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Squeeze flow behavior of (soft glassy) thixotropic material

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ABSTRACT

We study the flow behavior of a model soft glassy material – an aqueous suspension of Laponite – when it is squeezed between two circular parallel plates of different roughness. Aqueous suspension of Laponite shows a time dependent aging behavior as reflected in increased elastic modulus as well as yield stress, both of which however also decrease with an increase in the strength of deformation field thereby demonstrating typical thixotropic character. In a squeeze flow situation, under both force as well as velocity controlled modes; we find the behavior to be independent of the initial gap between the plates. In a constant force mode, the gap between the plates decreases until it reaches a finite limiting value, which is found to increase with an increase in age of the material as well as with a decrease in the applied force. In constant velocity experiments, at large gaps between the plates, normal force varies inversely with plate separation. The normal force is higher for a sample aged for a longer time as well as for a larger velocity of the top plate. We observe that the experimental behavior follows prediction of Herschel–Bulkley model solved for the squeeze flow (with different friction coefficients at the two plates) reasonably well under weak deformation fields. However, under strong deformation fields, experimental behavior deviates significantly from the prediction of Herschel–Bulkley model. This deviation arises due to melting or partial yielding of Laponite suspension under large deformation fields causing decrease in the viscosity, elastic modulus and the yield stress.

1. Introduction

Many biologically and commercially important soft materials undergo evolution of their structure and rheological properties as a function of time. Typically such materials demonstrate increase in viscosity and elastic modulus under quiescent conditions [1–3]. Under application of deformation field having sufficient strength, viscosity and elastic modulus decrease as a function of time [4–7]. In rheology literature this behavior is represented by a term: thixotropy [8,9]. Physically, the time dependent behavior is usually attributed to kinetic infeasibility of these materials to attain thermodynamic equilibrium over the experimental timescales. Consequently, the exploration of the available phase space in search of lower energy states drives the thixotropic materials to undergo evolution of their structure as a function of time. In the recent literature, this phenomenon is termed as physical aging [6,10–14], while decrease in viscosity and elastic modulus as a function of time under application of deformation field is termed as rejuvenation. Owing to the behavioral similarity, which this class of materials share with the structural glasses, these are represented as soft glassy materials [15]. The thixotropic behavior (or aging and rejuvenation) imparts strong history dependence and influence any commercial processing of these materials necessary to form a useful product. Therefore, better understanding of the rheological behavior of these materials, particularly through more realistic flow fields, is desirable.

Squeeze flow is an important flow field often encountered in materials processing [16] and biology [17]. Material is compressed between two parallel plates in a squeeze flow, either by applying a compressive force [18] or by maintaining a constant velocity [19,20] so that a radial flow is produced. Squeeze flows of inelastic and visco-elastic fluids have been extensively studied by theory, simulations and experiments [16]. This class of rheometry is cost effective and can be particularly useful for materials which are too stiff or viscous to be handled with conventional rheometer geometries. Due to its commercial importance and academic interest, the study of yield stress fluids under squeeze flow conditions has generated considerable interest. Though this rheological technique is operationally simple and convenient, the complexities of wall slip [21–23], unsheared regions between the plates [24–26], phase separation due to relative radial motion of various phases [27], can make interpretations more difficult than the conventional methods. Much of work on squeeze flow of yield stress fluids is associated with theoretically and experimentally analyzing the flow field vis-à-vis yielding surface which determines the size and shape of unyielded regions within the material which move as a plug [24,26,28], effect of the boundary characteristics like presence or absence of a slip between the plate–sample surface

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[29–31] and as a means of an estimation of various rheological properties of the material particularly the yield stress [28,32,33]. Most of the real fluids that possess the yield stress are thixotropic and show complicated rheological behavior for any simple mathematical model to work well. Nonetheless, in numerous studies it has been shown that many soft materials with yield stress can be satisfactorily modeled with Bingham or Herschel–Bulkley constitutive relation in which the material is modeled to possess an infinite viscosity until a threshold stress is reached beyond which it behaves like a ‘power law’ viscous fluid [18,20,32,34–37]. However, still in many cases these models prove to be rather simple in nature due to some important inadequacies. The most important one is that they are time independent in nature whereas yield stress fluids are more often than not thixotropic as well. The reason is that the same underlying phenomenon of presence/destruction of a microscopic structure gives rise to both [9]. Another difficulty is determining the exact value of the yield stress of the material, which has been a subject of much debate in the rheology community [38,39]. The reason for this is a strong dependence of the yield stress on the experimental procedure followed in order to determine it. Many models for thixotropic fluids have been proposed which take into account the evolution of a structural parameter with time which corresponds to the degree of flocculation, jamming or the fraction of particles trapped in energy wells [8,40–42].

In this work, we study squeeze flow with plates having different roughness. In industry plates having dissimilar roughness are used so that the material preferentially sticks to only one surface. Plates having dissimilar characteristics have also been used to study shear flow behavior of soft glassy materials [43]. We use an aqueous suspension of Laponite in the squeeze flow study. Laponite is an important additive used in the chemical and the food industry to control rheological behavior of the end product [44]. Aqueous suspension of Laponite not only shows yield stress [45], but also demonstrates various generic thixotropic characteristic features of soft glassy materials like: deformation field dependent and time dependent viscosity and elasticity (and therefore, relaxation time) [6,46], incomplete stress relaxation upon application of step strain [47], weak frequency dependence of modulus [48], etc. It is thus considered to be a model soft glassy material [7,14,45,49–56]. Apart from academic significance, understanding squeeze flow behavior of soft glassy (thixotropic) materials is important from an industrial point of view as well. Many commercial soft glassy materials such as highly filled polymer melt and wheat dough do undergo squeeze flow while forming useful products.

2. Material, sample preparation and viscometry

Laponite® is a synthetic hectorite clay and is composed of disk like particles with a diameter of 25 nm and thickness of 1 nm [44,57]. In this work, we have used a suspension of 3.5 wt.% Laponite RD (Southern Clay Products, Inc.) in water. The white powder of Laponite was dried for 4 h at 120 °C before mixing with water. The sample was prepared by mixing a calculated amount of Laponite RD with ultrapure water at pH 10 under continuous stirring. The suspension was stirred vigorously for 30 min and left undisturbed for 1 month in a sealed polypropylene bottle. In this work, we have carried out oscillatory shear experiments and both force controlled and velocity controlled squeeze flow experiments using Anton Paar Physica MCR 501 rheometer (parallel plate geometry, 50 mm diameter). Before starting each experiment, Laponite suspension was shear melted by first passing it through an injection syringe having 0.5 mm diameter needle and 4 cm length several times. Subsequently, after loading it in the parallel plate geometry of the rheometer, suspension was further shear melted by applying an oscillatory rotational shear stress which is higher than the yield stress. The aging time was measured after the shear melting was stopped. We have carried out most of the oscillatory as well as squeeze flow experiments using a rough sandblasted top plate (roughness: 5.74 μm), unless otherwise mentioned where a smooth polished top plate (roughness: 0.8 μm) was used. The bottom plate was a smooth polished plate (roughness: 0.8 μm) in all the experiments. The free surface of the sample between the plates was coated with low viscosity silicone oil to prevent evaporation and/or contamination with CO_2 [58]. All the experiments were carried out at 25 °C.

3. Results

Aqueous suspension of Laponite is known to undergo enhancement in elastic modulus and characteristic relaxation time as a function of time [7,48,59]. The suspension has paste like (soft solid) consistency and it also demonstrates yield stress [60]. We measure the yield stress of the suspension at various times elapsed after shear melting (aging time) by carrying out stress sweep oscillatory experiments at frequency $f = 1$ Hz. Fig. 1 shows that with increase in shear stress (σ_0), elastic modulus (G') decreases sharply over a very narrow range of stress amplitudes thereby enabling measurement of yield stress. We do not rule out the presence of thixotropy introducing an error in the determination of the yield stress but we expect it to be small as the rate of change of shear stress was high allowing completion of the whole experiment in a time span of the order of seconds. As the time required to carry out each stress sweep experiment was much smaller than the corresponding elapsed time (aging time), structural evolution (aging) that takes place during the course of experiment can be ignored. Interestingly, all the data obtained at different aging times show a superposition when elastic modulus is normalized by the elastic modulus associated with the linear response regime (G_0) and stress amplitude is normalized by the yield stress (τ_y). In the inset of Fig. 1, G_0 and τ_y are plotted as a functions of aging time (t_w). It can be seen that material demonstrates enhancement of modulus (G_0), and yield stress (τ_y) with the same dependence on aging time, $\tau_y,\ G_0 \propto t_w^{0.4}$ ($\tau_y \sim G_0$).

It has been shown that many soft glassy materials with yield stress can be satisfactorily modeled with Herschel–Bulkley constitutive relation given by [18,20,32,35]:

$$\underset{\approx}{\sigma} = \tau_y + k\dot{\gamma}^{n-1}\,\underset{\approx}{\dot{\gamma}}, \quad \text{if } (\underset{\approx}{\sigma} : \underset{\approx}{\sigma}/2 \geqslant \tau_y^2) \quad \text{and}$$

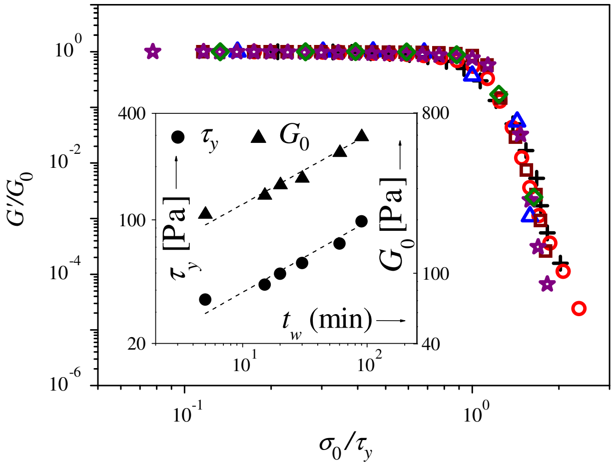

Fig. 1. Yielding behavior of Laponite suspension in a stress sweep oscillatory test at frequency $f = 1$ Hz for various aging times [square: 5 min, circle: 15 min, triangle: 20 min, diamond: 30 min, star: 60 min and plus: 90 min]. Inset shows yield stress (τ_y) and linear regime elastic modulus (G_0) plotted as a function of aging time (t_w). Both τ_y and G_0 follow the same dependence on t_w given by $\tau_y, G_0 \propto t_w^{0.4}$.