no signs of clumping or sedimentation. There were also no apparent signs of any chemical reactivity between any of the materials making up the bidisperse IFF. Furthermore, the two materials appeared to be well mixed with each other within the system. Table 1 shows the compositions of the particle sizes used for the suspension.

2.2. Equipment

The tests were carried out using a Paar Physica MCR300 rheometer with the 20MR magnetorheological cell; this employs parallel plates of diameter of 20 mm. A uniform magnetic field, generated by coils under the bottom plate, was applied perpendicularly to the plates. The sample was enclosed in a chamber consisting of iron elements to create a magnetic circuit. This test cell has previously been used in several studies on MRF (See and Chen, 2004; See *et al.*, 2004; Lim *et al.*, 2004). Fig. 1 shows a schematic of the experimental set up. All tests were performed using a constant gap setting of 1mm, a fixed frequency of 10 Hz and constant temperature of approximately 25°C. The frequency of 10 Hz was chosen as it is close to frequencies commonly encountered in magnetorheological applications (Iyengar and Alexandridis, 2004).

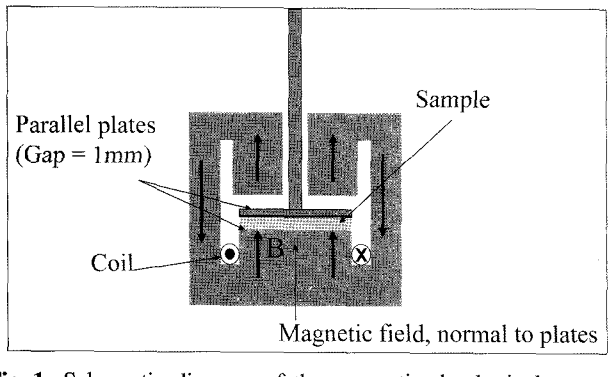

Fig. 1. Schematic diagram of the magnetic rheological test cell MR20 on the Anton Paar Physica MCR 300 rheometer.

2.3. Rheological test: oscillatory shear flow

In the absence of a magnetic field, the sample was initially subjected to a shear rate of $20s^{-1}$ for 10s to ensure the break up of any preformed aggregates. Following this, a field with a magnetic flux density of 0.54T, the highest which can be obtained with this system, was applied to the sample for 2 minutes in the absence of shearing to promote the formation of aggregates. After this, a sinusoidal strain was applied to the sample with a logarithmic increase over the strain range 10^{-4}-1, and the stress response of the material to this deformation was then analyzed. Two regimes exist for which this stress output analysis may be carried out, namely, the linear viscoelastic regime (corresponding to pre-yield) and the non-linear viscoelastic regime (corresponding to post yield).

In the linear viscoelastic regime, the material is subject to small oscillatory strains, and the resultant stress will oscillate with the same imposed driving frequency, but with a phase shift, δ. The behaviour of the material in this regime may be analyzed by measuring the storage and loss moduli (G' and G'' respectively).

At large strain amplitudes, however, the material progresses to the non-linear viscoelastic regime, and this is characterized by the appearance of odd multiples (or harmonics) of the driving frequency (Wilhelm *et al.*, 1995; Li *et al.*, 2003; Sim *et al.*, 2003). Fourier Transform (FT) analysis may then be used to obtain a more in-depth understanding of the stress response of the material, which may be represented as (Li *et al.*, 2003)

$$\tau(t) = \sum_{k=1,odd}^{N} \tau_k \sin(2\pi fkt + \delta_k) \tag{1}$$

where τ is the stress (Pa), k is the odd-harmonic number, f is the driving frequency (Hz), t is the time, and N is the number of harmonics being considered, δ_k is the phase angle of the k-th harmonic.

The intensities of the odd harmonics indicate deviations from linearity (Wilhelm *et al.*, 1995). Thus, it is possible to compare the intensities of the harmonics in this regime, and this will then give an idea of the yielding behaviour of the material. The approach taken in this paper is that we will monitor the yielding process by comparing the intensities of the 1st and 3rd harmonics as strain amplitude is increased. From these results it is expected that more light will be shed on the effects of bidispersity on the behaviour of IFF.

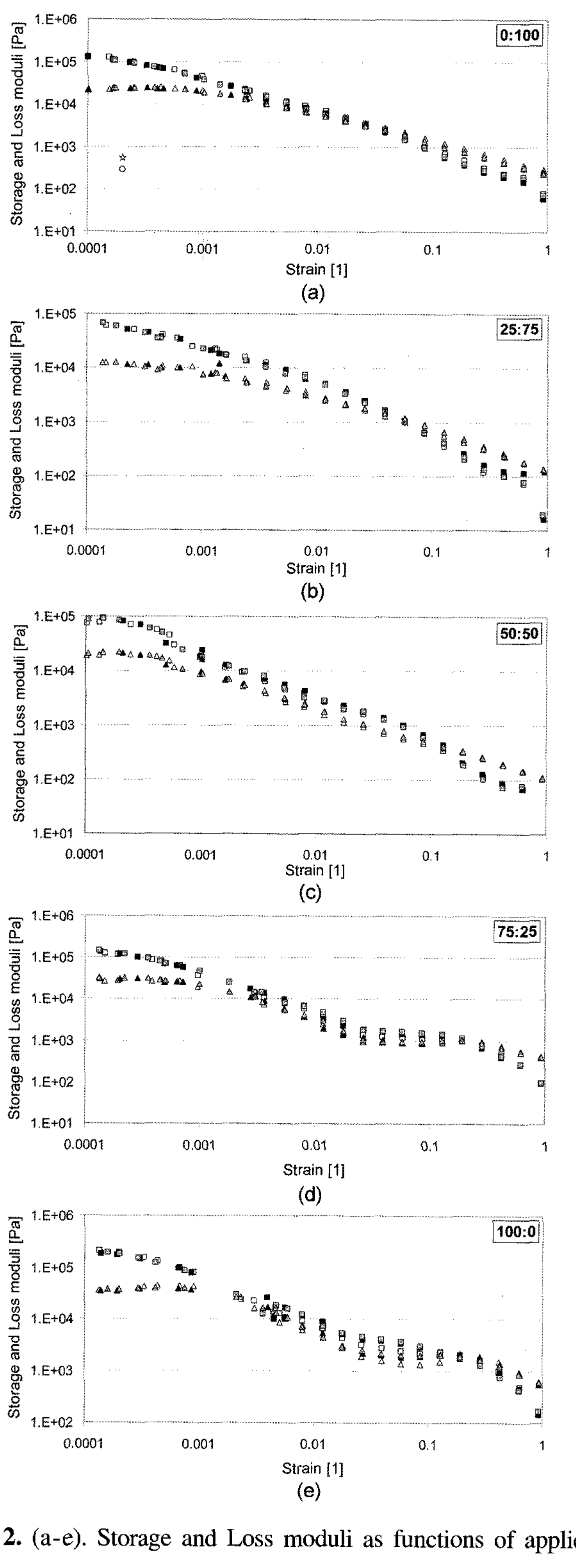

Fig. 2. (a-e). Storage and Loss moduli as functions of applied strain for the different compositions of bidisperse systems used in this test. Squares refer to storage moduli, G'; triangles refer to loss moduli, G''. The proportions of small to large particles in each figure are (a) 0:100; (b) 25:75; (c) 50:50; (d) 75:25; (e) 100:0. The hexagon and star on Fig. 2 (a) mark the storage and loss moduli respectively for the zero field (0T) case.

3. Results and discussion

3.1. Linear viscoelastic behaviour of bidisperse IFF

Figs. 2(a-e) show the fundamental storage and loss moduli for the different bidisperse systems as functions of the applied strain. The raw data points are deliberately shown on the graphs in order to show the reproducibility of each test. Note that each measurement was repeated twice (making a total of three measurements for each test).

Fig. 2a also shows the storage and loss moduli for the 0:100 monodisperse sample in the absence of a magnetic field. The values are ~300Pa and 600Pa for G' and G'' respectively - these are similar to the zero field values with the other suspension systems studied. Note that these values are orders of magnitude lower than the case under an applied magnetic field, thus showing that these IFF samples indeed show a dramatic increase in shear resistance under a magnetic field.

In all cases, G' is seen to be at least an order of magnitude higher than G'', and this shows that each of the bidisperse mixtures show predominantly elastic behaviour at small strains in the presence of the magnetic field. Also, G' begins to decrease at strains of $\sim 2\times10^{-4}$ regardless of the composition of the mixture, and this suggests that the yield strain is independent of size or composition for these bidisperse systems. Taking a look at G'', it appears to be less sensitive to the small strain deformations applied to the IFF, as the amplitude at which it begins to decrease is slightly higher than the yield strain for G' (typically $\sim 6\times10^{-4}$). The decrease in G' with increasing strain may be explained by the rupture of the aggregates within the system. As these aggregates are broken up, the elastic-like behaviour of the system is reduced as the system becomes more viscous-like in nature. These results seem to indicate that the point at which yielding occurs is independent of the composition of a bidisperse IFF.

Turning now to the magnitudes of G' and G'' observed, we may investigate the stiffness of a bidisperse IFF system by looking more closely at the values of the moduli at the small-strain limit of linear viscoelasticity. Table 2 shows these values for the different mixtures of small and big particles.

Table 2. G' and G'' values for an IFF composed of different ratios of small and large particles

Proportion of small particles	G' × [kPa]	G'' × [kPa]
100:0	200 ± 10	41 ± 2
75:25	140 ± 10	30 ± 2
0:100	120 ± 10	25 ± 1
50:50	90 ± 10	21 ± 1
25:75	50 ± 10	12 ± 0.2

From the table it is clear that the highest values of both G' and G'' occur for the monodisperse 100:0 system (all small), while the lowest values occur for the bidisperse 25:75 system. G' is a measure of the stiffness of the aggregates formed. The results also suggest that the value of G' depends on both the size of particles (comparing the two monodisperse systems), as well as the ratio of particle sizes present in a bidisperse system.

These results presented above for the monodisperse systems differ significantly from the observations of Saldivar *et al.* (2006), who found that the value of G' was independent of particle size. The differences in results could be explained by the differences in size ratio of the particles used for both works: the particles used by Saldivar *et al.* (2006) differ in size by a ratio of (3.7:1), while the particles used in our study differ in size by a ratio of (17.4:1). To further examine this point, we note that the trend observed in the value of G' as a function of composition for the bidisperse system is also different to the findings of Saldivar *et al.* (2006) for their polydisperse system, but similar to the findings of Wu and Conrad (1998) on bidisperse ERF. The similarity to the results of Wu and Conrad may be attributed to the fact that the size ratios of particles used in our work (17.4:1) is also similar to the size ratios used by Wu and Conrad (16.7:1). See *et al.* (2002) have suggested that, for the large size ratios used by Wu and Conrad (1994) for their work (and similar to that used in this present work), the bidisperse particles would be very highly packed, and as such, would occupy less volume. This could tend to reduce the overall stress contribution of this system, and as such, the value of G' would be lower for the system. This provides a possible explanation for the minimum observed in Table 2 for the 25:75 case. Further, these results seem to suggest that there would be a minimum size ratio for which G' ceases to be independent of particle size.

3.2. Nonlinear viscoelastic behaviour of IFF: Large strain amplitudes

Large strain oscillatory shear is often used to probe the yielding behaviour of materials which are believed to possess a yield stress. We now consider the effects of bidispersity on IFF in the non-linear viscoelastic region. As was mentioned earlier, the large strain behaviour of a complex material may be explored more fully by comparing the intensities of the odd harmonics as a function of the strain amplitude. Fig. 3(a-e) shows results of the Fourier analysis of the stress response of these bidisperse systems based on equation (1). The figure shows the intensities of the 1st and 3rd harmonics as functions of the applied strain. The data points are for average values of the harmonics for the 3 sets of tests for each bidisperse system. The error bars show the 95% confidence limits of the average values of the harmonics, and from the small size of the error bars it

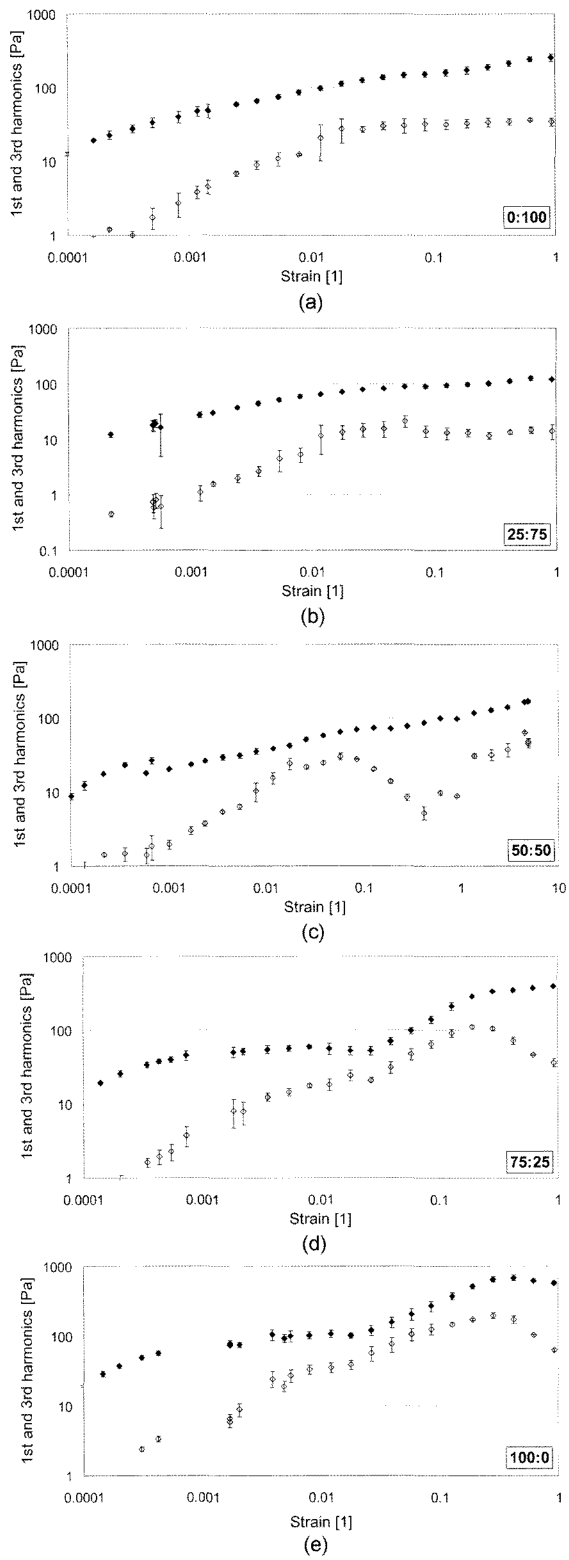

Fig. 3. (a-e). 1st and 3rd harmonics for the different compositions of bidisperse systems plotted as functions of applied strain. The filled symbols denote the 1st harmonic (I_1), while the open symbols denote the 3rd harmonics (I_3). The proportions of small to large particles in each figure are (a) 0:100; (b) 25:75; (c) 50:50; (d) 75:25; (e) 100:0.

can be seen that the results shown are very reproducible.

Now we consider the effects of bidispersity on the results shown in the non-linear regime in Fig. 3(a-e). The key quantity in this analysis would be the ratio (I_3/I_1) of the 3rd harmonic (I_3) to the 1st harmonic (I_1), and this corresponds to the proximity between the two curves on the logarithmic axis of the harmonics (since the difference between the two curves is equal to $\log(I_3)-\log(I_1)=\log(I_3/I_1)$). From Fig. 3(a-e), it is clear that the proximity of the 3rd harmonic to the 1st harmonic becomes more pronounced (i.e. I_3/I_1 is larger) as the proportion of small particles increases, and this proximity occurs for strains in the range 0.01-0.1. As the behaviour of a field induced material in the non-linear regime is believed to result from the continuous break down and reformation of clusters or structures which percolate through the system (Sim *et al.*, 2003; Parthasarathy and Klingenberg, 1995), it may be surmised that the behaviour of the 3rd harmonic depicts this mechanism, and this seems to be more likely to be happening in the monodisperse 100:0 (all small) system, and least likely to be happening in the monodisperse 0:100 (all large) system. The physical mechanism behind this is unclear at present. In an attempt to gain a better picture of the behaviour of the 3rd harmonic for these IFF, we may plot the phase angles for the different bidisperse systems as functions of strain amplitude. Here the phase angle is that of the fundamental of the stress signal which results from the input sinusoidal strain. This is done in Fig. 4 for the two cases of the monodisperse small and large particle system.

The fundamental phase angle can be used to describe the transition of the material from solid-like to viscous-like: when the material is predominantly elastic/solid-like, the phase angle is low; when the material is predominantly viscous, the phase angle is high; and at intermediate values, the material shows transitory behaviour between the two states. Thus, as expected, the profiles in Fig. 4 show that both systems move from being elastic-like materials at low strains to being viscous-like materials at higher strains, and this also agrees with the fact that these systems were observed to begin yielding at very small strains in Fig. 2. However, there is an obvious difference in the transition behaviours of the phase angles of both materials. The 0:100 system (all large) shows a smooth monotonic transition over the entire range of strains. On the other hand, for the 100:0 system (all small), at strains in the range 2×10^{-3}–0.04 the phase angle appears to decrease slightly, and then starts increasing again at strains >0.04. Looking back to Fig. 3, it is within this same strain range that the proximity of the 3rd and 1st harmonics is most pronounced. Interestingly, the phenomenon of decreasing phase angle with strain was equally noticeable in the 50:50 and 75:25 systems as well over the same strain range, while the 25:75 particle system had a monotonically increasing (but lower) slope, similar to the 0:100 system.

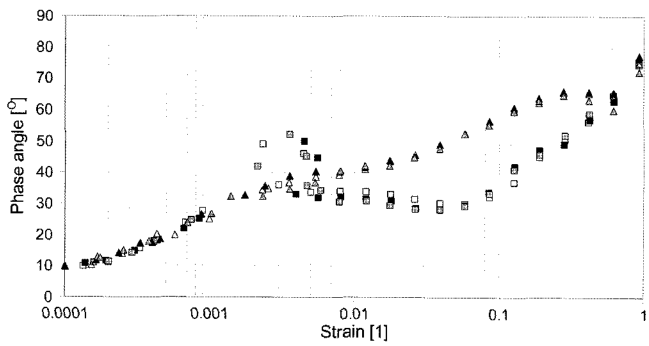

Fig. 4. Comparing the behaviour of the fundamental phase angles as a function of applied strain for the 100:0 (all small) and 0:100 (all large) IFF systems. The square symbols denote the 100:0 system (all small), while the triangles denote the 0:100 system (all large).

The behaviour observed in Fig. 3 (for the harmonics) and Fig. 4 (for the phase angle) for these systems is obviously size dependent, and may possibly be attributed to the manner in which breakage occurs in these systems. For the materials which show a monotonic increase in phase angle as well as small values of I_3/I_1 (0:100 and 25:75 particles), it is likely that the finite size of the gap relative to the size of the particles may lead to a more continuous and smooth breakage of the aggregates within the system during each cycle throughout the ramping of the strain amplitude. As a result, the system retains its sinusoidal nature throughout each cycle, but there is a smooth shift to more viscous-like behaviour (as shown by the steady increase in the phase angle) as the strain amplitude is increased.

On the other hand, the behaviour observed for the smaller particle systems which show larger I_3/I_1 (in Fig. 3), and the dip or plateau in the phase angle profile (Fig. 4) may be due to the fact that, over the corresponding range of strain amplitudes, the aggregates break and reform significantly through each cycle, and this leads to the increased non-sinusoidal shape of the stress signal. These signals would be expected to show larger I_3/I_1 and have more complex phase angle behaviour as a function of strain amplitude. It should be noted that similar non-sinusoidal waveforms have been reported by Li *et al.* (2003) for MRF, and by See *et al.* (1999) for ERF under oscillatory squeeze flow. The return to a waveform close to sinusoidal at even higher strain amplitudes for these systems reflects the fact that the aggregates exist in the ruptured or broken up state for a large part of each cycle, and hence the response is largely viscous in nature, with corresponding lower values of I_3/I_1 and phase angles approaching 90°, as seen in Figs. 3e and 4.

4. Conclusions

We have investigated the effect of particle size distribution on IFF in the linear and nonlinear viscoelastic regimes by mixing two monodisperse powders with different particle sizes, and subjecting this mixture to oscillatory shear flow. In the small strain linear regime, the results show that the monodisperse small particles had the highest storage modulus, while the 25:75 mixture of small to large particles had the lowest storage modulus. These results suggest that the stiffness of IFF under a magnetic field is very much dependent on the particle size (for monodisperse cases) as well as the ratio of particle sizes (for the bidisperse systems). The difference between these results and those of Saldivar *et al.* (2006) is most likely due to the differences in size ratios of particles used.

The results from the Fourier analysis obtained from the oscillatory tests at larger strain amplitudes also highlight the importance of particle size and size distribution for IFF. The ratio of 3^{rd} to 1^{st} harmonics was found to be least pronounced for the monodisperse large particles, and most pronounced for the monodisperse small particles. This ratio also increased with the proportion of small particles in the bidisperse system. Furthermore, the phase angles seemed to show that the transition of the IFF from solid-like to liquid-like behaviour was accompanied by a shift from smooth to more significant breakage and reformation of the aggregates as the particle size and size ratio decreased.

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References

Block, H. and J.P. Kelly, 1988, 'Electro-Rheology', *J. Phys, D: Applied Physics* **21**, 1661-1677.

Bombard, A.J.F., M. Knobel, M.R. Alcantara and I. Joekes, 2002, Evaluation of magnetorheological suspensions based on carbonyl iron powders, *J. Int. Mat. Syst. Struct.* **13**, 471-478.

Bossis, G. and E. Lemaire, 1991, Yield stresses in magnetic suspensions, *J. Rheol.* **35**, 1345-1354.

Claracq, J., J. Sarrazin and J. Montfort, 2004, Viscoelastic properties of magnetorheological fluids, *Rheol. Acta* **43**, 38-49.

De Gans, B.J., C. Blom, A.P. Philipse and J. Mellema, 1999, Linear viscoelasticity of an inverse ferrofluid, *Phys. Rev.* E **60**, 4518-4527.

De Gans, B.J., N. Duin, D. Van den Ende and J. Mellema, 2000, The influence of particle size on the magnetorheological properties of an inverse ferrofluid, *J. Chem. Phys.* **113**, 2032-2042.

Ekwebelam, C.C. and H. See, 2006, Determining the flow curves of an inverse ferrofluid (submitted).

Gamota, D.R. and F.E. Filisko, 1991(a), Dynamic mechanical studies of electrorheological materials: moderate frequencies, *J. Rheol.* **35**, 399-425.

Gamota, D.R. and F.E. Filisko, 1991(b), High frequency dynamic mechanical study of an aluminosilicate electrorheological material, *J. Rheol.* **35**, 1411-1425.

Gamota, D.R. and F.E. Filisko, 1991(c), Linear/non-linear mechanical properties of electrorheological materials, in Tao R. (ed), Proc. Int. Conf. on ERFs, World Scientific Publishing Co., New Jersey, 246-263.

Gamota, D.R., A.S. Wineman and F.E. Filisco, 1993, Fourier transfer analysis: nonlinear dynamic response of an electrorheological material, *J. Rheol.* **37**, 919-933.

Iyengar, V.R. and A.A. Alexandridis, 2004, Wear testing of seals in magneto-rheological fluids, *Tribology Transactions* **47**, 23-28.

Kittipoomwong, D. and D.J. Klingenberg, 2005, Dynamic yield stress enhancement in bidisperse magnetorheological fluids, *J. Rheol.* **49**, 1521-1538.

Klingenberg, D.J., 2001, Magnetorheology: Applications and Challenges, *AIChE J.* **47**, 246-249.

Kordonski, W.I. and D. Golini, 2000, Fundamentals of magnetorheological fluid utilization in high precision finishing, *J. Int. Mat. Syst. Struct.* **10(9)**, 83-89.

Lemaire, E., A. Meunier and G. Bossis, 1995, Influence of the particle size on the rheology of magnetorheological fluids, *J. Rheol.* **39**, 1011-1020.

Li, W.H., H. Du, G. Chen, S.H. Yeo and N. Guo, 2003, Nonlinear viscoelastic properties of MR fluids under large-amplitude-oscillatory-shear, *Rheol. Acta* **42**, 280-286.

Lim, S.T., M.S. Cho, I.B. Jang and H.J. Choi, 2004, Magnetorheological characterization of carbonyl iron based suspension stabilized by fumed silica, *J. Magn. Magn. Mat.* **282**, 170-173.

Magnac, G., P. Meneroud, M.F. Six, G. Patient, R. Leletty and F. Claeyssen, 2006, Characterisation of magneto-rheological fluids for actuators applications, ACTUATOR 2006, 10th International Conference on New Actuators, Bremen, Germany, 856-859.

Ota, M. and T. Miyamoto, 1994, Optimum particle size distribution of an electrorheological fluid, *J. Appl. Phys.* **76**, 5528-5532.

Otsubo, Y., M. Sekine and M. Katayama, 1992, Electrorheological properties of silica suspensions, *J. Rheol.* **36**, 479-496.

Parthsasrathy, M. and D.J. Klingenberg, 1995, A microstructural investigation of the nonlinear response of electrorheological suspensions (II): Oscillatory shear flow, *Rheol. Acta* **34**, 430-439.

Parthasarathy, M. and D.J. Klingenberg, 1996, Electrorheology: Mechanisms and Models, *Mater. Sci. Eng.* **R17**, 57-103.

Parthasarathy, M. and D.J. Klingenberg, 1999, Large amplitude oscillatory shear of ER fluids, *J. Non-Newt. Fluid Mech.* **81**, 83-104.

Popplewell, J., R.E. Rosenweig and J.K. Siller, 1995, Magnetorheology of ferrofluid composites, *J. Magn. Magn. Mat.* **149**, 53-56.

Popplewell, J. and R.E. Rosenweig, 1996, Magnetorheological fluid composites, *J. Phys, D: Applied Physics* **29**, 2297-2303.

Saldivar-Guerrero, R. Richter, I. Rehberg, N. Aksel, L. Heymann and O.S. Rodriguez-Fernandez, 2005, Liquid to solid transition of inverse ferrofluids, *Magnetohydrodynamics* **41**, 385-390.

Saldivar-Guerrero, R. Richter, I. Rehberg, N. Aksel, L. Heymann and O.S. Rodriguez-Fernandez, 2006, Viscoelasticity of mono- and polydisperse inverse ferrofluids, *J. Chem. Phys* **125**, (084907) 1-7.

See, H., 1999, Advances in modeling the mechanisms and rheology of electrorheological fluids, *Korea-Australia Rheol. J.* **11**, 169-195.

See, H., J.S. Field and B. Pfister, 1999, The response of electrorheological fluids under oscillatory squeeze flow, *J. Non-Newt. Fluid Mech* **84**, 149-158.

See, H., A. Kawai and F. Ikazaki, 2002, The effect of mixing particles of different size on the electrorheological response under steady shear flow, *Rheol. Acta* **41**, 55-60.

See, H. and R. Chen, 2004, The behaviour of a field responsive fluid under shear start-up, *Rheol. Acta* **43**, 175-179.

See, H., R. Chen, and M. Keentok, 2004, The creep behaviour of a field responsive fluid, *Colloid Polym. Sci.* **282**, 423-428.

Shih, Y. H. and H. Conrad, 1994, Influence of particle size on the dynamic strength of electrorheological fluids, *Int. J. Mod. Phys. B* **8**, 2835-2844.

Sim, H.G., K.H. Ahn and S.J. Lee, 2003, Three dimensional dynamics simulation of electrorheological fluids under large amplitude oscillatory shear flow, *J. Rheol.* **47(4)**, 879-895.

Skjeltorp, A.T., 1983, One and Two Dimensional Crystallization of Magnetic Holes, *Phys Rev Lett.* **51**, 2306-2309.

Stanway, R., J.L. Sproston and A.K. El-Wahed, 1996, Applications of electro-rheological fluids in vibration control: a survey, *Smart Mater. Struct* **5**, 464-482.

Trendler, A.M. and H. Böse, 2004, Influence of particle size on the rheological properties of magnetorheological suspensions, in Lu K., Shen R. and Liu J. (eds), Proc. 9th Int. Conf. of Electrorheological Fluids and Magnetorheological Suspensions, Beijing, China World Scientific, Singapore, 433-438.

Volkova, O., G. Bossis, M. Guyot, V. Bashtovoi and A. Reks, 2000, Magnetorheology of magnetic holes compared to magnetic particles, *J. Rheol.* **44**, 91-104.

Weiss, K.D., J.D. Carlson and D.A. Nixon, 1994, Viscoelastic properties of magneto- and electro-rheological fluids, *J. Int. Mat. Syst. Struct.* **5**, 772-775.

Wereley, N.M., A. Chaudhuri, J.H. Yoo, S. John, S. Kotha, A. Suggs, R. Radhakrishnan, B.J. Love and T.S. Sudarshan, 2006, Bidisperse magnetorheological fluids using Fe particles at nanometer and micron scale, *J. Int. Mat. Syst. Struct.* **17**, 393-401.

Wilhelm, M., D. Maring and H.W. Spiess, 1995, Fourier-transform rheology, *Rheol. Acta* **37**, 399-405.

Wu, C.W. and H. Conrad, 1998, Influence of mixed particle size on electrorheological response, *J. Appl. Phys.* **83(7)**, 3880-3884.