

LETTERS

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Theory of suspension segregation in partially filled horizontal rotating cylinders

Rama Govindarajan^{a)}

Fluid Dynamics Unit, Jawaharlal Nehru Centre for Advanced Scientific Research, Jakkur,
Bangalore 560 064, India

Prabhu R. Nott^{b)}

Department of Chemical Engineering, Indian Institute of Science, Bangalore 560 012, India

Sriram Ramaswamy^{c)}

Centre for Condensed-Matter Theory, Department of Physics, Indian Institute of Science,
Bangalore 560 012, India

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It is shown that a suspension of particles in a partially-filled, horizontal, rotating cylinder is linearly unstable towards axial segregation and an undulation of the free surface at large enough particle concentrations. Relying on the shear-induced diffusion of particles, concentration-dependent viscosity, and the existence of a free surface, our theory provides an explanation of the experiments of Tirumkudulu *et al.* [Phys. Fluids **11**, 507 (1999); **12**, 1615 (2000)]. © 2001 American Institute of Physics. [DOI: 10.1063/1.1418362]

The primary motivation for this work is to understand the experiments of Refs. 1 and 2, showing spontaneous segregation in sheared suspensions. In these experiments, mono-disperse polymer spheres several hundred μm in diameter were suspended uniformly in fluids about 1000 times as viscous as water, with the same mass density as the particles, so that there was no sedimentation. The initial experiments¹ were carried out with a suspension in a horizontal Couette cell, i.e., in the gap between two concentric cylinders, but in subsequent studies,² a single horizontal cylinder of radius R was used. In both sets of experiments the container was filled only partially, i.e., there was a free surface. Let us restrict our description to the experiments in Ref. 2 for simplicity. When the cylinder was rotated at a tangential speed $v_0 = R\Omega$ about its symmetry axis the initially uniform suspension was found to undergo a dramatic instability towards segregation into bands of high and low concentration, with wave vector along the cylinder axis. The surface profile was modulated as well, i.e., the thickness of the fluid layer varied along the axis (see Fig. 1), with thicker regions corresponding to higher concentration. No instability was seen if there was no free surface, i.e., when the cylinder was completely filled with suspension.

The phenomena reported in the experiments^{1,2} are

among the many intriguing effects known to arise in suspensions of non-Brownian particles in highly viscous fluids, driven by shear flow or sedimentation. The feature of these suspensions that is of relevance in this paper is that the particles diffuse³ even though their thermal Brownian motion is negligible. The microscopic explanation³⁻⁵ for this diffusion is that the hydrodynamic interaction between the particles renders their motion chaotic, even in the Stokesian limit where inertia of fluid and particles is ignored. The diffusive flux of particles has two parts, one driven by a gradient in the particle volume fraction ϕ , the other by a gradient in the deformation rate $\dot{\gamma}$. For situations in which the predominant variation is with respect to a single coordinate z (e.g., the axial coordinate of the cylinder in Refs. 1 and 2) and time t the local volume fraction $\phi(z, t)$ (integrated over the remaining directions) of particles obeys the conservation law

$$\frac{\partial \phi}{\partial t} = - \frac{\partial j}{\partial z}, \quad (1)$$

where the shear-induced current^{3,6} can be written as

$$j = -f_c(\phi)a^2\dot{\gamma}\frac{\partial \phi}{\partial z} - f_s(\phi)a^2\phi\frac{\partial \dot{\gamma}}{\partial z}. \quad (2)$$

In (2), a is the particle radius, and f_c and f_s are dimensionless functions of the particle volume fraction ϕ .^{3,6,7} Note that (2) says that particles can move in the absence of concentration gradients, or even *against* concentration gradients, if the gradient in the deformation rate is appropriately directed.

^{a)}Electronic mail: rama@jncasr.ac.in

^{b)}Electronic mail: prnott@chemeng.iisc.ernet.in

^{c)}Also with JNCASR, Bangalore 560 064, India. Electronic mail: sriram@physics.iisc.ernet.in

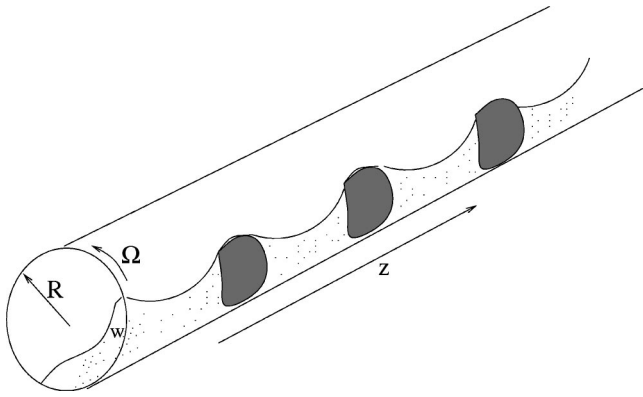


FIG. 1. Schematic of steady-state surface profile of a horizontal cylinder of radius R , rotating with angular speed Ω . The thickness w of the suspension layer dragged up, as well as the concentration of solute indicated by shading, are modulated with respect to the axial coordinate z (see Ref. 2).

These equations are an essential ingredient of our theory of the shear-induced segregation seen in Refs. 1 and 2.

The main result of our analysis is that Eqs. (1) and (2), when applied to neutrally buoyant Stokesian suspensions in horizontal rotating cylinders, predict precisely the instability seen in the experiments of Refs. 1 and 2, if the concentration is large enough. The growth rate Γ_q of the instability varies as q^2 for small wave number q . The parameters which govern the instability depend only on the volume fraction ϕ of the suspension and the fill fraction of the cylinder. At low rotation rates, Ω scales completely out of the problem: the range of unstable wave numbers is therefore independent of Ω . For ϕ just above the instability-onset value ϕ_c , Γ_q reaches a maximum at $q = q_* \sim \sqrt{(\phi - \phi_c)\rho g / \sigma}$, where σ is the surface tension of the suspension. For reasonable values of these parameters we find that the fastest growing mode has wavelength of order centimeters. We also explain why the instability disappears for large rotation rates.

We now obtain coupled equations of motion for the particle concentration and free-surface profile, and show that these lead naturally to the above results. Consider a homogeneous suspension with kinematic viscosity ν , filling a frac-

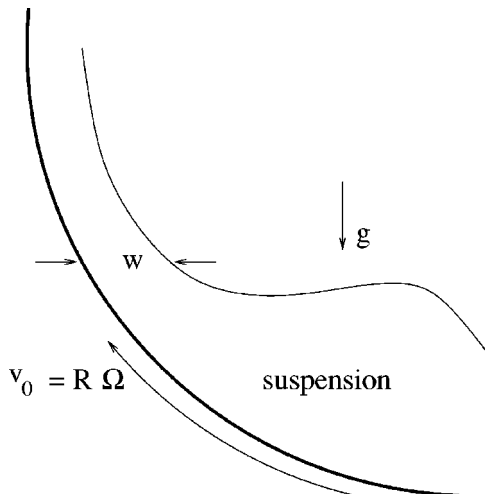


FIG. 2. Cross-sectional schematic of profile of fluid layer of characteristic thickness w and kinematic viscosity ν , dragged up against gravity g by a cylinder rising at speed v_0 .

tion F of the volume of a horizontal cylinder of radius R , rotating about its symmetry axis with a tangential velocity $v_0 = R\Omega$ (see Fig. 2). Recall first the results of Ref. 8: (a) The dimensionless combination $\beta \equiv F \sqrt{gR^2/\nu v_0}$, where g is the acceleration due to gravity, measures the relative importance of gravitational and viscous forces. (b) As v_0 is increased (i.e., as β is decreased), a fluid film of thickness \bar{w} is dragged up and coats the cylinder wall. (c) Since \bar{w} , for low speeds, is smaller than the depth of the residual pool of fluid at the bottom of the cylinder, the thickness profile has a “bump” at the bottom. (d) Once \bar{w} reaches a value $\approx FR$, which occurs for $\beta = \beta_c \approx 1.4$, the growth of \bar{w} saturates since all the available fluid then coats the cylinder more or less uniformly, and the bump disappears. (e) For higher speeds, i.e., for $\beta < \beta_c$, \bar{w} is effectively independent of the rotation speed, and is determined simply by the geometrical statement $\bar{w}/R = F$. The mechanism we propose below for the instability applies only when the thickness is determined by the rotation speed by an explicit balance between viscous and gravitational forces which is why, in Ref. 2, the instability disappears when the bump does.

Consider a general situation (Fig. 2) where the thickness $w(z, t)$ of the fluid film dragged up, as well as the volume fraction field $\phi(z, t)$ (and hence the viscosity), are varying in space and time. The component of the deformation rate that could vary in the axial direction is given by the velocity difference across the layer divided by the thickness:

$$\dot{\gamma}(z, t) \propto \frac{v_0 - \alpha_2 \dot{w}}{w}, \quad (3)$$

where α_2 is a pure number of order unity, independent of material parameters. The experiments of Refs. 1 and 2 are performed on highly viscous fluids, so that the Reynolds number is very small over the entire range of speeds and length scales studied. We shall therefore work in the limit of zero Reynolds number, where the inertia of particles and fluid are ignored. Accordingly, the balance of gravitational, viscous and interfacial forces per unit area of the layer tells us that

$$\rho g w(z, t) = \alpha_1 \eta(\phi) \frac{v_0 - \alpha_2 \dot{w}}{w} + \sigma \frac{\partial^2 w}{\partial z^2}, \quad (4)$$

and in particular that the layer thickness in the *steady, spatially uniform* state is

$$\bar{w} = \sqrt{\frac{\alpha_1 \eta(\phi) v_0}{\rho g}}. \quad (5)$$

In (4) and (5), ρ , $\eta(\phi)$, and σ are, respectively, the density, effective viscosity (as a function^{9,10} of the local particle volume fraction ϕ) and surface tension of the suspension, g is the acceleration due to gravity, and α_1 is another geometrical factor of order unity, with no dependence on any material parameter. The fill fraction F determines the angle made by the cylinder with the free surface of the pool of suspension, and hence the details of the flow in the fluid layer. This is

reflected in the parameters α_1 and α_2 in our model, but our conclusions are qualitatively insensitive to their precise numerical values.

Let us now perturb the thickness and concentration fields: $[w(z,t), \phi(z,t)] = [\bar{w} + \delta w(z,t), \phi_0 + \psi(z,t)]$. This will in turn lead to perturbations of the local values of $\dot{\gamma}$ and η , yielding closed equations of motion for the evolution of $\delta w(z,t)$ and $\psi(z,t)$ via (1) and (4). We work to linear order in δw and ψ . Let us work in terms of the nondimensional quantities $H \equiv \delta w / \bar{w}$, $\tau \equiv (v_0 / \alpha_2 \bar{w})t$, $\zeta \equiv z/a$. ψ is of course already dimensionless. Note that to write (1) and (4) in terms of thickness and concentration fluctuations, we must use (3) to express the local deformation rate in (2) in terms of the thickness perturbation, and replace a local viscosity perturbation by a local concentration fluctuation via $\delta\eta/\eta \approx \mathcal{N}\psi$ where

$$\mathcal{N} \equiv \frac{\partial \ln \eta}{\partial \phi}(\phi = \phi_0). \quad (6)$$

Although the procedure is straightforward, some care must be taken in obtaining the perturbation equation from (1): the perturbation of $\dot{\gamma}$ will involve \dot{w} , which must then once again be eliminated in favor of δw , ψ . Carrying out these steps, and Fourier-transforming with respect to ζ , i.e., considering spatial variation of the form $\exp i q \zeta$, we find that the Fourier components H_q , ψ_q obey

$$\begin{aligned} \frac{\partial}{\partial t} \begin{bmatrix} H_q \\ \psi_q \end{bmatrix} &= \mathbf{M} \begin{bmatrix} H_q \\ \psi_q \end{bmatrix} \\ &\equiv \begin{bmatrix} -(2 + \Sigma q^2) & \mathcal{N} \\ -Sq^2(1 + \Sigma q^2) & -(C - \mathcal{N}S)q^2 \end{bmatrix} \begin{bmatrix} H_q \\ \psi_q \end{bmatrix}, \end{aligned} \quad (7)$$

where

$$S \equiv \alpha_2 \phi_0 f_s(\phi_0), \quad C \equiv \alpha_2 f_c(\phi_0), \quad \text{and} \quad \Sigma \equiv \frac{\sigma}{\rho g a^2}. \quad (8)$$

The stability or otherwise of our sheared suspension is determined by the characteristic equation

$$\lambda^2 + (2 + Dq^2)\lambda + Eq^2 + C\Sigma q^4 = 0 \quad (9)$$

for the eigenvalues λ of the dynamical matrix \mathbf{M} in (7), where

$$D \equiv \Sigma + C - \mathcal{N}S, \quad E \equiv 2C - \mathcal{N}S. \quad (10)$$

For $q \rightarrow 0$, the solutions to (9) are

$$\lambda_1 \approx -\frac{E}{2}q^2, \quad (11)$$

$$\lambda_2 \approx -2 + \left(\frac{E}{2} - D\right)q^2. \quad (12)$$

We see from (11) that the uniform state is linearly unstable to segregation and thickness modulation if $E < 0$, with perturbations growing at a rate $\Gamma_q \sim q^2$ at small q , and particles concentrating in the thick regions. It is straightforward to show that λ_1 turns over at larger q , passing through zero at $q = \sqrt{-E/C\Sigma}$, with a peak located, for $E \rightarrow 0^-$, at

$$q_* \approx \sqrt{-E/2C\Sigma}, \quad (13)$$

which determines the observed wavelength of the initial instability.¹¹ For a given fill fraction, E can vary only with the volume fraction ϕ . If there is an instability, it must then be because E turns negative, in general as $\phi_c - \phi$, as ϕ crosses a critical value ϕ_c . This leads to the main result presented at the start of the paper. In terms of the parameters in (2) and (6), the instability criterion is

$$\left[\frac{\phi}{\eta} \frac{\partial \eta}{\partial \phi} \right]_{\phi_0} > \frac{2f_c(\phi_0)}{f_s(\phi_0)}, \quad (14)$$

which should in general happen in real suspensions at large enough ϕ_0 .

In more detail, note that the coefficient $f_c(\phi)$ originates^{3,6} from a direct shear-induced self diffusion as well as from a tendency to move down viscosity gradients. The latter tendency opposes the instability, as we shall now show. For a Newtonian suspension, η varies only if ϕ does. Thus, by the arguments of Leighton and Acrivos,³ we can write the current in (1) as

$$j = -a^2 \phi \left[M_s \phi \frac{\partial \dot{\gamma}}{\partial z} + \dot{\gamma} (M_c + M_\eta \mathcal{N}) \frac{\partial \phi}{\partial z} \right], \quad (15)$$

where M_c , M_s , and M_η are order-unity phenomenological quantities. Comparing (15) and (2) we see that $f_s = \phi M_s$ and $f_c = \phi (M_c + \mathcal{N} M_\eta)$, and the instability criterion (14) becomes $(M_s - 2M_\eta) \mathcal{N} > 2M_c$. The experiments of Refs. 3 and 6 have determined these coefficients for diffusion in the gradient direction, while diffusion in the problem we consider is in the vorticity direction. Further, a microscopic theory for determining them is also not available. Provided $M_s > 2M_\eta$, the growth of the viscosity with ϕ (Refs. 9 and 10) means that the instability should always arise at large enough concentration. An independent measurement of these coefficients is clearly called for.

We now assume that the uniform state is unstable and ask whether the typical wave number of the segregation changes from its initial value q_* , and whether nonlinear terms cause the exponential growth to saturate at long times. It suffices to look at the dynamics for the slow mode (eigenvalue λ_1 in the linearized limit). In that case H_q in (7) is “slaved” to ψ_q , $H_q \approx -\mathcal{N}\psi_q/(2 + \Sigma q^2)$. Expanding (2) about ϕ_0 , and retaining terms nonlinear in ψ_q , we obtain the effective equation of motion,

$$\frac{\partial \psi}{\partial t} = \frac{\partial^2}{\partial z^2} \left(\frac{E}{2} \psi + c_2 \psi^2 + c_3 \psi^3 \dots \right) - \frac{\mathcal{N}S}{4} \frac{\partial^4 \psi}{\partial z^4}, \quad (16)$$

where c_2 and c_3 are coefficients arising from the ϕ dependence of f_c , f_s , and η . Equation (16) is well-known in the domain-growth literature.^{12,13} In particular, it has been shown¹⁴ that the nonlinear terms in (16) with $E < 0$ cause the characteristic wavelength $L(t)$ of the pattern of segregation at time t to grow as $\ln t$ at long times. This extremely slow growth should in principle be testable by a patient experimenter.

We have shown that two classic properties of non-Brownian suspensions, viz., concentration-dependent

viscosity^{9,10} and shear-induced diffusion,³ lead to a natural explanation of the experiments of Refs. 1 and 2 on segregation in suspensions in partially filled, rotating horizontal cylinders. Our dynamical equations, at high enough concentration, display an instability towards axial segregation and a modulation of the free surface, with particles accumulating under the crests of the modulation. For parameter values, say, 10% past the instability, taking a plausible surface tension of 30 dyne/cm, (13) implies a wavelength of about 3 cm for the fastest growing mode at onset, which is consistent with the experiments of Ref. 2. Independent measurements of the parameters in (14) and (13), in transient experiments, should provide a stringent test of our theory, as should studies of the long-time behavior of the wavelength of the segregation pattern.

ACKNOWLEDGMENT

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