Korea-Australia Rheology Journal
Vol. 12, No. 1, March 2000 pp. 27-38

Experimental dilemmas in non-Newtonian fluid mechanics and their theoretical resolution

D. V. Boger* and K. Walters
Department of Chemical Engineering, University of Melbourne, Australia
Department of Mathematics, University of Wales, Aberystwyth, UK

1. Introduction

There is no doubt that non-Newtonian Fluid Mechanics has made significant strides in recent years and there is a growing belief that the many provocative experimental phenomena and dilemmas now have a realistic possibility of being explained theoretically. We intend to illustrate this optimism by appealing to three important benchmark problems in non-Newtonian Fluid Mechanics, namely contraction flows, settling and die swell.

Fig. 1 illustrates the process for the solution of viscoelastic fluid mechanics problems. In contrast to Newtonian fluid mechanics, non-Newtonian fluid mechanics has had to be concerned with the development of general constitutive equations for viscoelastic fluids. These constitutive equations should in principle lead to the definition of flow properties that need to be measured to define the viscoelastic fluid (rheometry) and to the development of the equivalent Navier Stokes equations for the solution of all possible boundary value problems. The process is completed by solution of the appropriate equations, where the methods of computational fluid mechanics have been required; analytical methods for complex flows of viscoelastic fluids are generally not useful.

The full story, illustrated in Fig. 1, then involves these various strands of activity and it will be necessary to consider at least four of them in some detail. For example, we shall need to be quite specific about the experimental conditions pertaining to the relevant phenomena. The flows are invariably complex and the 'experimental dilemmas clearly refer to *complex* flows, where the flow domain often involves abrupt changes in geometry, and where the flow strength is high enough to permit a terminology which majors on 'high Weissenburg numbers' and 'high Deborah numbers'. This is of course reasonable obvious, but it nevertheless needs to be stated.

So we want to address the question: "How do elastic liquids behave in complex flows?" and it is immediately apparent that the answer must involve a consideration of how the same liquids behave in *simple* flows, so that obtaining rheometrical data on the test liquids is an essential part of the exercise. Such data, when available, serve more than one useful purpose; they certainly provide a foundation set of data, which must be accommodated in the associated mathematical model for the test liquids. That is to say, the constitutive equation, which is an essential ingredient in any theoretical resolution of the experimental dilemmas, has to be consistent with the rheometrical data. Indeed, if the model cannot simulate behaviour in simple flows, what chance does it have in complex flows?!

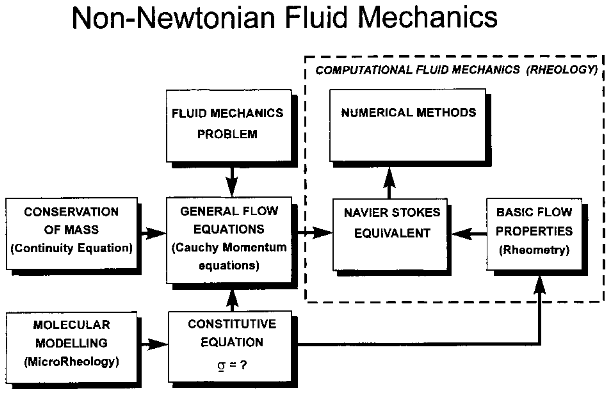


Fig. 1. The procedure for the solution of a Non-Newtonian Fluid Mechanics problem.

Clearly, the choice of constitutive equation is central to the whole operation and this choice is far from trivial or obvious. Indeed, a constitutive model which satisfies the dual constraints of tractability and quantitative (or even semi quantitative) prediction may not exist! But that shouldn't and doesn't prevent a search for this missing link'; but it is wise to be aware of the possibility of disappointment.

As is illustrated in Fig. 1, the constitutive model has to be solved in conjunction with the stress equations of motion and the equation of continuity, to predict and explain the experimental phenomena and dilemmas. Analytic solutions are out of the question so far as complex flows are concerned and Computational Rheology is now an established, if fairly recent science, which seeks theoretical answers to provocative experiments and phenomena. Computational Fluid Dynamics (CFD) has been an essential

*Corresponding author: d. boger@chemeng.unimeb.edu.au

part of the scene for at least twenty years and significant advances have been made in recent years, with accelerated progress reflecting the acceleration in the size and speed of computers.

So, the paper will need to address five topics:

(i) How do the test liquids behave in simple flows? i.e. Rheometry.

(ii) What constitutive equation should we associate with the test liquids? i.e. Constitutive Modelling.

(iii) What can Computational Rheology tell us about the way the test liquids ought to behave in complex flows? i.e. CFD within a non-Newtonian setting.

(iv) How do the liquids behave experimentally in complex flows? What flow features are distinctively rheological in nature? What experimental dilemmas emerge?

(v) Can the dilemmas of (iv) be resolved by the simulations of (iii)? If not, why not? Is it the deficiencies of the numerical codes or is it the poor choice of constitutive model? What is the current standing of Computational Rheology and what are its successes and failures? What are the outstanding problems?

In order to answer at least some of these questions, we shall be selective in our choice of experimental phenomena. We shall also restrict attention to a limited number of representative test liquids. So, for example, the so called Boger fluids will figure prominently in the discussion, since much of the experimental work (certainly on polymer *solutions*) has concentrated on this important sub class of elastic liquids. In the main, they are highly elastic liquids that exhibit a high resistance in stretching flows, their main attraction being their reasonably constant shear viscosity. This means that the well-known problems of defining dimensionless numbers for shear thinning fluids are avoided.

2. Some Representative Experimental Dilemmas

A better heading to this section might be some representative experimental *phenomena* since it might be pessimistic to talk of dilemmas and more correct to refer to challenges.

Highly elastic liquids are known to generate many extravagant effects not encountered in classical Newtonian Fluid Mechanics (see Boger and Walters, 1993) but we shall simply refer to three examples, which have all been used as important benchmark problems for workers in Computational Rheology.

2.1. Tubular Entry Flow

The first phenomenon refers to flow through axisymmetric contractions. The most provocative aspect of viscoelastic behaviour in these geometries is so called vortex enhancement, although the associated finding that, in the build up of vortex enhancement, an unexpectedly unsteady asymmetric flow regime can occur is also worthy of mention and study.

The basic elements of laminar flow through an abrupt entry circular contraction are illustrated in Fig. 2. The flow progresses from being fully developed at a plane some distance upstream from the contraction to being fully developed in the downstream tube at a distance, L_e, from the contraction plane. Depending on the Reynolds number of the flow and the fluid type, Newtonian or non-Newtonian, a secondary flow vortex may be present in the corner of the upstream tube, as is illustrated. A vortex may also grow from the re-entrant corner, which has been called the lip vortex. Conditions do exist for elastic liquids where both the lip and corner vortices are present at the same time. The shape of the vortex boundaries may be convex, concave or straight, with respect to the contraction corner. Characteristics of the corner vortex are of particular interest in the design of extrusion dies.

Figs. 3-9 all illustrate the influence of fluid elasticity in a circular entry on the flow in the absence of any shear rate dependent viscosity effects or fluid inertia. Fig. 3 illustrates the vortex growth observed for an elastic liquid in a circular 4 to 1 contraction, which stimulated the imagination of the Computational Rheology community. Fig. 3 illustrates the growth of the vortex and traces the increase as the shear rate and Weissenberg number increase. Above the Weissenberg condition ((d) in Fig. 3), the vortex becomes asymmetric in the tube and rotates about the tube wall. The frequency of this rotation increases with increasing flow rate until the periodic helical flow illustrated in Fig. 4 in a 7.67 to 1 contraction is observed. Figs. 3-9 illustrate the significant effects fluid elasticity has on the flow; without the elasticity of the liquid, no change in the flow field would be observed from the small corner vortex observed and predicted for inelastic Newtonian fluids, which is similar to that shown in Fig. 3(a) for the early stage of development of the flow phenomena.

Both a lip and corner vortex can be present in contraction

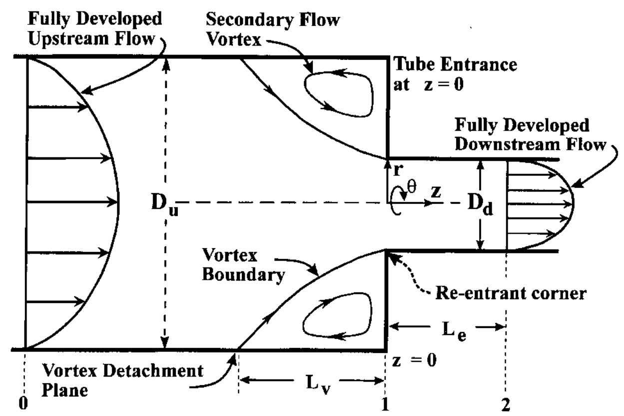
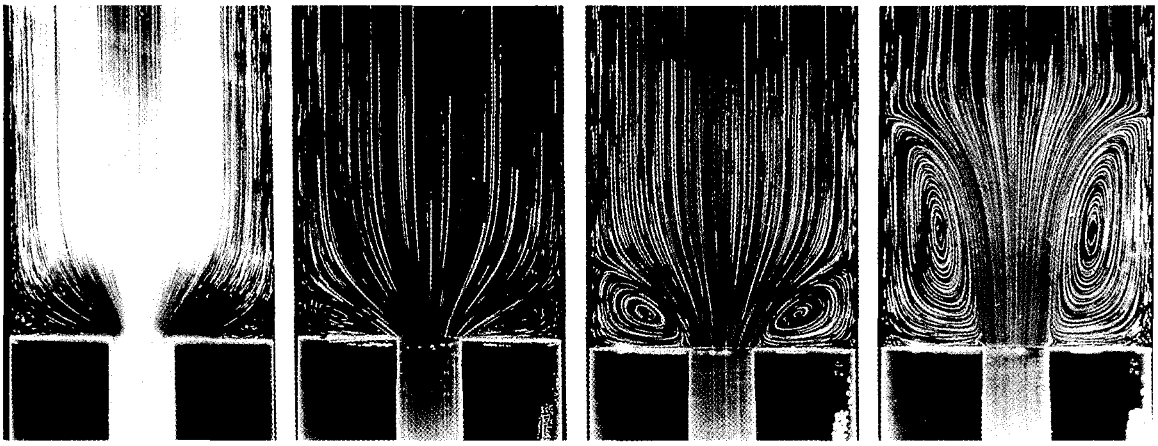
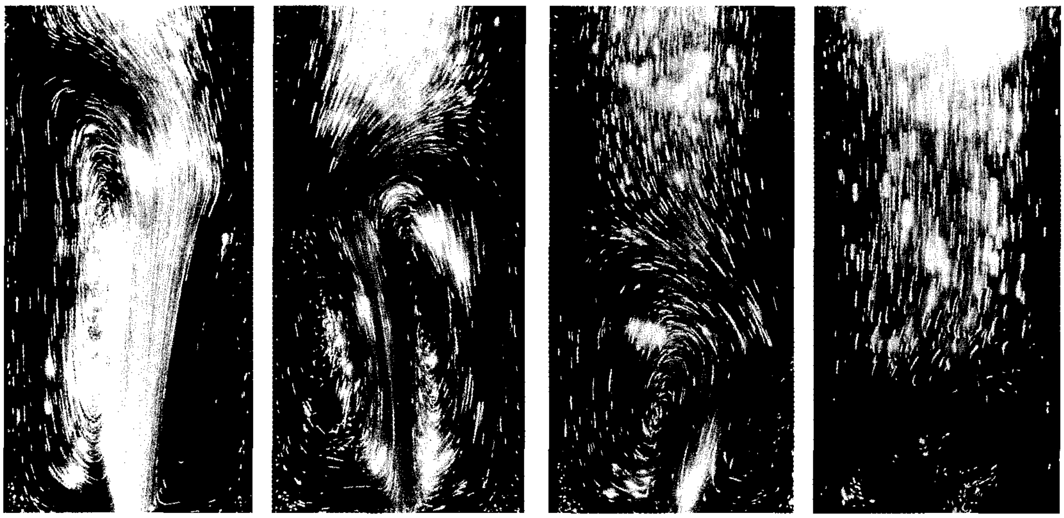


Fig. 2. Basic elements of an entry flow for flow from a large tube through an abrupt entry into a smaller tube.



(a) (b) (c) (d)

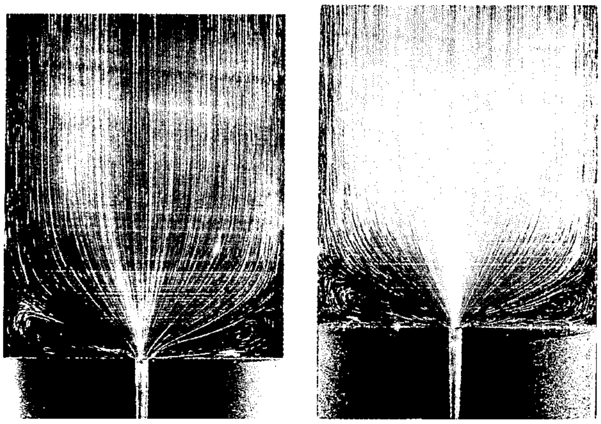
Fig. 3. Vortex growth for creeping flow in a 4 to 1 contraction for a Boger fluid (0.04% polyacrylamide (Separan AP30) in water and corn syrup solution). (a) $\dot{\gamma}=1.1\ s^{-1}$, $R_e=5.7\times10^{-4}$, $W_e=0.079$; (b) $\dot{\gamma}=3.4\ s^{-1}$, $R_e=1.76\times10^{-3}$, $W_e=0.120$; (c) $\dot{\gamma}=9.3\ s^{-1}$, $R_e=4.8\times10^{-3}$, $W_e=0.179$; (d) $\dot{\gamma}=24.2\ s^{-1}$, $R_e=1.25\times10^{-2}$, $W_e=0.204$. (From D.V. Boger, D.U. Hur and R.J. Binnington, *J. Non-Newt. Fluid Mech.*, **20**, 1986, 31, and reproduced from Boger and Walters, 1993.)



(a) (b) (c) (d)

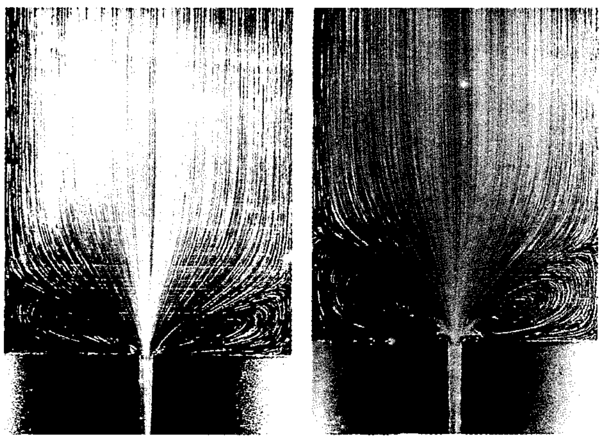
Fig. 4. Periodic helical flow in a 7.67 to 1 contraction for a Boger fluid (0.05% polyacrylamide (Separan MG500) in a glucose solution). $\dot{\gamma}=300\ s^{-1}$, $R_e=2.9\times10^{-2}$. (a) to (d) illustrate the sequence. The flow lines descent into the downstream tube like a tornado, i.e. when viewed in two dimensions, the points of contact between the flow lines at the wall, at fixed flow rate, gradually move downstream, with the secondary flow vortex diminishing in size until it disappears into the downstream tube, with the fluid in the larger upstream tube surging into the smaller tube. A large new vortex is then formed and the process is repeated. (From H. Nguyen and D.V. Boger, *J. Non-Newt. Fluid Mech.*, **5**, 1979, 353, and reproduced from Boger and Walters, 1993.)

flow at the same time time; this is illustrated in Fig. 5 for the international test fluid M1. A great deal of detailed information of the flow properties of fluid M1 is available (J.Non-Newtonian Fluid Mech., 35(2&3) 1989). In Fig. 5, notice the interaction of the two vortices, the straightening of the vortex boundary from sequence (a) to (b), while the re-attachment length of the cell, L_v, hardly changes. Note also the long photographic exposure time needed for definition of both vortices. As the shear rate is increased, the lip vortex devours the corner vortex, the vortex boundary becomes concave and the size of the cell increases, as is shown in Fig. 6. Close examination of the enlargement of Fig. 6 in Fig. 7 shows the start of a new lip vortex at the re-entrant corner. In a smaller contraction (4 to 1) for a Boger fluid of similar composition to Fluid M1, a pulsating lip vortex can also be observed where the



(a) (b)

Fig. 5. The co-existence of a lip and corner vortex for Fluid M1 (a 0.244% polyisobutylene solution in a mixed solvent consisting of 7% kerosene in polybutene) in a 22 to 1 circular contraction. (a) $\dot{\gamma}=89$ s^{-1}, R_e $\dot{\gamma}=0.0044$, 1 $\dot{\gamma}=6.8$, $X=0.21$ (exposure time = 16 min); (b) $\dot{\gamma}=94$ s^{-1}, $R_e=0.0054$, 1 = 7.2, $X=0.195$ (exposure time = 21 min). (From D.V. Boger and R.J. Binnington, *J. Non-Newt. Fluid Mech.*, **35**, 359, and reproduced from Boger and Walters, 1993.)



(a) (b)

Fig. 6. Vortex growth for Boger fluid M1 in a 22 to 1 contraction. (a) $\dot{\gamma}=170$ s^{-1}, $R_e=0.0085$, 1 $\dot{\gamma}=12.8$, $X=0.25$ (exposure time = 10 min); (b) $\dot{\gamma}=342$ s^{-1}, $R_e=0.017$, 1 $\dot{\gamma}=25$, $X=0.35$ (exposure time = 4 min). As the shear rate is increased the lip vortex devours the corner vortex, the vortex boundary becomes concave and the size of the cell increases. Close examination of (b) reveals the start of a new lip vortex at the re-entrant corner. (See the enlargement in Figure 7). (From D.V. Boger and R.J. Binnington, *J. Non-Newt. Fluid Mech.*, **35**, 1990, 359, and reproduced from Boger and Walters, 1993.)

lip vortex oscillates in size with a similar period (see Fig. 8). The growth of a lip vortex through the oscillatory stage is shown in Fig. 9. Fig. 10 illustrates in dramatic fashion

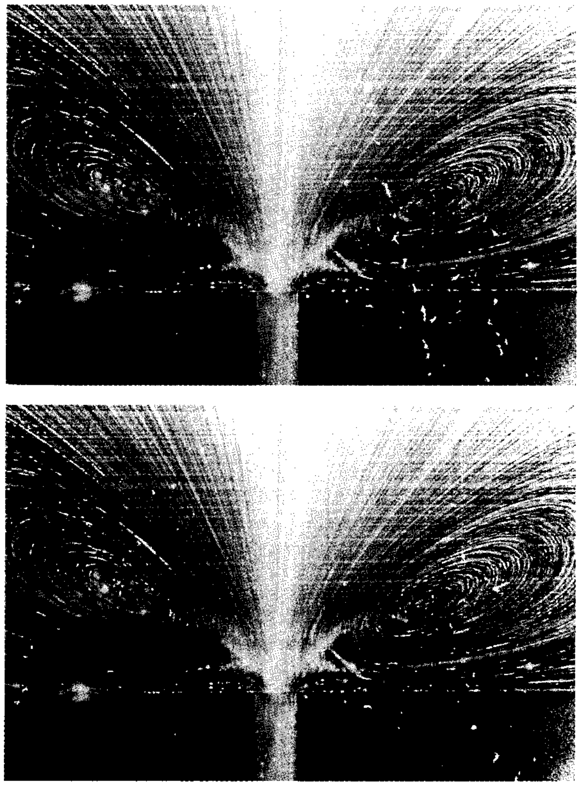
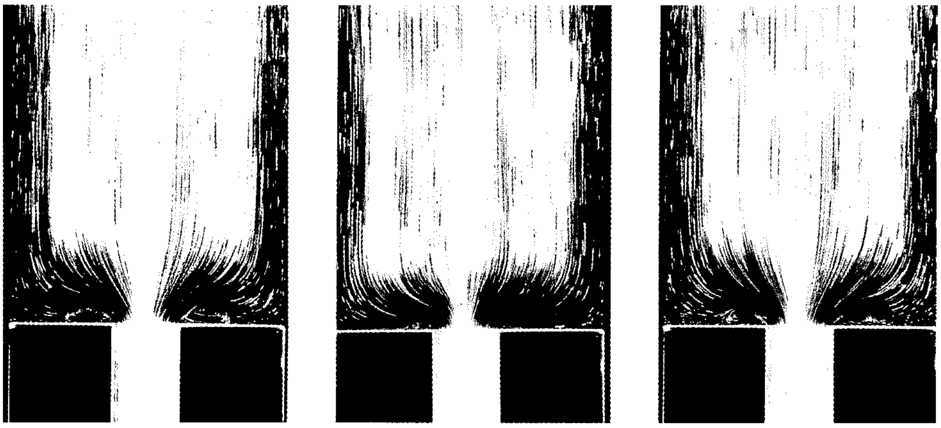


Fig. 7. The Formation of a new lip vortex for fluid M1 in the presence of a large recirculating vortex (enlargement of Figure 6).

the sensitivity of the lip vortex to the geometry of the re-entrant corner in a circular contraction.

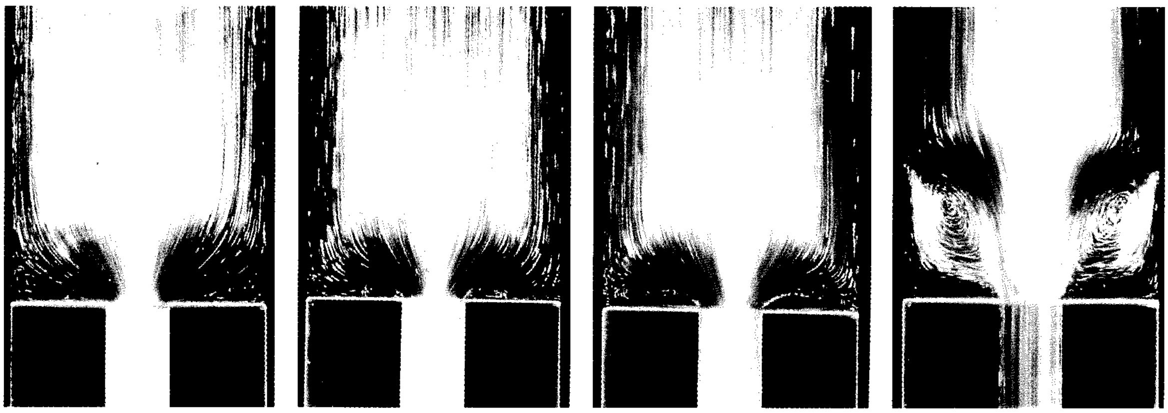
Comparable flow visualization experiments have been carried out in *planar* contractions, with intriguing similarities but also some provocative differences (see, for example Boger and Walters, 1993; Evans and Walters, 1986 and 1989). Salient corner vortices and lip vortices are again in evidence, vortex enhancement is now uncommon and contaction ratio is an important variable in determining the dominant flow features. However, the dependence on contraction ratio here sometimes shows qualitative differences from that found in flow through axisymmetric contractions and there are also other even more unexpected differences. For example, when constant-viscosity Boger fluids are used as test fluids, the vortices are, if anything *reduced* from the Newtonian case. Available evidence would seem to indicate that some shear thinning is essential if vortex enhancement is to occur in planar contractions. So it is evident that the whole subject of 'flow through a contraction' is immensely rich and provocative and has provided theoreticians with many daunting challenges.

Clearly, there are numerous and provocative experimental observations in tubular and planar entry flow for constant viscosity elastic fluids and where prediction of the



(a) (b) (c)

Fig. 8. Pulsating lip vortex for a Boger fluid (0.1% polyisobutylene in a polybutene and kerosene solution) in a 4 to 1 circular contraction. The flow field changes in a cyclic fashion from a-b-c to a-b-c etc. through a cycle of approximately 25 s. $\dot{\gamma} = 65.4\ s^{-1}$, $R_e = 0.014$, $W_e = 0.278$ (see also Figure 9). (Photograph courtesy of R.J. Binnington, Department of Chemical Engineering, The University of Melbourne, 1989, and reproduced from Boger and Walters, 1993.)



(a) (b) (c) (d)

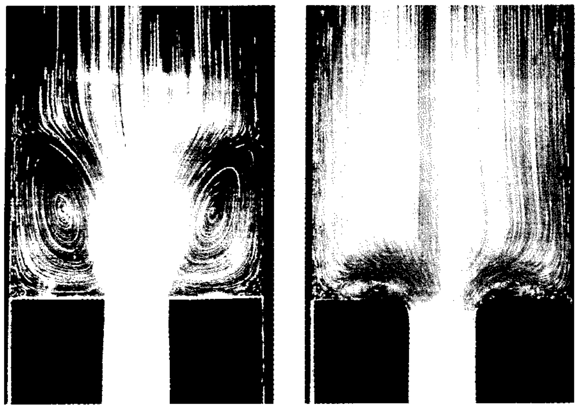
Fig. 9. Streakline photographs illustrating the growth of a lip vortex through the oscillating stage in a 4 to 1 contraction for a Boger fluid. (0.1% polyisobutylene and kerosene solution). (a) $1\ \dot{\gamma} = 1.56$; (b) $1\ \dot{\gamma} = 2.30$; (c) $1\ \dot{\gamma} = 2.40$; (d) $1\ \dot{\gamma} = 2.41$. The product $1\ \dot{\gamma}$ ultimately approaches a constant for this liquid. This limiting behaviour has been reached in (c) and (d), where a large change in the flow is associated with a very small change in $1\ \dot{\gamma}$. (From D.V. Boger, D.U. Hur and R.J. Binnington, *J. Non-Newt. Fluid Mech.*, **20**, 1986, 31, and reproduced from Boger and Walters, 1993.)

phenomena represent a considerable challenge.

2.2. Settling of a Single Sphere

The second example of a distinctively viscoelastic response is provided by the settling problem in the case of polymer solutions. In this, a sphere is released from rest in an expanse of liquid, which is contained in a cylindrical container. The sphere is dropped along the axis of the cylinder and, although there is some interest in the transient build up to steady state, the main concern is the terminal velocity and how this depends on the rheology of the test fluids and also the ratio ($\beta = a/R$), where a is the radius of the sphere and R is the radius of the cylindrical container. Experimental data are available on both shear thinning and constant viscosity elastic liquids, but our main concern here is the behaviour of Boger fluids in the settling experiment.

In the limit of an infinite expanse of test liquid (i.e. $\beta \rightarrow 0$), available experimental data for the drag coefficient relative to the Newtonian value show a rich diversity, with the behaviour depending on the physical chemistry of the dissolved polymer and related issues (see, for example, Solomon and Muller, 1996). While the drag coefficient for a Newtonian fluid for a sphere is only a function of the Reynolds number ($C_d = 24/Re$, where C_d is the drag coefficient of the sphere falling in an infinite medium and Re is the Reynolds number), the drag for a constant viscosity elastic liquid also depends on the Weissenberg



(a) (b)

Fig. 10. Streamlines for a Boger fluid (0.03% Separan MG500 in water and corn syrup) in a 4 to 1 contraction illustrating the sensitivity of the flow field to the geometry of the re-entrant corner. Each photograph is taken at the same conditions: $\dot{\gamma} = 8.3\ s^{-1}$, $R_e = 2.9 \times 10^{-3}$, $W_e = 0.0169$. (a) Abrupt re-entrant corner. (b) A re-entrant corner with a 2 mm radius into a 5.5 mm diameter downstream tube. A comparison between (a) and (b) graphically illustrates the importance of the shape of the re-entrant corner on the resultant flow pattern for an elastic liquid. (Photograph by R.J. Binnington, Department of Chemical Engineering, The University of Melbourne, 1991, and reproduced from Boger and Walters, 1993.)

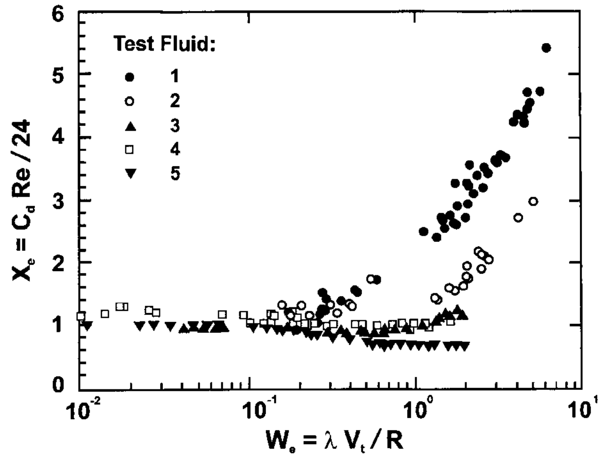


Fig. 11. The effect of Weissenberg number on the non-dimensional drag coefficient for five constant viscosity elastic liquids. (Reproduced from Boger and Solomon, 1996.)

number (W_e) of the flow. The Weissenberg number measures the relative strength of elasticity in the flow. In the flow past a sphere, $W_e = \lambda(V_t)/a$ where λ is the material relaxation time, V_t is the terminal velocity and a is the sphere radius. The effect of W_e on flow past a sphere in a constant viscosity elastic liquid can be examined by plotting the non-dimensional drag coefficient, X_e versus W_e. X_e is the ratio of the measured drag to that of a Newtonian fluid of the same viscosity. Fig. 11 shows X_e as a function of W_e for five different constant viscosity elastic liquids; the data are taken from the literature. Fluids 1, 2 and 4 are from Solomon and Muller (1996), fluid 3 is from Tirtaatmadja *et al.*(1990), and fluid 5 is from Chhabra et al. (1982). Measurements on constant viscosity elastic liquids similar to fluids 3 and 5 have been reported by Chiemlewski *et al.* (1990).

The diverse effects of elasticity on flow past a sphere are observed in Fig. 11. If the elasticity has no effect on the measured drag for a fluid then $X_e = 1$, independent of W_e. In fact, in Fig. 11 for $W_e < 1$, very little deviation from Newtonian behaviour is observed for any of the test fluids. However, at higher W_e the Boger fluids show drag reduction or drag enhancement relative to Newtonian fluids. What mechanism would cause these fluids, all of a constant viscosity, to exhibit divergent drag behaviour? The magnitude in effect is striking, with the fluid behaviour spanning from 500 percent enhancement to 30 percent drag reduction. Note that 500 percent drag enhancement corresponds to a factor of five reduction in the velocity of the sphere falling in a test fluid relative to a Newtonian fluid of the same viscosity.

The case $\beta = 0.5$ was chosen by *Computational* Rheologists as one of their benchmark problems. There is now general agreement that, with the benefit of hindsight, this was not the most judicious of choices, for reasons that we shall not elucidate. However, the problem has nevertheless provided workers in CFD with a convenient basis for comparison of the various computer codes. So far as experimental data are concerned, the evidence that is available for $\beta = 0.5$ (Walters and Tanner, 1991; Oh *et al.*, 1992; Jones *et al.*, 1994; Degard and Walters, 1995) would suggest that, for Boger fluids, there is very little change in drag with increases in the relevant dimensionless number. To be specific, if we define a suitable drag coefficient and Weissenberg number as above, then with increasing W_e, (which for a given test liquid can be accomplished by dropping heavier and heavier spheres of the same radius), the drag coefficient does not vary significantly from the Newtonian zero-Reynolds number value. This state of affairs is in marked contrast to that for b□*0*.

The relatively simple settling problem has therefore provided research workers in the field with a very provocative series of problems and one is probably justified in this case of referring to an experimental dilemma.

2.3. Extrudate Swell

The third and final problem we address concerns the well-known problem of extrudate swell. Although industrial extrusion problems can have a complex set of geometrical and flow conditions, the one favoured in esoteric studies is much simpler, with the test liquid exiting a long capillary tube (or slit). The relevant swell ratio, S_w, is defined by *100(D - a)/a percent,* where D is the radius of the extruded liquid and a is the radius of the capillary. In the case of Newtonian liquids, it is well-known that S_w can vary from approximately *+13%* for vanishingly small Reynolds numbers to approximately *-13%* for high Reynolds numbers. In the case of elastic liquids, the phenomenon of extrudate swell occurs with values of S_w in excess of 2 common place. Fig. 12 illustrates the phenomena for two fluids of identical and constant viscosity. Here the basic problem is well defined and the experiment is relatively easy to perform. However, as we shall see, industrial die swell problems are not so well defined, with large-scale entrance and memory effects; these add significantly to the complications and therefore the challenges of extrudate swell.

3. Rheometry

Modern advances in instrument and transducer technology have meant that many research groups have access to sophisticated rheometers that can readily provide a wide range of rheometrical information. A determination of the viscosity/shear rate response *over the relevant shear rate range* is of course sacrosanct and little meaningful progress can be anticipated unless there is a corresponding attempt to measure the first normal stress difference N_1 over the same shear rate range. This is a more difficult pursuit, especially if the shear rates of relevance are high, but it is nevertheless an important one.

For more than two decades, a study of the *second* normal stress difference N_2 was deemed unnecessary, even though numerous techniques for carrying this out were introduced in the late 60s. The decision was a pragmatic one and was made for numerous reasons. First, the measurement of N_2 was, and still is, very much more difficult than the measurement of N_1, so much so that it was concluded that if N_2 was that difficult to measure it could not be that important. There is clearly some truth in this position, especially since the data that were obtained on polymeric systems seemed to indicate that, in relative terms, N_2 was small. As a rule of thumb, $N_2 < 0$, $|N_2| < 0.1\ N_1$ was regarded as being appropriate for a number of test liquids, including some that had been the subject of detailed experiments in many laboratories. So, except in isolated flow situations like flow in a pipe of non circular cross section and various instability flows (e.g. that associated with the Couette Taylor-vortex instability), N_2 was regarded as unimportant and expendable. So much so, that constitutive equations for the liquids were and often still are constructed on the basis that $N_2 = 0$.

However, there has been a renewed interest in N_2 in recent years, and, in the 90s, evidence has begun to emerge that at least for some shear thinning polymer solutions and polymer melts, N_2 may not be as small as once thought. Indeed, values of N_2 three or more times larger than traditional wisdom would suggest have been found by respectable research groups carrying out careful experiments (Magda and Baek, 1994; see also Tanner and Walters, 1998, p. 139).

This must surely generate a rethink on the part of constitutive modellers, and at the very least, we might anticipate a renewed interest in the importance of N_2 in the task of solving theoretically unresolved experimental dilemmas.

Interestingly, current experimental evidence for constant viscosity Boger fluids is not inconsistent with the low estimates of N_2 mentioned earlier, i.e. $|N_2| < 0.1\ N_1$.

Small amplitude oscillatory shear measurements have a long history, but they have taken on renewed vigour in recent years for more than one reason. Certainly, modern rheometers now make the determination of the storage modulus G' and loss modulus G' a routine procedure and, not surprisingly, research groups are making use of this improvement in rheometer design and operation. But these improvements have also coincided with the growth of interest in Computational Rheology and the realisation that any preoccupation with *single* relaxation time constitutive models is now misplaced. So, more and more computer codes are being written for *multimode* differential models and for *integral* constitutive models which require a full relaxation spectrum (either continuous or discrete).

Therefore, small amplitude oscillatory shear measurements are viewed by many as indispensable tools in the task of constructing constitute models. The problem has now inevitably shifted to the problem of calculating the

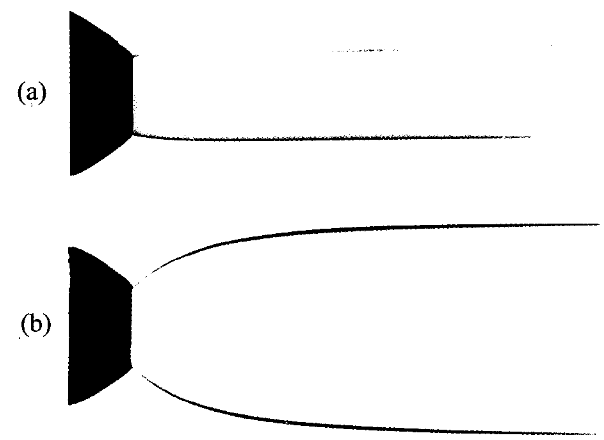


Fig. 12. Die swell for liquid extruded into a neutrally-buoyant medium constructed from a low viscosity silicone oil and carbon tetrachloride solution of matching density to the extruded flow medium. (a) Newtonian liquid of viscosity 11.6 Pa s being extruded ($R_e = 0.001$). (b) Boger fluid of viscosity 11.4 Pa s being extruded ($R_e = 0.0009$, $W_e = 0.272$). (Photographs by R.J. Binnington, Department of Chemical Engineering, Monash University, 1981, and reproduced from Boger and Walters, 1993.)

relaxation spectrum from the experimental dynamic data. The mathematical problem is known to be ill posed, but this hasn't prevented the emergence of a lively and competitive industry in computer packages for the purpose.

Another rheometrical function of increasing importance is the uniaxial extensional viscosity η_E. Its measurement is fraught with difficulties. In the case of polymer melts, the problem is one of maintaining the extensional deformation long enough for a steady state to be reached. In the case of polymer solutions, the problems are potentially more acute and revolve around the difficulty of generating experimentally a constant strain rate extensional flow. This hasnt prevented the development of a number of techniques, including variants of the basic spinning technique, stagnation flow devices, and flow through a contraction. Recent collaborative ventures involving round-robin tests on a number of test liquids such as M1, A1, S1, have highlighted the basic futility of the exercise. There has been little or no agreement between the various sets of data and, apart from some isolated recent experiments which have carefully adopted the spinning technique (see, for example, Orr and Sridhar (1999) and Spiegelberger and McKinley (1996)), no-one is now claiming that their technique provides unambiguous measurements of the steady-state extensional viscosity η_E.

However, many of the available techniques are naturally dominated by an extensional deformation component and can thereby fulfil a useful quality-control function. Of much more importance in the present context is the utility of these devices in a critical-experiment mode. So, for example, a constitutive model is constructed on the basis of steady-shear and oscillatory-shear data and this model is them solved taking full cognisance of the imperfect kinematics of the various techniques. The resulting predictions are next compared to the experimental results, and the agreement or otherwise between the predictions and the experimental data from the extensional devices is a test of the utility of the constitutive model.

This procedure has revealed a potentially depressing situation. Certainly, the various extensional rheometers can provide various functions, which we may very loosely call extensional viscosities, but their true utility must now be seen in their capacity of crucial experiments. It is obviously useful to test any constitutive model, which is based on shear, in a *relatively* simple flow which has a strong extensional component, before attempting to use it to resolve and explain experimental dilemmas, which are invariably associated with even more complex flows.

It would be wrong to view Rheometry as being important only within the context of constitutive modelling, but there is no doubt that, as process modelling becomes ever more popular, one of Rheometrys main applications will be in the construction of mathematical models for rheologically complex fluids.

4. Constitutive Modelling

It is fairly obvious that any theoretical resolution of experimental dilemmas must rely on the availability of a suitable constitutive model for the test liquids. Since the flows are *complex*, there is no possibility of being general in this respect and some approximation must be tolerated. The question then is this: is it possible to construct constitutive equations which are simple enough to permit numerical solutions for the complex flows of interest and yet general enough to have the required predictive capacity? The answer to this important question must depend in part on the test liquid under study, but for highly-elastic polymeric liquids, the construction of suitable constitutive equations is far from being a routine operation and there is no guarantee of success, even given the growing strength of present day rheological research.

The constitutive equations we are seeking must satisfy certain constraints which are shown schematically in Fig. 13. So, the equations have to satisfy certain formulation principles. These have been known for nearly half a century and are not controversial.

The form of the equations can be guided by a knowledge of the microstructure of the liquids and microrheology can have an important input into the formulation procedure. Indeed, some would argue that the way forward is to bypass the constitutive model step altogether and to proceed directly from a knowledge of the microstructure to flow simulation. We anticipate that much of the rheological research which will be carried out in the next five to ten years will be expended in this area.

As we have already intimated, available rheometrical data provide an indispensable input, and it is clearly essential that the rheometrical functions for the chosen constitutive model should match those obtained experimentally.

The final input into the process of constitutive modelling

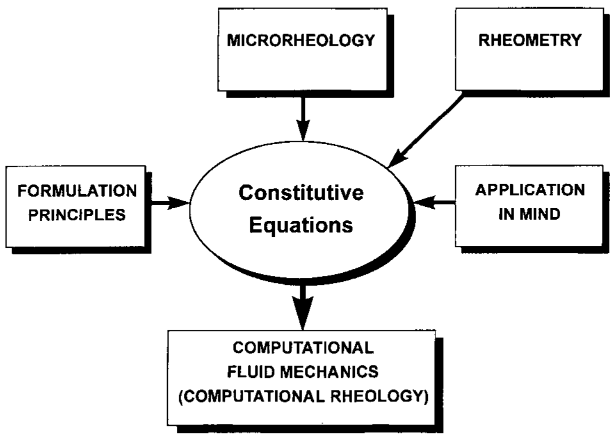


Fig. 13. Essential elements in the construction of a Constitutive Equation.

is the application in mind. In other words, we must have a horses for courses attitude to make use of any simplifying flow characteristic. Just one example will suffice to illustrate the point.

Any spinning flow will clearly involve an important *extensional* flow component and it is important in this case to be precise about the extensional viscosity prediction of the model, with *if necessary* a less rigorous attitude to some of the other rheometrical predictions of the model. This condition is not one which rheologists defend with any conviction or enthusiasm, and its very mention is an indication of the inherent difficulties in constructing constitutive models that have both utility and relative simplicity.

A cursory glance of the literature would soon highlight the plethora of constitutive possibilities and it is true to say that the popularity of many of these has been ephemeral. However, a fairly recent influential Workshop on Constitutive Equations led to a measure of agreement between the international experts present and a report of the Workshop has been published (Pearson, 1994).

So far as polymer melts are concerned, various forms of the so-called KBKZ model were favoured. These required damping functions to generate realistic rheometrical data and the forms suggested by Wagner (1978) and Papanastasiou *et al.* (1983) seem to be those in current favour. Seven or more relaxation times are considered necessary and it is here that the current preoccupation with the inversion of dynamic data to yield a discrete relaxation spectrum is seen to have an outlet and a motivation.

So far as dilute polymer solutions are concerned, variants of the FENE dumbbell models are currently in vogue and the so-called Chilcott-Rallison model has had an extensive following. However, the Workshop made it abundantly clear that any preoccupation with just *one* relaxation time was no longer tolerable, especially when attempts are made to predict quantitatively (or even semi-quantitatively) experimental data on complex flows.

It is evident that the important but complicated task of deciding on constitutive models for polymeric fluids has now reached a level of consensus. The atmosphere is not one of excessive optimism and the need for some compromise between tractability and generality is conceded, if grudgingly.

5. Computational Rheology

Computational Rheology is a relatively new field which came into prominence in the 70s as a result of two main factors. The first concerns the acknowledgement that in simulating complex flows of highly elastic liquids, analytic solutions are generally out of the question and that numerical methods are indispensable. The second refers to the growing availability of computer power in that period, which made numerical simulation a realistic possibility.

The basic wherewithal for solving problems for both differential and integral constitutive equations was soon in place and the frustrating delays caused by the ubiquitous high Weissenberg number problem were slowly resolved. A popular introduction to the subject entitled Computational Rheology: A new science has been written by Crochet and Walters (1993), and this paper highlights the significant advances that have been made in recent years. Numerous numerical techniques have been employed, most notably finite-elements, but the finite-difference and spectral methods have also their adherents. The finite-volume method is favoured by some.

It is probably true to say that the level of sophistication in the constitutive model now presents few fundamental problems and the field is expanding as fast as the increase in computer power will allow. An excellent recent review is that of R. Keunings (to be published in *Computational Fluid Dynamics Journal*, 2000).

6. Comparison of Numerical Simulations with Experimental Results

We are now in a position to refer the reader to the important comparison between simulation and experiment. In the case of contraction flow, there has been little difficulty in recent years in predicting vortex enhancement like that shown in Fig. 3 in a qualitative sense, but *quantitative* agreement between theory and experiment has been and still is elusive.

The ubiquitous disagreement between the contraction flow experiments of Boger and the innovative numerical simulations of Crochet was a well-known talking point at international conferences for ten years and has now entered scientific folklore. The initial breakthrough has also been well documented (Boger *et al.*, 1992). This involved the simple observation that the initial massive disagreement was due to nothing more sophisticated than different definitions of the appropriate Weissenberg numbers! With this in place, there was no difficulty in obtaining at least *semi* quantitative agreement between theory and experiment for vortex enhancement, but quantitative agreement remains elusive. This particular problem has important geometrical singularities at the re-entrant corner (see Fig. 10), and, although there have been noteworthy attempts to resolve the corner singularity problem, the implementation of these analyses has yet to be put in place.

While Fig. 3 shows that viscoelasticity does indeed generate re-circulating regions and that the qualitative nature of these re-circulating regions can be predicted, many outstanding problems remain to be solved. There is much experimental evidence that observed vortices are generated from the lip of the abrupt contraction and not from the enhancement of the Newtonian corner vortex. This phenomenon is graphically illustrated in Figs. 5, 6 and 7; few numerical results have yet been able to demonstrate this effect. It is also true that the maximum size of vortices calculated at the present time is much smaller than that of observations; the calculations seem to show saturation of the vortex size, which is not observed experimentally. In fact, some experiments (McKinley *et al.*, 1991) reveal an apparent bifurcation from steady state flow to a time dependent rotating flow beyond some value of the Deborah number. This phenomenon is illustrated in Fig. 8 and was beautifully quantified in the McKinley *et al.* (1991) paper. Despite recent efforts in this direction, bifurcations have not been detected in numerical simulations.

Lack of success in simulating these complex patterns observed in circular contraction flows is presently attributable to at least three possible causes: a misunderstanding of the behaviour of the fluid in the re-entrant corner, as is illustrated in Fig. 10; the inability of the constitutive equations to capture the behaviour of real fluids in strong complex flows; and the possible inadequacies of numerical codes. It is likely that three-dimensional time-dependent codes will be needed to tackle what would appear at first sight to be a steady axisymmetric problem.

Contraction flows clearly demonstrate that computational rheology is still not able to answer all the questions arising in non-Newtonian flow in complex geometries, but nevertheless it must be seen as an indispensable tool in the pursuit of basic understanding of the flow structure.

The entry flow problem has turned out to be far more complex than ever anticipated, yet the impact the problem has had in linking simulation to numerical prediction has been immense.

So here we have a case of slow progress, but significant challenges remain. The benchmark settling problem is in better shape and provides an excellent illustration of the successes and frustrations of the current situation. It is in fact an ideal vehicle to discuss the basic theme of this paper.

The availability of a benchmark problem was indispensable to progress in this area. This concerned the $\beta = 0.5$ situation for the upper Convected Maxwell (UCM) model, which was initially thought (somewhat naively perhaps) to be a useful first approximation for the Boger fluids used in the experimental work.

Walters and Tanner (1991) reviewed the state of the science in the early 90s and the result was both enlightening and depressing. Fig. 14 provides the simulations obtained by several respected research groups for the UCM model and $\beta = 0.5$. At that time, it would have been ludicrous to seek better agreement between theory and experiment through a more judicious choice of constitutive model. The problem was placed firmly at the door of the numerical analysts and their methods, who were fortunately up to the task. The basic difficulty was highlighted as the resolution of the thin stress boundary layers that were found to occur in this problem, particularly of course at high Weissenberg number. When these were accommodated, mainly through convenient mesh refinement, very commendable agreement was obtained up to a W_e *of 1.6*. The programs failed to converge at this point and $W_e = 1.6$ was seen as some kind of barrier and it was even conjectured that this barrier reflected a subtle change in the flow regime. However, as it became possible to refine the appropriate meshes yet further, it was soon conceded that $W_e = 1.6$ was no barrier at all and the appropriate W_e range was extended - to *3.0* in some cases. Fig. 15 contains a more recent set of simulations and readers may be excused a feeling of déjà vu! Undoubtedly the new lack of agreement is related to inappropriate mesh refinement and related problems in some of the simulations and there is every likelihood that within a short period of time the agreement between $W_e = 1.6$ and $W_e = 3.0$ will be as good as that shown in Fig. 15 for lower W_e values.

The important issue now is not the small disagreement between the different numerical schemes; it is rather the clear disparity between the sizeable drag reduction predicted by all the simulations and the experimental data, which we have indicated shows little change in the drag coefficient with Weissenberg number for some

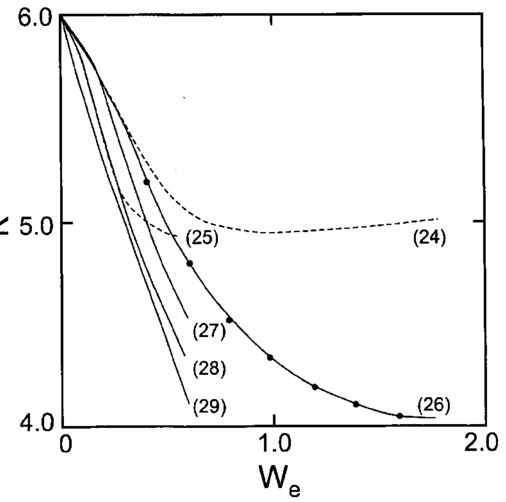


Fig. 14. Early viscoelastic drag force predictions using an Upper Convected Maxwell fluid for b = 0.5. The drag force in the limit of low Reynolds number is given by $F = 6\ K\ (\beta,\ W_e)\ p\ h\ a$. For a Newtonian fluid with b = 0 and $W_e = 0$, $K = 1$ and for b = 0.5 and $W_e = 0$, $K = 6$. A: Crochet (1988); B: Carew and Townsend (1988); C: Lunsmann *et al.* (1993); D: Zheng *et al.* (1990); E: Hassager and Bisgaard (1983); E: Suging and Tanner (1986).

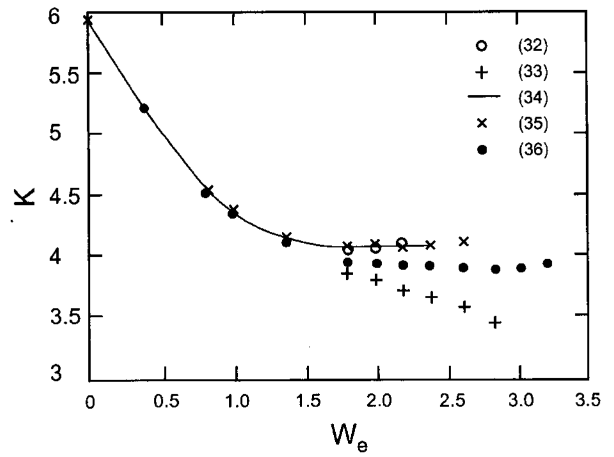


Fig. 15. Recent viscoelastic drag force predictions using an Upper Convected Maxwell fluid for b = 0.5. Only those results have been included that exceed a Weissenberg number of 2 and are obtained on the finest mesh reported. A: Jin *et al.* (1991); B: Luo (1996); C: Warechet (1997); D: Baaijens *et al.* (1997); E: Sun *et al.* (1996). (Reproduced from Baaijens, F.P.T., 1998.)

Boger fluids at least.

Since the basic accuracy of the numerical codes is no longer in doubt, the reason for the discrepancy is self evident -the UCM model is too simple for the Boger fluids used in the experiments and the settling problem can now be viewed as a convenient critical experiment to assess the utility of constitutive models for the Boger fluids. (see, for example, Satrape and Crochet, 1994).

The critical problem in regard to settling is the $\beta = 0$ case where the magnitude of the drag enhancement and reduction was striking, as was illustrated in Fig. 11. Since all of the researchers whose data are shown in Fig. 11 have taken into account all effects for the measured drag, the differences observed are significant. These differences suggest that more than one dimensionless group is required to determine the drag beha-viour for constant viscosity elastic liquids, a conclusion which perhaps is not surprising since the next most complicated constitutive equation beyond the upper Convected Maxwell model is the Oldroyd-B model which contains both a relaxation and retardation time; this would lead to two dimensionless groups, in contrast to the one for the UCM. The challenge now for creeping flow around a sphere is in fact to deal with the differences observed in Fig. 11, which is the subject of current research in non-Newtonian fluid mechanics.

We now pass on finally to the extrudate swell problem. It has long been possible to predict extrudate swell in a qualitative sense and the current state of the science concerning accurate prediction is best illustrated by referring to a specific example highlighted by Crochet and Walters (1993).

The experiments concern a high density polyethylene melt studies by Koopmans (1992). He used the experimental layout shown in Figure 16 In the circumstances of interest here, Koopmans obtained a die swell of about 180 percent. Fig. 17 contains numerical predictions obtained by Goublomme *et al.* (1992) using a powerful finite element code and a reasonable description of the rheometrical behaviour of the melt.

The top picture (Fig. 17a) shows the form of the free surface for the Wagner model, obtained on the assumption that the capillary tube is of infinite length. The computed swell ratio in this case was 144 percent, which is well below that found experimentally. When the same Wagner model was applied to the geometry shown in Fig. 17b, an extravagant die swell of the order of 800 percent was obtained, which reflects the dominance of the elastic character of the model under the conditions pertaining in the experiments. Clearly this prediction was unacceptable.

When Goublomme *et al.* (1992) used an irreversible version of the Wagner model and also introduced a *second* normal stress facility, they predicted a die swell of 186 percent, which was reasonable close to the experimental value. So, we have here, once again, an example of the application of the Scientific Method, with the comparison of the numerical simulations and the experimental data providing useful, indeed indispensable, information about the level of sophistication required in the chosen constitutive model.

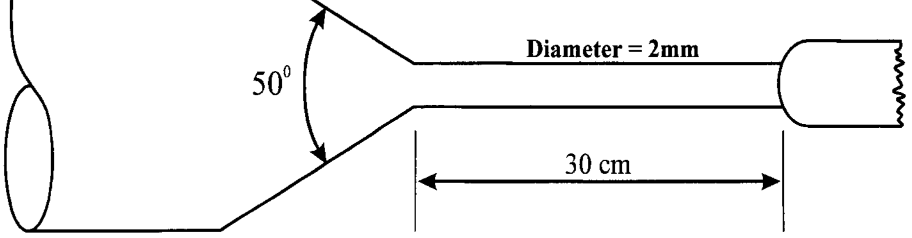


Fig. 16. The Koopmans experimental layout for die swell measurements on high density polyethylene (Koopmans, 1992).

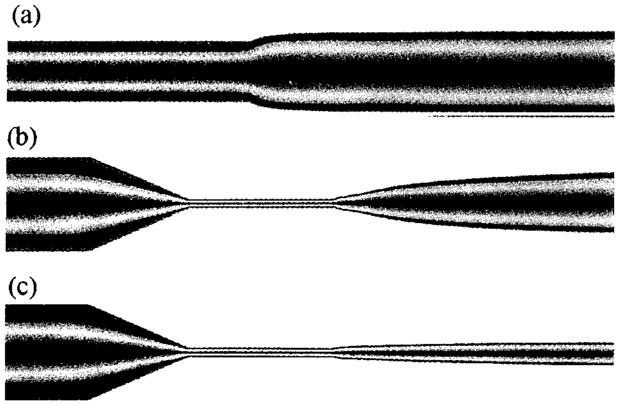


Fig. 17. Simulation of the swelling of a high density polyethylene with a Wagner model, (a) out of a long capillary tube, and (b) out of a die with a conical entry section. The results are very similar to experimental data when one uses a modified rheological model (c). (Reproduced from Crochet and Walters, 1993.)

7. Conclusion

The paper has highlighted some provocative experimental phenomena in the flow of highly elastic polymeric liquids in complex geometries. We have argued that many of them are now accessible to modern numerical simulation. Computational Rheology has come of age and it is to be viewed as an essential part of the basic Scientific Method, which is now being used with some success to resolve any remaining experimental dilemmas. At the present time, any outstanding problems are usually due to an inappropriate or oversimplified choice of constitutive model.

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