## Shock Dynamics in Particle-Laden Thin Films

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We present theory and experiments for thin film particle-laden flow on an incline. At higher particle concentration and inclination angle, a new phenomenon is observed in which a large particle-rich ridge forms at the contact line. We derive a lubrication theory for this system which is qualitatively compared to preliminary experimental data. The ridge formation arises from the creation of two shocks due to the differential transport rates of fluid and particles. This parallels recent findings of double shocks in thermal-gravity driven flow [A. L. Bertozzi *et. al.*, PRL, **81**, 5169 (1998), J. Sur *et. al.*, PRL **90**, 126105 (2003), A. Münch, PRL **91**, 016105 (2003)]. However, here the emergence of the shocks arises from a new mechanism involving the settling rates of the species.

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Thin films and coating flows have long been studied in industrial, biological and environmental contexts including microchip fabrication, the liquid lining in the lungs, and the wetting of contact lenses [1]. While extensive research has been conducted on clear fluids [2–6] and dry granular flows [7, 8], far fewer studies have been dedicated to particle-laden fluid films. In applications such as erosion and debris flow [9, 10], slurry transport, and mixing of pharmaceuticals, particles play a dominant role in the dynamics and cannot be neglected [11].

New and unexpected behavior is observed in these particulate systems owing to the introduction of a new time scale. In particle-laden, gravity-driven films [12], one time scale is associated with the viscous fingering that occurs at the contact line and a second is associated with particle settling. At low inclination angles, if these two time scales are widely disparate, the behavior of the suspension closely reflects that of clear fluids. However, new and interesting phenomena arise when the two time scales are comparable, resulting in nonuniform particle distributions.

In clear fluid experiments involving flow down or up an inclined plane, the large scale dynamics is described by convective theory in which the first order terms in the equations of motion yield quantitative information about the speed and shape of the front. Recent results [13–15] show that temperature gradient driven flow balanced by gravity can be described by a scalar conservation law with a nonconvex flux. The effect of surface tension in the shock layer is responsible for the emergence of 'nonclassical' shock patterns. In the present study we find similar dynamics in particle-laden films. New phenomena is observed experimentally in the limit of high particle concentration and inclination angle, and this data is compared qualitatively to predictions arising from shock theory.

The experimental apparatus consists of a  $30 \text{cm} \times 120 \text{cm}$ acrylic sheet with an adjustable inclination angle,  $\beta$ , ranging from 0° to 60°. A suspension of viscous fluid and polydisperse glass beads (250-425  $\mu$ m in diameter),



FIG. 1: Schematic of the experimental apparatus.

flows from a reservoir with an adjustable gate (see figure 1). Profiles of the flowing layer at fixed y, as functions of x and time, are measured using a laser light sheet and a high resolution digital camera (see figure 2). Experiments were conducted using both wetting (Dow Corning 200, 1000 cSt fluid) and partially-wetting (glycerol) fluids. Initial particle concentration was varied from 0% to 55% by volume.

As with clear fluid films, the propagating front undergoes a fingering instability (figure 2-Top). Unlike the clear fluid case, three distinct regimes were observed (figure 2-Bottom). At low inclination angles and concentrations, sedimentation perpendicular to the plate occurs rapidly and the particles are deposited in a uniform particulate front behind the advancing free surface. Clear fluid flows over the particulate matter and eventually fingers; the dynamics is precisely that observed in numerous clear fluid experiments ([e.g. 2, 16, 17]). At high inclination angles and particle concentrations, the evolution deviates strikingly from the clear fluid case and a large particle-rich ridge appears at the advancing free surface front. The ridge observed in these dense suspensions is considerably larger than that observed in clear fluids and may grow to several times the thickness of the trailing film. Between these two regimes, a transition re-



FIG. 2: (Top) Photographs of typical fingering instabilities in each regime. (Bottom-L) Phase diagram for 250-425  $\mu$ m beads in glycerol showing three regimes. (Bottom-R) Profiles of an advancing silicon oil front illustrating the widening ridge. Initial concentration  $\phi_0 = 45\%$  and inclination angle  $\beta = 42^{\circ}$ . Both axes are made dimensionless using *H*, the thickness of the film at the gate. Profiles are plotted every 30 sec.

gion appears characterized by regular fingers containing particles. Intuitively we expect the large ridge to arise if the particle settling velocity along the plate is faster than that of the propagating front. As the settling beads overtake the moving contact line, they accumulate, dramatically increasing the local viscosity.

The governing equations of motion for the mixture are

$$\nabla \cdot \mathbf{\Pi} + \rho(\phi)\mathbf{g} = 0, \quad \nabla \cdot \mathbf{j} = 0, \tag{1}$$

where  $\mathbf{j}$  is the volume averaged flux of the mixture,  $\mathbf{g}$  is the acceleration of gravity, and  $\mathbf{\Pi} = -p\mathbf{I} + \mu(\phi)(\nabla \mathbf{j} + (\nabla \mathbf{j})^T)$  is the stress tensor for the mixture. Here p is pressure,  $\rho(\phi)$  and  $\mu(\phi)$  are the effective density and viscosity respectively, and  $\mathbf{I}$  is the identity matrix. We consider low Reynolds number flows and neglect inertia of both the particulate and the fluid phase. The volume flux is related to the velocity by  $\mathbf{j}_p = \phi \mathbf{v}_p$ ,  $\mathbf{j}_f = (1 - \phi) \mathbf{v}_f$ and  $\mathbf{j} = \mathbf{j}_p + \mathbf{j}_f$ , where subscripts p and f refer to particle and fluid phase respectively and  $\phi$  is the local particle concentration (by volume). Hence,  $\mathbf{j}_p$  can be expressed in terms of the flux of particles relative to the fluid,  $\mathbf{j}_R$ , by  $\mathbf{j}_p = \mathbf{j}_R + \phi \mathbf{j}$ .

Particles move relative to the fluid phase via settling. This relative velocity is given by the Stokes settling velocity modified by two effects: (1) the presence of other particles in the dense suspension which hinders settling and (2) the presence of the no-slip boundary condition at the wall. To model the first, we adopt the Richardson & Zaki correction [18], valid for high concentrations,  $\bar{f}(\phi) = (1-\phi)^m$ , where *m* is empirically determined to be  $\approx 5.1$ . To capture the wall effect, we modify the Stokes settling velocity by a function that vanishes near the wall and approaches unity in the bulk flow [22]. Thus

$$\mathbf{v}_R = \frac{2}{9} \frac{\left(\rho_p - \rho_f\right) a^2}{\mu_f} \bar{f}(\phi) w(h) \mathbf{g}$$

where  $\mathbf{v}_R$  denotes particle velocity relative to the fluid phase, *a* is a characteristic particle radius,  $\mu_f$  is the viscosity of the fluid phase,  $\rho_f$  and  $\rho_p$  are the density of the fluid and particulate phase respectively, and  $w(h) \equiv \frac{Ah^2}{\sqrt{1+(Ah^2)^2}}$ . Here *h* is the local film thickness and *A* is a parameter that determines the sharpness of the transition from the no-slip wall to the bulk flow.

The effective mixture density  $\rho(\phi) = \phi \rho_p + (1 - \phi) \rho_f$ , and effective mixture viscosity [19]

$$\mu(\phi) = \mu_f (1 - \phi/\phi_{max})^{-2}$$
(2)

are both functions of the local concentration. The viscosity diverges as the concentration approaches  $\phi_{max}$  and the mixture becomes increasingly more solid-like.

At the substrate, we enforce a no-slip boundary condition. At the free surface, the normal stress balance is given by  $\mathbf{n} \cdot \mathbf{\Pi} \cdot \mathbf{n} = \gamma \kappa$  and the tangential stress balance by  $\mathbf{t} \cdot \mathbf{\Pi} \cdot \mathbf{n} = 0$  where  $\mathbf{n}$  and  $\mathbf{t}$  are respectively the unit outward normal and unit tangential vectors to the free surface,  $\gamma$  is the surface tension of the mixture (here assumed to be independent of particle concentration) and  $\kappa$  is the curvature of the free surface.

To simplify the analysis, we consider the 2D problem. neglecting variations in the y direction; i.e. we begin by analyzing the evolution of the film prior to the onset of fingering. Consider the following rescaling, motivated by the scaling adopted in gravity-driven clear fluid films [20]:  $x = \tilde{x}L \equiv \tilde{x} \left(\ell^2 H / \sin\beta\right)^{1/3}, z = H\tilde{z},$  $p = P\tilde{p} \equiv \frac{\mu_f L U}{H^2} \tilde{p}, \ t = \tilde{t}T \equiv (3\mu_f/\gamma)(\ell^2 L/H^2 \sin\beta)\tilde{t},$ and  $(j^x, j^z) = U(\tilde{j^x}, \epsilon \tilde{j^z})$ . Here  $\ell = \sqrt{\gamma/\rho_f g}$  is the capillary length, H is the thickness of the film far behind the moving front, dimensionless quantities are denoted by a tilde, and a characteristic velocity is defined as U = L/T. Equations (1) are rescaled using the above relations and we retain only the lowest order terms in  $\epsilon \equiv H/L = (3Ca)^{1/3}$  where  $Ca = \mu_f U/\gamma$  is the capillary number. If  $\epsilon Pe \ll 1$ , where Pe = UH/D is the Peclet number and D is the diffusion coefficient for the particulate phase, particles diffuse rapidly across the depth of the film creating a uniform distribution in z. Hence  $\phi$ may be approximated as  $\phi(x,t)$ . Following a standard long wavelength approach for thin films [e.g. 4], we solve for the velocities and pressures and depth-average the equations bearing in mind that there are two mass conservation conditions, one for the mixture and one for the particles. Dropping the tildes, this yields two coupled equations for the evolution of  $\phi$  and h:

$$\frac{\partial(\rho(\phi)h)}{\partial t} + \left\{\frac{\rho(\phi)}{\mu(\phi)}h^3h_{xxx} - D(\beta)\left[\frac{\rho(\phi)}{\mu(\phi)}h^3(\rho(\phi)h)_x - \frac{5}{8}\frac{\rho(\phi)}{\mu(\phi)}h^4(\rho(\phi))_x\right] + \frac{\rho(\phi)^2}{\mu(\phi)}h^3\right\}_x = 0$$
(3)

$$\frac{\partial(\phi h)}{\partial t} + \left\{\frac{\phi}{\mu(\phi)}h^3h_{xxx} - D(\beta)\left[\frac{\phi}{\mu(\phi)}h^3(\rho(\phi)h)_x - \frac{5}{8}\frac{\phi}{\mu(\phi)}h^4(\rho(\phi))_x\right] + \frac{\phi\rho(\phi)}{\mu(\phi)}h^3 + \frac{2}{3}V_s\phi hf(\phi)w(h)\right\}_x = 0$$
(4)

where  $V_s \equiv \frac{\rho_p - \rho_f}{\rho_f} \frac{a^2}{H^2}$ ,  $D(\beta) \equiv (3Ca)^{1/3} \cot(\beta)$ , subscripts denote partial derivatives, and we have absorbed the extra factor of  $(1 - \phi)$  into f by defining  $f(\phi) \equiv (1 - \phi)\bar{f}(\phi)$ .

A full lubrication model of the form (3-4) for particle laden flow has not been considered before in the literature. To determine macroscopic properties of the flow, it is instructive to investigate the underlying convective dynamics, described by the system of two conservation laws:

$$-\frac{\partial(\rho(\phi)h)}{\partial t} + \left\{\frac{\rho(\phi)^2}{\mu(\phi)}h^3\right\}_x = 0 \quad (5)$$

$$\frac{\partial(\phi h)}{\partial t} + \left\{ \frac{\phi \rho(\phi)}{\mu(\phi)} h^3 + \frac{2}{3} V_s \phi h f(\phi) w(h) \right\}_x = 0.$$
(6)

Consider as an initial condition a jump in the height from 1 to the precursor thickness b, and a constant concentration of  $\phi_0$ . That is, at time zero, h = 1 for  $x \leq 0$ , h = b for x > 0, and  $\phi = \phi_0$ . The above system has the form of a  $2 \times 2$  system of scalar conservation laws

$$\frac{\partial u}{\partial t} + [F(u,v)]_x = 0, \quad \frac{\partial v}{\partial t} + [G(u,v)]_x = 0, \quad (7)$$

where  $u = \rho h$  and  $v = \phi h$ . The initial condition gives us a Riemann problem i.e. both of the system variables u and v have a jump at the origin at time zero. Classical solutions of this problem, when they exist, are well-known in the theory of shock-waves [21]. An intermediate state  $(u_i, v_i)$  emerges separating the left state  $(u_l, v_l) = (\rho_f \rho(\phi_0), \phi_0)$ , from the right state  $(u_r, v_r) = (\rho_f \rho(\phi_0) b, \phi_0 b)$ . The values  $u_i$  and  $v_i$  determine the density of particles,  $\phi_i$ , and and the fluid thickness,  $h_i$ , in the ridge (see figure 4-Left).

A general  $2 \times 2$  system of conservation laws (7) is hyperbolic if the Jacobian matrix  $J(u, v) = \begin{pmatrix} F_u & F_v \\ G_u & G_v \end{pmatrix}$  has two real distinct eigenvalues  $\lambda_1(u, v)$  and  $\lambda_2(u, v)$ , with eigenvectors  $r_1, r_2$ . The problem is called genuinely nonlinear if  $r_i \cdot \nabla \lambda_i \neq 0$ , where the gradient is with respect to the variables u and v. For a large range of relevant physical parameters in (5-6), both conditions are satisfied provided we include the wall function w(h). Setting w(h) = 1 results in a change in hyperbolicity of the equations (5-6) at small h.

Under the assumptions of hyperbolicity and genuine nonlinearity, there is a methodology for determining the



FIG. 3: Full and reduced model numerical profiles for  $\phi_0 = 30\%$ . (Top) Comparison of height profiles of the full system (3-4) with the reduced system (5-6). (Bottom) Comparison of concentration profiles of the full system (3-4) with the reduced system (5-6). Profiles are plotted at 40 sec intervals.

intermediate values  $u_i$  and  $v_i$  in terms of the left and right states. Thus, for a fixed concentration in the reservoir and upstream film thickness, given the particle concentration and thickness of the precursor film, we can find the height, concentration and propagation speed of the ridge. A one parameter family of shock solutions, connecting  $(u_i, v_i)$  to  $(u_l, v_l)$  or  $(u_r, v_r)$  can be determined by recognizing that the weak form of (7) yields two Rankine-Hugoniot conditions for the shock speed both of which have to be satisfied. For the 1-shock, connecting  $(u_l, v_l)$ to  $(u_i, v_i)$ , we have

$$s_1 = \frac{F(u_i, v_i) - F(u_l, v_l)}{u_i - u_l} = \frac{G(u_i, v_i) - G(u_l, v_l)}{v_i - v_l} \quad (8)$$

which gives two equations in three unknowns. Analogous formulas hold for the 2-shock connecting  $(u_i, v_i)$  to  $(u_r, v_r)$ . This gives a total of four equations and four unknowns, namely  $s_1, s_2, u_i$ , and  $v_i$ . Generically there will be a locally unique solution, however these equations are also accompanied by entropy conditions which restrict whether a 1-shock or 2-shock is an admissible connection [21]. In our case, the entropy condition is satisfied and the shock connection problem can be solved numerically, providing an implicit formula for determining the intermediate state  $(u_i, v_i)$  that emerges from the Riemann initial data.

Numerical simulations show that the reduced model (5-6) captures the the large scale dynamics of the full equations (3-4), including the shock speeds  $s_1$  and  $s_2$  and ridge height  $h_i$  and ridge particle concentration  $\phi_i$ . Fig-



FIG. 4: (Left) Schematic diagram of the connection problem. (Right) Shock speeds calculated via the connection problem as a function of precursor thickness for  $\phi_0 = 15\%$  (filled symbols) and  $\phi_0 = 30\%$  (open symbols).

	Full Model	Reduced Model	Shock Theory
$h_i$	1.150	1.162	1.1619
$\phi_i$	0.359	0.362	0.3620
$s_1$	0.435	0.434	0.4556
$s_2$	0.498	0.493	0.4956

TABLE I: Comparison of shock speeds and intermediate states in model simulations and shock theory. Data shown for  $\phi_0 = 0.30, b = 0.1$ . Quantitative experimental data is not presented here owing to the sensitive dependence on precursor film as discussed in the text.

ure 3 shows a sample calculation with parameters m = 5,  $\phi_{max} = 0.67, \ \phi_0 = 0.3, \ b = 0.1, \ \beta = 90^\circ, \ \text{and} \ A = 0.5.$ The large spike at the leading edge of the full system solution corresponds to the capillary ridge due to surface tension, which is neglected in the the reduced system. In both solutions, an intermediate state appears connecting the left and right states via shocks. The trailing shock in figure 3 is smeared out due to numerical diffusion. The full lubrication theory, reduced model and shock connection problem all consistently predict the formation of a 4

large particle-rich ridge at high inclination angles and high concentrations as observed in the experimental data. The numerically observed values in the full and reduced model are compared with the analytic calculations from the connection problem in table I. All three models display a sensitive dependence on the precursor thickness making quantitative comparison with experimental data challenging. This presents a marked contrast to the clear fluid case in which the dependence on precursor is logarithmic. This sensitivity is illustrated in the shock speeds shown in figure 4-Right and is an interesting open question for future studies.

The strong influence of the precursor on the flow structure underscores the need for more detailed experimental measurements. Specifically, quantitative comparison between theory and experiment requires precise measurements of the precursor thickness and shock separation over time. Both of these are subjects of current study.

Shock theory for systems of conservation laws is well known in gas dynamics but has never before been applied to the physics of contact lines. This new theory suggests many avenues for further study including the dependence of the dynamics on contact line physics, and the extension of the theory to the transverse direction to address the development of fingers, which appear to be somewhat suppressed in the experiment by the formation of the particle-rich ridge.

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