REACTIVE RIMMING FLOW OF NON-NEWTONIAN FLUIDS

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Abstract: - Unsteady flow of a liquid polymer treated as a non-Newtonian fluid on the inner surface of a horizontal rotating cylinder is investigated. Since the Reynolds number of a thin liquid film is small, a simple lubrication theory is applied. Governing equations for non-steady power-law and Ellis fluids are solved numerically and the time of transition from non-steady to steady-state mode for various model parameters and flow conditions are defined. The stabilization effect of a chemical reaction within the polymeric fluid (reactive flow) is examined.

1 Introduction

The problem of rotational flow on the inner and/or outer wall of a hollow horizontal cylinder has been of interest for many years due to its wide range of applications in industry [1,2]. Moffatt [3] was the first to derive the condition of the maximal supportable load for a Newtonian liquid. Later Preziosi & Joseph [4] presented the same condition in another form and named it a run-off condition for coating and rimming flows.

The possible instability of the liquid film on a cylindrical surface is one the most challenging fundamental aspects of this problem. Experimental and theoretical investigations by Thoroddsen & Mahadevan [5], Hosoi & Mahadevan [6], and Melo [7] demonstrated that under certain conditions, the fluid settles into a steady two-dimensional flow. A highly unstable nature of rimming Newtonian flow was discussed in a number of recent publications [8-13]. For example, Benilov and O'Brien [8] and Benilov [9] examined the stability of solutions, accounting for inertia and surface tension, and concluded that including these higher order corrections to the governing equation for the liquid film thickness may cause the instability of the steady-state solution. They proved that inertia always causes instability, but viscosity can make the characteristic time of growth large enough to effectively stabilize the film. Benilov et al [11, 12] have shown that the system admits strongly unstable solutions, which develop singularities in a finite time.

Although the aforementioned investigations highlight the main characteristics of the rimming flow, given its importance, not enough has been done to show the effect of non-Newtonian properties on such flow. Only a few attempts have been made, in which power-law model [14], Carrea-Yasuda model [15], Ellis model [16], Bingham model [17], and visco-elastic model [18] were used and the corresponding equations for the liquid layer thickness were solved numerically. In the recent papers Fomin et al. [15] and Fomin [16] extended the estimates, which were made by Moffatt [3] for Newtonian fluids, for the case of a generalized Newtonian fluid. Scaling analysis provided in [16] for typical rotational moulding conditions shows that rimming flow is mainly dominated by the interaction of the gravitational and viscous forces. Most polymeric solutes used in rotational coating are non-Newtonian liquids, which exhibit shear-thinning behavior for moderate to high shear rates. Liquid polymers behave as Newtonian liquids near the free surface (at very low shear rates) and exhibit non-Newtonian characteristics above a certain transitional value of the shear rate, $\dot{\gamma}_t$. The importance of the non-Newtonian effects for the generalized Newtonian fluids is characterized by the shear-thinning number $Wi=\lambda\Omega/\delta$, where Ω is the characteristic angular velocity of the rotating cylinder, δ is the ratio of the characteristic thickness of the film and the radius of the cylinder, and $\lambda \approx (\dot{\gamma}_t)^{-1}$ is a typical time scale for liquid polymers, which is well documented [19] and normally stays in the range of 10^{-2} - 10^{-1} second. In some situations, e.g. higher speed of rotation, thinner liquid layer or smaller transition shear rate, the value of *Wi* can be quite large which illustrates the dominating role of the non-Newtonian effects. Results of numerical computations available in [15] show that a Carreau fluid exhibits power-law behavior for large values of *Wi*. For instance for *Wi*=8, the

results obtained with the Carreau and power-law models practically coincide. A similar conclusion was made in [16] for Ellis fluids. Hence, flows that can be characterized by high values of shear-thinning number can be modeled by the power-law constitutive equation.

Our main concern is the rotational molding of highly viscous polymers [1, 2] that exhibit non-Newtonian shear thinning or shear thickening behavior. We are particularly interested in eliminating possible instabilities and providing the criteria for the steady-state flow in order to obtain a continuous and smooth coating film on the wall of the horizontal cylinder. Our current research focuses on the analysis of rimming flow of highly viscous non-Newtonian polymeric fluid and the transition of this time-dependent flow to a steady-state mode. Analysis of non-steady, non-Newtonian rimming flow is based on the numerical solution of the governing equation for the liquid film thickness for power-law and Ellis fluids. The factors that affect the length of the transition period to the steady-state are defined, and behavior of the liquid film during this period is illustrated. For reacting rimming flow, the stabilizing effect of chemical reaction that increases the viscosity of the fluid is demonstrated.

2 Governing Equations

In the previous studies [3, 4, 5, 14], the condition of the maximum supportable load for a Newtonian liquid was formulated in a form of the inequality $q \leq q_{max}$, where q is the mass flux through the cross-section of the liquid layer on the wall of the cylinder, and q_{max} is the maximal supportable value. In this case, the fluid film thickness is a smooth function of the azimuthal coordinate. The horizontal rotating cylinder is assumed to be of infinite length and a cylindrical coordinates system (r, θ, z) is located with the z-axis coinciding with the axis of the cylinder so the flow picture is two-dimensional (Fig.1).

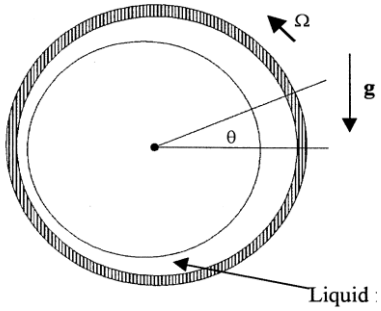

Fig. 1 A schematic of rimming flow on the inner wall of a horizontal cylinder

Non-dimensional mass flux $q>0$ can be readily computed from equation:

$$q = \int v_\theta dR\,, \tag{1}$$

where v_θ is the non-dimensional velocity along the wall of the cylinder, h is the non-dimensional thickness of the liquid film, and R is the non-dimensional radial coordinate (see Nomenclature). Expression for the fluid velocity v_θ in (1) can be obtained from the mass and momentum conservation equations. The characteristic scales for the variables in rimming flow are obvious and well documented [15, 16]. As a result of applying scale analysis to the governing equations and boundary conditions, three non-dimensional parameters are defined

$$\delta = \frac{h_0}{r_0} = \left(\frac{\mu_0 \Omega}{\rho g r_0}\right)^{1/2} << 1, \quad \mathrm{Re}\,\delta^2 \frac{\rho \Omega r_0^2}{\mu_0} << 1, \quad C_B = \delta^2 \frac{\sigma}{r_0 \mu_0 \Omega} << 1. \tag{2}$$

Parameter δ, which represents the ratio of characteristic thickness of the film h_0 and the radius of the cylinder r_0 , is obtained by equating the viscous and gravity forces. For rotational moulding, the value of □ is in the range of 10^{-2} to 10^{-1}. The Reynolds number, Re, characterizes the ratio of inertial to viscous forces and C_B is the inverse of the Bonds number which characterizes the ratio of capillary to gravitational forces. Exploiting the fact that □, Re, and C_B are very small (and to this end ignoring the terms of O(□, C_B, Re)), we may reduce the non-dimensional continuity and momentum equations in cylindrical coordinates to:

$$\frac{\partial v_R}{\partial R} + \frac{\partial v_\theta}{\partial \theta} = 0 \tag{3}$$

$$\frac{\partial P}{\partial R} = 0, \tag{4}$$

$$-\frac{\partial P}{\partial \theta} + \frac{\partial \tau_{R\theta}}{\partial R} = 0. \tag{5}$$

For the generalized Newtonian fluid, which is a purely viscous liquid, the components of the stress tensors reduce to the following form:

$$\tau_{R\theta} = \eta \frac{\partial v_\theta}{\partial R}, \quad \tau_{RR} = 2\eta \frac{\partial v_R}{\partial R}, \quad \tau_{\theta\theta} = 2\eta \frac{\partial v_\theta}{\partial \theta} \tag{6}$$

where η is a function of the second invariant of the rate of deformation tensor and of the concentration of reacting solute, *c*.

Stress balance conditions on the free surface $R = h$ reduce to $\tau_{\theta R} = 0$ and $p = 0$. On the wall of the cylinder $R = 0$, the fluid velocity vector is $v = (v_r, v_\theta) = (0, u(t))$, where $u(t)$ is the non-dimensional angular velocity of the cylinder. The kinematic condition on the free surface, which represents the fact that the normal velocity of particles at the surface matches the normal velocity of the surface itself, reduces to

$$\frac{\partial h}{\partial t} + v_R + v_\theta \frac{\partial h}{\partial \theta} = 0 \tag{7}$$

Combining equations (3) and (7) yields:

$$\frac{\partial h}{\partial t} + \frac{\partial q}{\partial \theta} = 0, \tag{8}$$

where *q* is defined by equation (1).

In the reacting rimming flow [2] of chemically initiated polymeric fluid, due to the transient nature of the reaction, viscosity increases with time and flow stops once the polymeric concentration reaches the gelling point. Assuming that mass diffusion is negligible and temperature is constant, the concentration field can be considered uniform, *c=c(t)*. For thin films and pre-mixed fluids these assumptions are reasonable [20]. The chemical reaction is initiated by raising the temperature beyond the gelation temperature for the given reacting liquid. We assume that the polymeric reaction can be described by a simple kinetic equation

$$\frac{dc(t)}{dt} = -\kappa c(t), \tag{9}$$

The non-dimensional initial condition is $c = 1$ at t=0.

Solution of the equations (4) and (5), subject to boundary conditions, is rather straightforward,

$$\tau_{\theta R} = (h - R)\cos\theta\,. \tag{10}$$

It follows from the formulae (6) with an accuracy of $O(\square)$, that

$$\tau_{\theta R} = -\eta(|\dot\gamma|, c)\dot\gamma\,, \qquad \dot\gamma = \frac{\partial v_\theta}{\partial R} \tag{11}$$

Without loss of generality, following Qi and Johnson [20], we use the viscosity model $\eta = \frac{1}{c(t)}\mu(\dot\gamma)$. Hence, accounting for the solution of equation (9), equation (11) can be rewritten as:

(12)

Introducing a function $G(x)$ as the inverse of function $\mu(x)x$, so that for $x>0$ equations $G[\mu[\mu(x) = x$ and $\mu(G(x))G(x) = x$ are satisfied, the stress-strain correlation (12) can be converted to its inverse form:

(13)

Accounting for the corresponding boundary conditions, the unknown velocity v_θ, with an accuracy of $O(\square)$, can be readily obtained as

$$v_\theta = [u(t) - \exp(-\kappa t)\mathrm{sgn}(\tau_{\theta R})\int_0^R \exp(\delta\delta R)G(|\tau_{\theta R}|)dR]\exp(-\delta R)\,, \tag{14}$$

Hence, the non-dimensional flux q can be presented as

$$q = u(t)h - \exp(-\kappa t)\mathrm{sgn}(\tau_{\theta R})\int_0^h (h - R)G(|\tau_{\theta R}|)dR \tag{15}$$

Substituting (15) into equation (8) gives the leading order governing equation which models the evolution of the liquid film

$$\frac{\partial h}{\partial t} + \frac{\partial}{\partial\theta}\left[u(t)h - \exp(-\kappa t)\mathrm{sgn}(\cos\theta)h^2\int_0^1 yG[h|\cos\theta|y]dy\right] = 0 \tag{16}$$

Unfortunately, the explicit representation of G is available only for a few models of non-Newtonian fluids [16]. Namely, for the power-law model, $\tau_{\theta R} = -|\dot\gamma|^{n-1}\dot\gamma$, obviously $G(x) = x^{1/n}$, and for the Ellis model, $\dot\gamma = -\tau_{\theta R}\left[1 + (Wi|\tau_{\theta R}|)^{1/n-1}\right]$, $G(x) = x(1 + x^{1/n-1})$, where and n and Wi are constant parameters.

For the Power-Law model, equation (16) converts to

$$\frac{\partial h}{\partial t} + \frac{\partial}{\partial\theta}\left[u(t)h - \exp(-\kappa t)\mathrm{sgn}(\cos\theta)|\cos\theta|^{1/n}\frac{n(h)^{\frac{2n+1}{n}}}{2n+1}\right] = 0\,, \tag{17}$$

and for the Ellis, the equation converts to

$$\frac{\partial h}{\partial t} + \frac{\partial}{\partial\theta}\left[u(t)h - \exp(-\kappa t)h^3\cos\theta\left(\frac{1}{3} + \frac{n(Wi|\cos\theta|h)^{\frac{1-n}{n}}}{2n+1}\right)\right] = 0 \quad (0 < n < 1) \tag{18}$$

3 Numerical Results and Discussion

Numerical solutions of equations (17) and (18) modeling the non-steady flow of liquid polymer are obtained for various flow conditions and values of characteristic parameters. Even though the governing equations for power-law and Ellis models have many common features, there are still some differences due to intrinsic properties of the constitutive equations. For example, as it was indicated in [15,16], most polymeric solutes used in rotational moulding exhibit non-Newtonian behavior for relatively high shear rates (near the wall of the cylinder) and behave as Newtonian liquids near the free surface (at very small shear rates). Such behavior can be well modeled by Ellis constitutive equation [16], especially when *Wi* is relatively low and flow index $n \square 1/3$. In some situations, e.g. higher speed of rotation or/and very thin liquid layer, the value of *Wi* can be quite large which corresponds to the dominating role of non-Newtonian effects. As it was proved in [15, 16], the flows, which are characterized by the high values of shear-thinning number, can be modeled by the power-law constitutive equation. Therefore, both power-law and Ellis equations can be used in modeling the process of rotational moulding for various polymeric solutes and technological conditions.

Defining an initial distribution of the liquid polymer $h(\theta) = h_{in}$, the governing equations (17) and (18) can be solved to demonstrate the evolution of the film over time.

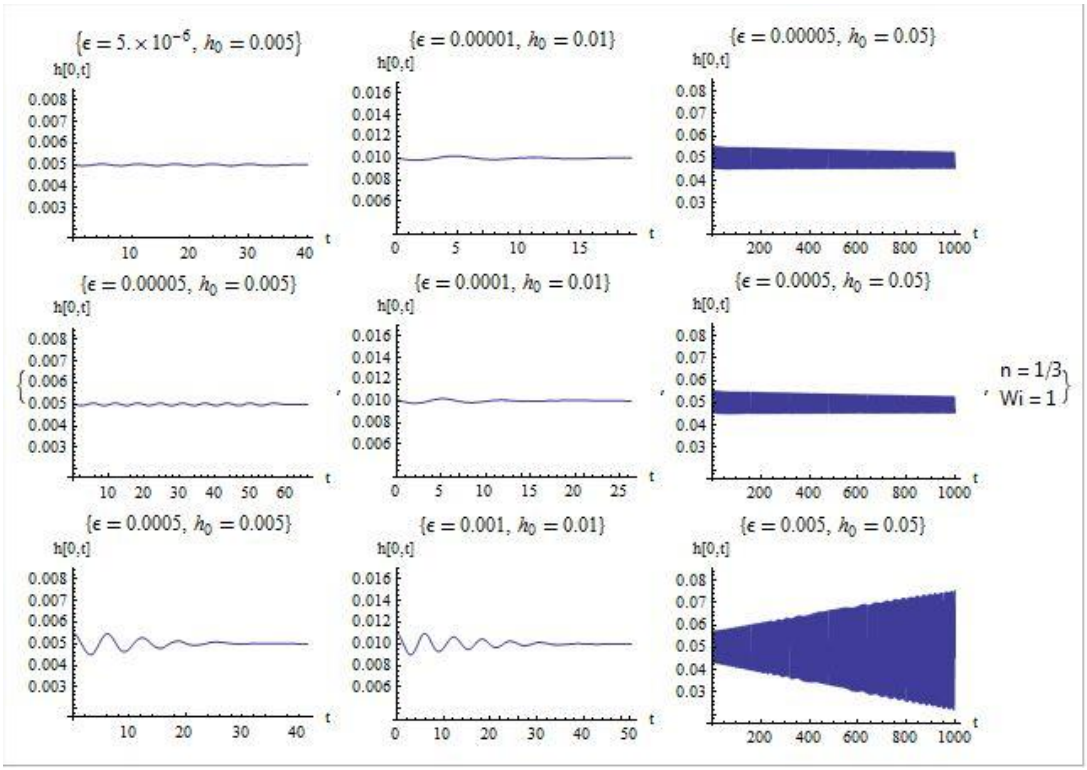

Fig. 2. Thickness of the liquid film in the rimming flow of the Ellis fluid without chemical reactions.

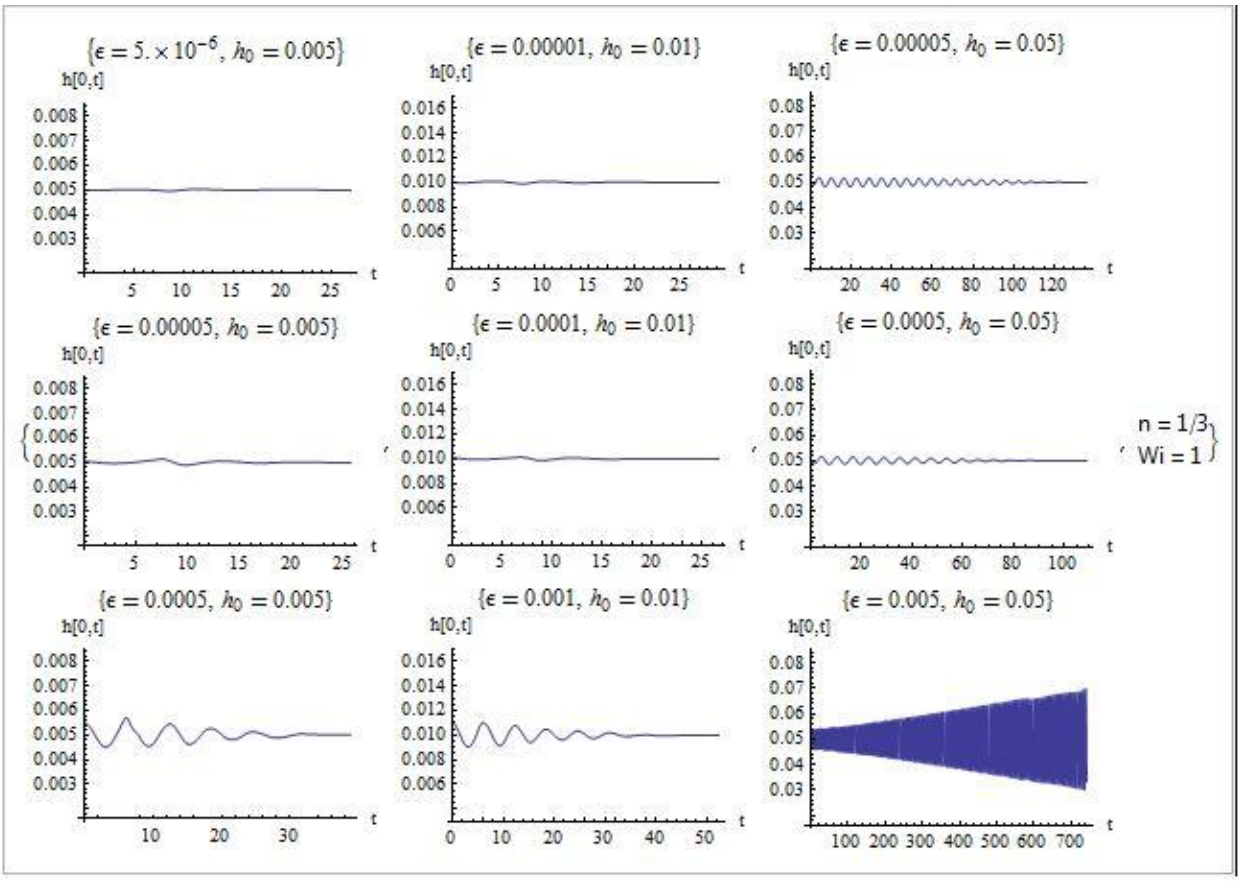

Fig. 3. Thickness of the liquid film in the rimming flow of the Ellis fluid with chemical reactions.

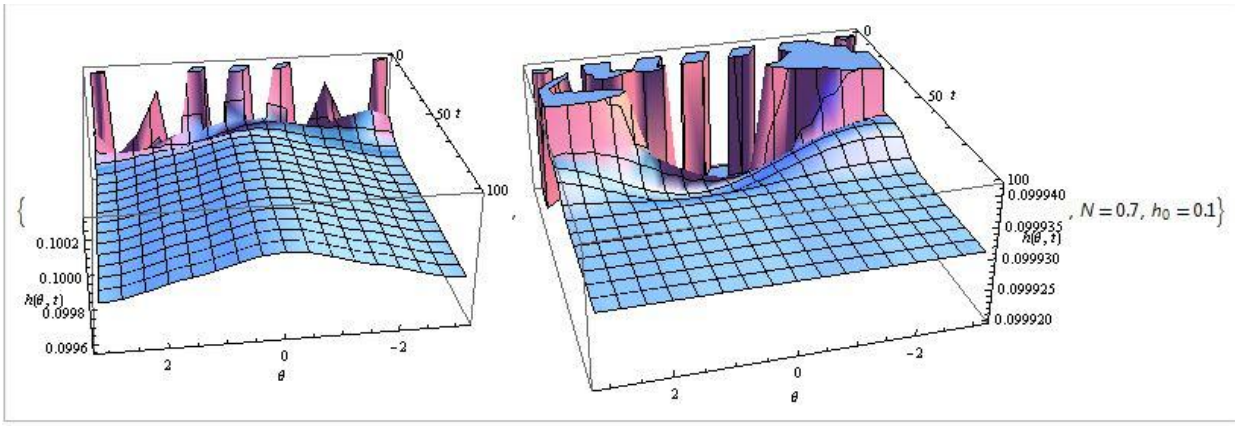

Fig. 4. Thickness of the liquid film in the rimming flow of the power-law fluid without chemical reaction.

Figures 2-4 illustrates the variations of the liquid film on the wall of the cylinder at the point $\theta = 0$ versus time. In order illustrate the rate at which the rimming flow can reach a steady-state

mode, the initial conditions are chosen in a form of the perturbed steady state solution $h_{st}(\theta)$ of equations (17) and (18), i.e. $h(\theta,0) = h_{st}(\theta) + \varepsilon\cos(f\theta)$, where ε is the amplitude of disturbance and f is the frequency. In Figs. 2-4 h_0 is the given value of $h_{st}(\theta)$ at $\theta = 0$, so that the boundary condition for the steady-state problem is $h_{st}(0) = h_0$. As it can be readily seen for the low amplitude of perturbation, after some initial fluctuations, the fluid reaches a steady state regime. However, if the amplitude is above certain value, the flow remains non-steady. This is attributed to the intrinsically non-stable character of the rimming flow [8-13].

Computations (made in absence of chemical reaction, $\kappa = 0$) demonstrate that the film thickness is not uniformly distributed along the wall of the cylinder even in the steady-state. In many industrial applications, the goal is to obtain a uniform coating on the surface. One suggested method to produce a uniform liquid layer is the implementation of a chemical reaction that increases the viscosity of the fluid. The polymeric reaction is modeled by the first-order kinetic equation (9). Due to the exponential growth of viscosity, in this case the governing equation for liquid film thickness reduces to $\frac{\partial h}{\partial t} + \frac{\partial h}{\partial \theta} = 0$ as $t \to \infty$, which results in a uniform liquid film for the given initial conditions. For the conditions studied, the reactive flow with increasing fluid viscosity yielded a solution that most closely resembled a uniform coating along the inside wall of the cylinder. Another advantage of the presence of chemical reaction is its stabilizing effect. Fig. 3 illustrates that for the reactive rimming flow, the transition time from non-steady to steady-state regime substantially reduces.

Conclusion

The non-steady flow of the power-law and Ellis fluids was analyzed numerically. The influence of characteristic parameters on the evolution of the liquid film has been investigated. Fluids with larger flow indexes take longer to reach steady flow. Ellis fluids with high values of *Wi* exhibit a longer period of non-steady flow before reaching a steady state. As expected, it takes longer time for larger volumes of fluid in the cylinder (larger values of *q*) to reach the steady state.

Viscosity-increasing chemical reactions cause the liquid to form a uniform film along the cylinder, making them desirable for the rotomoulding technology. The inclusion of these factors makes the model more physically accurate and realistic, and therefore more useful for a variety of industrial applications.

NOMENCLATURE

c	concentration of reacting solute, $c = c^*/c_0$
c_0	initial concentration of reacting solute
C_B	inverse to the Bond number
De	Deborah number
G	inverse function to $\mu(x)x$
g	gravity acceleration
H_0	mean thickness of the liquid layer
H_c	critical mean thickness of the liquid layer
h	thickness of the liquid layer, $h = h^*/h_0$
h_c	critical thickness of the liquid layer at $\square$=0

h_0	characteristic thickness of the liquid layer
k	consistency parameter in a power-law constitutive equation
n	flow index
p	pressure, $P = P^* / (\rho \Omega^2 r_0^2)$
q	mass flux
r	radial coordinate , $r = r^* / r_0$
r_o	radius of the cylinder
R	non-dimensional radial coordinate, $R = (1 - r)/\delta$
Re	Reynolds number as defined by equation
t	time , $t = t^* \Omega$.
v_R, v_θ	radial and azimuthal components of the fluid velocity,
W	total mass of the liquid
Wi	shear-thinning number, $Wi = \lambda\Omega / \delta$

Greek symbols

δ	ratio of the characteristic liquid layer thickness and radius of the cylinder , $\delta = h_0 / r_0$
$\dot{\gamma}$	shear rate
λ	relaxation time
κ	parameter in a kinetic equation that defines the rate of polymeric reaction
η	dynamic viscosity
η_0	characteristic viscosity ($\mu_0 = k(\Omega / \delta)^{n-1}$ for the power-law fluid)
θ	azimuthal coordinate
ρ	liquid density
σ	surface tension
$\tau_{\theta R}$, τ_{RR} , $\tau_{\theta\theta}$	non-dimensional components of the deviator of the stress tensor,
$u(t)$	angular velocity of the cylinder
ω	characteristic (maximal) angular velocity of the cylinder

Superscripts

*	dimensional quantities

Subscripts

0	characteristic quantity
θ, R	azimuthal and radial components, respectively

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