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Behaviour of field-responsive suspensions under oscillatory shear flow

Matti Keentok and Howard See[1]*
Engineering Support Group, GWEO, Bldg. N129, Defense Establishment, Orchard Hills, NSW 2748, Australia
[1]School of Chemical & Biomolecular Engineering, The University of Sydney, Sydney, NSW 2006, Australia
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Abstract

There has been considerable interest in recent years in field-responsive suspensions, which are of some importance in industry in many different applications. The microstructure of these materials is a significant issue which can be probed by rheological measurements. In this study, measurements were made of a magnetorheological fluid (MRF) under steady and oscillatory shear flow, with and without a magnetic field. Mathematical inversion was used to derive the relaxation time spectrum of the MRF from oscillatory shear data. Experimental evidence is presented of the gel-like properties of this MRF.

Keywords : Magnetorheological fluid, field-responsive suspension, microstructure, cross-linking, oscillatory shear flow, mathematical inversion, non-linear regularization

1. Introduction

Magnetorheological fluids (MRFs) typically consist of magnetisable (iron) particles in an oil (often silicone, sometimes mineral oil). When under a magnetic field, MRFs show a dramatic increase in shear resistance due to microstructural changes which arise from the induced dipole-dipole forces between particles leading to elongated aggregates in the field direction.

Various types of MRF have been constructed and studied: these include ferromagnetic fluids made with magnetite or with carbonyl iron. The solvent phase in a MRF can be an oil or silicone fluid (Rankin *et al.*, 1998; Park and Park, 2001), oil-in-water emulsion (Park *et al.*, 2001) or grease (Rankin *et al.*, 1999). Various other additives have been considered. Note that ferrofluids composed of nanoparticles of iron (Larson, 1999) are not suited for use as MRFs as their field-induced viscosity increment is small.

MRFs are of some importance in industry and now have a large number of applications in diverse fields (Klingenberg, 2001): these include in finishing/polishing machines, as MR dampers in buildings, automobiles and helicopters, and also in medical uses (Flores *et al.*, 1999; Liu *et al.*, 2001).

The structure of MRFs is an important issue, as seen by mentions in many reviews of magnetorheological research. Structure will be reviewed briefly here and will be a matter of discussion on the basis of results presented here. Besides the usual steady shear rheological measurements, oscillatory shear rheology has been undertaken here to study microstructure in MRFs. Also of relevance in these microstructural studies are electrorheological fluids (ERFs) which are electrical counterparts to MRFs, involving a suspension of electrically polarisable particles under an electric field. MRF and ERF have many physical similarities and are sometimes referred to as 'field-responsive suspensions'.

As part of rheological characterisation, it is now possible to generate relaxation time spectra from rheological data, using various mathematical procedures including non-linear regularization. From a literature survey, summarised below, it became clear that the relaxation time spectrum of MRFs has not been published and this is a gap in our knowledge which we aim to fill.

1.1. Literature

The rheology of MRFs has been reviewed many times and in particular by Larson (1999), Rankin and co-workers (Rankin *et al.*, 1998), Park and Park (2001), to name a few recent reviews. In recent times, there has been research on the rheology of viscoplastic media (*i.e.* a magnetic dispersion in a grease) by Rankin *et al.* (1999), on the rheology of MR suspensions by Chin *et al.* (2001), on the rheology of MR suspensions in oil-water combinations by Park *et al.* (2001) and measurements of creep and recovery by Li *et al.* (2002). Other rheological measurements on MRFs have been undertaken by Vekas *et al.* (2000), by See and Tanner (2003), and by the group of Choi (Choi *et al.*, 2007). Mostly, published data contains flow curves of shear stress against shear rate with very little oscillatory

*Corresponding author: howards@chem.eng.usyd.edu.au

shear data.

A small amount of oscillatory shear data has been obtained for MRFs (and ERFs). Chin *et al.* (2001) studied carbonyl iron and magnetite suspensions, although only one MRF was reported with oscillatory shear data. A magnetite suspension (volume fraction φ=0.20) showed a flat G" curve but increasing G' curve in the frequency range 1-10^3 rad/s at low magnetic fields. Thus while the MRF was elasticoviscous, it was not a gel. Further, no oscillatory shear data was provided for the zero field measurement. Another oscillatory shear test in ER, that of Chin and Winter (2002), showed gel-like structure in an ERF when under an electric field, but not without a field. Rankin *et al.* (1999) studied oscillatory shear characteristics of a MRF suspended in grease.

Relaxation time spectra have now been obtained for many types of materials, including polymer melts, polymer blends, polymer solutions and biomaterials such as bread dough (Keentok *et al.*, 2002; Keentok, 2001; Phan-Thien and Safari-Ardi, 1998). There do not appear to be any relaxation time spectra for MRFs (or ERFs for that matter). Approximate and exact methods are available for extracting relaxation time spectra from rheological data: among the exact methods are several types of mathematical inversion including non-linear regularization.

The dramatic increase in flow resistance of MRFs or ERFs under the external field is due to the structures formed by the particle aggregates. Much research effort has gone into studying the relationship between the type of structures formed and the materials' overall mechanical response. The problem is actually quite complex - the structures formed typically involve a large number of particles in complicated configurations. To simplify the theoretical analysis, idealised structures which are aligned with the field and span the gap are often assumed; for example single-width chains of particles or regularly-shaped columns of particles. There is now a large volume of modelling work in the literature, and it is beyond the scope of this paper to review this in detail - several articles which provide a useful overview are now available (Larson, 1999; Rankin *et al.*, 1998; Park and Park, 2001; Klingenberg, 2001; Halsey, 1993; Parthasarathy and Klingenberg, 1996, Klingenberg, 1998; Ginder, 1998; See, 1999). On the experimental side, the microstructure of dilute systems has been studied by optical transmittance and light scattering (Martin *et al.*, 1999; Melle *et al.*, 2002) as well as by direct microscopic observation (Klingenberg and Zukoski, 1990; Tao, 2001).

A common feature of many of the theoretical models developed to date is that they assume a linear structure which, before deformation is applied, is aligned in the field direction. This would perhaps be a reasonable model for dilute systems, but there is a question as to the appropriateness at moderate to high particulate concentrations. An interesting departure from this picture was presented by Pan and McKinley (Pan and McKinley, 1997), who, after examining the strain dependence of oscillatory shear data for an ERF, inferred that there was cross-linking of the structures in ERFs (*i.e.* shorter chain-like structures which link the chains/aggregates spanning the gap). They proposed that an anisotropic network model may be more appropriate for ERFs. Such a network structure would be expected to show rheological behaviour similar to a gel, and indeed this possible connection will be a focus of the present study.

2. Theory

The relaxation time spectrum is important and is required if one wishes to develop a multi mode constitutive model. The spectrum is also important as it can be used to calculate any viscoelastic material function (Mead, 1994; Weese and Friedrich, 1994) and may also be useful for polymeric materials in other calculations (Thimm *et al.*, 2000).

Relaxation time spectra may be generated from dynamic (oscillatory shear) data using non-linear regularization by Tikhonov regularization (Honerkamp and Weese, 1993). The program NLREG of Honerkamp and Weese (1993) was used to invert dynamic data to produce a relaxation time spectrum, which is expressed through the function $H(\lambda)$, defined as below. NLREG has been previously used here to compute relaxation time spectra for bread by Phan-Thien and Safari-Ardi (1998) and by Keentok *et al.* (2002). The mathematical definition of inversion is given by Phan-Thien and Safari-Ardi (1998) as follows:

$$G'(\omega)=G_E+\int_{-\infty}^{\infty}\frac{\lambda^2\omega^2}{1+\lambda^2\omega^2}H(\lambda)d\ln\lambda \tag{1}$$

$$\eta'(\omega)=\int_{-\infty}^{\infty}\frac{\lambda}{1+\lambda^2\omega^2}H(\lambda)d\ln\lambda \tag{2}$$

where $H(\lambda)$ is the value of the relaxation time spectrum at relaxation time, λ, G_E is the equilibrium shear modulus, and $G'(\omega)$ and $\eta'(\omega)$ have the usual meanings of storage modulus and the in-phase component of the complex viscosity, respectively.

3. Experimental

The samples used in the experiments were made from carbonyl iron particles (ISP Technologies INC Iron Micropowder Grade S-3700) dispersed in Dow Corning 200 silicone oil (100 cSt) made up to a volume concentration of 30% (φ=0.30). The diameter distribution of the carbonyl iron particles was determined by optical microscopy to be in the range 0.3-3 μm; the particles were not truly spherical. This diameter distribution is similar to that found in

other carbonyl iron-based magnetorheological suspensions (as reviewed in (Klingenberg, 2001) for example). The saturation magnetisation of these particles is approximately 200 emu/g (this mass magnetization in SI units corresponds to 200 Am^2/kg) (Bombard *et al.*, 2003).

The rheometer used for the MR experiments is a Physica MCR300 fitted with a MR system, the TEK 70MR magnetorheological cell. The TEK 70MR is a parallel plate system which uses a strong electromagnet under the bottom plate to apply a vertical magnetic field to the sample. Details of this apparatus have been previously reported (See and Tanner, 2003).

Measurements were undertaken at a room temperature around 25°C. Since MRFs are often based on oils or silicone (as this one is), they are inherently stable but can suffer from sedimentation particularly where the particle size is large: thus the sample must be stirred before use. For this sample, sedimentation was not noticeable over a period of a week (probably because the particle distribution is around 1 μm), however stirring was always undertaken.

4. Results

4.1. Steady Shear

The low molecular weight silicone fluid (100 cSt) was measured on its own on the Physica rheometer, to check for any instrument effects; the molecular weight for this fluid is about 20,000 (Keentok, 2001; Keentok, 1997). The viscometric measurements revealed no instrument effects.

Viscometry of the MRF was undertaken in the steady shear rate range 0-100 s^{-1} (0.8 mm gap) and with a range of magnetic field values of B=0, 0.07, 0.14, 0.29, 0.38, 0.57 Tesla (T).

Typical data is shown in Fig. 1. Reproducibility of the shear rate sweeps was excellent. When a magnetic field is applied, there is a significant increase in shear stress and yield stress. Under maximum field (B=0.57 T), there was a yield stress of 17 kPa, which is comparable to other literature (See, 2001).

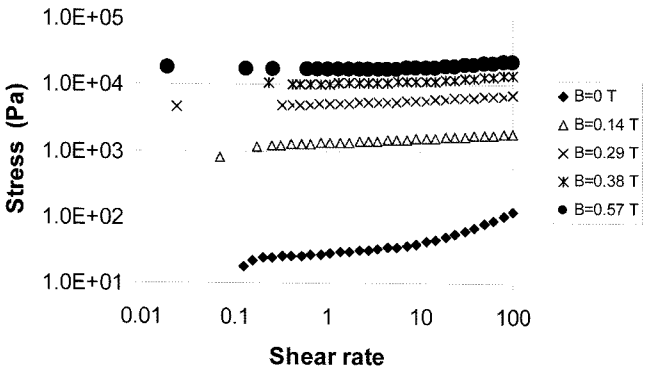

Fig. 1. Steady shear rheology of MRF as a function of magnetic field strength, B.

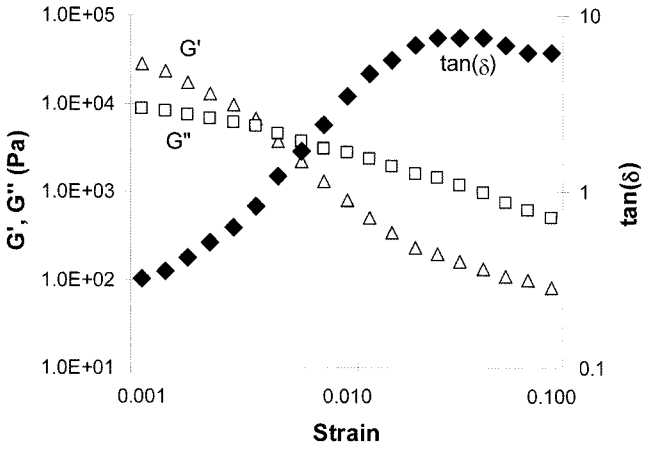

Fig. 2. Oscillatory shear rheology of MRF: strain sweep at fixed frequency ω=10 rad/s.

4.2. Oscillatory shear

Strain sweep: A strain sweep was performed at ω=10 rad/s (see Fig. 2) and this shows the differences in the strain dependence for G' and G". Note that G' drops off more rapidly with strain amplitude than G", as seen by the change in tan δ(=G"/G') which is also plotted. From this data, the strain for the limit of linear viscoelasticity was determined to be approximately 0.001. Comparable values for the linear viscoelastic limit have been obtained by Pan and McKinley (1997) and Chin *et al.* (2001). Hereafter all oscillatory shear measurements were undertaken at this limit.

Frequency sweep: Frequency sweeps at a strain amplitude of 0.001 were undertaken in the range given by ω: 1-100 rad/s for coil currents in the sequence I=0 A, 0.25A, 0.5A, 1.0A, 1.5A, 2.0A, corresponding to field strengths of B = 0, 0.07, 0.14, 0.29, 0.38, 0.57 Tesla. Typical data are shown in Fig. 3. Reproducibility of oscillatory shear data

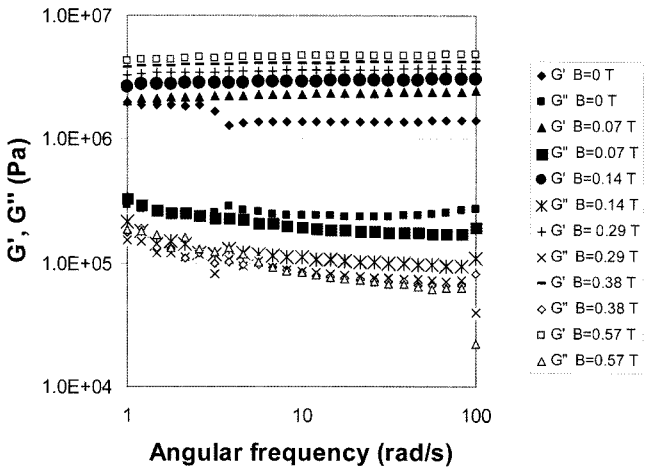

Fig. 3. Oscillatory shear rheology of MRF: frequency sweep as a function of magnetic field strength (constant strain amplitude of 0.001).

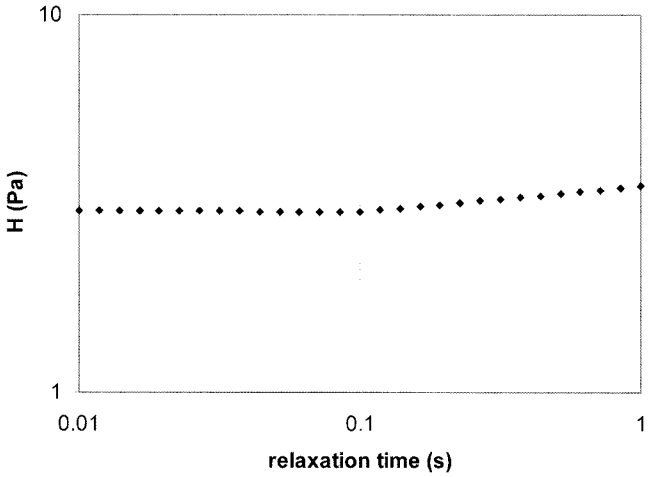

Fig. 4. Relaxation time spectrum of MRF under magnetic field of 0.57 T.

was satisfactory. Even at zero field, G'>G" and this becomes more pronounced when a magnetic field is applied (*i.e.* G'>>G", elasticoviscous behaviour).

Under the fields tested, there was almost a constant G' and G" with increasing frequency, indicating a gel-like consistency, similar to that seen in particulate gels ((Larson, 1999), see also Analysis). The gel-like consistency agrees with the yield stress seen both without and with the magnetic field (see Fig. 1) and suggests that this MRF is a semi-solid under small strain oscillatory shear.

The oscillatory shear data was also used to compute the relaxation time spectrum of this MRF with a magnetic field (see Fig. 4), using non-linear regularization implemented in the NLREG software of Honerkamp and Weese (1993). Results for weaker field strengths were of a similar form. Relaxation time spectra were previously obtained for bread dough using the same NLREG software (Keentok *et al.*, 2002; Phan-Thien and Safari-Ardi, 1998). The relaxation time spectrum shows log H as a function of relaxation time. The relaxation time spectrum is susceptible to the slightest noise in the experimental data, so the curve is the average of two datasets.

5. Analysis and Discussion

5.1. Steady Shear

It is clear from bi-linear plots of shear stress against shear rate, that the data do not follow a Bingham model (except in a very limited range of shear rates). From Fig. 1, we conclude that with the exception of the data for B=0 T, the data follow a Herschel-Bulkley model:

$$\tau=\tau_y+k\dot{\gamma}^n \quad (3)$$

where τ is the shear stress, τ_y is the field-induced yield stress, k is a constant, n is the power-law exponent and $\dot{\gamma}$ is the shear rate. The power law exponent n is generally about 0.23 (except for B=0 T). The yield stress varies significantly with magnetic field intensity from a zero field value of ~10 Pa to 17 kPa at 0.57 T (see Fig. 1). We see a thousand-fold increase in yield stress at maximum field and hence we have a good MR effect at volume fraction of φ=0.30. At the maximum field of Chin *et al.* (2001), they obtain an increase in yield stress of ~300, whereas our MRF shows an increase of ~100 to a comparable magnetic field (see also below). The maximum yield stress of our MRF (17 kPa at B=0.57 T) is comparable to MRF reported elsewhere (reviewed in (See, 2001)).

It is difficult to directly compare our data with the carbonyl iron data of Chin *et al.* (2001) since their largest magnetic field is approximately equivalent to our smallest magnetic field. Our carbonyl iron MRF shows larger shear stress $\tau(\dot{\gamma})$ both for zero and non-zero fields (by as much as a factor of 4 when comparing data for the same magnetic field, and by as much as a factor of 10 for zero field). This difference may be due to differences in the particle size distribution: our MRF has a broader particle diameter distribution with more particles of smaller diameter compared to Chin *et al.* (2001) (whose MRF had a narrow diameter distribution). It is worth noting Chin *et al.* (2001) also comment that under magnetic fields, both iron suspensions (*i.e.* magnetite and carbonyl iron) gave similar rheological properties. Thus, since the steady shear behaviour of two different MRFs is similar, oscillatory shear could prove a useful way to probe and differentiate microstructure, and this will now be explored.

5.2. Oscillatory shear

The frequency sweep data shows that there is almost a constant G' and G" with increasing frequency (even at zero field), indicating a gel-like consistency (see Experimental, Fig. 3). The strain dependence of G', G" and tan δ (Fig. 2) indicates that under small strain oscillations, this MRF behaves like a gel but becomes liquid-like under large strain oscillations as the aggregates are broken up. A large increase in tan δ as strain increases is evident also in the MRF data of Chin *et al.* (2001), but this was not commented on by them - this increase indicates the same behaviour as seen here. It is also worthwhile comparing our results with the behaviour predicted by particle-level dynamic simulations (Sim *et al.*, 2003) - interestingly we do not observe the slight increase in G" at the onset of non-linearity predicted by the simulations (possibly the frequency we used was too high to observe this overshoot phenomenon). Further, on more careful examination of the frequency sweep data, there is a slight trend of decreasing G" with increasing frequency at some values of the magnetic field strength (Fig. 3); interestingly this trend is also seen in the 'plateau' region of polymeric materials (*e.g.* Graessley, 1974). The field dependence is as expected for an MRF: as the magnetic field is applied and increased, G'

increases and G" decreases. This indicates increasing elasticity and decreasing viscosity (or increasing elasticoviscosity) with increasing magnetic field. However, the frequency dependence of G' and G" requires some explanation, as follows.

There have been many attempts to derive microstructural information from rheology, in particular oscillatory shear rheology (Ferry, 1980; Gras *et al.*, 2001). Almost flat G' and G" curves as a function of frequency are seen in crystalline polymers, in cross-linked polymers and polymer gels (Ferry, 1980) and in solid-like materials (Chap. 1 in (Larson, 1999)). The current sample is not believed to be crystalline or a solid, but possibly cross-linked or forming a network which is easily destroyed by large amplitude oscillations or steady shearing. Particulate gels also give rise to almost flat G', G" curves as a function of frequency (Larson, 1999), with identical trends in G' and G" as this MR fluid. Further, Pan and McKinley (1997) have compared an ERF with a particulate gel and found similarities in the rheology. Larson (Chap 7 of (Larson, 1999)) has commented that stabilization of a suspension to form a gel can be by electrostatic means or particle bridging, but it is suggested here that it may also be possible magnetostatically. Thus while it is possible that another microstructure could give rise to the characteristic G' and G" curves seen here, at this stage the gel-type network structure would seem to be the most likely candidate.

In addition, the decreasing trend in G" as frequency increases, together with almost constant G', is typically seen in the rubbery-plateau zone for entangled polymers (Graessley, 1974; Larson, 1988). That we should see this in an MRF is interesting and further suggestive of a cross-linked microstructure. Indeed, the low values obtained for tan δ indicate considerable cross-linking (Ferry, 1980). At this stage it is not certain whether there is cross-linking with MRFs, but Pan and McKinley (1997) found cross-linking and network structures in an ERF and also found experimental evidence for this based on microscopy.

The oscillatory shear data of Chin *et al.* (2001) shows that their MRF was elasticoviscous (G'>G", more so at high frequencies) when exposed to a magnetic field but there was no evidence of being gel-like with or without a magnetic field (Fig. 9 of (Chin *et al.*, 2001)). This difference could be due to their MRF being a lower volume fraction of 0.2. Being gel-like implies the existence of a combination of linear, gap-spanning aggregates and cross-links - that is, similar to structures one finds in polymer gels or in rubbers (*i.e.* cross-links or networks). This would be less likely to occur with lower particulate volume fractions.

A survey of previous rheometrical studies on MRFs indicates that there have been no suggestions of cross-linked structures for these systems. However, Pan and McKinley (1997) have found cross-linking and network structures in an ERF based on oscillatory shear rheology and on microscopy. Similarly, gel-like oscillatory shear rheology was also seen in an ERF by Chin and Winter (2002). For MRFs, there is evidence of the existence of this cross-linked structure from other types of data. Microscopy of MRFs exposed to magnetic fields has shown the linear chain (Klingenberg, 2001) and thicker columnar structures (Tao, 2001). However, in recent years, finer structure in MRF is evident from computer simulations: simulations of colloidal suspensions which demonstrate deviations from strictly linear aggregates are common. For example Martin *et al.* (1999) show both linear and curved structures of particles with side chains when an MRF is exposed to magnetic fields. Thus, there is indeed the possibility of cross-linking in MRF. There is also some support from computer simulations of ferrofluids, which show that at higher concentrations, as well as the linear clusters formed along the field, there is also aggregation perpendicular to the field (Satoh and Kamiyama, 1995). Of course, it should be kept in mind that the particles in ferrofluids are permanently magnetised and are much smaller (typically 10 nm) than those in MRFs.

Further, it should be noted that there is the possibility of remanent magnetisation in MRF particles, so there is a weak magnetically-induced microstructure even when the magnetic field has been turned off. This also may explain why we see a gel-like structure with no applied magnetic field.

That this MRF has a broad spectrum of relaxation times is a new finding not previously reported. The relaxation spectrum curve (log H, Fig. 4) is flat, indicating that in this range of relaxation times, the shear moduli, G_i, for each λ_i are the same (Phan-Thien and Safari-Ardi, 1998). This is somewhat unusual, and comparison may only be made with relaxation time spectra for polymeric systems and biomaterials. In the case of an end-linked gel, log H shows power-law behaviour (Mours and Winter, 1998) which is typical of polymers but different from this spectrum. For bread dough, again power-law behaviour is seen (Keentok *et al.*, 2002).

6. Conclusions

The importance of oscillatory shear data in probing the microstructure of samples is well known (*e.g.* in relation to bread dough (Gras *et al.*, 2001)) and this work has demonstrated another use in MRF. The existence of a gel-like structure in MRFs from small-strain oscillatory shear suggests that these fluids are not necessarily mobile liquids even when not exposed to a magnetic field, but could possibly be modelled in a similar fashion as a polymeric liquid with cross-linked microstructure. The existence of this microstructure in an MRF does not appear to have been noted previously (despite being known in ERF). It is rea-

sonable to say that cross-linking is plausible, based on microscopic and simulation studies, as well as rheology.

From consideration of the relaxation time spectrum, this MRF has a broad spectrum of relaxation times and this supports the above suggestion of a complex structure, with cross-linking. Thus this work has provided further insights into the microstructure of MRFs. To improve the relaxation time spectra, it would be desirable to increase the frequency range of the oscillatory shear measurements: besides giving a broader range of relaxation times, this would stabilise the mathematical inversion process used to extract the spectrum.

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