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Clouds of particles in a periodic shear flow

Bloen Metzger¹ and Jason E. Butler²

¹IUSTI-CNRS UMR 6595, Polytech Marseille, 13453 Marseille Cedex 13, France ²Department of Chemical Engineering, University of Florida, Gainesville, Florida 32611, USA

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We have investigated the time evolution of a cloud of non-Brownian particles subjected to a periodic shear flow in an otherwise pure liquid at low Reynolds number. This experiment illustrates the irreversible nature of particulate systems submitted to a shear. When repeating the cycles of shear, we have found that clouds of particles progressively disperse in the flow direction until reaching a threshold critical volume fraction that depends upon the strain amplitude; this critical volume fraction coincides with measurements of the threshold for reversibility found from experiments on homogeneous suspensions in periodic shear. Two distinct patterns, including a "galaxy-like" shape, are observed for the evolution of the clouds and the transition between the patterns is identified using a simple scaling analysis. Movies are available with the online version of the paper. © 2012 American Institute of Physics. [http://dx.doi.org/10.1063/1.3685537]

Half a century ago, Taylor proposed an experiment¹ which elegantly illustrates the reversibility of viscous fluid flows. A small blob of dye is inserted into a very viscous fluid contained in the gap between two concentric cylinders. The blob is first stretched into a long thin filament by rotating the inner cylinder several turns. Then upon reversing the rotation for the same number of turns, the drop astoundingly recovers its initial shape, except for some minor effects of molecular diffusion.

Inspired by Taylor's work, we perform a similar experiment on finite-sized clouds of non-Brownian particles. The key idea is to perform successive cycles of shear and measure the evolution of the shape of the clouds at the end of each cycle. This experiment is particularly advantageous for investigating the transition between reversible and irreversible dynamics in suspensions. In this stroboscopic measurement, the affine part of the particle motion, i.e., the advection motion, is eliminated. Hence, all departures from the initial cloud shape can be attributed to irreversible events caused by the interaction between particles.

The experiment also provides important clues regarding the nature of the interactions between particles that are very near to each other. The minimum separation distance between two approaching particles can be less than 10^{-4} of their radius,^{2,3} though the question of whether particles make contact is still a matter of debate. Clarifying this issue would substantially enhance our understanding of suspension behavior.

For instance, homogeneous suspensions in a periodic shear undergo a remarkable transition⁴ as the concentration and strain amplitude are varied. At each concentration, the suspension either transitions to a state where the particle positions are reversible or remain irreversible, if the strain amplitudes are below or above a critical value, respectively. Although, the phenomenology of this transition was captured by a numerical simulation within which particles that pass close to each other are given a small random displacement⁵ and the microstructures of the reversible states can be predicted from more detailed simulations,⁶ the physical origin of the irreversibility remains unclear.

Understanding the nature of any non-hydrodynamic particle interactions is also critical for modeling the rheology of suspensions and phenomena such as particle migration. For the latter case, a "revisited suspension balance model"⁷ was applied to capture the migration process of a suspension submitted to a rotating rod; the work suggests that contact stress governs the



FIG. 1. (Color online) Sketch of the experimental shear-cell. The acrylic transparent walled vessel has inner dimensions $2 \times 7 \times 50$ cm³ and was filled with fluid to a height of 5 cm to perform the experiments.

migration behavior.⁸ However, other models assert that the stresslet contribution arising from the non-symmetric microstructure⁹ is responsible, at least in part, for the migration.

The present work thus aims toward the challenging goal of getting a better grasp on how close particles interact in sheared suspensions. We have found that clouds of particles having an initially moderate volume fraction ($\phi = 20\%$ –40%) progressively disperse in the flow direction until they reach a threshold critical volume fraction. For larger volume fractions, clouds can exhibit a different pattern of dispersion. We found that clouds initially at the maximum packing fraction can evolve into a "galaxy-like" shape.

The experimental set-up, inspired by Rampall *et al.*,¹⁰ is sketched in Figure 1. A precision rotating stage (M-061.PD from PI piezo-nano positioning) with high angular resolution $(3 \times 10^{-5} \text{ rad})$ drove a transparent belt to create the shearing flow. The fluid was a Newtonian mixture of Triton X-100 (76.241 wt. %), zinc chloride (14.697 wt. %), and water (9.062 wt. %) having a viscosity of 3 Pa and a density of 1.18 g/cm³ at room temperature. The composition was chosen to match both the refractive index and density of the acrylic particles. Carefully matching the fluid and particle densities was critical since the sedimentation rate of a cloud of particles greatly exceeds that of a single particle.^{11,12} We achieved a density match that ensured that the cloud's settling velocity was lower than 10^{-7} m/s. A small amount of hydrochloric acid (≈ 0.05 wt. %) was added to the solution to prevent the formation of zinc hypochlorite precipitate, thereby significantly improving the optical transparency of the solution.

We used two different batches of spherical acrylic particles of density 1.18 g/cm³ having diameters of $200 \pm 50 \ \mu\text{m}$ and $450 \pm 50 \ \mu\text{m}$. Suspensions of volume fractions $\phi = 30\%$ and 40% were first prepared in a small container and then were transferred into the shear cell using a pipette of inner diameter 3 mm. Clouds were injected into the cell two centimeters below the free surface of the fluid as shown in Figure 1. Clouds at maximum packing were prepared by partially filling the pipette with pure fluid so that about 3 mm of its tip remained free of liquid. Then, dry particles were directly poured into that remaining space and the fluid in the pipette was slowly pushed through the tightly packed bed of particles.

Before preparing the suspensions, the particles were dyed with rhodamine 6G by letting them soak for 40 min at 60° C in a mixture of 50 wt. % water, 50 wt. % ethanol, and a trace amount of rhodamine. This dye fluoresces under illumination provided by a green laser diode. Such a laser



FIG. 2. (Color online) Evolution of clouds composed of particles with $d = 450 \ \mu \text{m}$ at an initial volume fraction 30%. Images were taken (a) initially and (b) after 40 cycles at a strain amplitude of $\gamma_0 = 6$. (c) Evolution of the cloud extension, $E(N) = e_N/e_0$, in the flow direction (enhanced online) [URL: http://dx.doi.org/10.1063/1.3685537.1].

(100 mW power and wavelength of 532 nm) with a combination of plano-cylindrical lenses were used to generate a horizontal laser sheet of thickness \approx 30 μ m. The fluorescing particles within the plane of the laser sheet were imaged from the top using a high resolution digital camera (Nikon 300s) and a high quality magnification lens (Sigma APO-Macro-180 mm-F3.5-DG). A 550 nm longpass colored glass filter was positioned in front of the lens to remove any light scattered by the particle surfaces.

The cloud was sheared forwards then backwards in a repeating cycle. An image of the cloud was recorded at the end of each cycle. The total accumulated strain experienced by the cloud over a run is thus $\gamma = 2\gamma_0 N$, where γ_0 is the strain amplitude and N is the number of cycles. One strain unit corresponds to a relative displacement of the cell walls equal to their separation distance.

When sheared, clouds composed of the larger particles ($d = 450\mu$ m) stretch along the flow as expected (cf. Video 1). Upon reversing the shear flow, the clouds do not reconstitute identically at the end of each cycle of shear, but instead progressively expand in the flow direction as seen in Figures 2(a) and 2(b). The dynamics of the cloud extension is shown in Figure 2(c) in terms of $E = e_N/e_0$, the cloud extension in the flow direction normalized by the initial size. The cloud initially extends rapidly and eventually reaches a steady state after a number of cycles that increases as the strain amplitude increases. After reaching steady state, the clouds become reversible and reform identically (within measurement capability) after each cycle.

The evolution of the cloud extension is similar to the self-organization into a reversible state observed from the periodic shear of homogeneous suspensions when the strain amplitude is small.^{4,13} Here the volume fraction of the clouds is free to vary. Consequently, as the cloud disperses into the surrounding fluid, the volume fraction decreases until diluting to the threshold critical value for reversibility at that strain amplitude. Figure 3 shows that this principle holds over volume fractions of 30% and 40% and strain amplitudes of $\gamma_0 = 2$ to 6. The initial volume fraction of the clouds is represented by thin symbols on the phase diagram. The volume fraction of the cloud in its reversible state was calculated using $\phi_r = e_0\phi_i/e_r$, where e_0 and e_r denote the cloud dimension in the flow direction in its initial state and in its reversible state, respectively. This estimate of the concentration is reasonable since the dispersion occurs mainly in the plane of shear, as verified experimentally. Similarly, as one can observe from Figure 2, dispersion in the gradient direction is small relative to the large dispersion in the flow direction.

We found that dispersion stops once the volume fraction decreases below a critical value that matches the threshold for reversibility as measured from studies on homogeneous suspensions⁴ as seen in Figure 3. The grey arrow in this figure indicates the evolution of a cloud initially prepared at $\phi = 30\%$ and periodically sheared with a strain amplitude of $\gamma_o = 4.5$. This cloud progressively disperses until reaching the reversible region of the diagram. Clouds follow that evolution independently of their initial volume fraction, particle size, and applied strain.

Clouds composed of the smaller diameter spheres are not represented in the diagram when $\phi = 40\%$ and for strains larger than 3. Clouds above this limit, represented in Figure 3 by the



FIG. 3. (Color online) Phase diagram (γ_0 - ϕ) for clouds initially prepared with volume fractions of 30% and 40%. Initial positions on the phase diagram are indicated by thin symbols and the corresponding positions of the reversible state are indicated with thick symbols. The grey arrow indicates the evolution of a cloud initially prepared at $\phi = 30\%$ and periodically sheared with a strain amplitude of $\gamma_o = 4.5$. This cloud progressively disperses until reaching the reversible region of the diagram. The critical curve, $\gamma_0 = C\phi^{-\alpha}$ (C = 0.14, $\alpha = -1.93$), is from experiments on homogeneous suspensions.⁴

thick dashed line, demonstrate a different mode of dispersion as pictured in Figure 4. These clouds produce a "galaxy-like" shape during large amplitude oscillations, wherein two thin tails of particles emanate from both sides of the cloud forming a slight angle with the direction of flow (cf. Video 2). During shear, the core of the cloud at maximum packing fraction rotates as a solid body (cf. Video in the supplementary material (Ref. 14)) and particles located at the cloud periphery shed into the surrounding fluid to form this pattern.

A simple model qualitatively reproduces the galaxy shape and provides insight into why particles do not return to their initial positions when shear is reversed. The core of the cloud is modeled as one large solid particle of radius a in a straining flow **E** generating a flow disturbance **u** at the position r,¹⁵

$$\mathbf{u}(\mathbf{r}) = \mathbf{r} \cdot \mathbf{E} \cdot \left(\mathbf{rr} \left(-\frac{5a^3}{2r^5} + \frac{5a^5}{r^7} \right) - \mathbf{I} \frac{a^5}{r^5} \right).$$
(1)

This expression corresponds to the leading order term (stresslet) of the flow disturbance generated by a rigid spherical particle embedded in a shear flow. Particles located at the cloud periphery are represented by passive tracers that are initially distributed randomly in a thin layer around the



FIG. 4. (Color online) Comparison between an experiment and model for the evolution of a cloud composed of the small particles, initially prepared at the maximum packing fraction, and oscillated at a strain amplitude of $\gamma_0 = 6$. Images (a₁) and (b₁) show the initial conditions for the experiment and simulation, respectively. For the simulation, passive tracers are initially distributed in a thin layer around the core, which is progressively reduced in size. The distribution predicted by the simulation after 20 cycles appears in (b₂) and is compared to the distribution from the experiment after 20 cycles as shown in (a₂). (c) Schematic for the pore pressure feedback mechanism (enhanced online) [URL: http://dx.doi.org/10.1063/1.3685537.2]; [URL: http://dx.doi.org/10.1063/1.3685537.3].

core and that follow the fluid motion, which includes the imposed shearing flow and disturbance flow.

If one keeps the core size constant (*a* fixed) throughout each cycle of the periodic shear, the tracers return to their initial positions owing to the reversibility of Stokes equations. However, the core size reduces during shear owing to the loss of particles at the edge of the cloud as represented in part by the tracers. Hence, the size *a* is reduced during the forward motion of each cycle, when the shear rate is positive. As a result, the flow disturbance produced by the core is not the same during the backward as forward portions of each cycle and the tracer particles do not return to their original positions. The progressive shrinking of the core size over multiple cycles of shear opens more and more streamlines that allow particles to escape from the cloud. Such clouds are thus intrinsically unstable. A pattern similar to the one seen in the experiment develops from the simulation as seen in Figure 4(b) (cf. Video 3). In this simulation, the core size was reduced at every half cycle such that the total loss after 20 cycles was 30%.

The experimental observations indicate that the transition between the stretching and the rotating modes depends both on the cloud volume fraction and the particle size. The onset of the rotating mode relies on the appearance of a "pore-pressure feedback" effect.¹⁶ The schematic in Figure 4(c) illustrates this mechanism. Densely packed granular material is known to dilate under an imposed shear.¹⁷ Thus, when shearing a tight bed of particles from configuration A to B (see Figure 4(c)), the pores in between the particles become larger. This induces a motion of fluid (represented by the arrows) from outside of the densely packed region toward the center of the cloud. This fluid motion creates a pore-pressure gradient that tends to press the grains together, enhancing friction.

This so-called pore pressure can be estimated and balancing the shear stress with this pore pressure effect provides a simple and accurate prediction of the transition point for the behavior of the cloud. If the viscous stress generated by the surrounding shear flow on the cloud ($\eta \dot{\gamma}$, where η is the fluid viscosity and $\dot{\gamma}$ is the shear rate) is larger than the stress created by the pore-pressure ($\mu_s P^p$, with μ_s the friction coefficient and P^p the pore pressure), then the cloud should stretch; if it is smaller, the cloud should rotate. This latter pore-pressure stress is estimated using Darcy's law, $\nabla P^p \propto \eta \delta U/d^2$, where δU denotes the relative velocity between the fluid and the particle phase and *d* is the particle diameter.

To capture the dilation of the cloud network, we use the dilatancy angle ψ :¹⁶ $\delta U = \dot{\gamma} d \tan(\psi)$ (see Figure 4(c)) which relates the normal and the transverse motion of the particles. The pore pressure at the center of the cloud is thus $P^p = \eta \dot{\gamma} \tan(\psi)D/d$, where *D* is the cloud diameter. This provides a criterion for the onset of rotation: if $D/d \ge 1/(\mu_s \tan(\psi))$, the cloud should rotate. This transition depends on the volume fraction through the dilatancy angle as $\tan(\psi) = K(\phi - \phi_{eq})$ which assumes that the dilatancy angle is proportional to the difference between the actual volume fraction, ϕ , and the steady state volume fraction ϕ_{eq} . Using the parameters from the experiment of $\phi \approx 0.63$ (maximum packing) and values provided in the literature of K = 4.09, $\phi_{eq} = 0.58$,¹⁶ and $\mu_s = 0.32$,⁸ we find that the onset for the rotating mode for $D/d \approx 15$; this prediction of the transition point for the two observed patterns is consistent with the experimental data. Indeed we found that at the maximum packing fraction, clouds composed of the smaller particles, where $D/d \approx 20$, do rotate, whereas clouds composed of the larger particles, where $D/d \approx 11$, stretch in the shear flow without rotating.

We have investigated the time evolution of a cloud of non-Brownian particles submitted to a periodic shear flow in an otherwise clarified, Newtonian fluid at low Reynolds number. Clouds at initially moderate concentrations ($\phi = 20\%-40\%$) progressively disperse until diluting to a threshold critical volume fraction. These dynamics and the existence of a threshold concentration, below which particles cease to fluctuate between cycles, suggest that the irreversible behavior arises from particle-particle collisions. Above the threshold, collisions between the particles create the irreversible motion; below the threshold, particles solely interact through hydrodynamic interactions and their motion is reversible. This result corroborates previous observations from numerical simulations that the long range hydrodynamic interactions are not a source of irreversibility.¹⁸ Stronger evidence of the importance of solid contacts would be to show that the irreversibility is dramatically reduced when the particle roughness is reduced. To tackle this issue, the same experiment will be performed with a stable emulsion at small capillary number.

Clouds initially prepared at the maximum packing fraction and composed of sufficiently small particles disperse into the surrounding fluid by producing a symmetric pattern that is reminiscent of the shape of a galaxy; this pattern differs from that observed for clouds made of larger particles. A simple model indicates that the formation of the galaxy pattern is directly related to the solid rotation of the core of the cloud. We also provided a dimensional analysis which suggests that the onset of the rotation of the cloud is caused by a "pore-pressure feedback effect" when D/d > 15. We plan to more fully test this hypothesis and resulting scaling for the transition by systematically varying D/d in future experiments.

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