Model polymer fluid systems

David V. Boger

Department of Chemical Engineering, The University of Melbourne, Parkville, Victoria, 3052, Australia

Abstract - The development of non-shear thinning elastic liquids and their impact in bridging the gap between predictions from continuum theory and practical reality in problems of interest in polymer processing is described. It is shown that the highly elastic, constant viscosity fluids represent a class of fluid behaviour consistent with molecular theory, which can be described at low shear rates by a constitutive equation developed by Oldroyd in 1950. Experimental observation with these materials has allowed elastic effects to be observed in flow fields for the first time in the absence of any effects due to a varying viscosity, and has also allowed direct comparison to be made with predictions from continuum theory in these flows. In squeeze film flows good agreement between observation and prediction is obtained. In exit flows (extrudate swell) significant elastic effects are observed and predicted but the agreement between observation and numerical simulation is poor at high levels of elasticity. In tubular entry flows (die entry flows) large vortex enhancement due to elasticity is observed which is not predicted by the appropriate continuum analysis. It is not clear why continuum theory is adequate in some flow and inadequate in others. Nevertheless great strides have been made in the solution of flow problems of importance in polymer processing.

INTRODUCTION

For those contemplating working on the experimental investigation and quantitative observation of the kinematics of viscoelastic fluid flows of relevance in polymer processing, difficulties arise at the outset in the choice of appropriate test fluids. Since a major practical and economic interest in these flows involves molten polymers, it would seem logical and appropriate to choose a molten polymer for the investigation. Such is not the general case. In addition to the difficulties of generating an isothermal flow at elevated temperatures and the lack of optical clarity of some polymers for visual study, even the simplest elastic property measurements, like the first normal stress difference, can rarely be obtained at shear rates exceeding $10 \, \text{s}^{-1}$ - a level usually too low to be of interest in the practical processing situation. Hence with a few exceptions, one being the work of White and Kondo on entry flows (ref. 1), most workers concerned with observing the kinematics of polymer melt flows have not attempted or have not been able to accompany their observations with any fundamental elastic property measurement at appropriate shear rates. By necessity then less understood empirical indications of fluid elasticity such as the end correction in a capillary rheometer (ref. 2), the exit pressure (ref. 3), and die swell (ref. 4) have been used. Although much has been learned in regard to the behaviour of specific polymers in certain complex flows of practical interest, little of this information has been helpful in developing and using constitutive equations for the solution of fluid mechanics problems for viscoelastic fluids in polymer processing flows. This is in contrast to inelastic Newtonian fluids where the Newtonian fluid constitutive equation with the equations of motion is sufficient to solve (in principle) any Newtonian fluid mechanics problem.

Therefore, many investigations, concerned with the more general problem of bridging the gap between experiment and theory and developing a fluid mechanics for viscoelastic fluids, have turned their attention to an examination of the behaviour of viscoelastic polymer solutions in flow fields of interest. These solutions can be used at room temperature and it is possible and relatively simple to measure at least the viscosity and first normal stress difference at shear rates ranging up to $1000~{\rm s}^{-1}$. Hence a minimal, although perhaps inadequate, characterization of these materials can be obtained. Polyacrylamide in water solutions have been the most popular choice. They are optically clear in solution at room temperature, are significantly elastic in so far as the ratio of the first normal stress difference to the shear stress is in the range expected for many molten polymers (ref. 1 and 5), and they are highly shear thinning (ref. 6) like most polymer melts. In addition there is evidence to suggest that they exhibit strain thickening in extension (ref. 7) which is typical of low

density polyethylenes and polystyrenes (ref. 8). However, their shear viscosities can be orders of magnitude lower than typical molten polymers as are their relaxation times calculated in the basis of a Maxwell constitutive equation (Eq. 1). The earlier experimental observations for solutions have therefore been made for shear thinning, low viscosity elastic liquids under conditions where fluid inertia (a variable not present in polymer processing flows) may be introduced in addition to the shear thinning and elastic characteristics of the fluid.

With the revolution in computation and our ability now to solve numerically very complex fluid mechanics problems, a considerable gap has developed between those who use constitutive equations for viscoelastic fluids with the general flow equations in an attempt to solve practical polymer processing problems and those who are attempting to experimentally verify the predictions from these very sophisticated numerical solutions. The major reason for the gap is that most of the numerical solutions examine elastic effects in the absence of shear thinning and fluid inertia, while the experimental observations are made for real fluids that are not only elastic but also shear thinning under conditions for polymer solution flows where fluid inertia may also be an important variable (ref. 9). In recent years however highly elastic non-shear thinning fluids (the so called Boger fluids (ref. 10-12)) have been developed and used by a number of researchers to study the effect of elasticity in flow fields in the absence of any significant shear thinning and fluid inertia (ref. 13-27). Much of this work has been motivated by a need to verify the results being generated from numerical solutions for polymer processing flow fields (ref. 28). Commercial software packages for polymer processing flow fields, like the Polyflow package developed by Professor M. Crochet in Belgium, are now available and used by many of the major chemical companies.

Other workers (ref. 29-33) are now concerned with gaining a better understanding of the flow properties of these fluid systems. The works of Gupta et al (ref. 30,31) and our own (ref. 29,34-36) have clearly shown that non-shear thinning elastic fluids represent a class of fluid behaviour entirely consistent with molecular and continuum theory and that such ideal elastic fluid systems can be constructed by dissolving a number of different high molecular weight polymers in a number of different viscous Newtonian fluid solvents. Possibly the most interesting fluid to emerge from these works is the organic Boger fluid composed of a high molecular weight polyisobutylene in a Newtonian solvent of kerosene and polybutene. The features of this fluid will be discussed in some detail. Constant viscosity elastic fluid behaviour is not confined to dilute solutions of polymers in a viscous Newtonian solvent, but has also been observed in low molecular weight polymethysiloxanes (ref. 37), in polycarbonates (ref. 38) and recently in suspensions of glass fibres in viscous Newtonian fluids (ref. 39).

The purposes of this paper are similar to those in References 29 and 34:

- (i) to reinforce the conclusion that a general class of real materials exist which exhibit both high elasticity and non-shear thinning characteristics,
- (ii) to demonstrate once again that such behaviour is consistent with dilute solution theory and that this class of fluid behaviour does not violate simple fluid theory,
- (iii) to clearly show that a three constant Oldroyd B constitutive equation is more appropriate to model the behaviour of this class of material than the convected Maxwell model, but to stress that the Oldroyd B equation is only applicable at low shear rates for these dilute solutions.
 - (iv) to demonstrate that direct comparison between theoretical prediction and experimental observation is now possible in a Weissenberg number region where significant elastic effects as observed in a non-viscometric flow,

and finally

(v) to stress that understanding this class of fluid behaviour is essential for future prediction of the behaviour of more complex polymers in processing flows.

NON-SHEAR THINNING ELASTIC LIQUIDS

Figure 1 shows dynamic and steady shear properties typical of the non-shear thinning elastic fluids constructed from polyacrylamide dissolved in corn syrup (ref. 10,34). The fluid (0.02% Separan AP 30, 4% water, 95.98% corn syrup) was prepared in our laboratory where shear stress, τ_{12} , and first normal stress difference, N_1 , measurements were made with an R-19 Weissenberg Rheogoniometer. The shear stress, τ_{12} , and the storage modulus, G', and the loss modulus, G'', were also measured in the laboratories of Professor Böhme and Professor Spurk at the Hochschule der Brundeswehr Hamburg and the University of Darmstadt respectively. Note that the shear stress data from the two laboratories agree and that the viscosity is essentially constant with shear rate. Also note that at shear rates less than 8 s^-1 the

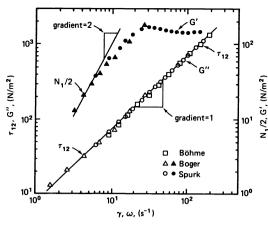


Fig. 1. Dynamic and steady shear property data illustrating that constant viscosity elastic liquids do not violate simple fluid theory.

first normal stress difference varies with the square of the shear rate. The identities of $\tau_{12}=G''$ and $G'=\frac{1}{2}N_1$ (for $\dot{\gamma}<8s^{-1}$) confirm the expectations from simple fluid theory (ref. 34) and indeed there is nothing unique about the low shear rate, low frequency behaviour of these fluid systems.

It is now quite clear that non-shear thinning and highly elastic fluids can be constructed by dissolving a high molecular weight polymer in a very viscous Newtonian solvent and that this ideal behaviour is not unique to the polyacrylamide corn syrup system where the behaviour was first observed (ref. 10,11,13). Such behaviour has also been observed for polyisobutylenes dissolved in polybutenes (ref. 29,30) and adequately explained within the framework of dilute solution theory (ref. 30), i.e., with the elastic dumbbell model.

Typical steady and dynamic shear properties for a polyisobutylene-polybutene fluid are shown in Figs. 2 and 3 (ref. 35). For this fluid the steady shear viscosity of the solution, n, and of the solvent, n_S, were both measured as was the first normal stress difference N₁, with an R-19 Weissenberg Rheogoniometer. The dynamic viscosity, n', and the storage modulus, G', were measured at the same temperature as the steady shear properties with a Mechanical Spectrometer in the Department of Chemical Engineering at the University of Minnesota. The fluid was constructed from 0.2% Vistanex MML-120 (Exxon Chemicals, M.W. \simeq 1.2 x 10^6) in 96% Hyvis 30 (BP Chemicals, M.W. \simeq 1300). 3.8% kerosene was used to dissolve the polyisobutylene before adding the polybutene. Also shown in Figs. 2 and 3 are the dynamic viscosity and storage modulus predicted from the steady shear property measurement using the Maxwell and Oldroyd B constitutive equations. λ and λ_1 are the Maxwell and Oldroyd B relaxation times,

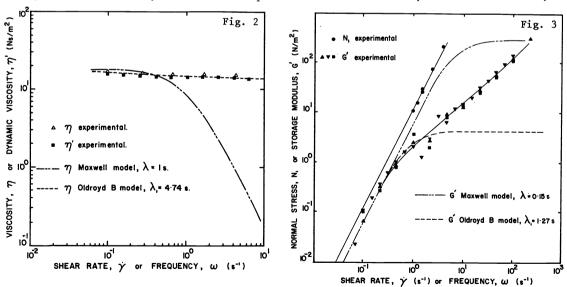


Fig. 2. Steady shear and dynamic viscosity for a polyisobutylene-polybutene fluid (0.2% Vistanex MML-120, 3.8% kerosene and 96% Hyvis 30) - comparison between experiment and prediction with Maxwell and Oldroyd B constitutive equations.

Fig. 3. First normal stress difference and storage modulus for the polyisobutylene-polybutene fluid as in Fig. 2 with similar comparison between experiment and constitutive equation prediction.

defined from the steady shear properties by

$$\lambda = \frac{N_1}{2\hat{\mathbf{y}}^2 \mathbf{p}} \quad , \tag{1}$$

and

$$\lambda_1 = \frac{N_1}{2\dot{\gamma}^2 \eta_p} , \qquad (2)$$

where $\dot{\gamma}$ is the shear rate and η_{D} is the polymer contribution to the viscosity, i.e.,

$$\eta = \eta_{S} + \eta_{p} . \tag{3}$$

Also shown in Figs. 2 and 3 are the dynamic viscosity and storage modulus predicted from the steady shear property measurements for a Maxwell fluid, where

$$G' = \frac{\eta \lambda \omega^2}{1 + \lambda^2 \omega^2}, \qquad (4)$$

and

$$\eta' = \frac{\eta}{1+\lambda^2 \omega'^2} \quad , \tag{5}$$

and for an Oldroyd B fluid, where

$$G' = \frac{\eta(\lambda_1 - \lambda_2)\omega'^2}{1 + \lambda_1^2 \omega'^2} , \qquad (6)$$

and

$$\eta' = \frac{\eta(1+\lambda_1\lambda_2\omega'^2)}{1+\lambda_1^2\omega'^2} . (7)$$

The retardation time, λ_2 , is defined in terms of the solvent and polymer contributions to the viscosity by

$$\lambda_2 = \left(\frac{\eta_s}{\eta_s + \eta_p}\right) \lambda_1 . \tag{8}$$

As has also been shown for the polyacrylamide corn syrup fluids (ref. 29,34), the Maxwell model is only applicable at very low shear rates while the Oldroyd B model does predict the observed deviation in quadratic behaviour in G' at the higher shear rates. However at shear rates greater than 1 s⁻¹ the Oldroyd B model also fails even through N_1 is still quadratic with shear rate at these conditions.

The conclusion that must be reached from the literature in general, from the papers of Gupta et al (ref. 30,31), Jackson et al (ref. 33) and our own work is that constant viscosity elastic fluids represent a real class of fluid behaviour. This behaviour can be explained (qualitatively) by elastic dumbbell theory (ref. 30). Effectively the materials are dilute solutions, constructed by dissolving small amounts of high molecular weight polymer in a very viscous Newtonian solvent. There is no magic recipe and many combinations are possible. In such systems possible shear thinning effects are masked by the presence of the high viscosity solvent. Furthermore, for the first time real materials exist where the measured continuum properties are consistent with both molecular and continuum theory. The constant viscosity elastic fluids are not *true* Oldroyd B fluids, but their steady and dynamic properties can be effectively predicted at low shear rates by this model. There is additional evidence that the second normal stress difference, N₂, may be zero for these fluids (ref. 33,41) which is also consistent with the dumbbell interpretation and the Oldroyd B model. However the available information on extensional behaviour (ref. 19,32) suggests that the Oldroyd B model may be far from adequate to represent the flow when there is a significant extensional contribution.

Dilute solutions of constant viscosity and high elasticity are available. It is now necessary to re-examine both molecular and continuum theories to establish a better constitutive equation for these simple fluids. The major requirement for this investigation is some good extensional viscosity measurements coupled with steady and dynamic shear property measurement on carefully prepared samples. This conclusion will be confirmed in the next section where we compare experimental observation to numerical simulation in flow fields of interest.

COMPARISON BETWEEN NUMERICAL SIMULATION AND EXPERIMENTAL OBSERVATIONS

Worldwide, there is now a great deal of interest in developing software packages for predicting the kinematics of flow fields of interest in polymer processing. In fact, the Fourth International Workshop on Numerical Methods in Non-Newtonian Fluid Mechanics, will be held in Belgium in June, 1985. The objective of these workshops has been to develop appropriate numerical methods for solution of the complex flow problems of interest in polymer processing, and to establish which constitutive equations are suitable to represent the flow behaviour of real polymer fluid systems. Experimental observations with non-shear thinning elastic fluids in flow fields of interest have allowed direct comparison between numerical prediction and experimental observation. Without this comparison little progress could have been made in applying the numerical methods to more complex polymer systems. Some of the results of the numerical and experimental interactions are summarized here.

Squeeze film flows

Possibly the first successful prediction of a significant observed elastic effect in a non-viscometric flow was in a squeeze film flow. The mechanics of a thin film being squeezed between two parallel plates is of considerable interest in that it models the action of a lubricant in a bearing under unsteady load. It is relevant in the interpretation of various plastometers and as Lee et al (ref. 42) point out the flow is of importance in polymer processing operations such as compression moulding:

The results of squeeze film flow measurements for a constant viscosity elastic fluid are presented in Fig. 5 where the dimensionless load

$$\omega = \frac{F}{\pi \eta V R^{4} / 4h_0^3} \tag{9}$$

is shown as a function of dimensionless time

$$\tau = \frac{tV}{h_0} \tag{10}$$

An Instron testing machine was used for the constant velocity, V, squeeze film measurements. A schematic diagram is shown in Fig. 4 where the appropriate symbols are also defined. The experimental technique was confirmed by measuring the load, F, as a function of time, t, for the Newtonian fluid solvent. The results agree well with the Stefan prediction (ref. 43). Futher details, additional data, and a comparison of the solutions of the constant velocity squeeze film flow problem for Maxwell and Oldroyd B fluids can be found elsewhere (ref. 44). As might be expected from the results shown in Figs. 2 and 3, the Maxwell constitutive equation prediction for the squeeze film flow are poor relative to the Oldroyd B predictions (ref. 44).

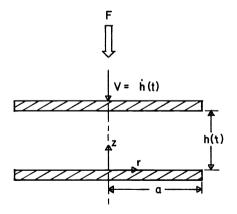


Fig. 4. Schematic diagram of the constant velocity squeeze film flow.

Basically the prediction for an Oldroyd B fluid demonstrates that the load lies between two asymptotic limits: a Newtonian $upper\ limit$ which corresponds to the squeezing flow of a Newtonian fluid of viscosity $\eta = \eta_S + \eta_p$, and a $lower\ limit$ which corresponds to squeezing flow of a Newtonian fluid of viscosity, η_S . The upper limit is reached in an asymptotic process when We \rightarrow 0. The lower one is reached when We \rightarrow ∞ . The Weissenberg number for the squeeze film flow of an Oldroyd B fluid is defined by

We =
$$\frac{\lambda_1 V}{h_0}$$
 (11)

The experimental results shown in Fig. 5 are in agreement with the theoretical prediction. The load is reduced as a result of fluid elasticity, the reduction seems to increase with increasing We as predicted by the theory, and for the range of Weissenberg numbers examined

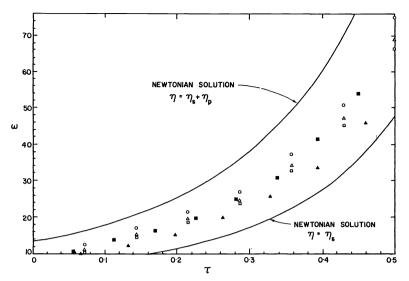


Fig. 5. Experimental observation of load as a function of time for a constant viscosity elastic liquid. Data obtained for We of O = 0.026, Δ = 0.066, ■ = 0.052, □ = 0.132 and ▲ = 0.121.

 $(0.026 \leqslant \text{We} \leqslant 0.132)$ the load does not exceed the lower asymptote predicted by the theory. The experiments were conducted at relatively low Weissenberg numbers, which corresponds to the shear rate region where the Oldroyd B constitutive equation is expected to be applicable for the fluid used in the experiments. For the results shown in Fig. 5 a polyisobutylene-polybutene fluid characterized by: λ_1 = 1.11 s, η = 74 Ns/m² and η_S = 34 Ns/m² was used.

The agreement between numerical prediction and experimental observation for the constant velocity squeeze film flow as observed in our work (ref. 44) and in the work of Lee et al (ref. 42) for a constant load squeeze film flow is encouraging. However as the flow fields became more complex this agreement becomes progressively worse.

Extrudate swell

Many recent papers have been devoted to the finite element calculation of extrudate swell for viscoelastic fluids (ref. 45-51), a problem of considerable importance in polymer processing. These works, generally completed for a Maxwell fluid, share the same convergence failure of the numerical scheme above some critical dimensionless number, $\lambda \mathring{\gamma}_{\omega}$, where λ is the relaxation time and $\mathring{\gamma}_{\omega}$ is the shear rate at the wall of the exit flow configuration. Tuna and Finlayson (ref. 51) reached the maximum value of $\lambda \mathring{\gamma}_{\omega}$ = 1.6 for a convected Maxwell fluid where they predicted a die swell of De/D = 1.386. Crochet and Keunings (ref. 49) were able to extend the value of $\lambda \mathring{\gamma}_{\omega}$ to 4.5 and generate a die swell of the order of 2 by use of a large entry length in the numerical scheme together with a retardation time in the constitutive equation. The addition of the retardation time in the constitutive equation. The addition of the retardation time in the constitutive equation was equivalent to solving the problem for an Oldroyd B fluid. This was the first solution for a complex non-viscometric flow which predicted a significant effect due to fluid elasticity. The die swell predictions of Crochet and Keunings for the Oldroyd B fluid are shown in Fig. 6 in comparison to the Tanner approximate theory (ref. 52), our experimental observations for a non-shear thinning elastic fluid (ref. 20,29), and some die swell observations selected by Crochet and Keunings for two polystyrenes (ref. 53,54).

At first glance one is tempted to conclude that the numerical solution for the Oldroyd B fluid is adequate to predict the observations for polystyrenes, but as Crochet and Keunings point out the polystyrenes are shear thinning elastic fluids which do not behave like Oldroyd B fluids, and that the values of $S_R = N_1/2\tau_\omega$ for a die swell experiment are normally not available from direct property measurement for molten polymers and are usually inferred by indirect methods. Therefore this good agreement may be fortuitous. Comparison of the predictions for the Oldroyd B fluid to the experimental observations for a constant viscosity elastic liquid (polyacrylamide in corn syrup) show a large discrepancy which increases as λ_1 and SR increases. Only at λ \uparrow ω < 1.25 does the data agree with the prediction. Unfortunately, the die swell in this region is little different from the Newtonian value of 1.13. The Crochet and Keunings calculations are for the Oldroyd B fluid with n_D/n_S = 8/9 while the experimental data were obtained for a dilute solution where η_p/η_s was more on the order of 1/9. The exact Oldroyd B parameters for the ideal elastic fluid used in the experiment, unfortunately, are not available. To date there has not been a direct comparison between numerical simulation and experimental observation. This is now possible by comparing die swell observations obtained with well characterized ideal elastic fluids to results now available or which can be generated with available numerical codes.

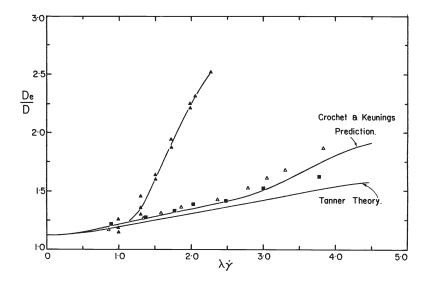


Fig. 6. Extrudate swell for a circular die; experimental data: \triangle -non-shear thinning elastic fluid, polystyrene- \triangle (Vlachopoulos) and - \blacksquare Racin and Bogue.

Creeping flow through an abrupt circular contraction

Creeping flow through an abrupt circular die represents one of the most important flows in polymer processing. It represents the most extreme flow encountered in an extrusion die and represents the flow fields used extensively for the measurement of the flow properties of molten polymers with the capillary rheometer. The circular entry flow is also one of the major test problems chosen for the International Workshops in Non-Newtonian Fluid Mechanics. The first observations of the influence of fluid elasticity in this flow field in the absence of any significant shear thinning or fluid inertia were made by Nguyen (ref. 11,14,19,22) in 1978 using the polyacrylamide corn syrup ideal elastic fluids. At this time it was not realized that the ideal elastic fluids behaved like Oldroyd B fluids and not like Maxwell fluids. Therefore Oldroyd parameters were not available and results were interpreted in terms of Maxwell fluid parameters.

The effect of elasticity on the secondary flow vortex in a 7.68 to 1 circular contraction is illustrated by the streak photographs shown in Fig. 7. The observations in Fig. 7 show that

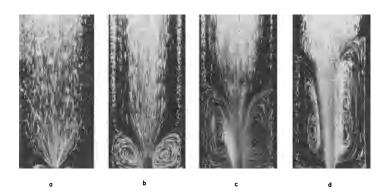


Fig. 7. Entry flow patterns in an abrupt entry circular die for a 0.057% Separan AP 30 in corn syrup, non-shear thinning elastic solution. (a) $\dot{\gamma}_{\omega}=1.8s^{-1}$, Re=1.36x10⁻⁴, We_t=0.112; (b) $\dot{\gamma}_{\omega}=9.5s^{-1}$, Re=7.8x10⁻⁴, We_t=0.218; (c) $\dot{\gamma}_{\omega}=22s^{-1}$, Re=1.9x10⁻³, We_t=0.263; (d) $\dot{\gamma}_{\omega}=40s^{-1}$, Re=3.5x10⁻³, We_t=0.28, where We_t= $^{\lambda V}/_{D}$

the secondary flow vortex increases in size as the Weissenberg number increases until at $\grave{\gamma}_\omega=40s^{-1}$ in Fig. 7d the flow becomes asymmetric. Following the asymmetry the flow becomes unstable and illustrates one of the forms of *melt fracture* as observed in low density polyethylene and polystyrene. For shear rates at the wall of the downstream tube in the quadratic region of behaviour of N_1 , the dimensionless reattachment length of the secondary cell (X = $^Lv/D_u$, where L_v is the reattachment length of the secondary cell and D_u is the upstream tube diameter) is observed to be 0.18 in comparison to the Newtonian value of 0.177. In this quadratic region of normal stress behaviour the cell size varies only slightly with flow rate and is virtually identical to the predicted value for a Newtonian fluid and for a Maxwell fluid in: circular 2 to 1 and 4 to 1 contractions (ref. 55-57). In fact the theoretical results for a Maxwell fluid show that the shape (but not the intensity) of the secondary flow vortex is virtually identical to the observed Newtonian behaviour. No significant vortex growth has been predicted for the Maxwell fluid.

The reattachment length of the secondary flow vortex as a function of Weissenberg number is presented in Fig. 8 (ref. 11,14,22,29) for six non-shear thinning and elastic polyacrylamide in corn syrup solutions, where the viscosity, η_{\star} and Maxwell (low shear rate) relaxation times ranged from 89 < η < 750 poise and 0.1 < λ < 0.9s. The Weissenberg number, We_t, is defined by

$$We_{t} = \frac{\lambda V}{D} \tag{12}$$

where V is the average velocity in the downstream tube of diameter D and λ is the relaxation time determined from the steady shear flow properties at the shear rate at the wall in the downstream tube. Also shown in Fig. 8 is the slight increase in cell size with Weissenberg number which was predicted by Viriyayuthakorn and Caswell (ref. 56) and by Crochet and Bezy (ref. 55), using finite element numerical simulation techniques, for a Maxwell fluid in a circular 4 to 1 contraction.

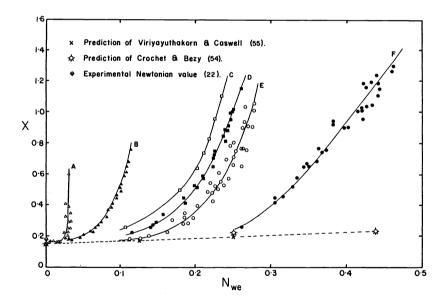


Fig. 8. The reattachment length as a function of Weissenberg number for creeping flow through circular contractions.

Note that the experimental and numerical results are in agreement only at the lower shear rate limit for each ideal fluid experiment. This is to be expected as the Maxwell model can only be expected to apply at extremely low shear rates (see Figs. 2 and 3 and ref. 29). At higher shear rates the upper convected Maxwell model predictions completely fail to predict (even qualitatively) the vortex enhancement observed, i.e. the flow field is hardly influenced by elasticity of the flow. It was hoped that the failure of the Maxwell model was due to the limited domain of elasticity where the calculations were completed. However the use of the Oldroyd B fluid rather than the convected Maxwell model allowed Keunings and Crochet (ref. 57) to reach relatively high values of elasticity where unfortunately no significant vortex enhancement was predicted. The question as to why the Oldroyd B model will predict observed elastic effects in squeeze film and die swell flows, but not in entry flows remains unanswered. This observation for the Oldroyd B fluid in tubular entry flows led Keunings and

Crochet to speculate that it is the constant viscosity and quadratic normal stress behaviour which may be the cause of this lack of deviation from Newtonian behaviour; a conclusion which hardly seems reasonable in light of the evidence available for squeeze film and die swell flows. Keunings and Crochet have predicted significant vortex enhancement in tubular entry flows with a Johnson and Segalman constitutive equation (ref. 58) which is a special case of the molecular model developed by Phan Thien and Tanner (ref. 59). In addition to elasticity this model also allows shear thinning and there is no available experimental data for comparison. However according to Keunings and Crochet this work (ref. 57) "reopens the acute question of why the iterative procedure fails under some sets of circumstances related to the mesh (used), the fluid and the problem".

CONCLUSION

Families of non-shear thinning elastic fluids are now available which allow elastic effects to be observed in the absence of significant shear thinning and inertial effects in flow fields of practical interest. The behaviour of these ideal fluid system is consistent with molecular theory and their continuum properties are well fit by the Oldroyd B constitutive equation at low shear rates. Observed elastic effects in a non-viscometric flow field (squeeze film flow) have been predicted from continuum theory for the first time. However the question remains as to why the finite element predictions are adequate in one flow field and are inadequate in another. It is clear that a better constitutive equation is required for this class of fluid behaviour. With such an equation and good experimental observations the gap between continuum theory and practical reality in polymer processing will be bridged.

ACKNOWLEDGEMENT

The help of my friend and colleague, R.J. Binnington, in the preparation of this manuscript is greatly appreciated. Our work in non-Newtonian fluid mechanics is supported by the Australian Research Grants Scheme.

REFERENCES

- 1. J.L. White and A. Kondo, J. Non-Newtonian Fluid Mech., 6, 41 (1977).
- K.S. Hyun, Polym. Eng. Sci., 14, 666 (1974).
- D.V. Boger and M.M. Denn, J. Non-Newtonian Fluid Mech., 6, 163 (1980). J. Vlachopoulos, Rheol. Acta, 13, 223 (1974).
- I. Jen Chen, "A Comparative Study of Rheological Models for Polymer Melts", Ph.D. Thesis, University of Tennessee, Knoxville, Tenn., USA (1971).
- D.V. Boger, <u>Nature</u>, <u>265</u>, 126 (1977). T.H. Nguyen, "The Influence of Elasticity on Die Entry Flow", Ph.D. Thesis, Monash University, Clayton, Victoria, Australia (1978).
- Y. Ide and J.L. White, <u>J. Appl. Polym Sci.</u>, <u>22</u>, 1061 (1978). P.J. Cable and D.V. Boger, <u>A.I.Ch.E. J.</u>, <u>24</u>, <u>869</u> (1978); <u>24</u>, 992 (1978) and <u>25</u>, 152 (1979).
- D.V. Boger, J. Non-Newtonian Fluid Mech., 3, 87 (1977/1978).
- 11. D.V. Boger and H. Nguyen, Polym. Eng. Sci., 18, 1037 (1978).
- 12.
- K. Walters, J. Non-Newtonian Fluid Mech., 5, 113 (1979).

 D.V. Boger and R. Binnington, Trans. Soc. Rheol., 21, 515 (1977).

 H. Nguyen and D.V. Boger, J. Non-Newtonian Fluid Mech., 5, 353 (1979).

 J.C. Chang and M.M. Denn, J. Non-Newtonian Fluid Mech., 5, 369 (1979).
- B. Mena, O. Manero and D.M. Binding, J. Non-Newtonian Fluid Mech., 5, 427 (1979). 16.
- 17.
- D.V. Boger and M.M. Denn, J. Non-Newtonian Fluid Mech., 6, 163 (1980).
 R.P. Chhabra, P.H.T. Uhlherr and D.V. Boger, J. Non-Newtonian Fluid Mech., 6, 187 (1980). 18.
- D.V. Boger, "Separation of Shear Thinning and Elastic Effects in Experimental Rheology", Rheology, Vol. 1, Edited by G. Astarita, G. Marrucci and L. Nicholais, Plenum Publishing Corp., New York, 195 (1980).
- R. Binnington and D.V. Boger, Proceedings of the 2nd National Conference on Rheology 20. (Australian Branch of the British Society of Rheology), Sydney, 117 (1981).
- T. Cochrane, K. Walters and M.F. Webster, Phil. Trans. R. Soc. Lond., A 301, 163 (1981). 21.
- D.V. Boger, "Entry Flows of Inelastic and Viscoelastic Fluids", Advances in Transport Processes, Vol. 2, pp. 43-104, Wiley International, New Delhi (1981). 22.
- 23. K. Walters and D.M. Rawlinson, Rheol. Acta, 21, 547 (1982).
 24. K. Walters and M.F. Webster, Phil. Trans. R. Soc. London, A 308, 199 (1982).
- 25. D.R. Oliver, A.W. Ninow, R.J. Mitson and K. Terry, Chem. Eng. Research and Des., 62(2), 123 (1984).
- 26. D. Sigli and A. Maalouf, J. Non-Newtonian Fluid Mech., 9, 191 (1981).
- R.K. Prud'homme and E. Shagfeh, A.I.Ch.E. J., 30, 485 (1984).
- 28. M.J. Crochet and K. Walters, Ann. Rev. Fluid Mech., 15, 241 (1983).

- D.V. Boger, "Dilute Polymer Solutions and their Use to Model Polymer Processing Flows", Interrelations between Processing Structure and Properties of Polymer Materials, Eds. J.C. Seferis and P.S. Theocaris, Elsevier Science Publishers, Amsterdam, 307 (1984) and in Proceedings of the Third National Conference on Rheology, Australian Society of Rheology, 11, Melbourne (1983).
- R.K. Gupta, G. Prilutski, T. Sridhar and M.E. Ryan, J. Non-Newtonian Fluid Mech., 12, 233 (1983).
- R.K. Gupta, T. Shridhar and M.E. Ryan, Proceedings of the Third National Rheology Conference, Australian Society of Rheology, 33, Melbourne (1983).
- L. Choplin, P.J. Carreau and A. Ait Kadi, Polym Eng. and Sci., 23, 445 (1983).
- K.P. Jackson, K. Walters and R.W. Williams, J. Non-Newtonian Fluid Mech., 14, 173 (1984).
- R.J. Binnington and D.V. Boger, "Constant Viscosity Elastic Liquids", In Press, J. Rheol. (1985)
- R.J. Binnington and D.V. Boger, "Misconceptions on Non-Shear Thinning Elastic Fluids", In Press, Polym. Eng. Sci. (1985)
- N. Phan-Thien, J. Dudek, D.V. Boger and V. Tirtaatmadja, "Squeeze Film Flow of Ideal Elastic Fluids", In Press, J. Non-Newtonian Fluid Mech. (1985).
- V.H. Giesekus, Rheol. Acta, 6, 339 (1967).
- M.G. Hansen and J.B. Jansma, Rheology, Eds., G. Astarita, G. Marrucci and L. Nicolais, Plenum Press, New York, p. 193 (1980).
- 39. D.U. Hur, Private Communication, Department of Chemical Engineering, University of Melbourne, January (1985).
- R.B. Bird, O. Hassager, R.C. Armstrong and C.F. Curtis, Dynamics of Polymer Liquids: 40. Vol. II - Kinetic Theory, p. 490, John Wiley and Sons, New York (1977).
- M. Keentok, A.G. Georgesen, A.A. Sherwood and R.I. Tanner, J. Non-Newtonian Fluid Mech., 6, 303 (1980).
- S.J. Lee, M.M. Denn, M.J. Crochet, A.B. Metzner and G.J. Riggins, J. Non-Newtonian Fluid 42. Mech., 14, 301 (1984). J. Stefan, Akad. Wiss. Math. Natur., 69, 713 (1874).
- 43.
- N. Phan-Thien, J. Dudek, D.V. Boger and V. Tirtaatmadja, "Squeeze Film Flow of Ideal 44. Elastic Liquids", In Press, J. Non-Newtonian Fluid Mech. (1985). K.R. Reddy and R.I. Tanner, J. Rheol., 22, 661 (1978).
- 45.
- 46. P.W. Chang, T.W. Patten and B.A. Finlayson, Comp. Fluids, 7, 267 (1979).

 47. M.J. Crochet and R. Keunings, J. Non-Newtonian Fluid Mech., 7, 199 (1980); 10, 85 (1982).
- 48. C.J. Coleman, J. Non-Newtonian Fluid Mech., 8, 261 (1981).
- 49. M.J. Crochet and R. Keunings, J. Non-Newtonian Fluid Mech., 10, 339 (1982).
- B. Caswell and M. Viriyayuthakorn, J. Non-Newtonian Fluid Mech., 12, 13 (1982).
- N.Y. Tuna and B.A. Finlayson, <u>J. Rheol.</u>, <u>28</u>, <u>79</u> (1984). R.I. Tanner, <u>J. Polym. Sci.</u>, <u>A2</u>, <u>8</u>, 2067 (1970). 51.

- 53. J. Vlachopoulos, Proc. 2nd World Congr. Chem. Eng., Montreal, p. 269 (1981).
 54. R. Racin and D.C. Bogue, J. Rheol., 23, 263 (1979).
 55. M.J. Crochet and M. Bezy, J. Non-Newtonian Fluid Mech., 5, 201 (1979); Rheology, Vol. 2, G. Astarita, G. Marrucci and L. Nicolais (Eds), Plenum Press, p. 3, New York (1980).
- 56. M. Viriyayuthakorn and B. Caswell, <u>J. Non-Newtonian Fluid Mech.</u>, <u>6</u>, 245 (1980).
- 57. R. Keunings and M.J. Crochet, J. Non-Newtonian Fluid Mech., 14, 279 (1984).
 58. M.W. Johnson and D. Segalman, J. Non-Newtonian Fluid Mech., 2, 255 (1977).
 59. N. Phan-Thien and R.I. Tanner, J. Non-Newtonian Fluid Mech., 2, 353 (1977).