

# Threshold of ribbing instability with Non-Newtonian fluids

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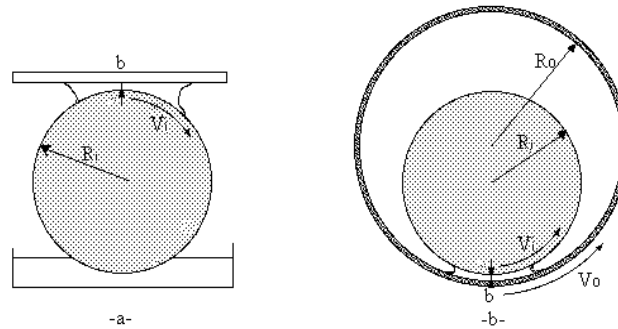
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In most industrial applications the coated fluids are complex and exhibit non Newtonian properties such as shear-thinning, viscoelasticity or thixotropy. However the mechanisms of the ribbing instability that often limits the operating ranges are not fully understood even with simple Newtonian fluid. Clearly there is still a need for fundamental research in this field, ever theoretical, numerical or experimental. With these ideas in mind we revisited the determination of the onset of ribbing in two model set-ups and with two classes of polymers solutions. The results are compared to the Newtonian case. The critical velocity for shear-thinning fluid is quite high, but when expressed in effective capillary number  $Ca^*$  is only slightly smaller than for Newtonian fluid. With viscoelastic fluid, the lowering of the onset is more important (a factor 10). Such results are in agreement with some previous experimental results<sup>1,2</sup> and predictions<sup>3</sup>.

## 1. EXPERIMENTAL SET-UP

We use two roll-coating set-ups. The first one corresponds to a Dural cylinder of radius  $R = 50$  mm and length 420 mm, which is partially immersed in the solution (Fig. 1a)<sup>4</sup>. The rotation of the cylinder drags the fluid out of the tank (if it is viscous enough and if it wet the metal). The fluid then fills the gap of adjustable thickness  $b$  that exists at the top of the set-up between the cylinder and an horizontal glass plate. If the velocity is large enough, the downstream meniscus (upper right on Fig. 1a) becomes wavy and the coated surface uneven. The second set-up (Fig. 1b) is a two-rolls coating set-up where one cylinder is inside the other allowing an easy visualisation of the meniscus (journal bearing geometry). The two glass cylinders, have a length of 420 mm and radius  $R_i = 33$  mm and  $R_o = 50$  mm respectively<sup>5</sup>. Thus the equivalent radius of curvature in the nip is  $R = R_o R_i / (R_o - R_i)$

97 mm. Various studies of the ribbing instability has been made in such set-ups with Newtonian fluids (Silicon oil)<sup>4,5</sup>. In the present paper we present new results when aqueous solution of polymers are used. Some are semi-rigid polymers (Xanthan) others are long chain polymers (AP 45). The first ones exhibit shear-thinning properties, the others shear-thinning and viscoelastic properties. The rheology of the fluid have been investigated and is presented in a companion paper<sup>6</sup>.



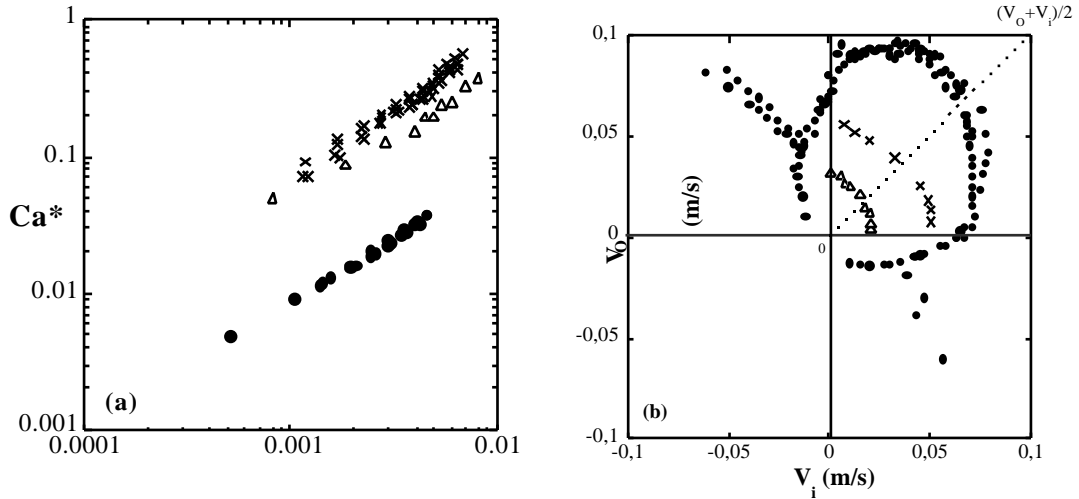
**Fig. 1** : Sketch of the experimental set-ups : (a) The cylinder/plane geometry. (b) The two-rolls cell.

## 2. EXPERIMENTAL RESULTS ON THE INSTABILITY THRESHOLD

When one cylinder is rotating, viscous drag coats the moving surface. In permanent conditions both meniscus (upstream and downstream ones) take well define positions which depend on the angular velocity. Above a critical velocity, the downstream meniscus becomes unstable i.e. develops sinusoidal perturbations. For Newtonian fluids the onset is characterised by a critical capillary number  $Ca = \mu V/T$  where  $\mu$  is the dynamical shear viscosity,  $T$  the air-liquid surface tension and  $V$  the tangential velocity. The critical capillary number depends on the aspect ratio of the cell i.e. of the ratio  $b/R$ . Most of the published results are known to collapse on a law  $Ca^* \approx 40$ .<sup>7</sup> Our results with PDMS (Silicon oil) also do. When shear-thinning fluids are used,  $Ca$  is no more well defined. Usually an effective capillary number is defined as  $Ca^* = \mu(\dot{\gamma})V/T$  where  $\dot{\gamma} = V/b$  is the estimated shear rate in the nip. In Fig. 2a we present our results on  $Ca^*$  versus  $b/R$  for Newtonian, shear-thinning and viscoelastic fluids. The critical velocity for shear-thinning fluid is very high, but when expressed in  $Ca^*$  is only slightly smaller than for Newtonian fluid. With viscoelastic fluid as AP45, the lowering of the  $Ca^*$  at onset is more important (a factor 10). Such results are in agreement with previous experimental results<sup>1,2</sup> and predictions<sup>3</sup>.

In the journal bearing geometry, both cylinders can rotate and the critical velocities at onset define a curve in the plane of the two tangential velocities (Fig. 2b). This curve for Newtonian fluids is almost a quarter of a circle for co-rotating cylinders ( $V_i$  and  $V_o > 0$ ). For Non-Newtonian ones the curves are different qualitatively and quantitatively, however, they are difficult to compare as there is no straightforward definition of effective capillary numbers. For example the shear, if defined in the gap as  $(V_i - V_o)/2b$ , is zero or very small for exact co-rotation of the cylinders, leading to an overestimation of the apparent viscosity  $\mu(\dot{\gamma})$ . More studies taking into account the shear induced by the pressure gradient in the gap are thus necessary.

In the next sections we examine the influence of shear-thinning and viscoelastic behaviours on pressure profile along the gap between cylinders. This procedure will give some insight on the modification of the basic flow and thus a first step in understanding the modification of the instability threshold.



**Fig. 2** : Comparison of the threshold of ribbing with Newtonian (× : PDMS) and Non-Newtonian fluids (the solutions are made of 3000 ppm polymers ( : Xanthan ; ● : AP45) in water/glycerol mixture) : (a) Critical capillary number  $Ca^*$  versus the aspect ratio of the cell in a log-log plot for the cylinder/plane geometry ; (b) Onset of the instability in the plane of the two tangential velocities in the two-rolls cell.

### 3. SHEAR-THINNING EFFECTS

The first aspect we should focus our attention is how shear-thinning behaviour modifies pressure conditions at the nip. For that purpose, we will briefly describe the approach followed by several authors<sup>8,9</sup>. All the quantities are made dimensionless (streamwise length by  $\sqrt{Rb}$ , transverse length by  $b$ , velocity by the wall velocity  $V$ ). Under lubrication approximation, for a power-law fluid satisfying

$$\mu = m \frac{V}{b} \left| \frac{u}{y} \right|^{n-1} \quad (1)$$

a highly non linear set of equations can be established between flow rate, pressure gradients and shear rate field<sup>10</sup>. Dien and Elrod<sup>11</sup> proposed an approximate solution to the problem by considering small pressure gradients along the nip, leading to a Reynolds-like equation, which can be integrated subjected to pressure end conditions: in our case, neglecting the effect of the menisci, this means,  $P(\pm \infty) = P(\pm \infty) = 0$  :

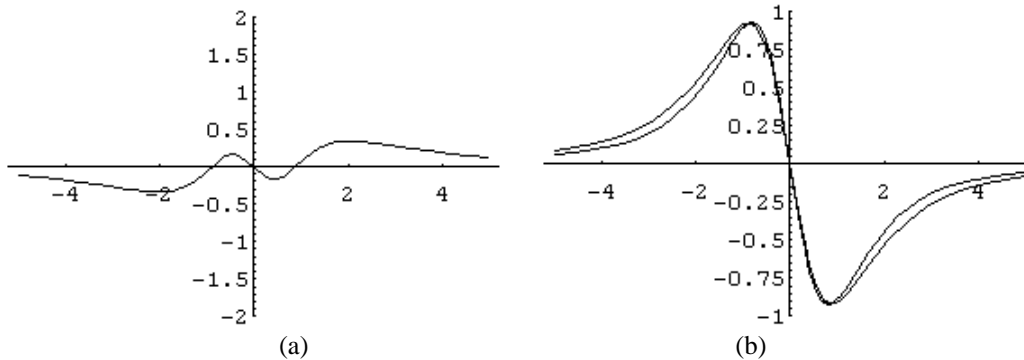
$$\frac{P}{x} = \frac{6n[h(x) - 2Q]}{h(x)^{n+2}} \quad (2)$$

where  $Q$  is the flow rate in the gap and  $h(x)$  the local thickness of the cell in  $x$ . The pressure has been made dimensionless by  $mV^n\sqrt{Rb}/b^n$ .

In order to visualise the effect of viscosity dependence on shear, a regular expansion of this expressions can be made :  $P(x) = P_N(x) + P(x)$  (N denotes Newtonian pressure and  $n = n - 1$ ). Considering a parabolic shape for the nip ( $h = 1 + x^2/2$ ) this leads to a pressure correction of the form :

$$P(x) = P_N(x) \frac{1}{3} - \text{Log} \left( 1 + \frac{x^2}{2} \right) \quad (3)$$

This pressure correction is the product of the Newtonian pressure profile by a function of the thickness profile (Fig. 3a). Modified pressure profiles for a (negative) value of  $n$  are shown on Fig. 3b. As it can be shown, shear-thinning effects do not change dramatically pressure profiles under this approximation. Nevertheless, it is clear that pressure does decrease (since  $n < 0$ ) for shear-thinning liquids) on the downstream zone, where ribbing takes place.



**Fig. 3 :** (a) First order pressure correction  $P$  (weak shear dependence viscosity) for a parabolic profile of the nip ; (b) Pressure profile  $P(x)$  ( $n = 0.75$ ) with respect to the Newtonian case. Pressure on downstream position is decreased for shear thinning fluids.

#### 4. VISCOELASTIC EFFECTS

As for shear dependent fluids, a regular perturbative expansion of pressure can be arranged for purely viscoelastic fluids, as it has been shown by Tichy in a recent paper<sup>12</sup> for the problem of blade lubrication. This assumption resides in the fact that for almost any coating application, the Deborah number ( $De = V / \sqrt{Rb}$ ,  $\tau$  is the relaxation time of the fluid) is less than unity. So, a regular expansion of the

problem leads to  $P(x) = P_N(x) + De P_{De}(x)$  with  $De$  as the perturbative parameter. This time  $P$  is made dimensionless by  $\mu V \sqrt{Rb} / b^2$ .

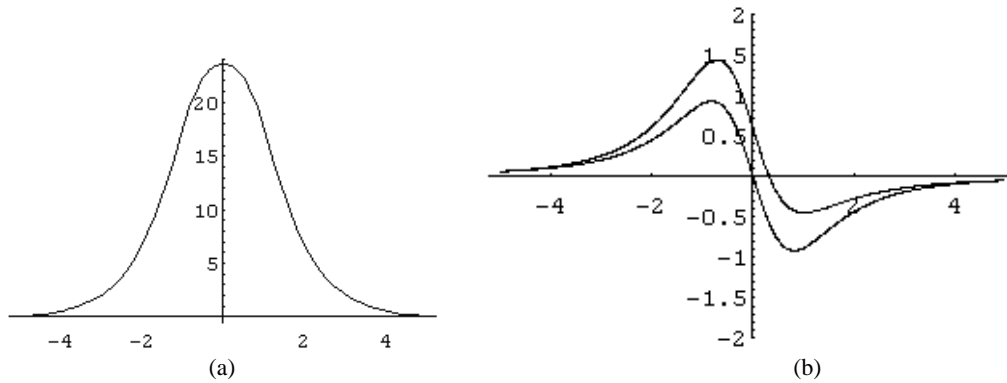
In order to take account of time-dependent effects, a convective Maxwell model is used, in order to keep track of deformation along the nip :

$$\rho \frac{d}{dt} \left( \frac{v_{ij}}{h} \right) = \mu \frac{d}{dx} \left( \frac{v_{ij}}{h} \right) \text{ with } L_v \left( \frac{v_{ij}}{h} \right) = -\frac{v_{ij}}{t} + v_m \frac{v_{ij}}{x_m} - \frac{v_m}{x_j} \frac{v_{ij}}{h} - \frac{v_m}{x_j} \frac{v_{ij}}{h} \quad (4)$$

This codeformational derivative introduces non linear terms in the set of equations, which has to be solved numerically. As in the previous section, a Reynolds-like equation can be found for the perturbative part of the pressure

$$\frac{d}{dx} \left( h^3 \frac{dP_D}{dx} \right) = h F_1(h) + \frac{h^2}{h} F_2(h) \quad (5)$$

( $F_1$  and  $F_2$  are functions of nip profile only). The most important aspect of this equation is that not only is a function of thickness profile but also of its derivatives. This second order equation for the profile can be solved by a Runge-Kutta boundary value algorithm, subjected to the usual restrictions ( $P(-\infty) = P(\infty) = 0$ ). The solution is shown on Fig. 4a for parabolic profile. A well defined pressure increment occurs when fluid traverses the minimum gap. This behaviour is in accordance with the results obtained by Tichy for positive curvature blades<sup>12</sup>. First normal stress difference usually increase shear (as it is shown in a companion paper<sup>6</sup>), which may explain this pressure excess on high shear zones.



**Fig. 4 :** Pressure profiles along the streamwise direction : (a) First order pressure correction  $P_{De}$  (weak viscoelasticity limit) for a parabolic profile ; (b) Pressure profile  $P(x)$  ( $De = 0.025$ ) compared to the newtonian case . Pressure is larger everywhere along the nip, especially near minimum gap.

## 5. CONCLUSION

Our first attempt in order to study ribbing instability for complex fluids consisted on the determination of the onset of ribbing as a function of minimum

gap between surfaces. For the case of journal bearing geometry, where both cylinders can rotate, a limit between stable and unstable regions on parameter space has been found.

For shear-thinning, inelastic fluids (Xanthan) threshold lies in the vicinity of the Newtonian case when expressed in effective capillary number. A regular expansion for weak shear-dependent viscosity showed little influence on pressure profiles. The same has been found for roll coating processes in other works<sup>12,13</sup>.

For viscoelastic fluids (PAAm polyacrylamide, POLYOX polyoxyethylene) a dramatic change in threshold has been found. Viscoelastic properties reduces ribbing threshold by a factor 10 in most cases, in accordance with previous results obtained by Bauman *et al.*<sup>1</sup>. Perturbative expansion of pressure (via convective Maxwell model) shows an increase in pressure everywhere along the nip, especially at higher shear zones. This pressure excess can be related to first normal stress difference increment with shear rate.

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1. Bauman, T. Sullivan, T. and Middleman, S. Ribbing Instability in Coating Flows : Effect of Polymer Additives, *Chem. Eng. Commun.* **14**, 35-46 (1982).
2. Grillet, A.M. Lee, A.G. and Shaqfeh, E.S.G. The stability of elastic fluid interfaces in coating flows, to appear in *J. Fluid Mech.*
3. Ro, J.S. and Homsy, G.M. Stability of viscoelastic interfacial flows between non-parallel walls : the ribbing instability of viscoelastic coating flows, preprint.
4. Bellon, L. Fourtune, L. Ter Minassian, V. and Rabaud, M. Wave-Number Selection and Parity-Breaking Bifurcation in Directional Viscous Fingering, *Phys. Rev. E* **58**, 565-574 (1998).
5. Michalland, S., Rabaud, M. and Couder, Y. The Instability of the Upstream Meniscus in the Directional Viscous Fingering, *J. Fluid Mech.* **312**, 128-148 (1996).
6. Pauchard, L. Varela López, F. Rosen, M. Allain, C. Perrot, P. and Rabaud, M. On the Effects of Non-Newtonian Fluids Above the Ribbing Instability, in this volume.
7. Coyle, D.J. Macosko, C.W. Scriven, L.E. *J. Fluid Mech.* **216**, 437-458 (1990).
8. Savage, M. D. Variable speed coating with purely viscous non newtonian fluids, *J. App. Math. Phys.* **34**, 358 (1983).
9. Sinha, P. Singh, C. Lubrication of a cylinder on a plane with a non newtonian fluid considering cavitation, *Trans. ASME J. Lubrication Technol.* **104**, 168 (1982).
10. Ross, A. B. Wilson, S. K. and Duffy, B. R. Blade coating of a power-law fluid, *Phys. Fluids* **11**, 958 (1999).
11. Dien, I.K. and Elrod, H. G. A generalized steady-state Reynolds Equation for non newtonian fluids, with application to journal bearings, *Trans. ASME J. Lubrication Technol.* **105**, 385 (1983).
12. Tichy, J. A. Non-newtonian lubrication with the convected maxwell model, *Trans. ASME J. Tribology* **118**, 344 (1996).
13. Tekic, M.N. and Popadic, V.O. Coating using pseudoplastic liquids, Ch. 35, Polymer rheology and processing, *Encyclopedia of Fluid Mechanics*, N. Cheremisinoff, Ed. (1990).