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## Motion of a solid object through a pasty (thixotropic) fluid

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For materials assumed to be simple yield stress fluids the velocity of an object should continuously increase from zero as the applied force increases from the critical value for incipient motion. We carried out experiments of fall of a sphere in a typical, thixotropic, pasty material (a laponite suspension). We either left a sphere falling in the fluid in different initial states of structure or vibrated the fluid in a given state of structure at different frequencies. In each case three analogous regimes appear either for increasing restructuring states of the fluid or decreasing frequencies: A rapid fall at an almost constant rate; a slower fall at a progressively decreasing velocity; a slow fall at a rapidly decreasing rate finally leading to apparent stoppage. These results show that the motion of an object, due to gravity in a pasty material, is a more complex dynamical process than generally assumed for simple yield stress fluids. A simple model using the basic features of the (thixotropic) rheological behavior of these pasty materials makes it possible to explain these experimental trends. The fall of an object in such a fluid thus appears to basically follow a bifurcation process: For a sufficiently large force applied onto the object its rapid motion tends to sufficiently liquify the fluid around it so that its subsequent motion is more rapid and so on until reaching a constant velocity; on the contrary if the force applied onto the object is not sufficiently large the fluid around has enough time to restructure, which slows down the motion and so on until the complete stoppage of the object. © 2004 American Institute of Physics. [DOI: 10.1063/1.1640372]

### I. INTRODUCTION

The motion of solid objects such as coarse particles through concentrated suspensions, emulsions or foams is a subject of practical importance since it for example conditions sedimentation or migration in various industrial processes involving such materials. For example, in order to drive rock debris within drilling fluids it is necessary that they do not settle through the flowing paste, sedimentation during the preparation of concrete may lead to heterogeneities which may significantly weaken the final structure, or the settling of cocoa or sugar particles during chocolate preparation would give it a bad taste.

In a Newtonian liquid the steady velocity ( $V$ ) of a sphere (of radius  $R$ ) falling through it under the action of gravity ( $g$ ) results from the balance between the drag force ( $6\pi R\eta V$ ) and the apparent weight of the sphere in the fluid:

$$V = \frac{2}{9} \frac{(\rho_s - \rho)gR^2}{\eta}, \quad (1)$$

where  $\rho_s$  and  $\rho$  are, respectively, the solid and liquid densities. Here the apparent viscosity of the fluid,  $\eta$ , is a constant. In shear-thinning liquids this parameter also depends on the shear rate,  $\dot{\gamma}$ , so that the fall velocity is a more complex function of the parameters but still differs from zero as soon as  $\rho_s - \rho \neq 0$ .<sup>1</sup> For yield stress fluids the apparent viscosity

tends to infinity as the shear rate tends to zero, so that the velocity in (1) may be equal to zero even for a finite density difference.

In practice it has been considered that the ability of pasty materials to keep denser particles without settling originates in their yield stress ( $\tau_c$ ), i.e., a critical stress which has to be overcome for the material to flow. This property is generally expressed with the help of a yielding criterion, such as the von Mises one,<sup>2</sup> which writes

$$\sqrt{-T_{II}} < \tau_c \Rightarrow \mathbf{D} = 0, \quad (2)$$

in which  $\mathbf{D}$  is the strain rate tensor and  $T_{II}$  the second invariant of the deviator of the stress tensor  $\mathbf{T}$ . In this context, unlike simple liquids, a yield stress fluid initially at rest *a priori* remains unperturbed beyond a finite distance from an object moving through it. Indeed the additional stress induced by the object motion decreases to zero away from the object and thus the criterion (1) is fulfilled at a specific distance in each direction, which forms a surface  $S$ . In the absence of inertia effects it follows from the momentum balance over the fluid volume within  $S$  that the drag force on the object moving in the direction of unit vector  $\mathbf{u}$  may be written

$$F_D = \int_S (\mathbf{T} \cdot \mathbf{n}) \cdot \mathbf{u} ds, \quad (3)$$

in which  $\mathbf{n}$  is the outer normal vector at any point on the surface  $S$ . We deduce from dimensional considerations that the drag force may be expressed as

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$$F_D = 4\pi R^2 \tau_c f(S), \quad (4)$$

in which  $f$  is an increasing, positive, dimensionless function of the extent of  $S$  relatively to the surface of the sphere. We obviously expect that, for a given sphere, the surface  $S$  increases as the object velocity increases, so that the drag force increases. On the contrary, when the velocity decreases to zero,  $S$  should tend to a critical value  $S_c$ , from which we can compute the drag force for incipient motion through a yield stress fluid:  $F_c = 4\pi R^2 \tau_c f(S_c)$ . Within this frame the object velocity should conversely decrease to zero as the force exerted on it decreases to  $F_c$ .

Previous works (see, for example, the review of Chhabra and Uhlherr<sup>3</sup>) in this field focused on the experimental measurement of the incipient force or the fall velocity under gravity, and the analytical computation or numerical simulation of the flow characteristics around a sphere moving through a yield stress fluid at rest, which in any case implies to determine  $f(S_c)$  or  $f(S)$ . However, there still lacks a consensus on the corresponding values. For example, for a sphere,  $f(S_c)$  was found between 0.8 and 3.5 depending on authors.<sup>4–7</sup> The different, recent, numerical simulations of the flow field, which all assume that the fluid has a very high viscosity at low shear rates instead of a perfect yielding behavior,<sup>7–9</sup> now provide a more complete, though numerical, solution of the problem under various conditions, but these predictions have not been compared in detail with experimental data. Note that all the theoretical works, which assume a simple yielding behavior, predict a particle velocity progressively decreasing to zero as the applied force tends to the critical value for incipient motion.

Although qualitatively in agreement with above expectations, i.e., object motion only beyond a critical density or volume, the experimental results presented in literature in fact raise several questions. A first critical observation concerns the very poor reproducibility of data for a settling velocity smaller than  $10 \text{ cm} \cdot \text{s}^{-1}$ .<sup>10</sup> Other authors<sup>11,12</sup> pointed out that reproducible results could be obtained only after releasing 4–10 spheres. A second critical observation is that all data in literature<sup>4,10,12–13</sup> correspond to relatively large settling velocities: The velocity to particle diameter ratio (characteristic shear rate) is never below  $1 \text{ s}^{-1}$  and is generally much larger. At last ultrasonic observations in bentonite suspensions show velocity increase as the object gets deeper in the fluid.<sup>14</sup> The final velocity is five times the initial value.

Such problems do not occur in simple liquids: For example, by increasing the fluid viscosity or decreasing the solid–liquid density difference one obtains a continuous set of fall velocities tending to zero. Relevant explanations for the above problems cannot either be found in the viscoplastic behavior of the fluids: As the solid density decreases or the yield stress increases towards critical values the settling velocity should reach any value down to zero with an analogous reproducibility and without any significant time effects. Above problems thus necessarily result from some uncontrolled time and flow-dependent mechanical properties of the fluid, which suggests that the possible thixotropic behavior of the pasty materials should also be taken into account for predicting the characteristics of the slow fall of particles

through them. Clearly in these regimes the Bingham or Herschel–Bulkley are insufficient to describe flow properties, which suggests that viscoplastic fluids are also significantly time-dependent at low velocities. This conclusion is in agreement with experiments under simpler flow conditions<sup>15</sup> which showed that the flow of a layer of yield stress fluid lying over an inclined plane is unstable: In contrast with the predictions of the viscoplastic models it starts to flow abruptly at a critical slope when inclining the plane and stops flowing abruptly at another, smaller, critical slope. In this frame the apparent viscosity should now be expressed as a function not only of the shear rate but also on the current state of structure, that we will simply describe here by a single variable  $\lambda$ . A fundamental consequence is that the motion of an object in a paste is a time-dependent phenomenon.

The aim of this work is to study, both experimentally and theoretically, the influence of thixotropic effects on the motion of solid objects under the action of gravity in pasty materials. In order to study these effects we chose a model, transparent, thixotropic fluid (a laponite suspension), which evolves over large characteristic times. In particular its apparent yield stress significantly increases after large durations at rest. In the first part we present experimental results concerning experiments of sphere fall in the fluid at rest under different restructuring times. Three regimes of fall were distinguished, apparently indicating either a negligible or significant influence of fluid aging. Experiments with the fluid at a fixed age but submitted to vibrations at different frequencies lead to similar regime observations. In the second part we propose a theoretical interpretation of these trends taking into account fluid thixotropy. In this aim we use a simple generic rheological model and propose a simplified theoretical approach of the flow problem which provides velocity vs time curves as a function of parameter values and initial conditions, in qualitative agreement with the experimental ones.

## II. EXPERIMENTS

### A. Materials and procedures

We used a transparent colloidal suspension made by mixing distilled water and Laponite, a synthetic clay (Laponite, Grade RD, *Rockwood Additives Limited*). The solid volume fraction was 1.1% and NaOH was added in the solution so as to have a pH close to 10. The suspension ( $\rho = 1017 \text{ kg} \cdot \text{m}^{-3}$ ) was mixed at a velocity of 2000 rpm during 8 hr, then left at rest during seven days. Such a preparation ensured that significant irreversible aging did not occur within the maximum duration of the experiments (a few hours).

Then the fluid was mixed again during 30 min and put into a cylindrical vessel with a depth of 21 cm and an inner diameter of 3.9 cm. This vessel had graduations every 1 cm. There the fluid was left at rest a given time during which it could restructure so that its apparent viscosity increased. Then an iron sphere ( $R = 1.5 \text{ mm}$ ;  $\rho_s = 7780 \text{ kg} \cdot \text{m}^{-3}$ ) was put with pliers and withdrawn in the fluid just beneath the free surface and close to the central axis of the vessel. When it was put carefully the sphere fell down vertically in the fluid. Then we recorded the time whenever the sphere came

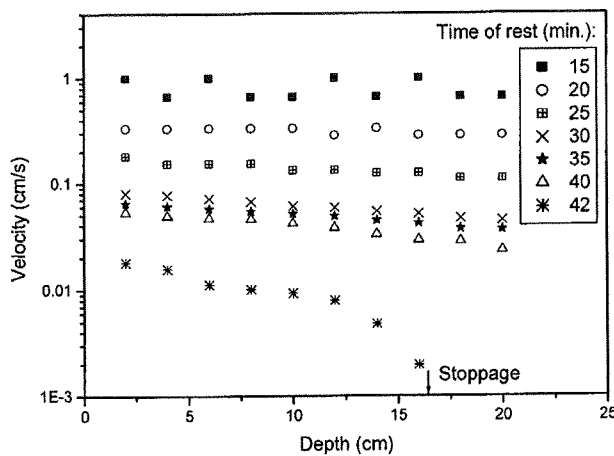


FIG. 1. Fall of a sphere in a laponite suspension (globally) at rest in different initial states of structure (time of rest after mixing): velocity as a function of the distance from the initial position of the sphere. For a time of rest of 45 min the sphere did not move at all over several hours.

in front of each graduation. Obviously the relative uncertainty on our measurements was larger for higher fall velocities but this does not affect our analysis since the velocity remains almost constant for rapid drops. The particle Reynolds number was always smaller than  $4 \cdot 10^{-3}$ , which indicates that the flow around the sphere was laminar.

We also carried out similar experiments by vertically vibrating the fluid. In this aim the vessel was fixed and shaken vertically over a horizontal, vibrating plate (Vibrator V403) controlled by a digital sine controller (DSC-4) and a power amplifier PA100E (Ling Dynamics System). With this setup the amplitude and the frequency of vibrations could be monitored separately by a piezo-electric accelerometer positioned onto the vibrator. The results presented here were obtained with one amplitude only (1 mm) and different frequencies. These experiments were carried out with the laponite suspension after two different times following its preparation and with a Newtonian liquid (a commercial honey).

## B. Results

### 1. Fall through the fluid at rest

The results for the sphere fall in the liquid at rest after different times of rest before the test are presented in Fig. 1. Three regimes appear:

- For sufficiently short times of rest the sphere falls at an apparently constant, large velocity so that it rapidly reaches the vessel bottom;
- for long times of rest the sphere either apparently does not move at all or starts to move but its velocity rapidly decreases so that it finally stops moving somewhere in the fluid before reaching the vessel bottom;
- for intermediate times of rest the sphere falls at an intermediate velocity which continuously decreases.

These results were reproducible to within 10% on average velocity and the main source of uncertainty comes from the control of the initial state of the fluid.

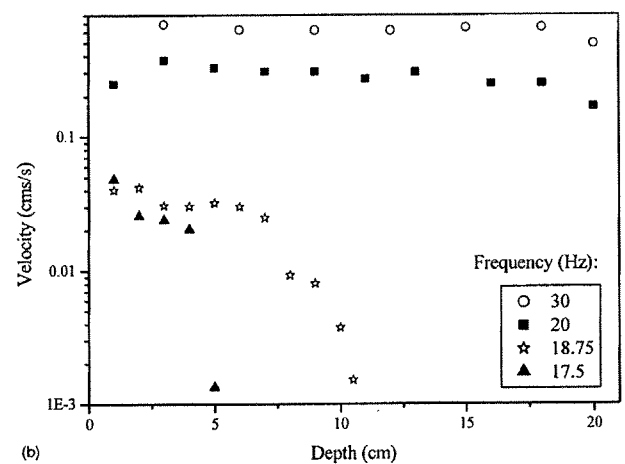
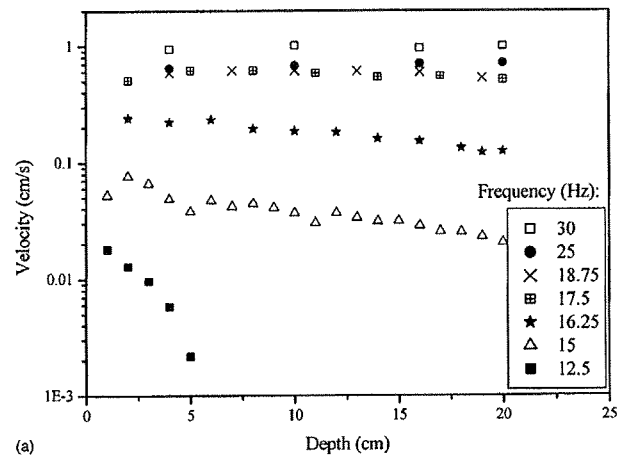


FIG. 2. Fall of a sphere in a vibrated laponite suspension at rest for different frequencies: Velocity as a function of the distance from the initial position of the sphere for the fluid after 45 min (a) and 60 min (b) of rest.

### 2. Fall in a vibrated fluid

For the vibration tests we first observed that the fall of the sphere in a Newtonian liquid (commercial honey) occurs at the same, apparently constant velocity with and without vibrations at any frequency.

With the laponite suspension the results of vibration tests at different imposed frequencies appeared to be similar to those for the fluid at rest under different initial restructuring times: The three regimes (a)–(c) are obtained, respectively, for high, low and intermediate frequencies (cf. Fig. 2).

## C. Discussion

Our results first suggest a possible explanation for the experimental problems mentioned in literature: the reproducibility of velocity measurements at low rates of fall is poor certainly because it may significantly depends on the fluid preparation and the position at which it is recorded in the fluid.

Our experiments also show that there is a strong effect of thixotropy on the fall characteristics. At this stage we can nevertheless suggest a simple, qualitative explanation for that. For a fluid initially sufficiently liquid (for a short time of rest) the sphere falls rapidly and the fluid has not enough

time to restructure during this fall, so that the fall velocity remains almost constant. For a fluid having initially a large viscosity (for larger times of rest) the sphere velocity is small and its slow displacement leaves to the fluid enough time for significantly further restructure. This leads to a viscosity increase which in turn decreases the fall velocity and so on. Such a process may obviously lead to the complete stoppage of the sphere. In this context the intermediate regime, in which the velocity continuously decreases, does not find a simple explanation yet.

Concerning the motion in the vibrated fluid it is first necessary to explain the sphere fall beyond a critical frequency whereas this sphere does not move at all before. We may first suspect a kind of (macroscopic) liquefaction of the whole fluid due to these vibrations, leading to a decrease of its viscosity. However, as long as its free surface remains horizontal, the fluid globally moves in mass and flows as a result of the sphere motion relatively to it. As a consequence there is no reason for which it should destructure as a direct effect of vibrations only. Another possibility is a liquefaction at the scale of the particles because of inertia effects due to vibrations. However, because of the very small size of these clay particles (disk-like particles of about 30 nm diameter and 1 nm thickness) it is easy to see that the corresponding inertia terms are several orders of magnitude lower than the viscous terms induced by some characteristic motion of a clay particle relatively to the interstitial liquid (water): The Stokes number is extremely small.<sup>16</sup> It results that the clay particles in general follow any motion in mass of the liquid imposed by the vibrations of the fluid, and thus no destructuring can be expected in such a way. Finally it seems to remain only one possible explanation: There is a complex interaction between the sphere motion and the fluid state at a macroscopic scale; the fluid flow that the sphere causes around it has an influence on its subsequent motion leading to a dynamical process with various possible evolutions. We suspect that this process has some similarity with that occurring for the sphere fall in the fluid at rest after different restructuring times. In the following we attempt to quantify these processes in order to understand more completely the different regimes by taking into account the basic mechanical processes playing a role and in particular the fluid behavior.

### III. THEORY

#### A. Rheological model

In literature various models have been proposed in the aim of describing thixotropy<sup>17-23</sup> but they have a common basic structure: an apparent viscosity depending on the shear rate and the structure parameter  $[\eta(\dot{\gamma}, \lambda)]$ , and a kinetic equation giving the time evolutions of  $\lambda$ , with the following, rather generic, shape:

$$\frac{d\lambda}{dt} = F(\lambda) - G(\lambda) \dot{\gamma}, \quad (5)$$

in which  $F$  and  $G$  are two material functions. The first term of the right hand-side of (5) expresses the rate of restructuring of  $\lambda$  while the second term expresses the destructuring rate which is proportional to the rate of shear. The form of

the apparent viscosity is more or less complex depending on authors but here we will use a very simple expression, only dependent of the material state

$$\eta = \eta_0(1 + \lambda^n), \quad (6)$$

in which  $\eta_0$  and  $n(\geq 1)$  are two constant parameters. For the kinetic equation we will use an extremely simple form of the type (5), by taking  $F = 1/\theta$  and  $G = \alpha\lambda$ , where  $\theta$  and  $\alpha$  are material parameters, which gives

$$\frac{d\lambda}{dt} = \frac{1}{\theta} - \alpha \dot{\gamma} \lambda. \quad (7)$$

Although this is one of the simplest representations of a thixotropic rheological behavior this model already appears capable to predict critical qualitative observations such as the viscosity bifurcation of various pasty materials under creep tests:<sup>15,24,25</sup> When a constant stress is applied their viscosity either evolves towards a given, low value or to infinity depending on the relative value of this stress and a critical value. This phenomenon was confirmed by MRI observations<sup>26,27</sup> or by direct observations in the case of transparent laponite suspensions<sup>28</sup> showing that these materials can flow in steady state only at a shear rate beyond a critical, finite value, associated to the critical stress; in the regions where the stress is smaller than the critical stress the shear rate progressively tends to zero. The practical consequence is a shear localization with a discontinuity in shear rate at the interface between two different regions. Finally the quantitative predictions of models (6) and (7) under various steady or transient conditions for a bentonite suspension were successfully compared with the local flow properties (as measured by MRI) [N. Roussel, R. Le Roy, and P. Coussot (submitted)].

#### B. Fall of an object in a thixotropic fluid

With this model, the velocity of a sphere through the fluid in a homogeneous state  $\lambda$  is simply deduced from the expression (1)

$$V = \frac{2}{9} \frac{(\rho_s - \rho)gR^2}{\eta_0} (1 + \lambda^n)^{-1} = \frac{W_0}{(1 + \lambda^n)}. \quad (8)$$

The parameter  $W_0 = 2(\rho_s - \rho)gR^2/9\eta_0$  expresses a reference fall velocity of the sphere as a result of its apparent weight through a completely destructured fluid ( $\lambda=0$ ). In fact this approach is valid only at the very first time of motion when the fluid can be considered in a homogeneous state. Indeed then the fluid close to the sphere destructures while the fluid far from it tends to restructure, thus leading to a heterogeneous, time-dependent distribution of  $\lambda$ . The complete solution of this problem would require a two-dimensional (2D)-numerical approach taking into account boundary and initial conditions, and from which the different flow regimes might not be easily distinguished. Here we only review simplified, analytical approaches intending to provide a qualitative description and understanding of major trends.

In this frame a very simple approach consists in assuming that the sphere "sees" at each time around it a fluid the state of which evolves as if it were at rest. Thus here we

simply take into account the restructuring but leave apart the destructuring resulting directly from the sphere motion. Under this assumption it follows from (8) and (7) in which we assume  $\dot{\gamma}=0$  that

$$V = W_0(1 + (\lambda_0 + t/\theta)^n)^{-1}, \quad (9)$$

where  $\lambda_0$  is the state of the material at  $t=0$ . Equation (9) shows that the velocity of a falling object should progressively decrease in time. Our MRI experiments with laponite and bentonite suspensions (unpublished results) show that the values of  $n$  that can be found by fitting the above rheological model to steady-state data are very close to 1. Note that this assumption is relevant because we only take into account the restructuring process but otherwise  $n$  must be strictly different from 1 for the observed trends (rheology and sphere fall) to be explained by the theory. Using here this specific value and the initial condition  $x(t=0)=0$  we can find by integrating (9) the expression for the velocity as a function of the distance  $x$  covered by the object from the initial time

$$V = \frac{W_0}{1 + \lambda_0} \exp\left(-\frac{x}{W_0\theta}\right). \quad (10)$$

Equation (10) predicts that the velocity does not differ strongly from  $W_0/(1 + \lambda_0)$  over the distance  $W_0\theta/2$ , then the velocity decreases to almost 1/10 of its initial value at the distance  $2W_0\theta$  and finally progressively tends to zero. This model thus predicts an apparent stoppage if the length of observation is sufficiently large, but this stoppage is much slower than in our experiments. However, the strongest disagreement between this model and our tests is that it predicts a master curve  $[\exp(-x/W_0\theta)]$  for the velocity scaled by its initial value as a function of the distance.

A further approach consists in considering also the fluid destructuring due to the flow induced by the motion of the object. For a Newtonian fluid we know that the disturbance due to the sphere extends to a considerable distance  $r$  from the sphere since the fluid velocity approaches zero at large distances as  $r^{-1}$ .<sup>29</sup> The corresponding second invariant of the strain rate tensor more rapidly decreases as  $r^{-2}$ . For a shear-thinning fluid, i.e., with a viscosity decreasing with this second invariant (equivalent to the shear rate in simple shear), the velocity will decrease much more rapidly so that in general a small volume (say few times the sphere volume) around the sphere should be affected by the sphere motion. For a fluid following the constitutive equations (6) and (7) this effect is amplified since the viscosity in the regions close to the particle tends to decrease as a result of particle motion, while the viscosity in the farther regions tend to increase due to restructuring under low rates of flow. Thus for a thixotropic fluid the volume significantly affected by sphere motion should generally be restricted to a volume of fluid of the order of that of the sphere.

Consequently we assume that, at a given time, only a limited cylindrical volume of fluid around the sphere is affected by its motion. We can expect some variations of the length of this cylinder as the particle velocity varies but as a first approximation we will neglect them. More precisely at a given time  $t$  we assume that the fluid is only affected by the

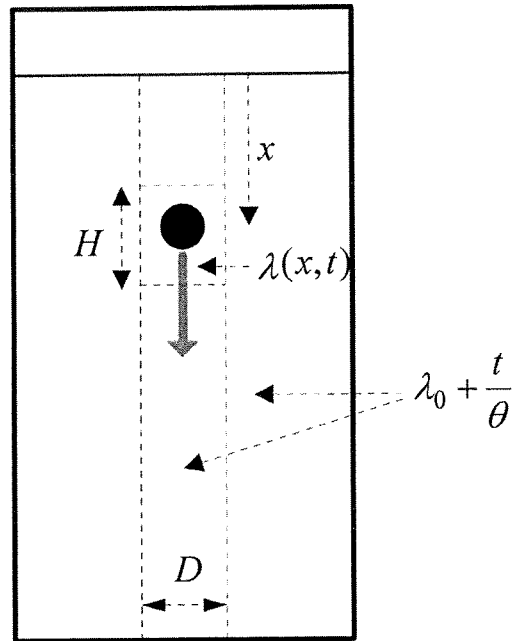


FIG. 3. Schematic representation of the different regions (see text) of fluid during the motion of a sphere through a thixotropic paste: The central region is a cylinder in which the fluid flows at some time while the rest of the fluid remains undisturbed.

sphere motion in a cylinder portion of length  $H$  around the sphere situated at a mean distance  $x(t)$  from a reference point (cf. Fig. 3). Now we can follow the change of the average value of the instantaneous state in this cylinder when the sphere moves of a distance  $dx$  in a time  $dt$ , from the material derivative of  $\langle\lambda\rangle = 1/\Omega \int_{\Omega} \lambda dv$ :

$$\frac{D}{Dt} \langle\lambda\rangle = \left\langle \frac{\partial\lambda}{\partial t} \right\rangle + \frac{1}{\Omega} \int_S \lambda \mathbf{u} \cdot \mathbf{n} ds, \quad (11)$$

in which  $\Omega$  and  $S$  are, respectively, the volume and surface of the cylinder portion,  $\mathbf{u}$  the velocity of the fluid relatively to  $S$  and  $\mathbf{n}$  the outer unit normal vector. In order to compute (11) we will make several assumptions:

- (i) The flow to which the fluid is submitted on average within  $\Omega$  induces an effect on  $\lambda$  equivalent to that induced by a simple shear at a shear rate  $\dot{\gamma} \approx V/e$ , where  $e$  is a characteristic thickness of shear which only depends on cylinder characteristics; if in addition we take into account (7) this leads to consider that  $\langle\partial\lambda/\partial t\rangle \approx 1/\theta - \alpha\dot{\gamma}\langle\lambda\rangle$ .
- (ii) On average there is no net vertical or radial fluid flow; thus the flux through the moving volume  $\Omega$  only results from the entrance of a fraction of yet undisturbed fluid from downstream in the cylinder portion [from (7) the state of this fluid writes  $\lambda_0 + t/\theta$ ] and from the exit of a fraction of disturbed fluid in the average state  $\langle\lambda\rangle$ .

Under these conditions, Eq. (11) now writes

$$\frac{d\lambda}{dt} = \frac{1}{\theta} + \left( \frac{1}{H} \left( \lambda_0 + \frac{t}{\theta} \right) - \left( \frac{\alpha}{e} + \frac{1}{H} \right) \lambda \right) V, \quad (12)$$

in which we dropped the average signs around  $\lambda$ . The complete set of equations describing the sphere motion is composed of (12) and (8) along with initial conditions.

It is first interesting to analyze the problem in the case where the time variations of the state of the undisturbed material in the cylinder can be neglected, i.e.,  $\lambda_0 + t/\theta \approx \lambda_0$ . Here Eq. (12) may be expressed in the simplified, dimensionless form:

$$\frac{d\lambda}{dT} = 1 + W \frac{(\lambda_0 - (1 + \beta)\lambda)}{1 + \lambda^n} = f(\lambda), \quad (13)$$

in which  $T = t/\theta$  (dimensionless time),  $W = W_0 H/\theta$ , and  $\beta = \alpha H/e$ . Note that  $W$  and  $\beta$  are two parameters independent of  $\lambda$ ,  $\lambda_0$ , and  $t$ . The minimum of  $f$  for positive values of  $\lambda$  is reached for a particular value  $\lambda_c > \lambda_0$ . If  $f(\lambda_c) > 0$ ,  $f(\lambda)$  remains positive and tends to 1 when  $\lambda \rightarrow \infty$ , which means that  $\lambda$  increases and tends towards infinity; the viscosity thus also tends towards infinity and the sphere eventually apparently stops moving. If  $f(\lambda_c) < 0$ ,  $f(\lambda)$  has two roots,  $\lambda_1$  and  $\lambda_2$  ( $\lambda_1 < \lambda_2$ ). In that case the sphere would evolve either towards steady state motion if  $\lambda_0 < \lambda_2$ , or towards apparent stoppage otherwise. Since we can check that  $\lambda_0 < \lambda_c < \lambda_2$  this is always the former situation which prevails.

Finally, for a given initial state  $\lambda_0$ , there are only two possible cases depending on the relative values of  $W$  and  $W_c$ , defined by  $f_{W_c}(\lambda_c) = 0$  ( $W_c$  is in fact solely a function of  $\lambda_0$  and material parameters):

- (i)  $W < W_c$ , the sphere progressively stops, its velocity scaling as  $t^{-n}$  at large times, like in the initial simple approach; in that case the restructuring of the fluid is the dominant process;
- (ii)  $W > W_c$ , the sphere evolves towards a steady motion at a velocity larger than a finite, critical value, i.e.,  $V_c = W_c / (1 + \lambda_c^n)$ ; in that case the destructuring due to flow balances the restructuring.

The variations of the velocity as a function of the dimensionless distance covered ( $X = x/H$ ) for different values of  $W$  of a given system are represented in Fig. 4. The bifurcation above described clearly appears. Conversely if we fix the value of  $W$  but start with a material under different (increasing) initial values of the state  $\lambda_0$ , all happens qualitatively as if we were decreasing the value of  $W$ , and effectively the corresponding curves (cf. Fig. 5) show a similar bifurcation. Note that we choose two different representations in these two figures: In Fig. 4 we used a linear scale for the distance in which we looked at the aspect of the bifurcation for small variations of  $W$  at this particular scale; in Fig. 5 we used a logarithmic scale which makes it possible to observe the complete bifurcation from the first instants (where the fluid viscosity, and thus the velocity, is solely a function of  $\lambda_0$ ) to long times.

What happens now if we take into account the time variations of the state of the undisturbed material in the cylinder, i.e., we no longer neglect the term  $t/\theta$  in (12)? The key parameter is the dimensionless fluid length  $d^* = d/V\theta$ , where  $d$  is the length of observation of sphere fall through the fluid. If initially we are in case (ii) and  $d^*$  is small, we

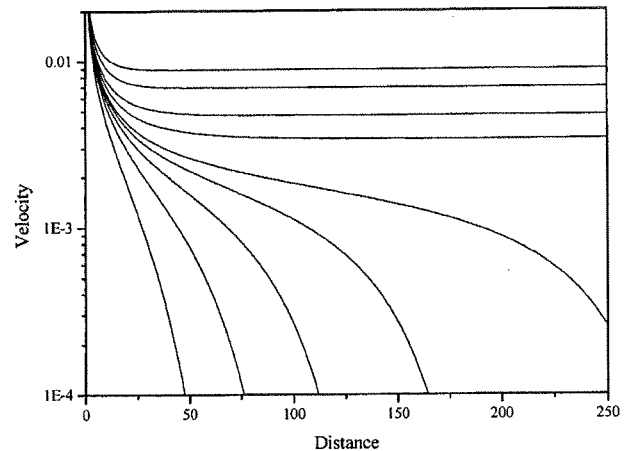


FIG. 4. Predictions of the model (see text) for the motion of a sphere through a thixotropic paste under the action of a force of different levels (from top to bottom:  $W = 0.60; 0.59; 0.58; 0.575; 0.57; 0.568; 0.565; 0.56; 0.55$ ), with the following parameters:  $\beta = 1$ ,  $n = 1.02$ , and  $\lambda_0 = 10$ .

remain in this case and might simply observe a very slight decrease of the velocity in time. For intermediate  $d^*$  we may observe some significant decrease of the velocity in time and for large  $d^*$  we reach the case (i) before the end of the fall within our experimental conditions and finally the sphere will stop moving. If we are initially in case (i) the restructuring of the undisturbed material will simply foster the sphere stoppage. Finally the complete model predicts three regimes of motion: A rapid fall at an almost constant velocity, a slower fall at a decreasing velocity and a rapid velocity decrease until complete stoppage. These regimes are in agreement with our experimental observations.

### C. Fall of an object in a vibrated thixotropic fluid

Let us now consider the same problem but with the fluid now submitted to vibrations at a frequency  $N$  and with an amplitude  $x_0$ . The exact solution of the flow problem is again rather complex but we will assume that the sphere falls through the fluid now undergoing a new pressure distri-

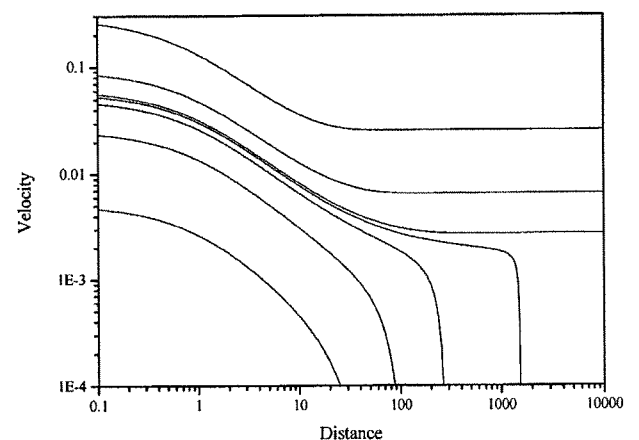


FIG. 5. Predictions of the model for the motion of a sphere through a thixotropic paste in different initial states (from top to bottom:  $\lambda_0 = 1; 5; 8; 8.5; 10; 20; 100$ ) under the action of a constant force, with the following parameters:  $\beta = 1$ ,  $n = 1.02$ , and  $W = 0.57$ .

bution resulting solely from vibrations. As long as the fluid remains static (motion "in mass") in the vibrated vessel the motion equations show that the pressure field for the vibrated fluid is simply equal to the hydrostatic distribution ( $p_a + \rho g x$ , where  $p_a$  is the atmospheric pressure) for the fluid at rest plus a term due to vibrations:

$$\rho(2\pi N)^2 x_0 x \sin(2\pi N t). \quad (14)$$

The motion of the sphere in this fluid is now simply dictated by Eq. (8) in which the liquid density must be replaced by an apparent density  $\rho^*$ , defined as

$$\rho^* = \rho(1 + \Omega \sin(2\pi N t)) \quad (15)$$

in which  $\Omega = (2\pi N)^2 x_0 / g$ . Under these conditions the vibrations simply tend to periodically decrease and increase the apparent liquid density, so that the new parameter  $W^* = W(\rho^*)$  will oscillate around its mean value  $W$  resulting from the gravity alone.

The consequence for a Newtonian fluid is that the fall velocity should oscillate around an average value corresponding to the velocity in the absence of vibration. Thus if  $N^{-1}$  is much larger than the time scale at which the velocity is measured one should record a constant velocity equal to that obtained in the fluid at rest. This is in agreement with our observations with the Newtonian liquid.

Let us now examine the consequences of these apparent density fluctuations for a thixotropic fluid. We will only consider one situation, that for which the characteristic duration of the fall is much larger than  $N^{-1}$ , so that the regime of fall is the result of a great number of oscillations. We also assume that the initial state is such that the sphere apparently set in the fluid very rapidly, i.e., after being put in the fluid it slightly moved then stopped. With the oscillations imposed to the fluid the parameter  $W^*$  will alternatively take values smaller than  $W$  for which the sphere has no particular reason to move and values larger than  $W$  for which it might move. Under these conditions we deduce that the influence of the vibrations is rather simple and may lead to different regimes, which directly derive from those found for the fluid at rest in different initial states:

- (1) For a sufficiently small value of  $\Omega$ , the large values of  $W^*$  remain insufficient for the sphere to move; it remains at the same position;
- (2) for a larger value of  $\Omega$ , the large values of  $W^*$  are sufficient for the sphere to move significantly but insufficient to overcome the subsequent restructuring of the fluid so that the sphere will appear to stop at a certain distance, similarly to regime (i);
- (3) for a sufficiently large value of  $\Omega$ , the large values of  $W^*$  are sufficient for the sphere to reach an almost constant, finite velocity as in regime (ii).

For the same reasons as above there might also exist some intermediate regime between (2) and (3) as between (i) and (ii) for which the sphere will move at a finite but significantly decreasing velocity over the distance of observation.

Thus we again find a scheme in qualitative agreement with our experimental observations.

#### IV. CONCLUSION

We have shown that the fall of an object in a typical, thixotropic, pasty material is more complex than usually assumed. Depending on the initial state of the material and the apparent weight to drag force ratio the object may fall under different regimes. In any case the fall is time-dependent and in some cases the object can even stop moving far from the vessel bottom after a significant motion. A simple model expressing the basic concepts of the rheological behavior of these pasty materials makes it possible to explain all the experimental trends in a static fluid in different states or in given fluid submitted to vibrations at different frequencies.

Since it has been shown<sup>15,24</sup> that pasty materials have generally a thixotropic behavior which may be qualitatively well represented by a model of type (6) and (7) our results concerning the fall velocity may be extrapolated to other pasty materials. Since for most of them the major restructuring process is rapid it is likely that mainly the two regimes (i) and (ii) may be observed in practice. This seems in agreement with existing literature data which mainly show significant velocity levels or no motion at all. This also seems in agreement with our common experience of the fall of objects in pastes: Indeed we can hardly imagine a slow and continuous motion of an object submitted to gravity through a mud or a foam. This finally shows that the motion of an object in a pasty material is a complex dynamical process which in particular, unlike a simple yield stress, governs its incipient motion or stoppage conditions.

A question naturally arises from this: what is the sense of the determination of the yield stress from usual observations of incipient motion in literature? Since in general it is considered that the object moves when its terminal velocity (close to the vessel bottom) is finite and reproducible it is likely that this corresponds to regime (ii). It thus effectively provides some information concerning the apparent viscosity for an almost steady, relative solid-fluid motion, but at relatively large velocities.

It nevertheless appears that the sphere fall through a paste can become a much richer test than considered until now, under the condition that the initial state of the material be well controlled and the sphere motion followed in detail: It will provide information concerning the thixotropic characteristics of the fluid. In this context it is worth noting that the vibration of the fluid is equivalent to increasing the apparent weight of the sphere so that this is a means to precisely and easily vary the apparent sphere density. It remains that the quantitative correspondence between the fluid characteristics thus determined and its exact rheological behavior is a difficult task which remains to be done.

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