Rheophysical classification of concentrated suspensions and granular pastes

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(Received 17 September 1998; revised manuscript received 19 November 1998)

On the basis of existing data along with rational arguments we propose an overview, in a conceptual diagram (solid fraction vs shear rate), of the dominant interaction type (Brownian, colloidal, and hydrodynamic) at a mesoscopic scale within flows of concentrated suspensions or granular pastes. Different flow regimes identified by the values of dimensionless numbers are thus distinguished. The main characteristics of the rheological behavior of suspensions within each regime are inferred. A specific domain corresponds to regimes for which dilatancy effects occur which may lead to frictional or collisional processes if an additional (normal) force is applied over the particles. [S1063-651X(99)09104-7]

PACS number(s): 83.70.Hq, 83.10.−y, 83.20.−d, 82.70.−y

I. INTRODUCTION

The main trends of the rheological behavior of simple dilute suspensions and colloidal dispersions, and some model concentrated suspensions, are relatively well understood from a physical point of view [1]. Concentrated suspensions or granular pastes [a clearer definition of these systems will result of this work (see Sec. VIII)], within which complex particles interact strongly, giving rise to a viscosity much higher than the viscosity of the (interstitial) suspending medium, have received less attention from physicists, probably because of the apparent higher complexity of the field. Nevertheless the flows of such systems are important in industry at different stages of design, preparation, production or use of various materials such as fresh concrete, cement or mortar, herbicides, drilling muds, foodstuffs, pharmaceutical and cosmetic pastes, inks, paints, etc. Flows of natural materials such as lavas, debris flows, landslides, avalanches, etc. are also concerned. Hinch [2] proposed an interesting overview of practical processes in which particulate systems are involved along with an attempt of unification of the corresponding physical phenomena.

The rheological behavior of concentrated suspensions and granular pastes has been dealt with mainly in three, apparently distinct, fields which consider different types of materials under various conditions: (a) rheology of (fluid) suspensions, (b) physics of granular matter, and (c) soil mechanics. These fields often ignored one another, but it is one of the objectives of our work to show that they might reconcile or overlap in some cases. Here we are mainly concerned with one-phase flows for which, within a representative volume, the average particle velocity is equal to the velocity of the surrounding fluid, but possible two-phase effects might occur due to short, local, particle migration, dilatancy, and so on.

The first field (a) was developed by physicists somehow extrapolating the approach of Einstein [3] concerning dilute suspensions of hard spheres to concentrated systems. Thus the viscosity of such systems is related to the viscosity of the

interstitial fluid (μ_0), the solid volume fraction (ϕ), and the maximum packing fraction (ϕ_m) through various formulas, going from models assuming only hydrodynamic effects [4] to models attempting to include colloidal interactions and Brownian motions at different levels by using a varying maximum packing fraction [5] or an effective volume fraction [6]. More recently some authors also attempted to predict the shear-thinning trend, i.e., apparent viscosity decrease with shear intensity, or the yield stress of concentrated suspensions through mesostructural approaches using fractal concepts [7] and transient network modeling originally developed for polymer dynamics [8]. In practice the Bingham model, for which the stress is the sum of a yielding term (τ_c), and a Newtonian-like term have most often been used in literature in the aim of flow modeling or as the type of behavior that some theoretical approaches should predict. However, it is now admitted that this model lacks physical sense and, contrary to a Herschel-Bulkley model ($\tau = \tau_c + q\dot{\gamma}^p$, where $\dot{\gamma}$ is the shear rate, and q and p are fluid parameters), it is incapable of representing data within a shear rate range of several decades [9]. In addition, these materials are often viscoelastic and thixotropic (the viscosity varies with flow time) [10,11]. Finally, the existing theories remain far from able to predict all the various and complex aspects of rheological properties of concentrated suspensions. In addition, it is worth noting that concentrated suspensions and granular pastes are precisely the most difficult systems to study with rheometers. Indeed various disturbing effects may develop during tests, such as wall slip (the most important effect), fracture, sedimentation, migration, evaporation, edge effects, etc. Some of these perturbating effects originate in the specific internal structure of the suspensions. This may lead to serious misinterpretation of rheometrical data [12], since in that case this is not the theoretical volume of homogeneous constant material which is sheared.

Concentrated systems have also been tackled by physicists within the framework of the study of granular systems (b), which have received widespread attention in recent years [13]. For predictions of the mechanical properties of flowing granular assemblies, mainly the kinetic theory and numerical models have been used [14] with only partial success. In particular these models remain unable to predict the various

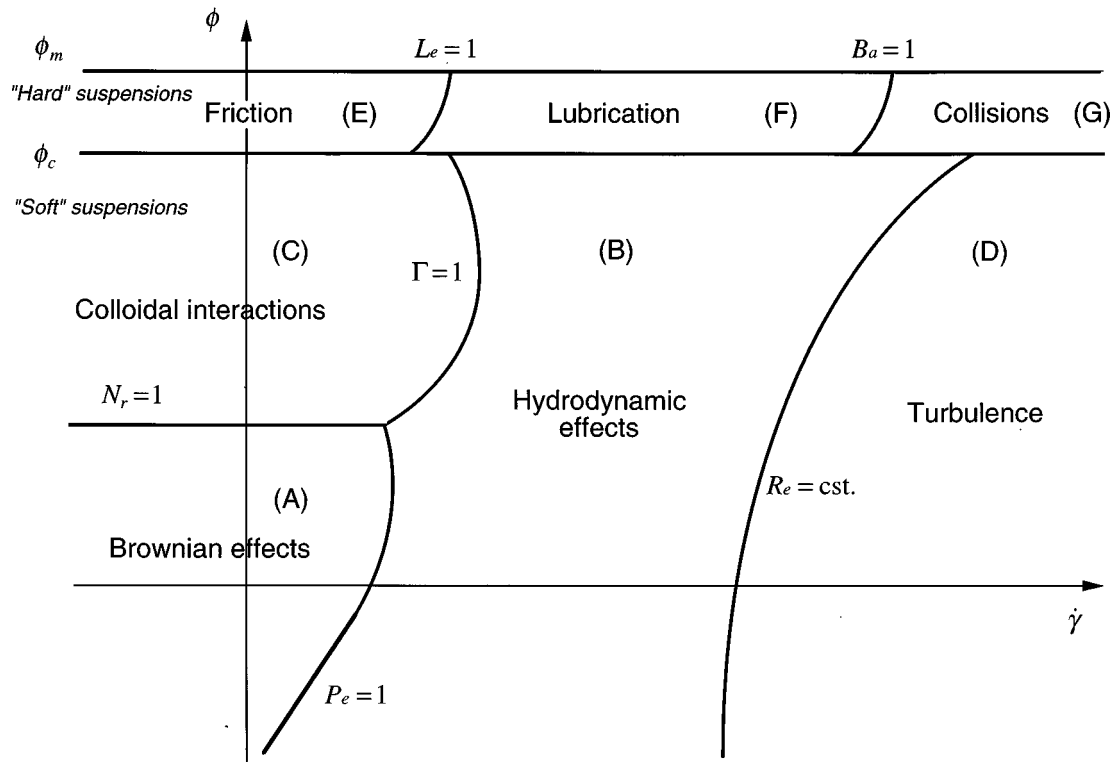*FAX: 01 40 43 54 85. Electronic address: philippe.coussot @lcpc.fr

FIG. 1. Conceptual classification of the rheophysical regimes of a suspension as a function of shear rate and solid fraction on a logarithmic scale. The other characteristics of the suspension are fixed, and the limiting curves do not correspond to strict transitions (see the text).

properties of dry granular systems in a wide range of conditions [15]. Moreover, they cannot be used for modeling slow or moderate flows of granular pastes yet.

The last field (c) in which such systems have been studied is soil mechanics. Here the constitutive equation is generally inferred using phenomenological relations concerning plastic dissipation and the so-called flow rule, which enables one to relate a strain increment to a stress increment. The most common theory is the so-called Cam-Clay model [16] which is used to describe the elastoplastic hardening behavior up to the failure. Alternative models, always based on plasticity potential and incremental formulation, have been developed, sometimes using a more refined framework (such as Cosserat media) [17]. In contrast with fields (a) or (b) and contrary to metal plasticity, domain (c) suffers from the absence of a physical interpretation (at the particle scale) of the plasticity mechanisms [18]. We can, however, quote some efforts which have recently expended to try to explain the plastic behavior of granular assemblies using homogenization techniques [19]. The lack of physical sense is obvious in many parameters used for identifying soil features. For instance, the so-called Atterberg limits (giving the water content of clay materials, respectively, for the plastic failure and flow beginning) are arbitrarily defined by an experimental protocol.

Here we essentially intend to provide, from rational arguments, a simplified conceptual diagram of predominant interactions within flowing concentrated suspensions (mainly under simple shear) as a function of shear rate and solid fraction (Fig. 1). As soon as one knows the appropriate dimensionless numbers governing the transition between the different regimes, it is easy to draw similar diagrams using other variables such as particle radius or aspect ratio, ionic strength, external (normal) force, etc. The present scale $(\phi, \dot{\gamma})$ is used for the presentation of results because it seems the most useful in practice: it contains two parameters providing rapid basic information concerning the density of the suspension and the flow intensity. In this diagram we assume that all other suspension parameters, such as ionic strength, particle diameter and shape, flow geometry, temperature, friction coefficient, viscosity of the interstitial fluid, fluid and particle density, external force, and boundary conditions, are constant. For example, for large grains, the curves corresponding to the transition from regime (A) or (C) toward regime (B) would be displaced toward infinitely small shear rates, which means that under most conditions hydrodynamic effects prevail. We also remark that the transition curves which have been drawn do not correspond to an exact limit between two regimes, but simply point out a region around which it can be expected that the type of predominant interactions progressively changes. Since this is a conceptual diagram we did not try to draw precisely the curves for a given set of parameters, but we represented all the main trends of these curves (in terms of slope) as appears from the viscosity and yield stress functions that we used.

Our intention is not to provide a complete framework for modeling the rheological behavior of the various systems under different conditions. This indeed requires taking into account the specific form of interactions at a mesoscopic scale, and may give rise to particular macroscopic properties of the suspension [8,11,20]. It is expected that a clearer distinction of significant interactions in different regimes constitutes a necessary first stage in order to understand these

systems better. Since it would take a great deal of space to describe all existing works and synthesize their results without losing ourselves in detail, we shall often adopt a global position. Nevertheless we think that such an analysis could provide a useful overview and might foster an understanding, in physical terms, of the relative situation of the different fields above cited with regards to concentrated suspensions.

In order to establish the diagram of predominance, we successively review the main types of interactions within a suspension of approximately monodisperse particles: Brownian effects, colloidal forces, viscous forces, and contacts (lubrication, friction, collision). We also provide some elements concerning the typical rheological behavior of suspensions in each regime. Since we aim at obtaining a rough overview of possible behavior and interaction types, we shall not detail the possible, more or less understood, specific behavior of some model systems (in particular uniform glass bead suspensions) [21].

II. GENERALITIES

We first consider mixtures of a Newtonian fluid with neutrally buoyant, solid, particles whose size is larger than 1 nm. We assume that from a colloidal point of view the suspension is stable. From a practical point of view, this mainly means that repulsive forces prevail if particles are colloidal, and that attractive forces remain negligible if particles are noncolloidal. We remark that in the following we shall often use, maybe improperly, the word ''colloidal'' to refer to any situation in which intermolecular or surface forces are important. Here we are concerned with various systems such as model systems (e.g., polystyrene lattices) as well as rather complex systems (e.g., clay suspensions, fresh concrete, etc.). As a consequence we shall not define repulsive and attractive forces in more detail. In addition we shall neither consider a specific particle shape nor assume a specific arrangement of the particles. We shall simply use two parameters to describe the geometrical structure: r, a characteristic length of the solid particles defined as the cubic root of the mean particle volume; and ϕ, the local, solid, volume fraction in a representative volume. From these parameters, it is possible to define b, a characteristic mean distance between the two centers of neighboring particles, as $r(\phi/\phi_m)^{-1/3}$. Clearly this simplified description only intends to present a unified view, but other types of variations of b may be found depending on the specific structure of the suspension under study [22].

III. EQUILIBRIUM STRUCTURE AND YIELD STRESS IN COLLOIDAL SYSTEMS

We shall start by analyzing colloidal systems, simply considering suspensions of large particles as a limiting case, i.e., with negligible colloidal forces but with possible, significant, direct contacts. Each particle is submitted to various forces, the largest of them under usual conditions resulting from van der Waals attraction, electrostatic forces, forces due to soluble polymers, Brownian effects, viscous forces, and inertia effects [1]. These two last effects are negligible for sufficiently slow flows. We first consider interactions between particles under such conditions, and we suggest the

following simplified, general, scheme for the particle interactions.

At a given time a particle is surrounded by other particles, each of them interacting with it to a various extent via colloidal interactions. Thus we can consider the potential energy (Φ) of the particle as a function of the position of the surrounding particles. For a particle, there exists a (instantaneous) position related to a minimum in potential energy [23], which should be situated more or less at an equal distance from the centers of neighboring particles. However, the particle needs a certain time to reach this position when it has been initially displaced. Thus the position of the particle relative to its instantaneous minimum of energy depends on the history of its motion, and thus on flow history. Moreover, Brownian motions of all particles constantly induce fluctuations of this equilibrium position in time. The particle can reach an equilibrium position only when the suspension has been left at rest during an infinite time. As a consequence, it is useful to refer to a mean potential minimum (in time and space), to which corresponds an approximate, average energy barrier (Φ_0) to overcome when extracting (extremely slowly) a particle from its environment (but keeping it in the fluid), after an infinite time of rest of the suspension. Taking the above remarks into account, it must be kept in mind that the potential energy of each particle, resulting from its interactions with other particles, can seriously deviate from this equilibrium value, and in particular can be much larger during a rapid flow, since particles do not have time to go back to an instantaneous, local position of minimum energy. (We remark that in this case the potential barrier to extracting the particle is smaller.) This potential energy obviously depends on the characteristics of the interactions and interparticle arrangement, and it might be expressed in terms of ionic strength, Debye length, solid fraction, etc., as for simple systems [1]. This scheme has some similarities with the initial scheme of Ogawa *et al.* [24], who suggested treating suspensions of charged, stabilized, colloidal particles by applying Eyring's transition state theory.

If the interaction energy between particles at a distance of the order of the mean separating distance between their surfaces ($h = b - r$) is negligible, the particles are free to move within the suspending medium as long as they do not approach each other too closely; the suspension is thus apparently Newtonian. Conversely, if this energy is significant, particles cannot easily move away from each other. In that case the suspension should clearly exhibit a yield stress, since a flow can be obtained only if a finite energy is provided to the system, which should make it possible to extract each particle from its instantaneous, local, potential well reached after an infinite time of rest, even extremely slowly. Typically such a phenomenon has been observed with suspensions of charged spheres when the Debye length becomes of the order of the separating distance between particles [1]. In the following we provide some elements in order to quantify these phenomena within the framework of our analysis of complex systems.

Roughly speaking, the colloidal interactions as described above are negligible compared to Brownian motion when N_r is much smaller than 1, with

$$N_r = \frac{\Phi_0}{kT}, \tag{1}$$

where k is the Boltzmann constant and T the temperature. (In this paper we shall drop the $\frac{1}{2}$ factor in the average, thermal, kinetic energy.) In that case, the system never reaches a specific equilibrium structure since the particles, even situated close to a local, instantaneous, equilibrium position, can be rapidly removed far away because of thermal agitation.

When N_r is much larger than 1, it will be more difficult for a given particle to extract itself from its local position between other particles. We remark that this is less difficult if the particle is far from its equilibrium position, in particular just after an intense flow. Conversely, at rest, one may expect that some forces will tend to draw the particles toward an equilibrium position. Thus, as time goes on, the probability for a particle to escape from its potential well by self-diffusion decreases toward zero. As a consequence it may be expected that there exists a well-defined equilibrium structure after an infinite time of rest for which each particle is embedded within a particle network. This network must be broken in order to impose a homogeneous flow on the suspension, each particle being extracted from its equilibrium position. Macroscopically this corresponds to an apparent yield stress.

It is worth noting that this scheme is valid only if the positions of equilibrium keep a local character even after a long time of rest. For suspensions with a marked thixotropy it is likely that slow collective rearrangements of the particles (due to Brownian and colloidal effects) occur at rest, giving rise to a particular configuration of particles throughout the suspension. Within this arrangement particles are in positions of an energy minimum which result from a collective arrangement. In that case the energy barrier can be significantly larger than Φ_0. This effect is probably at the origin of the marked thixotropy of some of these systems (in particular the strength of the particle network increases with time of rest), and we assume that it can be associated with a characteristic time (T_c^*). Within this framework the yield stress, as a unique suspension parameter, has a physical sense only if it is associated with the equilibrium structure reached after an infinite time of rest, which is in agreement with relevant experimental procedure recently developed for determining it [25]. Parallel to this, the time (T_c) is associated with the return of a particle toward its instantaneous, local, equilibrium position, under the action of colloidal forces and despite the viscous drag (see the quantitative definition below). In the following, for the sake of simplicity, we assume that T_c^* is much larger than T_c and T_h, the characteristic time of flow, or that collective rearrangements have a negligible effect on yield stress level. Under these conditions thixotropic effects will always be neglected, and the infinite time of rest will be a reference time equal to a few times T_c.

To sum up, for sufficiently slow flows, the curve $N_r = 1$ approximately corresponds to the transition between a regime (A) for which Brownian motions dominate, toward a regime (C) for which colloidal interactions dominate, and in that case the suspension clearly exhibits a yield stress associated with an equilibrium structure after a time of rest of reference. The transition curve is a horizontal line in our diagram since Φ_0 does not vary with the shear rate (Fig. 1).

For flocculated suspensions it has often been assumed that, during shear, the network breaks in flocs, the size of which decreases while the supplied energy increases due to increasing viscous drag [26]. This scheme does not seem appropriate here since the energy is mainly transmitted via the interactions between particles. Conversely, we assume that the energy required to deform the suspension of a critical value (γ_c) at an infinitely small velocity is used to move a certain number (which can be small) of particles out separately from their potential well. Clearly this is an approximation, since in fact the particles are not really moved far away from each other, and the potential well results from mutual interactions. This scheme seems nevertheless in global agreement with recent experimental observations with foams and emulsions [27,28]. γ_c depends on the network structure, but since it results from the rearrangement of some particles we can also assume that the induced deformation is directly linked to the number (n) by the unit volume of irreversibly rearranged particles, so that $\gamma_c = nb^3$. Under these conditions the energy dissipated by the unit volume deduced from mean mesoscopic considerations can be equated to that deduced from macroscopic considerations ($n\Phi_0 = \frac{1}{2}\tau_c\gamma_c$) and we obtain

$$\tau_c \approx \Phi_0 b^{-3}. \tag{2}$$

Equation (2) is quite useful since it provides a basic relationship between the (macroscopic) yield stress and the local energy barrier within the suspension. It is worth noting that the energy barrier (Φ_0) already contains a possible additional dependence on the concentration arising from long-range interactions, which, for simple systems, provide a dependence of τ_c on ϕ^2 [1]. This explains why in Eq. (2) the yield stress appears to be only proportional to the solid fraction (through b^{-3}). However, for a given material, it is likely that taking into account the particle (fractal) arrangement of some complex systems along with the detailed characteristics of pair interaction would provide other types of dependence closer to those obtained from experiments or theory [29,30], i.e., $\tau_c \propto (\phi - \phi_0)^p$ where p is associated with the fractal exponent of the structure and generally ranges from 2 to 3, and ϕ_0 is a critical solid fraction (percolation threshold). An exponential increase of τ_c with ϕ has also been observed in literature far from the percolation threshold [31]. When $\Phi_0(\phi)$ is known, Eq. (1) makes it possible to determine the approximate, critical, solid fraction (ϕ_0) beyond which colloidal interactions are significant, simply assuming that $N_r \approx 1$ in that case. In practice, this corresponds to a situation beyond which the suspension exhibits a yield stress. When thixotropic effects are neglected, the yield stress appearance beyond a critical solid fraction (or a critical ionic strength or pH) is reminiscent of glass transition. Indeed here, as solid fraction is increased, particles become more or less encaged though still fluctuating around their local equilibrium positions. However, the analogy is not complete because of the possible long time restructuration of the suspension at rest which could correspond to a long-range ordering.

IV. HYDRODYNAMIC VS BROWNIAN EFFECTS

The viscous force acting over the particle for some steady motion through a Newtonian fluid can be expressed as

$-K\mu_0 V$, where V is the relative velocity between the particle and a representative volume of fluid around it, and K is a coefficient which depends on the shape and size of the particle and on the orientation of the particle. For a sphere, K is equal to $3\pi r$ (Stokes equation). For a disk of diameter w, $K/\pi w$ ranges from 1.7 to 2.6 depending on its orientation relatively to the direction of particle motion. Other expressions can be proposed for rods or ellipsoids [32]. In a suspension, each particle will in fact perturb the velocity field at every other particle, a phenomenon which is not accounted for in the above expression. It has been shown that the viscosity of hard sphere suspensions can be correctly evaluated by simply considering that the particles are added progressively in a fluid of increasing viscosity as a result of previously added particles [33]. A mathematical rule for the relative viscosity (μ/μ_0) results, which makes it possible to determine the form of this function of the solid fraction (with an arbitrary parameter). The fundamental hypothesis behind this approach, which certainly fails at solid fractions close to ϕ_m, is that, when a particle is added in a suspension, it ''sees'' around it a homogeneous fluid. For a particle embedded in a suspension and moving relatively to it, it can be suggested, in analogy with the viscosity problem, that, during its motion, the particle more or less sees a fluid around it, whose viscosity is that of the geometrically identical suspension of non-Brownian, noncolloidal particles, of viscosity μ, so that the viscous force is expressed as

$$F_\nu = K\mu V, \tag{3}$$

where V is now the particle velocity relative to a representative volume of suspension. For a very dilute suspension the usual expression is recovered. Obviously expression (3) is approximate, but contains the appropriate parameters and variables. In particular it can be expected that the force is not exactly the same for a motion over a small distance, when the particle remains at some distance from neighboring particles, and for a motion over a long distance, when it needs to move over other particles. This problem is similar to that encountered with self-diffusion within a suspension: the short and long time-scale self-diffusion coefficients differ [34]. In addition the force also depends on the motions of the neighboring particles. In view of our simplified overview and considering that we shall deal with flow problems in which particles are locally constrained to move relative to each other in various directions, it is sufficient to use a single average expression (3), which reflects the expected variations of the average drag force as a function of main parameters for all types of relative motion of fluid and particles.

When a simple shear (of shear rate $\dot{\gamma}$) is imposed on our system, there is a finite, average, relative motion of particles, so that viscous effects can become significant. We consider that Brownian effects have a significant influence on viscosity when they are capable to move a particle of a distance of the order of the interparticle separation within the characteristic flow duration. In the case $N_r \ll 1$, this occurs when the ratio (Péclet number, P_e) of the hydrodynamic dissipations ($F_\nu b$) along a path of length b at the velocity $V=\dot{\gamma}b$, to the average, thermal kinetic energy (kT) is much larger than 1, with

$$P_e = \frac{K\mu b^2 \dot{\gamma}}{kT}. \tag{4}$$

In that case, one obtains the well-known shear-thinning effect for hard sphere suspensions resulting from the predominance of Brownian effects and consequently larger energy dissipations at low shear rates, and the predominance of mean hydrodynamic effects at higher shear rates [1,33]. The suspension can be considered as Newtonian both at low and high shear rates, but with two different viscosities. We remark that the Péclet number is also the ratio of a characteristic time ($T_b=K\mu b^2/kT$) of diffusion due to Brownian motion to the characteristic time of flow ($T_h=1/\dot{\gamma}$). The curve $P_e \approx 1$ thus corresponds to the transition between a regime (A) for which Brownian effects dominate toward a regime (B) for which mean hydrodynamic effects dominate. In the diagram (Fig. 1), we assume that the suspension viscosity for negligible colloidal interactions is given by the equation [33]

$$\mu = \mu_0 \left(1 - \frac{\phi}{\phi_m}\right)^{-(5/2)\phi_m}, \tag{5}$$

which has been proved to represent existing data correctly and relies on the rational theoretical approach discussed above. Equation (5) predicts that the viscosity tends toward infinity when $\phi \rightarrow \phi_m$ which is not realistic from a physical point of view. In fact, beyond a critical value of ϕ (such that $\phi_m - \phi$ is finite), colloidal interactions (see Sec. V) or contacts (see Sec. VI) may become predominant. We remark that the Péclet number is usually written with r^2 instead of b^2, which is formally similar, and even gives close values for moderately concentrated suspensions. However, the present form and the related demonstration seem more general, and might make it possible to superimpose all curves for different particle radii and solid fractions on a single master curve in a ($\mu/\mu_0, P_e$) diagram.

Now we suggest a physical scheme which makes it possible to explain in a more straightforward way the variations of the viscosity with shear rate. This leads to results similar to those obtained from dimensional analysis, but provides a physical sense to these results. We shall repeat such an approach for each transition considered in this paper. The relative viscosity of the suspension ($\eta = \mu/\mu_0$) at a given time is the result of the competition between diffusion processes due to Brownian motion and convection processes due to the macroscopic flow. As a consequence we can define a state of structure (λ) of the suspension as an instantaneous average amount of diffusing particles. The rheological behavior of the suspension depends on this amount and we have $\eta = \eta(\lambda)$. At leading order we can write the rate of change of λ as the difference between the number of particles tending to diffuse by a unit of time, and the number of particles tending to be convected by flow by a unit of time. The first term is proportional to a number (F) of ''available'' particles for diffusion and to the rate of diffusion ($1/T_b$). The second term is proportional to the number (G) of available particles for convection and to the rate of convection ($1/T_h$) so that we obtain

$$\frac{d\lambda}{dt} = \frac{F(\lambda)}{T_b} - \frac{G(\lambda)}{T_h}. \qquad (6)$$

In the steady state we have $d\lambda/dt = 0$, so that Eq. (6) reduces to

$$\lambda = H\left(\frac{T_b}{T_h}\right), \qquad (7)$$

from which we deduce that the steady state viscosity only depends on the Péclet number:

$$\eta = f(P_e) \quad \text{with} \quad f \to \eta_0 \quad \text{when} \quad P_e \to 0$$

$$\text{and} \quad f \to \eta_\infty \quad \text{when} \quad P_e \to \infty \qquad (8)$$

V. HYDRODYNAMIC EFFECTS VS COLLOIDAL INTERACTIONS

Consider the flow of a suspension in the case $N_r \gg 1$. In practice, for yield stress fluids, the yielding behavior basically takes the form of a flow curve (shear stress vs shear rate in a steady state) tending to a finite value at low shear rates. At high shear rates, for suspensions, the shear stress is proportional to the shear rate, indicating that a Newtonian behavior results from the predominance of hydrodynamic dissipations, i.e., energy dissipation due to the flow of the interstitial fluid, as in the case of moderately concentrated hard sphere suspensions at low or high shear rates. On the basis of this observation, it appears possible to estimate the importance of colloidal interactions compared to hydrodynamic effects from the value of the following dimensionless number [30,31]:

$$\Gamma^* = \frac{\mu \dot{\gamma}}{\tau_c}. \qquad (9)$$

Indeed Γ^* is the ratio of hydrodynamic dissipations within the equivalent suspension of noncolloidal particles to the suspension yield stress, which represents the main rheological effect of colloidal interactions as considered here.

It is possible to give this number a more straightforward physical sense by simply remarking that the energy required to move a particle of b at a velocity $V = \dot{\gamma} b$ (relative velocity of adjacent particle layers) from its local potential well is the sum of the approximate hydrodynamic dissipation ($F_\nu b$) and the energy barrier (Φ_0) which must be overcome. Thus colloidal interactions are predominant during flow when the following dimensionless number is smaller than 1:

$$\Gamma = \frac{K \mu \gamma b^2}{\Phi_0} = \frac{K}{b} \Gamma^*, \qquad (10)$$

which is an equivalent form of Γ^*. We remark that Γ is also the ratio of the characteristic time (T_c) (see Sec. III) for a particle to move of a distance approximately equal to b driven by colloidal forces though slowed down by hydrodynamic forces, to the characteristic time of flow (T_h), with

$$T_c = \frac{K \mu b^2}{\Phi_0} = \frac{K}{b} \frac{\mu}{\tau_c}. \qquad (11)$$

Experimental works with clay-water suspensions [31], coal slurries [12], and silica particles in silicone oil [30] showed that the simple shear behavior of suspensions for a wide range of concentrations can be superimposed on a master curve by simply scaling the shear stress by τ_c and the shear rate by τ_c/μ. Thus it was shown that colloidal interactions become negligible while hydrodynamic effects progressively become predominant when Γ^* overcomes and becomes much greater than 1. Other authors presented experimental data which could be aligned in a similar way [35]. Similar master curves were obtained [36] when scaling the shear stress with G and the shear rate with G/μ, where G is the elastic modulus, which is more or less proportional to τ_c. It has also been shown that for electrorheological fluids an analogous master curve can be obtained if one uses a similar dimensionless number [37]: the Mason number $\mu \dot{\gamma}/\varepsilon_L E^2$, where ε_L is the liquid permittivity and E the applied electric field, which has also been interpreted as the ratio of the characteristic time for two particles to come into contact under the action of the electrical field ($\mu/\varepsilon_0 E^2$) alone and the characteristic time of flow. The physical explanation of all these results appears clearly here, since from Eq. (11) the characteristic time of the material used in these works, μ/τ_c, mainly increases like T_c for an increasing solid fraction. Indeed, as a first approximation, K/b, which is simply proportional to $\phi^{1/3}$, can be considered as constant. Thus we are dealing with a transition from colloidal to hydrodynamic effects, which is equivalent to the transition from Brownian to hydrodynamic effects which clearly appears by the flow curve superimposition [1] when the Péclet number is used to plot the data. Similarly the transition is here governed by the ratio of two characteristic times (T_c and T_h).

As a consequence, as for the transition between the Brownian and the hydrodynamic regimes, we can consider that the instantaneous behavior of the suspension results from the competition between a tendency of the particles to drop into an instantaneous, local equilibrium position at a rate $1/T_c$ and the convection due to flow at a rate ($1/T_h$). Thus we can define a parameter λ which is an average (in space) instantaneous state of structure, and we can write, for $\lambda(\Gamma, t)$, an equation similar to Eq. (6), from which it results that a dimensionless shear stress ($T = \tau/\tau_c$ for example) may be expressed in steady state as a function of Γ alone:

$$T = f\left(\frac{T_c}{T_h}\right) = f(\Gamma) \quad \text{with} \quad f \to 1 \quad \text{when} \quad \Gamma \to 0$$

$$\text{and} \quad f \to \alpha \Gamma \quad \text{when} \quad \Gamma \to \infty, \qquad (12)$$

where α is a factor depending on material characteristics. The detailed evolutions of the stress in Eq. (12) result from its expression as the sum of separated contributions from hydrodynamic interactions and colloidal interactions.

The curve $\Gamma \approx 1$ thus approximately corresponds to the transition between regime (C), for which colloidal interactions dominate, and regime (B) (see Fig. 1). It is worth noting that the corresponding curve and the curves $N_r = 1$ and

$P_e = 1$ intersect in a single point for which Brownian, hydrodynamic, and colloidal effects more or less balance. In Fig. 1, in order to be consistent with most experimental data and theoretical considerations, we assumed $\tau_c \propto (\phi - \phi_c)^n$ with $n \approx 3$. For suspensions with colloidal interactions, the appearance of yield stress beyond a critical solid fraction is the equivalent of the yielding phenomenon occurring when reducing, for example, the ionic strength [38]. In that case, for model suspensions, the transition has been explained on the basis of a reduced volume fraction in Eq. (5) taking into account an effective volume of particles, including around them a volume within which colloidal interactions are strong [1,6].

It is usually considered that suspensions of noncolloidal particles remain Newtonian even at high solid fraction but with $\mu(\phi) \to \infty$ when $\phi \to \phi_m$. Under these conditions, we can take $\Phi_0 = 0$, which effectively indicates that hydrodynamic effects should be dominant as long as Brownian effects are negligible. In that case, regime (C) in Fig. 1 disappears. However even for dispersions of hard spheres assumed to be as such, slight colloidal effects might induce a yield stress at solid fraction close to ϕ_m [39]. For suspensions of larger particles, possible slight colloidal interactions can hardly affect the rheological behavior, which depends mainly on hydrodynamic effects and possible direct contacts (see below).

VI. CONTACTS

A direct contact occurs when two particles touch each other. It is in fact rather difficult to define the exact form that such a process should take. Indeed, because of possible slight colloidal interactions, particle roughness, and hydrodynamic effects, a ''true'' direct contact can hardly occur over a large surface. Contact mechanics generally involve various, complicated, processes, such as elastoplastic deformation of junctions, adhesion, and film lubrication [40]. A possible way of defining direct contact without dwelling on details is to consider it from its effects on particle dynamics rather than through its mechanisms. Thereby it is very usual to distinguish between collisional (brief duration) and frictional (sustained) contacts. This definition, which makes it possible to avoid considering the physics of contacts in detail, is quite appropriate in our case since, in order to determine the predominant interactions from a rheological point of view, we mainly need to quantify them. Thus we shall retain that, basically for a sliding frictional motion between two particles, the tangential force is proportional to the applied normal force, and that for a collision there is a momentum transfer proportional to the relative velocity of particles (for identical particles).

A. Squeezing effects

Let us assume that colloidal interactions are negligible. In that case a direct contact requires that we overcome the viscous force resulting from the flow between particles. At a small separating distance ($h \ll r$) the most significant force is the normal force due to fluid squeezing [41], which in the case of spheres, is expressed in leading order as

$$\mathbf{F} = -\mu_0 \left(\frac{3\pi r^2}{8h} \right) \mathbf{V}, \tag{13}$$

where $\mathbf{V}$ is the relative velocity of particles in the direction of the particle centers. Clearly this expression is not valid for an extremely small separating distance, since $|\mathbf{F}| \to \infty$ when $h \to 0$, which would preclude any direct contact. In fact, there exists a minimum distance below which the expression no longer holds. This may originate from various mechanisms, such as an elastohydrodynamic interplay [42], the increase in viscosity [43], and the shortcoming of continuum hypothesis [44]. As a first approximation, we shall only take the effect of particle roughness into account. As a consequence we consider that a direct contact occurs when h is of the order of the particle roughness (ε), since at this separating distance secondary flows within some channels formed by surface irregularities are significant, which tends to considerably slow down the rate of increase of $|\mathbf{F}|$. Taking $h = \varepsilon$ in Eq. (13) thus provides an estimate of the maximum force due to fluid squeezing. This assumption is in agreement with the experimental results of Smart and Leighton [45]. This approach might not be valid for particles which are nonplanar or spherical at short distances, for which it could be considered that there is more than one scale of surface irregularities, but here we shall neglect this problem. Considering that the energy dissipated through fluid squeezing becomes predominant at solid fractions close to ϕ_m, Frankel and Acrivos and other authors [41,46] computed the suspension viscosity but obtained various expressions. In addition, some authors with slightly different hypotheses obtained other expressions [47]. This is due to the fact that the result a great deal depends on the assumed, instantaneous, particle configuration [48]. All theories at least predict the singular behavior of viscosity when $\phi \to \phi_m$.

During flow, a direct contact between two particles cannot easily occur due to the large repulsive force developed as they approach each other. More precisely, in the absence of an external action on particles, during simple shear such a process cannot occur since the repulsive force (at least $\mu_0 \dot{\gamma} r^3 / h$) is much larger that the force resulting from flow (of the order of $\mu_0 \dot{\gamma} r^2$) when $h \ll r$. In that case two particles will remain lubricated by a thin fluid layer during their relative motion. This may be called a lubricated contact. If the particles are submitted to a sufficiently large external action such as pressure or gravity, direct contacts can occur.

The predominance of contacts (either lubricated or direct) necessarily results from the existence of a considerable amount of contacts throughout the suspension, so that we conclude that this situation can be associated with the existence of a continuous network of particles in contacts. Such mixtures will be referred to as granular suspensions. Since this phenomenon is associated with a percolation process, it occurs when the solid fraction is larger than a critical one (ϕ_c). Current knowledge in this field does not make it possible to specify to what extent ϕ_c depends on flow and suspension characteristics. At least slight fluctuations are expected, since the arrangement of the particle network can change under varying flow conditions. As a first approximation we shall assume that ϕ_c remains constant. Experimental and numerical results indicate that for dry uniform spheres ϕ_c should be situated between 0.5 and 0.55 [49]. In the fol-

lowing we shall examine successively the behavior of the suspension when each of the three types of contacts (lubrication, collision, and friction) dominate, and establish criteria for the transition between these regimes.

B. Lubricational regime

We emphasize that, if fluid squeezing effects are predominant at high solid fractions, the mean, shear-induced, relative motion of particle layers should also develop normal forces. Indeed, a particle in motion can no longer travel far away from neighboring particles but must more or less slide at a small distance between the particles of the surrounding layers (above and below it). As a consequence the suspension behavior is no longer Newtonian. Indeed, the particle configuration is no longer isotropic and constant, crowding effects inducing some organization or disorder, depending on shear rate, allowing relative motion, and probably tending to minimize energy dissipation [50]. An exemplary consequence of this phenomenon is the shear-thickening effect for uniform hard sphere suspensions at a critical solid fraction which has been attributed to an order-disorder transition beyond a critical shear rate [51], or to cluster formation [52]. Since this phenomenon is intimately related to a crowding effect which tends to dilate the granular phase [53], it has been referred to as dilatancy [54]. Dilatancy is known to occur with granular systems within which friction dominates, or for rapid dry granular flows [55]. Dilatancy in fact belongs to a wider range of phenomena, which could be referred to as steric effects, and which correspond to evolutions of the particle configuration under shear resulting from mutual obstruction of particles.

We deduce that there should exist a transition between regime (B) and a lubricational regime for which hydrodynamic effects are predominant but dilatancy effects also occur, possibly giving rise to non-Newtonian effects, approximately beyond a critical solid fraction (ϕ_c). If this effect is considered to be directly related to shear thickening, various data can be found in the literature concerning this critical value [21,49]. It is worth noting that, as soon as dilatancy occurs, the real solid fraction depends on flow and boundary conditions and can be smaller than the initial solid fraction. As a consequence the solid fraction in the corresponding domain in Fig. 1 must only be considered as a solid fraction of reference of the suspension. Moreover, the associated rheological behavior depends strongly on the instantaneous particle configuration, which depends on the flow history and boundary conditions. This lubricational regime occurs only at sufficiently large shear rates and for appropriate boundary conditions. In particular, below we discuss (Sec. VI C) the transition from a frictional regime toward a lubricated regime for increasing shear rate.

C. Lubricational vs frictional effects

When the suspension is free to dilate (which depends on boundary conditions and network permeability), the repulsive forces increase with the shear rate so that direct contacts remain negligible. When a given additional (normal) force is applied to the particles (for example due to gravity effects, which is in fact often the case in practice with granular systems), direct contacts can occur at low shear rates because

the repulsive forces are small. As the shear rate increases, the repulsive forces due to fluid squeezing may become sufficiently larger than the additional force, and preclude direct contacts. This phenomenon again occurs only under appropriate boundary conditions, and is, for example, at the origin of the so-called resuspension of granular suspensions [56,57]. In that case initially settled grains are found to disperse progressively through the suspension under shear.

This transition from a frictional to a viscous regime has also been observed for roughly homogeneous granular suspensions [58] under gravity. Experiments have been carried out in a vane rheometer, with suspension of glass beads in glycerol, air, or water (initially $\phi \approx \phi_m$, but the system is free to dilate slightly). At low rotation velocities the rheological behavior was governed by friction. Indeed, the measured shear stress was proportional to the suspension height in the bob and thus proportional to the mean normal stress due to gravity, and did not vary with the interstitial fluid or velocity. When the rotational velocity increases, the repulsive force increases and the gravity force, which acts vertically but transmits forces transversally through grain contacts, was now unable to maintain particles in direct frictional contact. This appeared from the fact that the suspension behaves as a Newtonian fluid within the range of largest shear rates. In addition, for a given particle size, all curves can be plotted along a master curve in a diagram [the ratio of wall shear stress to fluid height and the ratio of (repulsive) viscous force to normal force (C)]. The transition between the two regimes effectively occurs around $C=1$. However, the data for another particle size, though showing the same kind of transition, do not fall on this master curve [58]. This might reflect the well-known difficulty in dealing with the mechanical behavior of granular materials, because the strength of the percolating network of direct contacts can strongly vary, particularly with the ratio of particle size to material length. Neglecting this problem, the generalized expression for the repulsive viscous force to the normal force ratio (that we shall call Le) with any normal stress (N) also provides the dimensionless number governing the transition from a frictional (E) to a lubricational regime (F) (Fig. 1) at high solid fractions under free dilatancy conditions, which is similar to the parameter governing resuspension in Leighton and Acrivos's theory [56]

$$\text{Le} = \frac{\mu_0 \dot{\gamma} b}{N \varepsilon}. \tag{14}$$

It is easy to show that Le is also proportional to the ratio of a characteristic time (T_l) for the particle to enter into direct contact with another particle when moving through the fluid under the action of the external normal stress, with

$$T_l = \frac{\mu_0 b}{N \varepsilon} \tag{15}$$

to the characteristic time of flow (T_h). As a consequence, the scheme used for the other transitions can once again be applied in that case. Now the internal state of the suspension more or less corresponds to the amount of particles separated by the fluid film at a given time. Describing the evolutions of this state can be made with the help of an equation similar to

Eq. (6), which finally provides the following result for the dimensionless shear stress ($T = \tau/N$):

$$T = f(\text{Le}) \quad \text{with} \quad f \rightarrow Cst. \quad \text{when Le} \rightarrow 0$$

$$\text{and} \quad f \propto \text{Le} \quad \text{when } L_e \rightarrow \infty. \tag{16}$$

It should be kept in mind that the rheological behavior indicated by this approach is obviously approximate because of the aforementioned lack of knowledge in this field. For example, we simply retained Coulombian and Newtonian behavior types for the extreme regimes in Eq. (16).

D. Hydrodynamic or lubricational vs collisional effects

Bagnold [59] suggested that collisions between particles could be predominant for sufficiently large shear rates and/or solid fractions, giving rise to the so-called inertia regime. In fact the probability of occurrence of a collision seems in general extremely small except for high solid fractions or low viscous interstitial fluid. Indeed the repulsive viscous force should considerably damp particle inertia. We can evaluate this effect from the ratio (Ba, which is a modified Bagnold's number) of particle kinetic energy ($\rho_p r^3 \dot{\gamma}^2 b^2$, where ρ_p is the particle density) to the viscous energy dissipated when particles approach each other (proportional to $\mu_0 r^2 b^2 \dot{\gamma}/\varepsilon$):

$$\text{Ba} = \frac{\rho_p \dot{\gamma} r \varepsilon}{\mu_0}. \tag{17}$$

Taking a reasonable value of $\varepsilon = 10 \ \mu$m for the particles, we find that Bagnold's data correspond to Ba in the range [0.014; 6], for which it is not obvious that collision effects could have been very predominant. However, collisions can occur under other conditions. Indeed, for a dry powder of particles of 1 mm with a roughness of 1 μm sheared at a rate of 100 s^{-1}, one obtains Ba of the order of 100. Conversely, for a suspension of sand ($r \approx 1$ mm, $\varepsilon \approx 10 \ \mu$m) in water, Ba $= 10$ when $\dot{\gamma} \approx 400 \ \text{s}^{-1}$, which is a rather intense shear rate. This means in particular that collisions may occur within certain powder flows, but seldom occur within flows of most current granular pastes under usual flow conditions, which, as a consequence, cannot be modeled with the help of Bagnold's theory (see below).

We emphasize that the value of Ba only makes it possible to determine whether collisions occur or not under free dilatancy conditions and without additional forces. In particular, even if Ba is large, it is not possible to conclude that collisions are predominant. Conversely, if Ba is small, collisions may nevertheless occur if an additional force is applied to the particles or if dilatancy is not permitted. Moreover, in some cases, in particular for dry granular flows under gravity, we can expect that there is a direct transition from the frictional to the collisional regime.

Bagnold [59] also proposed an original approach to estimate the shear stress within a flowing suspension when collisions are predominant. It consists of considering that the collisional rate is proportional to the shear rate, and that the momentum transfer is proportional to the local relative velocity which is also proportional to $\dot{\gamma}$. It results that $\tau \propto \dot{\gamma}^2$,

and that normal forces develop proportionally to the shear stress as a result of similar momentum transfer processes in the direction perpendicular to shear plane. Bagnold's data were in agreement with this theory but, since this author used rather viscous interstitial fluids, we remark that this might also result from normal forces due to the fluid squeezing effects mentioned above. In addition, it is worth noting that if one uses the semiempirical expression (5) for computing the viscosity of the suspensions used by Bagnold, one finds Reynolds numbers [Eq. (20)] larger than 1000 for the data supposed to correspond to the so-called inertia regime. We can conclude that, in the inertia regime of Bagnold, macroscopic turbulence was fully developed, as mentioned by the author himself. This constitutes a crucial difference from most conditions of subsequent use of Bagnold's model by other authors in various fields (natural flows, powders, etc.). Various other experiments were carried out with wet or dry coarse particles flowing in a channel or in an annular shear cell [60–62]. On the whole, the corresponding results when $Ba \gg 1$ seem to confirm a square dependence of the shear stress on the shear rate, but several reported phenomena are disquieting. For instance, Savage and Sayed [61] reported the occurrence of secondary flows, or did not find the same stress when applying the same shear rate with different flow depths. Craig, Buckholtz, and Domoto [62] also showed the strong influence of boundary conditions on results. Bagnold's approach also initiated the development of kinetic theory for granular materials [63], which derives directly from the gas kinetic theory. In practice, as appears from existing data, the field of validity of these theories for suspensions seems nevertheless rather narrow, since it in general requires a rather strong agitation of particles and a very low viscous interstitial fluid.

E. Synthesis

Granular suspensions are characterized by the existence of a network of particles in contact. In that case steric effects (or dilatancy) occur, which may strongly affect the rheological behavior of the suspension. As far as the rheological behavior of these suspensions is concerned, it is also necessary to take into account the influence of additional forces over the particles and specific boundary conditions (in particular concerning dilatancy). Under some conditions the shear is localized along a few particle layers or the sample fractures. When dilatancy is permitted and appropriate additional forces are applied, the predominant contacts are successively of frictional, lubricational, and collisional types, as the shear rate increases. Considering the influence of additional parameters and the lack of knowledge concerning the transitions, in Fig. 1 we simply represent this domain in the form of a band comprised between ϕ_c and ϕ_m, neglecting the possible, slight variations of ϕ_c with shear rate and other parameters. In practice, and typically when gravity effects are important, another problem occurs with flows within this domain. The extent of dilatancy, which depends on the applied normal force, decreases with the depth within the suspension. As a consequence flows of highly concentrated suspensions under gravity are usually more or less heterogeneous, and do not a priori exhibit the behavior types presented above corresponding to homogeneous suspensions.

Leaving apart these problems, we suggest the following simplified scheme: beyond ϕ_c we are dealing with granular suspensions for which different flow regimes may be obtained depending on the relative values of two dimensionless numbers: (a) a frictional regime (E) for Le$\ll$1 and LeBa $\ll$1; (b) a lubricational regime (F) for Le$\gg$1 and Ba$\ll$1; and (c) a collisional regime (G) for LeBa$\gg$1 and Ba$\gg$1. As a first approximation, the suspension behavior in these regimes can be represented with the help of a Coulomb-like model (E), a Newtonian model (F) and a Bagnold-type model (G), respectively.

F. Fluid-solid coupling

Though it can provide some useful information concerning the origin of the behavior of a granular suspension, the two-phase character of the suspension has not yet been taken into account. The force required to shear the suspension is related to the deformation of the grain network and to the varying pore pressure, i.e., the pressure within the interstitial fluid. In soil mechanics the importance of this phenomenon and its qualitative effects have been recognized for a long time [64]. The macroscopic effects of this physical process are highly dependent on the instantaneous particle configuration (loose or dense samples in soil mechanics) and boundary conditions [additional (normal) force applied and possibility of dilatancy (drained or undrained samples in soil mechanics)]. It has also been suggested that the role of rapid pore pressure fluctuations during the motion of certain wet granular masses could be crucial [65].

From a more general point of view the extent of coupling between the solid and liquid phases may be estimated by considering the Stokes number (St) which is the ratio of the characteristic time ($T_R = \rho r^3/K\mu_0$) of the motion of a particle submitted to viscous drag and inertia, to the characteristic time of flow of the suspension:

$$\text{St} = \frac{\rho_p r^3 \dot{\gamma}}{K\mu_0}. \tag{18}$$

When St$\gg$1, the particle motion is not subjected to the interstitial fluid motion. Since in our case (granular suspensions) we hypothesized a large solid fraction, this means that the solid phase governs the behavior of the whole suspension. If $\rho_p \approx 1000$ kg m^{-3}, $\dot{\gamma} \approx 1$ s^{-1}, and $K \approx r$, this occurs for a suspension of grains with a diameter larger than 1 mm in air or larger than 1 cm in water. In that case we are dealing with what is often referred to as granular flows.

When St$\approx$1, the coupling between the two phases is weak. In that case the mixture may exhibit a two-phase behavior since each phase is partially independent of and dependent on the other phase.

When St$\ll$1, the particle motion is mainly dictated by that of the fluid phase. All happens as if the particles were a part of the fluid. This, for example, occurs for a suspension (other characteristics as above) of grains of diameter smaller than 1 μm in air and smaller that 10 μm in water. It is worth noting that the suspension behavior can nevertheless still be dictated by interactions between the solid particles. This material type corresponds to what is often referred to as granular pastes. In that case fluid flows through the porous medium formed by

the grain network are extremely difficult because they induce very large pressure gradients. Indeed, Darcy's law predicts that the pressure gradient for the laminar steady flow of a Newtonian fluid through a porous material at a mean velocity **U** is expressed as $\nabla p = -\mu_0 \mathbf{U}/k_0$, where k_0 is the permeability of the porous structure. It is instructive to recall that $k_0 = d^2/12$ for a simple channel made of two parallel planes separated by a distance d. More generally it has been shown that the permeability of a porous material increases with the size of pores. Thus, under fixed dilatancy effects, the induced pressure gradient decreases with the separating distance between particles, and increases with the solid fraction. In order to estimate k_0 for a bead pack, one may use the empirical formula of Kozeny-Carman, $k_0 = r^2(1-\phi)^3/45\phi^2$. As a consequence, for a shear flow inducing a net velocity through the porous network, the amplitude of the dimensionless pressure gradient can also be written

$$\frac{\nabla p}{\rho_p g} \approx \frac{1}{\text{St}}\left(\frac{10U\dot{\gamma}}{g}\right). \tag{19}$$

This result demonstrates that the tendency of granular pastes to dilate is strongly counteracted by the difficulty of flow through the porosity, which tends to induce large pressure gradients. Thus it is probable that, as long as the particle network is sufficiently loose, flow induces local particle rearrangements instead of grain network dilatancy. When the solid fraction is larger than a critical value, flow tends to induce dilatancy, which would give rise to large pressure gradients. Then, for the material, a solution to minimize energy dissipation is to deform along specific surfaces or even (in stretch flow) to separate into two distinct parts. This might be the explanation for fracture in highly concentrated granular pastes. It has effectively been observed that beyond a critical solid fraction, highly concentrated suspensions of fine particle (pastes) fracture when submitted to shear [31,66,67] after a critical deformation. In that case the fracture takes the form of a localization of deformation in a very thin material layer. Fracture also occurs for a paste stretched between two plates, but in that case it takes the form of a separation of the sample in two parts [68]. As already mentioned, these phenomena depend highly on boundary conditions. For example, in order to avoid fracturing, it is necessary to knead concentrated pastes such as modeling clay or pastry. During this operation, one provides additional energy to favor or balance large pressure gradients due to local or macroscopic fluid transfers resulting from local dilatancy. We remark that the same type of energy is required to squeeze a paste in order to withdraw the interstitial fluid. This also makes it possible to propose some elements for a physical interpretation of the Atterberg limits [18]. The so-called liquidity limit is arbitrarily defined as the minimum water content for which flow under particular initial and boundary conditions (vibrations) leads to close a crack (of given size) made at the material surface, and should thus be related to a critical yield stress of the suspension. The so-called plasticity limit is defined as the maximum water content for which material rods (of given size) submitted to an elongation break, and could thus be related to a dilatancy criterion under particular normal stresses (pressure arising from the hands of the experimenter).

VII. TURBULENCE

Usual inertia effects become predominant compared to hydrodynamic effects, giving rise to turbulent flow, when the Reynolds number (Re) is sufficiently large, with

$$\mathrm{Re}=\frac{\rho\dot\gamma L^2}{\mu},\qquad(20)$$

where L is a characteristic length of the macroscopic flow over which the average (in time) velocity significantly varies. Re is the ratio of inertia effects ($\rho\dot\gamma^2 L^2$) to hydrodynamic dissipations ($\mu\dot\gamma$). We remark that the suspension viscosity and not the fluid viscosity must be used in Eq. (20). For non-Newtonian fluids (such as yield stress fluids) the correct dimensionless number can include additional material parameters but, though some theories and experiments already exist [69], the range of Re for the transition toward turbulence has not yet been completely determined for complex fluids. As a first approximation it is sufficient in that case to use a generalized Reynolds number with the apparent viscosity ($\tau/\dot\gamma$) instead of μ. It is worth noting that, for suspensions, this approach relies on the doubtful assumption that there is a direct transition from the colloidal regime to the turbulent regime. When this is not the case the suspension in the hydrodynamic regime is Newtonian, and expression (20) is relevant. In addition, since turbulence is the result of an instability, its occurrence is conditioned by various factors such as the macroscopic characteristic length (L) of the flow, the roughness of boundaries, the form of the flow geometry, initial conditions, etc. As a consequence the range of Re corresponding to the transition to turbulence varies in a wide extent with the system under consideration.

With a suspension, other inertia effects can be distinguished [70]. These are turbulence within the interstitial fluid and fluctuations of particle motions around mean motions. Quantifying each of these processes requires using a dimensionless number which is the ratio of inertia resulting from the flow of fluid between neighboring particles (or from a relative motion of two close particles) to hydrodynamic dissipations. This in fact yields a single dimensionless number as follows:

$$\mathrm{Re}_p=\frac{\rho\dot\gamma b^2}{\mu_0},\qquad(21)$$

Though the range of Re_p for the transition toward the different corresponding regimes can be different, it is reasonable to consider that in general these effects occur more or less beyond the same critical conditions for a given system. Moreover the question remains open whether these inertia effects can occur independently of macroscopic turbulence. Indeed, we have

$$\mathrm{Re}_p=\left(\frac{b}{L}\right)^2\frac{\mu}{\mu_0}\mathrm{Re},\qquad(22)$$

so that, in general, Re_p, though formally similar to Re, is much smaller, which might mean that these effects should occur only when macroscopic turbulence occurs. In addition,

at sufficiently large Reynolds numbers, boundary layers can detach from the particles leading to complex effects if they interlace with the slipstreams [71]. Considering our poor knowledge of this topic, in Fig. 1 we used only the generalized, macroscopic, Reynolds number given by Eq. (20) in order to position the transition between regime (B) and regime (D), for which either macroscopic or microscopic turbulence is significant.

VIII. SYNTHESIS AND CORRESPONDING RHEOLOGICAL TRENDS

The present study makes it possible to define more clearly a concentrated suspension. We suggest that this simply corresponds to a suspension for which either colloidal interactions or contacts dominate (respectively, $\Gamma>1$ or $\phi>\phi_c$). Remark that this definition is related to a flow regime and not simply to suspension characteristics. Concentrated suspensions are thus obtained when particle interactions play a major role in the suspension behavior. For some silica or clay suspensions this may occur for solid fractions as low as 0.1%.

Our diagram (Fig. 1) makes it possible to have an overview of the possible rheological behavior of a given suspension type. For a dilute suspension we have a shear-thinning behavior when one increases the shear rate, since Brownian motions dominate at low shear rates. Sufficiently increasing the shear rate leads to turbulent flow. For a concentrated colloidal suspension, i.e., for which colloidal interactions dominate at low shear rates, we are dealing with a yield stress fluid. In addition it is viscoelastic, because elasticity can provisionally be stored when particles only climb along their local potential well without leaving it. It is thixotropic, because there are characteristic durations associated with the time for coming back to the bottom of its local, provisional, potential well, and the time for the long-range particulate structure to rearrange. However viscoelasticity and thixotropy are not always apparent since they are associated with time scales which can be much smaller than the flow time scale. As long as the shear rate increases, hydrodynamic effects play an increasing role and can become predominant. At this stage the suspension behavior is that of the equivalent suspension of noncolloidal particles with the same shape. Then turbulence may occur at larger shear rates. For all these regimes the suspension behavior may be determined from a local point of view.

The rheological behavior of granular suspensions (for $\phi>\phi_c$) was discussed in Sec. VI E. This region correspond to regimes for which the network formed by particles in lubricated, or direct contacts throughout the suspension plays a critical role. The mutual force between two neighboring particles is then always of a repulsive type, so that displacing one particle within the suspension requires a large force, making it possible to deform the whole network. As a consequence the suspension behavior depends on the possible, additional, external force and boundary conditions. The behavior of the granular suspension has fundamentally a non-local character. It could be referred to as a ''hard'' suspension in opposition to the ''soft'' suspensions which are obtained in the other regions.

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