Concentration band dynamics in free-surface Couette flow of a suspension

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Particle concentration banding has been studied by experiments in the flow driven by the inner cylinder of a partially filled concentric cylinder, or Couette, device. In this geometry, alternating bands of relatively concentrated and dilute particle fraction are observed along the axis. A small ratio of the inner to outer radius was used, with $R_i/R_o=0.29$, and $R_i=0.64$ cm, resulting in concentration bands which were confined to the vicinity of the inner cylinder. This work examines the dynamics of band formation and subsequent band motions for a range of filled fractions of the annular region, f, and angles of inclination of the Couette device relative to the horizontal, α. The majority of the experiments were performed at a bulk average particle volume fraction of $\phi=0.2$, although banding was observed over the range of concentrations $0.01\leqslant\phi\leqslant 0.4$. The focus of this work is on band dynamics, which have been analyzed by time-lapse video imaging and image analysis. At zero inclination angle, the concentrated bands at $f<0.65$ are narrow, and fluctuate both in position and in number; the intervening zones are dilute relative to the bands but only slightly less concentrated than the bulk average. At fill fractions above $f=0.65$ and zero inclination, the number of concentrated bands decreases while the bands become much longer in the axial direction, and the intervening regions are both narrow relative to the concentrated bands and very dilute, $\phi\approx 0$. This change in banding behavior occurs over a narrow range around $f=0.65$, which corresponds to suspension just covering the inner cylinder in the absence of flow. For inclination angles in the range $0.4°<\alpha<5.3°$, concentrated bands form at regular intervals at the elevated end of the device (shallow depth) and move intact at a nearly constant speed down the cylinder axis. [DOI: 10.1063/1.1460877]

I. INTRODUCTION

Despite its intrinsic interest to coatings applications, the interaction of a flowing bulk suspension with a free surface has received little attention. The usual approach in such flows has been to treat the suspension as an effective fluid in which the particles alter the rheology. Recent observations of a segregation phenomenon in the free-surface suspension flow generated by partial filling of a concentric-cylinder shear flow (Couette) apparatus with a viscous suspension have, however, demonstrated that the presence of a free surface in a particle-laden flow can have remarkable effects. In particular, we note the axial concentration banding in a partially filled horizontal Couette flow, recently examined for viscous flow conditions by Tirumkudulu, Tripathi, and Acrivos,[1] although photographs showing similar axial particle fraction variation in the same flow geometry at much larger Reynolds number were presented earlier.[2] A similar segregation has been observed in flow of a suspension partially filling a single rotating cylinder.[3,4] While axial segregation of dry granular materials flowing in the latter geometry is well-known, free surface-induced segregation in viscous suspensions occurs at vanishingly small inertia and is of a different origin.

The phenomenon of interest is the following: when a noncolloidal suspension partially fills the annulus between the cylinders in a Couette device to leave a free surface, rotation of the inner cylinder results in axial segregation of the suspended particles into alternating concentrated and dilute bands. The mechanism leading to this segregation remains an open question. In the initial observation in a Couette device,[1] the particles and fluid were of equal density (neutrally buoyant), but the occurrence of banding in free surface flows is not limited to suspensions of neutrally buoyant particles as was shown in experiments using heavy particles by Boote and Thomas[3] in a partially filled rotating single cylinder.

Shear-induced migration in concentrated suspensions leading to nonuniform particle concentration in fully bounded suspensions has been of considerable interest for over a decade,[5–8] with some of this work specifically considering the radial migration in wide-gap Couette flow.[7,8] It is natural to consider that shear-induced migration may play a role in the axial segregation under a free-surface, but segregation in a partially filled Couette device has previously been reported[1] to occur at a particle volume fraction of $\phi=0.05$, and in this work has been observed to occur in a suspension of $\phi=0.01$. The multiparticle interactions necessary to generate the changes in rheology leading to shear-induced migration scale as ϕ^2 in the dilute limit,[5,7,8] and a scaling analysis presented in Sec. IV suggests that this mechanism would yield a migration rate too slow to explain the observed behavior at larger ϕ as well.

The range of behaviors possible in the free-surface Cou-

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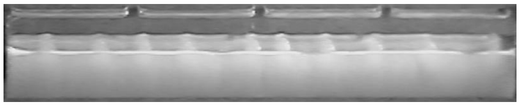

FIG. 1. Concentration bands formed in the Couette device with inner to outer cylinder ratio of $R_i/R_o=0.29$ at a fill fraction of $f=0.50$ and particle volume fraction of $\phi_{\text{bulk}}=0.2$. The rotation rate is 8 RPM and the particles are neutrally buoyant.

ette flow was not clear from the prior study. This work aims to expand understanding by considering free surface-induced segregation in a Couette flow under viscous-flow conditions differing in two ways from those of Tirumkudulu *et al.* (1999).[1] In that work, the bands bridged the entire gap between inner and outer cylinder. Because our interest is in examination of the dynamics of the bands, this bridging which restricts the motion of the bands is undesirable. Thus we study the free-surface flow in a Couette device with a ratio of inner to outer cylinder radii, $R_i/R_o=0.29$, which is markedly smaller than the value of 0.64 in the prior work. A photograph at a typical condition of 50% fill of the annular space with a suspension of bulk average particle fraction $\phi_{\text{bulk}}=0.2$ is shown in Fig. 1. Second, the inclination of the entire Couette device with respect to gravity has been allowed to vary. These changes are apparently simple ones, yet each opens a window to dynamics previously unobserved. The smaller R_i/R_o used here eliminates the interference by the outer cylinder, and also allows us to examine flows with free surfaces of qualitatively different form. Specifically, the free surface of the suspension may either cover the driving inner cylinder or not before flow commences, depending on the fraction of the cylindrical annulus filled with suspension. Previously unobserved phenomena occur at large fill fraction, meaning that the inner cylinder is completely covered prior to flow. Here, the segregation rate decreases with increasing fill fraction, but the concentrated bands are eventually separated by narrow zones essentially devoid of particles.

The most striking novel behavior reported here is due to inclination of the Couette device. Inclination breaks a basic symmetry found in prior studies,[1,3,4] and under inclination, concentration bands migrate down the axis away from the elevated end of the device. In general, the bands are identifiable over many hours: bands, periodically formed at the elevated end, flow intact with nearly uniform spacing down the entire length of the device.

The following section provides a description of our experimental methods and image analysis techniques. The number of parameters of the problem is large, and to provide context for the conditions probed by this work, a dimensional analysis of the suspension-flow problem in the Couette device is presented in Sec. II D. Results of our experiments are presented in Sec. III, with conclusions drawn from our investigations presented in Sec. IV. This final section includes a discussion of a proposed mechanistic basis for the onset of segregation.

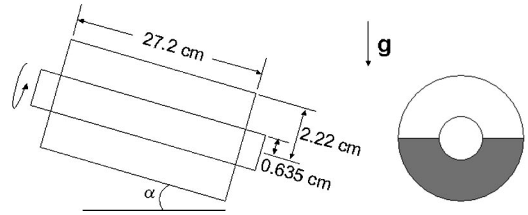

FIG. 2. Schematic in side and end views of the Couette device. The inner cylinder is stainless steel and has radius $R_i=0.6$ cm, while the outer Lexan cylinder has inner radius $R_o=2.2$ cm, and the annulus has length $L=27.2$ cm.

II. EXPERIMENTAL PROCEDURES

A. Suspensions and apparatus

Our experiments were performed in a concentric cylinder, or Couette, device shown schematically in side and end views in Fig. 2. The actual device is seen from the side in Fig. 1. This device consists of two concentric, 27.2 cm cylinders. The outer cylinder is a tube of Lexan (polycarbonate plastic), and the solid interior cylinder is stainless steel. The annulus is closed at each end by Lexan caps into which a sealed bearing is mounted, allowing the inner cylinder to turn while the outer cylinder is stationary. The cylinder motion is driven by a 1/15 horsepower electric motor. The device can be mounted horizontally (axis perpendicular to gravity) or in an inclined position; as seen in Fig. 2, this inclination is defined by the angle, α, which the cylinder axis makes with the horizontal. Angles studied were $0\leqslant\alpha\leqslant 6.5°$.

The radius of the solid inner cylinder is $R_i=0.64$ cm, while the outer cylinder has inner radius $R_o=2.2$ cm, and the annulus has length $L=27.2$ cm. The fill fraction occupied by the suspension is given by $f=V_{\text{susp}}/\pi(R_o^2-R_i^2)L$; the volume of suspension, V_{susp}, in the annulus was determined from the known suspension density and the measured mass of suspension placed into the device.

Two outer cylinders were used. For sufficiently small fill fraction, an outer cylinder with four 2.54 cm by 6.2 cm rectangular holes in the top was used, allowing easier filling and optical access. Larger fill fractions required, in order that the flow not be disturbed, a complete outer cylinder which was filled through two 0.6 cm radius circular holes.

Spherical poly-(methyl methacrylate) (PMMA; ICI Acrylics) particles were sieved to diameters in the range $2a=250-300\ \mu$m where we introduce a for the radius. The suspending fluid was a mixture of 76% of the surfactant Triton X-100 (Sigma), 16.2% zinc chloride ($ZnCl_2$), and 7.8% water, with the percentages based on mass. This mixture was density matched ($\rho=1.180$ g/cm^3) and roughly refractive index (RI) matched to the PMMA particles (RI$=1.491$ at $T=20$ °C). This suspending liquid mixture had a viscosity of $\mu=160$ Poise at $T=23$ °C. A few experiments used different ratios of the same components of the suspending liquid to vary the ratio of fluid to particle density (ρ_f/ρ_p) from unity. These will be noted where the results are described.

B. Particle concentration measurements

Particle concentration was measured in a limited number of experiments by direct sampling of the suspension from the Couette device. A variable volume pipette with opening of diameter 1.5 mm was used to remove approximately $m = 0.3$ g of suspension from a position in the flow. The suspending liquid was removed by flooding the suspension with warm water, and the particles were dried and weighed to find their mass, m_p. The particles and fluid were equal density in all sampled cases, yielding $\phi = m_p/m$. The method was accurate to ± 0.01 in the absolute value of ϕ based upon sampling from known-ϕ suspensions.

C. Band tracking

Band positions and velocities were determined for many of the experiments from time-lapsed video imaging of the Couette device taken for the duration of an experiment. This video was digitally downloaded to a personal computer. Individual images were analyzed by an algorithm, implemented using the Matlab Image Processing Toolbox, to determine positions of all bands, which were identified by characteristic image properties. For example, at small *f*, a concentrated band causes elevation of the free surface observable as a bright spot (see Fig. 1) reflecting more light to the camera. The algorithm was ''tuned'' such that positions determined computationally agreed with those found by eye; this was performed by setting the threshold level of light intensity for a position to be associated with a band. The information on band positions was further processed using a minimum spanning tree algorithm,[9] which identifies bands by linking close points to form lines (straight or curved) in a space–time plot of band positions. Groups with fewer than 10 points, determined by visual inspection of the video to be spurious in most cases, were discarded.

D. Dimensional analysis

The scope of our study is limited primarily to examination of the role of *f* and α upon the segregation phenomenon, with examination of limited ranges of certain other variables. Because numerous parameters are needed to fully characterize mixture flow in the Couette device, the number of relevant dimensionless variables in the general case is rather large. A dimensional analysis is presented here to delineate the factors which may influence the flow phenomena under the conditions studied.

We study the flow of a suspension with a very viscous suspending fluid. The bulk scale Reynolds number, $\mathrm{Re}_R = \rho_f 2\pi\omega R_i \Delta R/\mu$ with $\Delta R = R_o - R_i$, satisfies $\mathrm{Re}_R < 0.1$ for all conditions described and the Reynolds number based on the particle scale is much smaller, $\mathrm{Re} = (a/\Delta R)^2 \mathrm{Re}_R = O(10^{-6})$. Inertia is thus negligible. Particle sedimentation is negligible for the standard case of neutrally buoyant particle suspensions, although we have examined cases with weak positive and negative particle buoyancy. The particles are of noncolloidal scale at 250–300 μm in diameter and are readily dispersed in the surfactant solution used as suspending fluid. Brownian motion and nonhydrodynamic interparticle forces are therefore also negligible influences.

Geometric variables include the ratios $a/(R_o - R_i)$ and R_i/R_o, both of which are held fixed. Also fixed for our examination of the role of *f* and α is the bulk particle volume fraction, $\phi_{\mathrm{bulk}} = 0.2$. Our studies find that banding occurs in a horizontal device over the range $0.01 \leqslant \phi_{\mathrm{bulk}} \leqslant 0.40$. We have not performed experiments at ϕ outside this range.

The remaining dimensionless variables for a monodisperse suspension are the fraction of the annulus filled by suspension, *f*, the angle of inclination, α, and the ratio of gravitational to viscous stresses given by

$$G \equiv \frac{\Delta\rho g R_i}{\mu\omega},$$

where $\Delta\rho$ is the density difference between the suspension and the overlying fluid, ω is the angular velocity of the inner rod of radius R_i, and μ is the suspending fluid viscosity. A measure of the role of surface tension through a capillary number (viscous to surface tension effects) or Bond number (gravity to surface tension effects) is also needed based simply on dimensional considerations. Although it is expected that particle motions at the free surface may be influenced by surface tension,[10–14] we have not successfully examined this issue.

The primary variables considered here are the fill fraction, *f*, the angle of inclination, α, and the rotation rate of the inner cylinder, ω, expressed in rotations per minute (RPM). Because the experimental apparatus and physical properties of the suspension are fixed, we report dependence on the dimensional ω rather than the dimensionless quantity *G*.

III. RESULTS

As in prior experimental examination of free-surface Couette flow of a suspension,[1] we observe the formation of particle-rich bands separated in the axial direction by relatively dilute regions. However, we have also observed several features of the phenomenon not previously described, and we summarize these here. These include bands undergoing a fluctuational motion, coalescence of two bands to form one or simple disappearance of bands, and uniform-speed axial migration of the bands when the device is angled relative to the horizontal. We have also determined that the banding has different regimes depending upon whether the inner cylinder is covered by suspension without flow. The fill fraction which just covers the inner cylinder is defined as $f \equiv f_c$ with $f_c = 0.65$ for the apparatus of this study. For $f > f_c$, band formation rate was observed to slow dramatically, while the bands increased in width, and the number of bands was decreased.

The investigations reported here provide an experimental characterization of band formation and dynamics for a range of *f* and inclination angle, α. The majority of these were performed at $\phi_{\mathrm{bulk}} = 0.2$ and our standard rotation rate was $\omega = 8$ RPM; where it is not stated, these were the values used. We begin by considering in Sec. III A the influence of inclination angle upon the band dynamics, followed by a consideration of the influence of fill fraction in Sec. III B; the

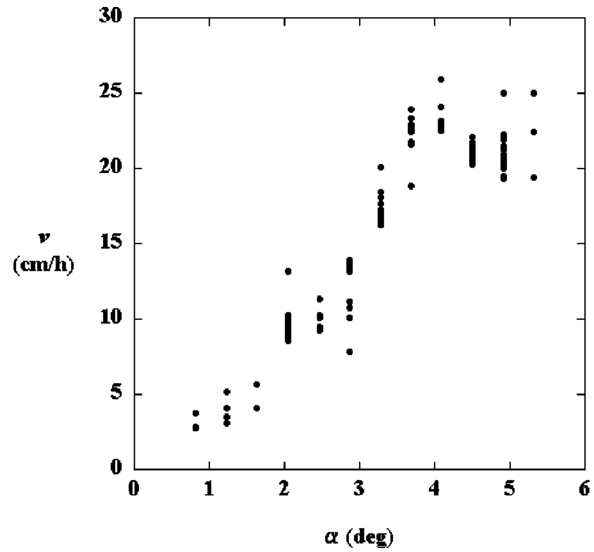

FIG. 3. Measured band velocity as a function of the incline angle α, for the conditions $\phi_{\mathrm{bulk}}=0.2$ and $f=0.50$, and inner cylinder rotation rate of 8 rotations/minute. The velocity plotted is the velocity of a particular band. The range of velocities determined for all bands measured is shown at each angle.

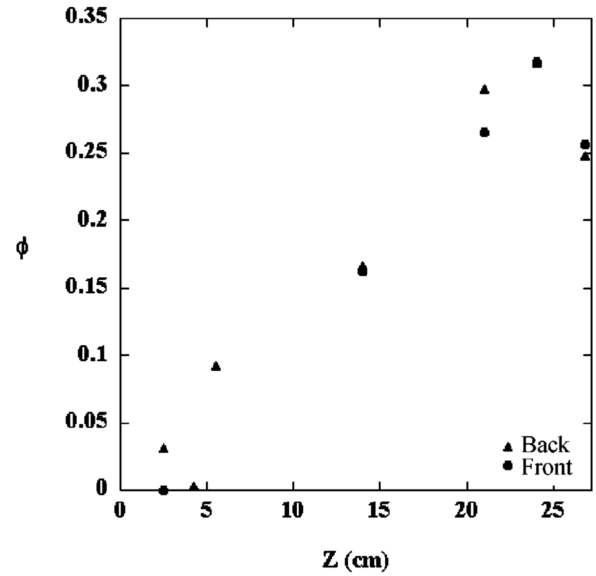

FIG. 4. Measured particle fraction along axis at $\alpha=6.5°$ for the conditions $\phi=0.2$ and $f=0.50$, and inner cylinder rotation rate of 8 rotations/minute. Here, ''front'' is defined as the side of the device on which the cylinder surface moves up, while ''back'' is defined as the side of the device on which the cylinder moves down.

experiments which probed these dependences are numerous and appear to adequately demonstrate the range of behaviors. We have also examined in more limited studies described in Sec. III C the influence of varying the rotation speed of the driving cylinder, the role of weak particle buoyancy, and the spreading of a drop of suspension; the latter study was performed to gain insight to drainage flows of suspensions.

A. Influence of inclination angle, α

Our experiments have shown that for appropriate conditions, the concentrated bands form and move at nearly constant speed, v, down the axis of the Couette for inclination angles $0.8° \leqslant \alpha \leqslant 5.3°$. In general, as shown by Fig. 3 for $\phi_{\mathrm{bulk}}=0.2$, $f=0.50$, and $\omega=8$ RPM the speed increases with increasing angle. At each value of α, several points are plotted. The velocity points plotted each correspond to the velocity of a particular band; because the band velocity varied slightly with time and with location of the band in the Couette, several fairly closely grouped values of the velocity are found for a single experiment. At $\alpha>5.3°$, bands are no longer observed to form. However, at these large angles, particles concentrate at the ''lower'' end of the Couette, i.e., at the end where the fluid is deeper. For $\alpha=6.5°$, $f=0.50$, and a bulk fraction of $\phi_{\mathrm{bulk}}=0.2$, the steady-state concentration was measured at a number of points along the axis, with the results shown in Fig. 4. It should be noted that for angles sufficiently large, $\alpha>4.5°$, the inner cylinder was raised completely out of the suspension at its elevated end and band formation and motion were clearly altered. This is the reason for the break in the linear increase of v with α seen in Fig. 3, but it is important to emphasize that even for angles $4.5° < \alpha < 5.5°$ bands continuously formed over the leftmost *wetted* portion of the inner cylinder.

From image analysis of the time-lapse video, the band positions have been identified. We consider first the band dynamics for zero and very small inclination angles. The axial positions of all of the identified bands as functions of time, $Z(t)$, are seen in Figs. 5(a) and 5(b), respectively, for representative experiments at $\alpha=0$ and $\alpha=0.4°$. All cases for which results are presented in Fig. 5 are at $\phi_{\mathrm{bulk}}=0.2$, $f=0.50$, and $\omega=8$ RPM. In the case of zero inclination, $\alpha=0$, the particle-rich band positions are found to fluctuate. Near the ends of the annulus, the band motion is biased by the endcaps (in a manner we do not understand), but in the center of the device the bands undergo an unbiased oscillatory motion with a characteristic time of 60–80 minutes; the motion is not sufficiently regular to define a period, as seen in Fig. 5(a). Similar oscillatory motion is observed for small angles, but is accompanied by an average motion of the bands from the elevated to the lower end of the device. Note also that there is correlation between the motions of adjacent bands; for example, consider $t=200$–500 minutes in Fig. 5(b), which illustrates the band dynamics for $\alpha=0.4°$. As noted the endcaps play a role, and for both $\alpha=0$ and $\alpha=0.4°$, the tendency of the particle-rich bands to move toward and ''die'' at the ends of the device is evident. In the case of $\alpha=0$, the bands near the center of the apparatus (between $Z=10$ and 20 cm) last essentially for the entire experiment, of duration greater than 24 hours, while those near the ends of the Couette are seen to form, migrate toward the wall, and vanish in 1 to 4 hours. At $\alpha=0.4°$, the migration of bands on average is down the incline, but is far from uniform for bands at different axial positions. Bands at the low end (to the right in the diagram) are observed to migrate much more rapidly than those at the elevated end, and as a result vanish into the boundary. This difference in the axial speed of the bands, coupled with the fact that bands vanish as they encounter the end cap, results in gaps larger than what may be termed the ''natural'' spacing of the bands, and

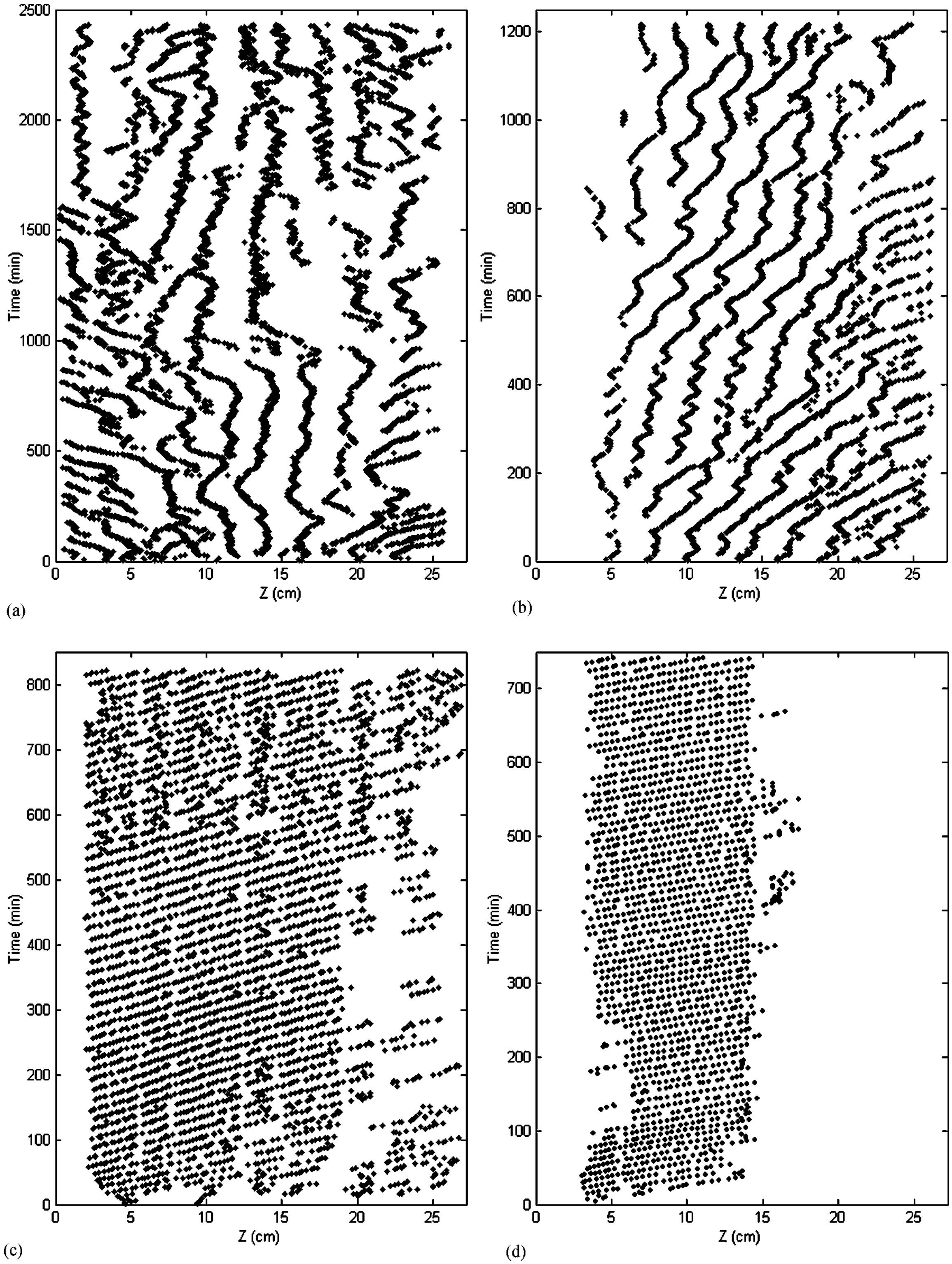

FIG. 5. Diagrams showing the concentration band positions as functions of time in the (a) horizontal Couette ($\alpha=0$), and at inclination angles of (b) $\alpha=0.4°$, (c) $\alpha=2.0°$, and (d) $\alpha=3.3°$. Time was set equal to zero when the presence of bands was first observed. In all cases, the suspension is at $\phi_{\text{bulk}}=0.2$ and the inner cylinder rotates at 8 rotations/minute.

new bands form between existing bands. This event is readily apparent in a video image, and can be seen in Fig. 5(a) at ($Z=9,t=420$) or ($Z=12,t=500$), and in Fig. 5(b) at ($Z=14,t=260$) or ($Z=14,t=350$).

We turn now to larger angles of inclination. For $0.8° \leqslant \alpha \leqslant 4.5°$, the bands migrate at a rate which is seen from Fig. 3 to be roughly linear in α: $v \approx k\alpha$ with $k \approx 5$ (cm/h)/degree of inclination. The band positions as a function of time for $\alpha=2.0°$ and $\alpha=3.3°$ are shown as Figs. 5(c) and 5(d), respectively. It is important to note that the bands are separate and do not form a continuous spiral structure. These plots illustrate that regular band formation occurs at the elevated end of the apparatus (left in the diagram) followed by regular motion of the bands toward the lower end. As the bands move down axis at these values of α, the interband spacing remains nearly constant, which is evident from parallel lines formed by the sequential positions of adjacent bands in Figs. 5(c) and 5(d). This spacing is found to decrease as α increases.

To conclude, we note that observation of the complete dynamics in this flow requires, in general, experiments of many hours duration. Both the band fluctuational motions at small α and the slow evolution of the bulk concentration field at large α take place over many cylinder rotations. For intermediate α, relatively short experiments provide information on the band formation and motion down the axis, although as the experiments progressed the appearance of new bands near $Z=0$ occurred at slightly longer intervals. This result is thought to be attributable to an apparatus-scale variation of the concentration similar to but weaker than that illustrated by Fig. 4 for $\alpha=6.5°$. It is interesting to take note of this last observation from a different perspective: despite the motion of the bands, there was only a very slow net transport of particles toward the lower end of the device.

B. Influence of filling fraction, *f*

We begin consideration of the influence of fill fraction by noting that the banding behavior depends qualitatively upon whether there is suspension completely covering the inner cylinder in the absence of flow. Note that all results in this section are for the horizontal configuration, $\alpha=0$. For $f<0.65$, the free surface of the mixture at rest lies below the topmost point on the inner cylinder. The depth of the suspension over the inner cylinder in the absence of flow, as a function of *f*, is illustrated in Fig. 6(a).

As *f* increases beyond $f\approx0.65$, the number of bands decreases, the width of a band increases, and the number of rotations required for band formation increases dramatically. To illustrate the difference in the band structure, bands formed at $f=0.50$ and $f=0.90$ are illustrated in Figs. 6(b) and 6(c), respectively.

The number of bands fluctuated during the course of an experiment and the observed range plotted as a function of *f* is presented in Fig. 7. For $f=0.50$, 8 to 11 bands are present, while at $f=0.90$ only three bands form. The abrupt change in number of bands at $f\approx0.65$, where as noted previously the inner cylinder is just covered by suspension, leads us to define $f_c=0.65$ as a critical fill fraction for $R_i/R_o=0.29$.

The concentration bands formed at $f>f_c$ are of qualitatively different structure from those at smaller *f*. The concentrated bands are wider, and for $f>0.8$, the regions between the bands are swept essentially free of particles after two to three days at 8 RPM (over 25 000 rotations). A similar complete removal of particles from the dilute regions has been observed for certain parameters in the segregation of a suspension in a partially filled and rotating single cylinder.[4] For the Couette flow at $R_i/R_o=0.29$ studied here, this is in sharp contrast to the dilute regions formed for $f<f_c$. For $f<f_c$, the concentrated bands are at ϕ well above $\phi_{\rm bulk}$, but because of the narrowness of these elevated-ϕ regions, it is

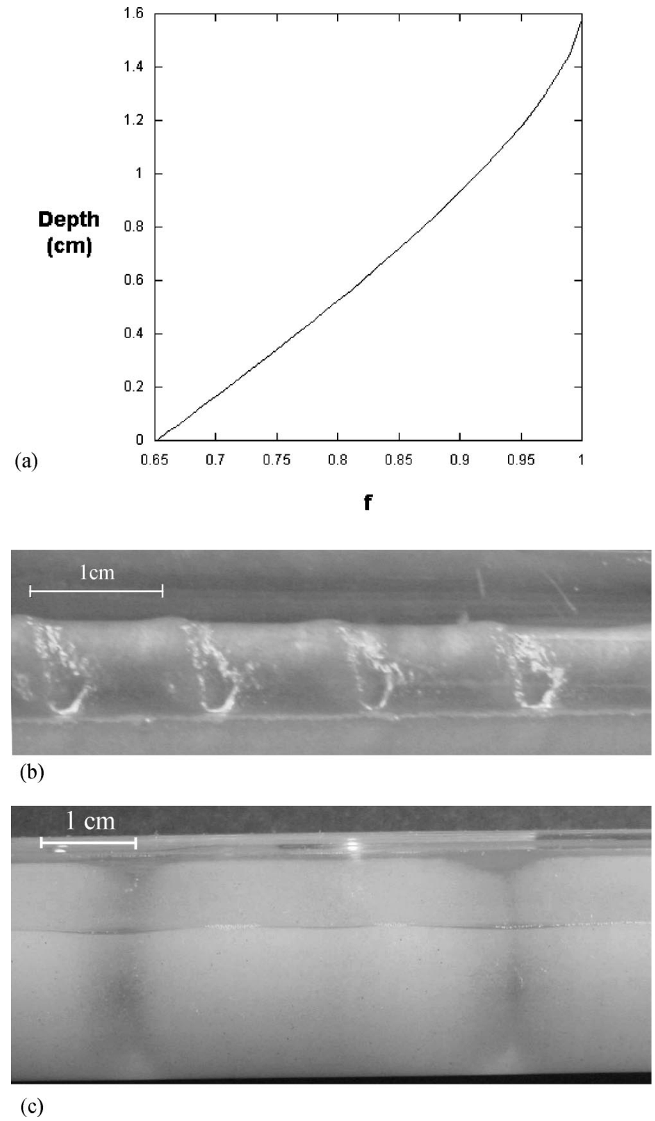

FIG. 6. In (a) is a plot of film thickness (when the inner rod is not rotating) above the inner cylinder of the device at several *f*; $f_c=0.65$ is the fill fraction at which the suspension just covers the inner cylinder. Photographs of bands at $f=0.50$ and $f=0.90$ at $\phi_{\rm bulk}=0.2$ and $\omega=8$ RPM are shown in (b) and (c), respectively. The bands in (b) are observed after roughly 200 rotations of the inner cylinder, while those in (c) were first observable after approximately 25 000 rotations, and the image was taken after about 50 000 rotations.

possible for the ''dilute'' regions to differ little from the bulk average concentration: we have made measurements of $\phi=0.35$ within a band for $\phi_{\rm bulk}=0.2$, while the separating regions were measured at $\phi=0.19$.

The characteristic time (or number of rotations) for band formation and fluctuational motion is also affected by fill fraction. It was noted above in the discussion of Fig. 5 that the time required for accessing the full dynamics of the bands was many hours at small fill fraction, and this time scale increased dramatically for $f>f_c$. Considering a suspension of $\phi_{\rm bulk}=0.2$, for $f=0.50$ bands are clearly apparent after about 200 rotations of the driving inner cylinder at a rotation rate of 8 RPM. By contrast, for the same volume fraction at $f=0.90$ and 8 RPM, there was no observable concentration variation after 12 000 rotations (roughly one

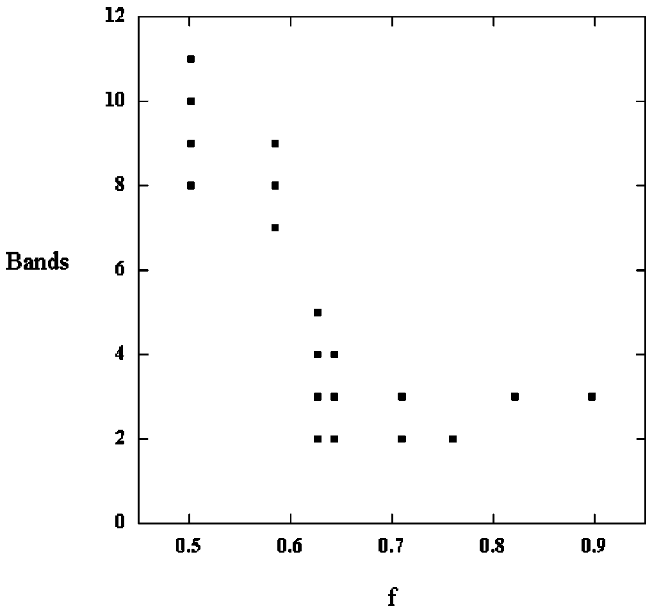

FIG. 7. Number of bands observed for the Triton X-100/$ZnCl_2$/water suspension as a function of fill fraction for $\phi_{bulk}=0.2$ at an inner cylinder rotation rate of 8 RPM. The symbols indicate the range of number of bands observed at a given *f*.

full day), definite bands were observed only after 25 000 rotations (two full days into the experiment), and ϕ in the dilute regions continued to decrease until it was essentially zero (after four days). The band motions described in Sec. III A for $f<f_c$ are not observed at elevated *f*. For $0.5<f<0.7$, the bands are found to migrate at $\alpha=0$, but this motion subsides rapidly with further increase in *f*. For $f>0.8$, the bands do not move measurably even over experiments of several days duration at the standard driving rate of 8 RPM.

C. Other results

Finite α at $f>f_c$: The behavior at small incline angle for high fill fraction was considered. This provided a test of the role of the film depth in the segregation process. At $\alpha=0.7°$ and $f=0.75$, the free surface lay above the inner cylinder at all axial positions before flow commenced, but the minimum depth of suspension over the cylinder at the elevated and lower ends was equivalent to that in a horizontal Couette where $f\doteq 0.7$ and $f\doteq 0.8$, respectively. The expectation based upon arguments presented in prior work[4] was that bands would form, if at all, first at the elevated end of the device where the ''film'' was of least depth. A single band was, in fact, observed at the elevated end of the device within roughly 300 rotations of the cylinder. This band became more pronounced over the course of an hour and another band formed toward the opposite end of the device after several hours. These bands did not, however, migrate down the axis.

Rotation rate: At $\alpha=0$, $f=0.5$, and $\phi=0.2$, the rotation speed was found to have the effects summarized in Table I. The most notable variation occurred in the range $\omega=4-6$ RPM, as the time for band formation was much larger at the slower rate and the number of bands was seven at $\omega=4$ RPM and increased to 12 at $\omega=6$ RPM. The formation time is an estimate based on the ability to clearly identify concentration bands on the videotape. The number of cylinder rotations for the formation of bands at 4 RPM was found to be over an order of magnitude larger than at 6 RPM (~1900 to ~160 rotations). Furthermore, the band structure for the lower rotation rate continued to evolve for a further 10 hours after the bands were initially observed. The number of bands was found to be essentially constant throughout an experiment for the 4 and 6 RPM conditions, but became quite variable as the rotation speed was increased to $\omega \geqslant 8$ RPM.

TABLE I. Band formation time and number of bands, reported as the range observed when variable, as a function of cylinder rotation rate, for $\phi_{bulk}=0.2$, $f=0.5$, and $\alpha=0$.

Rotation rate, ω (RPM)	Band formation time (min)	Number of bands
4	480	7
6	26	12
8	21	8–11
12	6	9–12
16	4	9–12
24	5	8–10
32		9–11

It is important to note that along with the abrupt change in segregation rate from $\omega=4$ RPM to $\omega=6$ RPM, there is a qualitative change in the band structure. At the slower rate, the bands are similar to those formed at large fill fraction, $f>0.65$ [see Fig. 6(c)], with broad concentrated bands separated by relatively narrow and very dilute regions. Note that the drainage from the inner cylinder is more thorough at the slower rate. This results in a very thin film of the suspension remaining on the inner cylinder, apparently too thin for band formation to occur here. Instead, the bands are observed to form throughout the entire radius of the annulus (from R_i to R_o) at $\omega=4$ RPM, rather than on or directly adjacent to the inner cylinder as seen at the higher rates.

A comparison of the role of ω seen in this work with prior observations shows that the separation between the bands in a Couette device with $R_i/R_o=0.64$ was found by Tirumkudulu *et al.*[1] to increase with rotation rate, and the same trend was seen in the banding in a single cylinder by Boote and Thomas.[3] The latter study was at a larger Reynolds number. For $\omega=4-6$, we observe the opposite trend, but with the noted change in band structure. For $\omega \geqslant 6$, our results show a weak trend similar to the earlier work, as the ''average'' separation between concentrated bands is observed to increase slightly with the driving rate: for a substantial fraction of the time the number of bands is at the lower end of the range shown.

Non-neutral buoyancy: By altering the ratios of components, the liquid density was varied while keeping the viscosity roughly constant. The same PMMA particles were used in all experiments. At 8 RPM, $\phi_{bulk}=0.2$, and $\alpha=0$, slightly less dense particles ($\rho_f/\rho_p=1.38$) resulted in a behavior indistinguishable from that of neutrally buoyant particles. A weak excess particle density ($\rho_f/\rho_p=0.89$) at these conditions resulted in formation of bands which then de-

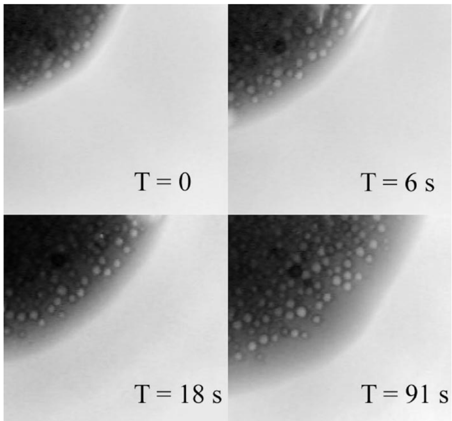

FIG. 8. Sequence of four images in the spreading of a dyed drop of the Triton X-100 suspension, of $\phi=0.2$, on a thin film of the suspending fluid. The times at which the images were taken are shown relative to an arbitrary zero time for the upper left image. The dark region free of particles results from the spread of the dyed suspending liquid.

tached from the inner cylinder and settled to the bottom of the annulus.

Spreading of a suspension drop on a wetted surface: The banding leads to elevated portions of the free surface, and prior work[4] has suggested that fluctuations of the free surface could be a factor in the segregation behavior. These fluctuations could be due, for example, to viscosity variation caused by variation of temperature or ϕ. Such fluctuations lead to gravity-driven (drainage) flows for which there is little understanding in the case of suspensions. To gain some insight, simple experiments on the spreading of a suspension drop were performed, using a suspension of neutrally buoyant particles and fluid. In these experiments, the Triton X-100 suspending fluid is dyed very dark purple (RIT liquid dye, Wine 10) and the particle surfaces are dyed black (RIT liquid dye, Black 15). The PMMA particles used were in this case not from a narrowly sieved fraction and the diameters ranged roughly from 100–300 μm, with the majority of the volume in the 250–300 μm fraction.

A drop of dyed suspension, volume approximately 0.1 cm^3, was dropped from an orifice at a height of 2 cm onto a horizontal plate covered by a submillimeter flat film of the undyed suspending liquid only. The viscous drop lost its inertia almost immediately upon impact. The subsequent spreading behavior is illustrated by the sequence of images in Fig. 8. These images are taken from a fixed vantage and focal point, with the first image, labeled by time $T=0$ s, taken at the instant the image first came clearly into focus (after about 1 s during which the initial curvature of the surface was reduced by spreading). This first image illustrates the formation of a leading ''foot'' of the dyed suspending liquid, which is observed as a dark region advancing ahead of the particles. The region of dyed fluid advances progressively farther ahead of the particles, which also continue to spread. The characteristic distance between the particle and fluid fronts is 600 μm, 1000 μm, 1400 μm, and 2000 μm (2 mm), respectively, at 0, 6, 18, and 91 s. Independent tests showed that diffusional spreading of the dye was essentially unmeasurable on this time scale. The advancing of the liquid front ahead of the particles indicates that there is ''differential drainage'' in the thin film: the film-averaged velocity of the particles is smaller than that of the liquid in the direction of spread. Differential drainage is discussed in connection with a proposed mechanistic basis for the onset of banding in Sec. IV.

IV. DISCUSSION

Particle segregation in the Couette device is apparently caused by repeated exposure of the suspension to a free surface. Our examination of this phenomenon has focused upon the role of fill fraction and angle of inclination, thereby exploring the behavior under conditions previously unstudied. Introduction of inclination breaks a basic symmetry of the system relative to prior studies, and the dynamics at finite inclination include a highly regular down-axis motion of evenly spaced bands. The observation of regular motion of long-lived bands prompted consideration of the general characteristics of the band motions, and the bands have been shown to undergo sustained fluctuational motion in the horizontal device.

Such fluctuational motions of the concentrated bands have not been described previously, and this may be related to the geometry of the Couette device used. The device used for this work was a wide-gap Couette of radius ratio $R_i/R_o=0.29$, and while we find qualitative behavior similar to that found[1] at $R_i/R_o=0.64$, the bands formed at $R_i/R_o=0.29$ have a different appearance at small fill fractions, being narrow, directly adjacent to the inner cylinder rather than spanning the annulus, and separated by regions only slightly diluted relative to the mean ϕ. Because of the small ratio of R_i/R_o, there was no bridging of the gap by suspension over the inner cylinder as seen in prior work in this geometry, leaving the free surface undisturbed by the outer wall. The smaller R_i/R_o allowed examination of the flow for fill fractions well above and below the value of f which provides complete coverage of the inner cylinder, specifically $f_c=0.65$.

The summary observation is that f_c separates two regimes of behavior. The band number and structure change abruptly at $f_c\approx0.65$ for $\alpha=0$. The primary influence of the fill fraction appears to be due to its relation to the thickness of the ''film'' of suspension covering the inner cylinder. At $f<f_c$, this film is generated by the cylinder carrying the suspension over to the opposite side of the device and thus drainage competes with the driving rotation to establish a film thickness. This thickness is generally thinner than the film depth generated in the case when $f>f_c$. There appears to be a lower limit to the film thickness for which the banding occurs due to flows on the inner cylinder; very slow rotation rates at $f<f_c$ yield such a thin film that banding occurs in the entire radial extent of the annular region rather than on the inner cylinder.

The velocity field is also influenced by fill fraction. For $f<f_c$ the mixture is forced to return predominantly under the cylinder. The coupling of the particle fraction to the velocity field warrants further study, as the return flow under the inner cylinder is known to be a factor in the centrifugal instability

leading to recirculation eddies and pattern formation of pure liquids in the partially filled Couette geometry.[15,16] However, our studies are at vanishing inertia and the results obtained for varying f and α have shown that banding occurs for a range of base state flows, supporting the view that instability of the subsurface flow is not a cause of the concentration banding.

In regard to the cause of the banding, it has been suggested[3,17,18] that shear-induced migration leads to the segregations. A scaling analysis shows that for the migration distances in the Couette flow, which are comparable to the inner cylinder radius, the time required is $t \sim R_i^2/D_{\mathrm{sh}}$, where $D_{\mathrm{sh}} = \dot{\gamma} a^2 \mathcal{D}(\phi)$ is the shear-induced diffusivity and $\mathcal{D}(\phi)$ is a dimensionless function of particle fraction. Simulation and experiment have been performed to determine $\mathcal{D}$ and an upper estimate from experimental work[19] (smaller values are found in simulation[20]) for the axial component is $\mathcal{D}_{zz}(\phi = 0.2) \approx 0.02$. The shear rate in the thin film is estimated as $\dot{\gamma} \sim \Delta \rho g h/\mu$. Consider the time predicted for shear-induced migration for particles of $a = 150\ \mu$m and a film prior to banding of $h = 0.2$ cm in thickness for $\omega = 8$ RPM. Using the pure fluid viscosity, the full gravitational acceleration, and assuming the suspension is subjected to the film shear rate at all times overestimates the rate of migration. However, even with these values for parameters, $t \sim 6 \times 10^4$ s, or almost 17 hours, during which time the cylinder would undergo roughly 8000 rotations at $\omega = 8$ RPM. Development of bands occurs in 200 rotations in most cases for $f < f_c$ (see Table I), or 40 times more rapidly than this prediction. We further note that migration was observed in a suspension of $\phi_{\mathrm{bulk}} = 0.01$ after several hours at 8 RPM in the Couette at $f = 0.5$, and the procedure above yields an estimate for shear-induced migration on the order of several *hundred* hours for this condition. Hence, there is strong evidence against shear-induced migration as the mechanism responsible for the observed segregation.

Experimental results from this work suggest that the minimum film depth is a controlling factor in the rate and structure of the observed banding. For fill fractions $f < 0.65$ in which the film is thin at the driving rates of this study, about 10 narrow bands of elevated ϕ are formed within 200 rotations of the inner cylinder, with the intervening regions only slightly diluted relative to ϕ_{bulk}. The bands formed at these low fill fractions are mobile, and their positions fluctuate, leading to coalescence and vanishing at the axial endcaps at zero inclination, $\alpha = 0$. For small values of α, the band formation rate and migration speed are observed to be regular. As the fill fraction increases beyond $f_c = 0.65$, the time scale, or number of rotations of the driving cylinder, required for banding increases rapidly. From less than 200 rotations to establish bands at $f = 0.50$, the number increases to about 4000 at $f = 0.71$, and to at least 25 000 rotations at $f = 0.90$. At large f, the concentrated bands become wider and the dilute regions, as seen in Fig. 6(c), tend toward $\phi = 0$. The behavior for $f > 0.90$ remains unknown.

The mechanistic basis which appears most plausible for the earliest stages, or onset, of axial segregation to concentrated bands is differential rates of drainage of particles and fluid by gravity-driven flows from fluctuations in surface elevation. We interpret the arguments presented here to be consistent with the discussion of banding segregation in the partially filled single cylinder presented by Tirumkudulu *et al.* (2000).[4] In that study, the particle size relative to the film thickness in the drainage flow down the cylinder wall was noted to be a controlling factor in the degree of segregation. The central idea upon which our proposed mechanism rests is that in gravity-driven flow in a thin film of suspension the fluid and particle phase film-averaged velocities will differ. Results of the drop spreading experiments described in Sec. III C support this notion. If we consider fluctuations of the free surface of a Couette flow along the cylinder z axis of form $\delta h = \epsilon \cos(z/\lambda)$ at uniform ϕ, the more rapid fluid drainage would result in a slight increase of the local ϕ at the crests of the surface. Such a mechanism is self-propagating in the sense that increased ϕ leads to an increase in the local effective viscosity, which in turn will cause the free surface to remain elevated at this position on subsequent rotations. The primary factors which cause "differential drainage" of the phases appear to be the following: (a) the layer of material directly adjacent to the free surface is liquid if the particles are wetted, and has the largest gravity-driven velocity, and (b) the motion of a particle within a thin film will be hindered by the presence of the solid (and to a lesser degree the free) surface. Both factors will result in fluid flowing ahead of the particles in an experiment such as the drop-spreading tests described in Sec. III C. Both factors also have relatively less influence at film depths which are many times the particle size. For this reason, the segregation is expected to be most rapid in regions of small film depth, and under limited testing we find this expectation holds. As noted in Sec. III C, at an overall $f = 0.75$ and small inclination, banding occurs most rapidly in the elevated end of the Couette where the film is shallow.

The arguments above are only intended to hold for the onset of the segregation. The subsequent evolution of the band structure involves complex secondary flows owing to the structure of the particle fraction field. The flow in the Couette device becomes three dimensional when the particle concentration varies axially, and in our experiments we have observed—after the banding is well-developed—pronounced extensional flows which pull suspension into the bands on the upflow side of the driving cylinder. Other structural features, including a "triplet" band structure where an individual concentrated band is made up of one central and two side bands, have been described in recent work on the single cylinder banding segregation by Thomas *et al.*[17] An understanding of how structural features of the band couple to the mixture velocity appears to require a detailed analysis of the velocity field.

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