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Guillaume Ovarlez, Sylvie Cohen-Addad, Kapil Krishan, Julie Goyon, Philippe Coussot

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G. Ovarlez[a,*], S. Cohen-Addad[b,c], K. Krishan[d], J. Goyon[a], P. Coussot[a]

[a] *Université Paris Est, Laboratoire Navier (UMR CNRS 8205), 2 allée Kepler, 77420 Champs-sur-Marne, France*
[b] *Univ. Paris 6, UMR 7588 CNRS-UPMC, INSP, 4 place Jussieu, 75252 Paris cedex 05*
[c] *Université Paris-Est, LPMDI, 5 Bd Descartes, 77454 Marne-la-Vallée, France*
[d] *P&G Kobe Technical Center, 1-17 Koyocho Naka, Higashinada-ku, Kobe, 6580032 Hyogo, Japan*

Abstract

Materials such as foams, concentrated emulsions, dense suspensions or colloidal gels, are yield stress fluids. Their steady flow behavior, characterized by standard rheometric techniques, is usually modeled by a Herschel-Bulkley law. The emergence of techniques that allow the measurement of their local flow properties (velocity and volume fraction fields) has led to observe new complex behaviors. It was shown that many of these materials exhibit shear banding in a homogeneous shear stress field, which cannot be accounted for by the standard steady-state constitutive laws of simple yield stress fluids. In some cases, it was also observed that the velocity fields under various conditions cannot be modeled with a single constitutive law and that nonlocal models are needed to describe the flows. Doubt may then be cast on any macroscopic characterization of such systems, and one may wonder if any material behaves in some conditions as a Herschel-Bulkley material. In this paper, we address the question of the existence of a simple yield stress fluid behavior. We first review experimental results from the literature and we point out the main factors (physical properties, experimental procedure) at the origin of flow inhomogeneities and nonlocal effects. It leads us to propose a well-defined procedure to ensure that steady-state bulk properties of the materials are studied. We use this procedure to investigate yield stress fluid flows with MRI techniques. We focus on nonthixotropic dense suspensions of soft particles (foams, concentrated emulsions, Carbopol gels). We show that, as long as they are studied in a wide (as compared to the size of the material mesoscopic elements) gap geometry, these materials behave as 'simple yield stress fluids': they are homogeneous, they do not exhibit steady-state shear banding, and their steady flow behavior in simple shear can be modeled by a local continuous monotonic constitutive equation which accounts for flows in various conditions and matches the macroscopic response.

Keywords: Yield stress fluid; Viscoplastic flow; Local rheology; Herschel-Bulkley

1. Introduction

Materials such as dense suspensions, colloidal gels, microgel suspensions, concentrated emulsions or foams, are yield stress fluids: below a yield stress τ_y, they do not flow[1]. Their steady flow behavior in simple shear is characterized in standard rheological experiments, e.g. in a Couette or a cone-and-plate geometry [1]. It is usually well accounted for by a Herschel-Bulkley equation $\tau(\dot{\gamma}) = \tau_y + \eta_{HB}\dot{\gamma}^n$, the parameters of which depend on the details of the material microstructure [2]. The ability of such models to help predicting flows in more complex configurations, e.g. in extrusion flows [4], then relies on the robustness of the characterization in simple shear.

However, the recent emergence of techniques [2, 5] that allow the measurement of local flow properties (velocity and volume fraction fields) has led to revisit the behavior of pasty materials. The reason is that, in rheometric experiments, one only measures macroscopic quantities (torque $T(\Omega)$ vs. rotational velocity Ω). The material constitutive law relating local quantities (shear stress $\tau(\dot{\gamma})$ vs. shear rate $\dot{\gamma}$) is then derived under the assumption that the flow and the material are homogeneous, and that the material can be modeled as a continuum with a local constitutive law. Local measurements have shown that these conditions are actually not met for some materials. The observations of steady flow inhomogeneities (shear banding) in homogeneous stress fields have in particular shown that, in addition to their yield stress τ_y, some materials are also characterized by a critical shear rate $\dot{\gamma}_c$ below which they cannot flow steadily [6, 7, 8]. Moreover, in some materials, it was found that flows under various conditions cannot be described by a single local constitutive law [9, 10], which led to propose nonlocal modeling of their behavior [9, 11]. These observations contrast with standard laws inferred from purely macroscopic measurements, such as that of Herschel-Bulkley, which are local and allow steady flow at any low shear rate $\dot{\gamma}$.

Doubt may finally *a priori* be cast on any macroscopic characterization of yield stress fluids, and one may wonder if any material behaves in some conditions as a Herschel-Bulkley material. The question we address in this paper is whether such a simple yield stress fluid behavior exists or not. In Sec. 2, we first focus on the shear banding issue; then, in Sec. 3, we investigate the consistency between yield stress fluids flows and a local modeling of their behavior. Both sections are structured

*corresponding author: guillaume.ovarlez@ifsttar.fr

[1] We neglect slow creep flows driven by coarsening dynamics in foams and emulsions[3].

in the same way. We first review experimental studies of yield stress fluid flows in the literature: we present the main sources of shear banding (Sec. 2.1) and of discrepancy between macroscopic and local behavior (Sec. 3.1). This leads us to distinguish three classes of materials: thixotropic materials (such as colloidal gels), nonthixotropic dense suspensions of rigid particles (such as noncolloidal suspensions and colloidal glasses), and nonthixotropic dense suspensions of soft particles (such as foams, concentrated emulsions and Carbopol gels). We discuss the role of thixotropy and shear-induced migration in the studied complex behaviors for the first two classes of materials. We then focus on nonthixotropic dense suspensions of soft particles (Secs. 2.2 & 3.1), whose case is yet unclear as contradictory observations are reported in the literature. We first show that the above mechanisms should not be at play in these materials. We then point out issues related to the procedure used to study their flows. We propose a well-defined procedure to ensure that steady-state bulk properties of the materials are studied, and we investigate the flows of several foams, concentrated emulsions, and a Carbopol gel, with MRI techniques (Secs. 2.3 & 3.2). We show that, as long as they are studied in a wide (as compared to the size of the material mesoscopic elements) gap geometry, these materials behave as 'simple yield stress fluids': they are homogeneous, they do not exhibit steady-state shear banding, and their steady flow behavior in simple shear can be modeled by a local continuous monotonic constitutive equation which accounts for their flows in various conditions and matches their macroscopic response.

2. Shear banding

In this section, we address the question of shear banding in yield stress fluids. We discuss the phenomenon and the origins identified in two classes of such materials: structural inhomogeneities in thixotropic systems; volume fraction inhomogeneities in dense suspensions of rigid particles. We then investigate flow stability in nonthixotropic dense suspensions of soft particles. We discuss the relevance of the above mechanisms of shear banding in these systems. We use a procedure designed to circumvent possible artefacts and show that these materials do not display any steady-state shear banding, in contrast with several results of the literature. Possible explanations of these discrepancies are proposed at the end of the section.

2.1. Phenomenon and possible origins

Shear banding in yield stress fluid flows here refers to the coexistence of liquid-like (sheared) and solid-like (unsheared) bands at steady-state in a homogeneous shear stress field[2]. It is observed in some materials when a macroscopic shear rate $\dot{\gamma}_{\text{macro}}$ lower than a critical value $\dot{\gamma}_c$, which depends on the material, is applied. In these conditions, the material basically splits into a region flowing at $\dot{\gamma}_c$ and a non flowing region, whose relative extent ensures that the shear rate spatial average equals $\dot{\gamma}_{\text{macro}}$ [12, 13, 7]. The critical shear rate $\dot{\gamma}_c$ is the shear rate below which no stable flow is possible. It appears to be a new mechanical characteristics of these systems, in addition to their yield stress τ_y: at steady state, flow implies both $\tau \geq \tau_y$ and $\dot{\gamma} \geq \dot{\gamma}_c$. In an inhomogeneous stress field, flow localization occurs with any yield stress fluid; the signature of shear banding is then that the material flows at a nonzero shear rate $\dot{\gamma}_c$ at the interface between the flowing region (where the shear stress τ is higher than τ_y) and the non-flowing region (where $\tau < \tau_y$) [7]. This phenomenon is generally attributed to the fact that the theoretical material steady-state flow curve is non-monotonic [14].

Although shear-banding materials cannot flow steadily at a local shear rate smaller than $\dot{\gamma}_c$, any macroscopic shear rate $\dot{\gamma}_{\text{macro}} < \dot{\gamma}_c$ can be applied to such system by the relative motion of two boundaries. This implies that their macroscopic characterization may fail to represent their actual behavior. Indeed, typical constitutive laws $\tau(\dot{\gamma}_{\text{macro}})$ found near the yield stress in macroscopic experiments are such that $\dot{\gamma}_{\text{macro}}$ tends continuously to zero as τ approaches τ_y; this is the case of standard laws such as the Herschel-Bulkley law. This contrasts with the locally observed behavior of shear-banding materials where $\dot{\gamma}_{local}$ tends towards a critical shear rate $\dot{\gamma}_c \neq 0$ as τ approaches τ_y. This issue is illustrated in Fig. 1.

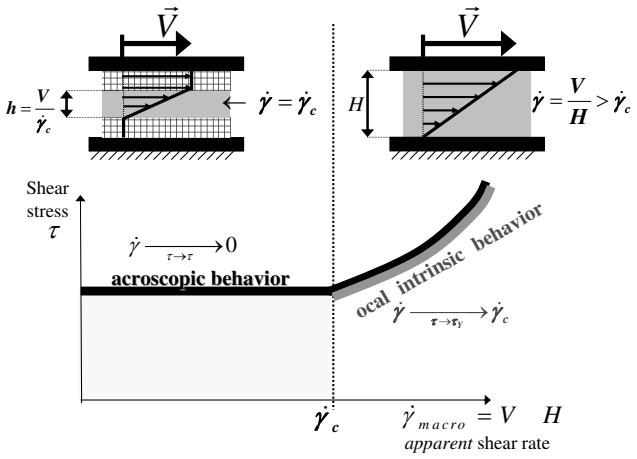

Figure 1: Illustration of the difference between the material intrinsic constitutive law (grey line) and the flow curve measured macroscopically in a homogeneous stress field (black line) in a shear-banding material. At high macroscopic shear rate ($\dot{\gamma}_{\text{macro}} > \dot{\gamma}_c$), flow is homogeneous and the macroscopic flow curve matches the local behavior. At low macroscopic shear rate ($\dot{\gamma}_{\text{macro}} < \dot{\gamma}_c$), shear banding occurs: the local shear rate in the flowing material – $\dot{\gamma}_c$ – differs from the macroscopic shear rate and the macroscopic flow curve does not match the local behavior.

Identifying which materials exhibit shear bands and which do not is thus of high importance; only the last ones may behave like simple yield stress fluids and be characterized using standard rheometric experiments. Two possible origins of shear-banding in yield stress fluids have already been identified experimentally: structural inhomogeneities in thixotropic systems, and volume fraction inhomogeneities in dense suspensions of rigid particles.

[2] It is also sometimes called 'discontinuous' shear banding to avoid confusion with other kinds of flow inhomogeneities.

2.1.1. Structural inhomogeneities in thixotropic systems

Shear banding in yield stress fluids seems to be observed in all thixotropic colloidal gels [12, 15, 13, 16, 7]. In these materials, it is attributed to the competition between aggregation, which is due to attractive interactions and thermally activated structuration, and shear, which tends to break the aggregates. Starting from a liquid state, when the applied shear rate $\dot{\gamma}$ is too low, the material basically splits between a liquid-like region (where it is a suspension of independent aggregates) and a region that gets continuously structured under shear until it jams (i.e., a percolated network forms). Shear banding is also observed in dense suspensions of rigid noncolloidal particles, where it is attributed to competition between sedimentation-induced contact formation and shear-induced resuspension [17, 18]. When the local shear rate is too low, the shear resuspension mechanism is not sufficiently efficient to prevent contact formation between the particles and the material jams as a percolated network of particles form.

In these two kinds of systems, shear banding seems to result from shear/structure coupling, and from the development of two bands of different structures, similarly to what is observed in wormlike micellar systems [19]. Several models based on the idea that the material properties depend on its structure and that the structure depends on shear history [20, 21, 22] have indeed predicted shear banding in simple shear, as a result of the instability of some solutions of homogeneous structure, and describe correctly the flow inhomogeneities observed in thixotropic systems [2]. Such models can be an explanation for shear banding in a given material only if a macroscopic thixotropic behavior – i.e., dependent on shear history [23] – is found experimentally.

2.1.2. Volume fraction inhomogeneities in suspensions of rigid particles

Colloidal glasses are not much thixotropic: they get structured at rest and destructured at flow start-up, but their flow behavior does not depend significantly on their flow history (see e.g. [24]). The above mechanism is thus not expected to be at play in such systems. However, shear banding has recently been observed in these materials and explained to result from the development of an inhomogeneous volume fraction profile [25]. The basic idea is that a small change in the volume fraction may result in a significant change in the rheological properties, in particular in the yield stress value, which is an increasing function of ϕ. In a homogeneous stress field, at low applied shear rate, regions of lower ϕ (and thus of lower τ_y) may then flow while regions of higher ϕ (and thus of higher τ_y) are jammed. The development of an inhomogeneous volume fraction profile in a homogeneous stress field suggests an instability induced by a shear-concentration coupling [26, 25]. Banding resulting from volume fraction inhomogeneities has also been observed in dense rigid noncolloidal suspensions [17, 27], where shear-induced migration [28, 29, 30] creates jammed regions of volume fraction ϕ higher than the jamming packing fraction ϕ_m.

It is worth noting that these systems are made up of rigid particles. Indeed, although hydrodynamic interactions may play a role, recent theoretical developments [30] supported by experimental findings [27] show that the shear-induced migration efficiency is strongly enhanced by direct rigid contact forces between the particles. This characteristic may thus play a crucial role in the emergence of shear banding in these systems.

2.2. Nonthixotropic dense suspensions of soft particles

The case of nonthixotropic dense suspensions of soft particles (foams, concentrated emulsions, microgels) is less clear, as contradictory observations are reported in the literature. Bécu *et al.* [31] reported shear banding in adhesive emulsions, whereas Ovarlez *et al.* [32] did not observe any shear banding in the same system as well as in four other adhesive and nonadhesive emulsions. Rodts *et al.* [33] reported shear bands in 3D sheared foams, but Ovarlez *et al.* [34] did not observe any shear banding in the same system and in three other foams. In 2D foams (bubble rafts) Gilbreth *et al.* [35] reported shear banding whereas Katgert *et al.* [10] did not. This suggests that subtle differences in the materials, setup, or procedures, may exist, and have to be identified. In the following, we first discuss the possible relevance of the mechanisms mentioned in Sec. 2.1 in these systems.

2.2.1. Nonthixotropic behavior

The systems we deal with are basically nonthixotropic: their flow behavior does not show significant history dependence. This is illustrated in Fig. 2a, where the response of an emulsion, a foam, and a Carbopol gel to consecutive up and down stress ramps is displayed. Such experiment is classically used to assess the thixotropic behavior of a material [2]. During the up-ramp, as long as the shear stress is below the (static) yield stress, the material is strained in its solid regime; this explains why the strain rate is roughly constant and proportional to the ramp rate in a first stage [2]. Once the material flows, there is no significant difference between the responses to the up- and down-ramps: these materials can be considered as nonthixotropic. This contrasts with the hysteretic response observed in a thixotropic material (bentonite suspension, Fig. 2b), which is the signature of strong dependence of the behavior on flow history, i.e., of thixotropy.

This suggests that the mechanisms discussed in Sec. 2.1.1 should not be at play in the Carbopol gel, the foam and the emulsion, as their structure should remain homogeneous when they flow. Structuration at rest is nevertheless observed in some of these systems [39, 3, 40], i.e., their structure in their solid and liquid regimes may differ. E.g., after a preshear, foams progressively recover their elasticity at rest (G' increases in time) as coarsening induces structural rearrangements [39, 3]. Structuration at rest can lead to differences between the static and dynamic yield stress [40]. As detailed in Sec. 2.3.1, this can lead to transient flow inhomogeneities but not to steady-state bands.

Recently, models of soft-jammed systems such as concentrated emulsions (a nonlocal model [22] and the SGR model [21]) have been extended to account for possible bulk shear banding. Both models predict macroscopic shear banding associated with structural inhomogeneities. We stress that, in these models, the behavior depends on internal variables that depend

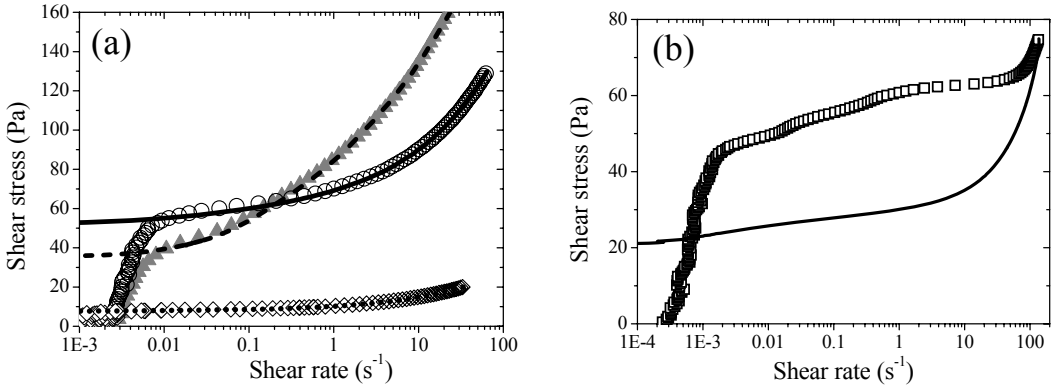

Figure 2: Shear stress τ vs. shear rate $\dot{\gamma}$ when consecutive up (symbols) and down (lines) stress ramps are applied, with a constant rate α, (a) to a foam (up triangles and dashed line, α=1 Pa/s), a Carbopol gel (empty circles and full line, α=0.5 Pa/s) and a concentrated emulsion (empty diamonds and dotted line, α=0.1 Pa/s), and (b) to a thixotropic (bentonite) suspension (α=0.5 Pa/s). A 4°angle cone-and-plate geometry is used, with serrated surfaces (60 mm diameter) for the foam, with sandblasted surfaces (40 mm diameter) for the other materials. The emulsion is composed of 1 μm diameter dodecane droplets dispersed at a 73% volume fraction in water stabilized by Sodium Dodecyl Sulfate at a 1 wt% concentration (see [36] for details). The Carbopol gel is made up of Carbopol 980 dispersed and neutralized at a 0.3% volume fraction in water (see [37] for details). The foam (Gillette Regular) consists of 27 μm diameter bubbles at a 92% volume fraction. The bentonite suspension is made up of (smectite) clay particles of length of order 1 μm and thickness 10 nm suspended at a 6% volume fraction in water [38].

on shear history, i.e., the system is supposed to be thixotropic: they cannot apply to *nonthixotropic* suspensions of soft particles.

2.2.2. *Material homogeneity*

As discussed in Sec. 2.1.2, volume fraction inhomogeneities may lead to shear banding. One may wonder whether they can be responsible for some of the reported shear bands in dense suspensions of soft particles. Shear-induced migration is well documented for rigid particles [28, 29, 17, 30, 27]. Migration of deformable particles, and particularly drops, has been much less studied. It is found that single droplets migrate away from rigid walls, due to asymmetric flows around deformed droplets [42, 43], but binary collisions tend to homogenize the systems for $\phi \geq 10\%$.

To test the possible emergence of volume fraction inhomogeneities in dense suspensions of soft particles, we study the evolution in time of the bubble or droplet volume fraction profiles when foams or concentrated emulsions are sheared in an inhomogeneous stress field (where particles migrate towards the low shear zones in suspensions of hard spheres). We use a wide gap Couette geometry, where the shear stress distribution is:

$$\tau(R) = \tau(R_i)R_i^2/R^2 \quad (1)$$

where R is the radial position in the gap and R_i is the inner cylinder radius. The materials and methods are described in detail in Ovarlez *et al.* [32, 34]. The average bubble diameter of the studied foam is 73 μm, its gas volume fraction is 89% [34]. Five emulsions are studied; their droplet diameter and droplet volume fraction are: (0.3 μm,75%), (1 μm,75%), (6.5 μm,75%) and (40 μm,88%); both adhesive and nonadhesive emulsions are studied [32]. The inner cylinder radius of the Couette cell is 4.1 cm; the gap is 1.9 cm wide. The systems are sheared at various constant rotational velocities Ω, ranging from 1 to 100 rpm, during long times (from 1 to 24 h, depending on the system). The Couette cell is inserted in a Magnetic Resonance Imager [44] and droplet or bubble volume fraction profiles are measured as in [17, 32].

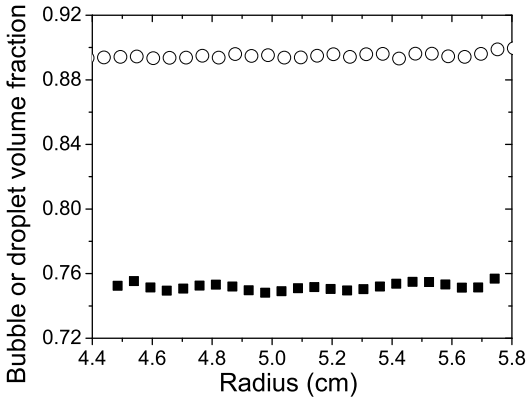

Figure 3: Concentration profiles observed within the gap of a Couette geometry by MRI techniques after shearing a concentrated adhesive emulsion (squares, average applied strain: 70000), and a foam (open circles, average applied strain: 12000). The emulsion is composed of 6.5 μm diameter silicone oil droplets dispersed at a 75% volume fraction in a mixture of 50 wt% glycerine and 50 wt% water stabilized by Brij and trimethyl tetradecyl ammonium bromide at a 6.5 wt% concentration [32]. The foam is composed of 73 μm diameter bubbles at a 89% volume fraction in a SLES foaming solution described in [34].

Typical results are shown in Fig. 3. We find that concentrated emulsions and foams remain homogeneous within ±0.2%, even when large strains are applied; in the experiments shown in Fig. 3, strains are larger than 10000. The same result is found with all emulsions studied in [32]; no other foam is studied. This is in contrast to the behavior of dense suspensions of rigid particles. In particular, the strain to reach a stationary inhomogeneous state can be as low as 20 in the suspension of rigid particles studied in [27], of particle size similar to the droplet

and bubble size of an emulsion and the foam we have studied. The absence of observable migration in concentrated emulsions and foams may thus be due to the very different nature of interparticle interactions in these systems: indeed, Lhuillier [30] has recently pointed out the major role of direct rigid contact forces in migration. Other factors may play a role; in particular, we emphasize that the dispersed phase volume fraction of all the soft materials studied here is well above the jamming transition, which may hinder local increase of the particle volume fraction.

From the absence of observable migration for very large strains, we conclude that this process, if it exists, would be much too slow and inefficient to lead to observable shear banding in dense suspensions of soft particles at the usual experiment timescales.

Finally, for nonthixotropic dense suspensions of soft particles, thixotropy and migration should not be at the origin of shear banding. To explain the apparently contradictory observations of the literature, one may thus wonder whether a yet unknown mechanism is at play in some of these systems only, or if differences in the setup or procedure are involved. In the following, we present an experimental investigation of flow stability in various materials with a well-defined procedure designed to circumvent possible artefacts.

2.3. Investigation of flow stability in dense suspensions of soft particles

We propose to study the possible shear banding in nonthixotropic dense suspensions of soft particles as quantified by an intrinsic bulk property, the critical shear rate below which flow is unstable. In order to design appropriate setup and procedure, we first deal with possible artefacts that may lead to observe flow inhomogeneities that are not steady-state bulk shear bands.

2.3.1. Possible artefacts

Finite size effects. In systems of small gap to dispersed elements size ratio, structural inhomogeneities are sometimes observed. Indeed, the local shear stress / shear rate relationship $\tau(\dot{\gamma})$ in concentrated emulsions [9, 45] and 2D foams [10] in confined geometries is found to depend on the system size and on the boundary conditions, suggesting that nonlocal laws are needed to describe their behavior [9, 11] (this is discussed in Sec. 3). The material's apparent viscosity $\tau/\dot{\gamma}$ near the walls for a given applied stress can in particular be smaller than that of the bulk material [9, 45]: the material is locally 'fluidized' by a rough surface. This mechanism could *a priori* lead to observe coexisting liquid-like and solid-like bands (note however that no shear-rate discontinuity is reported in [9, 10, 45]); this would not reflect a bulk property.

Transient flow inhomogeneities. Divoux *et al.* [46] have recently observed long-lived transient flow inhomogeneities during the flows of Carbopol gels in a thin gap Couette cell. These inhomogeneities always disappear at steady state [46]: at any rate, steady-state flow is homogeneous. However, given the very long lifetime of these inhomogeneities at low shear rate (they may last as long as 1 day at a 0.1 s^{-1} rate), one may wonder if all bands reported in the literature are really steady-state bands. It is thus crucial to understand in which condition such transient inhomogeneities are generated; this may allow us to propose a procedure to circumvent this possible artefact.

It is worth noting that a slight structuration at rest is observed in the studied systems [46, 40]: their elastic modulus increases with the resting time, and their static and dynamic yield stress differ. Moreover, in each experiment, flow is imposed on the material initially at rest: the transient bands characterize flow start-up. A first basic explanation for the development of these bands can be given in the framework of a simple model, illustrated in Fig. 4a. Let us consider a material characterized by a simple yield stress fluid behavior $\tau = f(\dot{\gamma})$ in its liquid regime, with $f(\dot{\gamma}) \to \tau_{y_d}$ when $\dot{\gamma} \to 0$, which defines its dynamic yield stress τ_{y_d}. In addition, we suppose that flow can be induced on the material initially at rest only when $\tau \geq \tau_{y_s}$, with $\tau_{y_s} > \tau_{y_d}$, which defines its static yield stress τ_{y_s}; this model accounts for the possibility of a specific structuration at rest. If a macroscopic shear rate $\dot{\gamma}_{\text{macro}}$ is applied on the material initially at rest in a slightly inhomogeneous shear stress field (as in a thin gap Couette geometry), two cases are found, depending on the value of $\dot{\gamma}_{\text{macro}}$. If $f(\dot{\gamma}_{\text{macro}}) \geq \tau_{y_s}$, after the solid/liquid transition, the material simply flows homogeneously at the applied shear rate $\dot{\gamma}_{\text{macro}}$ and the resulting stress is $\tau = f(\dot{\gamma}_{\text{macro}})$. If $f(\dot{\gamma}_{\text{macro}}) < \tau_{y_s}$ (Fig. 4a), although the material should be able to flow at a stress lower than the static yield stress, the stress applied on the material in its solid regime has first to reach τ_{y_s}. Consequently, just after the solid/liquid transition, shear must localize in a band in the region of higher stress (near the inner cylinder in a Couette cell): the local shear rate in the band $\dot{\gamma}_{\text{band}}$ has to be higher than the macroscopic shear rate $\dot{\gamma}_{\text{macro}}$ to ensure that the resulting stress $\tau = f(\dot{\gamma}_{\text{band}})$ equals τ_{y_s}, i.e., $\dot{\gamma}_{\text{band}} = f^{-1}(\tau_{y_s})$. Note that the mechanism proposed by Moorcroft *et al.* [47] in the framework of the SGR model is basically based on the same idea.

Of course, it is only a rough explanation of the Divoux *et al.* [46] observations[3]. However, the above simplified model probably captures the basic reason for the existence of a banded flow when flow is initiated on a structured state: stresses at flow start-up have to be higher than in steady state. It also suggests that other loading procedures can be used to avoid generating transient bands; in the following, we propose such procedure (Fig. 4b).

2.3.2. Setup and procedure

We are here interested in steady-state shear bands. As defined in Sec. 2.1, these should result from flow instability at low applied rate. We stress that, to investigate their possible development, a homogeneous flow of the material in its liquid regime should first be imposed at high shear rate; next, the

[3] It does not explain what happens subsequently to the band, i.e., how the material at rest is progressively eroded by the flowing band. Moreover, the full story of flow initiation is far more complex, as shown by Divoux *et al.* [40]; in particular, flow is initiated by full slippage at the rotor of the Couette cell just after the solid/liquid transition.

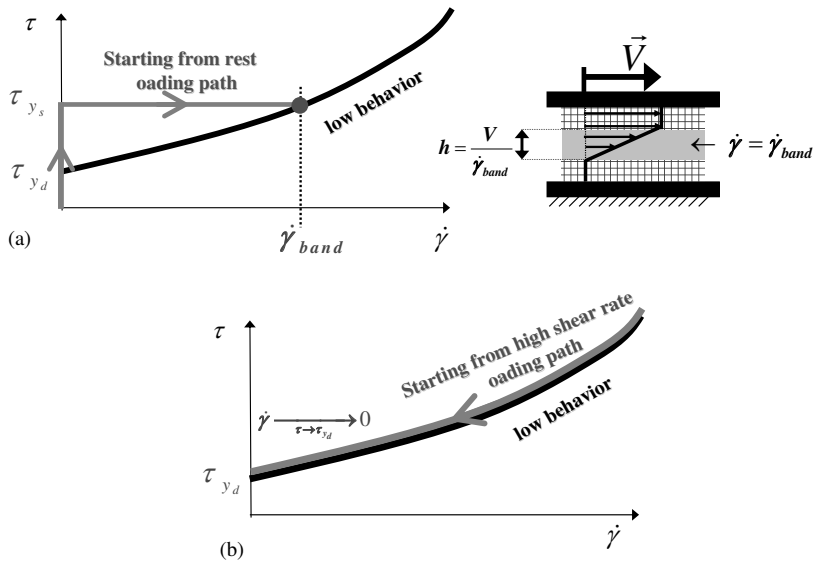

Figure 4: Illustration of the procedure-dependent behavior in a material whose static and dynamic yield stresses differ: (a) a band may form at flow start-up (Sec. 2.3.1); (b) an initially homogeneous flow may remain stable and homogeneous at any rate when the applied shear rate is decreased (Sec. 2.3.2).

shear rate should be decreased continuously (see Fig. 4b). In these conditions, transient flow inhomogeneities due to specific structuration at rest (Sec. 2.3.1) should not be observed: this procedure is thus preferable to applying a given shear rate on the material initially at rest, which may lead to wrongly report shear bands if experiments are not long enough (we recall that transient bands can last for 1 day). Then, if the material is not a shear-banding material, homogeneous flow should be observed at any low rate. In contrast, if the material is a shear-banding material, flow instability should develop at low shear rate. Note that this procedure also avoids observing the history-dependent flow inhomogeneities recently predicted by Cheddadi *et al.* [48] for materials exhibiting significant normal stress differences; this point is discussed in more detail below.

We focus on the behavior of dense suspensions of soft particles: concentrated emulsions, Carbopol gel, and foams, described in detail in [32, 49, 34]. To better show the difference between these materials and thixotropic systems, we also study the behavior of a colloidal gel (bentonite suspension), described in [7]. The materials are sheared in the same wide gap Couette geometry as in Sec. 2.2.2. This ensures that the bulk behavior, free from the finite size effects discussed in Sec. 2.3.1, is studied (this is shown in detail in Sec. 3). Consistently with the above procedure, we first apply a high rotational velocity Ω to the inner cylinder of the Couette cell, and we decrease progressively the value of Ω. Steady-state azimuthal velocity profiles $V(R)$ are then measured as a function of the radial position R in the gap using MRI techniques described in [50, 44], for various constant rotational velocities Ω.

2.3.3. *Experimental results and analysis*

All the studied materials exhibit similar behavior; an example is shown in Fig. 5. We observe that the whole sample is sheared at high rotational velocity Ω. When decreasing Ω, below a critical value that depends on the material and is of order 40 rpm in Fig. 5, the material is sheared only in a fraction of the gap: $V(R)$ vanishes (within the measurement uncertainty) at some radius $R_c(\Omega)$ inside the gap, which decreases when Ω decreases.

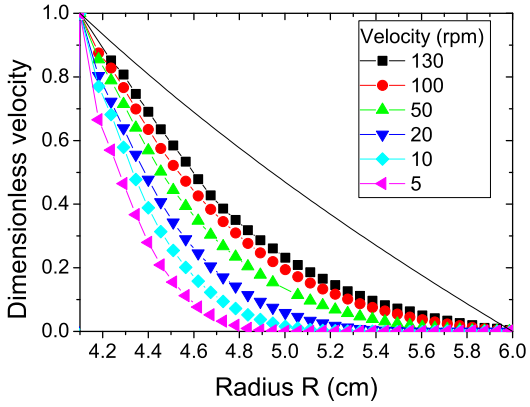

Figure 5: Dimensionless velocity profiles $V(R)/V(R_i)$, measured by MRI techniques, for the steady flows of a concentrated emulsion in a wide gap Couette geometry, at various rotational velocities ranging from 5 to 130 rpm (see legend); the solid line is the theoretical profile for a Newtonian fluid. The emulsion is composed of 1 μm diameter silicone oil droplets dispersed at a 75% volume fraction in water stabilized by Sodium Dodecyl Sulfate at a 8.5 wt% concentration [32]. Figure from Ovarlez *et al.* [32].

Flow inhomogeneity here originates from the shear stress field inhomogeneity (Eq. 1). Shear localization should then not be here confused with the shear banding observed in homogeneous stress fields: it is a feature of any yield stress fluid flow in a Couette geometry. At low Ω, flow stops at a radius R_c within the gap where the local shear stress $\tau(R)$ equals τ_y (i.e., $R_c = R_i\sqrt{\tau(R_i)/\tau_y}$). The decrease of $R_c(\Omega)$ as Ω is decreased

then arises from the rate dependence of the constitutive law at the approach of τ_y.

Although it induces shear localization for both shear-banding and non-shear-banding materials, the stress inhomogeneity can be used advantageously to analyze the possible shear-banding behavior from the shape of the localized velocity profiles. Indeed, from the constitutive behavior point of view, the question of shear banding can be posed as follow (Sec. 2.1): what is the value of the shear rate as τ tends towards the dynamic yield stress? In shear-banding materials, it should tend towards a nonzero value $\dot{\gamma}_c$; in simple yield stress fluids, it should tend towards zero. When flow is localized, τ approaches τ_y when R tends to R_c. As the local shear rate is

$$\dot{\gamma}(R) = V/R - \mathrm{d}V/\mathrm{d}R \tag{2}$$

the local velocity in a shear-banding material should thus tend to zero with a nonzero slope $|\mathrm{d}V/\mathrm{d}R| = \dot{\gamma}_c$ at the interface between the sheared and the unsheared regions, as in a homogeneous stress field [7]. In contrast, the slope of the velocity profile at the interface between the sheared and the unsheared regions should be equal to zero in a non-shear-banding material.

In the above analysis, we have assumed that the yield criterion only involves the shear stress τ, whose value thus determines the position of the interface between the sheared and the unsheared regions. If the material displays significant normal stress differences in its solid regime (as e.g. in dry foams [51]), those may enter in the yield criterion and things can be different. Whereas steady-state normal stresses in the flowing region depend only on the shear rate and are thus independent of the initial conditions, different nonzero normal stresses can be stored in the material at rest in the nonflowing region: their value depends on shear history, i.e. on preparation. Some sample histories may then lead to a discontinuity of the normal stress differences at the interface between the sheared and unsheared regions [48]. When taken into account in the yield criterion, this discontinuity implies that the shear stress at this interface is higher than in the case of continuous normal stress differences and that $\dot{\gamma}$ at this interface is nonzero [48]. This leads to a slope discontinuity in the velocity profile that appears as a critical shear rate which is not an intrinsic property of the material, but rather reflects the loading procedure. We point out that such discontinuities are not expected to take place with the loading procedure we use, as the solid-like zone is progressively formed by decreasing continuously the inner cylinder rotational velocity. We thus think that even with materials exhibiting significant normal stress differences, only intrinsic shear banding will be evidenced.

To analyze the possible shear banding, we now plot localized velocity profiles obtained for various materials, and characterized by almost the same position R_c (here $\simeq 5.4$ cm) of the interface between the sheared and the unsheared regions (Fig. 6a). We focus on the shape of the velocity profiles at this interface. From this plot, it clearly appears that (i) the thixotropic system (bentonite suspension) exhibits an abrupt transition from flow to rest, as expected from the shear-banding behavior of these materials observed in cone-and-plate geometry [12, 7], whereas (ii) concentrated emulsion, foam and Carbopol gel exhibit a smooth transition from flow to rest and thus seem to be not shear-banding materials.

To better illustrate these differences and their link with the constitutive behavior of the materials, we combine the local stress data $\tau(R)$ (Eq. 1) and the local shear rate data $\dot{\gamma}(R)$ (Eq. 2) extracted from the localized velocity profiles (Fig. 6a,b); this allows the reconstruction of the local constitutive law $\tau(\dot{\gamma})$ at the proximity of the yield stress τ_y. These laws are depicted in Fig. 6c. A clear difference between the thixotropic system and the nonthixotropic dense suspensions of soft particles is again observed: when $\tau \to \tau_y$, $\dot{\gamma}$ tends towards a finite value – the critical shear rate $\dot{\gamma}_c$ (here of order 6 s^{-1}) – in the bentonite suspension, and towards zero in dense suspensions of soft particles. We have observed this smooth transition from flow to rest, with the same procedure, in several other emulsions [32] and foams [34].

We conclude that none of the studied dense suspensions of soft particles is a shear-banding material. Of course, due to finite experimental resolution, we can only provide upper bounds for the value of the critical shear rate – if any. Note that the accuracy of the shear rate measurement depends on the spatial resolution and on the MRI signal to noise ratio, which depends itself on the material and on external factors. We finally obtain typical upper bound ranging between 0.01 s^{-1} and 0.3 s^{-1} [32, 49, 34].

2.4. Probable explanation of previous observations

From MRI observations on various systems, we claim that nonthixotropic dense suspensions of soft particles do not show any bulk steady-state shear banding. To strengthen this claim, we have to check that previous observations of shear rate discontinuities in similar systems in the literature can be explained. In the following, we review these observations and identify possible origins of the discrepancy between these results and ours.

Finite size effects. Denkov *et al.* [52] have reported shear banding in 3D foams, in a parallel plate geometry of gap to bubble size ratio of order 10. Gilbreth *et al.* [35] have reported shear banding in bubble rafts, in a geometry of gap to bubble size ratio of order 20. Such ratios are probably much too small to ensure that a bulk property is studied (see Sec. 3). It is worth noting that Katgert *et al.* [10] reported nonlocal effect in bubble rafts of similar gap to bubble size ratio as in [35]. It is thus possible that the [35, 52] shear rate discontinuities are due to finite size effects as explained in Sec. 2.3.1.

Transient effects and loading history. Transient flow inhomogeneities may last for hours at the vicinity of the jamming transition [46]: this casts doubt on band observations obtained by applying a given shear rate on the material initially at rest. Moreover, procedure-dependent steady flow inhomogeneities are expected at flow start-up due to normal stress differences trapped during the loading procedure in materials which exhibit significant normal stress differences [48]. Much care thus has to be taken when analyzing inhomogeneities observed at flow

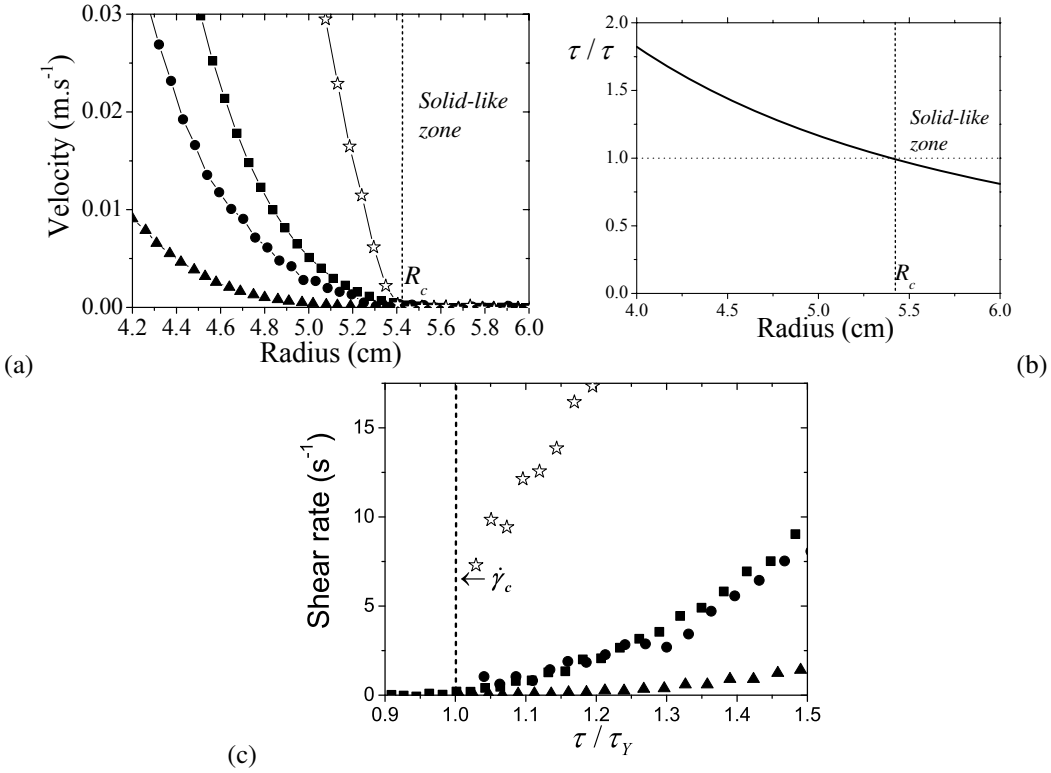

Figure 6: (a) Comparison between localized velocity profiles of a concentrated emulsion (squares), a foam (circles), a Carbopol gel (triangles), and a bentonite suspension (empty stars). (b) Dimensionless shear stress field $\tau(R)/\tau_y$ corresponding to the velocity profiles of Fig. 6a. (c) Shear rate $\dot{\gamma}$ vs. shear stress extracted from the velocity profiles of Fig. 6a. The emulsion is composed of 1 μm diameter silicone oil droplets dispersed at a 75% volume fraction in water stabilized by Sodium Dodecyl Sulfate at a 8.5 wt% concentration [32]. The foam is composed of 45 μm diameter bubbles at a 92% volume fraction in a SLES foaming solution described in [34]. The gel is a hair gel (Vivelle Dop, France), which is mainly made up of Carbopol in water [49]. The bentonite suspension is made up of (smectite) clay particles of length of order 1 μm and thickness 10 nm suspended at a 4% volume fraction in water [38].

start-up. In the Bécu *et al.* [31] experiments, shear was applied on a concentrated emulsion initially at rest; bands were observed while the shear stress was decreasing for 2 hours as in the Divoux *et al.* [46] experiments. We did not observe a shear-banding behavior on the same material, when applying the procedure of Sec. 2.3.2 (progressive decrease of the applied shear rate). The bands reported by Bécu *et al.* [31] were thus probably transient flow inhomogeneities. The observations of Rouyer *et al.* [41] on dry foams may also find the same explanation, as these were made when shear was applied on the material initially at rest, and as the experiment time was rather short (the applied strain was of order 1).

Uncontrolled sources of thixotropy. A simple yield stress fluid can be easily turned into a thixotropic material. E.g., Ragouilliaux *et al.* [15] have shown that tiny amounts of colloidal clay particles dispersed in the continuous phase of a simple emulsion can form bridges between droplets and lead to thixotropic effects and to shear banding as detailed in Sec. 2.1.1. The same might happen with a foam. As we did not observe any shear banding in the same foam as Rodts *et al.* [33], using the same procedure and setup, it is suggested that the occurrence of shear banding in their experiments might be due to uncontrolled traces of impurities in the system, e.g. clay particles, which were also studied with the same equipment.

Other artefacts. Let us finally note that many observations of flow inhomogeneities in 2D foams are now attributed to viscous damping at the glass boundary [53, 54, 55, 56, 10] when the bubbles are confined by one (or two) glass plate. Wall drag cannot explain observations in bubble rafts with free surface; in these last systems, as stated above, the main possible source of such inhomogeneities stands in nonlocal effects.

All results thus seem to be consistent with the absence of steady-state bulk shear banding in dense suspensions of soft particles. Of course, all of the above explanations still have to be checked in new experimental investigations.

3. Consistency between macroscopic and local behavior

In this section, we address the question of the existence of a single intrinsic local constitutive law to describe steady-state flows of yield stress fluids. If the material can be characterized by such law, it is expected to account for what happens in any flow geometry, independently of the boundary conditions. Once shear banding has been dealt with, the main source of rheometric artefact is removed. The macroscopically measured behavior in rheometric experiments should then reflect the local behavior; it should thus *a priori* be trusted and used to predict flows in other configurations.

In the following, we review data from the literature where local and macroscopic behavior do not seem to be consistent. We discuss the main possible origins of this inconsistency: thixotropy, volume fraction inhomogeneities, and nonlocal effects. We then present experimental results obtained in non-thixotropic dense suspensions of soft particles, and show that, under appropriate conditions, in a wide gap geometry, they are characterized by a single local law that matches the macroscopic one. We discuss the conditions needed to obtain such result.

3.1. Local observations in the literature

The ability of the macroscopically measured stress/strain rate relationship to predict the flow behavior can be accurately tested by performing local measurements of material velocity profiles $V(\vec{r})$ in geometries of controlled stress inhomogeneity $\tau(\vec{r})$ (Couette geometry, Poiseuille flow, inclined plane flow). In such case, a theoretical velocity profile $V_{th}(\vec{r})$ is easily derived from the macroscopic relationship $\tau_{\text{macro}} = f_{\text{macro}}(\dot{\gamma}_{\text{macro}})$ as the local shear rate should be $\dot{\gamma}_{th}(\vec{r}) = f^{-1}_{\text{macro}}(\tau(\vec{r}))$; theoretical and experimental profiles can then be compared. An alternative consists in deriving the local shear rate field $\dot{\gamma}(\vec{r})$ from the velocity profile; $(\tau(\vec{r}), \dot{\gamma}(\vec{r}))$ data obtained at various positions $\vec{r}$, under various boundary conditions, can then be combined to build the locally derived constitutive behavior, which can be compared to macroscopic observations.

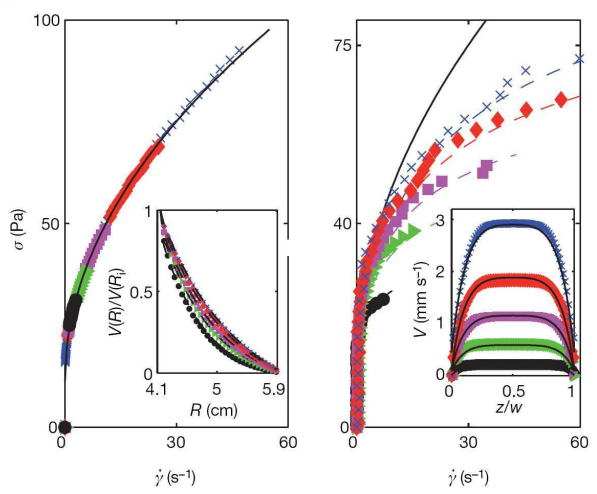

Figure 7: Laws $\tau(\dot{\gamma})$ measured locally in the gap of a 1.9 cm wide gap Couette geometry (a) and in a $w = 250$ μm wide microchannel (b), during the flow of a concentrated emulsion of 75% droplets of 6.5 μm diameter. Different symbol shapes and colors correspond to different imposed rotational velocities of the inner cylinder (a) or different imposed pressure drops (b). The full line is a Herschel-Bulkley equation $\tau = \tau_y + \eta_{HB}\dot{\gamma}^n$, with $n = 0.5$, $\tau_y = 11.6$Pa and $\eta_{HB} = 11.2$Pa.s^n. The insets show the velocity profiles from which these laws are extracted. The lines in Fig. 7b inset are fits of the velocity profiles to the nonlocal behavior Eq. 3. Figure from Goyon *et al.* [9].

Such local measurements in concentrated emulsions have yielded surprising results [57, 31, 9]: in some emulsions, the locally measured constitutive law does not match the macroscopically measured one. Moreover, it seems impossible to find a single local constitutive law $\tau(\dot{\gamma})$ compatible with all flows. The apparent $\tau(\dot{\gamma})$ law needed to describe the velocity profile for a given boundary condition does not match those needed for other boundary conditions: it depends on the velocity at the inner cylinder in a Couette cell [57, 31] or the pressure gradient in a Poiseuille cell [9] (see an example in Fig. 7b). The same features have been observed in dense noncolloidal suspensions [58] and 2D foams (bubble rafts) [10] sheared in Couette geometries.

Three main reasons can been mentioned to explain the apparent absence of a single local constitutive law: transient effects, shear-induced migration of the dispersed elements, and nonlocal effects.

3.1.1. Impact of thixotropy

In a thixotropic material, the shear stress τ is a function of the shear rate $\dot{\gamma}$ and of shear history. A single relationship between τ and $\dot{\gamma}$ can be defined only in steady state. During a transient, the shear rate can take many values for a given applied stress τ, depending on time and on shear history. When local measurements are not strictly performed in steady state, it may then leave the false idea that different laws are required to describe the flow of the sample. This is illustrated in Fig. 8 where local laws $\tau(\dot{\gamma}, t)$ measured in a thixotropic material at various times t during a transient are displayed. It is clearly observed that the local flow behavior depends on shear history, until a steady-state is reached.

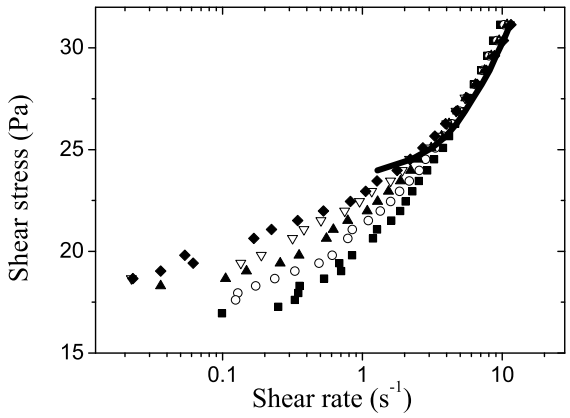

Figure 8: Laws $\tau(\dot{\gamma}, t)$ measured locally at various times t (squares: 30 s, empty circles: 90 s, up triangles: 150 s, empty down triangle: 210 s, diamonds: 270 s, full line: 1 h) during the flow of a thixotropic mud (see [15] for details on the material) in a wide gap Couette geometry. The material is first sheared at a rotational velocity $\Omega = 50$ rpm during 10 min; the velocity is then decreased abruptly at $\Omega = 15$ rpm and 2 velocity profiles are recorded per minute during 1 h, which allows reconstructing the time-dependent local law $\tau(\dot{\gamma}, t)$.

Although dense suspensions of soft particles are usually found to be nonthixotropic, it has been shown that long times can be needed to reach a steady state at flow start-up in some of these systems [46] (Sec. 2.3.1). This seems to be due to a specific structuration mechanism at rest; the structure formed at rest may then take time to be erased at low shear rate. Two situations should thus be distinguished: (i) when the material is

at rest, it gets slightly structured and its behavior at flow start-up may first depend on flow history during long time (as long as 24 h in the Divoux *et al.* [46] experiments); (ii) when the material is flowing at steady state, its subsequent flow behavior has no significant dependence on flow history as long as it is continuously strained in its liquid state. It contrasts with usual thixotropic systems such as colloidal gels, where competition between structuration and destructuration is observed in the liquid state.

These structuration effects might explain the observations of Bécu *et al.* [31], who cannot account for the flows of an adhesive emulsion with a single constitutive law. Their experiments are indeed performed at flow start-up. Their observation of flow inhomogeneity, whereas we do not observe shear banding – on the same material – when slowly decreasing the applied shear rate from high to low values (see previous section), then suggests that their local measurements are performed during a transient, consistently with the Divoux *et al.* [46] observations.

3.1.2. Impact of particle migration

Particle migration can also be a cause for the apparent lack of a local constitutive law [17]. Indeed, when various $(\tau(\vec{r}), \dot{\gamma}(\vec{r}))$ data are combined to build a constitutive law, a hidden hypothesis is that the material is homogeneous, i.e., that the same material is dealt with at any position $\vec{r}$ in the flow. If there is an inhomogeneous volume fraction field $\phi(\vec{r})$ in the material, one has to be careful when inferring a constitutive law from velocity profiles. To illustrate the problem, let us consider a volume fraction dependent constitutive behavior $\tau = g(\phi, \dot{\gamma})$. Under two different boundary conditions denoted b_1 and b_2, a same local value $\dot{\gamma}_0$ of the shear rate can be reached at two positions $\vec{r}_{b_1}$ and $\vec{r}_{b_2}$ where the local ϕ values differ. When reconstructing a constitutive law from all local data, this may lead to associating two different stress values $\tau_{b_1} = g(\phi(\vec{r}_{b_1}), \dot{\gamma}_0)$ and $\tau_{b_2} = g(\phi(\vec{r}_{b_2}), \dot{\gamma}_0)$ to the same shear rate $\dot{\gamma}_0$, thus leading to the – erroneous – conclusion that flow under different boundary conditions is not compatible with a single constitutive behavior.

This phenomenon explains the apparent lack of a single constitutive law to account for all flows in dense noncolloidal suspensions of rigid particles [58]. Further studies of the same system have indeed shown that it is inhomogeneous [17] due to shear-induced particle migration. A local constitutive behavior compatible with all flows can nevertheless be built [17, 27] by combining $(\tau(\phi(\vec{r}_0), \dot{\gamma}(\vec{r}_0)), \dot{\gamma}(\phi(\vec{r}_0), \dot{\gamma}(\vec{r}_0)))$ data measured at a fixed position $\vec{r}_0$ under various boundary conditions: this provides the constitutive behavior at a well defined volume fraction $\phi(\vec{r}_0)$ for various local shear rates $\dot{\gamma}(\vec{r}_0)$. By studying this local behavior at various positions $\vec{r}_0$, a ϕ-dependent local behavior is finally built. The observations of flow inhomogeneity, and thus of apparent viscosity inhomogeneity, in a colloidal glass sheared in a homogeneous stress field [25] is also accounted for by volume fraction inhomogeneities.

This phenomenon is not expected to occur in dense suspensions of soft particles: we have shown in Sec. 2.2.2 that these systems remain homogeneous under shear. Consequently, it probably cannot explain the Goyon *et al.* [9] and Katgert *et al.* [10] observations.

3.1.3. Nonlocal behavior

The apparent absence of a single local constitutive law can also be the signature of nonlocal phenomena. Indeed, the local $\tau(\dot{\gamma})$ relationship observed in dense emulsions in microchannels by Goyon *et al.* [9] was found to depend on the boundary conditions (pressure drop, surfaces roughness), which is the hallmark of nonlocal behavior (Fig. 7b).

A nonlocal modeling [9, 11] has been shown to successfully account for all flows in the Goyon *et al.* [9, 45] experiments; fair agreement has also been found with the Katgert *et al.* [10] experiments in bubble rafts. In jammed materials, flow basically occurs locally via a succession of elastic deformation and irreversible plastic events. The main idea of the nonlocal model [59] is that a localized plastic event induces relaxation of the stress in the whole system, and may thus significantly affect the behavior in its neighborhood – its zone of influence being quantified by a length ξ, called 'flow cooperativity length'. Consequently, the rate of plastic rearrangements in a given zone depends on the rate of rearrangements in its neighborhood. It will then be different in a strongly inhomogeneous stress field, i.e., if the stress varies significantly over ξ, than in a homogeneous stress field far from boundaries (in this last situation, the bulk behavior is obtained). Nonlocal effects are thus predicted to take place in the presence of high stress gradients. In its simplest form, the Bocquet and Colin [11] model is

$$[\dot{\gamma}/\tau](z) = [\dot{\gamma}/\tau]_{bulk}(z) + \xi^2 \partial_z^2 [\dot{\gamma}/\tau](z) \quad (3)$$

where $[\dot{\gamma}/\tau]_{bulk}$ is the bulk apparent inverse viscosity of the material (also called 'fluidity') and z is the direction of the stress gradient [9]. This nonlocal model also accounts for the impact of the wall roughness, when the material displays a different behavior near the boundary than in the bulk [9, 45]. In a homogeneous stress field, the length over which the behavior changes from surface to bulk behavior is then predicted to be of the order of a few ξ.

These two features explain the Goyon *et al.* [9, 45] observations in microchannels of small size. From a fit of experimental data to the nonlocal model (Fig. 7b inset), they have shown that ξ is zero below the jamming volume fraction ϕ_m, and increases continuously with ϕ above ϕ_m. E.g., ξ is found to be 5 droplet diameters in a monodisperse emulsion of 85% volume fraction. Consistently, discrepancy between the local and macroscopic behavior is found in 10 to 40 droplets wide geometries but not in a 3000 droplets wide geometry [9] (see Fig. 7). ξ is found to be of the order of 3 bubble diameters in the bidisperse bubble raft studied by Katgert *et al.* [10], which is 20 bubbles wide; no large scale study is reported for rafts.

In nonthixotropic dense suspensions of soft particles, the effects discussed above can *a priori* be dealt with. Indeed, migration is not an issue (Sec. 2.2.2) and significant time-dependent behavior may be found only in specific conditions (Sec. 2.3.1); moreover, the impact of nonlocal effects should be minimal in wide gap geometries. In the following, we propose to study the local behavior of such materials in well-defined conditions where the above mechanisms should not be at play.

3.2. Nonthixotropic dense suspensions of soft particles in a wide gap geometry

Our goal is to see if, under some conditions, a simple local law accounts for yield stress fluid flows and matches their macroscopic response. Given the effects discussed above, appropriate setup and procedures must be used.

3.2.1. Setup and procedure

In order to ensure that the bulk behavior, free from the finite size (nonlocal) effects discussed in Sec. 3.1.3, is studied, the flow geometry that is used should have specific characteristics. The nonlocal model (Eq. 3) and the observations of Goyon *et al.* [9] first imply that the gap has to be at least tens of cooperativity length ξ wide, i.e., more than one hundred dispersed elements diameters wide to minimize the impact of the boundaries. Moreover, from Eq. 3, smooth stress spatial variations (of a maximum order of a few % over ξ) are required.

Here, we use the same wide gap Couette geometry as in Secs. 2.2.2 & 2.3. In a Couette geometry, the stress is inhomogeneous (Eq. 1) and the stress relative variation over a length $\xi << R_i$, where R_i is the inner cylinder radius, is $|\tau(R_i + \xi)/\tau(R_i) - 1| \simeq 2\xi/R_i$. Assuming a value of ξ of the order of 5 dispersed elements diameter d, as found by Goyon *et al.* [9] in some systems, smooth stress spatial variations impose R_i to be at least equal to several hundreds of d. Note that this is only a rough estimate as these are the variations of the 'fluidity' $\dot{\gamma}/\tau$ that have to be smooth. We thus have limited our studies to materials whose dispersed elements diameter is less than 100 μm to meet the above requirements (with R_i = 4.1 cm and gap= 1.9 cm).

To ensure that steady-state properties are studied, the long-lived transient effects discussed in Sec. 2.3.1 and their possible consequence on the local behavior (Sec. 3.1.1) should be avoided. That is why we use the same procedure as in Sec. 2.3.2, i.e., we first impose a homogeneous flow of the material at high shear rate, before decreasing progressively the applied shear rate on the flowing – liquid-like – material (see Fig. 4b).

The same materials as in Sec. 2.3 are studied: concentrated emulsions, Carbopol gel, and foams, described in detail in [32, 49, 34]. After a 5 min preshear at 50 to 100 rpm (depending on the material), we measure both the steady-state azimuthal velocity profiles $V(\Omega, R)$ with MRI techniques (see Sec. 3.2.1) and the torque $T(\Omega)$ with a Bohlin C-VOR 200 rheometer, for various constant rotational velocities Ω of the Couette cell inner cylinder. In all systems, the stationary velocity profiles are found to develop within a few seconds and to remain stable for long durations (as long as hours in the emulsions), which means that these systems are not thixotropic [2]. For the Bécu *et al.* [31] emulsion, this contrasts with the long time evolution of the velocity profiles they observe. We argue that this comes from the differences in the procedures used, as discussed in Sec. 2.3.2; this validates the procedure we use to study a behavior free from transient effects.

3.2.2. Experimental results and analysis

Typical steady-state velocity profiles are shown in Fig. 5. The constitutive laws of the materials accounting for their velocity profiles can be built by using both the velocity profiles and the torque measurements. The stress distribution $\tau(R)$ at a radial position R within the gap is obtained from Eq. 1; we recall that $\tau(R_i) = T/(2\pi R_i^2 H)$ where H is the height of the inner cylinder. The local shear rate $\dot{\gamma}(R)$ in the gap is inferred from the velocity profiles $V(R)$ thanks to Eq. 2. Both measurements performed at a given radius R for a given rotational velocity Ω provide a local data point of the constitutive law $\tau = f(\dot{\gamma})$. Note that, as Eqs. 1 & 2 are valid whatever the boundary conditions are, this analysis is not affected by a possible wall slip. As the materials are homogeneous, we are allowed to combine the $(\tau(R), \dot{\gamma}(R))$ data measured at various positions R (see Sec. 3.1.2): they are *a priori* representative of the same material. The local constitutive laws extracted from the experiments performed at various rotational velocities Ω on an emulsion, a foam, and a Carbopol gel, are plotted in Fig. 9.

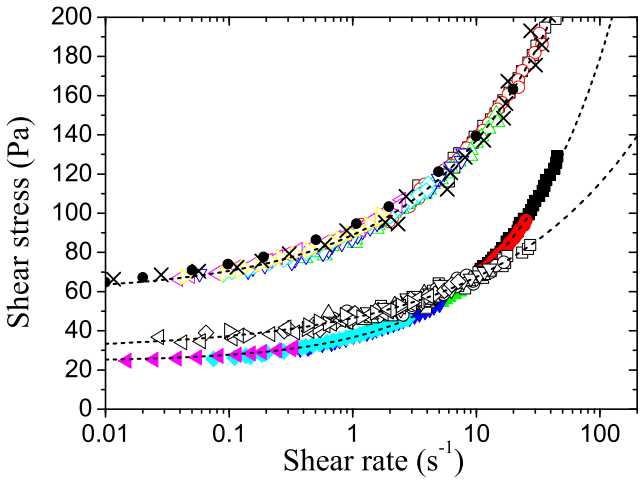

Figure 9: Constitutive laws of (from bottom to top) a concentrated emulsion, a foam, and a Carbopol gel, measured locally in a wide gap Couette cell using MRI techniques. Macroscopic data measured in the Carbopol gel in the Couette geometry (crosses) and in a cone and plate geometry (filled circles) are also shown. The dotted lines are Herschel-Bulkley fits to the data $\tau = \tau_y + \eta_{HB}\dot{\gamma}^n$ with: τ_y = 24.3 Pa, η_{HB}=12.3 Pa s$^{0.55}$, and n = 0.55 (emulsion); τ_y = 30.3 Pa, η_{HB}=16.2 Pa s$^{0.36}$, and n = 0.36 (foam); τ_y = 59.3 Pa, η_{HB}=29.6 Pa s$^{0.42}$, and n = 0.42 (Carbopol gel). The emulsion is composed of 6.5 μm diameter silicone oil droplets dispersed at a 75% volume fraction in a mixture of 50 wt% glycerine and 50 wt% water stabilized by Brij and trimethyl tetradecyl ammonium bromide at a 1 wt% concentration [32]. The foam is composed of 45 μm diameter bubbles at a 92% volume fraction in a SLES foaming solution described in [34]. The gel is a hair gel (Vivelle Dop, France), which is mainly made up of Carbopol in water [49].

A first important result is that, for each material, all the shear stress vs. shear rate data fall along a single curve. This means that, for a given material, data obtained under different boundary conditions (different Ω) effectively reflect the behavior of a single material with a given local intrinsic constitutive law in simple shear. This observation contrasts with the Goyon *et al.* [9] observations, on the same emulsion, consistently with the predictions of the nonlocal model discussed in Sec. 3.1.2:

nonlocal effects are not expected in the geometry we use as the gap is 3000 droplets wide for this material (thus ensuring that both surface effects and stress inhomogeneities have no observable impact on the behavior). We also contrast our observations on 3D foams with those on rafts by Katgert *et al.* [10]; again, this is consistent with what can be expected from the nonlocal model as the gap size is 400 bubbles wide in our experiments, and 20 bubbles wide in the Katgert *et al.* [10] experiments. Our measurements performed on the Bécu *et al.* [31] emulsion contrast with their observations too. In the Bécu *et al.* [31] study, however, the gap being 1mm for 0.3μm droplets, the nonlocal effects discussed in Sec. 3.1.3 cannot be the reason for the apparent absence of a single constitutive law accounting for all flows. As already pointed out above, we think that the main difference between the two experiments stands in the procedure as the Bécu *et al.* [31] experiments were performed at shear start-up on a structured material. This tends to show that, provided an appropriate procedure is used, a single local behavior is recovered.

It is worth noting that data are obtained at local shear rates as low as 10^{-2} s^{-1}, the lower measurable value depending on the resolution on the velocity measurement (thus on its derivative), and also on the coarsening rate for foams [3]. This observation is consistent with the absence of observable shear banding (Sec. 2.3). All observed behaviors are finally consistent with a continuous local law $\tau(\dot{\gamma})$ with $\dot{\gamma} \to 0$ when $\tau \to \tau_y$. Good agreement is found with a Herschel-Bulkley equation $\tau(\dot{\gamma}) = \tau_y + \eta_{HB}\dot{\gamma}^n$ for all materials in a [10^{-2} s^{-1}, 40 s^{-1}] range of shear rates; the fitting parameters (in particular the index n) depend on the material, and are given in the figure caption.

We now get back to the macroscopic behavior. The torque data $T(\Omega)$ obtained in the Couette geometry can be analyzed using standard rheometric analysis (without using the velocity profiles) to yield the macroscopic constitutive law $\tau = f_{\text{macro}}(\dot{\gamma})$ of the material; this analysis has to be performed carefully to account for shear localization [2, 32, 49] when computing the macroscopic shear rate from the value of the inner cylinder rotational velocity. These macroscopic data, displayed in Fig. 9 for the Carbopol gel, are in good agreement with the local data. Good agreement has also been found in the case of an emulsion [32]. To push the analysis further, we have also characterized the Carbopol gel with a 40 mm diameter 4° angle sandblasted cone-and-plate geometry. In such geometry, the stress field is homogeneous and cannot be a source of nonlocal effects; however, the gap still has to be wide enough to measure bulk properties. In the used geometry, the gap varies between 1.4 mm at the edges and 150 μm (truncature height); the size of the dispersed elements of the studied material should thus be smaller than $\approx 20\mu$m. The macroscopic data obtained in the Carbopol gel with this geometry are shown in Fig. 9. Good agreement is obtained with the macroscopic and local data measured in the Couette geometry. This shows that a single intrinsic local behavior indeed characterizes the flows of this material. This validates the use of macroscopic tools to study the behavior of these systems, provided the gap is wide enough and a procedure similar to the one we propose (shearing continuously from high to low shear rate) is used.

Altogether, these results show that, in the strict conditions discussed in Sec. 3.2.1, a simple yield stress fluid behavior exists: at steady-state, all the studied nonthixotropic dense suspensions of soft particles are indeed characterized by a single, intrinsic, local, continuous and monotonic constitutive law in simple shear. For all materials, it is moreover consistent with a Herschel-Bulkley equation, as usually found in standard rheological experiments.

4. Conclusion

Although many complex behaviors have been observed in yield stress fluid flows, we have shown that, under well-defined conditions, a class of materials, namely nonthixotropic dense suspensions of soft particles, display a simple yield stress fluid behavior: at steady-state, they are characterized by a single, intrinsic, local, continuous monotonic constitutive equation $\tau(\dot{\gamma})$ in simple shear with $\dot{\gamma} \to 0$ when $\tau \to \tau_y$ (i.e., they are not shear-banding materials).

To obtain these results, the main material characteristics are that:

- they have to be nonthixotropic. More precisely, they should not display time-dependent behavior in their liquid state, otherwise they will show shear banding. Possible structuration at rest is not an issue if an appropriate characterization procedure is used (see below).
- they should be made up of soft particles. Dense suspensions of soft particles have been shown to remain homogeneous when sheared, in contrast to suspensions of rigid particles.

Materials complying with these requirements are foams, concentrated emulsions, and Carbopol gels.

As regards the setup and procedure, two points have to be respected:

- the geometry where flow is studied should have a wide gap, and should be characterized by smooth stress inhomogeneities to ensure that nonlocal effects are not present. We recall that the relevant lengthscale to consider in this problem may be a 'cooperativity length' rather than the dispersed elements size, as found in concentrated emulsions.
- the following procedure should be applied: flow has first to be imposed at high shear rate to erase the memory of possible structuration at rest; afterwards, the material flow should be studied by progressively decreasing the applied shear rate, without passing by a resting period. This ensures that possible transient shear banding is avoided and that the local flow behavior is consistent with the macroscopic one.

In these conditions, a simple yield stress fluid behavior is finally shown to exist. For the studied materials, the steady

flow behavior in simple shear can be modeled by a Herschel-Bulkley equation, as found in standard rheological experiments. A macroscopic characterization can then be trusted as it reflects the local intrinsic behavior of the material. It can *a priori* be used to predict the flow behavior in other configurations, provided the studied flow also complies with the above requirements (large flow scale, problem different from flow start-up at low shear rate).

Of course, when confined flows or slow start-up flows are studied, all the complex effects discussed throughout the text will possibly occur; they thus still have to be studied in depth in fundamental studies to help predicting what happens in such situations. In some materials, other effects such as elastic effects should also be incorporated in the modeling as they may be of crucial importance in 3D flows.

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