Abstract

We measure the drag force on a sphere moving slowly through a 6% by weight suspension of bentonite clay particles in water. The steady state drag force depends only weakly of the speed of the sphere, indicating that yield stress effects dominate the drag in the range of pulling speeds studied. The drag force increases as a function of time as the suspension gels. The yield stress of the suspension as a function of suspension age is determined from the zero-velocity intercept of our force-pulling speed data. Both the initial transient build-up of the force and its relaxation when the motion stops are described by two exponential time constants, each of which shows an inverse power law dependence on pulling speed. We also determine the yield stress from the force remaining after the relaxation; it is significantly smaller than that determined from the steady-state force data due to the disruption of the structure of the suspension by the motion of the sphere.

Keywords: Rheological aging; Yield stress; Thixotropic gel; Bingham number

1. Introduction

Bentonite is a smectite clay mineral with many applications that take advantage of its interesting rheological properties in water-based colloidal suspensions [1]. The clay particles are platelets, typically nanometers in thickness and a few microns in diameter. They are positively charged at the edges and negatively charged on their faces. When they are suspended in water at concentrations of a few percent, electrostatic and van der Waals interactions among the particles lead to the formation of a thixotropic gel. This behavior makes bentonite an important component of materials such as drilling muds, paper coatings, and pharmaceutical products. Clay suspensions and related materials are also important in many other contexts, including mud and debris flows [2] and the flow of mine tailings. It is thus of considerable interest to understand the behavior of bentonite suspensions, and from a fundamental point of view to relate their bulk properties to structure and dynamics on the scale of the individual particles.

Bentonite suspensions are pseudoplastic, that is, when sufficiently concentrated they have a yield stress \( \sigma_y \) which results from the colloidal interactions among the particles. Below the yield stress a suspension behaves as a soft elastic solid, while above \( \sigma_y \) it flows with a shear-thinning behavior. The simplest model for this behavior is the Bingham model [3],

\[
\sigma = \sigma_y + K\gamma.
\]

Here \( \sigma \) is the shear stress, \( \gamma \) the shear rate, and \( K \) the consistency or Bingham viscosity. The Bingham number \( Bi = \sigma_y d/K \nu \), where \( d \) is the diameter of the sphere, gives the relative importance of the two terms in Eq. 1. Since the material does not flow if the local stress is less than \( \sigma_y \), a moving object will be surrounded by a fluidized region in which \( \sigma > \sigma_y \) and the material remains unsheared and solid-like [4].

The motion of a sphere through a yield stress fluid has been studied both experimentally and theoretically by several groups [4–18]. The slow motion of a sphere through a Bingham fluid was studied numerically by Beris et al., who determined the size and shape of the sheared region around the sphere [4]. Further numerical studies taking into account factors such as finite container size and wall slip have been reported in Refs. [5–7].

Early experimental studies of drag in yield-stress fluids include Refs. [8,9]. Atapattu et al. studied the creeping motion of a sphere through Carbopol, a polymer dispersion with a yield stress [10,11]. They studied the effect of nearby walls on the ter-
minal velocity of a falling sphere [10] and used particle imaging to study the size and shape of the sheared region and the depen-
dence of the drag coefficient on velocity and fluid parameters
[11]. Briscoe et al. studied the drag coefficient of spheres falling
at their terminal velocity through bentonite clay dispersions [13],
and found good agreement with the numerical results of Bera et
al. [4]. Jossic and Magnin studied the drag force on objects mov-
ing slowly through carbopol solutions, with particular emphasis
on the force required to overcome the yield stress at zero veloc-
ity. They also considered the effects of surface roughness and
wall slip [14]. Fertor et al. focussed specifically on the effects
of thixotropy on the motion of a sphere through suspensions of
laponite [18], and developed a theoretical model for these ef-
fects.

The study of bentonite suspensions is complicated by their
thixotropy, which involves a time-dependence – or more cor-
crctly a dependence on the shear history of the material – of the
rheological properties. This behavior is not encompassed by the
simple Bingham relation given in Eq. (1), and various attempts
have been made to model thixotropy [18–20]. In addition, the
properties of our bentonite suspensions change over long times
as the material ages, presumably due to slow chemical reactions
or dissolution of the clay [21]. Measurements of the rheological
properties of bentonite have been reported in a number of publi-
cations [19,22–28]; Ref. [1] is a review. Alderman et al. studied
the yield stress of bentonite as a function of concentration and
noted that σy increased with suspension age [23]. Cussot et
al. studied the rheology of a bentonite suspension and analyzed
their results in the context of a theoretical model for yield stress
behavior [19]. Laponite, a synthetic clay which, like bentonite,
forms a thixotropic gel when suspended in water, has been stud-
ied using a variety of scattering and rheometric techniques [29–
36], as have suspensions of other clays with similar properties
[2,37].

In this paper, we report on experiments on the drag force
on a sphere moving slowly through a 6% by mass bentonite
suspension. From the transient changes in force that occur when
the motion of the sphere starts and stops, we get information
about the processes involved in the break-up and reformation
of local structure in the material. The steady-state drag force
provides an estimate of the yield stress and Bingham viscosity.
Since our measurements are done at very low velocities, yield-
stress effects are much larger than viscous contributions to the
drag. We also study the changes in behavior over time as the
suspension gels.

2. Experiment

We measured the force on a steel sphere while and after it
moved at constant speed through a suspension of bentonite clay
in water. The experimental apparatus is similar to that described
in Ref. [38]. A 0.0254 m steel sphere was attached to a calibrated
load cell by a length of monofilament nylon thread [39]. The load
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2. Experiment

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The clay mixture was contained in a PVC cylinder 0.152 m in diameter and 0.605 m tall, closed at the bottom but open at the top. The cylinder was used without any surface treatment, but is large enough that wall effects are not expected to be important [4,10,11]; this issue is discussed further below. The sphere was sequentially raised and lowered through the mixture at a given velocity, with v varied between 7.6 × 10−3 m/s and 1.02 × 10−2 m/s. A typical run involved 15 cycles of raising and lowering. The length of time the sphere was in motion varied with the velocity to keep the distance travelled in the range of 0.07–0.1 m. Between periods of raising and lowering the sphere was held at rest for 30 s. When the sphere was in motion the load cell voltage was recorded approximately three times per second. Once the motion was stopped it was recorded every 1.5 s.

The suspension used in the experiments consisted of 6% by weight (2.4% by volume) laboratory grade bentonite (Fisher Scientific B235-500) in deionized water. The maximum particle diameter was approximately 4 μm as determined by the sedimentation method [28]. The clay was slowly added to deionized water and mixed for 45 min with an Arrow 1750 mixer. After the initial mixing, the suspension thickened significantly over a time scale of order 20 min as a gel formed. Over the longer term (that is, over several months), the properties of the suspension continued to change slowly, as discussed in the Appendix A. The results presented here were obtained with a suspension that had been prepared 9 months prior to the experiments, then kept in a sealed container. Before each trial, the suspension was remixed for 20 min with the same mixer in an effort to ensure consistent initial conditions. The sphere was positioned on the axis of the cylinder, roughly 0.1 m (four sphere diameters) from the bottom. The bentonite suspension was poured into the cylinder to a depth sufficient to eliminate any surface effects at the high point of the sphere’s motion, and the run commenced essentially immediately.

3. Results

Fig. 2 shows the measured force on the sphere as a function of time for one complete cycle of a typical run. Initially the sphere is at rest. When it starts moving upwards the force increases due to the drag on the sphere, but it takes a few seconds for the drag force to reach a steady state value, F_s. When the sphere stops moving, the force on the sphere decays, again over a few seconds, but to a value higher than its initial value. In the second half of the cycle the sphere is lowered and the force decreases, since the direction of the drag force is now upwards, but otherwise the behavior is the same as in the first half of the cycle. In what follows we will not differentiate between data from the raising and lowering parts of the cycle. After lowering, the material is again allowed to relax for 30 s before the cycle is repeated.

Fig. 2 shows the force as a function of time for a full 15-cycle run. The magnitude of the steady state drag force, F_s, increases over the course of a run as the material properties change. From fits to the individual raising and lowering segments, as described
Here, we obtain values for $F_s(T)$, where $T$ is the age of the material measured from the start of the experiment, taken as the midpoint time of each segment. $F_s(T)$ can be described by a simple exponential approach to a constant value with a time constant $\tau_F$:

$$F_s(T) = F_s(0) e^{-T/\tau_F} + F_y,$$

(2)

A fit to this function is shown by the dashed envelope function in Fig. 2, $\tau_F$ is a measure of the time scale for changes in the material properties as it forms a gel. It shows no systematic dependence on pulling speed $v$, and has a mean value of $540 \pm 150$ s.

$F_y(0)$ and $F_y(\infty)$, the steady state drag force at zero and infinite time from the start of the run, were found as a function of $v$ in Fig. 3. The measurements at different pulling speeds were performed in random order over a 2-day period. We observed a noticeable dependence of the drag force on the order of measurement; that is, the drag force tended on average to increase from one run to the next, despite the fact that the suspension was remixed before each run, and the variations due to sample aging were at least as large as those due to variations in $v$. This long term aging is discussed further in the Appendix. In an effort to correct for this, the force data were adjusted to a common reference time by subtracting a term linear in time determined from a fit to the data. The results are presented in Fig. 3. The magnitude of the steady state drag force at $T = 0$ increases slightly with $v$ over the range studied here. Within the accuracy of our measurements the variation is linear, and a fit gives $F_y(0) = (3.8 \pm 0.3) \times 10^{-2} + (1.6 \pm 0.6) v$, with $F_y$ in N and $v$ in m/s. At infinite age the drag force is larger, but is constant as a function of $v$ within the experimental uncertainty; a fit gives $F_y(\infty) = (7.4 \pm 0.7) \times 10^{-2} + (0.2 \pm 1.5) v$, N. These results indicate that the force on the sphere is almost entirely due to the yield stress, and the viscous contributions are at most 30% in this range of $v$.

We can estimate the yield stress of this suspension from the zero-velocity intercepts of the plots in Fig. 3. Beris et al. showed numerically that, for a sphere at rest in a Bingham fluid, the force $F_y$ required to overcome the yield stress was given by [4]:

$$F_y = 4\pi r^2 \sigma_y,$$

(3)

Applying this result to our data gives a yield stress of $5.3 \pm 0.4$ Pa at $T = 0$ and $10.4 \pm 0.9$ Pa as $T \to \infty$.

Since in our work the viscous contributions to the drag force are small, the Bingham number $Bi$ is expected to be large. As a result, the same conversion from force to stress should be reasonably accurate for the moving sphere as well. We estimate the shear rate to be $\dot{\gamma} = v/d$, where the sphere diameter
\[ F = C_D \frac{\rho v^2}{2} \]  

Since \( F \) is only weakly dependent on \( v \), \( C_D \) is approximately proportional to \( \frac{\rho v^2}{2} \). A dimensionless quantity that attempts to account for both the yield-stress and viscous contributions to the drag has been used in Refs. [9] and [11]. It is defined by

\[ \frac{Q}{\tau} = \frac{R_e}{\tau_1 + 4\tau_2} = \frac{\rho v^2}{R_e \eta D + \tau_1 \eta} \]  

where \( R_e \) is a modified Reynolds number and \( k \) is a constant given in Ref. [9] as \( \tau_1 \eta/24 \). We plot \( C_D \) as a function of \( Q \) for the zero-age data in Fig. 4. \( C_D \) is proportional to \( Q^{-0.1} \) over the range of our data, which again reflects the fact that the drag in this case is dominated by the yield stress while the viscous contribution is small. These results are in excellent agreement with those of Refs. [9, 11].

The transient approach of the force to its steady state value when the sphere starts moving is well described by the sum of two exponential terms,

\[ F(t) = F_1 e^{-t/\tau_1} + F_2 e^{-t/\tau_2} + F_0, \]  

\[ \tau_1 = \frac{\tau}{2} + \frac{\tau}{2} \]  

\[ \tau_2 = \frac{\tau}{2} + \frac{\tau}{2} \]  

where \( \tau \) is the time (measured from the start of the sphere’s motion for each segment), \( \tau_1 \) and \( \tau_2 \) are time constants, \( F_1 \) and \( F_2 \) are the amplitudes of the exponential terms and \( F_0 \) is the force measured when the sphere is at rest at time zero. The steady-state drag force acting on the sphere is \( F_s = F_1 + F_2 \). Fits to this function for both the raising and lowering segments of the data are shown as dashed lines in Fig. 1. Except as described below, a function with only one exponential term is not adequate to describe the transient, and the presence of two time constants indicates that there are two distinct processes involved in the build-up of the force.

The relaxation of the drag force when the motion of the sphere stops is also well described by a sum of two exponential terms.

We fit the relaxation data to the form

\[ F(t) = R_1 e^{-t/\tau_1} + R_2 e^{-t/\tau_2} + R_0, \]  

where \( \tau_1 \) and \( \tau_2 \) are the time constants for the relaxation, \( R_1 \) and \( R_2 \) are the corresponding amplitudes, and \( R_0 \) is force as \( t \) goes to infinity. As above, the presence of two time constants indicates that two processes contribute to the relaxation. Fits of Eq. (7) to the decaying segments of the data are also shown in Fig. 1.

Within the experimental scatter, the two time constants \( \tau_{11} \) and \( \tau_{12} \) which characterize the transient build-up of the drag force remain constant over the course of a run, indicating that they are not affected by the gelation or aging of the material. At high \( v \), only the shorter time constant \( \tau_{11} \) is required to describe the transient; the term with the longer time constant goes to zero as described below. In addition, the force transient for the first raising segment in each run has only the shorter time constant, indicating that the character of the fluid when freshly poured is qualitatively different from that even a minute later: \( \tau_{11} \) and \( \tau_{12} \) are plotted as a function of \( v \) in Fig. 5. Each point in this figure is an average over the 30 upward and downward segments of a run. Both \( \tau_{11} \) and \( \tau_{12} \) show an inverse power law dependence on pulling speed (or equivalently, on shear rate) over the range studied.
The time constant \( \tau_{r0} \), the longer time constant, is approximately inversely proportional to \( v \); a fit gives \( \tau_{r0} = (5.5 \pm 2.0) \times 10^{-3} \) s (with \( v \), here and elsewhere, in m/s). We identify this as the time scale for the establishment of the sheared region around the sphere when it starts to move [38]. The flow induced by the moving sphere disrupts the local structure of the suspension out to some distance \( \delta \), and the time scale for this disruption to develop is just \( \delta/v \), the time it takes for the sphere to move \( \delta \). Our results indicate that \( \delta \approx 0.011 \) m, slightly smaller than the radius of the sphere. At the start of the first segment of each run, the suspension is already fully fluidized from the mixing and from being poured into the experimental container. There is thus no need for the sphere to establish a sheared region around itself in this case — the material is already sheared. This explains the absence of the \( \tau_{r0} \) term for this first segment. At later times, the suspension has begun to gel and this term appears.

The shorter time constant \( \tau_{r1} \) has a weaker dependence on pulling speed \( v \); a fit gives \( \tau_{r1} = (3.2 \pm 0.3) \times 10^{-3} \) s. The strength of the material depends on interactions among the suspended clay platelets, which in turn depend on local variations of the particle positions and orientations. One would expect these to adjust diffusively to changes in the local environment, and we suggest that \( \tau_{r1} \) is the time scale for the response to changes in the structure of the material in the region immediately around the sphere.

For the first raising segment of each run, \( F_2 = 0 \) since, as noted above, only the \( F_1 \) term is needed to describe the force transient. Thereafter, the relative size of the \( F_1 \) term decreases slightly with time while that of the \( F_2 \) term (due to shear) increases, indicating that more work is required to fluidize the material as its gelation progresses. Fig. 6 shows the relative sizes of \( F_1 \) and \( F_2 \) as a function of pulling speed \( v \) at a particular time. The relative magnitude of the shear term decreases as \( v \) increases, while that of the \( F_1 \) term increases. For \( v \gtrsim 5 \times 10^{-3} \) m/s, our fitting program is unable to discern the \( F_2 \) term and the transient is well described by a single exponential term with the time constant \( \tau_{r0} \). The disappearance of the term due to shear is likely because the structure of the region around the sphere is sufficiently disrupted at high \( v \) that it is not necessary to reestablish the sheared region in each cycle of the experiment.

The time constants \( \tau_{r1} \) and \( \tau_{r2} \) characterizing the relaxation of the drag force are plotted as a function of \( v \) in Fig. 7. These data were also averaged over all cycles of each run. Despite that fact that the sphere is now motionless, both depend on the previous velocity, and by inference are related to processes taking place within the sheared region created by the previous motion of the sphere. As in the case of motion, the shorter time constant \( \tau_{r1} \) behaves approximately as \( v^{-1/2} \): \( \tau_{r1} = (2.9 \pm 0.2) \times 10^{-3} \) s. As before we identify this as the time constant for relaxation of stress due to diffusive changes in the structure of the material in the sheared region around the sphere. The shorter magnitude in this instance is likely a result of the fact that the motion of the sphere through the suspension has disrupted the structure of the material, reducing the viscosity and so the response time below that at the onset of shear.

\( \tau_{r2} \), the larger relaxation time constant, also decreases with increasing velocity, but with a significantly weaker dependence on \( v \). A fit gives \( \tau_{r2} = (0.94 \pm 0.02) v^{-0.3} \) s. The associated stress relaxation process is likely related to some sort of healing of the fluidized region, although its exact nature is unclear.

Fig. 8 shows the relative contributions of the \( R_1 \) and \( R_2 \) terms to the relaxation of the drag force as a function of \( v \) at a particular time. Although the variation with (previous) pulling speed is weaker than was the case for the build-up of the force, the relative importance of the \( R_1 \) term increases slightly and that of the \( R_2 \) term decreases as \( v \) increases.

The stress remaining on the sphere after the relaxation process has finished should be simply equal to the yield stress itself, since it is the stress measured at zero shear rate [40]. The force remaining after relaxation, \( F_0 \), was obtained from fits to Eq. (7) and as before converted to a yield stress using Eq. (3). As with other quantities, \( \sigma_y \) determined in this way is a function of the time \( T \) from the beginning of the experiment, and the variation with \( T \) is well described by an exponential function similar to Eq. (3).
Our own measurements suggest that the effect is significantly smaller than this. Buoyant forces in yield-stress fluids have not been considered in much detail but if we assume that the pressure within our suspension is simply hydrostatic, then the buoyant force on the sphere will be the same for both upward and downward portions of the experiment and will not affect our measurements.

The existence of a yield stress results from an internal structure within the fluid — in the present case, due to interactions among the suspended clay particles. Shear disrupts this structure and fluidizes the material, reducing its viscosity. On the cessation of shear (and neglecting aging effects), the internal structure reforms and the fluid properties return to their original values. This restructuring takes time; this is the origin of the thixotropy of the suspension. The reformation of structure following shear is distinct from, but presumably closely related to, the gelation of the material over time, which takes place in the unsheared material.

The bentonite suspension was remixed before each run, completely disrupting any pre-existing structure in the material. As the experiment progresses, the properties of the suspension change as structure reforms and the material gels. Both the steady-state drag force and the yield stress determined after relaxation of the drag force show an exponential approach to a constant value, with a time constant of 540 ± 150 s in the first case and 500 ± 130 s in the second case. These values are the same within our uncertainties. No systematic variation of this gelation time with experimental parameters or with the age of the sample was observed, although the uncertainties are fairly large due to scatter from run to run. The drag force and yield stress increase with time, despite the fact that the motion of the sphere periodically disrupts the structure in the region around it. This suggests that the formation of structure in the sheared region around the sphere is influenced by the state of the suspension in the surrounding, undisturbed material. In scattering experiments on laponite suspensions, Kroon et al. showed that gelation involves the partial development and then decay of orientational order among the clay platelets [33,34]. They found the gelation time to decrease exponentially with the mass fraction of the clay. Extrapolating their data to a mass fraction of 6% gives a gelation time of order 100 s, a factor of five lower than observed here.

We obtained estimates of the yield stress of our suspension in two ways: first, from the zero-velocity intercept of our force versus velocity data, as in Fig. 3, and second from the force remaining on the sphere after relaxation. As noted above, the values obtained after relaxation are lower than those from the steady-state drag force. The motion of the sphere through the material disrupts the existing structure and so reduces the yield stress, and as a result the yield stress measured at the end of a cycle when the motion stops is less than at the beginning of a cycle. The implication of this is that, although the overall gelation time is of order 500 s, the local structure in the sheared region around the sphere heals significantly over the 30 s that the sphere is at rest between segments.

A number of measurements of the yield stress of bentonite suspensions have been reported [19,22–28]. Although the value of the yield stress will depend on the details of the material used and on how the measurements were done, it is nonetheless useful to compare our results to the previous work. Our result from the zero-velocity intercept of the force–velocity data is 5.3 ± 0.4 Pa immediately after remixing, and increases as the
measurements. In addition, there are two potentially significant
from analysis of the flow of a sheet of the suspension down an
rheometry or the value of 2.2 Pa for a 6% suspension obtained
50
σy
shear flows in thixotropic fluids, we would expect our values of
the state of the entire sample. While the same is true of simple
carbopol, a difference of about 40% was observed between mea-
moving (but not when it is at rest). In similar experiments using
affect our measurements of the drag force when the sphere is
ond, wall slip may be present at the surface of the sphere and
that the Bingham model describes this material adequately. Sec-
tions [4], is correct for our system, and indeed the assumption
v
the force–velocity data.
Our measurements show a linear increase in drag force as a
function of v for the freshly mixed suspension. At later times as
the suspension gels, the drag force becomes independent of v
and so is due entirely to the yield stress. Equivalently, the stress
is independent of shear rate over the range covered. Reliable
measurements of the Bingham viscosity K in the literature are
few and are based on measurements done over a much wider
range of shear rates, and the uncertainties in our values of K are
large, so a meaningful comparison with our results is difficult.
Jossic and Magnin observed that the drag force they measured
in carbopol was independent of speed for low speeds [14], and
a velocity-independent drag force has also been observed in a
dry granular medium [41].
When the sphere starts to move in the suspension, one would
expect the medium to respond elastically (i.e., linearly) until a
yield strain is exceeded and the material fluidizes. In fact, this
elastic regime is not particularly evident in our force–time data,
probably because we do not have sufficient time resolution at
the beginning of each cycle to distinguish it. Both the transient
development of the stress at the onset of motion and its relax-
when the motion is stopped are nonexponential in time and
can be fitted by functions with two exponential terms. This indi-
cates that two distinct processes are involved in the build up and
relaxation of stress in this suspension. All four time constants
— two for build-up and two for relaxation — show an inverse
power law dependence on v (or, equivalently, on shear rate).
In that

\[ \tau_{22} \] is roughly inversely proportional to shear rate. In simi-
lar experiments on aqueous foam [38], the build-up of the drag
force was characterized by a single time constant which was also
inversely proportional to \( \dot{\gamma} \). As in that work, we associate this
time scale with the time required for the sheared region around
the sphere to develop as the sphere moves. The shorter of the
two time constants for motion and relaxation, \( \tau_{22} \) and \( \tau_1 \), re-
spectively, are both approximately proportional to \( 1/\sqrt{v} \) and
have about the same magnitude, so it is reasonable to infer that
they are due to the same process, and that this process is sim-
ilar whether the sphere is moving or not. The gelation process
in clay suspensions involves positional and orientational diffu-
sion of the clay platelets [33], and we suggest that these time
scales are associated with a diffusive response to the local dis-
ruption — in the case of the build up of the drag force — or
the reforming — in the case of relaxation — of orientational or
positional organization in the material. Since as shown in Fig.
3 the stress is roughly constant as a function of shear rate, the
viscosity of the suspension is inversely proportional to v,
and so the diffusion constant \( D \propto v \). The time for diffusion over a
distance \( \lambda \) would be \( \lambda^2/Dv \) and thus inversely proportional to v.
However, one could imagine that the distance scale \( \lambda \) could in-
crease weakly with v, since a faster-moving object would cause a
more serious disruption of the local structure. In this case, the
\( v \) dependence of the diffusive time scale would be weaker than
linear, as observed. The longer time scale for the relaxation of
the stress, \( \tau_2 \), is proportional to \( v^{-0.3} \). The same \( v \) dependence
was observed in one component of the relaxation of the drag
force in similar experiments on foam [38], in which case the re-
 laxation was related to local restructuring of the foam following
the cessation of shear. Here again this process must be related
to the redevelopment of structure and relaxation of stress in the
sheared region, although further study is required to determine
its exact nature. These time constants are all significantly shorter
than the relaxation and restructuring times, as defined in the the-
oretical model of Coussot et al. found for a bentonite suspension
in Ref. [19]. We note also that the creep compliance of bentonite
suspensions has also been found to involve two time constants
[25].

Rheological aging in so-called soft glassy materials has re-
cently been studied both theoretically [42,43] and experimen-
tally [44–47]. In this phenomenon, the structure and properties
of the material change slowly with time but, since the relaxation
time itself increases with sample age, the system never reaches
a steady state. Our experiment is not particularly well suited to
the study of rheological aging and we note that our measured
drag force does appear to approach a steady state value at long
times. Harden et al. have studied aging in suspensions of laponite
clay using a combination of scattering techniques and observed a
complex aging behavior [44–46]. At concentrations high enough
that the laponite formed a gel, they observed a fast relaxation of
the scattering correlation function which they attribute to diffu-
sion, and a much slower process which they attribute to processes
that relax stress locally [46]. These relaxation processes may be
related to those seen here.

It is interesting to compare the results presented here to those
obtained in similar experiments on an aqueous foam [38]. In that
case, the transient build-up of the drag force was well described by a single exponential term, with a time constant inversely proportional to shear rate. This time constant is apparently analogous to \( \tau_{\text{m2}} \) in the present case, but \( \tau_{\text{m1}} \), which here we attribute to a time scale for adjustments in the position or orientation of the clay platelets, is not present in the foam case. This is reasonable, since there is no analogous process in the foam. Three time constants were required to adequately describe the relaxation of the drag force in foam, compared to two in the present case. The shortest of these was of order one second and decreased weakly with the previous shear rate as \( \gamma^{-0.3} \). As discussed above, this is the same shear rate dependence as shown by \( \tau_{\text{2}} \) in the present work and is likely related to a healing of the material structure in the previously sheared region surrounding the sphere. The diffusive time scale is again absent from the relaxation in the foam, with the two longer time scales in that case being roughly independent of shear rate and of order 10 and a few hundred seconds. These times were associated with the time between local bubble rearrangements and coarsening of the bubble size distribution, respectively, both processes that do not occur in the clay suspension.

5. Conclusions

We have studied the drag force on a sphere moving through a 6% by mass bentonite suspension. The build-up and relaxation of the force are nonexponential, indicating that two distinct processes contribute to the disruption and reforming of structure within the sheared region surrounding the sphere. The steady state drag force increases weakly with the speed of the sphere for the freshly remixed, and not at all for a suspension which had time to gel. This indicates that the drag force in this material is almost entirely due to the yield stress. The yield stress increases with time as the suspension forms a gel, with a gelation time of roughly 520 s.

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Appendix A. Suspension aging

The bentonite suspension studied shows long-term aging effects in addition to the shorter-term time dependence due to gelation described above. This slower aging is likely due to slow chemical changes in the material as a result of its exposure to air [21], and is also distinct from the rheological aging mentioned above [42-43]. The effect of this (chemical) aging is illustrated in Fig. A.1, which shows \( F_v(\infty) \), the infinite-time steady-state drag force, as a function of the age of the sample for a pulling speed of \( v = 5.0 \times 10^{-3} \text{ m/s} \). The data were obtained from two different suspensions that were prepared identically. The point at age zero was obtained immediately after the suspension was prepared. All other data were obtained from a second suspension that had been prepared and then covered for approximately 6 months before any measurements were taken. Fig. A.1 shows a steady increase in the drag force as the material ages over a period of a few months, followed by a more sudden increase at an age of approximately 265 days. This increase corresponds to a period of more frequent experimentation over a 1-week period. The repeated mixing and pouring of the suspension during this time exposed it to the air much more than when it was sitting in a covered container and accelerated the chemical changes occurring. To minimize the effect of this chemical aging on our results, the data reported in this paper were collected over a 2 day period at an age of 267 days.

References


