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Shear banding in polymer solutions

Michael Cromer,^{1,2} Michael C. Villet,³ Glenn H. Fredrickson,^{1,2,4}
 and L. Gary Leal^{1,4,5}

¹*Department of Chemical Engineering, University of California, Santa Barbara, California 93106, USA*

²*Materials Research Laboratory, University of California, Santa Barbara, California 93106, USA*

³*DSM Research, 6160 MD Geleen, The Netherlands*

⁴*Department of Materials, University of California, Santa Barbara, California 93106, USA*

⁵*Department of Mechanical Engineering, University of California, Santa Barbara, California 93106, USA*

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The current theoretical belief is that the steady-state shear banding in viscoelastic liquids requires a non-monotonic constitutive relationship between shear stress and shear rate. Although existing rheological studies conclude that the constitutive equation for entangled polymers is monotonic, recent experimental evidence suggests shear banding can nevertheless occur in polymer solutions. In this work, we predict, for the first time, steady state shear banding with a realistic monotonic constitutive theory for polymeric liquids. The key is that a proper account must be taken of the coupling of polymer stress to polymer concentration. We also predict multiple steady states at some shear rates as seen experimentally, with shear banding if the flow is ramped quickly enough from rest, but homogeneous linear shear flow otherwise.
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In this paper, we address an outstanding fundamental theoretical question concerning the steady shear flow behavior of polymer solutions: Are steady state shear banded velocity profiles possible, given a constitutive model in which the relationship between the shear stress and the shear rate is monotonic? A related issue is the validity of the assumption of a homogeneous fluid that is common to nearly all studies of flow in polymeric liquids.

Recent experimental studies exhibit what seems to be evidence of steady state shear banding in highly entangled polybutadiene solutions¹ and in DNA as a model monodisperse polymer solution.² Furthermore, these studies also suggest that there may be conditions in which one can realize either a linear velocity profile or a shear banded profile, starting from a rest state, depending on the rate at which the flow device is ramped up to the final nominal shear rate. In spite of these observations, however, the debate over whether a steady inhomogeneous shear banded state can exist for polymeric fluids is still very much ongoing.^{3–5}

From a theoretical perspective, it is known that a non-monotonic constitutive curve is linearly unstable,⁶ and that this instability results in an inhomogeneous, banded velocity profile in shear. However, there is no rheological evidence that polymer solutions can exhibit non-monotonic constitutive behavior. As a consequence, it has been suggested that experimental observations of “shear banding” must either represent a long-time transient, or be a consequence of the curvature of the velocity profile when the observations are made in Couette devices.^{7,8} In this paper, we demonstrate that shear banding can be predicted with a monotonic constitutive equation.

The key is to recognize the possibility that the polymer concentration can become inhomogeneous in a flow. Existing theories typically assume that polymer concentration remains uniform in flow. However, it was shown 20 years ago by Helfand and Fredrickson⁹ that there is a mechanism for polymer solutions, involving coupling between stress and concentration, that can drive concentration

non-uniformities in flow. We shall see that if this mechanism is taken into account, shear banding can be predicted using a realistic monotonic constitutive theory.

It was shown almost two decades ago that simple shear flow can be linearly unstable even with a monotonic constitutive curve¹⁰ if polymer concentration is coupled to stress. However, this study was largely discredited because it assumed that the osmotic pressure was a function of shear rate,¹¹ and neglected the Helfand-Fredrickson (HF) mechanism. Later, both experimental and theoretical studies^{12–16} explored the coupling between stress and concentration in the context of worm-like micelles. Specifically, detailed linear stability analysis was carried out using a diffusive (“non-local”) version of the Johnson-Segalman (J-S) model coupled to concentration through the Helfand-Fredrickson mechanism, which showed that it is possible for regions of increasing shear stress to be linearly unstable.^{14,15} The coupling to concentration serves to enhance the mechanical instability in the regions of decreasing stress while broadening the instability window to include portions of the curve where the overall shear stress is increasing. These latter regions were attributed to a shear-induced demixing (SID) instability as opposed to a mechanical instability.¹⁵ It has been shown numerically¹⁶ and experimentally¹² that this SID instability leads to inhomogeneous flow.

Although the work of Olmsted and Fielding¹⁶ thus indicates that shear banding can be achieved from a monotonic constitutive curve, there are a number of issues that make the relevance of these predictions questionable for shear banding in polymeric liquids. First, with the parameter values used, the standard J-S model predicts non-monotonic dependence of stress on shear rate. To produce a monotonic dependence, the authors add a Newtonian contribution to the stress in addition to the contribution of the solvent that they identify as due to the fast Rouse modes, which decreases with micelle concentration ϕ more slowly than the modulus $G(\phi)$ of the standard model. Hence for low enough concentrations, the combined equation for the stress is monotonic, but the underlying direct J-S contribution to the stress is always non-monotonic.¹⁶ Second, and more generally, the Johnson-Segalman model is known to provide unrealistic rheological predictions for entangled polymeric solutions.¹⁷ Furthermore, if the model parameters are chosen to produce a non-monotonic stress, as in the work cited above, a major consequence is that it is necessary to introduce ad hoc stress diffusion terms into the J-S constitutive equations to produce a unique, stable, shear-banded solution.¹⁸ The fact that stress diffusion is required to obtain a unique steady banded solution even with stress-concentration coupling,¹⁶ is evidence that the concentration coupling is modifying the underlying constitutive instability, rather than causing the instability of the flow. Finally, existing experimental evidence suggests that the constitutive curve for polymer solutions is monotonic at all concentrations. Thus, it remains to be established¹¹ whether a flow-concentration instability can ultimately lead to a unique steady shear-banded flow within the standard model of inhomogeneous polymer solution dynamics,^{9,19–24} using a realistic and well-established (monotonic) constitutive theory, and without the need for “ad hoc” constructs such as the stress diffusion terms in the J-S model.

In this paper, we address this issue. We show that all of the experimental observations of shear banding by Wang *et al.* can be qualitatively reproduced, including the existence of multiple steady states seen for different start-up conditions. To do so, we have investigated shear flow using the well-known Rolie-Poly (RP) constitutive model, with concentration/stress coupling incorporated through a two-fluid model approach pioneered by earlier studies,^{19–24} in which the polymer and the solvent are considered as two interpenetrating fluids. In the following, we briefly introduce the model, but refer the reader to the above references for further details.

The two-fluid model consists of two sets of equations, expressing conservation of mass and momentum for the solvent and polymer, treated as two interpenetrating continua. In the following, dimensional variables are denoted with a prime. The two fluids are coupled by a phenomenological friction force, which has traditionally been assumed of the form $\zeta'(\phi)(\mathbf{v}'_s - \mathbf{v}'_p)$, where ζ' is a friction coefficient and ϕ is the local volume fraction of polymer. In addition to friction, the Newtonian solvent is acted upon by viscous stress with viscosity μ_s and by the hydrostatic pressure, while the polymer is acted upon by the polymer stress σ'_p and by the osmotic pressure π'_ϕ . In the analysis that follows, we assume that the mean volume fraction of polymer, ϕ_0 , is small enough that the solvent velocity can be approximated by the overall solution velocity $\mathbf{v}' \approx \mathbf{v}'_s$. In addition, we assume that both the solvent and polymer inertia are negligible.

The model equations presented below are in non-dimensional form. The concentration, ϕ , is normalized by the mean polymer volume fraction, ϕ_0 . As a characteristic length, we use the intrinsic length scale $\ell_c = \sqrt{D\tau_{d0}}$ at which concentration relaxation by diffusion and stress relaxation by reptation occur on the same time scale. Here $D = \phi_0^2/\chi_0\zeta_0$ is the cooperative diffusion coefficient, with χ_0 the osmotic susceptibility and τ_{d0} and ζ_0 , the reptation time and friction coefficient at the mean polymer concentration ϕ_0 . We scale time by τ_{d0} and velocities by the ratio ℓ_c/τ_{d0} . Material parameters that depend on ϕ are non-dimensionalized using their values at ϕ_0 .

With polymer inertia neglected, the momentum equation for the polymer can be expressed as $\mathbf{v}_p = \mathbf{v} - \zeta^{-1}(\nabla\pi_\phi - E\nabla\cdot\boldsymbol{\sigma}_p)$, where E is the ratio of elastic to osmotic stress, $E = \chi_0 G_0/\phi_0^2$, with $G_0 \equiv G'(\phi_0)$ the plateau modulus. The remaining equations governing conservation of polymer mass and conservation of solution (solvent) mass and momentum are

$$\frac{\partial\phi}{\partial t} = -\mathbf{v}\cdot\nabla\phi + \nabla\cdot\left[\frac{\phi}{\zeta}(\nabla\pi_\phi - E\nabla\cdot\boldsymbol{\sigma}_p)\right], \quad (1)$$

$$\varpi\nabla^2\mathbf{v} + \mathbf{T}\cdot(\nabla\cdot\boldsymbol{\sigma}_p) = 0, \quad (2)$$

where \mathbf{T} is the transverse projection operator to enforce incompressible flow, and the ratio of solvent to polymer viscosity is denoted as $\varpi = \mu_s/G_0\tau_{d0}$.

We obtain the osmotic pressure π_ϕ from a square gradient approximation for the mixing free energy:

$$F_{\text{mix}}[\phi] = \int d\mathbf{r} \frac{1}{2} (\phi^2 + \xi^2 |\nabla\phi|^2) \equiv \int d\mathbf{r} f_{\text{mix}}(\mathbf{r}), \quad (3)$$

where $\xi = \xi'/\ell_c$ is the dimensionless polymer correlation length, which is a function of the local concentration. The osmotic pressure is then defined as $\pi_\phi = \phi\delta F_{\text{mix}}/\delta\phi - f_{\text{mix}}$.

Closure requires a constitutive model for the polymer stress $\boldsymbol{\sigma}_p$. We use a simplified Rolie-Poly constitutive equation²⁵ in which the polymer configuration is specified by a configuration tensor $\mathbf{Q} = 3\langle\mathbf{R}\mathbf{R}\rangle/R_0^2$, where \mathbf{R} is the chain end-to-end vector and R_0 is the equilibrium chain length, and the non-dimensionalized stress is related to \mathbf{Q} according to $\boldsymbol{\sigma}_p = G(\phi)(\mathbf{Q} - \mathbf{I})$ in which $G(\phi)$ is the scaled plateau modulus, G'/G_0 , that depends on ϕ . The polymer configuration evolves according to the Rolie-Poly equation²⁵

$$\overset{\nabla}{\mathbf{Q}} = \tau_d^{-1}(\phi)(\mathbf{I} - \mathbf{Q}) - 2\theta(1 - \lambda^{-1})[\mathbf{Q} + \lambda^{-1}(\mathbf{Q} - \mathbf{I})], \quad (4)$$

where $\lambda = \langle R^2 \rangle/R_0 = \sqrt{\text{tr}\mathbf{Q}/3}$ is a stretch parameter, $\theta \equiv \tau_{d0}/\tau_R$, where τ_R is the Rouse time, and we assume that polymer stress is convected by the polymer velocity according to

$$\overset{\nabla}{\mathbf{Q}} = \frac{\partial\mathbf{Q}}{\partial t} + \mathbf{v}_p\cdot\nabla\mathbf{Q} - (\nabla\mathbf{v}_p)^T\cdot\mathbf{Q} - \mathbf{Q}\cdot\nabla\mathbf{v}_p. \quad (5)$$

This model includes relaxation due to reptation, convective constraint release, contour length fluctuations, and chain stretch, and in the terminology of the general model²⁵ we have set $\beta = 1$ and $\delta = -1/2$, which means that the shear stress-shear rate relationship is strictly monotonic. We further assume a power-law dependence of the dimensionless material functions $\tau_d(\phi) \sim \phi^{3/2}$, $G(\phi) \sim \phi^{9/4} \equiv \phi^\alpha$, $\xi \sim \phi^{-3/4}$, and $\zeta \sim \phi^{3/2}$, where we have used theoretical exponents for a good solvent. Finally, in considering shear flow, the scaled shear rate is given by the Weissenberg number, $\text{Wi} \equiv \dot{\gamma}\tau_{d0}$, which will appear in the magnitude of the boundary velocity.

The Rolie-Poly model is chosen for its ability to accurately capture the shear rheology of polymer solutions for a wide range of strain rates.²⁵ For slow flow, the RP model predicts the expected Newtonian flow response: stress proportional to strain rate. As the separation of reptation and Rouse time scales increases, the model predicts a broader and flatter plateau region corresponding to shear thinning. However, we emphasize that in the form used here, the RP model has a strictly monotonic constitutive relationship between shear stress and shear rate, i.e., $\partial\sigma_{xy}/\partial\dot{\gamma} > 0$.^{25,26}

We now show that this polymer solution model exhibits steady-state shear banding. One preliminary result, which we do not reproduce here due to lack of space, is that a linear shear flow with a homogeneous concentration distribution is linearly unstable for a range of Weissenberg numbers

at each θ above a minimum relaxation time ratio that depends on the parameter E . In agreement with previous results,¹⁵ the instability is driven by polymer migration *up* elastic stress gradients towards regions of higher stress (larger polymer volume fraction), in opposition of diffusion. Of course, it is not obvious that a linear instability will necessarily lead to shear banding. We therefore investigate how this linear instability will manifest itself in the nonlinear equations, by solving the full system in the flow generated between infinite parallel plates in which the bottom plate is held fixed and the upper plate is moved from rest up to a constant velocity in a finite time period.

Unlike most studies, we have used the intrinsic length scale $\ell_c = \sqrt{D\tau_{d0}}$ rather than the channel width H' to non-dimensionalize spatial variables. We denote the dimensionless channel width by $H = H'/\ell_c$. We expect the transition in the shear banded velocity profile to occur over a dimensionless distance of $O(1)$. For many common experimental situations, one would expect $\ell_c = O(\mu\text{m})$ and $H' \leq O(\text{mm})$, thus H can be rather large, making spatial resolution difficult in numerical simulations. As our objective is to demonstrate the existence of shear banding, for ease of simulation we use a very small (likely non-physical) value $H = 5$ and choose the following parameter set, for which the base flow has a window of instability: $E = 0.33$, $\xi_0 = 0.3$, $\theta = 10^3$, $\varpi = 0.0004$. To consider the effects of how the upper plate is ramped up to the desired velocity from rest, a hyperbolic tangent function is used to control how quickly the wall velocity gets up to speed. In particular, the velocities at the two walls are given, via the no-slip condition, as $v|_{y=0} = 0$, $v|_{y=H} = H \text{Wi} \tanh(at)$. The ramp speed parameter, a , is introduced to control the wall acceleration. For $a = 1$, the final steady wall velocity is obtained in a dimensionless time of $O(1)$, i.e., a dimensional time scale of $O(\tau_d)$. For example, for $a \ll 1$, the time it takes for the wall velocity to attain the desired value is sufficiently long that the polymer can fully relax to its steady state configuration at each shear rate along the way to the final shear rate, thus allowing the system to achieve homogeneity via diffusion. The zero flux of concentration boundary condition at the walls, $\mathbf{n} \cdot (\phi \mathbf{v}_p) = 0$, is imposed by

$$\frac{\partial \phi}{\partial y} = \frac{\partial \pi_\phi}{\partial y} = 0 \text{ and } \frac{\partial Q_{yy}}{\partial y} = 0, \quad (6)$$

which collectively satisfy the desired condition; this is necessitated by the need for four (as opposed to two) boundary conditions on ϕ due to the fourth order derivative that appears in Eq. (1) via the term involving π_ϕ . A second order diffusion-like term also appears in $\nabla \cdot \sigma_p$ due to the fact that it is the polymer velocity that enters the upper convected derivative, thus providing a “natural” source of stress diffusion and so further boundary conditions are required. The polymer velocity component in the flow direction, v_p , can be written as

$$v_p = v + \frac{E}{\zeta} \frac{\partial \sigma_{xy}}{\partial y} = v + \frac{E}{\zeta} \left[G \frac{\partial Q_{xy}}{\partial y} + Q_{xy} \frac{\partial G}{\partial y} \right] \quad (7)$$

with $\partial G/\partial y = \alpha \phi^{-1} G \partial \phi/\partial y$. We can thus obtain an additional boundary condition by specifying v_p at the boundaries. We choose to assume $v_p = v$ at $y = 0, H$, and thus enforce

$$\frac{\partial Q_{xy}}{\partial y} = 0, \quad (8)$$

though it is clear that we could explore the effect of polymer slip with a modification of this condition. We assume that the solution is initially in a quiescent state $\mathbf{Q} = \mathbf{I}$, $v = 0$. In order to assess the nonlinear effects due to concentration gradients, we impose an initial defect in the polymer concentration

$$\phi = 1 + \delta \cos(\pi k y/H), \quad (9)$$

where the magnitude of the defect is chosen to be $\delta = 0.01$; for larger δ the phenomena discussed below occur at earlier times. Additional numerical tests reveal that the initial shape of the concentration profile (e.g., random distribution) does not affect the steady state. The values $k = 1$ and 5 have been chosen for the examples shown in this paper. The nonlinear system of partial differential equations is solved by an adaptive domain decomposition spectral method.²⁷

For slow flow (i.e., for Wi below the critical value for linear instability), the concentration defect is damped and the steady state flow is the normal linear profile. We first consider two cases

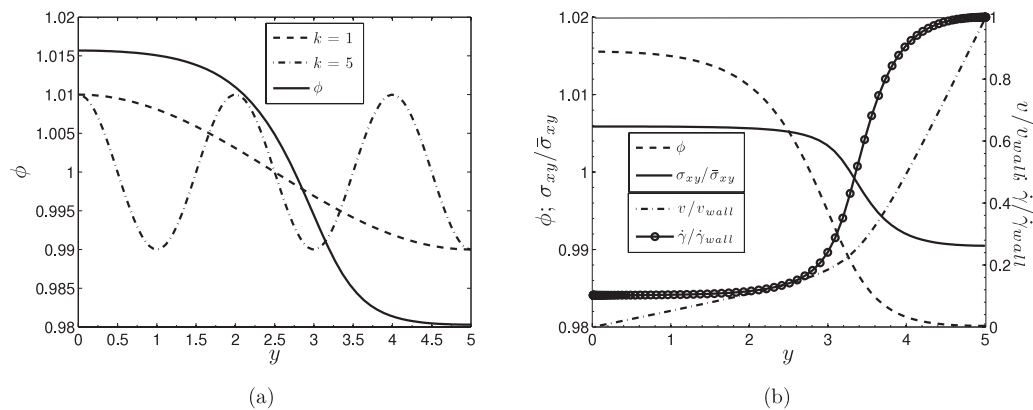


FIG. 1. $Wi = 10$. (a) Concentration profiles at $t = 0$ and at steady state for wavenumbers $k = 1$ and 5. (b) Left y-axis: Concentration profile and polymer shear stress profile normalized by the average shear stress. Right y-axis: Steady velocity and shear rate profiles normalized by respective wall values. To view the complex temporal evolution of the velocity and concentration profiles, see Figure 2.

for $Wi = 10$, which is within the linear instability window for our chosen model parameters. We use a ramping speed of $a = 1$, and initial concentration profiles defined by $k = 1$ and 5. In both cases, the velocity and concentration distribution undergo a nontrivial temporal evolution but the velocity profile eventually reaches a banded steady state. For a full appreciation of the complex dynamics, it is important to view the online movies—the respective steady-states for $k = 1, 5$ are reproduced in Figures 2(a) and 2(b); the temporal evolution is especially remarkable in the case $k = 5$. In Figure 1(a), we show only the two sets of initial profiles and the final banded concentration profiles, which are identical in the two cases.

In the transient approach to steady state, the flow field initially is linear in space and the concentration defect begins to dampen. In the case $k = 1$, the shear stress overshoots, and the defect is amplified driving concentration down the shear-rate gradient (towards the fixed wall) such that the velocity profile splits into two distinct linear profiles with different slopes. Near the fixed wall, the flow slows down due to the influx of polymer, whereas there is a fast flow regime towards the moving wall. After about 100 strain units, the flow settles into a steady shear-banded profile (Figure 1(b)). Correspondingly, there are two regions of constant shear rate connected by a transition region. As noted above, the evolution of the profiles is much more complicated in the case $k = 5$, despite the fact that the final solution is identical.

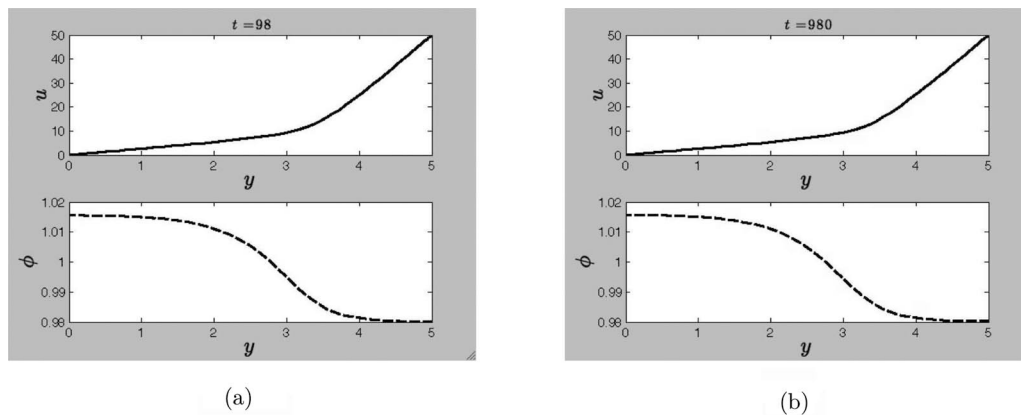


FIG. 2. $Wi = 10$. Stead-state velocity, u , and concentration, ϕ , profiles for (a) $k = 1$ and (b) $k = 5$. To view the complex temporal evolution of the velocity and concentration profiles, see the movies (enhanced online). [URL: <http://dx.doi.org/10.1063/1.4805089.1>] [URL: <http://dx.doi.org/10.1063/1.4805089.2>]

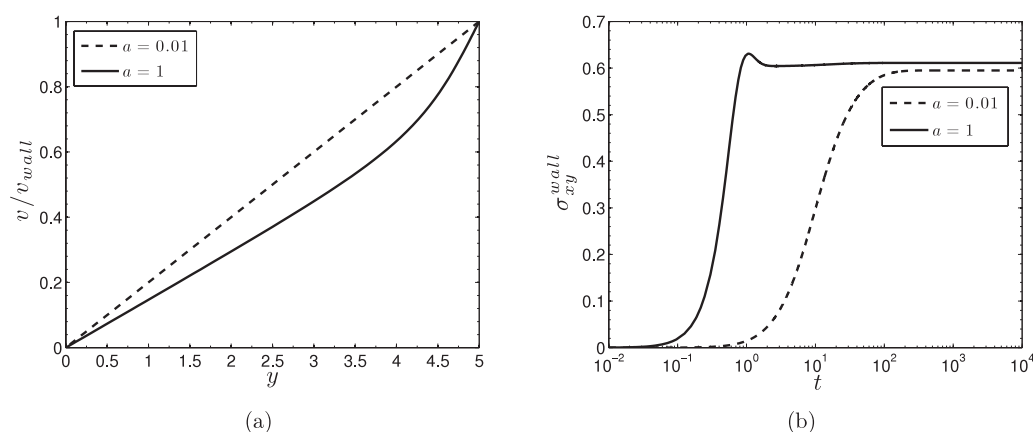


FIG. 3. $Wi = 3.9$. (a) Steady normalized velocity profiles for two ramp speeds $a = 0.01, 1$ showing multiple steady-state behavior. (b) Transient evolution of wall polymer shear stress for two ramp speeds $a = 0.01, 1$ showing the overshoot for fast ramps resulting in inhomogeneous flow profile. Note the non-uniqueness of the steady wall shear stress.

Referring to the final steady state profiles, it should be noted that the width of the transition region is $O(\sqrt{D\tau_{d0}})$. Hence, in the present case where the dimensionless channel width is only $H = 5$, the transition appears relatively smooth. However, as H is increased, the transition will appear sharper relative to the channel width. Additionally, there is a jump in the first normal stress difference N_1 , (not shown) and also in the polymer shear stress (of course, the total shear stress is constant across the gap as required by the momentum balance) (Figure 1(b)). The important thing to notice is that the polymer shear stress $\sigma_{xy} = \phi^\alpha Q_{xy}$ undergoes a jump across the transition from the high shear rate to low shear rate region, which is the distinguishing mark of shear banding. From the rheology above, we know that Q_{xy} increases with shear rate, thus it is the depletion of polymer in the high shear rate regime that produces a sufficient decrease in the shear stress such that shear banding is possible.

Finally, experimental results^{28,29} indicate that it is possible to reach multiple steady states (homogeneous and inhomogeneous profiles) depending upon the wall ramp speed. Such behavior is clearly impossible if the homogeneous flow profile is linearly unstable, and hence the inhomogeneity cannot be explained by non-monotonicity of the constitutive curve. Indeed, for appropriate material parameters, we find that our model exhibits a subcritical, nonlinear instability in which steady state flow can be either homogeneous or inhomogeneous depending on the ramping speed. In Figure 3(a), we show that for parameters close to, but outside the linear instability window (namely, $Wi = 3.9$, other parameters as specified above), it is possible to find multiple steady-states depending upon the ramp speed, a . For ramp speeds sufficiently large that the shear stress undergoes an overshoot, the resulting steady profile is inhomogeneous. On the other hand, a homogeneous profile can be recovered if instead the ramp speed is sufficiently slow that the shear stress overshoot is circumvented (Figure 3(b)).

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