

On the effects of Non-Newtonian fluids above the ribbing instability

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The topology of the flow is of most importance for the final properties of coated surfaces. This topology is mainly controlled by the viscosity of the liquid sheet. However, in most coating systems this viscosity is not a constant and depends on a variety of experimental parameters, the most important being the mechanical stress the fluid is exposed to. Thus, for Non-Newtonian fluids in roll coating geometry, several changes are observed in the manifestation of the instability. In this article, we present first the rheological properties of some Non-Newtonian fluids used, then the experimental results on the shape of the unstable meniscus observed above onset in a journal bearing geometry described in a previous paper¹.

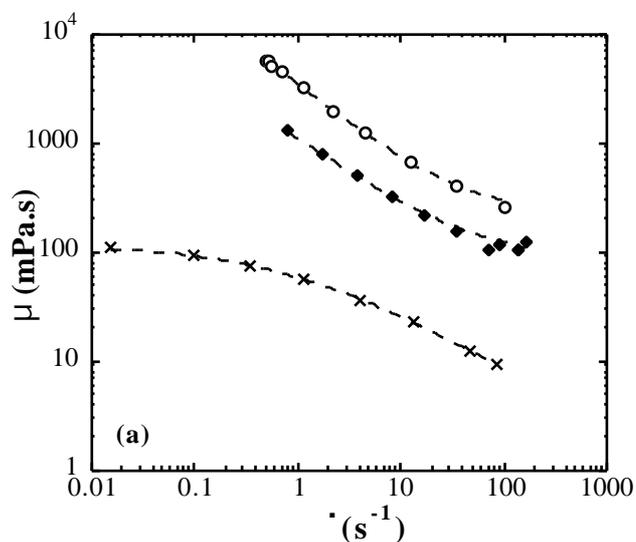
1. RHEOLOGICAL PROPERTIES

The liquids used are semi-dilute solutions of two polymers in a mixture of water and glycerol (usually 10% and 90% respectively). One fluid is a solution of a semi-rigid polymer, a polysaccharide (Xanthan), exhibiting strong shear-thinning properties. The second fluid is a solution of a flexible polymer, an hydrolysed polyacrylamide (Separan AP45), exhibiting shear-thinning but also strong elastic properties. For each solutions, the apparent shear viscosity μ was measured in a large range of shear rate $\dot{\gamma}$ at 20°C, using a constant stress rheometer (Rheological Stress-Tech) in a cone-plate configuration. The steady state shear viscosity decreases with the shear rate and increases with the polymer concentration (Fig. 1a). These measurements are characteristic of a shear-thinning behavior and well fitted by a Carreau law : $\mu = \mu_s + \frac{\mu_0 - \mu_s}{1 + (\beta \dot{\gamma})^2}$ where μ_s is the viscosity of the solvent, μ_0 is the low shear viscosity of the solution and with the exponent $0 < \beta < 1$.

In addition, a very important effect for the flow of concentrated solutions of flexible polymers is the existence of large normal stress. Indeed when such solution is sheared in the xy plane by a velocity gradient dv_x/dy , the pressure on a wall located in the xz plane is augmented by the first normal stress difference, noted N_1 . When in the lagrangien frame of reference of a fluid particle the flow is non stationary, the structure of the flow is extremely sensible to the existence of such

normal forces. One of the most classical phenomenon characterising elastic polymer solutions is die swell effect. Such liquids emerging from a capillary tube at low Reynolds number form an expanding jet which diameter increases to a maximum value downstream of the exit. The maximum diameter may be several times larger than the capillary inner diameter, depending on the imposed flow rate and thus on the wall shear rate (Fig. 2). The first normal stress difference can be directly measured with our rheometer or deduced from the swell ratio using the Tanner's model² a method that allows to reach larger flow rate. The variations of N_1 as well as the normal stress coefficient Ψ_1 (defined as $\Psi_1 = N_1 / \dot{\gamma}^2$) versus the permanent shear rate $\dot{\gamma}$ are presented on Fig. 1b for the polyacrylamide solution. For the Xanthan solutions, no significant normal stress can be measured.

In a cylinder/plate geometry or in a journal bearing one induced by the walls, the effective shear rate can be estimated as planar Couette shear, thus as the ratio V/b , V is the difference of tangential velocities of the two surfaces and b the clearance between them ; such estimation neglects all the shear induced by the pressure gradient along the flow. With the selected fluids, the instability occurs when the shear rate is in the range 10 to 100 s^{-1} , thus in the region where the shear-thinning behaviours are significant. The experimental results for the onset values of the ribbing instability are presented in a companion paper¹.



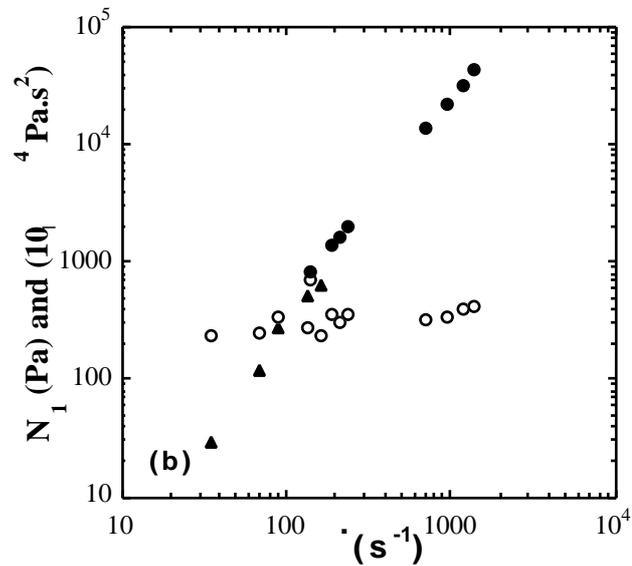


Fig. 1 : (a) Variations of the shear viscosity μ versus the shear rate : (○) Xanthan at a concentration of 3000 ppm in weight in a mixture of 90 % glycerol and 10 % of water ; (◆) AP45 at 3000 ppm in the same solution and (×) Xanthan at 1000 ppm in pure water. Dashed lines are adjustment by Carreau laws with respective exponent $n = 0.71, 0.82$ and 0.55 .

(b) First normal stress difference N_1 and coefficient β_1 versus the shear rate for AP45 : (▲) N_1 measured with the rheometer ; (●) N_1 deduced by the die swell and (○) β_1 calculated with the previous points. For the Xanthan solutions, N_1 is so small that it cannot be measured ($N_1 < 5$ Pa).

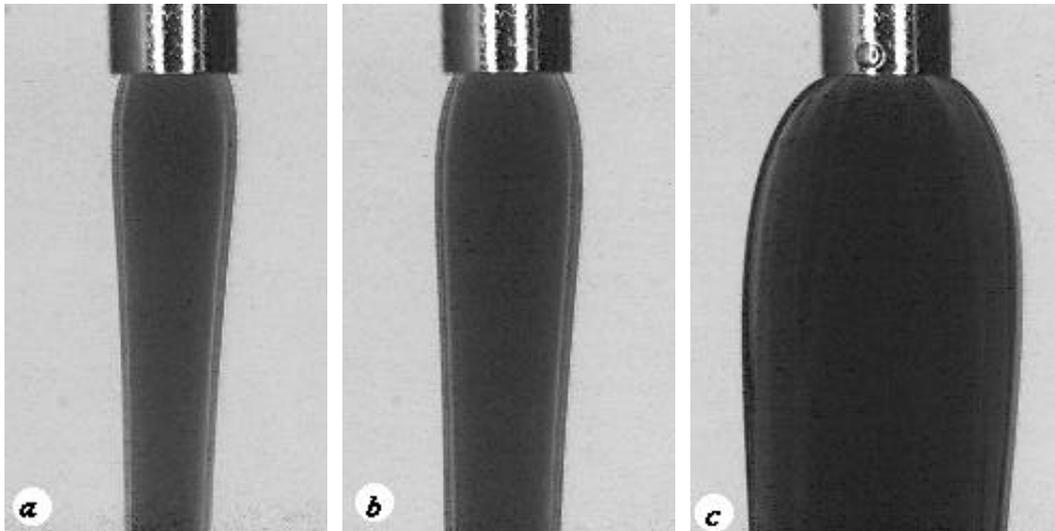


Fig. 2 : Photographs of the die swell jet of a 3000 ppm for AP45 in a mixture of 90 % glycerol and 10 % of water, for increasing values of the wall shear rate ($\dot{\gamma} = 29, 140$ and 714 s^{-1}). Note that the maximum diameter of the jet is obtained at a distance of the orifice. This distance is related to the short time of relaxation of the solution ; this time is here of the order of 0.1 s but decreases with $\dot{\gamma}^{-2}$.

2. SHAPE OF THE AIR FINGERS ABOVE THRESHOLD

In a roll coating geometry, when the angular velocity of a roll exceeds a critical value, a steady state of a stable fingering pattern with characteristic wavelength is formed. Above onset, the amplitude of such deformation of the interface, i.e. difference between the front and the back of the fingers, were measured as a function of distance to threshold $\Delta V = (V - V_c)/V_c$ (Fig. 3a). On Fig. 3b the evolution of the wavelength is plotted versus distance to threshold. As for newtonian fluids the wavelength decreases as ΔV^{-3} . Figure 4 shows pictures of Xanthan-air and AP45-air interfaces as well as the shape of a newtonian oil-air interface for comparable ΔV . With Xanthan as well as with AP45, the shape of the air fingers evolves continuously to an angular shape comparable to a gothic arch with almost an angle at the tip of the finger. Such evolution was never observed with newtonian fluids, but is observed also for Boger fluids⁴. Thus this change of the form is not only due to the shear-thinning properties of our fluids.

On figure 5 we present such evolution of the shape of the finger when increasing ΔV for a Xanthan solution.

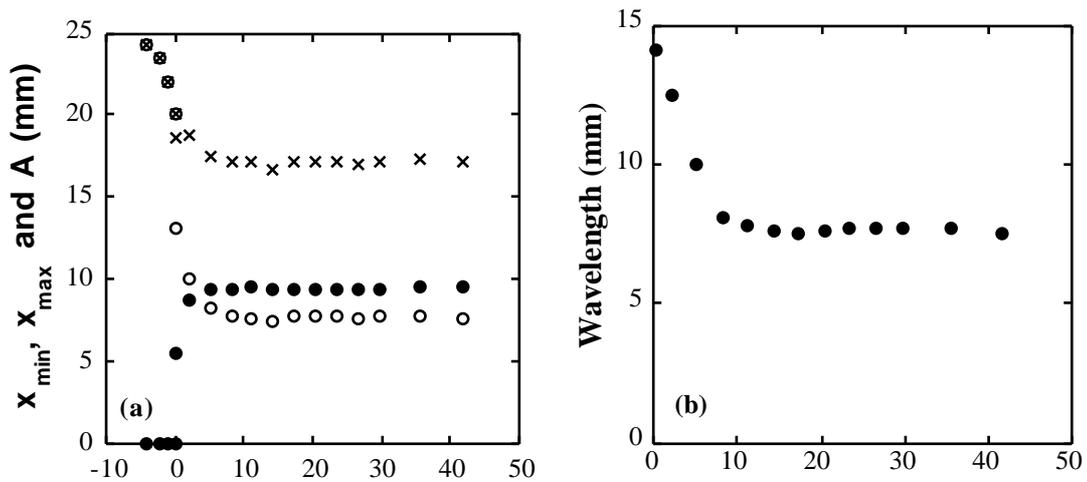


Fig. 3 : For a 3000 ppm Xanthan solution : (a) Evolution of the position of the meniscus $x_{m \text{ back}}$ (X) and $x_{m \text{ front}}$ (O) below and above threshold ($V_c = 30$ mm/s) as well as the amplitude A of the finger $A = (x_{m \text{ front}} - x_{m \text{ back}})$ above threshold (●) and (b) evolution of the wavelength of the pattern as a function of ΔV .

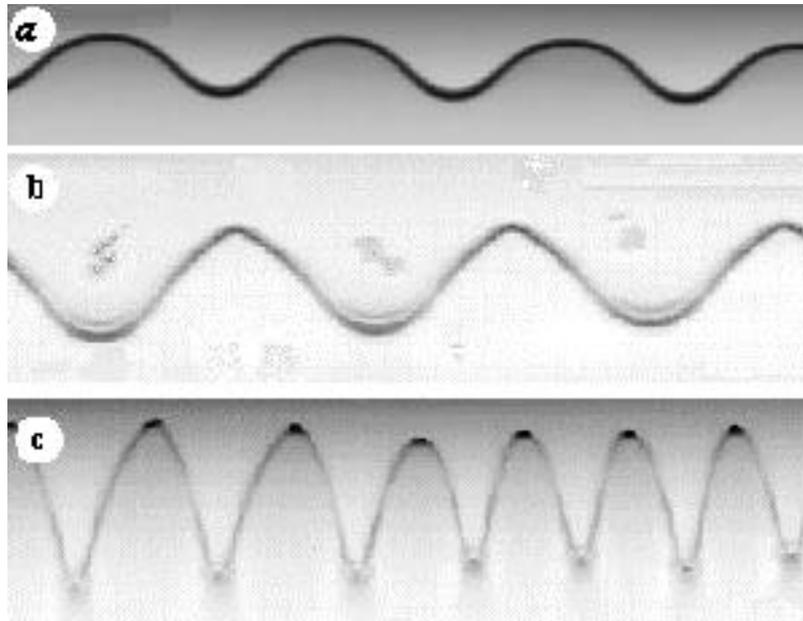


Fig. 4 : Three photographs of the steady state of the downstream meniscus for a velocity of the inner cylinder above the critical value (the outer cylinder being fixed), at about the same distance 0.2 from threshold. In each picture liquid is above and air below : (a) newtonian fluid (Silicon oil) ; (b) Xanthan and (c) AP45.

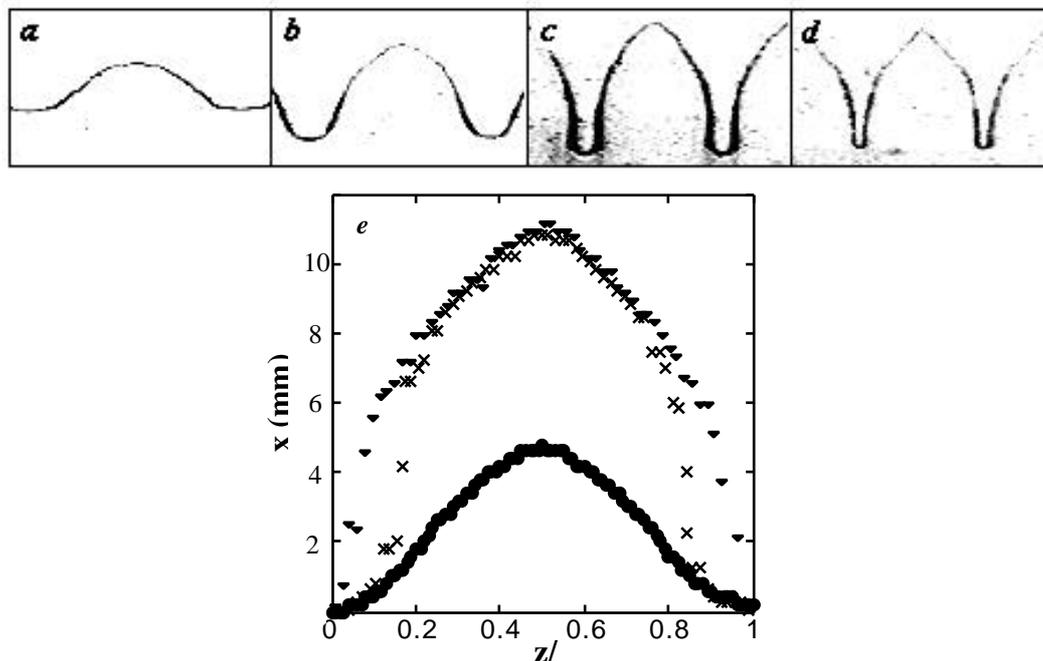


Fig. 5 : Photographs of the interface with a Xanthan solution (3000 ppm) for increasing : (a) $\lambda = 0.75$; (b) 1.25 ; (c) 3.5 ; (d) 6.0 and (e) plot of the azimuthal position x of the interface versus the dimensionless axial position z/λ where λ is the wavelength of the finger : (●) $\lambda = 0.75$, (×) $\lambda = 3.5$ and (▼) $\lambda = 6.0$.

3. CONCLUSION

For shear-thinning solutions we determined the main rheological properties and shown that above threshold the interface is still formed of air fingers penetrating in the liquid, but contrary to newtonian cases, these fingers become sharp above the onset. This shape evolution exists for non elastic as well as for strongly elastic fluids.

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