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ORIGINAL CONTRIBUTION

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The creep behaviour of a field-responsive fluid

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Abstract Many particulate suspensions show a dramatic but reversible increase in flow resistance when subjected to an external magnetic or electric field, with the material often modeled as having a field-induced yield stress τ_y. Creep tests on a suspension of carbonyl iron particles dispersed in oil under a constant magnetic field reveal that the material will undergo deformation over a prolonged period, even if the applied shear stress is considerably smaller than τ_y. This indicates that the rheological behaviour is more complex than can be captured by the current simple constitutive formulations.

Keywords Field-responsive fluid · Particle suspension · Creep response · Linear viscoelasticity · Magneto-rheology

Introduction

Recent years have seen the development of a new class of engineering materials, the so-called ‘field-responsive fluids’, which display dramatic changes in mechanical properties when subject to an external electric or magnetic field: the corresponding fluids are often referred to as electro-rheological fluid (ERF) or magneto-rheological suspension (MRS), respectively. Both systems typically comprise of micron-sized solid particles dispersed in a carrier liquid. When the external field is applied, the following process is believed to take place: electric (magnetic) polarisation is induced in each particle in the ERF (MRS), due to the mismatch in polarisabilities between the particle material and the carrier liquid, and the resulting dipole-dipole interaction force between the particles leads to the formation of elongated aggregates aligned in the field direction. As illustrated in Fig. 1, the presence of these aggregates in the flow field gives rise to the increased resistance to flow. This tunable flow behaviour has many potential engineering applications, such as in active vibration damping systems. There are several review articles available now on these materials [2, 5, 6, 8, 9, 18, 20, 21, 22, 26].

The mechanical behaviour of these systems is typically evaluated in the shearing mode, with the external field applied normally to the shearing planes. The tests under steady shear are usually performed at moderate values of the shear rate, and it is often found that the variation of the shear stress τ with can be closely fitted by the Bingham fluid equation:

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

Here τ_y is the field-induced yield stress due to the presence of the particulate aggregates, and η_p is the plastic viscosity. Although extensive experimental verification of this behaviour under steady shearing has been reported for a range of electro-rheological fluids [3, 7] and magneto-rheological suspensions [1, 6, 19], these tests have typically tended to focus on the response at shear rates over 1 s^{-1}.

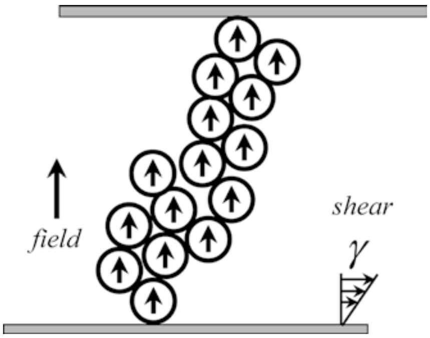

Fig. 1 Schematic diagram of a column of polarised particles formed under an external field. A shear stress τ_0 is applied to the column, and it deforms as shown (strain γ). In the unstrained state the column is aligned in the field direction

The creep behaviour of these materials, whereby the sample is subject to a constant (possibly small) shear stress while the strain is monitored, is likely to be more complex than described by this Bingham fluid model, since the latter simply assumes that the pre-yield material does not deform at all if the applied stress is less than the critical value τ_y. This paper will focus on the response of a magneto-rheological suspension in a creep test, and in particular will examine to what extent and over what timescales the sample will deform when a small shear stress is continually applied. It should also be noted that data on the behaviour under slow deformations are still not widely available in the literature for field-responsive fluids, and so this information will prove valuable in the ongoing effort toward the construction of a general constitutive framework for these materials.

Although creep tests have the potential to illuminate many of the mechanisms behind the flow behaviour of field responsive fluids, there have only been a few related reports in the literature. Otsubo and Edamura [17] studied the creep behaviour under dc electric fields of polymer composite particles dispersed in silicone oil. They found that, for a constant field of 1 kV/mm, under small shear stresses the material initially displayed an elastic response followed by a steady viscous flow, but when the applied stress was increased to 70 Pa the strain was observed to instantaneously jump to an equilibrium value without viscous flow at all, which is the opposite of the expected behaviour. At the still higher shear stress of 100 Pa, the strain was observed to increase with time in a stepwise fashion, suggesting that continual rupture and reformation of the structure under shearing was taking place. More recently, Li et al [13] have studied the creep behaviour of a commercially available magneto-rheological suspension under a fixed magnetic flux density of 0.34T. They found that under small applied stresses the sample would initially show an elastic response and then undergo a transition to a steadily flowing, viscous regime. The possibility that the response curves to different applied stresses may collapse to one curve corresponding to a linear viscoelastic material did not seem to be explored. This possibility and the field dependence of the creep behaviour are the issues focused upon in the present paper.

It is appropriate at this stage to briefly review the microstructure-based rheological models of these systems, which seek to account for the physical mechanisms beyond the simple Bingham fluid equation Eqn.1. In general, the models developed to date assume an idealised microstructure of the particle aggregates and use a simplified description of the interaction forces between particles. Review articles of advances in the theoretical modelling of these systems are available [18, 22]. Although the details of the particle interactions and configurations may differ, theoretical models based on idealised microstructures, such as particles arranged in linear chains or in a regular lattice, generally predict that the shear stress under small strains will vary linearly with the degree of deformation of the structures from their initial equilibrium state. Thus, if we focus on the creep response after the application of a constant stress τ_0, these models would predict that if τ_0 is small compared to the stress for yielding, there will be an almost instantaneous jump to the corresponding strain value (γ_0) with the system remaining motionless thereafter. As will be seen from the experimental results, the actual behaviour seems somewhat more complex than this.

This paper is organised as follows. In the next section, we describe the materials and apparatus used and the rheological measurement procedure. In the following section, we present and discuss the results of the series of creep tests. In the final section we give some concluding remarks.

Experimental

Materials

The magneto-rheological suspension was a commercial sample supplied by Fuchs-DEA Schmierstoffe GmbH & Co KG (Germany). It consisted of carbonyl iron powder of typical size 4–6μm, dispersed in a paraffinic spindle oil of kinematic viscosity 6.5 x10^{-6} m^2s^{-1}. The particle volume fraction was 30%. The sample bottle was vigorously shaken before placing the fluid in the rheometer. At the conclusion of each series of tests the samples were closely inspected, and no significant separation of the particles from the oil was observed.

The dependence of shear stress on shear rate (the "flow curves") for the same MRS under various magnetic flux densities has been reported in detail elsewhere [25]. To provide a brief summary here, we note that it was confirmed the sample showed the Bingham fluid-like response often reported for MRS under steady shearing [1, 6, 10, 11, 12]. Typical values of the shear stress under a magnetic flux density of 0.57T were 9700 Pa at 1 s^{-1}, and 12,050 Pa at 100 s^{-1}. Prior to the present series of tests, measurements of the shear stress under steady shear flow and a range of magnetic flux densities were carried out, and the results were confirmed to be close to those previously obtained.

Apparatus

The rheometer was an Anton Paar Physica MCR300, operating in the controlled stress mode. A detailed description of this set-up has been given in other publications [23, 24], and so just a brief overview will be provided here. Parallel plates of diameter 20 mm were used, which were specially constructed from a stainless steel of low magnetic permeability. For all shearing tests a gap of 1 mm was used. Following the standard convention, the shear strain and shear rate values quoted in this paper will be those at the outer radius of the plate, although it should be kept in mind that what is being measured is in fact a response averaged over the linearly varying shear field from the plate centre to its outer edge.

The magneto-rheological test cell attached to the rheometer was the commercially available Physica TEK 70-MR. This consisted of a unit under the bottom plate, which housed the lower half of a magnetic circuit constructed from iron elements. This unit also contained a coil of 495 windings through which a dc electric current was passed to generate the magnetic field. On the upper side, there was a removable cylindrical block which sat on the bottom stage and completely enclosed the two plates, with a central hole to pass the rheometer shaft. This block was also made up of iron elements and constituted the upper half of the magnetic circuit, thus enabling a uniform magnetic field to be applied perpendicularly across the plates. The magnetic flux density across the plates was controlled by the rheometer software which adjusted the electric current through the coil, taking into account the air gap immediately above the upper plate, and the characteristic permeabilities and dimensions of the other elements comprising the magnetic circuit. The maximum magnetic flux density across the plates achievable with this system was 0.57T. It should be noted that there is actually a lack of magnetic field immediately below the shaft, but this effect is not considered to be large since the shaft diameter is only 4 mm.

Further, it is pointed out that tests were performed on magnetically inert silicone oils to confirm that the application of the magnetic field in itself did not affect the rheometer's torque measurement system.

Rheological measurements

Prior to the rheological measurements, the samples were initially sheared without a magnetic field at a shear rate of 50 s^{-1} for 1 min to ensure the particles were uniformly dispersed. The shearing was then stopped and for 5 min the required magnetic field was applied across the plates with the sample subject to no deformation, so that the field-induced aggregates could form in the sample. To confirm that 5 min is sufficiently long for the structures to form, a series of measurements was undertaken whereby the steady shear viscosity at 1 s^{-1} was measured after different intervals of time following the application of the field. For all magnetic field strengths it was found that after 3 minutes, and often much earlier, the viscosities reached a steady state, which would indicate that the aggregate structure had fully formed. Thus the standard period of 5 min was chosen as the waiting time for aggregate formation. A detailed study of the transient response after application of a field is currently being carried out, and will reported in the future.

The creep measurements under various magnetic flux densities were carried out by applying a constant shear stress to the sample and monitoring the strain. A typical response to a creep test is illustrated schematically in Fig. 2: following an initial solid-like response, there is a transition region (sometimes called the "retarded elastic response"), and finally there is often a steady flow region at very long times. The transient behaviour after the removal of the applied stress at time t_{off} is also examined. If the material is responding in the linear viscoelastic regime, the quantity $\gamma(t)/\tau_0$ is generally known as the creep compliance $J(t)$, and ideally the data

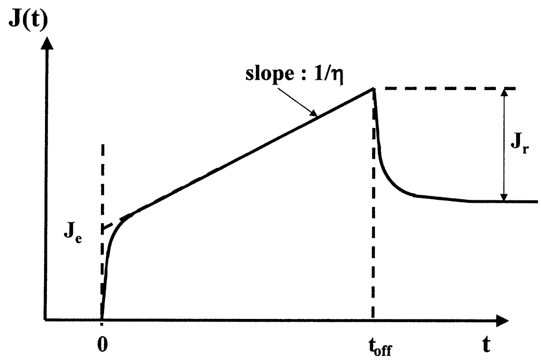

Fig. 2 Schematic diagram of the typical behaviour of the strain $\gamma(t)$ when a constant shear stress τ_0 is applied to a sample in the creep test. From time t_{off}, the stress is removed and the recovery behaviour of the sample is monitored

in the small strain limit should collapse to this function. Under these conditions, as indicated on Fig. 2, the following parameters can be determined: J_e, J_r and η [15]. J_e accounts for the elastic response immediately after the application of the stress behaviour and the transient response (up to the steady flowing stage), and J_r represents the so-called "elastic recovery" immediately after the stress is removed. For an ideal linear viscoelastic material, it can be shown that $J_e = J_r$ [15]. If the stress is applied for long times, the sample may eventually continue to undergo a permanent deformation, and the viscosity η at the corresponding shear rate is given by the inverse of the slope of the $J(t)$ curves in this steady flow region.

The duration of the creep tests was fixed at 1000 s for consistency. It was confirmed that this provided enough time for the sample's transient deformation behaviour to be established under the range of conditions studied.

Results and discussion

Figs 3a and 3b show the typical behaviour of the strain $\gamma(t)$ when a shear stress is applied to the sample, with all tests performed under a constant magnetic flux density of 0.57T. The values of the applied stress varied from 500 Pa to 5000 Pa. We observe that there is an initial jump in strain (the elastic response) followed by a very gradual transition region, lasting of the order of hundreds of seconds. When the stress is removed, the sample shows an immediate recovery, followed by a slower recovery process. Some data collapse of $\gamma(t)/\tau_0$ is obtained for curves where the applied stress is less than 3000 Pa, indicating that the behaviour is close to the linear regime at these stress levels. In terms of strain magnitude, Fig. 3a indicates that the linear response occurs for strains of order 10^{-3} or smaller. It is noted for comparison that the dynamic yield stress extrapolated from the steady flow curves under the same flux density of 0.57T was 9500 Pa [25], so we are indeed focusing on the "pre-yield" regime with regard to the stress magnitudes. It should also be noted that, for several of the tests, in addition to the transient behaviour at the start

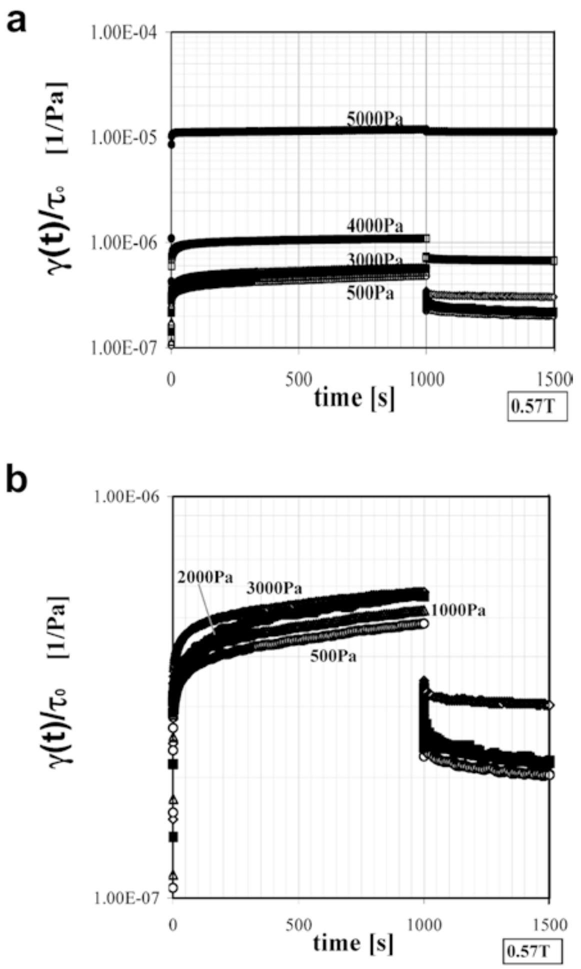

Fig. 3 Creep response curves for the case of a magnetic flux density of 0.57T, which was applied throughout the test. **a** The applied shear stresses are : 500 Pa (open circle), 1000 Pa (open triangle), 2000 Pa (black square), 3000 Pa (open diamond), 4000Pa (open square) and 5000 Pa (black circle). Logarithmic scale on the vertical axis has been used to clarify the initial transient behaviour, and the applied stresses were removed after 1000 s. **b** A close-up of the data under the lower applied stresses

of the measurement, extremely slow flow appears to occur at long times, as indicated by the fact that some of the curves in Fig. 3 do not become perfectly horizontal but continue to rise slightly. However, it is considered that the very low values of the shear rate involved (typically $10^{-5}s^{-1}$) make this part of the measurement susceptible to possible sample slippage and instrument noise effects, and so no analysis of this portion of the curves was undertaken.

A series of these tests was carried out to investigate the creep response under different magnetic flux densities. Similar to the observations in Fig. 3, for each flux density there was a range of applied shear stresses which corresponded to the linear response regime. Within this range we determined J_e and J_r by extrapolation using linear-linear plots, and we have plotted the variation of these parameters with magnetic flux density in Figs 4 and 5, respectively. The values obtained for different applied stress levels have been plotted in Figs 4 and 5, and it can be seen that the data collapse is reasonably good, which is expected if the response is in the linear regime. Note that data from several separate tests under each condition have been plotted together on these figures, and this does lead to some scatter, but nevertheless the trends appear to be clear and reproducible. Comparing the two quantities J_e and J_r, we note the qualitative similarity of the dependence on the magnetic flux density of J_e and J_r : both are significantly reduced as the field is increased. In fact, it was found that the field dependence of the data could be best fitted by a power law relationship, $J \propto B^{\alpha}$ where B is the magnetic flux density and α = -4.4 for J_e and -2.7 for J_r. In

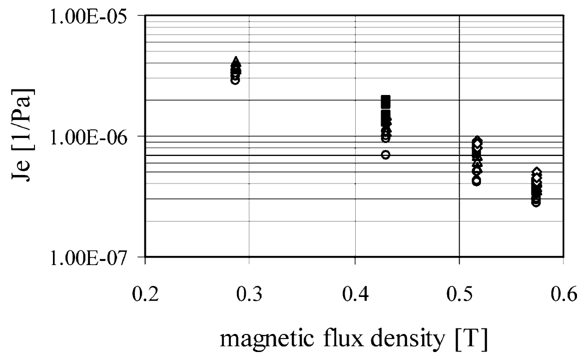

Fig. 4 Variation of the creep curve parameter J_e for linear response with magnetic flux density. J_e indicates the initial strain response. Measurements obtained for different applied shear stresses within the linear regime are plotted : 500 Pa (open circle), 1000 Pa (open triangle), 2000 Pa (black square), 3000 Pa (open diamond)

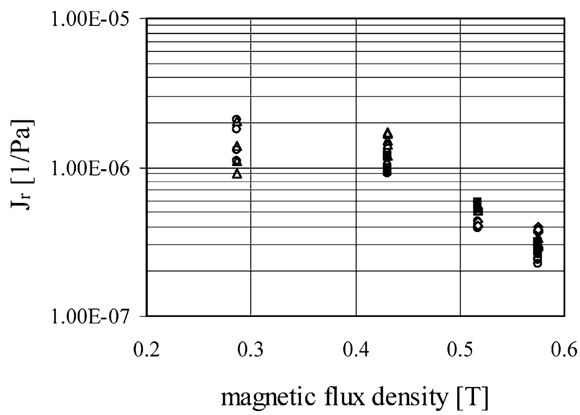

Fig. 5 Variation of the creep curve parameter J_r for linear response with magnetic flux density. J_r indicates the 'elastic recovery' response immediately after the removal of the shear stress. Measurements obtained for different applied shear stresses within the linear regime are plotted : 500 Pa (open circle), 1000 Pa (open triangle), 2000 Pa (black square), 3000 Pa (open diamond)

general, the significant decrease in J_e and J_r with increasing field is expected since the structures become stiffer and provide a greater resistance to shearing as the field strength is raised, due to the enhanced interparticle forces. However, there is some discrepancy with the simple "dipole-dipole interaction model": theoretically, since the interparticle forces due to induced dipoles scale as B^2 (see for example [18, 22]), for a fixed aggregate structure it would be expected that J_e and J_r would vary inverse quadratically with B (in other words with an exponent of -2). The difference between this prediction and the experimentally observed exponents (for J_e in particular) may reflect the fact that as the magnetic flux density is increased the nature of structures themselves undergo a change; for example the columns may significantly increase in thickness as the field is raised. Indeed this growth in column thickness under high fields has been directly observed in a model magneto-rheological suspension, as reported by Liu et al [14], who monitored structure growth in an emulsion of oil droplets in a ferrofluid. Thus it is possible that the response under creep is governed not just by the forces between particles, but by the nature of the aggregates that the particles find themselves in.

Although reasonable data collapse is observed in Figs 4 and 5, close inspection reveals that there appears to be a slight but systematic dependence of J_e and J_r on the magnitude of the applied stress : the lower stresses tend to produce lower J_e and J_r values. This departure from ideal linear viscoelastic behaviour is perhaps not unexpected, given the fact that the microstructure of these fluids is dynamically changing – for example, internal rearrangement of particle positions within aggregates have been observed in computer simulations of field responsive fluids (for instance [4]). Thus, at the level of the dynamics of the microstructure, these fluids seem to behave in a fundamentally different way to polymeric solutions for example, for which linear viscoelasticity is known to hold up to strains of order 20% (see for example [16]).

A feature of these results is that, even in the case of a strong magnetic field and small applied shear stress (for example the case of a 500 Pa shear stress under 0.57T flux density), the material was observed to undergo deformation over an extended period. As can be seen from Fig. 3, this appears to occur even if the applied shear stress is significantly smaller than the 'yield stress' obtained from steady shearing measurements (which is approximately 9500 Pa for this sample and 0.57T magnetic flux density). This suggests that the pre-yield material is unlike an elastic solid, for which the response to the applied stress would be a near-instantaneous jump in the strain, but that instead it undergoes very gradual deformation, presumably involving continual internal rearrangement and flow within the particle aggregates. This behaviour is certainly more complex than can be captured by the Bingham fluid equation Eqn.1 or indeed by the theoretical models based on idealized particle chains/lattices described in the Introduction. Possibly the key feature missing from these theories is the concept that when an external macroscopic stress is applied, the irregular configuration of the particles within the aggregates may lead to local concentrations of stress, in turn leading to local rearrangements of the particle positions.

Conclusions

We have examined the creep behaviour of a magneto-rheological suspension under a fixed magnetic flux density. It was found that even if the applied shear stress was considerably less than the 'dynamic yield stress' value extrapolated from steady shearing measurements, the material would show very gradual deformations at long times, although the strains involved are very small. This effect occurred even under the highest magnetic flux densities. This indicates that the mechanical behaviour of these materials is more complex than can be captured by the simple constitutive formulations developed to date.

The creep data obtained under low shear stresses showed reasonable collapse when plotted to test linear viscoelasticity, although, as expected, there was dependence of the viscoelastic parameters (J_e J_r) on the external field strength. This suggests that a model based on linear viscoelasticity may furnish a possible theoretical approach to describing the behaviour of these systems under very small strains. To place this on a firmer foundation, however, there is a need for a systematic series of tests to examine the behaviour of these field-responsive fluids under other small deformation modes, such as small amplitude oscillations, stress relaxation after a step strain, and the response under non-shearing deformations (for instance squeeze flow). These tests are currently being carried out on electro-rheological fluids and magneto-rheological suspensions, and the results will appear in future reports.

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