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Determining the flow curves for an inverse ferrofluid

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Abstract

An inverse ferrofluid composed of micron sized polymethylmethacrylate particles dispersed in ferrofluid was used to investigate the effects of test duration times on determining the flow curves of these materials under constant magnetic field. The results showed that flow curves determined using low duration times were most likely not measuring the steady state rheological response. However, at longer duration times, which are expected to correspond more to steady state behaviour, we noticed the occurrence of plateau and decreasing flow curves in the shear rate range of 0.004 s^{-1} to $\sim$20 s^{-1}, which suggest the presence of non-homogeneities and shear localization in the material. This behaviour was also reflected in the steady state results from shear start up tests performed over the same range of shear rates. The results indicate that care is required when interpreting flow curves obtained for inverse ferrofluids.

Keywords : inverse ferrofluid, flow curve, shear localization

1. Introduction

Field-responsive fluids are suspensions of micron-sized particles which show a dramatic increase in flow resistance upon application of external magnetic or electrical fields. The two main types are electrorheological fluids (ERF) which respond under electric fields and magnetorheological fluids (MRF) which respond under magnetic fields. In both cases, the reason for this behaviour is that application of the external field leads to the formation of field-aligned dipoles within each particle. The subsequent dipole-dipole interactions between the particles then leads to the formation of elongated aggregates or clusters in the direction of the applied field, and this in turn brings about the increase in the flow resistance. There are review articles available providing more details on these physical mechanisms and materials (Block and Kelly, 1988; Parthasarathy and Klingenberg, 1996; See, 1999). The tunable flow properties of these fluids lend themselves to a variety of engineering applications, including damping and high precision polishing devices (Klingenberg, 2001; Magnac *et al.*, 2006; Stanway *et al.*, 1996; Bombard *et al.*, 2002).

As a model of these suspensions, the "inverse ferrofluid" (abbreviated to IFF), has recently been receiving some attention (Skjeltorp, 1983; Bossis and Lemaire, 1991; Popplewell *et al.*, 1995; de Gans *et al.*, 1999a, 1999b, 2000; Volkova *et al.*, 2000; Saldivar-Guerrero *et al.*, 2005; Ekwebelam and See, 2007), and will be the focus of this work. An IFF comprises a magnetisable carrier liquid, otherwise known as a ferrofluid, which is a colloidal dispersion of nano-sized magnetite particles. Into this we disperse micron-sized non-magnetic solid particles, thereby producing an IFF. The difference in particle size between the larger non-magnetic particles and those comprising the ferrofluid carrier means that the larger particles would behave as if they were in a continuum. Under a magnetic field, the non-magnetic particles are observed to form chains in the field direction (Skjeltorp, 1983), and this is due to essentially the same induced dipolar interactions as arises in ERF and MRF. Due to these interactions and structures, IFF show a field-induced increase in shear resistance analogous to ERF and MRF (Bossis and Lemaire, 1991; Popplewell *et al.*, 1995; de Gans *et al.*, 1999a, 1999b, 2000; Volkova *et al.*, 2000; Saldivar-Guerrero *et al.*, 2005; Ekwebelam and See, 2007). A significant advantage of this kind of system is that it allows us to probe the effects of using a wide range of particle sizes and geometries-these studies will be reported in future. This work is an attempt to establish the basic rheological behaviour of these systems, and so we have focused on the simplest IFF comprising monodisperse spherical particles in a ferrofluid and examined the response under steady shear flow.

In general, the rheological behaviour under steady shearing of many materials is described by a plot of shear stress versus shear rate, otherwise known as a flow curve. For MRF and ERF it is found that the flow curves under an external field will take the form of a 'Bingham fluid' described by the following equation:

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$$\tau = \tau_y + \eta_p \dot{\gamma} \quad (1)$$

Here τ is the instantaneous stress (units of Pa), τ_y is the yield stress (Pa), η_p is the plastic viscosity (Pa·s), and $\dot{\gamma}$ is the applied shear rate (s^{-1}). That is, these materials show a yield stress, corresponding to the minimum shear stress necessary to rupture the aggregates produced under the given applied field. Numerous examples of flow curves exist in literature for ERF (Klingenberg and Zukoski, 1990; Park and Park, 2001; Sung *et al.*, 2004; Park *et al.*, 2005; Espin *et al.*, 2006) and MRF (Bombard *et al.*, 2002; Ashour *et al.*, 1996; Rankin *et al.*, 1999; Stanway, 2004; Claracq *et al.*, 2004; Choi *et al.*, 2005; Wereley *et al.*, 2006). In all of these works, curves similar to the Bingham fluid curve were observed. It should also be noted that for flow curves to provide unambiguous characterization of the rheological properties of a fluid, they ideally should be measuring the response of the material at steady state at each shear rate (Coussot, 2005).

The rheological behaviour under steady shearing of IFF under magnetic fields has been reported by several workers, including Popplewell *et al.* (1995), de Gans *et al.* (1999a, 1999b, 2000), and Saldivar-Guerrero *et al.* (2005). For example, de Gans *et al.* (2000) determined the flow curves of an IFF composed of nanometer-sized non-magnetic particles, but their small size means that they would be susceptible to considerable thermal motion effects, and hence the behaviour would be expected to differ from the micron-sized systems we study in this work. Popplewell *et al.* (1995) explored the effect of shear rates of up to 400 s^{-1} on an IFF which consisted of composites of hollow glass beads dispersed in a ferrofluid. Saldivar-Guerrero *et al.* (2005) also explored the field dependence of polystyrene particles dispersed in a ferrofluid under steady shear flow. These workers all report field-induced Bingham-type behaviour in their flow curves. However, none of these workers have reported the time allowed for the test, despite this being an essential consideration when interpreting flow curves (Coussot, 2005).

To date, there seem to be no reports which explore in detail whether the flow curve measurements for IFF based on micron-sized particles, are indeed being taken at steady state. It is expected that the system will approach steady state as we increase the duration time of the test. *The main point of this work, therefore, will be to explore the effects of the duration times (or equivalently, the ramp-up rate of the shear rate) used for determining the flow curves of IFF.*

In an earlier paper (Ekwebelam and See, 2007; hereinafter referred to as Paper 1), we reported on the behaviour of essentially the same IFF system under oscillatory shear flow, with particular focus on the effects of mixing two particle sizes. This present paper extends our investigations of this IFF system to the behaviour under steady shear flow, particularly as large strains are reached.

The structure of this paper is as follows: the materials, experimental methods and procedures used are described next; the tests carried out are flow curve and shear start up tests. In the following section we display the results from these two rheological tests, and discuss the implications of these results. The results are plotted in the form of 'shear stress versus shear rate' (*i.e.*, flow curves) and 'shear stress versus shear strain' curves. The paper ends with a conclusion section.

2. Experimental

2.1. Material

The ferrofluid was supplied by Ferrotec Inc. USA, (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} Pas at 27°C, a density of 1.21 g/ml, and a remanent magnetism of 400 gauss. Into this ferrofluid we dispersed polymethylmethacrylate (PMMA) spherical particles with an average particle size of 4.6 μm, to a volume fraction of 30%, thereby making up the inverse ferrofluid (IFF). This ferrofluid and the PMMA were used in Paper 1 for studies on the yielding behaviour of bidisperse mixtures of IFF (Ekwebelam and See, 2007). It is noted that the density of the PMMA particles is less than that of the disperse phase (~1.08 g/cc), and as such, there is the potential for flotation of the particles in suspension. However, the IFF was observed under a microscope before and after each test, and showed no apparent signs of flotation. Further, there was also no apparent sign of any chemical reaction having taken place.

2.2. Equipment

A Paar Physica MCR300 rheometer was used with the 20MR magnetorheological cell. This employs parallel plates of diameter 20 mm, and a constant gap of 1 mm was used. 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 soft

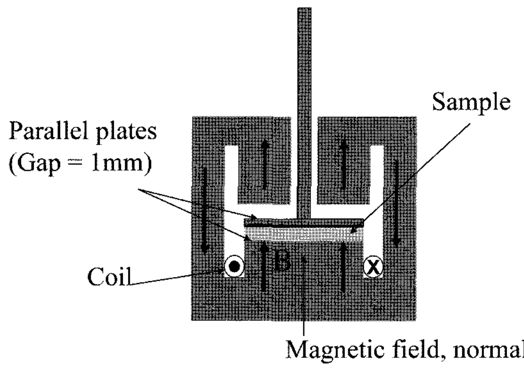

Fig. 1. Schematic diagram of the magnetic rheological test cell (Anton Paar Physica MCR300).

iron elements to create a magnetic circuit. This test cell has been used in several previous studies on MRF (See *et al.*, 2002a; See and Chen, 2004; See, 2004) and was also used in Paper 1 to study the yielding behaviour of IFF (Ekwebelam and See, 2007). Fig. 1 shows a schematic of the experimental set up. For all tests performed the temperature was kept constant at approximately 25°C.

2.3. Rheological test conditions

Test 1: Flow curve test

The sample was initially subjected to a shear rate of 20 s^{-1} for 10 s, with no magnetic field applied, to ensure the break up of any aggregates which may have pre-formed. After this, a field with magnetic flux density of 0.54 T, the highest which can be obtained with this system, was applied to the sample for a period of 120 s in the absence of shearing to promote the formation of aggregates. Following this, shear rates ranging from 0.001 to 100 s^{-1} were applied to the sample, with a logarithmic increase over a period of 10 minutes (this period was varied). From this test, the instantaneous shear stress was plotted as a function of the applied shear rate in order to generate the flow curves. It should be noted that tests performed over a duration of several minutes are typical of flow curve measurements on complex micro-structured materials (see for example p 96 of Coussot, 2005; p 70 of Brummer, 2006).

The speed at which the shear rate is increased when obtaining the flow curves is an important quantity here. This speed will be expressed as the "*ramp-up rate*", defined as the "*number of orders of magnitude of shear rate per unit time*" (note that the shear rate is increased logarithmically in our tests, so it is appropriate to speak of "orders of magnitude per unit time"). For example, the 10 min duration for the 0.001 to 100 s^{-1} test described previously corresponds to a ramp-up rate of 0.5 orders of magnitude/min (*i.e.*, 5 orders of magnitude of shear over 10 min). For brevity, we will henceforth write this as 0.5 order_mag/min. In addition to the 0.5 order_mag/min test, flow curves tests were carried out at the same flux density of 0.54T for ramp-up rates of 0.24 order_mag/min, 0.17 order_mag/min, and 0.014 order_mag/min.

In addition to these flow curve tests, further useful information can also be extracted from a shear start-up test which was also carried out, as described next.

Test 2: Shear start up test

Essentially, similar conditions to those used for the flow curve test prior to the application of the magnetic field were used for this test. Following the application of the field, a constant shear rate was applied to the sample and the shear stress was measured as a function of the strain on the sample. The attainment of steady state for the test would be indicated by a plateau of the stress-strain curve. The shear rates used were 0.003, 0.01, 0.1, 1, and 10 s^{-1}. For reproducibility of each individual test, two repeat experiments were carried out at each constant shear rate, making a total of three test results altogether. It should be noted that a similar test was reported by de Gans (2000), but for the case of nano-sized non-magnetic particles and under only one shear rate of 0.0103 s^{-1}.

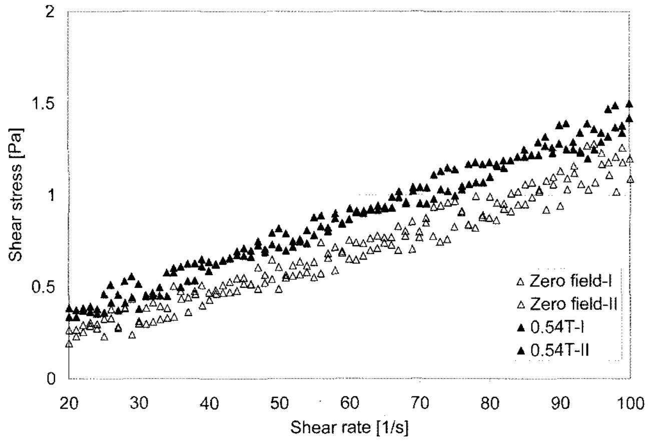

Fig. 2. Typical flow curves for the ferrofluid tested under conditions of zero field (0 T) and under a flux density of 0.54 T. The curves show two tests carried out on the same sample under each condition. The ramp-up rate used was 0.5 order_mag/min.

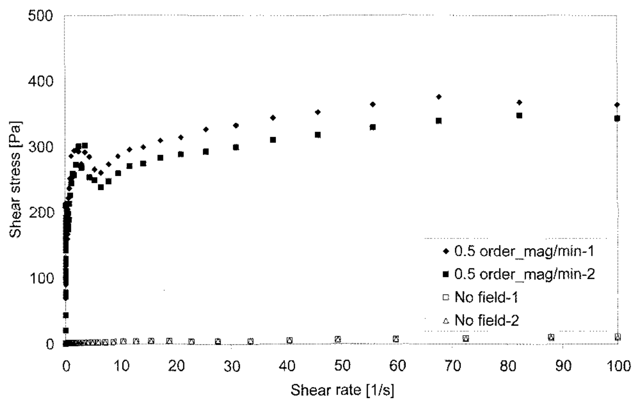

Fig. 3. Typical flow curves for IFF tested under conditions of no field (0 T) and under a flux density of 0.54 T. The curves show two tests carried out on the same sample under each condition. The ramp-up rate was 0.5 order_mag/min.

3. Results and Discussion

3.1. Flow curves for 0.5 order_mag/min ramp-up rate

Figs. 2 and 3 show typical flow curves for the ramp-up rate of 0.5 order_mag/min for the ferrofluid and for the IFF respectively. In both figures, the shear stress is plotted as a function of the applied shear rate, and it is clear from both figures that the results are quite reproducible. In both cases, data points are plotted for the case of no applied field as well, for comparison.

In Fig. 2 the shear rate axis is plotted from 20 s^{-1}, as values of shear stress below this shear rate were found to approach the resolution of the MCR300 rheometer. The figure shows that there is an almost negligible increase in the shear stress of the ferrofluid as the applied field is increased from 0 T to 0.54 T, thereby suggesting that the applied field has only the slightest effect on the rheological response of the ferrofluid. The stress magnitudes in Fig 2 are significantly smaller than those of the inverse ferrofluid.

Turning now to the behaviour of the IFF shown in Fig. 3, it can be seen that the material does indeed respond similarly to conventional MRF in that there is definitely a very significant field-induced increase in the shear stress values. Furthermore, the curve displays Bingham-fluid type behaviour, as expected for an inverse ferrofluid. Using the standard method of extrapolating to the shear stress (y-axis), as required by the Bingham model (Eq. 1), the yield stress was found to be in the range 200-300 Pa (in carrying out the extrapolation, we included the data values generated at the highest shear rates used for the test). At very low shear rates, there is a clear decrease in the shear stress values (as can be seen for the data points on the left side of Fig. 3) and if just these points were extrapolated the "yield stress" would take a lower value. This issue will be explored next.

It is known that many materials which appear to show a yield stress actually show quite complex flow behaviour at low shear rates, and this has been highlighted in a series of papers by Barnes and Walters (1985) and Barnes (1999). Along these lines, we explore the low shear rate behaviour of this IFF by transforming the shear rate axis from a linear to logarithmic axis, as seen in Fig. 4. The advantage of this transformation is that it highlights any behaviour of the curve which might otherwise be masked by the linear plot, specifically the low shear rate behaviour. Interestingly, Fig. 4 shows that there is a steady increase in the shear stress over the entire range of the applied shear rates, with the stress values starting at a non-zero value and increasing monotonically with shear rate.

From Fig. 4, it can be seen that the behaviour of this material suggests that it may not actually have a yield stress, and so it could possibly fit into the class of materials described by Barnes and Walters (1985) as flowing at all shear rates, which is different to the strict definition of a Bingham fluid. The fact that the IFF shows this behaviour is not surprising, as essentially it consists of particles and aggregates which, although held together by field-induced forces, are still able to break and slide over each other to some extent once bonds are ruptured.

The behaviour shown in the two figures above are for the ramp-up rate of 0.5 decades/min. However, different behaviour to this was found for the other ramp-up rates explored. The results for the lowest case of 0.014 order_mag /min are described next.

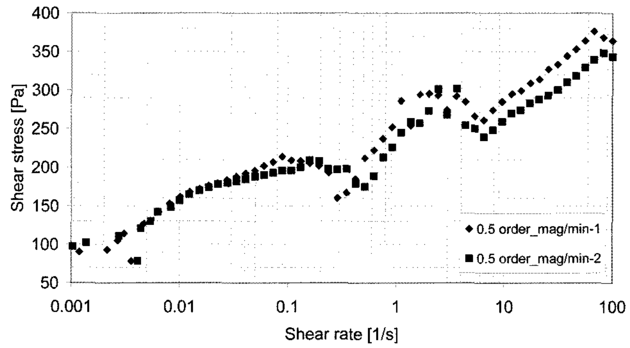

Fig. 4. Logarithmic plot of shear stress as a function of shear rate for IFF under an applied magnetic flux density of 0.54 T. The ramp-up rate was 0.5 order_mag/min.

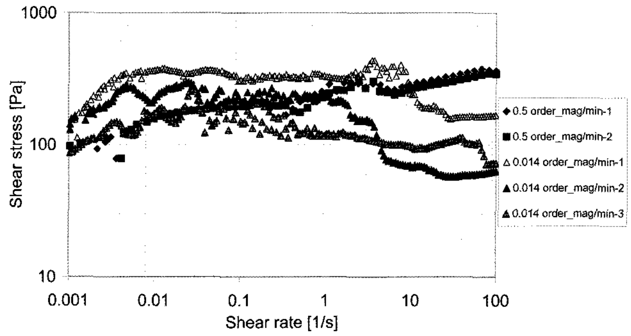

Fig. 5. Comparing the flow curves for an IFF determined using ramp-up rates of 0.5 order_mag/min and 0.014 order_mag/min. The magnetic flux density was 0.54 T for all tests.

3.2. Effect of varying the 'ramp-up rate' on the flow curves – the case of a ramp-up rate of 0.014 order_mag/minute.

To probe the effect of varying the ramp-up rate on the flow curve, the same test was carried out using a ramp-up rate of 0.014 order_mag/minute. Fig. 5 shows the resulting behaviour of the sample, with the results obtained previously for the higher ramp-up rate of 0.5 order_mag/min also shown for comparison.

As seen from Fig. 5, there is a very significant difference in the behaviour of the sample under the two ramp-up rates. As stated earlier, there is a continuous increase in the shear stress with shear rate for the ramp-up rate of 0.5 order_mag/min. However, when the ramp-up rate is reduced to 0.014 order_mag/min, the shear stress initially appears to increase at very low shear rates, and then continuously fluctuates between 100 and 400 Pa over 2-3 orders of magnitude of shear rate. This is followed by a reduction in the shear stress when the shear rate is further increased in the range 0.1 s^{-1}-20 s^{-1}. When the shear rates exceed ~20 s^{-1} the curve shows a tendency to plateau and remain constant but at significantly lower values than those prior to the dip. These trends can be seen for all three curves obtained at 0.014 order_mag/min ramp-up rate in Fig. 5, although there is some variation in the stress values as well as in the onset of the different phases as shear rate is increased. There is clear contrast with the 0.5 order_mag/min case, with its monotonically increasing curves. The difference between the 0.5 order_mag/min case and the 0.014 order_mag/min case also indicates that the assumption that the system has reached steady state in the former needs to be carefully considered. Certainly, it would seem that the 0.5 order_mag/min case is not a steady state measurement, but whether the 0.014 order_mag/min case actually corresponds to steady state or not is yet to be conclusively determined, although this is expected to be more likely than the 0.5 order_mag/min case. Indeed, the shear start up tests (reported in the next subsection) were carried out to shed light on this. In any case, as pointed out earlier, the 0.5 order_mag/min test has a total duration of 10min and is similar to the tests which have been commonly used to date for IFF, but our results would seem to indicate that these are unlikely to have been at steady state.

The observed shape of the flow curves for the shear rate of 0.014 order_mag/min, with the possible plateau and clearly decreasing shear stress with increasing shear rate, is an unusual rheological phenomenon. Indeed, it is known that a decreasing flow curve cannot occur for a homogeneous material in a shear test (Coussot, 2005; Møeller *et al.*, 2006). These regions of the flow curve are believed to correspond to the presence of instabilities within the system, and these may take the form of shear localization, where the shear rate is concentrated in a layer of the sample (Coussot, 2005; Møeller *et al.*, 2006). The phenomenon of decreasing flow curves has been reported by a significant number of authors working with ERF under dc fields, including Schubring and Filisko (1995), Kawakami *et al.* (1999), Aizawa *et al.* (2000), and See *et al.* (2002b). See *et al.* (2002b) in particular compared the behaviour of an ERF composed of sulfonated polystyrene particles dispersed in silicone oil in both d.c. and a.c. fields, and found that while the shear stress decreased with increasing shear rate for the case of the d.c. field, for the case of the a.c. field the flow curves behaved as was expected *i.e.* increasing shear stress with increasing shear rate. To the best of our knowledge, this is the first time either plateau flow curves or decreasing flow curves have been reported for an IFF composed of micron-sized particles, but it is presently not clear what the nature of the non-homogeneities would be in these flow regimes. Obviously, further work is necessary to understand the mechanisms behind this behaviour. It should also be noted that de Gans *et al.* (2000) indicate the possibility of shear-induced non-homogeneities in their nanometer-sized dispersions but do not probe this possibility in any detail. Thus it is possible that this phenomenon occurs over a range of particle sizes in IFF.

Note that the flow curve test was also carried out using ramp-up rates of 0.25 order_mag/min and 0.17 order_mag/min. The same trend was seen in the case of 0.25 order_mag/min as was seen for 0.5 order_mag/min *i.e.* the monotonically increasing shear stress with shear rate as shown Fig. 4. The 0.17 order_mag/min case, on the other hand, closely resembled the behaviour observed for the case of 0.014 order_mag/min in Fig. 5.

These results indicate that even when tests are performed at low ramp-up rates with the aim of probing the steady shear behaviour of these systems, the flow curves obtained need to be regarded with considerable caution, as there is the possibility that the material is responding in a non-homogeneous fashion. Hence the flow curve alone does not necessarily provide a useful and unambiguous characterization.

In addition to the flow curve tests, we carried out shear start up tests on the same IFF in order to further explore the shear response of the material as a function of test duration. The advantage of this test is that the onset of steady state for the test is marked by a leveling out of the stress-strain curve *i.e.* the stress curve reaches a plateau with increasing strain, and so the steady state shear stress may be determined from this plateau value. These results are presented next.

3.3. Shear start up tests

Figs. 6 and 7 show the results obtained from plotting the shear stress as a function of the strain for different values of applied shear rates. All tests were performed under the constant magnetic flux density of 0.54 T.

Fig. 6 shows that for all the low shear rates, there is an initial increase in shear stress with strain (corresponding to an elastic or solid-like response), then a plateau region

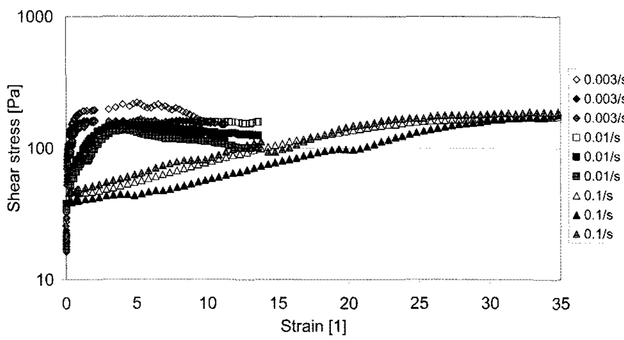

Fig. 6. Curves obtained for the shear start up test applied at low constant shear rates (values indicated in legend). Note that there is an overlap between some of the data points for the 0.003 and 0.01 s^{-1} shear rates. The magnetic flux density was 0.54 T for all tests.

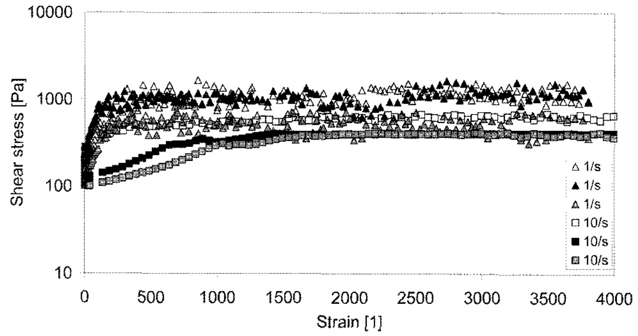

Fig. 7. Curves obtained for the shear start up test applied at higher constant shear rates. The magnetic flux density was 0.54 T for all tests.

where the yielded material undergoes steady flow at the imposed shear rate. The curves for the 0.1 s^{-1} case are intriguing in that they show a slow increase of stress with strain and this was found to be reproducible - the reason for this long strain transitional behaviour is unclear at present. The key feature of these curves which is of particular interest to this study is the final plateau values reached by the curves at large strains, as they are expected to correspond to "steady state" behavior. For the shear start up tests, we have deliberately left the raw data points on the plots so that the reader can get an idea of the reproducibility.

Fig. 7 shows that for the higher shear rates of 1 s^{-1} and 10 s^{-1}, we again see the same solid-like pre-yield behaviour, followed by a transition region where the curves plateau and the IFF starts behaving more viscous-like.

We now consider Figs. 6 and 7 in light of the behaviour observed in the flow curves *i.e.* for low and high ramp-up rates for the flow curve test (Fig. 5). In Fig. 6, the stress-strain curve can be seen to reach a plateau at stresses between 100 and 200 Pa for the lower shear rates of 0.003, 0.01, and 0.1 s^{-1}. This value of shear stress is similar in magnitude to the stress values found in the flow curves for shear rates in the same range *i.e.* ~0.003-0.1 s^{-1} for the low ramp-up rate case (0.014 decades/min). However it should be noted here that while the magnitudes are roughly correct, there appears to be no order to the plateau values in Fig. 6 as a function of applied shear rate - that is, we would expect that the higher shear rates would be associated with higher values of stress, but this is not the case for this material.

Turning now to the shear rates of 1 and 10 s^{-1}, the plateau values of the shear stress in Fig. 7 are seen to fluctuate between 300 and 2000 Pa. Looking at the flow curves in Fig. 5 for the low ramp-up rate case of 0.014 decades/min, on the other hand, it can be seen that it is within this region of shear rates (*i.e.* between 1 s^{-1} and 20 s^{-1}) that the issue of decreasing flow curves becomes most pronounced. Thus we see that there is no agreement in the values of the shear stress between the two tests at the higher shear rates. Furthermore, in Fig. 7 we again observe, similar to the low shear rate cases seen in Fig. 6, that the plateau values of the shear stress are not in the expected order for the shear rates used *i.e.* we would expect the plateau value of the shear stress to increase with the applied shear rate. Indeed, from Fig. 7 it seems that the plateau values for the 10 s^{-1} shear rate are somewhat lower than those for 1 s^{-1}. Quite possibly this fact, as well as the relatively large fluctuations seen in the Fig 7 data, are reflecting the onset of the instabilities/non-homogeneities which were seen in the flow curves, although the stress values in the shear start up test overall are much higher than the corresponding ones in the flow curve test. The reason for this is unknown at present.

At this point we draw attention to a rheological fitting model recently developed by Cho *et al.* (2005), and which has subsequently been used to fit the behaviour of some polymer based ERF materials which show decreasing flow curves. While this model captures features of the behaviour of these materials, it does not necessarily fully explain the instabilities exhibited by these field responsive fluids, and the reasons behind these instabilities remain an on-going area of research.

4. Conclusions

The question of whether or not the characterization of the flow behaviour of a material has been performed under steady state conditions is a key one when interpreting rheometrical tests. Inverse ferrofluids containing micron-sized particles have been the object of some considerable attention in recent years as model systems for magneto- or electrorheology, but reports of the flow curves of these materials do not seem to have considered this question of steady state behaviour of the test. Thus, in this work we have explored this issue experimentally by using a range of ramp-up rates of the shear flow in a flow curve test under a constant magnetic field. The results seem to indicate that measurements at ramp-up rates comparable to those reported in literature are possibly not corresponding to steady state rheological behaviour of the material, and thus caution is advised when interpreting these results. Furthermore, at lower ramp-up rates (which would be expected to more closely correspond to steady state response), the material showed the unusual phenomena of plateau shear stresses as well as decreasing shear stresses with shear rate, suggesting the possible existence of non-homogeneities and shear localization within the sample. If this is the case, care must be exercised in extracting any rheological information from the flow curves. An additional complication emerging from the flow curve tests was that there did not appear to be a unique value of the "yield stress" in the sense that the material appeared to continue to flow even at very low shear rates - this is a phenomenon widely reported for materials which are often modeled, in a simplistic way, as following the Bingham flow curve with a fixed yield stress. The shear start-up tests carried out also reflected the complex behaviour of this material at low and higher shear rates, in particular via the plateau values of shear stresses observed at high strains. The question as to whether similar phenomena are observed in actual ERF and MRF systems is an important one, which could be the focus of future rheometrical studies. Furthermore, recent advances in the modeling approaches for these field-responsive suspensions will give us additional insights into the micro-structural mechanisms governing the complex rheological behaviour (for example, see the reviews Parthasarathy and Klingenberg (1996) and See (1999)) - these modeling studies are currently being carried out and will be reported in future publications. A particularly useful approach here is particle-level computer simulations, whereby the time evolution of a system of spherical particles, under an externally applied flow field and a magnetic field, is determined via numerical integration of the force balance equation for each particle.

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