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The mechanics of yield stress fluids: similarities, specificities and open questions

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Abstract

A wide range of materials encountered in our everyday life, such as clay suspensions, foams, concentrated emulsions, cement pastes, paints, glues, purees, creams, can flow like simple liquids under certain conditions and behave like solids under other conditions. This is the specificity of yield stress fluids which makes them so useful in various applications. In their "liquid regime" these materials exhibit typical flow properties of simple fluids such as a transition to turbulence, the roll wave instability, the hydraulic jump, etc. The specific properties occur when the solid regime is involved, either in a part of the material or as a whole. In that case one may for example observe plug flow, flow stoppage over steep slopes, no sedimentation of dense particles, cylindrical drips, Saffman-Taylor instability at vanishing velocity, etc. In addition yield stress fluids are often thixotropic, i.e. their viscosity may vary in time. The physical origin of this phenomenon and the mechanical model appropriate for describing it remain the most challenging aspects of these fluids.

Introduction

Besides simple (Newtonian) fluids such as air and water which condition our everydaylife, we find a wide range of industrial or natural materials which are strongly non-Newtonian. A non-exhaustive list of such materials include clay suspensions, foams, concentrated emulsions, drilling fluids, cement pastes, paints, glues, purees, creams, ceramics, mayonnaise, hair gel, etc. Their main specificity relies in the fact that under some circumstances they can flow like simple liquids, so that they are generally considered as fluids, while under other circumstances they can behave as solids. For example mudflows in mountain streams, drilling fluids in conduits, fresh concrete during casting, foams coated over skin, mayonnaise extruded from a tube, appear as simple, though viscous, flowing fluids. However mudflows also form thick deposits over steep slopes, drilling fluids can support denser particles in suspension, shaving foams remain fixed on a vertical surface after coating, the mayonnaise keep its shape in the plate. In those cases these materials behave as solids, i.e. they undergo finite deformation when submitted to a stress below a critical value. This is the specificity of **yield stress fluids**, which makes them so useful in various applications. From a general point of view the description of these fluids is rather difficult. Here we first review some aspects of their behaviour in the liquid regime, i.e. when they appear as simple fluids; then we describe some implications of the existence of a solid regime. At last we discuss the most complex aspect of these materials, namely the fact that their apparent viscosity or strength, may significantly vary in time due to a spontaneous internal evolution of their microscopic structure.

Behavior modelling

Yield stress fluids are materials which can flow only when the applied stress overcomes a critical value, namely their yield stress τ_c. At first sight they resemble ideal plastic materials but they are in fact *viscoplastic*, since the resulting flow rate increases with the difference between the applied stress and the yield stress. In this context usual models describing such a behavior in simple shear steady state express as follows:

$$\tau < \tau_c \Rightarrow \dot{\gamma} = 0 \tag{1}$$

$$\tau > \tau_c \Rightarrow \tau = \tau_c + f(\dot{\gamma}) \tag{2}$$

in which τ and $\dot{\gamma}$ are the shear stress and shear rate amplitudes [1-2]. A "solid" and a "liquid" regimes thus respectively correspond to (1) and (2). A fundamental assumption of usual yield stress models is that f is an increasing function tending to zero when $\dot{\gamma}$ tends to zero. This means that there is a continuous transition from the solid to the liquid regime in terms of shear rate: as the shear stress decreases to the yield stress, the shear rate progressively tends to its value in the solid regime, i.e. zero. In other terms the apparent viscosity ($\eta = \tau/\dot{\gamma}$) of the material is finite in the liquid regime and progressively tends to infinity (its value in the solid regime) as the applied stress tends to the yield stress. In practice the Herschel-Bulkley model, for which $f(\dot{\gamma}) = K\dot{\gamma}^n$, in which K and n are two material parameters, and the Casson model, for which $f(\dot{\gamma}) = K\dot{\gamma} + 2\sqrt{K\tau_c\dot{\gamma}}$, are the most often used models. Note that the Bingham model is recovered from the Herschel-Bulkley model with $n = 1$. Although the Casson's model is often used in industry because it involves only two parameters, the Herschel-Bulkley model appear to be capable to very well represent usual shear stress vs shear rate data over a wide range of shear rates (several decades) [3-4]. The following three-dimensional expression is generally used:

$$\sqrt{-T_{II}} < \tau_c \Leftrightarrow \mathbf{D} = 0 \tag{3}$$

$$\sqrt{-T_{II}} > \tau_c \Rightarrow \mathbf{\Sigma} = -p\mathbf{I} + \tau_c \frac{\mathbf{D}}{\sqrt{-D_{II}}} + F(D_{II})\mathbf{D} \tag{4}$$

in which $\mathbf{D}$ is the strain rate tensor, $\mathbf{\Sigma}$ the stress tensor, p the pressure, and D_{II} and T_{II} the second invariant of the strain rate and stress tensors. Note that $F(D_{II}) = 2^n K / \left(\sqrt{-D_{II}}\right)^{1-n}$ for a Herschel-Bulkley fluid. Also note that although the simple shear expression (1-2) was deduced from experimental data, expression (3-4) is only extrapolated.

The equations (1-2) or (3-4) assume that the fluid is perfectly rigid below the yield stress. More sophisticated models were also proposed to take into account the viscoelastic behavior in the solid regime. For example a basic approach consists to replace equation (1) by a viscoelastic solid model such as the Kelvin-Voigt model for $\gamma < \gamma_c$, i.e. $\tau = G\gamma + \mu\dot{\gamma}$, in which G and μ are two material parameters and γ is the deformation from the initial (rest) state. Note that in this frame the solid-liquid

transition is expected for the critical deformation γ_c, and the minimum stress value making it possible to reach this value is $\tau_c = G\gamma_c$.

Figure 1 illustrates this behavior: when the stress applied to the material initially at rest, one observes a progressive increase of the apparent shear rate, which in fact simply reflects the instantaneous flow in the solid regime (and is thus dependent on the rate of increase of the stress); then we enter the liquid regime beyond a critical deformation associated with a critical stress (the yield stress); when decreasing the stress while the material is in its liquid regime the material progressively stops flowing as the stress reaches the yield stress, and we find the typical stress plateau of yield stress fluids in a shear stress vs shear rate diagram.
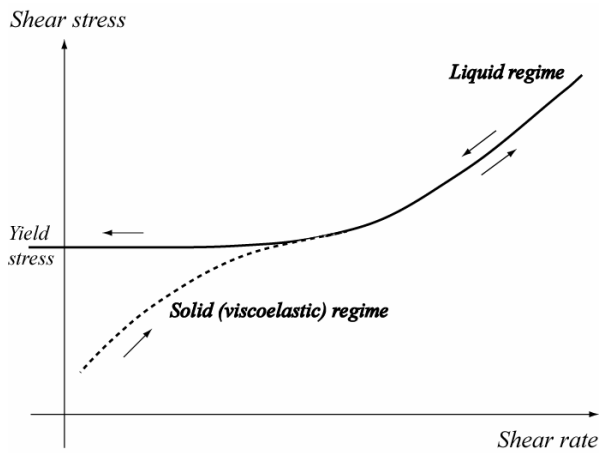

Figure 1: Typical aspect of an apparent flow curve of a yield stress fluid during a stress increase-decrease sweep.

The physical origin of this behavior lies in the fact that concentrated emulsions, foams, colloidal suspensions or polymeric gels are **jammed systems** [5] since they are made of a great number of elements (droplets, bubbles, particles, polymer chains) in strong (direct or at distance) interaction in a liquid. These elements form a continuous network of interactions which has to be broken for flow to occur. The strength of this network, from which the yield stress derives, is related to the current distribution of positions and states of the elements within the liquid.

Similarities with simple fluids
In their liquid regime yield stress fluids often look like simple fluids. Indeed for example in conduit flows the flow rate increases with the pressure drop [2] or in free surface flows the flow rate increases with the flow thickness as in usual hydraulics [6]. Other aspects typical of usual simple fluid flows may be observed in free surface flows: a hydraulic jump separating a sub-critical and a super-critical regimes [7] (see Figure 2), the equations describing this phenomenon being rather similar to equations in conventional hydraulics; flow around an obstacle (see Figure 3); roll waves [6].

Also one expects a transition to turbulence for sufficiently fast flows [8-13] but so far the criterion governing this transition is not well-known and the experimental data able to confirm theoretical approach are scarce. In this context it is sufficient to consider that the turbulent regime occurs when the Reynolds number, in which the viscosity is replaced by the apparent viscosity, is larger than a critical value. Obviously such an approach is relevant only at sufficiently large stress or flow rate for which the apparent viscosity is almost constant (i.e. independent of the shear rate).

Figure 2: "Laminar" hydraulic jump in the free surface flow of a mud in an inclined channel (width=20cm). The flow direction is indicated by the small arrow. The fluid from upwards (supercritical) has a large velocity and small thickness, it penetrates the fluid downwards flowing with a larger thickness in the subcritical regime.

Figure 3: Channelized flow of a mud: flow around an obstacle, parallel lines have been drawn upstream on the free surface, these lines deform was the fluid passes over the obstacle.

In fact the apparent fluid behaviour under sufficiently large stresses of yield stress fluids is rather easily explained by physical considerations. Indeed in general such materials are made of a lot of elements jammed within a given volume of liquid. The elements may develop various types of interactions through the liquid, which in particular govern the solid regime. In addition hydrodynamic interactions take place. The forces developed by the element interactions, e.g. electrostatic forces, van der Waals attractions, steric repulsion between surfaces with adsorbed polymers, droplet or bubble deformation forces, grain frictions, do not vary much with the local shear rate amplitude. In contrast the viscous dissipation due to the flow of liquid increases with the shear rate. At low shear rates the element interaction dominate, we are in the **pasty regime**. At sufficiently large shear rates the viscous dissipation due to liquid flow is dominant and the material behaves as a suspension of non-interacting elements [3]. In this **hydrodynamic regime** the material is expected to exhibit a constant viscosity depending essentially on the concentration and shapes of elements.

Specificities

The main specific aspect of yield stress fluids is obviously due to the existence of the solid regime in their behaviour: under sufficiently low stress (relatively to their yield stress) they behave as solids. Thus, in contrast with simple liquids defined as materials taking the shape of the container, they can remain at rest with the shape they were initially given, over some solid surfaces. For example, when they are poured over an inclined plane, they form deposits of significant thickness, an effect which is not due to surface tension effects. Indeed the theory predicts that for example for a uniform fluid thickness over an inclined plane with a slope i, the final thickness after flow stoppage will be $h_c = \tau_c / \rho g \sin i$ (in which ρ is the fluid density and g the gravity). As long as the fluid thickness is larger than h_c the material flows, an effect illustrated in Figure 4. This phenomenon is used in many applications (shaving foams, mayonnaise, etc) and explains the formation of mudflow deposits over steep slopes. A more thorough examination of such deposits shows that it is possible to relate the shape of the deposit from with the yield stress [14].
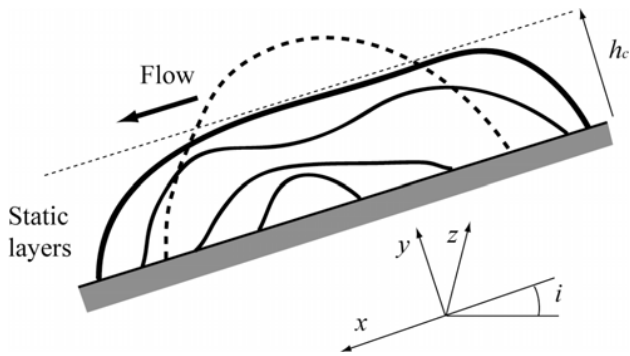

Figure 4: Different possible aspects (in 2D) of layers (dark lines) of a yield stress fluid on an inclined plane. The critical height is represented by a dotted line parallel to the solid plane. The (virtual) heap represented by the dashed line cannot remain at rest because its thickness is somewhere larger than the critical thickness.

Another typical aspect of yield stress fluid flows is the coexistence of a solid and a liquid regimes within the material. Indeed except in viscometric flows the stress distribution is in general strongly heterogeneous so that in some regions the stress is larger than the yield stress while in some other regions it is smaller than the yield stress. This for example leads to the well-known plug effect (see Figure 5) in conduit flows: the fluid is sheared along the wall but close to the center of the conduit the stress falls to zero so that the fluid is unsheared. There is thus a plug region around the conduit axis, which is transported by the rest of the fluid without being deformed.
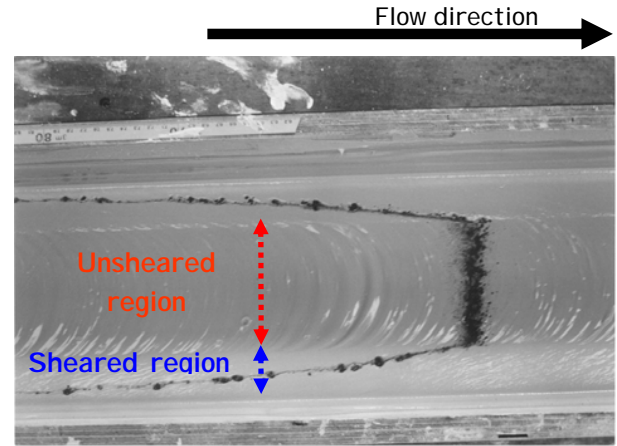

Figure 5: Coexistence of the two regimes (solid and liquid) in the free surface flow of a mud: the central part (plug) is unsheared while the lateral parts are sheared.

Note that the coexistence of a liquid and a solid regimes makes the numerical simulations of yield stress fluid flows a challenging problem. It indeed implies to be able to determine at each step of the simulation the boundary between the two regimes, which obviously can evolve. Usually, for the sake of simplicity, one assumes that the material is highly viscous below a critical shear rate, which avoids considering two strictly different regimes and makes it possible to reasonably approach real flow characteristics.

An original property may be observed with yield stress fluids: a flow instability at vanishing velocity. The Saffman-Taylor instability occurs when a fluid of low viscosity pushes, in a confined space, a fluid of larger viscosity, at a velocity larger than a critical value which depends on the width of the interface between the fluids. This instability is due to a competition between viscous and surface tension effects: at sufficiently large velocity it is energetically more efficient for the low viscous fluid to penetrate through the other one in the form of fingers than to push the fluid along a straight front. Obviously this fingering disappears when the velocity decreases. For a yield stress fluid the instability can occur even at vanishing velocity because its apparent viscosity tends to infinity in that case. The theory [15] also predicts the size of the fingers as a function of the yield stress, for example in the limit of vanishing velocity we have:

$$\lambda_m = 2\pi \sqrt{\frac{3\gamma_{LG} b}{\tau_c}}$$

in which γ_{LG} is the interfacial tension between the two fluids, and b half the separating distance between the plates in which the fluids flow. Moreover, since we are dealing with yield stress fluids, as soon as the fingers have been formed they keep their shape. This instability is usually observed when separating two solid surfaces between which there is a thin layer of paste (see Figure 6). This may be observed in our everyday life for example with glues, purees, paints, etc. In that case, when the layer is sufficiently thin, the air rapidly penetrates the gap between the plates, which induces a flow similar to that obtained when air pushes the fluid between two fixed plates.
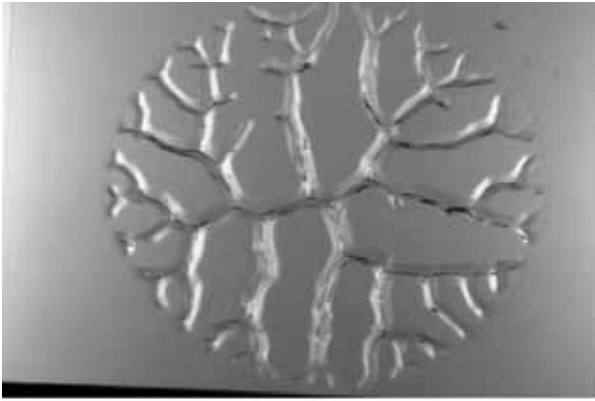
Figure 6: Tree-like aspect of the viscous fingering after separation of two plates initially in contact via a thin layer of a yield stress fluid.

Open questions

We already mentioned the difficulty to simulate complex flows involving both regimes. Another question concerns the effective existence of a solid regime. Some authors suggested [16] that the yield stress fluids in fact always flow even under low shear stresses. This is however mainly a philosophical problem because it is recognized that in practice, within the usual experimental times, yield stress fluids exhibit a true apparent yield stress.
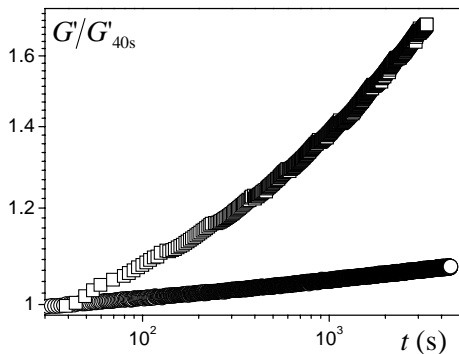

Figure 7: Elastic modulus (scaled by the value reached at 40s) of the pure (circles) and loaded (squares) emulsions as a function of time.

A much more important problem lies in the fact that we are dealing with out of equilibrium system generally made of small (colloidal) elements in interaction and submitted to thermal agitation. As a consequence the structure at rest slightly evolves in time so that its elastic modulus increases (see Figure 7). This is a typical property of thixotropic materials. Moreover, in the liquid regime, the apparent viscosity may take some time before reaching its steady value. This is for example observed from local measurements in a fluid with the help of Magnetic Resonance Imaging velocimetry in a Couette flow (see Figure 8) [17]: the profile of tangential velocity in the gap progressively evolves towards its steady value after a sudden change in the rotation velocity of the inner cylinder. This effect is also typical of thixotropic materials and is supposed to be due to a progressive evolution of the structure during flow, for example an increase or a decrease of the size of the flocs formed by the elements (when they are able to somewhat aggregate) with the flow intensity.
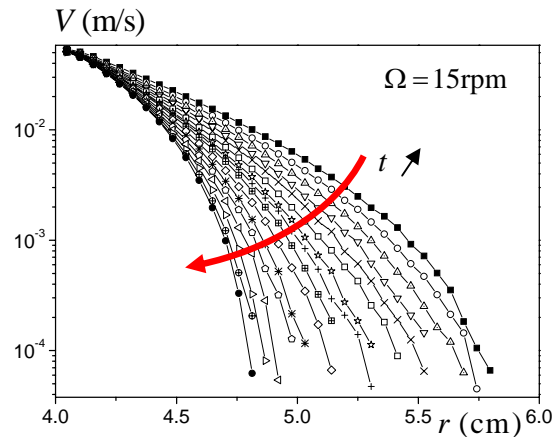

Figure 8: Velocity profiles at different times in a wide-gap Couette geometry as measured by MR velocimetry for the loaded emulsion. Up to the initial time the material was presheared at a high velocity, at the initial time the rotation velocity is abruptly decreased to 15rpm. The different times at which the velocity profiles are recorded are: (from right to left) 30, 60, 90, 120, 150, 180, 210, 240, 270, 330, 390, 450, 540, 660, 780, 900s.

In fact thixotropic effects can be very different depending on the elements interactions. For example if we consider two model concentrated water-in-oil emulsions, either pure or loaded with clay particles, we get two very different behaviours: in the solid regime the restructuring of the material is almost negligible with the pure emulsion and significant with the loaded emulsion (see Figure 7); in the liquid regime the apparent yield stress of the materials significantly differ but the flow curve of the loaded emulsion fall along the flow curve of the pure emulsion (see Figure 9). Moreover for a stress below the yield stress for the loaded emulsion the flow progressively stops, so that there is a critical shear rate ($\dot{\gamma}_c$) below which no steady-state can be obtained, in contrast with what is generally assumed for simple yield stress fluids. This implies that if one imposes an apparent shear rate, $\dot{\gamma}$, smaller than $\dot{\gamma}_c$, to the loaded emulsion it will develop a shear localization, leaving in steady-state some regions unsheared, and shearing the rest of the material at $\dot{\gamma}_c$ so as to obtain $\dot{\gamma}$ on average. Here this effect is likely due to the progressive aggregation of droplets via clay particle links. In the initial state, i.e. just after a preshear this aggregation is negligible and the two materials have a similar behaviour (see Figure 9).

One of the first difficulty due to thixotropy is this tendency to shear-localization in steady-state [18-19]. Another important difficulty is the fact that so far we hardly can identify clear rheological parameters making it possible to provide a relevant characterization of the materials. This is related to the fact that the modelling of thixotropic fluid flows remain an extremely difficult problem, and there is as yet no experimental study providing a precise and relevant set of data under a wide range of conditions not only in the liquid but also in the solid regimes of the materials and at the transition between the two regimes. This is finally also related to our poor understanding of these materials from a physical point of view. A basic analogy with glass behaviour and in particular glass aging was recently emphasized, but it does not make it possible to significantly progress in that field.
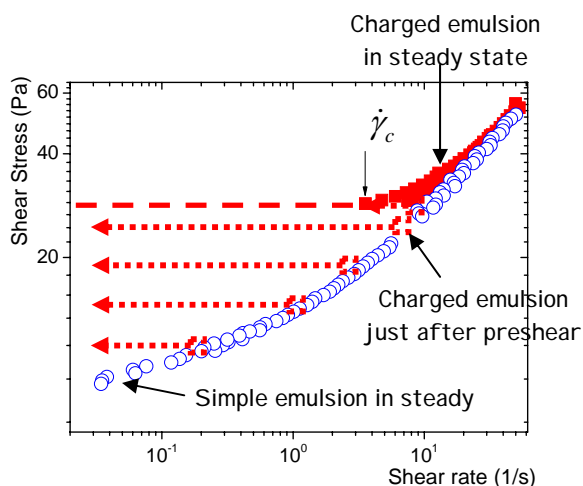
Figure 9: Steady-state shear stress vs shear rate for the pure (circles) and loaded (squares) emulsions as deduced from local measurements within the wide-gap rheometer under different torque values. The dotted squares correspond to the initial behavior of the loaded material, and the dotted lines show its evolution in time.

Conclusion

Yield stress fluids constitute a very interesting field in which one may at the same time find some usual characteristics of simple fluids and some simple solids. However these materials also offer great challenges for their modelling especially when they are in addition strongly thixotropic, i.e. with a time-dependent behaviour. The field of thixotropic fluids is one the main wide-open field in non-Newtonian fluid mechanics.

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