Models of non-Newtonian Hele-Shaw flow

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We study the Saffman-Taylor instability of a non-Newtonian fluid in a Hele-Shaw cell. Using a fluid model with shear-rate dependent viscosity, we derive a Darcy's law whose viscosity depends upon the squared pressure gradient. This yields a natural, nonlinear boundary value problem for the pressure. A model proposed recently by Bonn *et al.* [Phys. Rev. Lett. **75**, 2132 (1995)] follows from this modified law. For a shear-thinning liquid, our derivation shows strong constraints upon the fluid viscosity— strong shear-thinning does not allow the construction of a unique Darcy's law, and is related to the appearance of slip layers in the flow. For a weakly shear-thinning liquid, we calculate corrections to the Newtonian instability of an expanding bubble in a radial cell. [S1063-651X(96)51611-9]

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One reason for the enduring interest in Newtonian fluid flow in Hele-Shaw cells is its close analogy to quasistatic solidification. The Saffman-Taylor (ST) instability of the driven fluid-fluid interface plays the same role as the Mullins-Sekerka instability of the solidification front [1]. Features usually associated with solidification, such as the growth of stable dendritic fingers and sidebranching, have also been observed in fluids with an imposed anisotropy, say by scoring lines on the plates of the cell [2]. However, experiments using non-Newtonian or anisotropic fluids, such as liquid crystals, have shown that ''solidification'' structures can be induced by the bulk properties of the fluid itself [3–5]. The precise mechanisms of generating such dendritic fingers with stable tips are unknown. One of our interests is in liquid crystal flows, which are characterized by complicated hydrodynamics [6]. We conjecture that stable tip propagation in these materials is a consequence of shear thinning associated with flow induced realignment of the liquid crystal director. In this paper we focus on this single property, and consider an expanding gas bubble in a radial Hele-Shaw cell containing a shear-thinning liquid.

In recent work on polymeric fluids, Bonn and co-workers [7] proposed modeling the Hele-Shaw flow of a non-Newtonian fluid by positing the modified Darcy's law

$$\mathbf{u}=-\frac{b^2}{12\mu(|\mathbf{u}|^2/b^2)}\nabla p, \quad \nabla\cdot\mathbf{u}=0, \tag{1}$$

where $\mathbf{u}$ is the velocity, b is the gap width, p is the pressure, and the viscosity μ depends on the shear rate ω ($|\omega|\approx|\mathbf{u}|/b$): $\mu(\omega^2)=\mu_0(1+\alpha\omega^2\tau^2)/(1+\omega^2\tau^2)$, $0<\alpha\leqslant 1$. For $\alpha<1$, μ is a decreasing function of ω^2, and the fluid is shear-thinning with asymptotic viscosities μ_0 and $\mu_\infty=\alpha\mu_0$. Here τ is a single characteristic relaxation time of the polymer.

A similar approach is found in the injection molding literature [8]. In this derivation, velocity and viscosity are averaged over the transverse direction, giving rise to equations similar to Eq. (1), but taking into full account the viscosity dependence on the transverse shear.

In this Rapid Communication, we give several results. We derive the modified Darcy's law from first principles, using a simple non-Newtonian fluid model. We show further that Eq. (1) follows from a more basic version of Darcy's law, where the viscosity depends instead upon the squared pressure gradient. This law gives a natural boundary value problem (BVP) for the pressure, as in the Newtonian case. A result of particular interest is that for a shear-thinning fluid, it is not always possible to define a unique viscosity in the Darcy's law. This is associated with the appearance of negative effective viscosities in the non-Newtonian Navier-Stokes equation, and the possibility of solutions with discontinuous shear. Such solutions may give a basis for interpreting observed ''spurt'' behavior in liquid crystal flows [9]. For a shear-thickening liquid, the viscosity in Darcy's law is unique, but the BVP can change from elliptic to hyperbolic. In other systems this is associated with the appearance of shock waves. For a weakly shear-thinning liquid, we calculate corrections to the Newtonian ST instability of an expanding bubble in a radial cell. Finally, we note some discrepancies between our results and those of Bonn and coworkers.

Derivation and constraints. The simplest model of an incompressible, isotropic but non-Newtonian fluid is given by the generalized three-dimensional (3D) Navier-Stokes equations:

$$\rho\frac{D\mathbf{v}}{Dt}=-\nabla p+\nabla\cdot(\mu(|\mathbf{S}|^2)\mathbf{S}), \quad \nabla\cdot\mathbf{v}=0, \tag{2}$$

where $\mathbf{S}$ is the rate-of-strain tensor, and $|\mathbf{S}|^2=\mathrm{tr}(\mathbf{S}^2)=\Sigma_{ij}S_{ij}^2$ [(μ can also depend on det($\mathbf{S}$), but this is negligible in Hele-Shaw flow] [10]. We choose z to be the coordinate across the gap, and x and y as the lateral coordinates. No-slip is assumed on the plates: $(u,v,w)|_{z=\pm b/2}=0$.

There is an apparent limitation on applying this model. Consider simple Poiseuille flow $\mathbf{v}=(u(z,t),0,0)$, driven by a constant pressure gradient a. For unit density, Eq. (2) reduces to

$$u_t=-a+(\mu(u_z^2)u_z)_z=-a+\eta(u_z^2)u_{zz}, \tag{3}$$

where the *effective viscosity* $\eta(\zeta)=\mu(\zeta)+2\zeta\mu'(\zeta)$ is the coefficient of u_{zz}. If in a shear-thinning liquid $\mu(\zeta)$ decreases rapidly enough, then η is negative for some range of $\zeta>0$. The condition $\eta(\zeta)>0$, necessary for classical well-posedness, arises naturally in our derivation below. If $\eta(\zeta)<0$ in some range, the existence and nature of solutions is a fascinating question, intimately related to the appearance of slip layers in the flow. One well-grounded approach to such a situation is to consider additional (regularizing) physics, a point to which we will return.

In the small gap limit, the Reynolds number is small and inertial terms can be neglected:

$$\nabla p-\nabla\cdot(\mu(|\mathbf{S}|^2)\mathbf{S})=0, \quad \nabla\cdot\mathbf{v}=0. \tag{4}$$

These Stokes equations are further simplified by making use of the large aspect ratio of the cell. Let L be some lateral length scale, rescale the coordinates by $x=Lx'$, $y=Ly'$, and $z=bz'$, and let $\epsilon=b/L\ll 1$. Velocities are scaled correspondingly. We note that $|\mathbf{S}'|^2=\epsilon^{-1}\mathbf{u}'_{z'}\cdot\mathbf{u}'_{z'}+O(1,\epsilon)$, where $\mathbf{u}=(u,v)$. Retaining only lowest order terms yields reduced Stokes equations

$$\nabla_2 p-\partial_z[\mu(|\mathbf{u}_z|^2)\mathbf{u}_z]=0, \quad \partial_z p=0, \tag{5}$$

where $\nabla_2=(\partial_x,\partial_y)$. These equations, with a ''power-law'' viscosity $\mu(\zeta)=M(\zeta/\zeta_o)^{(n-1)/2}$ $(0<n<2)$, have been used to study the linear stability of interfaces in the channel and radial geometries [11].

We continue without further approximation. As p depends only upon x and y, Eq. (5) integrates to

$$z\nabla_2 p=\mu(|\mathbf{u}_z|^2)\mathbf{u}_z. \tag{6}$$

For simplicity, we consider velocity profiles symmetric about $z=0$. We wish to find $\mathbf{u}_z$ as a function of $\nabla_2 p$, as in the usual Darcy's law. Squaring Eq. (6) gives an implicit equation for $|\mathbf{u}_z|^2$ in terms of $z^2|\nabla_2 p|^2$. The invertibility of this equation, or lack thereof, is a central issue. For $f(\zeta)=\zeta\mu^2(\zeta)$, $f(0)=0$ and $f'(0)=\mu^2(0)>0$, and so there is local invertibility around $\zeta=0$. A sufficient condition for finding $\zeta=|\mathbf{u}_z|^2$ *uniquely* in terms of $z^2|\nabla_2 p|^2$ is that $f'(\zeta)>0$, implying that

$$\eta(\zeta)=\mu+2\zeta\mu'(\zeta)>0, \tag{7}$$

for all $\zeta>0$. This is the condition of having positive effective viscosity in Eq. (3). For the power-law or a shear-thickening viscosity μ, this inequality is always satisfied. For a general shear-thinning liquid this constraint will not be satisfied for every μ. For example, if $\mu(\zeta)=\mu_0(1+\alpha\zeta/\zeta_o)/(1+\zeta/\zeta_o)$, with $0<\alpha\leqslant 1$, then μ satisfies inequality (7) only for $\alpha>1/9$. Figure 1 shows $f(\zeta)$ for $\alpha=1/2$ and 1/20. Assuming that inequality (7) holds, then Eq. (6) can be inverted uniquely, to give

$$\mathbf{u}_z=\frac{z\nabla_2 p}{\tilde{\mu}(z^2|\nabla_2 p|^2)}, \quad \text{or} \quad \mathbf{u}=\int_{-b/2}^{z}dz'\,\frac{z'\nabla_2 p}{\tilde{\mu}(z'^2|\nabla_2 p|^2)}. \tag{8}$$

Here, if μ is a strictly decreasing or strictly increasing function of its argument, then so is $\tilde{\mu}$. The gap-averaged velocity

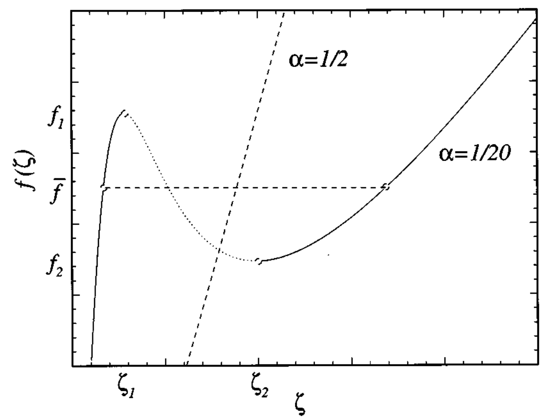

FIG. 1. $f(\zeta)$ for $\alpha=1/2$ (dashed) and 1/20 (solid).

is $\overline{\mathbf{u}}(x,y)=(1/b)\int_{-b/2}^{b/2}dz\ \mathbf{u}(x,y,z)$. Gap averaging Eq. (8) and the divergence-free condition yields

$$\overline{\mathbf{u}}=\frac{-b^2}{12\overline{\mu}(|\nabla_2 p|^2)}\nabla_2 p, \quad \text{and} \quad \nabla_2\cdot\overline{\mathbf{u}}=0 \tag{9}$$

where $1/\overline{\mu}=(12/b^3)\int_{-b/2}^{b/2}dz[z^2/\tilde{\mu}(z^2|\nabla_2 p|^2)]$. The subscript on ∇ and bar on $\mathbf{u}$ are now dropped. Equation (9) is our first result. Figure 2 shows $\overline{\mu}(|\nabla p|^2)$, for various α's, including the limiting value of 1/9.

Equation (1) used by Bonn and co-workers now follows by squaring Eq. (9), and expressing $|\nabla p|^2$ in terms of $|\mathbf{u}|^2$. In this case, the functional form is such that this inversion can always be accomplished. We consider Eq. (9) to be the more natural form of the flow equations; it leads to a BVP for p, as in the Newtonian case.

We have thus derived a non-Newtonian Darcy's law for the bulk fluid. Now consider a finite patch of fluid, denoted by Ω with boundary Γ, surrounded by a gas at uniform pressure. Then p must solve the nonlinear BVP,

$$\nabla\cdot\left(\frac{1}{\overline{\mu}(|\nabla p|^2)}\nabla p\right)=0 \ \text{ in } \Omega, \quad p|_\Gamma=-\gamma\kappa, \tag{10}$$

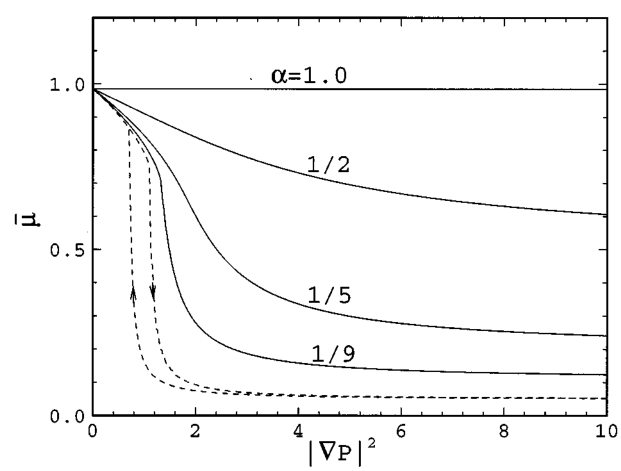

FIG. 2. The effective viscosity $\overline{\mu}$ for various α. Possible hysteretic behavior in $\overline{\mu}$ for $\alpha=1/20$ (dashed).

where for simplicity the traditional Laplace-Young condition at Γ has been taken, with γ the surface tension parameter and κ the curvature of Γ. Nonlinear BVPs of this form arise in many other physical contexts, such as gas dynamics [12] and elasticity [13].

If $\overline{\mu}$ is constant, then p is harmonic, and the BVP (10) can be solved by boundary integral methods [14]. For $\overline{\mu}$ not a constant, the shear-thinning and shear-thickening problems must be considered separately.

Shear thinning. We assume that $\overline{\mu(\zeta)}$ is monotonically decreasing, $0<\overline{\mu(\infty)}\leqslant\overline{\mu(0)}<\infty$. Then the BVP (10) can be interpreted as the Euler-Lagrange equation to find the minimizer of

$$\mathcal{F}[p]=\int_\Omega F(|\nabla p|)dA, \text{ with } p|_\Gamma=-\gamma\kappa, \tag{11}$$

where $F(s)=\int_0^{s^2}ds's'/\overline{\mu(s'^2)}$. Then, $F''(s)=[\overline{\mu(s^2)}-2s^2\overline{\mu'}(s^2)]/\overline{\mu(s^2)}^2>0$. And so, $F(s)$ is monotonic and convex. Under these conditions, the BVP (10) is strongly elliptic and has a unique solution [15].

Shear thickening. We assume now that $\overline{\mu(\zeta)}$ is monotonically increasing with $0<\overline{\mu(0)}\leqslant\overline{\mu(\infty)}<\infty$. The corresponding variational problem now has an integral density $F(s)$ (still monotonically increasing) that can possibly lose convexity. Loss of convexity is associated with a transition from elliptic to hyperbolic behavior, which, in the compressible flow context, is associated with the appearance of shock waves [12]. In the context of Hele-Shaw flow, it is unclear to us what would be the physical manifestation of such a transition.

The flow problem. The flow problem we consider is that of an expanding gas bubble in a radial Hele-Shaw cell. The typical scenario for the development of the ST instability is the appearance of growing petals, whose radius of curvature increases until it is comparable to the wavelength of an unstable mode. The petal tip then splits, engendering new petals, and the process continues, giving rise to a dense branching morphology [16]. We expect qualitative differences in shear-thinning fluids, and conjecture that shear-thinning prevents the growth of the radius of curvature, preventing splitting and stabilizing the tips. Such behavior has been observed both in liquid crystals [4,5] and in polymer solutions [3].

The flow problem requires solving the BVP (10) for the pressure. The interface Γ must move with the fluid: If $\mathbf{V}$ is the velocity of Γ, then $\mathbf{n}\cdot\mathbf{V}=\mathbf{n}\cdot\mathbf{u}|_\Gamma$, where $\mathbf{n}$ is the normal to the interface. The fluid domain Ω (outside of the gas bubble) is unbounded, with a finite mass flux at infinity. The boundary conditions on p then become

$$p|_\Gamma=\gamma\kappa, \quad p\to -A\ \ln r \text{ as } r=\sqrt{x^2+y^2}\to\infty. \tag{12}$$

Here $A=6\mu_0 S_t/\pi b^2$, and S_t is the rate of bubble area increase. A unique solution still exists to the BVP with unbounded Ω [19].

The flow problem simplifies in the limit of weak shear thinning ($\alpha=1-\delta$, $\delta\ll1$), and we can calculate corrections there to the Newtonian ST instability [5]. For a bubble of radius $R(t)$, the instantaneous growth rate σ of an azimuthal disturbance with wave number $m>1$ is

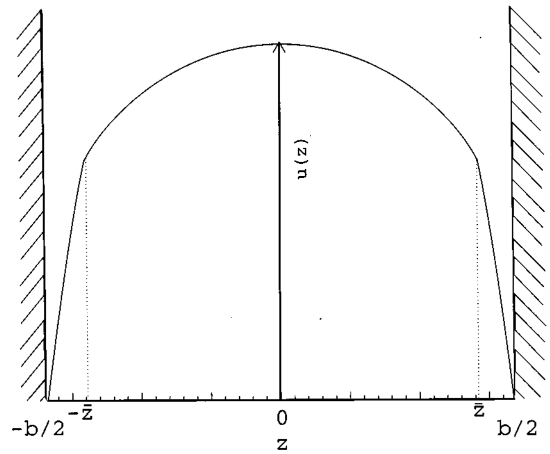

FIG. 3. A possible steady-state solution with a slip layer at the walls.

$$\sigma=\frac{\dot R}{R}\left[-2+m\left(1+C\delta\frac{m-1}{m+1}\right)\right]+\gamma\frac{b^2}{12\mu_0R^3}m(1-m^2)\left(1+C\delta\frac{2m}{m+1}\right), \tag{13}$$

where $C>0$ is a constant. Shear thinning both increases instability due to driving (the first term), and increases the stabilization due to surface tension (the second term). The net effect, at small surface tension, is to *decrease* the band of unstable modes, and *increase* the growth rate of the most unstable mode. (This is consistent with numerical calculations for general α, the details of which will be presented elsewhere.) Perhaps this enhanced wavelength selection is related to the formation of stable tips in shear-thinning fluids.

Negative effective viscosity and loss of uniqueness. We noted that loss of unique invertibility in Eq. (6) was equivalent to the appearance of a region of negative η in Eq. (3). Figure 1 shows $\zeta=|\mathbf{u}_z|^2$ versus $f(\zeta)=\zeta\mu^2(\zeta)$, for $\alpha=1/20$ (the solid curve). Since $f'(\zeta)=\mu(\zeta)\eta(\zeta)$, there is a range (ζ_1,ζ_2) where $\eta<0$. For small pressure gradient, or $f<f_2$ for $|z|\leqslant b/2$, there is a unique symmetric shear flow solution with positive η. However, as the pressure gradient is increased (''ramped up''), f can become greater than f_2, and negative η is possible.

While the general initial value problem is then ill-posed, formal steady states can be constructed which join solutions in the positive diffusion regions, under the constraints of continuous velocity and stress. One such solution is shown in Fig. 3, where the wall region has a ''slip layer'' arising from the velocity gradient at the wall jumping to the lower viscosity branch. Such slip layers may already have been observed [17]. The jump is shown by the dashed line at $\overline{f}$ in Fig. 1. Since f is the squared viscous stress, such a horizontal jump corresponds to its continuity. In this construction, however, $\overline{f}=\overline{z}^2|\nabla p|^2$ is not fixed, and so neither is the point of matching $\overline{z}$. We suspect that in the steady state $\overline{f}$ might be given by the Maxwell construction, where the areas between the curves of $\sqrt{\overline{f}}$ and $\sqrt{f(\zeta)}$, as a function of ζ, are equal.

Slip layers have been discussed by Malkus, Nohel, and Plohr [18] who consider a Johnson-Segalman-Oldroyd fluid, rather than one described by Eq. (2). Their set of constructed steady states is the same, but apparently without the problem of possible ill-posedness. They use dynamical systems tools to show that in the low Reynolds number limit, the extra stress equations pick the matching point $\overline{z}$ dynamically by fixed point stability. They show further that the system exhibits hysteretic behavior, but that for a pressure gradient being (slowly) ramped up, the selected $\overline{z}$ corresponds to $f=f_1$.

Such an approach would allow the construction of shear flows, characterized by a pressure gradient, in situations where our model Eq. (2) may be inappropriate. However, in constructing the viscosity $\overline{\mu}$, the possibility of hysteretic behavior would have to be allowed. A possible hysteresis loop in $\overline{\mu}$, for $\alpha=1/20$, is shown in Fig. 2.

In summary, for fluids whose viscosity depends on shear rate, we derive a Darcy's law whose gap-averaged viscosity depends upon the pressure gradient. Linear stability analysis suggests that one effect of shear thinning may be the stabilization of tips. Our derivation assumes positivity of an effective viscosity. Loss of positivity is associated with slip layers appearing in the flow, and nonuniqueness of the gap-averaged viscosity.

Bonn and co-workers argue that p is essentially harmonic. They apply the divergence free condition to Eq. (1) to find

$$\mu\nabla^2 p-\mu'\,\boldsymbol{\nabla}(\mathbf{u}\cdot\mathbf{u})\cdot\boldsymbol{\nabla} p=0.$$

As $\boldsymbol{\nabla}(\mathbf{u}\cdot\mathbf{u})$ scales like the convective derivative in the Navier-Stokes equation, they argue that it can be neglected. Unfortunately, smallness of the Reynolds number reduces the Navier-Stokes equations to the Stokes equations, but it is not available for such duty afterwards. Bonn and co-workers use the assumption of harmonicity to find the interface velocity. The exact non-Newtonian pressure can be computed for the circular expanding bubble, to find

$$\nabla^2 p=\frac{-24\dot{R}\mu_0}{b^2RD^2[1+(r/DR)^2]^2}(1-\alpha),\qquad D=\frac{\tau\dot{R}}{b}.$$

If $\alpha=1$ (constant viscosity), the pressure is harmonic; but for many shear-thinning fluids, $\alpha\ll 1$ (as in Bonn and co-workers). One can show that using $\nabla^2 p=0$, for a circular bubble, gives $O(1)$ relative errors in the interface velocity. These errors might be smaller in a channel geometry, as considered by Bonn and co-workers.

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