Orientation of long bodies falling in a viscoelastic liquid

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Abstract

New experiments on the orientation of a cylinder settling in viscoelastic and pseudoplastic fluids are described in an attempt to identify the main mechanisms which control the orientation of the cylinder as it falls.

1. Introduction

Liu and Joseph [1993a] have done experiments on the settling of long cylinders in aqueous solutions polyox and polyacrilamide, and in solutions of polyox in glycerin and water. The tilt angles of long cylinders and flat plates falling in these viscoelastic liquids were measured. The effects of particle length, particle weight, particle shape, liquid properties and liquid temperature were determined. In some experiments, the cylinders fall under gravity in a bed with closely spaced walls. No matter how or where a cylinder is released the axis of the cylinder centers itself between the close walls and falls steadily at a fixed angle of tilt with the horizontal. A discussion of the tilt angle may be framed in terms of competition between viscous effects, viscoelastic effects and inertia. When inertia is small, viscoelasticity dominates and the particles settle with their broadside parallel or nearly parallel to the direction of fall. When inertia is large the particles settle with their broadside perpendicular to the direction of fall. The tilt angle varies continuously from 90°, when viscoelasticity dominates, to 0°, when inertia dominates. The balance between inertia and viscoelasticity was controlled by systematic variation of the weight of the particles and the composition and temperature of the solution. Particles will turn broadside-on when the inertia forces are larger than viscous and viscoelastic forces. This orientation occurred when the Reynolds number $\mathbb{R}$ was greater than some number not much greater than one in any case, and less than 0.1 in Newtonian liquids and very dilute solutions. In principle, a long particle will eventually turn its broadside perpendicular to the stream in a Newtonian liquid for any $\mathbb{R}>0$, but in a viscoelastic liquid this turning cannot occur unless $\mathbb{R}>1$. Another condition for inertial tilting is that the elastic length λU should be longer than the viscous length ν/U where U is the terminal velocity, ν is

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the kinematic viscosity and $\lambda = v/c^2$ is a relaxation time where c is the shear wave speed measured with the shear wave speed meter (Joseph, [1990]). The condition $M = U/c > 1$ was provisionally interpreted by Liu and Joseph [1993] as a hyperbolic transition of solutions of the vorticity equation analogous to transonic flow. They showed that strong departures of the tilt angle from q=90° begin at about $M = 1$ and ends with q=0° when $1 < M < 4$.

The fluids used in the experiments of Liu and Joseph [1993a] are standard viscoelastic test fluids with non-zero normal stresses and power law exponents between 0.38 and 0.54. The more elastic fluids are also more shear thinning with a smaller exponent. All these fluids climb a rotating rod strongly and have substantial normal stresses in shear. We were persuaded that elasticity rather than shear thinning was the controlling factor at small speeds when the cylinders turn their long side parallel to gravity, in the direction of fall. Such turning is predicted by asymptotic analyses by Leal [1975] and Brunn [1977] for slow motions which perturb rest with inertia neglected. This kind of analysis leads asymptotically to a second order fluid for which shear thinning is absent. We were and are persuaded that the orientation of long bodies is a key to understanding flow induced microstructures of spherical bodies, and we found that when long bodies put their nose down, spheres chain to form long bodies, and that when the long bodies are titled broad-side on by inertia, the chain of spheres break up and tend to disperse into cross-stream arrangements.

B. Van den Brule and G. Gheissary [1993] saw a videotape of our experiments on sedimenting spheres, which form long chains in all viscoelastic liquids at the slow fall speeds in which long bodies rotate their long side parallel to the fall. This stimulated them to undertake experiments on sedimenting spheres of their own. They attempted to isolate the effects of shear thinning and normal stresses by using test fluids which have one and not the other of these properties. They dropped spheres in aqueous polyacrilamide which is an ordinary viscoelastic fluid with large normal stresses and strong shear thinning and found results identical to ours. Then they did experiments in "shellflow" which is an aqueous Xanthan solution which has no measurable normal stresses in shear but is strongly shear thinning. They found that the spheres chained in this fluid in much the same way that they did in the aqueous polyacrilamide. This suggests that shear thinning is the important parameter. They then did experiments in a Boger fluid which they prepared with small amounts of polyacrilamide (100 ppm) in glycerin and water. This is a very viscous fluid with large but saturated normal stresses, which lead to constant values of the recoverable shear at high rates of shear. They did not observe chaining in this Boger fluid, and they concluded that shear thinning and not elasticity is the mechanism controlling the chaining of spheres.

The aforementioned results do seem to contradict theoretical results for very slow motions, and they are not consistent with results which can be observed in other pseudoplastic and Boger fluids which

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we have studied and which will be discussed here. Nevertheless, the results of Van den Brule and Gheissary are of considerable interest and need further interpretation. According to our idea their observations ought to be extended to the problem of the tilting of sedimenting the cylinder which was discussed by Liu and Joseph [1993a]. These considerations have led us to undertake new experiments and the ones on the tilting of sedimenting cylinders, together with interpretations will be reported here.

2. Material and dimensionless parameters

The material parameters which were measured in the liquids used in the experiments are the density ρ, viscosity $\eta = k\dot{\gamma}^{n-1}$, where $\dot{\gamma}$ is the shear rate in reciprocal seconds, the climbing constant $\hat{\beta}$ measured on a rotating rod viscometer (Beavers and Joseph [1975]) and wave speed c. To compute $\hat{\beta}$ from measured values of the climb we need the interfacial tension which we measured with a spinning drop tensiometer (Joseph, *et al* [1992]). The value of $\hat{\beta}$ is insensitive to a small change of surface tension (chap.16 in [J1990]). In this paper we shall also present graphs of the dynamic moduli and other standard rheological properties which may be obtained on an RSF2 Rheometrics fluids rheometer. Tables and graphs of material parameters will appear where they are first used.

The climbing constant $\hat{\beta}$ is related to the limiting (zero shear) value of the first and second normal stress differences

$$(n_1, n_2) = \lim_{\dot{\gamma} \to 0} (N_1(\dot{\gamma}), N_2(\dot{\gamma}))/\dot{\gamma}^2 \tag{2.1}$$

by

$$\hat{\beta} = \frac{1}{2}n_1 + n_2 . \tag{2.2}$$

The climbing constant

$$\hat{\beta} = 3\alpha_1 + 2\alpha_2 \tag{2.3}$$

may also be expressed in terms of quadratic constants

$$(\alpha_1, \alpha_2) = (-\frac{1}{2}n_1, n_1 + n_2) \tag{2.4}$$

of the second order fluid. $\alpha_2/|\alpha_1|$ is the ratio of quadratic constants and

$$[\alpha_1, \alpha_2] = [-m, 2m-2]\hat{\beta}/(m-4) \tag{2.5}$$

where $m = 2\alpha_1/(2\alpha_1 + \alpha_2) = -n_1/n_2$ is the ratio of the first to second normal stress difference. It can be argued (§17.11 in [J1990]) that $m = 10$ is a reasonable value for our polymer solutions. Then

$$\frac{\alpha_2}{|\alpha_1|} = \left|\frac{2(1-m)}{m}\right| = 1.8 \tag{2.6}$$

is a constant and α_1 and α_2 are determined by the measured values of the climbing constant $\hat{\beta}$. The value of n_1 we get from measuring $\hat{\beta}$ is not sensitive to the value of the ratio n_2/n_1 as long as n_2 is relatively small and negative (see §17.11 in [J1990]).

The measured value of the climbing constant together with the assumption that the second normal stress difference is -1/10 as large as the first, allows us to evaluate Roscoe's [1965] formula

$$T_{11} - T_{22} = 3\acute{s}\eta_o + 3(\alpha_1 + \alpha_2)\acute{s}^2 \tag{2.7}$$

for the extensional stress difference where $\acute{s}$ is the rate of stretching in the direction x_1 and η_o is the zero shear viscosity. Using (2.6) and $\alpha_1 = -n_1/2$ we get

$$T_{11} - T_{22} = 3\acute{s}\eta_o + 1.2n_1\acute{s}^2 \tag{2.8}$$

The zero shear value of the first normal stress difference $n_1 = \dfrac{2m}{m-4}\hat{\beta} = \dfrac{10}{3}\hat{\beta}$ and the zero shear quadratic correction $4\acute{s}\hat{\beta}$ of Troutons viscosity, $3\eta_o$, increase with $\hat{\beta}$.

The study of supercritical flow around bodies can be expressed in terms of a Reynolds number $\mathbb{R}$ and a Weissenberg (Deborah) number W. The Reynolds number is given by

$$\mathbb{R} = \frac{U\ell}{\eta(\acute{\gamma})} \tag{2.9}$$

where U is the terminal velocity and ℓ is the hydraulic diameter (4 times the projected area over the projected perimeter where the projection is on the plane perpendicular to gravity).

The Weissenberg number will be defined by

$$W = \frac{\lambda U}{\ell} \tag{2.10}$$

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which is appropriate for models like Maxwell's with a single time of relaxation. For these models

$$\underset{\mathbb{R}}{W} = \frac{U^2}{(\eta/\lambda\rho)} = \frac{U^2}{c^2} = M^2 \qquad (2.11)$$

The relaxation time $\lambda = \dfrac{\eta}{\rho c^2}$ taken from wave speed measurements is much smaller than the values obtained on conventional rheometers which typically filter out the high frequency response.

The parameters $\mathbb{R}$ and W are convenient for studies in which hyperbolicity and change of type are not relevant. When change of type is an important issue, it is better to use M and either

$$E = \frac{\eta\lambda}{\rho\ell^2} \qquad \text{(elasticity number)} \qquad (2.12)$$

or W as the fundamental parameters. The parameters are related, $W = \mathbb{R}E$, $\mathbb{R} = M/\sqrt{E}$.

3. Description of the experiments

Particles were dropped in a liquid-filled channel, called a sedimentation channel. Six channels were used (see Table 1).

	Channels	Liquids						
No.	Dimensions (inches)	Polyox	Polyacrylamine	Xanthan	Carbopol	STP	S1	shampoo
1	0.44x6.5x25	tested	tested			tested		tested
2	0.275x4x23	tested				tested		
3	0.5x6.7x25.5			tested	tested			
4	1x7x30						tested	tested
5	1x1.63x28					tested		
6	2.76x29 (cylinder)	tested	tested					

Table 1.

We used different test particles to vary the weight, size and shape as shown in Table 2.

The motion of sedimenting particles in our three dimensional bed is basically two dimensional. The central axis of the cylinder usually aligns itself in the plane midway between the closely spaced sidewalls, no matter what the initial condition (see Figure 1). Liu and Joseph [1993a] report that even a thin wire needle whose diameter (0.017 inches) is over 20 times smaller than the gap (0.44 inches) will center itself in this way. The central position of particles is an effect of close sidewalls, and it also depends on the fluid. Apart from the drag, the motion of the central plane is apparently independent of the gap size. Small cylinders dropped in large tanks of circular or square cross-section behave exactly as they do in the beds with close side walls; the tilt action is localized in a vertical plane defined by the cylinder axis.

material	density g/cm^3	diameters D, inches	lengths L, inches	shapes
plastic	1.318	0.1, 0.15, 0.25, 0.35, 0.4	0.4, 0.6, 0.8, 1.0	f, c, r
teflon	2.176	0.25	0.4, 0.6, 0.8, 1.0	f, c, r
aluminum	2.699	0.1, 0.15, 0.25, 0.35, 0.4	0.4, 0.6, 0.8, 1.0	f, c, r
titanium	4.487	0.25	0.4, 0.6, 0.8, 1.0	f, c, r
tin	7.288	0.1, 0.15, 0.25, 0.4	0.4, 0.6, 0.8, 1.0	f, c, r
stainless steel	7.834	0.017, 0.25	0.4, 0.6, 0.8, 1.0	f, c, r
brass	8.478	0.1, 0.15, 0.25, 0.3, 0.35, 0.4	0.4, 0.6, 0.8, 1.0	f, c, r
lead	11.338	0.312	0.4, 0.6, 0.8, 1.0	f, r
tungsten carbide	15.778	0.312, 0.625	0.4, 0.6, 0.8, 1.0, 2.6, 3.53	f

Table 2. Test particles. The Particles had cone ends, round ends and flat ends designated by c, r and f respectively.

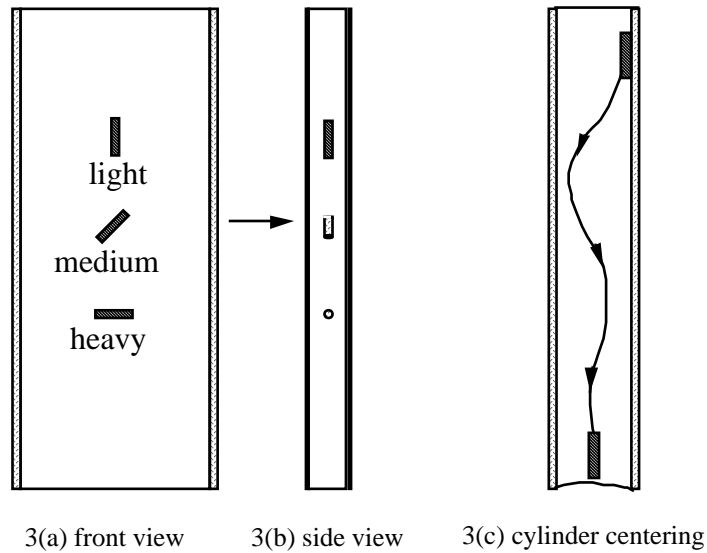3(a) front view   3(b) side view   3(c) cylinder centering

Figure 1. Cylinders always center themselves to the plane midway between the closely spaced sidewalls no matter what the initial condition and the final orientation.

The centering of particles falling in a viscoelastic liquid between close walls is not a universal phenomenon, but depends on the fluid and other factors. We find centering in all the fluids except STP for the spheres and cylinders used in our experiments. Even in STP the particles will center at modest to high speeds but not at the lowest speeds. We might expect problems of centering at low speeds of fall in other very viscous Boger fluids. The centering phenomenon requires effects from both walls. We have shown Liu and Joseph [1993b] that a sphere falling parallel to a wall in a viscoelastic liquid will be attracted to the wall if the distance between the wall and sphere is less than a critical value which depends on the fluid. We have seen flat plates settle in polyox solutions with their broadside parallel to gravity titled in the plane of falling in such a way as to put one thin edge near one wall and the other edge near the other wall. Each edge was in the region of attraction. Generally these flat plates center, so that the positioning of plates and other bodies is an involved subject which deserves study.

Velocities and tilt angles were measured with a Kodak Spin Physics SP2000 Motion Analysis System which can take pictures at 2000 frames per second (fps). The image is replayed at a speed of 60 fps. Images can also be played forward or backward at the rates of 2/3, 1, 3/2 or 3 fps, or one frame at a time. There are movable reticule lines which allow spatial measurements of the image. The elapsed time may be observed while a recording is being made and replayed. These functions allow one to measure the fall speed and tilt angle of sedimenting particles.

4. Settling of Long Cylinders in Polymeric Liquids

The motion of particles in liquids is influenced by viscosity, shear thinning, extensional stresses, normal stresses in shear and memory effects, to name some entries in a possibly longer list. It is useful to compare the rheometrical properties of liquids which are similar in some ways and different in other ways.

A very excellent comparison between 1.5% and 2% solutions of aqueous polyacrilamide (Magnaflox E10 supplied Allied Colloids) and 2% and 3% solutions of aqueous Xanthan gum Keltrol F (supplied by Kelco) was made by Walters, Bhatti and Mori [1990]. They arranged solutions with nearly the same viscosity values over a large range of shear rates and compared different measures of elasticity. They found that the Xanthan solutions had much lower normal stresses than the polyacrilamide, but had larger values of the storage modulus. They noted that Xanthan is a semi-rigid molecule and Polyacrilamide is a flexible molecule. They reasoned that Xanthan solutions are highly elastic near the rest state with a gel-like structure but this structure is easily broken down by shear. This is consistent with the observation that the dynamic properties of Xanthan solutions can be significantly affected by pre-shearing. The effect of such pre-shearing does relax but recovery can take a day. They noted that the Xanthan solutions are extensional thinning for shear rates which are apparently in excess of a value near 3 sec^{-1} but the polyacrilamide is strongly extensional thickening.

We are going to show that the tilt transition in semi-dilute solutions of Xanthan and water (0.3%) undergo tilt transitions just like those seen in the standard aqueous polyacrilamide and polyox solutions in which nonlinear elastic effects and shear thinning are present simultaneously. The shared properties of these standard test liquids with our semi-dilute Xanthan is the combined effect of shear thinning and a long memory. This idea will be explored in subsequent sections. We first are obliged however to take cognizance of the fact that these same particle migration and orientation effects are found in Boger and near Boger liquids, sometimes in weaker form, when viscous effects are huge compared to elastic ones as in the usual preparations of Boger fluids. The effects of elasticity on particles in liquids which are weakly pseudoplastic are evidently enhanced when the liquids are simultaneously more mobile and more elastic, as in S1 (section 4.1)

We next note that the effects of shear thinning on the migration of particles can be better understood by comparing two shear thinning fluids with effectively zero first normal stresses but different memory properties. For this, we obtained interesting results by comparing semi-dilute aqueous solutions of Carbopol and Xanthan (see section 4.2).

4.1 Sedimentation in STP and S1

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STP is a solution of polysolutilene in petroleum oil which was used extensively in early studies of rod climbing (see Joseph, [1990]). S1 is a solution of 50.0g of 5% W/W of PBI in decalin plus 50.0g polybutene oil. It is a world-wide standard test fluid which is being characterized by different laboratories in many countries. We mixed our own samples according to procedures laid down by Professor J. Ferguson of the University of Glasgow. Our homemade solutions have nearly the same properties as the premixed samples given to us.

The viscosity of these two polymer solutions was measured as a function of the shear rate $\dot{\gamma}$ on the RSF2 Rheometries fluid rheometer and is shown in Figure 2. The viscosity of STP is nearly constant for shear rates less than 100. The viscosity of S1 decreases with increasing $\dot{\gamma}$, but the decrease is very slow for shear rates less than 10. The viscosity of S1 is an order of magnitude smaller than STP; it is a much more mobile liquid. Both solutions climb a rotating rod, but the STP is not a good climber; the climbing constant is about one dyne/**in**. We can say that STP is a Boger fluid with very weak normal stresses. The climbing constant of S1 at 20$^\mathrm{o}$C is approximately 31 gm/cm and S1 can be said to resemble a Boger fluid with larger normal stresses, especially at low rates of shear.

Values for the dynamic moduli of STP and S1 are shown in Figure 3. The loss modulus for STP is an order of magnitude higher than S1. The storage modulus of S1 is larger than STP for shear rates less than about 10 seconds, and the shear rate at which the loss modulus falls below the storage modulus is much lower than S1. It is clear that S1 is a much more mobile and somewhat more elastic liquid than STP.

The mean shear wave in STP at a temperature between 24$^\mathrm{o}$ and 25$^\mathrm{o}$ is 286 cm/sec. The mean shear wave speed in S1 at a temperature near to 24$^\mathrm{o}$C is 71 cm/sec (see Table 3). The highest terminal velocity that could be achieved in S1 was about 30cm/sec along with a yet slower maximum velocity in STP for the heaviest cylinder. Since U is decisively smaller than the wave speed c in all cases, we did not expect to see and did not see inertial turning of the cylinder from straight down to broadside-on. The straight down orientation occurred for every particle from nylon to tungsten falling in S1 independent of the initial launching orientation.

h (mm)	1.181	1.956	2.959
$\bar{c}$ cm/sec	70.0	76.2	71
ϑ cm/sec	6.74	2.96	1.32

Table 3. Shear wave speeds in S1. The wave speed c is calculated from the ratio h/T where h is the gap size and T is the measured time of transit. The mean speed is an average over 10 trials and ϑ is the standard deviation.

Place Figure 2 here

Place Figure 3 here

The fall behavior in STP was more complicated. Heavier cylinders and flat plates falling in STP put their longside parallel to the flow independent of the initial orientation of the body. This is exactly like S1 and exactly like all the other viscoelastic fluids tested by us (aqueous polyox in concentrations ranging from 0.6% to 1.5%, 2% aqueous polyacrilamide, 8% polyox in glycerin and water, M1). The orientation of cylinders settling in STP cannot be specified unambiguously. The final orientation seems to depend on initial conditions and other factors over which we have no control. Light particles which settle slowly in STP do not always center; they interact with the wall and this and leads to orientations which vary from run to run and cannot be predicted. More satisfactory results can be obtained in a square channel, where nose-down orientation appear more frequently, but not every time. There is no evidence for inertial turning in the settling of long particles in STP.

The tilting of long particles into positions with the long side parallel to gravity in STP cannot be due to shear thinning (see Figure 2). We have also shown (Liu and Joseph [1993b]) the spherical particles falling STP exhibit side-by-side attraction if the initial distance is smaller than a critical value; a spherical particle is attracted weakly to a wall if it is close enough initially. Spherical particles falling in

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STP also form chains, though chaining is weaker than in other viscoelastic fluids which have greater levels of elasticity and also more shear thinning.

The tilting of long particles into positions with the longside parallel to gravity in S1 could be due to shear thinning, at least in part, since S1 does shear thin. However, the shear thinning is not great and is almost zero at small rates of shear (Figure 3). It is possible that shear thinning could be responsible for the robust turning we see in light cylinders which settle slowly. A more likely explanation, in our opinion, is that the elasticity is what is most strongly involved, like STP, but with more elasticity and less viscosity. S1 exhibits side-by-side attraction, sphere-wall attraction and robust chaining (see Figure 4) consistent with our notions about the importance of normal stresses.

4.2 Sedimentation in Xanthan and Carbopol Solutions

We attempted to isolate the role of shear thinning suppressing both normal stresses and elasticity by using a solution of 0.4% Carbopol 690 (Goodrich) in a 50/50 glycerin/water mixture in our sedimentation experiments. The viscosity versus shear rate for this Carbopol solution is plotted in Figure 4, and the dynamic moduli are plotted in Figure 5. Carbopol is thought to be a pseudoplastic fluid without elasticity. Since our Carbopol solution has a non-zero storage modulus, it cannot be said to have no elasticity. The presence of small elasticity in Carbopol solutions has been noted before; for example, Hartnett and Kostic [1989] have noted that an aqueous solution of 1000 wppm Carbopol exhibits a phase shift between the input shear stress response of less than $\pi / 2$ radians, and that there is evidence that aqueous Carbopol solutions experience strong secondary motions in laminar flow in noncircular channels, but they do not reduce drag. There is no evidence that Carbopol 690 in 50/50 glycerin/water has a measurable value of the first normal stress difference and it does not climb a rotating rod.

To determine the effects of shear thinning with a strong memory but no normal stresses, we used a solution of 0.3% Xanthan (Kelco) in water. The graph of viscosity versus shear rate is shown in Figure 4, and the variation of the storage and loss moduli with frequency is shown in Figure 5. This Xanthan solution is very shear thinning and it apparently has normal stresses. We could not register a first normal stress difference on the Rheometrics fluid rheometer and the 0.3 solution would not climb a rotating rod. On the other hand, this fluid has a high storage modulus and can be said to be linearly elastic.

We turn next to a description of our experiments on the settling of long cylinders in aqueous solutions of Carbopol and Xanthan.

Place Figure 4 here

Place Figure 5 here

Place Figure 6 here

Place Figure 7 here

Place Figure 8 here

Despite the fact that our 0.3% solution of Xanthan has no measurable normal stresses in shear and will not climb up a rotating rod, it tilts from longside-on to broadside-on in the usual way, with all straight-down falling dominated by inertia ($\mathbb{R}>1$) under supercritical conditions ($M\geq1$). In fact, the data shown in Figure 7 suggests a more close correlation with $M=1$ than need, in fact, be the case. It could be argued that the relevant speed to be used in the interpretation of the tilt transition is the falling speed of the cylinder when it settles with its long side parallel to gravity, when its fall speed is greatest. The fall speed after turning is much slower, and it has been used in computing the numbers shown in Figures 6 through 12. If we had used the faster fall velocities, the Reynolds and Mach numbers larger than one would shift slightly to the right.

We could conclude that the turning in Xanthan has nothing to do with normal stresses in shear and possibly with any other nonlinear measure of elasticity. But we think that it would be wrong to conclude that the subcritical nose-down orientation is controlled by shear thinning alone. In section 5, we will present an argument to show how these dynamical effects may be pulled into play when shear thinning is put together with a strong linearly-elastic response, as it is in the Xanthan solutions.

The tilt transition diagrams for semi-dilute Xanthan (Figures 6 and 7) are just like the aqueous Polyox diagrams given by Liu and Joseph [1993a] and reproduced here as Figures 8 and 9. The Polyox solutions are elastic and shear thinning, like Xanthan, but they also have substantial normal stresses and climb rotating rods. The main difference between Xanthan solutions and Polyox solutions is associated with normal stresses which produce a phenomenon, termed by Liu and Joseph [1993a] as shape tilting which causes sharp edge cylinders to tilt slight when round and cylinder align with gravity. The shape tilting does not occur in Xanthan; all the particles align smartly with gravity under subcritical conditions.

We were surprised by the finding that light cylinders which fall slowly in Carbopol also tilt nose-down, with their long side parallel to gravity. These fluids in other respects are nearer to Newtonian than to viscoelastic. Spherical particles dropped in our 0.4% Carbopol do not exhibit side-by-side attraction, they repel each other when they are initially together or nearly together as in Newtonian fluids. They are repelled by a wall, do not chain and do not exhibit anomalous rolling or chaining, just as in Newtonian fluids.

The nose-down tilting of cylinders in Carbopol does not appear to be a supercritical transition; it occurs when $\mathbb{R}>1$ for Mach numbers say $M = 0.1$ or larger, (see Figures 8 and 9). We do not think that Carbopol is strongly elastic, even in the rest state, but it does have a non zero storage modulus (Figure 5). Data for shear wave speed is given in Table 4. We lose the signal in the larger gaps (>3mm) so that the decay of the signal with distance is more pronounced than in more elastic fluids with the same viscosity. The weak signals that are received are erratic, giving evidence to diffusion. However, the

data in the smaller gaps shown in Table 4 are consistent, and we can find no internal inconsistency which would cause us to reject this data.

Long bodies settling in Newtonian liquids will always turn broadside-on; no matter how small the Reynolds number inertia will eventually have its way. In these cases a Mach number criterion for turning cannot be relevant. There are surely intermediate cases of small non-Newtonian effects where some effects of the fluid's small elasticity are present together with Newtonian-like behavior. This may in fact be the case for the Carbopol solutions.

h (mm)	0.748	1.003	1.956
$\bar{c}$ cm/sec	16.8	17.2	15.7
ϑ cm/sec	0.79	1.47	1.06

Table 4. Mean wave speed $\bar{c}$ for 0.4% Carbopol in 50/50 glycerin/water solutions. The speeds measured in the 2.959 mm gap where erratic ranging from 22 cm/sec to 71 cm/sec with many signals too unstable to make a clear reading.

h (mm)	2.959	1.956	1.181	1.003
$\bar{c}$ cm/sec	12.170	12.000	14.960	12.450
ϑ cm/sec	0.630	0.700	0.850	0.790

Table 5. Shear wave speeds in 0.3% aqueous solution of Kelco Xanthan.

5. Tilt transition in semi-dilute Xanthan solutions

In this section we wish to discuss some conjectures about the mechanisms which control nose down orientations of cylinders falling in 0.3% Xanthan under subcritical conditions ($M<1$) and broadside-on orientations under supercritical conditions ($M>1$, see Figure 7). The fluid is strongly pseudoplastic (Figure 4) and has relatively high levels of linear viscoelasticity (Figure 5). The measured

values of the mean shear wave speed are shown in Table 2. We recall that the first normal stress difference is effectively zero at all shear rates and that this fluid will not climb a rotating rod.

Surprisingly also is the similarity in the properties of particle migrations in 3% Xanthan with standard Polyox and Polyacrilamide solutions, which have large normal stresses and also are strongly pseudoplastic, and with S1, which is moderately elastic and weakly pseudoplastic. All these fluids and STP give rise to critical attractive distance for side-by-side and sphere-wall attractions. All of them chain, with STP (the Boger fluid) the weakest chainer. All these fluids support anomalous rolling, with Xanthan exhibiting the weakest rolling. None of these phenomena occur in Newtonian fluids or in our Carbopol solution which shear thins.

Evidently a combination of memory with shear thinning is required and may be enough to induce nose-down turning and the related chaining of spheres. We introduce the idea that shear thinning alone does all these tricks to particles much less because though it shear thins, the thinning is not persistent and decays very rapidly. The Xanthan solution remembers the place where the viscosity was reduced, so that the back part of a nose-down cylinder, or the spheres behind the lead sphere in a chain, experience a smaller viscosity than the leading end of the cylinder or leading sphere. We might think that corridors of reduced viscosity are marked on the fluid by shear thinning as a particle drops in the fluid and persist for a time before they relax.

The idea of the last paragraph could be tested with standard test liquids provided that they shear thin and have long memories, whether or not they exhibit normal stresses or climb rods.

The existence of relaxing corridors of reduced viscosity marked on the fluid by the shear thinning induced by a falling ball is consistent with the observations of Cho and Hartnett [1979] and Cho, Hartnett and Lee [1984]. They did falling ball rheometry, measuring the drag on balls which were dropped in the test liquid in specified and definite intervals of time. They found the memory effects to which we have alluded, and they were particularly evident in 10^4 wppm solution of aqueous polyacrilamide (Separan, AP-273) which is a highly viscoelastic and highly shear thinning liquid. The measured terminal velocity depended strongly on the time interval between the dropping of successive balls in the cylinder. Balls launched after only a short wait period would fall up to nearly twice as fast as the speed of the initial ball, and it took intervals of thirty minutes or more for the memory of the corridor of reduced viscosity to relax.

We can imagine the trailing spheres in a chain or the trailing end of a long particle setting itself in a corridor of reduced viscosity. For this, shear thinning and the memory of shear thinning is required. It is prudent at this point to recall that similar effects in weaker form occur in our constant viscosity fluid (STP) and in stronger form in S1 where the degree of shear thinning is small. So shear thinning plus

memory cannot explain everything. The experiments with semi-dilute Xanthan are interesting because shear thinning and memory are present, but many other mechanisms which could come into play are absent.

Now we seek an explanation of why a tilt transition which turns particles which fall nose down when $U < c = \sqrt{n/\lambda\rho}$ broadside-on under supercritical conditions $U > c$. We may remark here that the relaxation time $\lambda = \eta/\rho c^2$ which we get by measuring η and c on our meter, need not have anything whatever to do with the relaxation of a corridor of reduced viscosity.

The part of our explanation which seems secure is that the tilt to broadside-on settling is an effect of inertia. the transition from nose-down to broadside-on settling occurs over a small interval of velocities and is easy to identify. We think that this turning is due to high pressures at the stagnation point on the front face of the falling cylinder. Huang, Feng and Joseph [1993] did a direct, two-dimensional simulation of an ellipse falling in Newtonian fluid. they identified the "stagnation point" as the point on the front of the cylinder where the shear stress vanishes and showed that the high pressure at such a point always gives rise to a couple turning the long side of the ellipse across the stream. This couple is larger than the couples due to shear stresses or the viscous part of the normal stress. We think that this same inertial mechanism which is associated with high pressures at the point on the front face where the shear stress vanishes operates in the viscoelastic case.

In the case of Newtonian fluids this inertial mechanism comes into operation immediately for any Reynolds number larger than zero. At very small Reynolds numbers the turning to a broadside on orientation takes a long time, but a cylinder which is dropped with its broadside perpendicular to the flow will not nose down, as it always does in viscoelastic fluid. Moreover, the tilting from nose-down to broadside-on orientation does not occur until the Reynolds number is greater than one in all the cases we have studied and the viscoelastic Mach number was also greater than one in all these cases, except Carbopol.

It did not at first occur to us that the change of orientation of long bodies sedimenting in different polymeric liquids could be framed as a change of type with features resembling those already seen in studies of delayed die swell (see [J1990] and Joseph and Christodoulou [1993]). The cylinders settle vertically, more or less, when the viscoelastic Mach number is less than some number not too much greater than one. When this Mach number is larger than about three or four the particles have all turned their broadside to the stream, evidently controlled by inertia.

The orientation of a long body settling in a liquid under gravity is equivalent to the steady flow past a stationary long body. This latter problem has been treated in works of Ultman and Denn [1970], Joseph [1985], Crochet and Delvaux [1990], Hu and Joseph [1990] and Fraenkel [1988,1991]. These

studies and related matters are discussed in [J1990]. The nonlinear studies were based on the upper convected Maxwell model, but the linearized studies were basically model independent. In the linearized problem the flow goes supercritical when the viscoelastic Mach number $M = U/c$ passes through one; the vorticity equation of the steady flow changes type from elliptic when $M < 1$ to hyperbolic when $M > 1$. In the supercritical case there is a Mach cone of vorticity. In the front of the cone there is a "region of silence," actually potential flow with vortical flow in the cone behind the shock. In the nonlinear problem there can be a subcritical region near the body even when $M > 1$, as in transonic flow; but away from the body linearized dynamics will prevail. The supercritical transitions do seem to correspond to flow transitions observed in the experiments on the flow over wires reported by James [1967], James and Acosta [1970] and by Ambari, Deslouis and Tribollet [1980] as well as in the flow features observed in the experiments on flow over flat plates of Hermes and Fredrickson [1967].

The experiments on anomalous transfer of heat and mass in the flow over small wires were carried out in dilute drag reducing solutions of polymers in water, say 10 to 100 ppm of polyethylene oxide in water. The critical issue posed by these experiments is the comparison of the critical values of the velocity above which anomalous behavior occurs with measured values of the wave speed. Some few values for wave speeds for very dilute solutions are presented in the tables at the end of the book of Joseph [1990], but they are not reliable. We need to find the techniques to do reproducible measurements of wave speeds in dilute solutions. Gas dynamics would look good on paper, but only there, if we could not measure the speed of sound. Fortunately reliable measurements of the wave speeds of solutions in which tilt transitions occur can be made. In every one of many cases the transition commences when the fall velocity exceeds the wave speed, in all cases without exception.

Goldshtik, Zametalin and Shtern [1981] gave an interesting interpretation for the criterion for the onset of drag reduction in a Maxwell fluid. They said that there are two lengths to consider, a viscous length $\nu/u*$ where ν is the kinematic viscosity and $u*$ is the friction velocity, and an elastic length $\lambda u*$ where λ is the relaxation time. Onset occurs when the elastic length exceeds the viscous one.

$$\lambda u* > \nu/u*. \tag{1.1}$$

We can apply the same idea to the tilt transition by claiming onset when

$$\lambda U > \nu/U. \tag{1.2}$$

The question is what λ should be used. Our answer is that $\lambda = \nu/c^2$ which we get by measuring ν and c (shear wave speed) on our meter. Then, $M^2 = (U/c)^2 > 1$, correlating unrelated dynamic data from many different experiments.

It is clear from the arguments just given that a key element of our description is the capacity of a fluid to transmit waves of vorticity into a region of rest faster than the fall velocity of the body. In fact, this description is not strictly accurate. Near the body, the problem is not linear and the condition for a change of type is not as simple as M>1. We are exploring the idea that when $\lambda U > \upsilon / U$ the viscoelastic effects at the front of the body are confined to a layer through which the high pressure in the essentially irrational flow at the "stagnation point" on the front of the body will turn the body broadside-on. A cartoon of the scenario we have in mind is shown in Figure 12.

Having now described a possible scenario giving rise to a tilt transition from a change of type, we note that the required propagation of shear waves into a region at rest is supported by the linearly elastic semi-dilute Xanthan solution, even though the normal stresses in this fluid are effectively zero.

The mechanism which keeps the cylinder aligned with its axis parallel to gravity in the subcritical case is unclear. This orientation cannot be maintained by the mechanisms discussed by Leal [1975] and Brunn [1977] which turn long bodies already at second order because the second order contribution to the stress is effectively zero in semi-dilute Xanthan. The extensional flow mechanism for turning at second order which is based on Roscoe's formula and is discussed by Liu and Joseph [1993a] also cannot operate in 0.3% aqueous Xanthan. The results of Walters, et al [1990] indicate that the aqueous Xanthan in much higher concentrations, 2% and 3%, give rise to extensional thinning at higher extension rates, which is incompatible with the turning mechanism based on Roscoe's formula. We noted already that the Roscoe formula gives no result for 0.3% Xanthan because the second order coefficients vanish, but Xanthan does have a small second order region in the aqueous solutions (500, 1000, 2000, and 4000 wpmm) of Rhodigel 23 (manufactured by Rhone Poulenc) used by Allain, Cloitre, Perrot and Quemada [1993] and by Joseph and Christodoulou [1993] in studies of delayed die swell, and for the more concentrated Kelco solutions studied by Walters et al [1990]. The observations, and the logic of it, lead us to consider the possibility that all the observations of the tilt transition (Figures 6 and 7) in 0.3% Xanthan can be explained by corridors of relaxing viscosity created by shear thinning plus memory and a transition to the dominance of inertia at fall speeds which exceed the speed of propagation of shear waves into rest.

An example of how all the mechanisms we have discussed can be made to appear in the White-Metzner model of a viscoelastic by choosing appropriate forms for the viscosity and relaxation functions of the shear rate $\overset{o}{\gamma} \overset{def}{=} \frac{1}{2} \text{trace} \, \mathbf{D} \, [\mathbf{u}]$, where $\mathbf{D}$ is the symmetric part of the velocity gradient.

The White-Metzner model is an upper convected Maxwell model $\lambda \overset{\Delta}{\boldsymbol{\tau}} + \boldsymbol{\tau} = 2 \eta \mathbf{D}[\mathbf{u}]$ in which $\lambda \left(\overset{o}{\gamma} \right)$ and $\eta \left(\overset{o}{\gamma} \right)$ are functions of the second invariant of $\mathbf{D}$. Suppose that λ and η are relating functions of the form given in Figure 9.

Place Figure 9 here

Figure 9. Relaxation or viscosity function for the WM model

Let us suppose that $\lambda\left(\overset{o}{\gamma}\right) \to 0$ already at small $\overset{o}{\gamma}$ and such the $\eta\left(\overset{o}{\gamma}\right)$ is still substantial.

Then $\quad \boldsymbol{\tau} = 2\eta(\gamma)\mathbf{D}[\mathbf{u}]$ is shear thinning. On the other hand in the regime of linear viscoelasticity, perturbations of uniform motion or small oscillations

$$\overset{\Delta}{\boldsymbol{\tau}} \Rightarrow \frac{\partial \boldsymbol{\tau}}{\partial t} + U\frac{\partial \boldsymbol{\tau}}{\partial x} \overset{\text{def}}{=} \overset{o}{\boldsymbol{\tau}}$$

and

$$\lambda(o)\overset{o}{\boldsymbol{\tau}} + \boldsymbol{\tau} = 2\eta(o)\mathbf{D}[\mathbf{u}]$$

This gives rise to the usual $c = \sqrt{\eta(o)/\rho\lambda(o)}$ for wave speeds and to change of type when $U>c$.

Barnes and Robert [1992] have given some forms for $\lambda\left(\overset{o}{\gamma}\right)$ and $\eta\left(\overset{o}{\gamma}\right)$ which fit our idea, and they can give rise to an extensional viscosity which first increases with $\overset{o}{\gamma}$, as would be expected when the climbing constant is positive, and then decreases as in the measurements of Walters, et al [1990].

Place Figure 10 here

Place Figure 11 here

Place Figure 12 here

References

Ambari, A., Deslouis, C. and Tribollet, B. 1984 Coil-stretch transition of macromolecules in laminar flow around a small cylinder. *Chem. Eng. Commun.* **29**, 63-78.

Armour, S.J., Muirhead, J.C. and Metzner, A.B. 1984 Filament formation in viscoelastic fluids. *IX International Congress on Rheology*.

Barnes, H.A. and Robert, G.P. 1992 A simple emperical model describing the steady-state shear and extensional viscosities of polymer melts. *Journal of Non-Newtonian Fluid Mechanics*, **44**, 113-126.

Beavers, G.S. and Joseph, D.D. 1975 The rotating rod viscometer. *J. Fluid Mech.* **69**, 475-511.

Brunn, P. 1977 The slow motion of a rigid particle in a second-order fluid. *J. Fluid Mech.* **82**, 529-550.

Chiba, K., Song, K. and Horikawa, A. 1986 Motion of a slender body in quiescent polymer solutions. *Rheologica Acta* **25**, 380-388.

Cho, K., Cho, Y.I. and Park, N.A. 1992 Hydrodynamics of a vertically falling thin cylinder in non-Newtonian fluids. *J Non-Newtonian Fluid Mech*. **45**, 105-145.

Cho Y.I. and Hartnett, J.P. 1979 The falling ball viscometer--a new instrument for viscoelastic fluids. *Letters in Heat and Mass Transfer*, **6**, 335-42.

Cho, Y.I., Hartnett, J.P. and Lee, W.Y. 1984 Non-Newtonian viscosity measurements in the intermediate shear rate range with the falling-ball viscometer. *Journal of Non-Newtonian Fluid Mechanics*, **15**, 61-74.

Cox, R.G. 1965 The steady motion of a particle of arbitrary shape at small Reynolds numbers. *J. Fluid Mech*. **23**, 625-643.

Crochet, M.J. and Delvaux, V. 1990 Numerical simulation of inertial viscoelastic flow, with change of type. In *Nonlinear Evolution Equations that Change Type* (eds. B. Keyfitz and M. Shearer). Springer-Verlag, New York.

Fraenkel, L.E. 1988 Some results for a linear, partly hyperbolic model of viscoelastic flow past a plate. In *Material Instabilities in Continuum Mechanics and Related Mathematical Problems* (ed. J.M. Ball). Clarendon Press, Oxford.

Fraenkel, L.E. 1991 Examples of supercritical, linearized, viscoelastic flow past a plate. *J. Non-Newtonian Fluid Mech*. **38**, 137-157**.**

Goldshtik, M.A., Zametalin, V.V. and Shtern, V.N. 1982 Simplified theory of the near-wall turbulent layer of Newtonian and drag-reducing fluids. *J. Fluid Mech*. **119**, 423-441.

Hartnett, J.P. and Kostic, M. 1989 Heat transfer to Newtonian and non-Newtonian fluids in rectangular ducts. *Advances in Heat Transfer*, **19**, 297-354.

Hassager, O. Negative wake behind bubbles in non-Newtonian liquids. 1979 *Nature* **279**, 402-403.

Hermes, R.A. and Fredrickson, A.G. 1967 Flow of viscoelastic fluids past a flat plate. *AIChE J*. **13**, 253-259**.**

Hu, H.H. and Joseph, D.D. 1990 Numerical simulation of viscoelastic flow past a cylinder. *J. Non-Newtonian Fluid Mech*. **34**, 347-377.

James, D.F. 1967 Laminar flow of dilute polymer solutions around circular cylinders. Ph.D. Thesis. Cal. Inst. Tech. Pasadena.

James, D.F. and Acosta, A.J. 1970 The laminar flow of dilute polymer solutions around circular cylinders. *J. Fluid Mech*. **42**, 269-288.

Joseph, D.D. 1985 Hyperbolic phenomena in the flow of viscoelastic fluids. In *Viscoelasticity and Rheology* (eds. A.S. Lodge, J. Nohel, and M. Renardy). Academic Press.

Joseph, D.D. 1990 *Fluid Dynamics of Viscoelastic Liquids*. Springer-Verlag, New York.

Joseph, D.D. 1992 Bernoulli equation and the competition of elastic and inertial pressures in the potential flow of a second order fluid. *J. of Non-Newtonian Fluid Mech*. **42**, 385-389.

Joseph, D.D., Arney, M.S., Gillberg, G., Hu, H., Hultman, D., Verdier, C. and Vinagre, T. M. 1992 A spinning drop tensioextensiometer. *J. Rheology* **36**, 621-662.

Joseph, D.D. and Christodoulou, C. 1993 Independent confirmation that delayed die swell in a hyperbolic transition. *J. of Non-Newtonian Fluid Mech.*, to appear.

Joseph, D.D., Nelson, J., Hu, H.H. and Liu, Y.J. 1992 Competition between inertial pressures and normal stresses in the flow induced anisotropy of solid particles. In *Theoretical and Applied Rheology* (eds. P. Moldenaers and R. Keunings). Elsevier Science Publishers B.V., 60-65.

Leal, L.G. 1975 The slow motion of slender rod-like particles in a second-order fluid. *J. Fluid Mech*. **69**, 305-337.

Liu, Y.J. and Joseph, D.D. 1993a Sedimentation of particles in polymer solutions. *J. Fluid Mech.* (to appear), see AHPCRC reprint.

Liu, Y.J. and Joseph, D.D. 1993b Aggregation and dispersion of particles falling in viscolelastic liquid. Submitted to *Journal on Non-Newtonian Fluid Mechanics*.

Roscoe, R. 1965 The steady elongation of elasto-viscous liquids. *Brit. J. appl. Phys*. **16**, 1567-1571.

Ultman, J.S. and Denn, M.M. 1970 Anomalous heat transfer and a wave phenomenon in dilute polymer solutions. *Trans. Soc. Rheol.* **14**, 307-317.

Walter, K., Bhatti, A.Q. and Mori N. 1990 The influence of polymer conformation on the rheological properties of aqueous polymer solutions. Found in *Recent Developments in Structured Continua*, Volume 2, D. DeKee and P.N. Kaloni, eds., New York: Wiley Publishers.