

Open Problem

Open mathematical problems regarding non-Newtonian fluids

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Abstract. We present three open problems in the mathematical modelling of the flow of non-Newtonian fluids. The first problem is rather long standing: a discontinuity in the dependence of the rise velocity of a gas bubble on its volume. This is very well characterised experimentally but not, so far, reproduced either numerically or analytically. The other two are both instabilities. The first is observed experimentally but never fully predicted analytically or numerically. In the second instability, numerical studies reproduce the experimental observations but there is as yet no analytical or semi-analytical prediction of the linear instability which must be present.

1. Introduction

The major open question regarding non-Newtonian fluids is very simple: what equations should I use to model this fluid? This is not a mathematical question but addresses issues of physics, chemistry and engineering. However, it lies at the root of many of the mathematical problems presented in this paper.

Each of the problems which follows is essentially of the same form: a physical phenomenon which we have not yet successfully modelled. In one case the phenomenon has been reproduced by numerical simulation, which allows us to be certain that we have chosen the “correct” equations (though there is almost certainly more than one “correct” set); nonetheless, we do not have a mathematical (and hence physical) understanding of what is going on. In other cases, the open problem includes selection of a fluid model as well as solving the resultant equations.

Any incompressible fluid model can be specified using the following equations of motion, in which we introduce the variables \mathbf{u} , the fluid velocity; p , the pressure; \mathbf{s} , the stress tensor; and \mathbf{s}^p , the polymer extra stress tensor; and the parameters ρ (fluid density) and η_s (solvent viscosity, which may be set to zero). The variables may all depend on both position and time, t . We have mass and momentum conservation:

$$\nabla \cdot \mathbf{u} = 0, \quad \rho [\partial_t + (\mathbf{u} \cdot \nabla)] \mathbf{u} = \nabla \cdot \mathbf{s}, \quad (1)$$

and the definition of the polymer stress (in which \mathbf{I} is the identity tensor and $\mathbf{E} = \frac{1}{2}(\nabla \mathbf{u} + \nabla \mathbf{u}^\top)$ the velocity gradient tensor):

$$\mathbf{s} = -p\mathbf{I} + 2\eta_s\mathbf{E} + \mathbf{s}^p. \quad (2)$$

To complete the model we need the *constitutive equation* which governs the evolution of \mathbf{s}^p as a function of the flow history, and determines the fluid properties. The

examples we see in this paper (which are all appropriate for dilute polymer solutions rather than melts) may all be represented by the following generic equation:

$$\mathbf{s}^p + \lambda \overset{\nabla}{(f\mathbf{s}^p)} = - (\eta_p g \mathbf{I}) \quad (3)$$

in which λ is the relaxation time, η_p the polymer viscosity, and the upper-convected tensor derivative is defined as

$$\overset{\nabla}{\mathbf{A}} = [\partial_t + \mathbf{u} \cdot \nabla] \mathbf{A} - (\nabla \mathbf{u})^\top \cdot \mathbf{A} - \mathbf{A} \cdot \nabla \mathbf{u}. \quad (4)$$

We can extract many standard models from this generic form: in particular, the Oldroyd B (OB) and Upper Convected Maxwell (UCM) models (UCM is the special case of OB with $\eta_s = 0$), and two FENE models based on Finitely Extensible Nonlinearly Elastic dumbbells: FENE-P and FENE-CR. The parameters for these three models are given below.

$$\text{OB:} \quad f = g = 1 \quad (5)$$

$$\text{FENE-P:} \quad f = \frac{g}{a} = \frac{L^2 - 3}{L^2 + (\lambda/a\eta_p) \text{tr}(\mathbf{s}^p)} \quad a = \frac{L^2}{L^2 - 3} \quad (6)$$

$$\text{FENE-CR:} \quad f = \frac{L^2 - 3}{L^2 + (\lambda/\eta_p) \text{tr}(\mathbf{s}^p)} \quad g = 1 \quad (7)$$

where, in the FENE cases, L is the dimensionless length of the finite dumbbell.

The upper-convected derivative (4) is the derivative appropriate for line elements: an infinitesimal line segment \mathbf{dl} whose ends advect passively with flow will produce a tensor $\langle \mathbf{dl} \mathbf{dl} \rangle$ whose upper-convected derivative is zero. This is exactly equivalent to the usual material derivative (as seen on the left hand side of the momentum equation, for example), which is zero for the position of a passively advecting fluid point. As such (4) appears naturally in fluids which are derived from microscopic models involving dumbbells. In particular, the OB model can be derived by treating each polymer molecule as an isolated dumbbell of two beads connected by a spring (and neglecting hydrodynamic interactions between the beads) with a Hookean spring law.

The OB model [1] is one of the simplest of constitutive models to be a reasonable model for some specific polymer solutions. Under shear flow, it has a constant shear viscosity $\eta_s + \eta_p$, and a positive first normal stress difference (defined as $N_1 = s_{xx} - s_{yy}$ if the flow is in the x -direction with flow gradient in the y -direction) which depends quadratically on the shear rate. These two facets are matched rather well by so-called *Boger fluids*, in which a polymer is dissolved at low concentrations in a solvent consisting of short-chain polymers or monomers of the same material. However, in extensional flows OB has a serious problem: at a critical flow rate the resistance to flow diverges, and at higher rates an anomalous negative viscosity is produced, which is unphysical.

The FENE models shown in equations (6–7) were introduced to address this problem, which results from the Hookean dumbbells of the OB model being stretched to their full extent. The function f describes a nonlinear spring law with a finite maximum extension L , and the FENE-P model [2] is derived directly from the dumbbell model with this nonlinear spring law. The derivation is no longer perfect (a preaveraging assumption needs to be introduced to close the set of equations) but this is the logical next modelling step after OB. In shear flow it shows mild shear-thinning (the shear viscosity decreases with shear rate, which is a common physical phenomenon), and in extensional flow the anomalous negative viscosity has

been eliminated; the viscosity increases with flow rate but does not asymptote to infinity at a finite flow rate.

Finally, the FENE-CR model [3] is slightly more empirical. The function g , which naturally corresponds (apart from a scaling factor) to the nonlinear spring law f in the derivation of FENE-P, is modified to artificially eliminate shear-thinning. The model still avoids the unphysical problems of OB in extensional flow, and although it is not microscopically derived, is very useful in modelling to separate the effects of shear-thinning from truly elastic effects such as the first normal stress difference and a high extensional viscosity.

1.1. Dimensionless groups

In seeking to clarify the mechanism of any phenomenon, it is helpful to make the governing equations dimensionless. If we choose a lengthscale \mathcal{L} , velocity scale U , and timescale T (which may not be equal to L/U) based on the physical scenario, we have a natural viscous stress scale $\eta U/\mathcal{L}$ (introducing $\eta = \eta_s + \eta_p$ for the total zero shear viscosity). We can then form several standard dimensionless groups related to the flow, shown in table 1.

Reynolds number	$Re = \rho U \mathcal{L} / \eta$	Ratio of inertial to viscous forces
Weissenberg number	$We = \lambda U / \mathcal{L}$	Ratio of elastic to steady viscous terms
Deborah number	$De = \lambda T$	Elasticity relative to flow time
	$\beta = \eta_s / \eta$	Ratio of solvent to total viscosity
	L	Dimensionless length of FENE dumbbell

Table 1: Dimensionless groups in viscoelastic fluid mechanics.

Of these groups, the Reynolds number Re will be familiar to those working in Newtonian fluid mechanics. This will be the least important group in the discussions which follow, as I have chosen phenomena which cannot be reproduced using Newtonian fluids and thus cannot be driven by inertia alone.

2. The rise velocity jump discontinuity for a gas bubble

Consider a gas bubble rising through a viscoelastic fluid. Whatever initial transients there are, there will be a long-term average rise velocity which one can plot against bubble volume. The resultant plot shows a very sharp rise which effectively represents a discontinuity, as shown in the experimental results of figure 1a. This phenomenon was first studied by Astarita & Apuzzo in 1965 [4] and has been the focus of intense study ever since.

There is good experimental evidence of a correlation between the size at which we see the velocity discontinuity, and two related observations: a cusp on the trailing end of the bubble [5], and a *negative wake* behind the bubble [6]. The negative wake is a recirculation behind the rising bubble in which the fluid is locally moving downwards relative to the quiescent fluid far from the bubble. These two phenomena appear at around the same time, but cannot be inextricably linked as the negative wake can also be created behind a solid sphere [7]. The negative wake alone does not cause the discontinuity, as falling solid spheres show no such phenomenon. However, the most recent work on this subject, by Lind & Phillips [8], suggests that the cusp alone is also

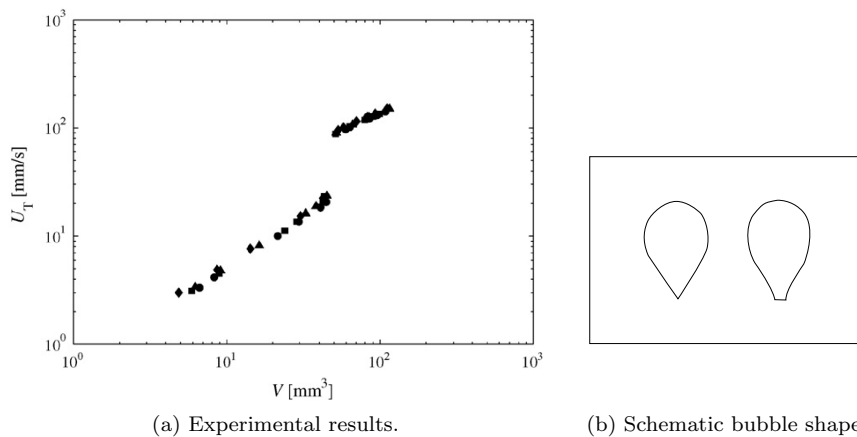


Figure 1: (a) Measured rise velocity U_T of a gas bubble in a dilute polymer solution, plotted against bubble volume V . Taken from figure 6 of [5] with permission. (b) Schematic of the generic shape of a cusped bubble. From the side the cusp is sharp; from the front it has a 2D, blade-like profile.

insufficient to predict the velocity discontinuity: their simulation method captures many aspects of bubble dynamics, including the trailing cusp, but its formulation explicitly excludes the formation of a negative wake and they did not see any velocity discontinuity. This work [8] uses the UCM model (5); another recent paper [9], using the OB model (like UCM except that $\eta_s \neq 0$) did predict the jump discontinuity using a numerical scheme which captures both the cusp and the negative wake. The introduction to [8] provides an excellent and thorough review of the history and current state of play for this phenomenon.

Because of the essential presence of a free surface to reproduce the velocity discontinuity, there is an extra ingredient to this problem compared to the other two. In addition to the governing equations (1–7), we need to specify the jump in normal stress across the air-fluid interface. In general, this jump depends not just on the stress in the fluid matrix, but also on surface tension, described with coefficient σ . The final boundary condition for this problem becomes:

$$\mathbf{s} \cdot \mathbf{n} = [-p_g + \sigma\kappa] \mathbf{n} \quad \text{at the interface,} \quad (8)$$

in which \mathbf{n} is the unit normal to the interface, κ the curvature of the interface, and p_g is the pressure inside the bubble. The introduction of an extra physical parameter, σ , introduces a further dimensionless group in addition to those shown in table 1: the Capillary number, $Ca = \eta U / \sigma$, which is the ratio of viscous stresses to surface tension.

To some extent the cause of the sharp cusp behind the bubble is intuitively obvious: polymer molecules which become stretched by the flow around the bubble tend to align behind it, naturally “pulling” the soft gas surface of the bubble backwards. Surface tension acts to counteract this effect, which accounts (very loosely) for the size dependence, as the polymer stretch will increase with increasing bubble size (and hence speed), whereas surface tension effects are larger for smaller bubbles with a smaller radius of curvature. However, there is little physical understanding of the negative wake, nor of the acceleration that takes place when the trailing cusp first appears.

3. Helical instabilities upstream of a contraction flow

Contraction flows occur in many polymer manufacturing processes, as molten polymer is extruded to form (ideally) a smooth, uniform thread. However, like so many flows, contraction flows can be subject to instabilities. This iconic, and as yet unexplained, instability was first observed by Nguyen & Boger in 1979 [10] using a dilute polymer solution: in figure 2a we see images from page 361 of that paper (which also appear in [11]), taken upstream of a cylindrical 8:1 contraction. A steady, axisymmetric flow would be left-right symmetric in the laser-sheet images shown here. In the first image the vortex on the left dominates; in the second, that on the right dominates. The third and fourth images do not show the large vortices. Essentially an asymmetric pattern as seen in the first figure is rotating about the central axis: the final two images show a different cross-section of the flow pattern from the first two. The fluid used in their experiments [10] was a dilute polyacrylamide solution, which should be well modelled by a FENE dumbbell model such as those presented in (6–7). A more recent observation of the same phenomenon may be seen in [12].

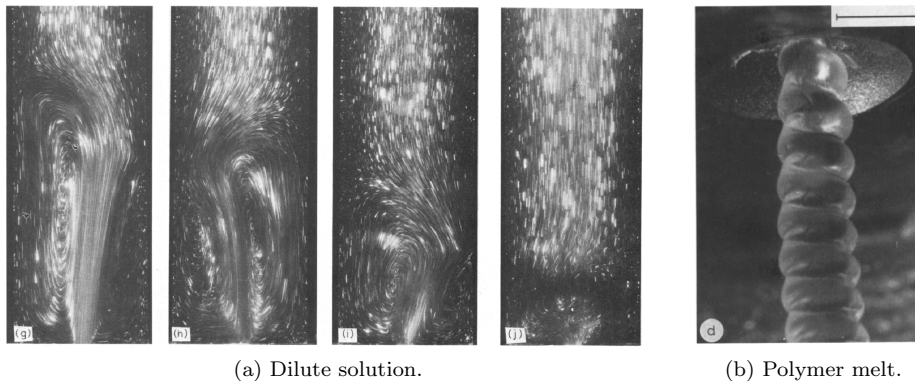


Figure 2: Helical instabilities in extrusion. (a) Helical instability in a dilute PAM solution upstream of an 8:1 contraction. The images are taken at different times. Taken from [10], page 361. (b) Helical extrudate following extrusion of a linear silicone oil through a cylindrical orifice die. Taken from figure 6 of [13].

There is a related, but possibly different, instability in polymer melts (which have a very different fluid rheology). As flow speed is increased in die extrusion, the first bulk (rather than surface) distortion seen in the extrudate is often helical in form, as seen in figure 2b. It has long been proposed (White [14, 15]) that the origin of this helical distortion is a spiral fluid instability upstream of the contraction.

Perhaps it is because of the inherently 3D nature of these instabilities (helical disturbance around an axisymmetric flow), but as yet there has been no analytical or numerical reproduction of either instability. There is a large literature of planar simulations of entry flows, but to my knowledge no fully three-dimensional studies which could capture this phenomenon. The critical mechanism must depend on the elastic stresses created in the polymer fluid; at this stage it is not possible to speculate further on the mechanism (or mechanisms) of instability.

4. Symmetry-breaking instability in the cross-slot device

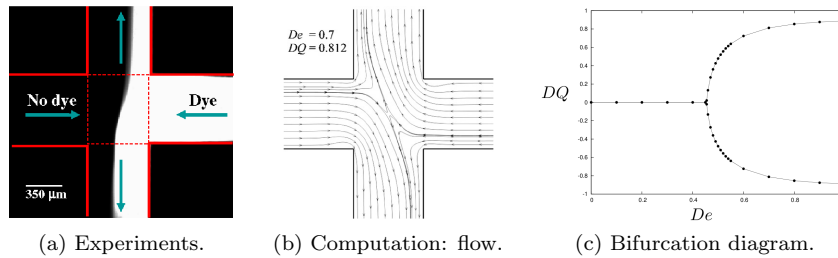


Figure 3: Asymmetry in a cross-slot device. (a) Experiments with a polyacrylamide fluid, taken from figure 1 of [16]. The fluid entering from the left is preferentially leaving by the top outlet. (b) Numerical simulation using the FENE-CR model, taken from figure 4(d) of [17]. (c) Numerical calculation of the dependence of the asymmetry parameter DQ on dimensionless flowrate De for the FENE-CR model with $\eta_p = 9\eta_s$, $L^2 = 100$ (as in (b)). The Deborah number is defined here as $De = \lambda U/d$ where U is the average velocity across each of the inlet channels, and d is the width of each channel. Data taken from table 2 of [17].

The phenomenon here is a symmetry-breaking instability. It was first observed by Gardner *et al* [18], and forgotten for many years, but recently rediscovered by Arratia *et al* [16] using a polymer solution in a micro-scale apparatus (in which inertia can be neglected). The geometry is a cross-slot device, as illustrated in figure 3, in which fluid enters through two opposing channels (say, at the left and right) and leaves through the other two (say, top and bottom). At very low flow rates the flow is steady and symmetric; at higher flow rates it is observed to become asymmetric, as shown in figure 3.

This instability has been reproduced numerically by Poole and coworkers [19, 17] using both the UCM model of (5), which is an unsatisfactory model for any extensional flow, and the two FENE models described in (6–7). If we denote by δ the proportion of the fluid leaving the top outlet which originated in the left inlet, the asymmetry was parametrised using $DQ = 2\delta - 1$; the results look like a textbook pitchfork bifurcation. However, as yet there is no prediction (numerical or analytical) of the corresponding linear instability beyond the bifurcation point. The closest attempt to date is an early work by Lagnado *et al.* [20], who found an instability in the idealised flow at the centreline (plane strain without the influence of the walls) for an OB/UCM fluid, but only for relatively short waves out of the plane of flow, whereas the perturbations seen here all lie within that plane.

Physically, it seems that the driving force for the symmetry-breaking is the natural need of the system to find the least dissipative flow solution. The highly-extended polymers produced at the stagnation point of the steady, symmetric flow can store elastic energy and produce a central strand which feels almost solid to the surrounding fluid, effectively separating the outgoing channel into two half-width channels, causing much more viscous dissipation than the equivalent flow through a single side channel. The asymmetric states have two advantages. First, the outflow is effectively split into a wide channel and a narrow channel in each slit, which can (between them) produce a greater flow for the same viscous pressure drop than two half-width channels. Second,

and perhaps more important, the stagnation point changes its nature and becomes the centre of a small region of shear flow, which produces much lower elastic stresses than steady strain for the dumbbell models used here. Thus at least two physical mechanisms can be postulated by which the system flows more effectively after the bifurcation. Which is the correct one remains an open question.

5. Discussion

This paper has presented a small selection of open problems in non-Newtonian fluid mechanics. It is not intended to provide a comprehensive list, or a really full description of each problem; it is simply an overview of my personal selection of interesting problems. Arguably, some of these are physical rather than mathematical problems; nonetheless, they all have an applied-mathematical aspect: the problem lies in constructing a predictive mathematical model that captures these phenomena. I hope that the descriptions I have given here will prompt some readers to follow up the references and bring their own expertise to this exciting area.

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