Effects of Shear Thinning on Forward Roll Coating

R. W. Hewson and N. Kapur

School of Mechanical Engineering, University of Leeds, Leeds. LS2 9JT. UK.

Abstract

This paper described the forward mode roll coating process of generalised non-Newtonian fluids characterised by the Ellis model. The fluid in the coating bead and the free surface formation are described by the lubrication approximation and the stability is also considered using a perturbation analysis of the downstream meniscus. Results highlight the complex behaviour of this coating process and volume flow rate and film thickness results are obtained for a range of operating conditions. The stability of the downstream meniscus is observed to improve with increasing level of shear thinning when $\tau_{\frac{1}{2}}$ (the shear stress at which the viscosity is half that of the zero shear stress viscosity) is small, however as $\tau_{\frac{1}{2}}$ increases the meniscus stability decreases with increasing levels of shear thinning.

Keywords: Roll coating, Generalised Newtonian fluid, Ellis viscosity model, fluid mechanics, mathematical modelling.

1. Introduction

The application of liquid layers onto a solid substrate is a commonly encountered industrial process. The long list of coated products includes

Email address: r.w.hewson@leeds.ac.uk (R. W. Hewson and N. Kapur)

packaging material, printed media, photographic film, magnetic media such as video and audio cassettes and optical media such as compact discs. As such, an almost equally wide range of equipment is used to manufacture these coated products, including roll coaters, where the transfer of fluid to the substrate is via a series of fluid coated rollers. The simplest geometry is a two roll coater of which there are two modes of operation of roll coaters - forward or reverse roll coating, these modes are shown in figure 1. In forward roll coating the two rolls pass through the coating bead in the same direction (i.e. the rolls are counter-rotating), in the reverse case the opposite is true with the two rolls passing through the coating bead in the same direction (i.e. co-rotating). Both forward and reverse roll coating may be operated with a flooded or starved inlet [27]. These definitions correspond to the film thickness entering the coating bead relative to the minimum gap between the rolls. When the thickness of the liquid film entering the coating bead is of the same order or greater than the gap between the rolls a rolling bank of fluid may form and be returned to the coating pool [6]. When the liquid film is significantly thinner than the roll separation, the upstream meniscus plays an important part in determining the pressures throughout the coating pool and on the entire coating process in general [5, 11, 12, 10]. In the current work the flooded forward roll coating case is examined, a problem encountered in a range of coating processes, for example the transfer of fluid from a metering to applicator roll. Invoking the lubrication approximation forward roll coating has been analysed by numerous authors [3, 14, 13, 26, 7]. As part of these analyses a range of pressure boundary conditions were applied at the upstream and downstream menisci. These conditions vary in com-

plexity and their ability to describe surface tension effects. The propensity of coating fluids that exhibit shear thinning behaviour has led to the analysis of the forward roll coating process when operated with such fluids. Coyle et al. [8] analysed the lubrication problem for shear thinning fluids obeying the power law and compared the lubrication theory derived solutions to a full two dimensional finite element formulation of the problem for shear thinning fluids obeying the more general Carreau-Yassuda fluid model. Applying the lubrication approximation and the Prandtl-Hopkins condition [20, 14] they obtained an elegant relationship for the ratio of film thicknesses leaving the bead to that of the roll speeds:

$$\frac{T_1}{T_2} = S^{\frac{n}{n+1}}, \quad (1)$$

where T_1 and T_2 are the film thicknesses on the upper and lower rolls respectively, S is the ratio of speeds of the top to bottom rolls and n is the power law index. In both the finite element analysis and lubrication model the effect of an increase in shear thinning behaviour (a decrease in n) was to increase the flow rate through the coating bead and move the film split location further from the point of minimum separation of the two rolls.

One of the major difficulties associated with assessing the effect of shear thinning on a coating process is in defining the viscosity that characterises the process. In the case of the analysis of Coyle et al. [8] neglecting the surface tension effects, and thereby solving the problem for an infinite capillary number (ratio of viscous to surface tension forces), avoided this problem. Considering surface tension forces in addition to viscous ones, as is undertaken in the work presented here, requires a capillary number to be defined

as a function of a characteristic viscosity. Consequently any assessment of shear thinning effects will depend on this relationship and the rheological model.

A number of computational studies of visco-elastic fluids have been undertaken. The visco-elastic nature of the fluid together with the requirement to establish the free surface location as part of the solution make such problems complex. In addition to the conservative equations, the general conformation tensor based constitutive equation must be solved, from which the polymeric contribution to the stress tensor is obtained [18, 4]. Romero et al. [24] and Bajaj et al. [2] studied slot coating of visco-elastic fluids onto a moving substrate. Similarly, Quintella et al. [21] examined both computationally and experimentally the displacement of a visco-elastic fluid filled capillary tube by a semi-infinite slug of air. The complexity of the computational models for visco-elastic fluids illustrates the need for a simplified model capable of capturing the physics of the fluid film deposition process, one such model was developed by de Ryck and Qur [9] who used a very elegant lubrication based asymptotic analysis - assuming a Poiseuille velocity profile - to obtain an equation describing the residual fluid film deposited on a wire withdrawn from a fluid filled gap. With only a single adjustable parameter, obtained from experimental data, the analysis provides a useful tool to enable the influence of the different parameters effecting the process to be examined. A similar analysis was undertaken by Ashmore et al. [1] who developed a more general analysis for a range of different geometries in the limits of when the fluid rheology is either dominated by shear thinning effects or elastic effects. In common with the analysis of [9] a quadratic velocity profile is assumed as

the constitutive equations lacked an analytical solution. Their model showed excellent qualitative agreement with the experiments they undertook, with quantitative agreement observed for the shear-thinning dominated flows. In order to fully analyse the effects of shear thinning the current analysis is restricted to generalised Newtonian fluids only (i.e. when the Weissenburg number goes to zero), furthermore it permits a stability analysis to be undertaken.

1.1. Scope of Paper

This paper examines the forward roll coating process of a fluid obeying the Ellis model [22, 16] by means of the usual lubrication approximation. The Ellis model was chosen as it is able to describe shear thinning fluid behaviour at low shear stresses - essential if free surface flow is considered [17]. Importantly for forward mode roll coating where meniscus stability can be an issue the stability of the coating bead is analysed by examining the restoring forces resulting from a perturbation in meniscus location [25].

2. Coating Model

The model is divided into two components illustrated in figure 2; (i) the lubrication model of the flow between the two rolls together with an upstream boundary condition $p_\infty = 0$, consistent with a fully flooded inlet [12], (ii) the film splitting model that describes the downstream meniscus.

2.1. Lubrication Flow

By making the lubrication assumption that the flow is unidirectional [23] the balance of pressure and viscous forces is written:

$$\frac{d}{dY} \left[\bar{\eta} \frac{dU}{dY} \right] = \frac{dP}{dX}, \quad (2)$$

where $\bar{\eta}$ is the local fluid viscosity, Y is the coordinate perpendicular to the near parallel roll surfaces and X the direction parallel to the local roll surface, P is the local pressure. For Ellis fluids the local viscosity is a function of shear stress, $\bar{\tau}$:

$$\bar{\eta} = \frac{\bar{\eta}_0}{1 + \left| \frac{\bar{\tau}}{\bar{\tau}_{\frac{1}{2}}} \right|^{\alpha-1}}, \quad (3)$$

where $\bar{\eta}_0$ is the zero shear stress viscosity, $\bar{\tau}_{\frac{1}{2}}$ the **shear stress** at which the viscosity is half the zero shear viscosity and α is a constant analogous to the inverse of the power law index when modelling the flow as a power law fluid. Note that as the zero shear stress viscosity (as defined in the Ellis model) tends to infinity the Ellis model collapses to the power law model and $\alpha \rightarrow \frac{1}{n}$. The dimensionless form of equations (2) and (5) used in the subsequent analysis are,

$$\frac{d}{dy} \left[\eta \frac{du}{dy} \right] = \frac{dp}{dx}, \quad (4)$$

$$\eta = \frac{1}{1 + \left| \frac{\tau}{\tau_{\frac{1}{2}}} \right|^{\alpha-1}}, \quad (5)$$

using the following non-dimensional scalings,

$$p = \frac{PH}{\bar{\eta}_0 U_1} \quad \tau = \frac{H\bar{\tau}}{\bar{\eta}_0 U_1} \quad [x, y] = \frac{[X, Y]}{H} \quad (6)$$

where H is the minimum roll-to-roll gap and U_1 is the lower roll peripheral speed. From the lubrication assumptions the shear stress is the product of the local viscosity and velocity gradient, $\tau = \eta \frac{du}{dy}$. This permits equation (4) to be written as,

$$\frac{d\tau}{dy} = \frac{dp}{dx} \quad (7)$$

which on integration with respect to y results in

$$\tau = \frac{dp}{dx} y + c_1 \quad (8)$$

where c_1 is the shear stress at $y = 0$. Multiplying the Ellis equation (5) describing the fluid by the velocity gradient $\frac{du}{dy}$ and rearranging gives,

$$\frac{du}{dy} = \tau + \tau \left| \frac{\tau}{\tau_{\frac{1}{2}}} \right|^{\alpha-1}, \quad (9)$$

into which equation (8) can be substituted resulting in,

$$\frac{du}{dy} = \frac{dp}{dx} y + c_1 + \frac{1}{\tau_{\frac{1}{2}}} \left(\frac{dp}{dx} y + c_1 \right) \left| \frac{dp}{dx} y + c_1 \right|^{\alpha-1}. \quad (10)$$

Equation (10) can now be solved for u , taking care to correctly consider the absolute terms,

$$u = \frac{1}{2} \frac{dp}{dx} y^2 + c_1 y + \frac{\left| \frac{dp}{dx} y + c_1 \right|^{\alpha-1} - |c_1|^{\alpha-1}}{\tau_{\frac{1}{2}}^{\alpha-1} \frac{dp}{dx} (\alpha + 1)} + u_0. \quad (11)$$

Further integration of equation (11) results a relationship between the flux, q , and the pressure gradient, $\frac{dp}{dx}$.

$$q = \frac{1}{6} \frac{dp}{dx} h^3 + \frac{1}{2} c_1 h^2 + \frac{1}{\tau_{\frac{1}{2}}^{\alpha-1} \frac{dp}{dx} (\alpha + 1)} \left[\frac{\left(\frac{dp}{dx} h + c_1 \right) \left| \frac{dp}{dx} h + c_1 \right|^{\alpha+1} - c_1 |c_1|^{\alpha+1}}{\frac{dp}{dx} (\alpha + 2)} - |c_1|^{\alpha+1} h \right] + u_0 h. \quad (12)$$

The film thickness, h , is calculated by the following parabolic approximation for the two rolls,

$$h = 1 + \frac{x^2}{2R_1} + \frac{x^2}{2R_2}, \quad (13)$$

where R_1 and R_2 are the non-dimensional radii of upper and lower rolls (throughout the paper $R_1 = R_2 = 500$).

2.2. Free Surface Lubrication Flow

The second part of the model is that which describes the film splitting at the downstream meniscus. Here those equations describing the free surface flow of an Ellis model fluid and those describing the local pressure due to the meniscus radius of curvature are solved. As the rolls are operated at different speeds the equations were solved for each roll surface with appropriate boundary conditions used to describe the point of film splitting. The problem is divided into the upper and lower roll films and the problem is defined along the free surface. The lubricating flow on the upper and lower rolls is

first considered. Using the usual lubrication assumptions and by assuming the air phase has negligible density and/or viscosity leading to zero shear stress at the free surface ($y = 0$ with y increasing in towards the substrate) equation (10) becomes,

$$\frac{du}{dy} = \frac{dp}{dx}y + \frac{1}{\tau_{\frac{1}{2}}} \left(\frac{dp}{dx}y \right)^\alpha. \quad (14)$$

The absolute terms are removed as the pressure gradient can be assumed to be positive, resulting from an assumed progressively decreasing film thickness from the point of film splitting to the final film thickness. Equation (14) can then be used to obtain the velocity of the flow with the boundary condition $u_2 = S$ at $y_2 = h_2$ or $u_1 = 1$ at $y_1 = h_1$ for the upper and lower rolls respectively¹,

$$u_1 = \frac{\left(\left(\frac{dp}{dx} \Big|_1 y \right)^{\alpha+1} - \left(\frac{dp}{dx} \Big|_1 h_1 \right)^{\alpha+1} \right) \tau_{\frac{1}{2}}^{1-\alpha}}{\frac{dp}{dx} \Big|_1 (\alpha + 1)} + \left(1 + \frac{1}{2} (y_1^2 - h_1^2) \frac{dp}{dx} \Big|_1 \right), \quad (15)$$

$$u_2 = \frac{\left(\left(\frac{dp}{dx} \Big|_2 y \right)^{\alpha+1} - \left(\frac{dp}{dx} \Big|_2 h_1 \right)^{\alpha+1} \right) \tau_{\frac{1}{2}}^{1-\alpha}}{\frac{dp}{dx} \Big|_2 (\alpha + 1)} + \left(S + \frac{1}{2} (y_2^2 - h_2^2) \frac{dp}{dx} \Big|_2 \right). \quad (16)$$

¹Subscripts 1 and 2 denote the lower and upper rolls respectively.

Further integration across the film provides the flow rate on each roll,

$$q_1 = \frac{-3\tau_{\frac{1}{2}}^{1-\alpha} h_1^{\alpha+2} \left. \frac{dp}{dx} \right|_1^\alpha - h_1 \left(\left. \frac{dp}{dx} \right|_1 h_1^2 - 3 \right) (\alpha + 2)}{3\alpha + 6}, \quad (17)$$

$$q_2 = \frac{-3\tau_{\frac{1}{2}}^{1-\alpha} h_2^{\alpha+2} \left. \frac{dp}{dx} \right|_2^\alpha - h_2 \left(\left. \frac{dp}{dx} \right|_2 h_2^2 - 3S \right) (\alpha + 2)}{3\alpha + 6}. \quad (18)$$

By applying Newton's method to the flux equations (17) and (18) the pressure gradients can be obtained iteratively from,

$$\left. \frac{dp}{dx} \right|_1^{\{i+1\}} = \left. \frac{dp}{dx} \right|_1^{\{i\}} + A \left(\left. \frac{dp}{dx} \right|_1^{\{i\}} \right), \quad (19)$$

$$\left. \frac{dp}{dx} \right|_2^{\{i+1\}} = \left. \frac{dp}{dx} \right|_2^{\{i\}} + B \left(\left. \frac{dp}{dx} \right|_2^{\{i\}} \right), \quad (20)$$

where,

$$A \left(\left. \frac{dp}{dx} \right|_1^{\{i\}} \right) = \left. \frac{dp}{dx} \right|_1^{\{i\}} \frac{3q\alpha - 3hS\alpha + h^3 \left. \frac{dp}{dx} \right|_1^{\{i\}} \alpha + 3\tau^{-\alpha+1} \left(\left. \frac{dp}{dx} \right|_1^{\{i\}} h \right)^\alpha h^2 + 2 \left. \frac{dp}{dx} \right|_1^{\{i\}} h^3 - 6Sh + 6q}{\left(h \left. \frac{dp}{dx} \right|_1^{\{i\}} \alpha + 3\tau^{-\alpha+1} \left(\left. \frac{dp}{dx} \right|_1^{\{i\}} h \right)^\alpha \alpha + 2 \left. \frac{dp}{dx} \right|_1^{\{i\}} h \right) h^2}, \quad (21)$$

$$B \left(\left. \frac{dp}{dx} \right|_2^{\{i\}} \right) = \left. \frac{dp}{dx} \right|_2^{\{i\}} \frac{3q\alpha - 3h\alpha + h^3 \left. \frac{dp}{dx} \right|_2^{\{i\}} \alpha + 3\tau^{-\alpha+1} \left(\left. \frac{dp}{dx} \right|_2^{\{i\}} h \right)^\alpha h^2 + 2 \left. \frac{dp}{dx} \right|_2^{\{i\}} h^3 - 6h + 6q}{\left(h \left. \frac{dp}{dx} \right|_2^{\{i\}} \alpha + 3\tau^{-\alpha+1} \left(\left. \frac{dp}{dx} \right|_2^{\{i\}} h \right)^\alpha \alpha + 2 \left. \frac{dp}{dx} \right|_2^{\{i\}} h \right) h^2}. \quad (22)$$

In addition to the balance of viscous and pressure forces as described by equations (19) and (20) the local pressure can be obtained from the curvature

of the free surface,

$$\left. \frac{d\theta}{ds} \right|_1 = Ca p_1, \quad (23)$$

$$\left. \frac{d\theta}{ds} \right|_2 = Ca p_2, \quad (24)$$

where Ca is the capillary number and is the ratio of viscous to surface tension forces given by,

$$Ca = \frac{\bar{\eta}_0 U_1}{\sigma}, \quad (25)$$

where σ is the liquid-gas interfacial surface tension. From conservation of mass it is clear that,

$$\left. \frac{dq}{ds} \right|_1 = 0 \quad \text{and} \quad \left. \frac{dq}{ds} \right|_2 = 0, \quad (26)$$

where s is the distance along the free surface. The final equations required to close the problem are geometric considerations, the first being the relation between x and s :

$$\left. \frac{dx}{ds} \right|_1 = \cos \theta_1 \quad \text{and} \quad \left. \frac{dx}{ds} \right|_2 = \cos \theta_2, \quad (27)$$

and the relation between h and s :

$$\left. \frac{dh}{ds} \right|_1 = \sin \theta_1 \quad \text{and} \quad \left. \frac{dh}{ds} \right|_2 = \sin \theta_2, \quad (28)$$

Noting that $\frac{dp}{ds} = \frac{dp}{dx} \frac{dx}{ds}$, equations (19), (20), (23), (24), (26), (27) and (28) are able to describe the change in film thickness from the point of film splitting to

the fully developed film. In order to make the equations tractable boundary conditions are required. The boundary conditions appropriate to the problem are:

1. $\theta_1 = 90^\circ$ at $s_1 = 0$ (location of film splitting),
2. $\theta_2 = 90^\circ$ at $s_2 = 0$,
3. $q_1 + q_2 = q$, i.e. the total flux on both roll surfaces is equal to that passing between the rolls,
4. $p_1 = p_2$ at $s_1 = s_2 = 0$
5. $p_1 \rightarrow 0$ as $s_1 \rightarrow \infty$,
6. $p_2 \rightarrow 0$ as $s_2 \rightarrow \infty$,
7. $\theta_1 \rightarrow 0^\circ$ as $s_1 \rightarrow \infty$,
8. $\theta_2 \rightarrow 0^\circ$ as $s_2 \rightarrow \infty$,
9. $q_1 \rightarrow h_1$ as $s_1 \rightarrow \infty$,
10. $q_2 \rightarrow Sh_2$ as $s_2 \rightarrow \infty$,

Subscript 1 denotes the boundary condition is at the location of film splitting, subscript 2 refers to a distance of $s = 75$ from the location of film splitting (see figure 2), this was considered far enough downstream for the film to be fully developed. This was confirmed checking independence of solutions for $s > 75$ for the lowest capillary number cases considered. With these boundary conditions it was possible to solve all 10 ODEs and obtain a solution for the film splitting problem.

2.3. Stability Analysis

The small disturbance stability of the downstream meniscus was analysed using the stability criteria of Pitts & Greiller[19] and Savage[25]. The analysis examines the forces acting on the meniscus when subjected to a small

perturbation. Whether the force acts to restore the original meniscus location or increases the perturbation determined the stability. This is given by the following condition:

$$\left. \frac{dp}{dx} \right|_{\text{Lubrication}} - \left. \frac{dp}{dx} \right|_{\text{Meniscus}} < 0, \quad (29)$$

where $\left. \frac{dp}{dx} \right|_{\text{Lubrication}}$ is the full film regime at the point of film splitting and $\left. \frac{dp}{dx} \right|_{\text{Meniscus}}$ is the change in capillary pressure due to the perturbation. The pressure gradient $\left. \frac{dp}{dx} \right|_{\text{Lubrication}}$ is taken directly from the lubrication approximation for the flow at the point of film splitting. The pressure gradient $\left. \frac{dp}{dx} \right|_{\text{Meniscus}}$ was obtained numerically by calculating the location and pressure of the meniscus at a flow rate 0.1% above and below the equilibrium condition.

2.4. Numerical Implementation

A shooting method was used to obtain the flow rate for which the pressure at the downstream meniscus was the same as that obtained from the free surface model described in the next section. In order to achieve this the pressure gradient is required as a function of the flow rate. A hill climbing bracketing procedure was adopted [15] to obtain the flux which gives consistent pressures throughout the coating bead (based on zero fluid pressure far upstream of the coating bead). A further complication was that that c_1 is also unknown, to solve this variable a further search (a further bracketing procedure) was used to obtain the upper roll speed when $y = h$ is substituted into equation (11). From an initial flux the pressure at the downstream meniscus is calculated using the equations describing the film splitting. Equations

tions (19), (20), (23), (24), (26), (27) and (28) were solved using Matlab's boundary value problem solver *bvp4c* and used to give the pressure at the downstream meniscus. This was then compared with the pressure at this downstream location as determined by the lubrication equation in the fluid coating bead. The pressure distribution in the coating bead was obtained by searching for the shear stress, c_1 and pressure gradient that were consistent with equations (11) and (12). The search for the flux through the coating bead was then repeated until the downstream pressures were entirely consistent (within an error of $\Delta p < 1 \times 10^{-6}$). Each calculation took less than one minute on a (3GHz desktop PC). Checks for mesh independence and convergence of the searches were checked and the flow rate is estimated to be within 1% of those values presented here.

2.5. Domain Space

Following a preliminary study of the coating process, an array of parameters were defined for consideration, see table 1.

3. Results

An example of the free surface profile (illustrating the differing film thicknesses on the two rolls), together with the pressure distribution along the film surface is shown in figure 3. Pressures are identical at the film split point and become zero far downstream of the film split where the film is planar.

3.1. Flow Rate Through the Coating Bead

In analysing the data produced by the model let us first turn to the volume flow rate passing through the coating bead as shown in figures 4

and 5. Figures 4(a) to 4(d) illustrate how the predicted volume flow rate passing through the coating bead varies with capillary number and α . It is clear that as the capillary number increases then the flow rate through the coating bead decreases, a result of the less negative capillary pressure at the downstream meniscus “sucking” fluid through the coating bead. When considered in conjunction with figure 5 the importance of shear thinning on the coating bead can be examined. It is clear that as the level of shear thinning (as described by α) increases then the level of fluid passing through the coating bead increases. This is as would be expected, as the capillary number is based on the fluid’s zero shear viscosity and an increase in shear thinning leads to the local fluid’s local viscosity to be lower than that on which the capillary number is based, resulting in the same effect as reducing the capillary number. A further feature that can be seen from figures 4 and 5 is that the effect of shear thinning increases as $\tau_{\frac{1}{2}}$ decreases. The free surface condition that the shear stress is zero (based on the lubrication approximation applied here) leads to the local viscosity in the vicinity of the free surface to be close to the zero shear viscosity, however the higher shear stresses in the region of minimum gap between the two opposing rolls leads to a lower local viscosity in this region. As $\tau_{\frac{1}{2}}$ decreases more of the domain is affected by the fluid’s shear thinning. It is proposed that this is the basis for the increasing sensitivity to shear thinning as $\tau_{\frac{1}{2}}$ decreases.

3.2. Film Thickness Ratio

The film thickness ratio, as defined by the ratio of the film thickness on the top roll divided by that on the lower roll exhibits some interesting features, as shown in figures 6 and 7. As with the flow rate passing through the coating

bead, the greatest sensitivity of film thickness ratio to α is observed as $\tau_{\frac{1}{2}}$ decreases. Decreasing fluid viscosity (as described by decreasing capillary number and a decreasing $\tau_{\frac{1}{2}}$) also leads to an increase in the film thickness ratio. Comparison with equation (1) derived by Coyle et al. [8] is provided in figure 8. The equivalent power law index has been used, $n = \frac{1}{\alpha}$ and is based on the collapse of the Ellis model onto that of the power law when $\tau_{12} \rightarrow 0$. From the results, it can be seen that the best agreement between Coyle's equation and that of the model presented here is best for high capillary numbers and low values of $\tau_{\frac{1}{2}}$, this is entirely as expected as Coyle's equation does not consider surface tension effects and applied the power law model to describe the fluid viscosity, the results of which is that the viscosity tends to infinity as the shear stress goes to zero. Despite this the agreement is surprisingly good with the level of agreement deteriorating for low capillary numbers and high values of $\tau_{\frac{1}{2}}$.

3.2.1. Stability

Figure 9 shows the critical capillary number (the upper limit) above which it would be expected that the ribbing instability would be encountered. The left column describes the capillary number as a function of the lower roll capillary number, Ca_1^* , while the right column shows the critical capillary number of the upper roll, $Ca_2^*(=SCa_1^*)$, this effectively describes the maximum speed the coated substrate, wrapped around the upper roll, can pass through the coating bead (and provides a measure of coating productivity). It is interesting to note that for $\tau_{\frac{1}{2}} = 0.1$ as the level of shear thinning increases (as described by an increasing α) then the critical capillary number also increases, indicating an increase in stability of the coating bead. The

decrease in fluid viscosity leads to a greater influence of surface tension derived forces – increasing the stability. However, as $\tau_{1/2}$ is increase further the stability of the downstream meniscus decreases with increasing levels of shear thinning. It is hypothesised here that the reason for this is that the highest shear regions, the region of the smallest gap between rolls, will exhibit lower viscosities, increasing the volume flow rate passing through the coating bead. This in turn results in the downstream meniscus lying further downstream, decreasing the free surface curvature and pressure and importantly the restoring forces from a free surface perturbation. The lower shear stresses experienced by the fluid in the region around the free surfaces lead to relatively higher viscosities, and pressure gradients, thereby increasing the instability of the menisci. The transition from a regime where an increasing level of shear thinning leads to an improved stability to one where the reverse is true occurs at around $\tau_{\frac{1}{2}} = 0.2$.

An increase in speed ratio leads to a decrease in stability of the coating bead, as measured by the lower roll capillary number, however, the reverse is true when the top roll is considered. The reason for this is that as the speed ratio increases then for a constant lower roll speed the viscous forces increase, decreasing the stability of the coating bead. The stability increases when measured by the top roll's capillary number because for a given upper roll capillary number, increasing the speed ratio leads to a decrease in the lower roll's peripheral speed, decreasing viscous forces overall. This indicates that as the speed ratio continues to increase the area of coated film produced increases. A major practical limitation of this is that in order to flood the coating bead a large enough volume of fluid must enter the bead on the lower

roll, however as only the top roll's speed is increasing with increasing speed ratio the constant speed lower roll is delivering a constant volume of fluid to the coating bead. It would therefore be expected that as the speed ratio increases further the coating regime becomes that of the starved or meniscus roll coating process, whereby the upstream meniscus must be accounted for and is beyond the scope of this paper.

The film thickness deposited on the upper roll for a range of critical capillary numbers is shown in figure 10. It is interesting to see that the film thickness does not vary significantly from figure 10(a) to figure 10(b) despite the significant variation in the bead stability between these two conditions. This reflects the relative insensitivity of the film thickness ratio to the fluid properties, when compared to the sensitivity of the stability of the process to fluid properties.

4. Conclusions

The model of forward roll coating of shear thinning fluids has highlighted the complex response of the coating process to changes in operating parameters. Perturbation analysis of the downstream meniscus allows the stability to be assessed and a predictions to be made. In particular:

- A decrease in the fluid viscosity, as prescribed by decreasing the capillary number or increasing the level of shear thinning leads to an increase in the volume of fluid passing through the coating bead.
- The ratio of film thicknesses on the upper roll surface to that of the lower roll surface is most sensitive to shear thinning for low values of

$$\tau_{\frac{1}{2}}.$$

- The stability of the downstream meniscus as measured by the upper roll's capillary number (Ca_2^*) improves as the speed ratio increases.
- The effect of α on the meniscus stability is dependent on $\tau_{\frac{1}{2}}$, an increase in α improves stability for low $\tau_{\frac{1}{2}}$ and worsens stability as $\tau_{\frac{1}{2}}$ increases beyond $\tau_{\frac{1}{2}} \approx 0.2$. This suprising result illustrates that there is not a clear characteristic viscosity for the problem, and that the shear stress variation throughout the coating bead needs to be considered.

The ability of the model to predict the coating characteristics and the stability of the coating bead makes it a useful tool in developing forward roll coating processes. This is especially true of the stability predictions which can be provided in terms of the top roll critical capillary number, providing a useful measure of the maximum productivity of a coating line depositing a fluid with specific characteristics.

- [1] Ashmore, J., Shen, A. Q., Kavehpour, H. P., Stone, H. A., McKinley, G. H., 2008. Coating flows of non-Newtonian fluids: weakly and strongly elastic limits. *Journal of Engineering Mathematics* 60, 17–41.
- [2] Bajaj, W., Prakash, J. R., Pasquali, M., 2008. A computational study of the effect of viscoelasticity on slot coating flow of dilute polymer solutions. *Journal of Non-Newtonian Fluid Mechanics* 149 (1-3), 104–123.
- [3] Banks, W. H., Mill, C. C., 1954. Some observations on the behaviour of liquids between rotating rollers. *Proceedings of the Royal Society of London A* 223, 414.

- [4] Behr, M., Arora, D., Coronado, O. M., Pasquali, M., 2005. GLS-type finite element methods for visco-elastic fluid flow simulations. In: Bathe, K. (Ed.), Third MIT Conference on Computational Fluid and Solid Mechanics. pp. 586–589.
- [5] Benkreira, H., Edwards, M. F., Wilkinson, W. L., 1982. Ribbing instability in the roll coating of Newtonian fluids. *Plastics and Rubber Processing and Applications* 2, 137–144.
- [6] Coyle, D. J., 1997. Knife and roll coating. In: Kistler, S. F., Schweizer, P. M. (Eds.), *Liquid Film Coating*, 1st Edition. Chapman and Hall, pp. 539–571.
- [7] Coyle, D. J., Macosko, C. W., Scriven, L. E., 1986. Film-splitting flows in forward roll coating. *Journal of Fluid Mechanics* 171, 183–207.
- [8] Coyle, D. J., Macosko, C. W., Scriven, L. E., 1987. Film-splitting flows of shear-thinning liquids in forward roll coating. *AIChE Journal* 33 (5), 741–746.
- [9] de Ryck, A., Quéré, D., 1998. Fluid coating from a polymer solution. *Langmuir* 14 (7), 1911–1914.
- [10] Gaskell, P. H., Kapur, N., Savage, M. D., 2001. Bead-break instability. *Physics of Fluids* 13 (5), 1243–1253.
- [11] Gaskell, P. H., M.D., S., 1995. Meniscus roll coating. In: Kistler, S. F., Schweizer, P. M. (Eds.), *Liquid film coating*. Chapman and Hall.

- [12] Gostling, M. J., Savage, M. D., Wilson, M. C. T., 2001. Flow in a double-film-fed fluid bead between contra-rotating rolls part 2: bead break and flooding. *European Journal of Applied Mathematics* 12, 413–431.
- [13] Greener, J., Middleman, S., 1983. Reverse roll coating of viscous and viscoelastic liquids - response. *Industrial and Engineering Chemistry Fundamentals* 22 (4), 507–507.
- [14] Hopkins, M. R., 1957. Viscous flow between rotating cylinders and a sheet moving between them. *British Journal of Applied Physics* 8, 442–444.
- [15] James, G., 2008. *Modern Engineering Mathematics*, 4th Edition. Prentice Hall.
- [16] Matsuhis, S., Bird, R. B., 1965. Analytical and numerical solutions for laminar flow of non-Newtonian Ellis fluid. *AIChE Journal* 11 (4), 588–595.
- [17] Myers, T. G., 2005. Application of non-newtonian models to thin film flow. *Physical Review E* 72 (6).
- [18] Pasquali, M., Scriven, L. E., 2002. Free surface flows of polymer solutions with models based on the conformation tensor. *Journal of Non-Newtonian Fluid Mechanics* 108 (1-3), 363–409.
- [19] Pitts, E., Greiller, J., 1961. The flow of thin liquid films between rollers. *Journal of Fluid Mechanics* 11, 33.

- [20] Prandtl, L., March 1928 1928. Motion of fluids with very little viscosity. Technical memorandum of the national advisory committee, NACA.
- [21] Quintella, E. F., Mendes, P. R. S., Carvalho, M. S., 2007. Displacement flows of dilute polymer solutions in capillaries. *Journal of Non-Newtonian Fluid Mechanics* 147, 117–128.
- [22] Reiner, M., 1960. *Deformation, Strain and Flow*, 2nd Edition. H. K. Lewis & Co. Ltd., London.
- [23] Reynolds, O., 1886. On the theory of lubrication and its application to mr beauchamp tower’s experiment, including an experimental determination of the viscosity of olive oil. *Philosophical Transactions of the Royal Society of London* 177, 157–234.
- [24] Romero, O. J., Scriven, L. E., Carvalho, M. S., 2006. Slot coating of mildly viscoelastic liquids. *Journal of Non-Newtonian Fluid Mechanics* 138 (2-3), 63–75.
- [25] Savage, M., 1977. Cavitation in lubrication. part1. on the boundary conditions and cavity-fluid interfaces. *Journal of Fluid Mechanics* 80 (4), 763–755.
- [26] Schneider, G. B., 1962. Analysis of forces causing flow in roll coaters. *Transactions of the Society of Rheology* 6, 209–221.
- [27] Summers, J. L., Thompson, H. M., Gaskell, P. H., 2004. Flow structure and transfer jets in a contra-rotating rigid-roll coating system. *Theoretical and Computational Fluid Dynamics* 17 (3), 189–212.

Figure 1: Schematic of roll coaters; a) forward, b) reverse.

Figure 2: Definition of problem domain showing the two regions; (i) the lubrication flow between the two rolls and (ii) the film splitting of the fluid film onto the upper and lower rolls.

(a) Free surface geometry

(b) Pressure profile

Figure 3: Example of a typical film forming results for parameters: $Ca = 0.1$, $S = 3$, $a = 1.5$, $\tau_{\frac{1}{2}} = 1$, $R_1 = R_2 = 500$. A reasonably large speed ratio has been used to demonstrate the different pressures and free surface profiles on the upper and lower rolls.

Figure 4: Contours of volume flow rate passing through the coating bead against capillary number and shear thinning parameter, α .

Figure 5: Contours of volume flow rate passing through the coating bead against capillary number and shear thinning parameter, $\tau_{\frac{1}{2}}$.

Figure 6: Contour plots of the ratio of the fluid film thickness on the top roll to that on the bottom roll against capillary number and shear thinning parameter α .

(a) $S = 2, \alpha = 2.0$

Figure 7: Contour plots of the ratio of the fluid film thickness on the top roll to that on the bottom roll against capillary number and shear thinning parameter $\tau_{\frac{1}{2}}$.

Figure 8: Contour plots of the ratio of the fluid film thickness on the top roll to that on the bottom roll against speed ratio, S , and shear thinning parameter, α . The dashed lines are obtained from equation (1) for the limit of $\tau_{1/2} \rightarrow 0$.

(a) $Ca_1^*, \tau_{1/2} = 0.1$

(b) $Ca_2^*, \tau_{1/2} = 0.1$

(c) $Ca_1^*, \tau_{1/2} = 0.2$

(d) $Ca_2^*, \tau_{1/2} = 0.2$

(e) $Ca_1^*, \tau_{1/2} = 0.5$

(f) $Ca_2^*, \tau_{1/2} = 0.5$

(g) $Ca_1^*, \tau_{1/2} = 2.0$

(h) $Ca_2^*, \tau_{1/2} = 2.0$

Figure 9: Stability contours of critical lower and upper roll capillary number for a range of speed ratios and fluid properties.

Figure 10: Contours of film thickness at the critical capillary number for a range of speed ratios and fluid properties.

S	$1 \rightarrow 2$
α	$1 \rightarrow 2$
Ca	$0.01 \rightarrow 1$
$\tau_{\frac{1}{2}}$	$0.1 \rightarrow 2$
R_1	500
R_2	500

Table 1: Parameter space investigated in this study.