Film-Splitting Flows of Shear-Thinning Liquids in Forward Roll Coating

Asymmetric film-splitting of shear-thinning liquid between rotating cylinders in forward roll coating is analyzed with the Carreau model of shear-sensitive liquid. The mass and momentum conservation equations for steady, two-dimensional flow are solved by means of the Galerkin finite-element method. A simpler model is also developed from the lubrication approximation and its range of validity is established. The major results are that the greater the shear thinning, the farther out of the gap the splitting meniscus moves, the higher the flow rate through the gap, and the more symmetrical the film split than with Newtonian liquid. Experiments reported in the literature confirm some of these predicted trends.

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Introduction

Many common coating liquids are non-Newtonian, and most of those are shear-thinning. It is important to know the effect of shear thinning on the flow rate and film-splitting of liquid passing between counterrotating cylinders as in forward roll coating and related liquid metering devices. In this paper shear thinning is described by the Carreau viscosity equation, which accounts for Newtonian behavior at low shear rates and at high shear rates, and for power-law behavior in an intermediate range of shear rates.

Here the flow in the film-splitting region is taken as two-dimensional and steady, although ribbing and other patterning is encountered in many circumstances (Pitts and Greiller, 1961; Mill and South, 1967; Greener et al., 1980). First a limiting case is analyzed, that of flow for which the lubrication approximation is adequate. The range of applicability of this approximation is established subsequently by solving the general equations of mass and momentum conservation together with the Carreau model. The results are compared with available experimental data.

Lubrication Theory for Shear-Thinning Liquids

In setting up a lubrication model of roll coating flows it is convenient to define the dimensionless coordinates; see Figure 1:

$$\theta = \tan^{-1}(X/\sqrt{2RH_o}) \tag{1}$$

$$\eta = 2\frac{Y - H_1(X)}{H_2(X) - H_1(X)} \tag{2}$$

so that $-\pi/2 < \theta < \pi/2$ and $0 < \eta < 2$, and the roll surface profiles although circular are approximated by parabolas. The viscosity of a power-law liquid in the lubrication approximation is

$$\mu = \mu_o \dot{\gamma}_o^{1-n} \left| \frac{\partial U}{\partial Y} \right|^{n-1} \tag{3}$$

where μ_o is the viscosity at a reference shear rate $\dot{\gamma}_o$. The dimensionless flow rate through the gap is defined by

$$\lambda = \frac{\frac{1}{2}\int_{H_1}^{H_2} U\,dY}{\overline{V}H_o} \tag{4}$$

where $\overline{V}$ is the average velocity of the roll surfaces. Then the balance of x momentum can be integrated once introducing an arbitrary constant C, where $\eta = C$ is the location of zero velocity gradient. Because fractional powers of negative numbers are inadmissible, appropriate equations must be derived for each of the four distinct (in θ) flow regions, defined by whether the pressure gradient is positive or negative and whether $u(y)$ has a minimum (maximum) or not.

For each case, the momentum equation is integrated a second time and one boundary condition of no slip at the wall is imposed, resulting in an expression relating the velocity profile to the pressure gradient and the integration constant C. Impos-

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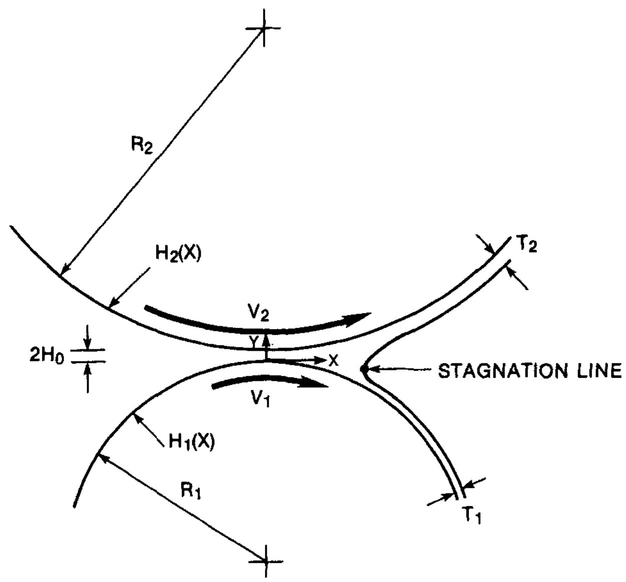

Figure 1. Asymmetric film-splitting in forward roll coating.

ing the second no-slip boundary condition gives an expression for the pressure gradient in terms of C.

In none of the four regions can the integration constant be found explicitly; instead the pressure gradient must be written as a function of C, and this expression has to be substituted into an equation for the dimensionless flow rate λ obtained by integration of the velocity profile across the gap. This gives C, and thus p_θ, v. θ for a given λ, so that the pressure profile can be numerically integrated up to the film-split location θ_m, where the calculated pressure is compared to the capillary pressure. The value of λ for which the pressure boundary condition is satisfied is found by a secant method. The expressions for pressure gradient are derived in detail by Coyle (1984).

Two important analytic results can be obtained directly from these pressure gradient and flow rate expressions. If the film is assumed to split at the first stagnation line downstream of the gap (Hopkins, 1957; Coyle et al., 1986), then the point $C = \eta_m$ represents a stagnation line. Setting the velocity there to zero and eliminating the pressure gradient from the resulting expression gives, after some algebra,

$$\eta_m = \frac{2}{1 + V^{n/(n+1)}} \tag{5}$$

where V is the speed ratio V_2/V_1. With this and much more algebra, the film thickness ratio is found to be the simple expression

$$\frac{t_2}{t_1} = V^{n/(n+1)} \tag{6}$$

The last two equations are the generalizations to a power-law liquid of the simple results for Newtonian liquids derived elsewhere (Eqs. 2.24 and 2.26 in Coyle et al., 1986).

To complete the model, the pressure distribution must be found by numerical integration. At a speed ratio of unity the model reduces to that proposed by Greener and Middleman (1975) for the symmetric case. As is also true in the lubrication theory for Newtonian liquid, the free-surface model is inadequate at low capillary number (Coyle et al., 1986). Thus only the predictions at infinite capillary number (negligible surface tension) are presented and compared to the accurate theory described next.

Finite Element Analysis of Two-Dimensional Free Surface Flow

In general, roll coating flows are bounded on several sides by sharply-curved liquid/air interfaces whose location is unknown *a priori* and under which the flow is inherently two-dimensional. While the lubrication approximation is adequate for the flow in the center of the gap, the free surface flow region requires a much more detailed analysis.

This section summarizes the Galerkin finite element method used to calculate the predictions of steady two-dimensional viscous free surface flows of liquids with shear-rate dependent viscosity. Only the essentials are presented; details are given by Kistler and Scriven (1983, 1984), Kistler (1983) and Coyle et al. (1986).

For steady, isothermal flow of an incompressible liquid, conservation of momentum and mass is expressed by the Navier-Stokes system in terms of a total stress tensor $\underline{\underline{T}}$. The simplest non-Newtonian fluid model is one with a viscosity that depends upon shear rate but with stress still directly proportional to the rate of deformation. In dimensionless form this is

$$\underline{\underline{T}} = -p\underline{\underline{I}} + \mu^*\underline{\underline{D}} = -p\underline{\underline{I}} + \mu^*[\nabla\underline{u} + (\nabla\underline{u})^T] \tag{7}$$

The finite element calculations in this paper used the Carreau viscosity model (Carreau, 1968; Bird et al., 1977), which has Newtonian plateaus in the viscosity at high and low shear rates, the two plateaus being connected by a power-law region. In dimensionless form the model can be expressed as

$$\mu^* = \frac{\mu}{\mu_o} = (1 - \beta)[1 + (\alpha\dot{\gamma})^2]^{(n-1)/2} + \beta \tag{8}$$

Here n is similar to the power-law index, $\beta = \mu_\infty/\mu_o$ is the ratio of infinite to zero shear-rate viscosities, $\alpha = (\overline{V}/H_o)/\dot{\gamma}_o$ is the ratio of the characteristic shear rate of the process to the characteristic shear rate of the liquid (transition from μ_o to power law), and $\dot{\gamma}$ is the second invariant of the rate-of-deformation tensor, which for two-dimensional flow can be written

$$\dot{\gamma} = \left[2\left(\frac{\partial u}{\partial x}\right)^2 + \left(\frac{\partial u}{\partial y} + \frac{\partial v}{\partial x}\right)^2 + 2\left(\frac{\partial v}{\partial y}\right)^2\right]^{1/2} \tag{9}$$

No-slip boundary conditions are specified at solid boundaries, while no-traction boundary conditions are specified at outflow planes. The downstream end of the free surface is usually taken at an outflow plane, where the appropriate boundary condition for the free surface is that it is parallel to the solid substrate. Inflow boundary conditions arise from matching a one-dimensional lubrication flow model to the two-dimensional flow.

At the free surface between the liquid and a gas (considered here as inviscid and without inertia) the normal stress in the liquid must balance the capillary pressure, which results in an expression relating the normal stress in the liquid to surface tension and curvature of the meniscus, and which also requires the shear stress to vanish. In addition, the kinematic boundary condition is also imposed there, which requires that there be no mass flow through the interface, i.e., the free surface is a streamline.

The Galerkin finite element method is used to solve the conservation equations. The system is nonlinear owing to the presence of free surfaces in addition to the nonlinear viscosity function. The formation of the weighted residual equations is the same as in the case of a Newtonian fluid (Coyle et al., 1986), except that the stress is given by Eq. 7 where the dimensionless viscosity is a function of the velocity gradients, Eqs. 8 and 9. Quadrilateral elements are employed, with nine-node biquadratic basis functions for the velocity field and four-node bilinear functions for the pressure field. The free surface is handled directly by the use of isoparametric mapping such that its location is calculated at the same time as the flow field.

The resulting set of nonlinear algebraic equations is solved using Newton's method, which converges quadratically over the entire parameter range and has the added bonus that the Jacobian matrix contains information about the stability of the flow, a point that is exploited elsewhere (Coyle, 1984; Coyle et al., 1987). When the viscosity is a function of the shear rate, the Jacobian can still be derived analytically by applying the chain rule to the stress term in the momentum weighted residual, along with the definitions given in Eqs. 8 and 9, which allow μ^* to be written explicitly in terms of the velocities (Coyle, 1984). An important point is that the entire flow field and free surface are calculated simultaneously. The addition of a shear-rate dependent viscosity does not require any type of successive approximation scheme to be introduced.

The full Newton system is assembled and solved by the frontal technique developed by Hood (1976, 1977), with the addition of the variable frontwidth modification suggested by Walters (1980). The solution for one set of parameter values is used as an initial estimate for a not too different set of parameters.

General Analysis of Forward Roll Coating for a Shear-Thinning Liquid

The finite element analysis of film-splitting flow is here generalized to shear-thinning liquids as described above. Upstream of the gap is considered to be flooded and the liquid is modeled by the power law; so the lubrication equations developed earlier determine the boundary conditions at the inlet of the finite element domain ($x = 0$). The main difference is that the pressure at the nip must be calculated by numerical integration rather than a simple Newtonian expression. Newton's method is used to determine the flow field, free surface, and flow rate simultaneously.

Typical finite element discretizations are shown in Figure 2.

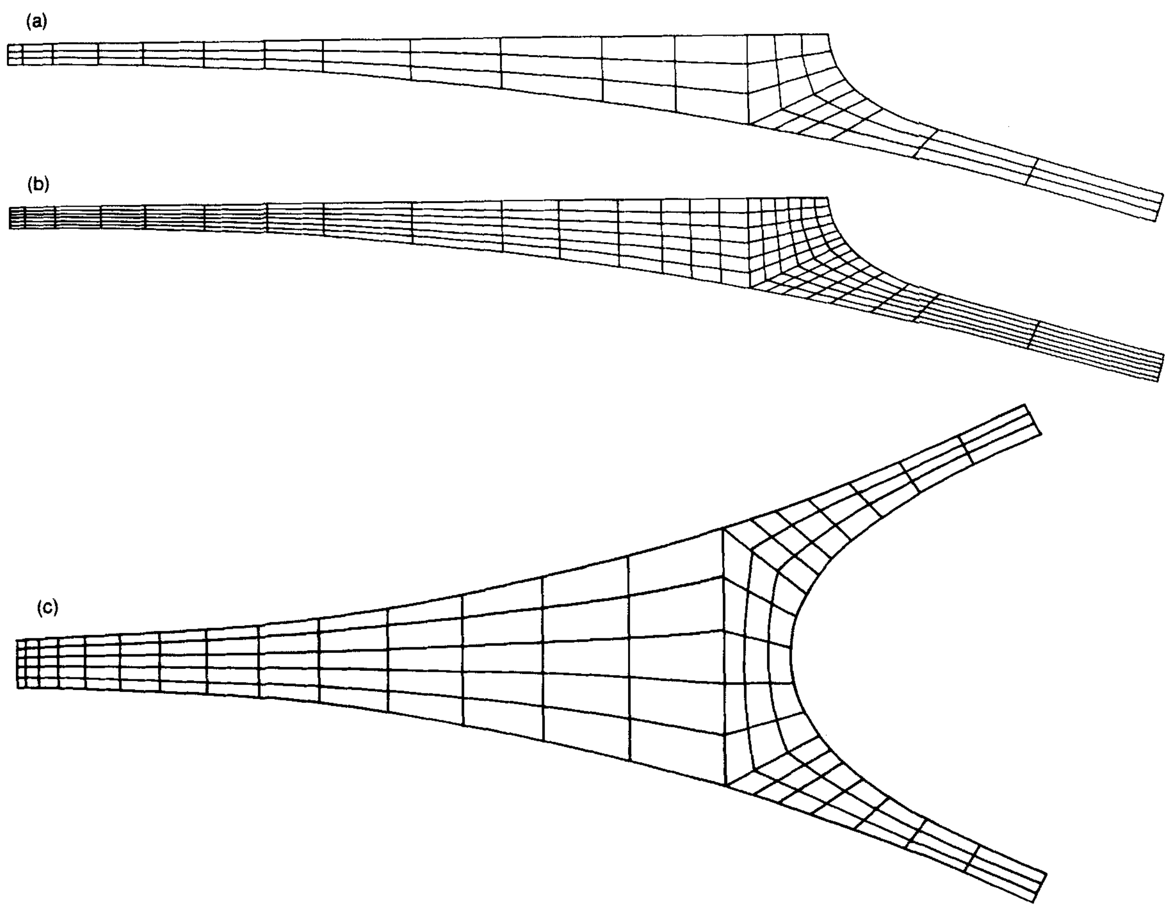

Figure 2. Finite element discretizations for flow in forward roll coating.
a. Symmetric, coarse
b. Symmetric, fine
c. Asymmetric

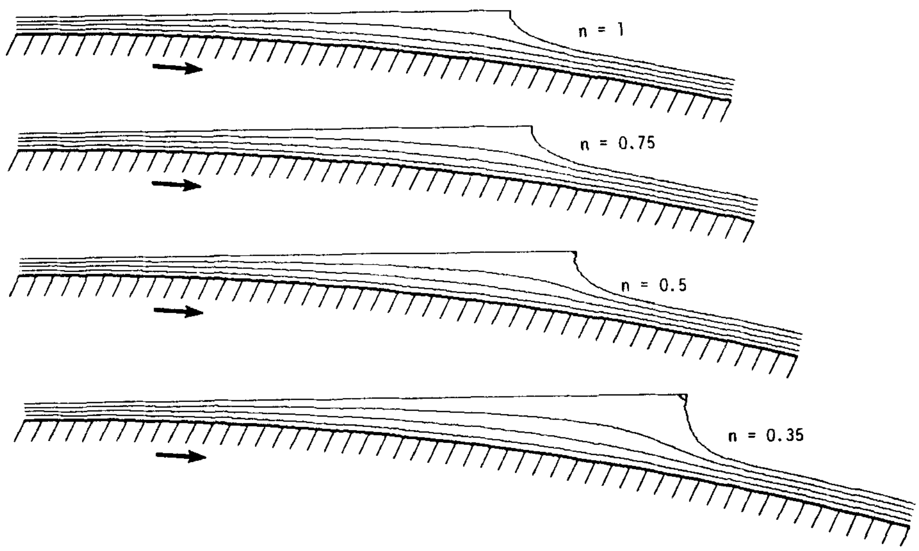

Figure 3. Calculated streamlines for symmetric film-splitting flow as a function of power-law index *n*.
$Ca = \infty, Re = 0, St = 0, R/H_o = 200, \alpha = 100, \beta = 0$

Fine-mesh solutions of the symmetric flow (Figure 2b, 1,775 unknowns) are within 0.1% of those obtained with a coarse mesh (Figure 2a, 711 unknowns), which indicates that the solutions are converged with respect to mesh refinement.

Figure 3 shows the calculated streamlines for symmetric forward roll coating of a shear-thinning liquid. At infinite capillary number there are no recirculations in the flow but there is a small "dead space" of very slow flow near the intersection of the free surface and the symmetry line. Decreasing the power-law index causes the meniscus to move out of the gap and the dead space to enlarge.

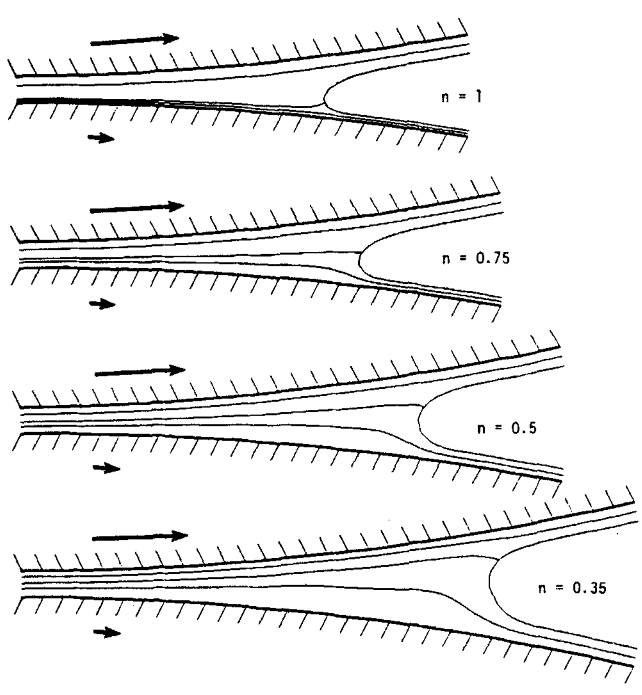

Figure 4. Calculated streamlines for asymmetric film-splitting flow as a function of power-law index *n*.
$Ca = \infty, Re = 0, St = 0, R/H_o = 200, V_2/V_1 = 5, \alpha = 100, \beta = 0$

Figure 4 shows the calculated streamlines of the asymmetric flow for the case of infinite capillary number and a speed ratio of 5. In addition to increasing the dead space, decreasing the power-law index causes the film to split much more symmetrically than it would if the liquid were Newtonian. At a power-law index of 0.35, the film-split is nearly symmetric even though the speed ratio is 5.

Asymmetry due to different roll radii was also investigated. The Navier-Stokes solutions indicate that the larger roll carries with it a slightly thicker film. For example, when the ratio of roll radii is 5, the film thickness ratio is 1.06 when the speed ratio is one. Thus for practical ranges of roll radius ratios, the lubrication model is accurate in predicting negligible effect.

The effect of α, the ratio of a characteristic shear rate of the flow (V_1/H_o) to the shear rate where the liquid viscosity begins to enter the power-law region ($\dot{\gamma}_o$), is negligible for a value greater than about 10, which is usually the case in experimental setups. This is due to the fact that the flow rate is metered and the pressure profile is controlled in the narrow gap region where the shear rates are the highest. For $\alpha < 1$, the liquid is essentially Newtonian (low shear rate viscosity plateau of the Carreau model). For $\alpha > 10$, the shear rate is in the power law region over the majority of the flow domain. Further increasing α does not change the flow significantly, although there may be minor changes in the slow-flowing recirculation zones near the meniscus.

Figure 5 shows the predicted flow rate and film-split location at infinite capillary number compared with the lubrication model. As is the case for a Newtonian liquid, the lubrication model performs poorly in predicting both λ and $\tan\theta$, and poorly too in predicting their sensitivity to parameter changes. Flow rate predictions of lubrication theory and two-dimensional finite element analysis are in reasonable agreement when $n > 0.6$, but the lubrication model overestimates the flow rate at lower values of power-law index. Film-split location predictions disagree at all values of power-law index, with the lubrication model overesti-

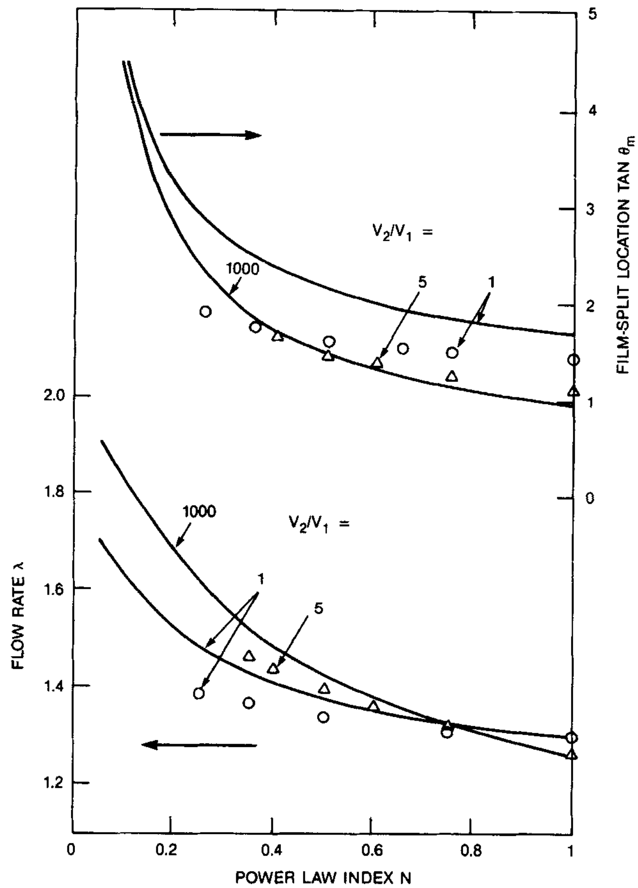

Figure 5. Effect of power-law index n on flow rate through the gap, λ, and film-split location, tan θ_m.

— Prediction of lubrication model
○△ Prediction of finite element analysis
$Ca = \infty$, $Re = 0$, $St = 0$, $R/H_o = 200$, $\alpha = 100$, $\beta = 0$

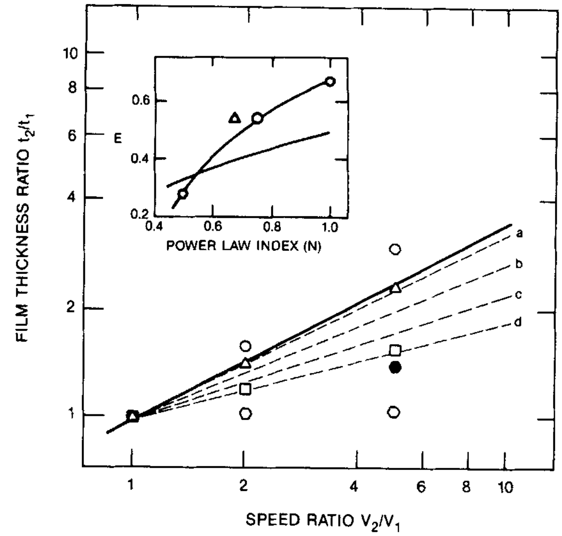

Figure 6. Effect of power-law index on film thickness ratio in assymetric forward roll coating.

$Ca = \infty$, $Re = 0$, $St = 0$, $R/H_o = 200$, $\alpha = 100$, $\beta = 0$
○ $n = 1$; △ $n = 0.75$; □ $n = 0.5$; ⬭ $n = 0.35$; ● $n = 0.35$, $\beta = 0.01$
— Data of Benkreira et al (1981)
--- Predictions of lubrication model (Eq. 6)
a. $n = 1$; b. $n = 0.75$; c. $n = 0.5$; d. $n = 0.35$.

Inset: Same results where E is the exponent defined by $t_2/t_1 = (V_2/V_1)^E$.

-o-o- Finite element analysis
— Lubrication model
△ Data

mating tan θ_m—especially at low values of the power-law index. Even so, such differences may be difficult to resolve experimentally.

Figure 6 shows how the film thickness ratio, at any speed ratio, is significantly lowered by shear-thinning. In general, the trend is confirmed qualitatively by the data of Benkreira et al. (1981), who showed that the split ratio for a liquid of power-law index $n = 0.67$ is proportional to $V^{0.54}$. The reduction of the film thickness ratio with decreasing power-law index is similar in form, although much stronger in magnitude, than predicted by the lubrication model for a power-law liquid in Eq. 6. This discrepancy is not due to the different models of shear-sensitive viscosity (Carreau vs. power law), because when the power-law model was used in the finite element calculations the predictions did not change. The difference stems solely from the presence of the free surface, which the finite-element calculation accurately incorporates. As was shown in the Newtonian case (Coyle et al., 1986), it is the two-dimensional flow near the free surface that determines how the film splits, not the one-dimensional flow upstream.

Figure 6 also shows that the split ratio can be sensitive to the infinite shear rate viscosity $\mu_\infty(\beta = \mu_\infty/\mu_o)$, with higher β tending to preserve more of the asymmetry in the film-split than would occur with a Newtonian liquid. This is reasonable because $\beta = 0$ gives the maximum shear-thinning behavior while $\beta = 1$ gives a Newtonian liquid. But it emphasizes that if the shear rates in the coater are very high, then the power-law type results are not applicable and the high shear rate behavior of the liquid cannot be neglected although it may often have gone unappreciated.

Other than the film-split ratio, the only aspect of these predictions that might be tested against available experimental data is the trend toward higher flow rates with shear-thinning liquids (see the data of Greener, 1979). But this aspect cannot be corroborated since Greener's experiments were restricted to such low speeds (to avoid the ribbing instability) that gravity effects dominated. At this writing there are no extensive data pertaining to the flow rate, film-split ratio, and their sensitivity to shear thinning; thus no more than qualitative comparisons can be made.

Conclusions

The general theory of steady, two-dimensional film-splitting flows in forward roll coating has been extended to include shear-thinning liquids. The theory based upon the lubrication approximation is only qualitatively correct, while the finite-element-based theory predicts the complete flow fields with free surfaces. Increasing the degree to which the liquid is shear-thinning, i.e., decreasing the power-law index n, causes the meniscus to move out of the gap between the rolls and the flow rate to increase. In addition, decreasing the power law index causes the film to split much more symmetrically than would a Newtonian liquid at the same speed ratio. This general trend is confirmed by the experiments of Benkreira et al. (1981), but more extensive data (including more complete rheological characterization of the liquids) are needed before quantitative comparisons can be made.

The theoretical work presented here sets the stage for accurate three-dimensional stability analysis to predict flow instabilities such as ribbing (Coyle et al., 1987). It also provides a base for separating the effects of shear thinning from those due to viscoelasticity, which can now be done by comparing experiments using viscoelastic liquids to the theoretical predictions for shear-thinning inelastic liquids.

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Notation

C = integration constant
Ca = capillary number (surface tension/viscous force ratio)
$\underline{\underline{D}}$ = rate of deformation tensor
H_o = half the minimum distance between rolls
H_1, H_2 = Y coordinate of surface of rolls 1 and 2
n = power-law index
p = dimensionless pressure
R, R_1, R_2 = average roll radius $[1/R = \frac{1}{2}(1/R_1 + 1/R_2)]$, radius of rolls 1 and 2
Re = Reynolds number (inertia/viscous force ratio)
St = Stokes number (gravity/viscous force ratio)
$\underline{\underline{T}}$ = stress tensor
T_1, T_2 = coating thickness on rolls 1 and 2
t_1, t_2 = dimensionless coating thicknesses $(T_1/H_o, T_2/H_o)$
U, u = fluid velocity in X direction
v = dimensionless y velocity
$V_1, V_2, \overline{V}$ = speed of rolls 1 and 2, average speed
V = speed ratio (V_2/V_1)
X, Y = Cartesian coordinates (flow direction, crossflow direction)
x, y = dimensionless Cartesian coordinates

Greek letters

α, β = constants in Carreau viscosity model
$\dot{\gamma}$ = shear rate
θ, η = dimensionless coordinates (x, y)
λ = dimensionless flow rate
μ, μ^* = viscosity (μ^* is dimensionless)
μ_o, μ_∞ = zero, infinite shear-rate viscosity

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