

## Avalanche Behavior in Yield Stress Fluids

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We show that, above a critical stress, typical yield stress fluids (gels and clay suspensions) and soft glassy materials (colloidal glasses) start flowing abruptly and subsequently accelerate, leading to avalanches that are remarkably similar to those of granular materials. Rheometrical tests reveal that this is associated with a bifurcation in rheological behavior: for small stresses, the viscosity increases in time; the material eventually stops flowing. For slightly larger stresses the viscosity decreases continuously in time; the flow accelerates. Thus the viscosity jumps discontinuously to infinity at the critical stress. We propose a simple physical model capable of reproducing these effects.

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Fluids that do not flow unless they are subjected to a certain load are widely used in both cosmetic and industrial applications; well-known examples are toothpaste, fresh concrete, paints, shaving foam, and drilling fluids. The flow behavior of these so-called yield stress fluids can be defined as the stress (the shear force per unit area) going to a finite nonzero value, the yield stress, if the shear rate (the velocity gradient) goes to zero. As a consequence, when subjected to a shear stress, the response of an ideal yield stress fluid is a slow shear flow provided the stress is slightly beyond the yield point: its steady-state viscosity (the ratio of shear stress and shear rate) diverges in a continuous fashion when the yield stress is approached from above (Fig. 1) [1–3].

In practice, however, the yield stress is mostly due to the microstructure of the fluid that resists large rearrangements: the system is jammed [4] and stops or starts flowing abruptly. When submitted to flow, this microstructure is partly destroyed, which is generally observed in rheological tests as a viscosity that decreases in time: the system is said to be thixotropic. In addition, for most of these systems at rest the microstructure reforms or evolves spontaneously: the system is said to age. If the microstructure reestablishes at rest one observes an increase of an apparent yield stress with time [5]. The mechanical behavior of these systems consequently results from the competition between aging and progressive “rejuvenation” (destruction of the microstructure) by the shear flow.

In this Letter we show that these observations are at the basis of the experimental difficulties one encounters when measuring a yield stress [3]: very different answers can be obtained depending on the experimental protocol. Because of the effect of flow on the microstructure, for different “typical” yield stress fluids, it is in fact impossible to unambiguously define a yield stress. We show that typical yield stress fluids (gels and clay suspensions) and soft glassy materials [6] (colloidal glasses) do not start flowing slowly. The incipient flow destructures the materials, entailing a viscosity decrease, which in turn accelerates

the flow and so on: avalanche behavior results that is remarkably similar to what is observed for granular matter. Combined with the spontaneous restructuration at rest, we show that a bifurcation in the rheological behavior occurs: for a given load, the fluid either stops flowing altogether or fluidizes, leading to rapid flows. The rheological measurements show that the viscosity jumps in a discontinuous way to infinity at a critical stress, contrary to the continuous divergence anticipated for yield stress fluids. In addition, due to the aging, this critical stress is not an intrinsic property, but depends on the (shear) history of the sample. All these observations show that yield stress is ill-defined.

The simplest mechanical test to determine the yield stress of a given fluid is the inclined plane [7]. Yield stress fluids can be defined as fluids that can support their own weight; as a consequence, a heap on an inclined plane will not flow if the slope is below a critical value. In such inclined plane tests, a large amount of the material is

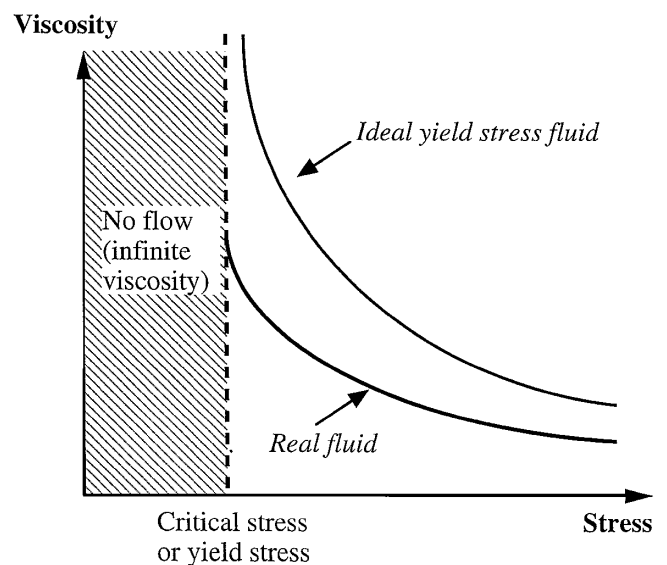


FIG. 1. Schematic of a typical steady-state flow curve (viscosity vs shear stress) for an ideal yield stress fluid and a real fluid.

deposited on an inclined plane (of slope  $\theta$ ) and flows under gravity. When the thickness of the resulting layer is much smaller than its extent over the plane, the shear stress distribution at rest is [8]:  $\sigma = \rho g(h - y) \sin(\theta)$ , where  $y$  is the height above the plane. For an ideal yield stress fluid the flow stops when the maximum shear stress (along the plane) becomes equal to the yield stress  $\sigma_y$ , i.e., when the thickness  $h$  equals the critical thickness  $\sigma_y / \rho g \sin(\theta)$ .

We performed inclined plane tests on an aqueous clay suspension (bentonite) at a solid concentration of 4%. This system is weakly flocculated [5,7]: the microstructure is that of a colloidal gel. The experiments (Fig. 2) reveal that for a given thickness of the initial deposit there exists a critical slope (corresponding to a critical stress) below which the restructuration (the aging) dominates over the destruction of the microstructure (the shear rejuvenation): the fluid stops moving more rapidly than would an ideal yield stress fluid. On the other hand, for a slope that is only slightly steeper, the shear rejuvenation fluidizes the sample and the fluid flows much faster. The shear rejuvenation leads to a stronger flow, leading in turn to a decrease in viscosity, accelerating the flow and so on: an avalanche results, transporting the fluid over large distances (Fig. 2).

This in fact reproduces at a smaller scale the spectacular liquefaction of so-called “sensitive clays,” clayey soils in Canada and Scandinavia for which landslides were reported over long distances [9]. The shear rejuvenation therefore accounts for the hitherto unexplained observation that clayey soils can liquefy abruptly if they are subjected to small variations of stress.

The “horseshoe” form remaining at the top of the inclined plane and the detailed form of the deposit are remarkably similar to those observed for avalanches in granular materials [10]: if exactly the same experiment is performed using sand, the same patterns are obtained.

This indicates that the common origin of the avalanches is indeed the “unjammed” of a jammed system. This suggests that, as is the case for granular systems, steady-state viscosity is infinite already for a finite stress and does not diverge continuously when the flow velocity goes to zero, as it would for an ideal yield stress fluid.

That this is indeed the case, and that the phenomenon is general for yield stress fluids, follows from rheometrical tests on the bentonite clay suspension, a polymer gel and a colloidal glass. As a typical polymeric yield stress fluid we use a commercial hair gel: its microstructure is that of a physical gel. As a colloidal glass, we use a 3% colloidal suspension of Laponite, a synthetic colloid, dissolved in water [11,12]. This glass does not have any structuration at large length scales, making its microstructure fundamentally different [12]. For the rheology, we used parallel plate and vane geometries [7]. The vane induces a flow very close to that of coaxial cylinders but avoids wall slip [7]. For the same reason, the outer cylinder was covered with sandpaper. For the parallel plate geometry, both surfaces were covered with sandpaper. For the two geometries the ratio of both the free and the sheared surface to sheared volume differ; therefore important perturbing effects (such as edge effects, evaporation, wall slip, etc.) would yield different apparent viscosities. Good quantitative agreement was found between the two different geometries proving the validity of the data.

For each test the material was presheared and left at rest for a given fixed time to obtain a reproducible initial state. Starting from this state, we observe for the three different materials that above a critical stress, the viscosity decreases to reach a low steady-state value after a long time. For a stress smaller than this critical value the viscosity increases indefinitely. Therefore, the flow either stops completely or evolves through the avalanche effect discussed above

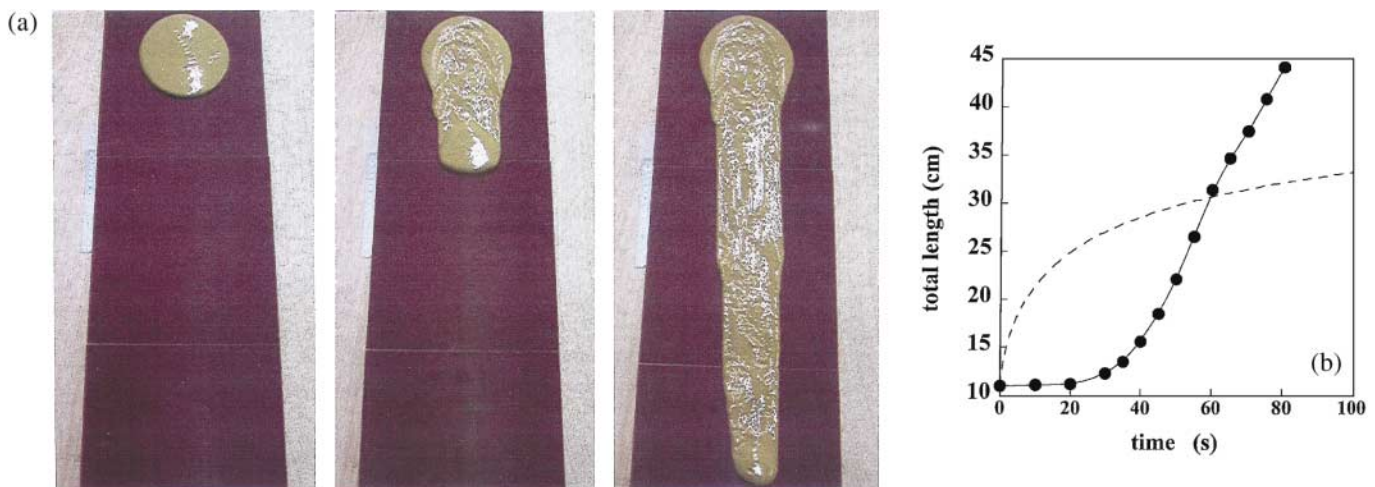


FIG. 2 (color). (a) Avalanche flow of a clay suspension over an inclined plane covered with sandpaper. The suspension was presheared and poured onto the plane, after which it was left at rest for 1 h. The pictures are taken at the critical angle for which the suspension just starts to flow visibly. (b) Distance traveled by the deposit as a function of time compared to that of a theoretical ideal yield stress fluid (dashed line).

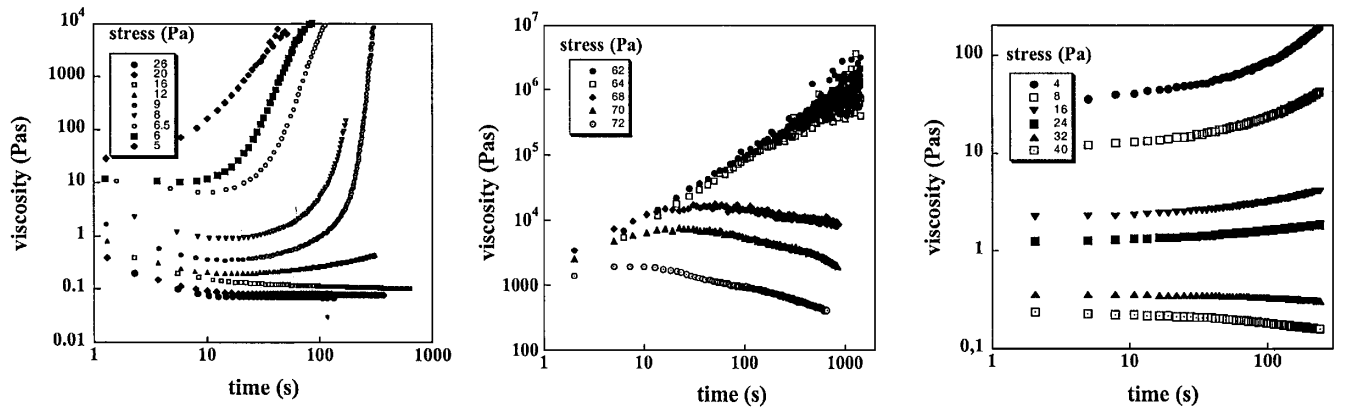


FIG. 3. Bifurcation in the rheological behavior: viscosity as a function of time for three completely different systems: from left to right, a weakly flocculated clay suspension: bentonite (solid fraction: 4%) in water; a polymer gel: a commercial hair gel (Vivelop) and a colloidal glass: 3% Laponite RD in water.

towards a rapid flow. The competition between aging and shear rejuvenation thus shows up as a bifurcation in the viscosity (Fig. 3). The important observation is that for stresses slightly smaller than  $\sigma_c$ , the viscosity increases in time, until the flow is halted altogether: the steady-state viscosity is infinite. On the other hand, for a stress only slightly above  $\sigma_c$ , the viscosity reaches a (low) steady-state value  $\eta_0$ . This implies that at the critical stress, the steady-state viscosity jumps discontinuously from infinity to a finite and low value at  $\sigma_c$ , which is indeed observed in the rheological experiments. This is in striking contrast with the ideal yield stress fluid of Fig. 1 but agrees with the conclusions from the inclined plane test.

A simple model that describes the competition between aging and rejuvenation captures the essentials of our results. We suppose that the degree of jamming can be described by a single parameter  $\lambda$ .  $\lambda$  represents for instance the degree of flocculation for clays [13], is a measure of the free energy landscape for glasses [14], or gives the fraction of particles in potential wells for colloidal suspensions [7]. For an aging system, at rest  $\lambda$  increases at a constant rate  $1/\tau$  where  $\tau$  is the characteristic time of evolution of the structure. The rate of decrease of  $\lambda$  under shear is assumed proportional to both the shear rate and the degree of jamming, leading to an evolution equation for  $\lambda$ :  $\frac{d\lambda}{dt} = \frac{1}{\tau} - \alpha\lambda\dot{\gamma}$ ,  $\alpha$  and  $\tau$  being system dependent constants,  $\dot{\gamma}$  the shear rate, and  $t$  the time. To relate flow and structure, as a general viscosity function we consider  $\eta = \eta_0(1 + \lambda^n)$ ; when the structure is entirely destroyed,  $\eta$  tends towards an asymptotic value  $\eta_0$ .

In steady state it follows that  $\lambda = 1/\alpha\tau\dot{\gamma}$ , so that the shear stress writes  $\sigma = \eta\dot{\gamma} = \eta_0\dot{\gamma}[1 + (\alpha\tau\dot{\gamma})^{-n}]$ . When  $n < 1$ ,  $d\sigma/d\dot{\gamma}$  is always positive: the flows are stable, but the fluid has no yield stress since  $\sigma \rightarrow 0$  when  $\dot{\gamma} \rightarrow 0$ ; it is a simple shear-thinning fluid. Yielding behavior ( $\sigma$  does not tend to 0 when  $\dot{\gamma} \rightarrow 0$ ) appears for  $n \geq 1$ ; it is especially interesting to remark that ideal yield stress fluids ( $\sigma$  tends to a finite value when  $\dot{\gamma} \rightarrow 0$ ) appear only as a marginal case for  $n = 1$ .

When a constant shear stress is applied the evolution equation writes  $\frac{d\lambda}{dt} = \frac{1}{\tau} - \alpha\lambda\sigma/\eta_0(1 + \lambda^n)$ . The variations of  $\lambda$ , and thus the viscosity variations, are dictated by the relative values of the two terms of the right-hand side of this equation. For  $n > 1$ , the time-dependent solution of this simple model qualitatively reproduces the bifurcation observed in the experiments (Fig. 4). In time the viscosity tends either towards infinity or to a small constant value depending on whether the stress is larger or smaller than a critical value  $\sigma_c$ , which depends on the initial state of the structure,  $\lambda_0 = \lambda(t=0)$ , where  $t=0$  is the moment the stress is first applied, i.e.,  $\sigma_c = \eta_0(1 + \lambda_0^n)/\alpha\tau\lambda_0$ .

Moreover when  $n > 1$ , in steady state  $\sigma$  decreases ( $d\sigma/d\dot{\gamma} < 0$ ) for shear rates smaller than a critical value. In that case flows are unstable, as can be shown from a simple linear stability analysis [10]. As a consequence the maximum viscosity that can be reached is  $\eta_0 n/(n-1)$ , which corresponds to the viscosity of the fluid at the minimum value of shear rate [ $\dot{\gamma}_c = (n-1)^{1/n}/\alpha\tau$ ] for which

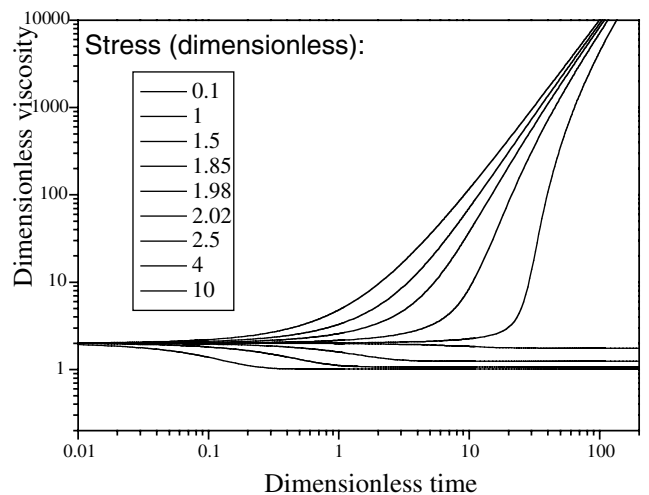


FIG. 4. Dimensionless viscosity  $\eta/\eta_0$  vs dimensionless time  $t/\tau$  as predicted by the model for different applied dimensionless stresses ( $\sigma\tau/\eta_0$ ). Here we used  $n = 2$ ,  $\alpha = 1$ , and  $\lambda_0 = 1$ .

stable flows may be obtained, i.e.,  $(d\sigma/d\dot{\gamma})_{\dot{\gamma}=\dot{\gamma}_c} = 0$ . The model thus shows that the competition between aging and shear rejuvenation directly leads to a viscosity that does not diverge when the flow velocity goes to zero but jumps discontinuously to infinity at a critical stress, in accordance with the rheological measurements. Consequently, when a constant shear rate rather than a constant stress is applied to the material, stable homogeneous flows can occur only when  $\dot{\gamma} \geq \dot{\gamma}_c$ . For smaller shear rates the flows are unstable; in practice, the material will either fracture or produce shear banding instabilities (shear localization) [7,15,16]. The latter was indeed observed experimentally for both Laponite and bentonite studied: for shear rates smaller than a critical value, shear localization was observed [16], showing that their rheological behavior is fundamentally different from that of an ideal yield stress fluid.

In conclusion, yield stress cannot be considered separately from thixotropy. We show here for a number of typical yield stress fluids that both are strongly interconnected: they are the result of the jamming and unjamming of the microstructure of the materials. It is likely that the same holds for granular flows, which undergo similar transitions between different states; this in particular leads to avalanche processes, equilibrium angle hysteresis, and sudden stoppage below a critical thickness [6,17] all of which is reproduced here using yield stress fluids. Moreover we show using a colloidal glass that the response to flow of yield stress fluids and glassy systems is similar, since they can undergo aging and rejuvenation and their mechanical behavior depends on the history of the sample [18,19]. The observation of very similar flow behavior for such different classes of systems lends some credence to the “generalized phase diagram” for jammed systems such as granular, gel-like, and glassy systems [20]: a common framework for the physical characterization of these very different systems is provided by their rheological properties. In addition, this highly nonlinear behavior should be taken into account for the formulation and handling of paints, inks, cement, muds, etc.

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