

# **DELAYED DIE SWELL AND SEDIMENTATION OF ELONGATED PARTICLES IN WORMLIKE MICELLAR SOLUTIONS**

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May, 1998.

## **ABSTRACT**

It has been recently proposed that the combined action of inertia and non-linear viscoelasticity may be the origin of very peculiar behaviors with dramatic changes of flow type. Two examples are the problem of delayed die swell and the orientation of elongated particles sedimenting in solutions of wormlike micelles. These solutions give rise to well defined viscoelastic properties which can be tuned precisely by changing the surfactant weight fraction. Our analysis does not rely on any constitutive equation and our results strongly support the interpretation that delayed die swell and sedimentation of long bodies in wormlike micellar solutions are ruled by a change of flow type from subcritical to supercritical.

## I. INTRODUCTION

Liquids with non-linear rheological behavior in complex flows exhibits a great variety of intriguing effects : die swell, rod-climbing, recirculation in contraction flows... Usually, it is considered that the Reynolds number is infinitely small and that inertia can be totally neglected. This is not often the case and recently, it has been proposed that the combined action of inertia and non-linear viscoelasticity may be at the origin of very peculiar behaviors with dramatic changes of flow type<sup>1, 2, 3</sup>. Two examples which will be addressed in the following are the problem of delayed die swell and the orientation of elongated particles in polymer solutions.

When a viscoelastic fluid is extruded from a die, the section of the die usually increases, which is the so-called die swell phenomenon. At high rates of extrusion, it may happen that the jet keeps a constant diameter after the die exit and swells far downstream after a delay which may be quite long. To our knowledge, all observations of delayed die swell have been made in polymer solutions<sup>4</sup>. An explanation of delayed die swell has been given by Joseph, Matta and Chen<sup>5</sup> who proposed that delayed die swell is a general non-linear phenomenon associated to a change of type of vorticity from elliptic to hyperbolic.

The sedimentation of elongated particles involves another flow which possibly exhibits a change of type<sup>6</sup>. Cylinders fall straight-down in viscoelastic liquids under the action of normal stresses. When inertia is not negligible, they turn broadside-on. It has been proposed that an inertial mechanism may turn the body in supercritical flow and therefore may cause orientation tilting.

In this paper we report and analyze delayed die swell and changes of flow type during the sedimentation of oriented particles in solutions which are not polymer solutions of wormlike micelles. These solutions give rise to well defined viscoelastic properties which can be tuned very precisely by changing the surfactant weight fraction. Our analysis does not rely on any constitutive equation and our results strongly support

the interpretation that delayed die swell and sedimentation of elongated particles in viscoelastic liquids are ruled by a change of flow type from subcritical to supercritical.

The paper is organized as follows. In section II, we present the rheological properties of micellar solutions as measured by cone and plate rheometer and wave-speed meter. In section III, we describe the extrusion experiment of micellar solutions. In section IV, we focus on delayed die swell and we discuss our experimental results in relation with the possible existence of a change of flow type in the extrusion of micellar solutions. In section V, we show that the orientation of elongated particles sedimenting in micellar solutions may tilt under some conditions, which is also consistent with a change of flow type.

## **II. RHEOLOGICAL PROPERTIES OF MICELLAR SOLUTIONS MEASURED BY WAVE-SPEED METER AND CONE AND PLATE RHEOMETER**

### **II.1. Preparation of the micellar solutions.**

Surfactant solutions were prepared by mixing cetylpyridinium chloride (CPyCl) and sodium salicylate (NaSal) in brine (NaCl at 0.5mol/l). The total weight fraction of surfactants,  $\Phi$ , ranges from 0.5% to 2%. To make the jets visible in the extrusion experiment, eosin is added to the solutions at a concentration of 2000ppm. The weight ratio  $W=m_{\text{NaSal}}/m_{\text{CPyCl}}$  is an important parameter which can be varied and has to be specified. Rehage and Hoffman<sup>7</sup> demonstrated that for constant CPyCl concentration and varying NaSal content, the viscosity rises steeply at a particular value of  $W$  for which the solutions exhibit strong analogies with entangled polymer. We used this criterion to determine that  $W=0.3$  corresponds to the formation of wormlike micellar solutions. We used this value in all our experiments. The solutions must be prepared and stored at a temperature greater than 22°C to prevent surfactant crystallization ; in practice, all the experiments reported in the following have been conducted at 25°C.

## I.2. Linear and non-linear rheology as a function of concentration.

The rheological properties of wormlike micelles made of CPyCl and NaSal have been investigated by many authors both in the linear and non-linear regimes<sup>8, 9, 10</sup>. All these studies have used purified surfactants in pure brine. Our solutions were prepared from commercially available products without further purification and they contain eosin as an additional component. This led us to check that the dynamic properties of our micellar solutions are effectively those of wormlike micelles. This study is restricted to the range of low concentrations where particular non-linear phenomena like shear-induced phase transitions do not occur<sup>9, 10</sup>. First, we carried out measurements of the storage modulus and of the loss modulus versus the frequency with a cone and plate apparatus. A typical result is reported in Figure 1 for  $\Phi = 0.02$ . The experimental points can be nicely fitted by a Maxwellian model, yielding a single relaxation time just as expected for solutions of wormlike micelles. This particular dynamics is confirmed by transient rheological measurements which indicate that the shear stress increases and decreases exponentially after the onset and cessation of steady-state flow. The high frequency plateau values of the storage modulus,  $G_0$ , and the relaxation time,  $\tau$ , are reported in table 1 as a function of the weight fraction of surfactants. The data reported in table 1 are in good agreement with the theoretical predictions<sup>11</sup> available for entangled wormlike micelles, according to which  $G_0$  and  $\tau$  should vary with  $\Phi$  as :  $G_0 \sim \Phi^{9/4}$  and  $\tau \sim \Phi^{5/4}$ .

With the same apparatus, we have measured the viscosity of the solutions as a function of the shear rate. The results for the three concentrations under consideration are reported in Figure 2. At low shear rates, the viscosity is constant reflecting linear equilibrium properties ; the zero-shear limit of the viscosity,  $\eta_0$ , is given in table 1. At higher shear-rates, the viscosity of the solutions decreases with  $\dot{\gamma}$ . The onset of shear

thinning,  $\dot{\gamma}_0$ , can be used as an other determination of the relaxation time of the solutions :  $\tau=1/\dot{\gamma}_0$ . Since the relaxation time is very small for  $\Phi=0.005$ ,  $\dot{\gamma}_0$  is very large, which explains why, at this weight fraction, solutions exhibit Newtonian behavior over the whole range of shear rates investigated. We have used the power-law model to characterize the shear rate dependence of the viscosity in the shear thinning region :  $\eta = m\dot{\gamma}^{n-1}$  ; the values of the exponent n are given in table 1.

### II.3. Measurements of the velocity of elastic shear waves with the wave-speed meter.

The wave speed meter that we used has been described by Joseph et al<sup>12, 13</sup>. The apparatus uses a Couette device with coaxial cylinders which may rotate independently. The gap is filled with the liquid to be studied. The outer cylinder is moved impulsively at time  $t=0$ . A shear wave propagates towards the inner cylinder which is set into motion after a certain time  $t$ , which is the transit time. For a Newtonian fluid, the transit time is the diffusion time of vorticity and it depends directly of the shear viscosity. For an elastic fluid, the transit time is the time of first reflection of the shear wave ; it must be proportional to the gap size, with the same constant of proportionality for sufficiently small gap sizes, the reciprocal of this constant being the wave speed. We have measured the wave speed velocity of micellar solutions at different solutions very carefully. For each concentration, measurements of the transit time have been made for different gap sizes ranging from 1.91mm to 0.57mm and we have checked that they all give the same result for the wave speed. The variations of the wave speed with the concentration are shown in Figure 3. c increases with  $\Phi$ , reflecting the fact that the solutions become more and more elastic as we increase the surfactant concentration. On the same graph, we have plotted the shear-wave speed calculated from the plateau values of the storage modulus,  $G_0$  :  $c = \sqrt{G_0 / \rho}$ , where  $\rho$  is the density of the solutions. The wave speeds measured with the wave speed meter are slightly greater than the values which are calculated from  $G_0$ , but the two sets of data fall on parallel

straight lines, the slope of which is close to 1. This value is in perfect agreement with what is expected for wormlike micelles. Indeed, table 1 shows that  $G_0$  is a quadratic increasing function of the weight fraction of surfactants. The shear wave speed  $c$  being proportional to the square root of the plateau modulus, we expect that  $c \sim \Phi$ , which is well verified in Figure 3.

### III. EXTRUSION EXPERIMENTS

#### III.1 Extrusion apparatus.

In our experiments, the micellar solutions are extruded vertically from stainless needles of different diameters. The extrusion takes place in a liquid bath in order to make interfacial effects negligible and to prevent the jet from breaking into tiny droplets. The shape of the jet is sensitive to the difference between the density of the micellar solutions and that of the bath. This effect will be analyzed in detail in section II.2 and the working conditions will be asserted precisely. The flow in the capillary is created by a Harvard programmable syringe pump; the maximum pressure exerted by the pump on the piston of the syringe is very high (200psi) so that viscous solutions can be extruded easily. The needles are 50mm long; their inner diameters,  $d_i$ , range from 0.31mm to 1.0mm. Long, flexible, teflon capillaries connect the needles to the syringe ; the inner diameter of these capillaries is of the order of 1mm. The long distance between the exit of the needle and that of the syringe ensures fully developed flow conditions at the needle exit. For the sake of stability, the micellar solutions are maintained at a constant temperature of 25°C as they flow from the syringe to the bath. The jet at the needle exit is observed with a CCD camera equipped with a macrophotography 55 mm lens which is connected to an image processing system. The jet profile is determined using a local binarisation algorithm. Typical jet profiles are represented in Figures 4.a and 4.b. The jet in figure 4.a exhibits the so-called die swell phenomenon : its diameter increases at the die exit and reaches its maximum value,  $d_m$ , at the terminal distance  $z_m$ .

In practice, die swell takes place during a characteristic swell time  $\lambda$ , which is

associated to the relaxation of the first normal stress difference, and which is a function of the shear rate in the die<sup>14</sup>. We can express the terminal swell distance as :  $z_m \approx \lambda(\dot{\gamma})U$ , where U is the area averaged velocity in the die. Little is known about the relaxation of the normal stresses but experiments<sup>15</sup> show that they relax to zero more rapidly as the shear rate  $\dot{\gamma}$  in the preceding steady shear flow is increased. In our experiments, the shear rate in the needle is quite high ( $10\text{s}^{-1} < \dot{\gamma} < 1000\text{s}^{-1}$ ) and the characteristic swell time associated to the relaxation of normal stresses is expected to be short,  $z_m$  to be small, so that the jet should swell immediately at the exit of the needle. Experimentally,  $z_m$  is found to be of the order of magnitude of  $d_i$ . The jet in Figure 4.b exhibits the so-called delayed die-swell phenomenon: the swell of the extrudate does not occur immediately at the die exit but at some distance downstream. In addition to  $d_m$  and  $z_m$ , it is convenient to define the position of the section where the jet profile changes its curvature,  $z_c$ , and the jet diameter at this section,  $d_c$ . The area-averaged velocities at  $z_m$  and  $z_c$  are  $U_m$  and  $U_c$ .

### III.2 Control of jet shape by gravity adjustment of the buoyancy bath.

In a typical experiment, a liquid is extruded with vertical velocity parallel to gravity into a lighter liquid of smaller density. As demonstrated several years ago by Joseph et al<sup>16</sup>, in this configuration, the fluid dynamics of the jet may be strongly affected by the gravitational body forces arising from the difference of density between the liquid in the jet and the ambient liquid, and by the entrainment of the ambient liquid by viscous action. In view of this, we have undertaken a systematic experimental study to elucidate the role of gravity and of fluid entrainment on the jet shape and to assess their effects on the die swell phenomenon in micellar solutions. This has been done by extruding micellar solutions in water baths containing different concentrations of sodium chloride, varying the surfactant weight fraction and the flow rate.

A typical result is given in Figure 5. We see clearly that the shape of the jet depends on the density difference  $\Delta\rho$  between the micellar solution and the external

bath ; in particular the diameter at maximum swelling changes drastically with  $\Delta\rho$ . A complete analysis requires to take into account the swelling of the jet under the action of normal stresses. In the following, we shall focus on the region  $z>z_m$  where viscoelastic effects are negligible ; in practice,  $z_m$  is of the order of magnitude of the needle diameter so that this asymptotic analysis is expected to be valid not too far from the exit. An equation giving the radius of the jet,  $r$ , can be obtained from the conservation of momentum. In the approximation where the  $r$  variations of the velocity are negligible, we get :

$$\frac{d}{dz} \left( \rho r^2 u^2 - r^2 \frac{du}{dz} \right) = \Delta\rho g r^2 + \mu' r \frac{du'}{dr}$$

$u$  and  $u'$  are the velocities inside the jet and in the external bath ;  $\mu$  and  $\mu'$  are the viscosities of the extruded liquid and of the ambient liquid. The left-hand side of this expression gives the variation of momentum as the jet flows down. The right-hand side has two terms : the first one can be interpreted as the weight of the jet per unit length while the second one represents the shear friction exerted by the ambient fluid on the jet boundary. Both terms increases with the flow rate since  $r$  and  $u'$  are increasing functions of  $Q$  and it is not clear under which conditions they compensate. Experimentally, we have found that for any flow rate there exists a range of values of the density difference for which the jet keeps a constant diameter as it flows down, indicating that there is a balance between gravity and entrainment effects. In figure 5, this occurs when the concentration of the external bath in sodium chloride is  $C=0.3\text{mol/l}$  ( $\Delta\rho=7.5\times10^{-3}\text{g/cm}^3$ ). When  $C=0\text{ mol/l}$  ( $\Delta\rho=1.3\times10^{-2}\text{ g/cm}^3$ ), the jet diameter decreases under the action of gravity forces which accelerate the jet. For  $C=0.45\text{ mol/l}$  and  $C=0.48\text{ mol/l}$  (respectively  $\Delta\rho=2.1\times10^{-3}\text{ g/cm}^3$  and  $\Delta\rho=1.2\times10^{-3}\text{ g/cm}^3$ ), the friction of the jet dominates over gravity, which makes the jet slow down and causes an increase of the jet diameter. It is to note that the balance of gravity and entrainment effects is obtained for a fairly large difference of density between the

velocity of extrusion is high. In all the experiments reported in this study, we have considered that the densities of the solution and of the bath are correctly matched when the diameter of the jet remains constant as the jet flows downwards. When gravity and entrainment effects cancel, the shape of the jet is entirely determined by the value of the shear rate before exit, indicating that the swelling dynamics under the action of normal stresses is the leading phenomenon. This appears clearly in figure 6 where we have represented jets obtained for different extrusion velocities and different needle diameters but for equal shear rates. Once the  $r$  and  $z$  coordinates are rescaled by the inner diameter of the needle which is the only characteristic length of the problem, the jets superimpose perfectly.

#### **IV. DELAYED DIE SWELL**

##### **IV. 1 Review of delayed die-swell in polymer solutions.**

In the phenomenon of delayed die-swell, the swell of the extrudate does not occur immediately at the die exit but at some distance downstream. To our knowledge all the observations of delayed die swell has been made in polymer solutions. A convenient and detailed discussion of the history of research on delayed die swell can be found in the book on the fluid dynamics of viscoelastic liquids by Joseph<sup>4</sup>. As a first level of interpretation, it is tempting to associate delayed die swell to a competition between die swell and the readjustment of the velocity profile from pipe flow inside the die to uniform flow downstream. We have seen previously that die swell is completed at the terminal swell distance  $z_m \approx \lambda(\dot{\gamma})U$ , where  $U$  is the mean velocity in the die. The readjustment of the velocity profile is induced by a discontinuity of the boundary conditions : no slip on the wall, no shear on the surface of the extrudate. Before exit, the vorticity profile varies linearly : it is the greatest at the wall and it is zero at the center of the die. Downstream, it is impulsively reduced to zero at the exit lip and the region of zero vorticity consumes more and more of the jet as the distance to the exit increases. The readjustment of the velocity profile and the relaxation of vorticity to zero is triggered by elastic waves which propagate inwards from the surface of the jet, with a

velocity  $c$ . The time that the elastic waves take to propagate from the surface to the axis of symmetry of the jet is  $d_m/2(c-V)$  where  $V$  is the outward velocity of fluid particles which can be estimated as  $V \approx (d_m - d_i) / 2\lambda(\dot{\gamma})$ . During this time, a fluid element in the jet has moved vertically by a distance  $z_a \approx Ud_m / 2(c - V)$ . If  $z_m < z_a$  or  $c\lambda(\dot{\gamma}) > d_m - d_i / 2$ , the jet swells immediately at the die exit ; in the opposite case, it should swell far downstream because of inertia., Since the diameter at maximum swelling,  $d_m$ , only depends on the action of normal stresses, the shear rate in the capillary should be the parameter controlling the dynamics of delayed die swell, in the frame of this interpretation.

Joseph, Matta and Chen<sup>5</sup> promoted a different interpretation according to which the delay is a critical hyperbolic phenomenon, analogous to a hydraulic shock, associated with a change of type of the vorticity equation, like in transonic flows. Yoo and Joseph<sup>1</sup> demonstrated that the vorticity equation for an upper convected Maxwell model in a channel switches from elliptic to hyperbolic type, in a region situated near the center of the channel, when the centerline velocity of the Poiseuille flow exceeds the velocity of vorticity waves,  $c$ . Ahrens, Yoo and Joseph<sup>2</sup> extended this result to pipes. Chen<sup>3</sup> has derived a similar criterion for two viscoelastic liquids which are extruded from a pipe. This explanation of delayed die-swell has been supported and some new understanding has been added by a numerical study of the plane jet of a viscoelastic liquid modeled by the upper convected model with a small retardation time, by Delvaux and Crochet<sup>17</sup>. The main new result is the existence of a « breakout of the region of hyperbolic vorticity ». When the centerline velocity in the channel is slightly supercritical, i.e. somewhat larger than the wave-speed, the hyperbolic region in the channel has a small extent downstream but never touches the free surface of the jet. When the velocity increases, the hyperbolic first touches the jet boundary, then consumes more and more of it. The change of shape of the free surface of the jet is associated with the breakout of the hyperbolic region. This simulation shows that the delay does not take place at small supercritical values of the velocity but requires larger <sup>near critical values</sup>

The defect of these analysis is that they are based on a particular constitutive model, namely the upper convected Maxwell model and it is not clear whether more general models would also lead to similar critical hyperbolic phenomena. Joseph reported that the onset of delayed die swell in a variety of polymer solutions correlated perfectly with the values of wave speed which can be measured on the wave speed meter<sup>4, 5, 18</sup>: the extrusion velocity at the exit of the pipe is always greater than the shear wave speed, the velocity at the position of maximum swelling is always smaller than the shear wave speed. The results reported in the following for solutions of wormlike micelles, and the corresponding discussion, are independent of constitutive assumptions ; we show than several experimental observations fit the interpretation that delayed die swell is a general non linear phenomenon and we point out some new features of delayed die swell.

#### IV.2 Delayed-die swell experiments.

We have performed a systematic study of the delayed-die swell of micellar solutions at different weight fractions ranging from  $\Phi=5\times10^{-3}$  to  $\Phi=10^{-2}$  ; the diameter of the needles has been varied from 0.31mm to 0.84mm. A sequential description of the phenomenon is shown in Figures 7a and 7b ; the surfactant concentration is  $\Phi=5\times10^{-3}$  and the diameter of the needles is respectively 0.31mm and 0.60mm. To characterize the experimental conditions, it is convenient to introduce the Reynolds number,  $Re=\rho Ud/\eta$ , and the Weissenberg number,  $We=U\tau/d_i$ . We take the area averaged velocities of the fluid for  $U$ . These two parameters are independent and any other composition of these two can be used. For instance, we can define the viscoelastic number,  $M=\sqrt{ReWe}=U/c$ , and the elasticity number,  $E=We/Re=c^2\tau^2/d_i^2$ . The Mach number compares the fluid velocity and the shear wave speed. The elasticity number depends on material parameters and needle diameter but is independent of flow. The values of the flow rate,  $Q$ , the Reynolds number,  $Re$ , and the Mach numbers,  $M$ ,

$M_C$  and  $M_m$  are reported in table 2.  $M (= U/c)$  is the Mach number before the exit of the needle,  $M_C (= U_C /c)$  is the Mach number in the section of the inflection point, and  $M_m (= U_m /c)$  is the Mach number in the section where the jet reaches its maximum (see figure 4.b). At low flow rates, the jet exhibits simple die swell. When the extrusion velocity is raised above a critical flow rate, delayed die swell appears in a dramatic form : first, the shape of the jet flattens at the exit, then a point of inflection appears. The distance to the exit of the inflection point and the terminal swell distance increases with the flow rate. At very high rates of extrusion, delayed die swell ends up by the smoothing of the jet and in extreme cases, the degree of smoothing is so great that the location of the inflection point cannot be determined accurately. The data reported in table 2 show that the Mach number at the exit is always greater than 1, that the Mach number downstream after the swell is smaller than 1, and that the Mach number in the section of the inflection point is of the order of 1. We also see that the critical Mach number at which delayed die swell first appears is the smallest for the largest needles. At larger surfactant concentrations, i.e. for larger relaxation time and larger elastic modulus, delayed die swell occurs in a similar fashion. The only difference is that the smoothing of the jets at high flow rates is less apparent at large  $\Phi$  ; instead the jets become unstable, the point of delay exhibiting oscillations between some extreme values.

To analyze quantitatively the delayed die swell phenomenon, we have identified the point of delay with the inflection point where the jet changes its curvature and we have measured  $z_C$ . In figure, we plot  $z_C/d_i$  as a function of the Mach number at the exit for two surfactant concentrations and three needle diameters. The points fall on lines which are nearly parallel and which intersect the horizontal axis at the critical Mach number. This representation shows that the extrusion velocity, or the Mach number, is the key parameter controlling delayed die swell. This contradicts the simple argument based on a competition between die swell and inertia developed in the previous section, which predicts that the shear rate should be the control parameter. To go further, the existence of a shock layer and the fact that the Mach number relaxes to a value always

larger than 1 at the exit to a value smaller than 1 downstream strongly support the idea that delayed die swell is a non linear phenomenon associated to a change of type from supercritical to subcritical flow.

An intriguing result is that, at the onset of delayed die swell, the fluid velocity at the exit can be much larger than the speed of vorticity waves and that the critical Mach number is a decreasing function of the diameter of the needle. This result has already been mentioned by Joseph, Matta and Chen in their paper on delayed die swell<sup>5</sup>. A possible interpretation relies on the role of the elasticity number. In the steady flow of an upper convected Maxwell fluid through a channel, it has been shown<sup>4</sup> that the thickness of the region of hyperbolic vorticity depends on the elasticity number as  $r^* \approx E^{-1/2}$ . This shows that  $r^*$  is proportional to the diameter of the needle. Thus, for a given fluid, when the diameter of the needle is small, the hyperbolic region is small and it is likely that the hyperbolic region never touches the free surface of the jet, except at very high flow rates. This explanation is in qualitative agreement with our experimental observations. However from figure 8, the data does not seem to correlate with the elasticity number but simply with the diameter of the needle. More data would be necessary before giving a definite answer to this question.

## V. ORIENTATION OF CYLINDERS FALLING IN MICELLAR SOLUTIONS

We have shown that delayed die swell of micellar solutions is not exceptional : the dynamics of the swell is of the type typically observed in polymer solutions. In view of this, it is of interest to compare the behavior of micellar an polymeric solutions in other flows in which a dramatic change of flow type occurs as the Mach number is increased past a critical value. The orientation of cylinders falling in polymeric solutions exhibits just such a transition.

Two kinds of experimental situations have been investigated in the literature. Liu and Joseph<sup>19</sup>, Joseph and Liu<sup>20</sup>, Joseph<sup>21</sup> addressed the change in orientation of falling cylinders of the same size and shape but different weights falling in each of many

viscoelastic fluids in terms of the Mach and Reynolds numbers. The light cylinders lined up with the flow, but the heavy cylinders turned broadside-on. This change from vertical to horizontal falling occurs when the stagnation pressures at the front of the falling body are dominated by inertia. This happens when the body fall faster than diffusion ( $Re>1$ ) and faster than shear waves ( $M>1$ ).

An apparently similar but actually different set of experiments were carried out by Chiba, Song and Horikawa<sup>22</sup>. They studied the settling of one cylinder in an aqueous solution varying the concentration rather than the weight of the particle. They demonstrated that a slender body fall with its long axis perpendicular to gravity in dilute solutions and parallel to gravity in concentrated solutions. At intermediate concentrations, tilted cylinders with side drift were observed. Such tilted orientations of long bodies, neither horizontal or vertical, have also been observed by Huang, Hu and Joseph<sup>23</sup> in their numerical study of an ellipse settling under gravity in an Upper Convected Maxwell Fluid. They are due to transient effects arising from walls, from shear thinning or from the existence of normal stresses.

The difference between the experiments where we change the concentration of the solution and those in which particle weight in a given solution was varied was recently explained by Huang, Hu and Joseph<sup>23</sup>. The explanation relies on the existence on a critical elasticity number and a critical Mach number. Both numbers mark borders in which the balance between inertia and viscoelasticity changes. The experiments carried out by Chiba, Song and Horikawa<sup>22</sup> correspond to changes in the elasticity number. The experiments by Liu and Joseph<sup>19</sup> and Joseph and Liu<sup>20</sup> represent a different physics where the critical Mach number is changed.

We have measured the orientation of cylinders with different weights falling in solutions of wormlike micelles. We used flat-ended and round-ended cylinders. The particles, released with their broadside initially parallel to the direction of fall, were dropped in a 64cm×16cm×1.25cm test bed and viewed with a video camera connected to a computer. Particle velocity was estimated by measuring the time of fall through the final 30cm of the bed height. Tilt angles were measured with image analysis software

with an accuracy of  $\pm 3^\circ$ . The particles were of mass 0.19g-18.15g, length 1.0cm-3.25cm, and diameter 0.25cm-1.0cm. The results are plotted against the Reynolds number in figure 9.a and against the Mach number in figure 9.b. These plots look like the ones shown in the paper by Liu and Joseph<sup>19</sup>. We find that there is a tilt angle transition like in polymer solutions and that the transition correlates with the Reynolds number and the viscoelastic Mach number. The first condition  $Re>1$  indicates that to make the cylinders turn broadside-on the inertial forces have to be greater than the viscous forces. The second condition  $M>1$  tells us that shear waves cannot propagate upstream in the fluid at rest so that inertia dominates viscoelasticity.

## VI. CONCLUSION

The scope of this paper is two-fold. First, we have shown that delayed die swell and orientation tilting during the sedimentation of elongated particles are not exceptional and may occur in non-polymeric viscoelastic solutions. Secondly, our results which have been discussed without the help of any constitutive equation, strongly support that some inertial mechanisms causing a change of flow type from subcritical to supercritical are at the origin of these phenomena.

Concerning die swell, we have found that the critical Mach number at which delayed die swell first appear strongly depends on the diameter of the die. It is probable that the extrusion through needles of very small diameters will never give rise to delayed die swell. A possible explanation is that the elasticity number, which decreases with the diameter, determines the spatial extent of the hyperbolic vorticity. At small elasticity numbers, the region of hyperbolic vorticity is small and it does not touch the jet boundary except at very large Mach numbers. However, this interpretation seems to be partially contradicted by the fact that the critical Mach number seems to depend only on the diameter and not on the elasticity number.

$\Phi(g/g)$	$G_0(Pa)$	$\tau(s)$	$\eta_0(mPa.s)$	n	$\tau'(s)$	c(cm/s)
0.005	0.5	0.007	9.8	1	<0.01	3
0.01	2.2	0.05	128	0.6	0.04	5.9
0.02	8.5	0.10	850	0.1	0.1	10.4

Table 1 : variations of the rheological properties of micellar solutions as a function of the weight fraction of surfactants.

$d_i$ (mm)	Q(ml/h)	Re	$M=U/c$	$M_c=U_c/c$	$M_m=U_m/c$
0.31	50	29	6.5	*	0.5
	70	41	9.2	1.6	0.5
	90	53	11.8	1.2	0.5
	130	77	17.0	1.4	0.8
	150	88	19.6	1.6	1.1
	170	100	22.2	*	*
0.60	150	44	4.9	*	0.3
	210	62	6.9	1.3	0.3
	270	79	8.8	1.4	0.6
	350	103	11.4	1.4	0.7
	400	118	13.1	1.6	0.9

Table 2 : measured values of flow rates, Reynolds numbers and Mach numbers for the experiments depicted in Figure 7.

## FIGURE CAPTIONS

Figure 1 : Storage modulus  $G'$  and loss modulus  $G''$  as a function of the angular frequency  $\omega$  for a micellar solution (the weight fraction of surfactants is  $\Phi=0.02$ ). The inset is a Cole-Cole plot of  $G'$  and  $G''$ . The experimental data can be nicely fitted by a Maxwellian model (continuous lines).

Figure 2 : Variations of the viscosity versus the shear rate for micellar solutions at different weight fractions. From top to bottom:  $\Phi=0.02$ ,  $\Phi=0.01$ ,  $\Phi=0.005$ .

Figure 3 : Variations of the shear wave velocity as a function of the weight fraction of surfactants. The data measured with the wave speed meter (open circles) are in good agreement with those obtained from the high frequency plateau value of the storage modulus (solid circles). The line has the slope +1.

Figure 4 : Cartoon of die swell (a) and delayed die swell (b).

Figure 5 : Shapes of jets when the difference of density between the micellar solution and the external bath is varied. The concentration of the bath in sodium chloride and the corresponding density difference are, from the axis of symmetry to the outside :  $C=0$  mol/l and  $\Delta\rho=1.3\times10^{-2}$ ,  $C=0.3$  mol/l and  $\Delta\rho=7.5\times10^{-3}$ ,  $C=0.45$  mol/l and  $\Delta\rho=2.1\times10^{-3}$ ,  $C=0.48$  mol/l and  $\Delta\rho=1.2\times10^{-3}$ . The density differences have been measured with an accuracy better than 10%.

Figure 6 : Jets obtained for different needles and different flow rates are identical provided that the shear rates before exit are equal. Note that the coordinates  $r$  and  $z$  have been rescaled by the inner diameter of the needle. The shear rate in the needle is  $248\text{ s}^{-1}$

; the experimental conditions are :  $d_i=0.84\text{mm}$  and  $Q=60\text{ml/h}$ ,  $d_i=0.60\text{mm}$  and  $Q=22\text{ml/h}$ ,  $d_i=0.41\text{mm}$  and  $Q=7\text{ml/h}$ .

Figure 7 : Delayed die-swell in micellar solutions ; the weight fraction of surfactants is 0.005 ; the diameter of the needle is  $d_i=0.31\text{mm}$  in (a) and  $d_i=0.60\text{mm}$  in (b). The measured values of flow rates, Reynolds numbers and Mach numbers are given in table 2. The elasticity numbers are respectively  $E=0.5$  (a) and  $E=0.1$  (b).

Figure 8 : Vertical location of the inflection point of the jet profile versus the viscoelastic Mach number before exit. Each symbol refers to a different experiment : (open diamonds)  $d_i=0.60\text{mm}$ ,  $\Phi=0.005$ ,  $E=0.1$  ;  $\blacklozenge$   $d_i=0.60\text{mm}$ ,  $\Phi=0.007$ ,  $E=5$ ;  $\circ$   $d_i=0.31\text{mm}$ ,  $\Phi=0.005$ ,  $E=0.5$  ;  $\bullet$   $d_i=0.31\text{mm}$ ,  $\Phi=0.005$ ,  $E=20$  ;  $\Delta$   $d_i=0.40\text{mm}$ ,  $\Phi=0.005$ ,  $E=0.2$ .

Figure 9 : Tilt angle versus Reynolds number (a) and viscoelastic Mach number (b) for flat-ended and round-ended cylinders settling in micellar solutions. The cylinder type and weight fraction of surfactants are :  $\circ$  flat-ended,  $\Phi=0.01$  ;  $\blacktriangle$  round-ended,  $\Phi=0.01$  ;  $\times$  flat-ended,  $\Phi=0.00125$  ;  $\square$  round-ended,  $\Phi=0.00125$ .

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