NON-NEWTONIAN FLUID MECHANICS

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Contents

Summary

Non-Newtonian Fluid Mechanics is a field of study which is growing in prominence and importance as the years progress, not least because many of the fluids one encounters in everyday life are non-Newtonian in their behavior.

To discuss the various aspects of research in the non-Newtonian fluid mechanics field, we shall divide the text into five basic sections:

(i) A study of the fluids in *simple* flow situations, such as those found in Rheometry.
(ii) The construction of suitable constitutive equations for the non-Newtonian fluids under test.
(iii) The use of these constitutive equations in the prediction of behavior in *complex* flows of practical importance.
(iv) An experimental study of the behavior of non-Newtonian fluids in the complex-flow situations discussed in (iii).
(v) A comparison of the theoretical predictions of (iii) with the experimental results of (iv).

1. Introduction

It is self evident that non-Newtonian Fluid Mechanics must be closely related to the well known field of Newtonian Fluid Mechanics, and we must therefore begin the present chapter with a brief consideration of this classical field.

As the name suggests, it originates from the research of the famous British scientist, Sir Isaac Newton, published in 1687. Considering what we would now identify as steady simple shear flow (see Figure 1), Newton essentially proposed the following equation:

$$\sigma = \eta \frac{d\gamma}{dt}, \tag{1}$$

where σ is the shear stress, $d\gamma / dt$ the rate of strain, and η is the (constant) coefficient of viscosity. It is well known that air and water are two very common fluids that obey Newton's postulate.

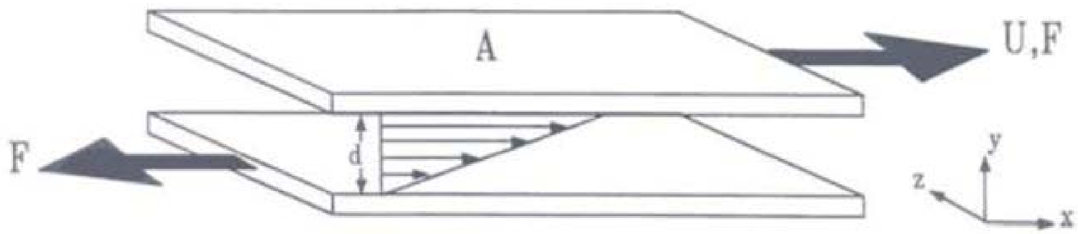

Figure 1. Steady simple shear flow. Two parallel planes are located at $y = 0$ and $y = d$, the intervening space being filled with sheared liquid. The upper plate moves with a relative velocity U and the lengths of the arrows between the plates are proportional to the local velocity in the liquid.

It wasn't until over a century later that Navier and Stokes independently developed a consistent three-dimensional theory for a "Newtonian" viscous fluid and the so-called Navier-Stokes equations form the basis for what is now universally known as Newtonian Fluid Mechanics (see, for example Tanner and Walters 1998).

However, from the 19th century onwards, it became abundantly clear that many fluid-like materials could not be described as "Newtonian" and this clearly provided the motivation for the upsurge of interest in the field known as *non*-Newtonian Fluid Mechanics. This is now seen as an important scientific discipline in its own right, with its own ethos and dedicated research journal. (The "Journal of non-Newtonian Fluid Mechanics", published by Elsevier).

Before we leave generalities, it is important to point out that although (most) non-Newtonian fluids possess a viscosity which varies with shear rate and therefore violates Eq. (1), it is not appropriate to define a non-Newtonian fluid simply as one that does not obey Newton's postulate – for the simple reason that *some* elastic fluids (like the so-called Boger fluids (Boger 1977/78)) do in fact possess a constant viscosity and would therefore satisfy Eq. (1) in a steady simple shear flow. So, a non-Newtonian fluid is accordingly defined as one whose behavior cannot be described by the Navier-Stokes equations.

As we shall see, there are *esoteric* reasons for a study of non-Newtonian fluid mechanics, but there are also more practical reasons, driven by the fact that many of the fluid-like materials one meets in everyday life are non-Newtonian in their behavior. One could cite many such examples, ranging from liquid detergents, multigrade oils, paints and printing inks to more viscous materials like bitumen and molten plastics, but this list is by no means complete. For this reason, if for no other, non-Newtonian fluid mechanics must be viewed as a very important scientific discipline.

2. The Various Strands of Research Activity in Non-Newtonian Fluid Mechanics

Any systematic study of non-Newtonian fluid mechanics must involve the following components:

(i) A systematic study of the behavior of the non-Newtonian fluids of interest in simple flow situations, such as those found in conventional rheometers.
So "Rheometry" must play an important role and the flows of interest necessarily include steady simple shear flow, small-amplitude oscillatory shear flow and extensional flow. In the former, the normal stress differences as well as the shear stress are of interest.

(ii) The construction of suitable constitutive Eqs. (rheological equations of state) for the non-Newtonian fluids under test. Such equations must necessarily satisfy the well known mathematical constraints arising from a consistent application of the Principles of Continuum Mechanics. Further, their general form can often be deduced from a consideration of the fluid's microstructure. It goes without saying that any proposed equations must be able to simulate, in simple flows, the rheometrical data provided in (i).
We shall see that there is no difficulty in satisfying these conditions *in principle*, but some pragmatism is usually required to meet the dual constraints of tractability and predictive capability.

(iii) The prediction of the behavior of non-Newtonian fluids (particularly highly-elastic non-Newtonian fluids) in *complex* flows of practical importance. To all intents and purposes, this now involves Computational Fluid Dynamics (CFD) applied within a non-Newtonian framework. Here, the constitutive Eqs. (arising from (ii)) have to be solved in conjunction with the familiar equations of motion and continuity, subject to appropriate boundary conditions, which can sometimes be more taxing than those arising in Newtonian fluid mechanics.

(iv) Quite naturally, the fourth component in the research program has to involve an *experimental* study of the behavior of the non-Newtonian elastic fluids in the complex flows already studied theoretically in (iii). Clearly, the experimental program can be driven by any provocative conclusions arising from the theoretical predictions, but, more often, the experimental work in (iv) provides the motivation for the theoretical work, rather than vice versa.

(v) The final component of the research program necessarily involves a realistic comparison of the predictions from (iii) with the experimental data of (iv). This can be seen as an essential part of the Scientific Method.

If the experimental data are reliable and there is a lack of agreement between (iii) and (iv), two courses of action are usually considered. In the first place, the reliability of the theoretical results has to be addressed. In these days when CFD is a dominant influence, there are important numerical complications that are not present in the solution of the Navier-Stokes equations for Newtonian fluids; there is sufficient evidence in the recent computation rheology literature to make this abundantly clear (see, for example, Owens and Phillips 2002, Walters and Webster 2003).

If and when the theoretical (especially numerical) techniques are shown to be reliable and there is still disagreement between theory and experiment, the rheologist then has to look into the choice of constitutive model from area (ii). Of course, this is not a "luxury" which is possible in classical fluid dynamics, where the Navier-Stokes equations are taken as sacrosanct.

For the rheologist, there are many cases where there is encouraging agreement between computation and experiment in complex flows. However, there are also famous cases where this agreement is very, very, elusive (see, for example, Walters 2006).

In setting out the above program of research, we are not suggesting that it is unique or flawless and we would have sympathy, for example, with an argument in favor of a major input from *micro* rheological considerations. However, the research program we have outlined can be defended as at least providing one possible coherent attempt to predict and understand the unusual and often bizarre behavior of elastic non-Newtonian fluids as these are made to flow in complex geometries.

Before concluding this section, we need to mention that in non-Newtonian fluid mechanics, it is convenient to define (at least) two non-dimensional numbers, R and W to characterize the flows. The usual Reynolds number R is given by

$$R = \frac{\rho U L}{\eta}, \qquad (2)$$

where η is an appropriately chosen viscosity, ρ is the density, U is a characteristic velocity and L is a characteristic length. The elasticity number W, which is often called the Weissenberg number or (sometimes) Deborah number, is given by

$$W = \lambda \frac{U}{L}, \qquad (3)$$

where λ is a characteristic relaxation time.

For most non-Newtonian elastic fluids, it is far from a trivial matter to arrive at an appropriate η and λ. However, for the important sub class of fluids called Boger fluids (Boger 1977/78), which we shall major on for illustration purposes, there is no ambiguity in defining η, since the viscosity of this class of fluids is effectively constant. Furthermore, λ is often defined consistently from the viscometric data of the fluids at low shear rates.

3. Rheometry

There are two (not necessarily unrelated) objectives in Rheometry. The first simply involves a straightforward attempt to determine the behavior of non-Newtonian fluids in a number of simple (rheometrical) flows, using suitably defined material functions. The simple desire here is to seek a correlation between material properties and molecular structure or, alternatively, between material properties and observed behavior in practical situations. In this latter connection, "Quality Control" is often an important driving force.

The second objective is concerned indirectly with the prediction of the behavior in non-simple flows of practical importance from the results of simple rheometrical experiments. As we shall see in section 4, the rheometrical data here are an important aid in the construction of constitutive equations for the non-Newtonian fluids, which may then in turn be used to predict behavior in more complex practical situations. The more accurate the rheological data the more accurate will be the constitutive model, so that a very detailed research program is required to meet this objective.

So, in one sense, the two objectives of Rheometry essentially take us from "Quality Control" to "Process Modeling".

Three basic rheometrical flows need to be considered. The first and most well known is steady simple shear flow, something we have already referred to in connection with Newton's postulate.

At this point, it is important to emphasize that for non-Newtonian fluids an indicial notation is essential. Specifically, we need to introduce the so-called stress tensor σ (Figure 2).

So, if we concentrate on the top surface of the volume in the figure, there will be a stress in the normal direction denoted by σ and shear components in the plane of the surface denoted by σ and σ. In this notation, the first index is linked to the orientation of the material surface and the second to the direction of the stress. The usual sign convention is such that a positive σ is a "tension".

Let us now consider a steady simple shear flow, in which there is flow only in the *x* direction, depending simply and linearly on the y coordinate. Here, we have written $\dot{\gamma}$ for the so-called rate of shear (or shear rate).

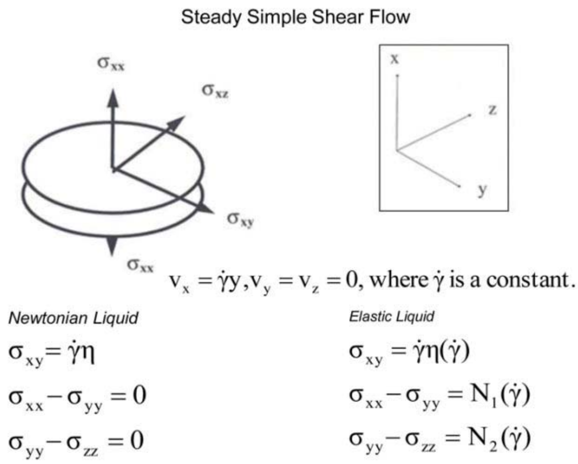

Figure 2. Steady simple shear flow for Newtonian and non-Newtonian elastic liquids.

As we have seen, for a Newtonian fluid, the stress distribution simple involves one material constant – the coefficient of viscosity. This can of course depend on the pressure and temperature, but it is constant so far as dependence on $\dot{\gamma}$ is concerned.

For a non-Newtonian elastic fluid, the stress distribution is more complex. The shear stress is now a non-linear function of the shear rate and there are two normal stress differences, the first and most important being N. N is the less important "second normal stress difference".

It is customary to show schematically the various types of behavior so far as the shear stress is concerned (now written σ for convenience), as in Fig. 3.

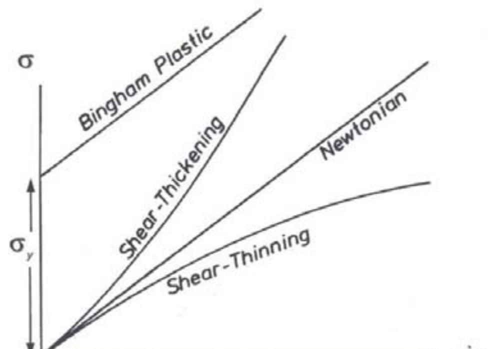

Figure 3. A schematic representation of the various kinds of response in a steady simple shear flow.

We see that there can be shear thinning, shear thickening, or even a constant viscosity (as in Boger fluids). Note also the possibility of so-called plastic behavior, where the material will not flow until a critical stress, called the yield stress, is exceeded.

For the majority of non-Newtonian elastic fluids, shear thinning will be in evidence and Fig 4 contains a typical response, where the viscosity falls from a so-called zero shear value η_0 to a lower "second Newtonian" plateau η_∞. The region where the viscosity falls is usually quite well described by a power law, i.e. by a straight line on a suitable log/log plot. For some non-Newtonian fluids, the difference between η_0 and η_∞ can be very severe.

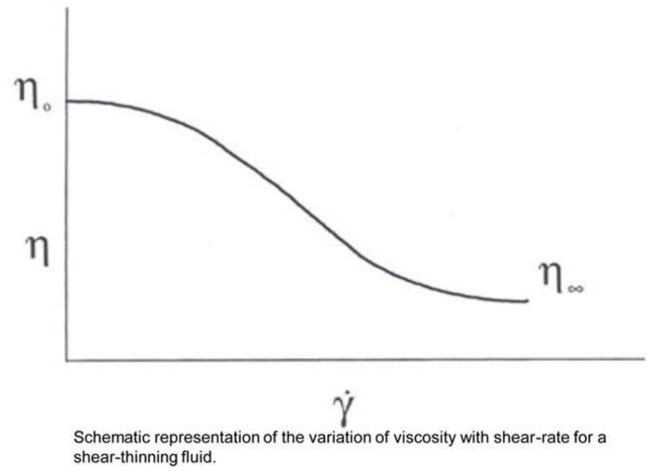

Figure 4. A schematic representation of shear-thinning behavior.

Turning now to the normal stress differences, Figure 5 shows typical data for a 1% aqueous solution of polyacrylamide. Note that σ and N_1 are both well represented by a power-law response. Note also that N_1 is about ten times larger than σ.

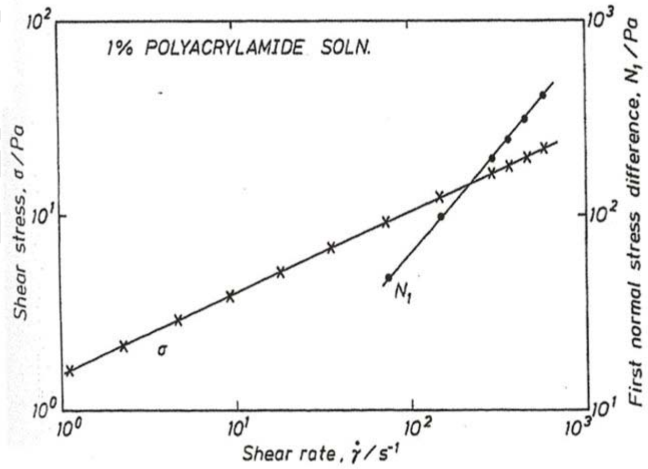

Figure 5. Viscometric date for a 1% aqueous solution of polyacrylamide at 20ºC (cf. Barnes et al 1989, Figure 4.2).

In many ways, the first normal stress difference N_1 can be considered to be a convenient measure of the level of "viscoelasticity". It is certainly important in flows like mixing and extrudate swelling. In contrast, the second normal stress difference N_2 is usually relatively small. For many polymeric systems, it is found to be negative and about one tenth of N_1 in magnitude, (see, for example, the data shown in Figure 6). However, for some polymer melts, $|N_2|$ has been reported to be sometimes as high as 25% of N_1 (cf. Tanner and Walters 1998).

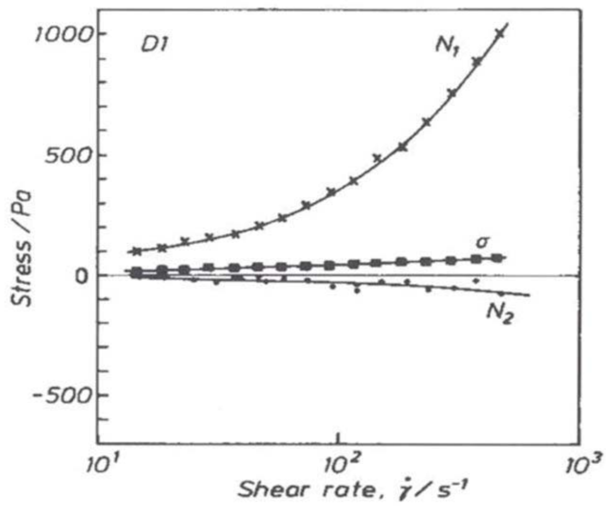

Figure 6. Viscometric data for a 2% w/v polyisobutylene (oppanol B200) in dekalin at 25ºC. (See, for example, Barnes et al 1989, Figure 4.6).

It is possible to point to some isolated circumstances where it is important to take N_2 seriously, e.g. in the case of shear fracture in rotational rheometers and other kinds of instabilities, in extrusion swelling, and in pressure-driven flow through pipes of *non*-circular cross section.

We now pass on to consider another very popular means of determining information about the viscoelastic properties of non-Newtonian fluids, namely that associated with small-amplitude oscillatory-shear flow, with the flow in the *x* direction in Fig 1 now involving the time as well as the *y* coordinate. α is a small amplitude in Eq. (4), small enough in fact for non-linear terms to be neglected.

$$v_x = \dot{\gamma}\alpha\omega\cos\omega t, v_y = v_z = 0, \qquad (4)$$

$$\sigma_{xy} = \alpha\omega\left[\eta'\cos\omega t + \frac{G'}{\omega}\sin\omega t\right]. \qquad (5)$$

In this case, the only stress component of relevance is σ_{xy}, which can be expressed in terms of two functions η' and G', both of which are functions of the imposed frequency ω . η' is called the dynamic viscosity and G' the dynamic rigidity.

In the literature, there are alternative but equivalent ways of displaying the data. For example, in place of η', many rheologists choose to use instead the so-called loss modulus G'', given by

$$G'' = \eta'\omega. \tag{6}$$

Figure 7 contains a schematic representation of one of the various choices.

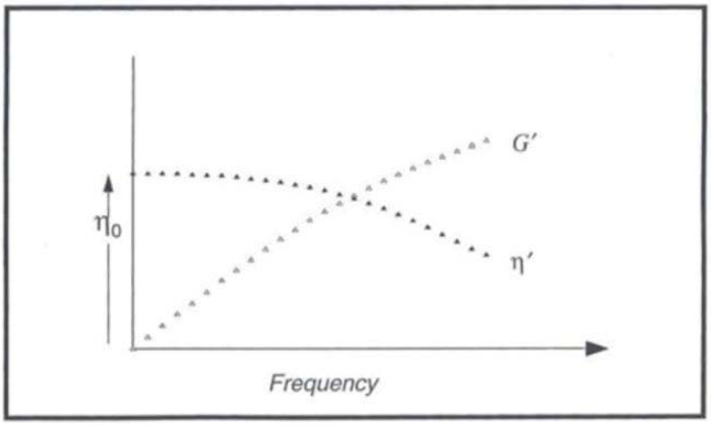

Figure 7. Schematic representation of the dynamic viscosity η' and dynamic rigidity G' as functions of the frequency.

When the so-called "dynamic data" are used in the second objective of Rheometry (i.e. the construction of constitutive equations for the test fluids), it is very important that the frequency range is as broad as possible, since the data have to be inverted to yield the so-called relaxation spectrum (or relaxation function) as a function of relaxation time. This is now seen as a non-trivial problem for the analyst, not least because the procedure is "ill posed" and the reliability of the relaxation spectrum on the time axis depends critically (but not simply) on the available range of the experimental dynamic data on the frequency axis (see, for example, Davies and Anderssen 1997).

We now need to consider the third basic rheometrical flow, namely "extensional flow". In the late 1960s, there was a dramatic surge of interest in extensional-viscosity measurement and there does not seem to be any indication of a fall off of interest in the subject – on the contrary!

The relevant deformation is described by:

$$v_x = \dot{\varepsilon}x, v_y = -\frac{\dot{\varepsilon}y}{2}, v_z = -\frac{\dot{\varepsilon}z}{2}, \tag{7}$$

$$\sigma_{xx} - \sigma_{yy} = \sigma_{xx} - \sigma_{zz} = \dot{\varepsilon}\eta_E\left(\dot{\varepsilon}\right). \tag{8}$$

In this case, the uniaxial extension is in the x direction, with contraction in the y and z directions. Here, the normal stress differences are important and these are expressed in terms of a so-called extensional viscosity η_E, which is a function of the extensional strain rate $\dot{\varepsilon}$. For a Newtonian fluid, it is easy to show that, as defined, η_E is simply three times the shear viscosity, a result first obtained by Trouton in 1906. So, rheologists speak of the Trouton ratio T_r, which is 3 for a Newtonian fluid. For non-Newtonian elastic fluids, particularly polymeric systems, T_r can be very high indeed, with values in excess of 30, 300 or even 3,000! For this reason, if for no other, the extensional viscosity is an important rheometrical variable and must be taken very seriously in both Rheometry objectives.

We conclude this section on Rheometry with a consideration of some of the limiting relationships between the various rheometrical functions we have introduced. It must be emphasized that these are not empirical, but based on sound continuum mechanics (see, for example, Barnes et al 1989, p80).

$$\eta(\dot{\gamma})\big|_{\dot{\gamma}\to 0} = \eta'(\omega)\big|_{\omega\to 0}, \tag{9}$$

$$\frac{N_1(\dot{\gamma})}{2(\dot{\gamma})^2}\bigg|_{\dot{\gamma}\to 0} = \frac{G'(\omega)}{\omega^2}\bigg|_{\omega\to 0}, \tag{10}$$

$$\eta_E\left(\dot{\varepsilon}\right)\big|_{\dot{\varepsilon}\to 0} = 3\eta(\dot{\gamma})\big|_{\dot{\gamma}\to 0}, \tag{11}$$

$$\frac{d\eta_E}{d\dot{\varepsilon}}\bigg|_{\dot{\varepsilon}\to 0} = \frac{3}{2\dot{\gamma}^2}\left[N_1\left(\dot{\gamma}\right) + 2N_2\left(\dot{\gamma}\right)\right]\bigg|_{\dot{\gamma}\to 0}. \tag{12}$$

Equation (11) implies that, whatever happens at finite strain rates, the limiting value of η_E for non-Newtonian fluids must satisfy the Trouton limit.

The final relationship (12) is an interesting one. From what we know of the steady-shear behavior for both polymer solutions and polymer melts, the term in brackets is invariably positive, so we should expect an initial non-zero positive slope in the extensional viscosity/strain rate graph, whatever happens at finite strain rates.

In conclusion, we note that we have introduced three basic kinds of rheometrical functions – those arising in steady simple shear flow (3 in all), those emanating from small amplitude oscillatory shear flow (2 in all) and the uniaxial extensional viscosity. All six functions are of importance to the rheologist as he seeks to construct constitutive equations for his test fluids.

There are other related rheometrical flows that could be added to the above list, such as *finite*-amplitude oscillatory shear flow and *planar* extensional flow, to mention just two. But the rheologist is usually satisfied with the three basic flows we have concentrated on, with any other tests being seen as "critical experiments" to assess the validity or otherwise of constitutive equations based on the three foundational rheometrical flows.

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Bibliography

Alves M.A, Oliveira P.J. and Pinho F.T. (2004) *On the effect of contraction ratio in viscoelastic flow through abrupt contractions.* J non Newtonian fluid Mechanics **122**, 117-130. [An important computational-rheology paper, which contains simulations of a complex rheological flow problem.]

Barnes H.A., Hutton J.F. and Walters K. (1989) *An introduction to rheology*. Elsevier. [A very popular introductory text.]

Bird R.B., Armstrong R.C. and Hassager O. (1987) *Dynamics of polymeric liquids* Vol 1 *Fluid mechanics.* 2nd Edition. John Wiley and Sons, Inc. [A very influential research text book.]

Boger D.V. (1977/78) *A highly elastic constant-viscosity fluid*. J. non-Newtonian Fluid Mechanics **3**, 87-91. [Introduces the constant-viscosity elastic fluid which is now known as the "Boger fluid".]

Boger D.V. and Walters K. (1993) *Rheological Phenomena in Focus.* Elsevier. [Contains a comprehensive set of flow-visualization pictures for non-Newtonian fluids.]

Boger D.V. and Walters K. (2000) *Experimental dilemmas in non-Newtonian fluid mechanics and their theoretical resolution*. Korea-Australia rheology journal **12**, 27-38. [The title of the paper says it all.]

Crochet M.J., Davies A.R. and Walters K. (1984) *Numerical simulation of non-Newtonian flow*, Elsevier. [An influential early book on computational rheology.]

Crochet M.J. and Walters. K. (1983) *Numerical methods in non-Newtonian fluid mechanics*. Ann. Rev. Fluid. Mech. **15**, 241-260. [A chapter on computational rheology in the influential "Annual Review" series.]

Crochet M.J. and Walters. K. (1993) *Computational rheology: a new science*. Endeavour **17**, 64-77. [A "popular" introduction to the subject.]

Davies A.R. and Anderssen R.S. (1997) *Sampling localization in determining the relaxation spectrum*. J. non-Newtonian fluid mechanics **73**, 163-179. [Introduces the now well-known "π/2 theorem".]

Evans R.E. and Walters K. (1986) *Flow characteristics associated with abrupt changes in geometry in the case of highly elastic liquids*. J. non-Newtonian fluid mechanics **20**, 11-29. [Provides planar contraction-flow pictures for shear-thinning polymer solutions.]

Nigen S. and Walters K. (2002) *Viscoelastic contraction flows: comparison of axisymmetric and planar configurations*. J. non-Newtonian fluid mechanics **103**, 343-359. [Contains information on the flow structure and the Couette correction].

Oldroyd J.G. (1950) *On the formulation of rheological equations of state*. Proc. Roy. Soc. **A200**, 523-541. [Arguably the most important paper on theoretical rheology.]

Owens R.G. and Phillips T.N. (2002) *Computational rheology*. Imperial College Press. [A comprehensive recent research text on the subject, which includes work done after the publication (in 1984) of the Crochet et al text.]

Phan Thien N. and Tanner R.I. (1977) *A new constitutive equation derived from network theory*. J. non-Newtonian Fluid Mechanics **2**, 353-365. [Introduces the well-known PTT model].

Pipkin A.C. and Owen D.R. 1967 *Nearly viscometric flows*. Phys. Fluids **10**, 836-843. [Derives the general equations for near-viscometric flows.]

Tanner R.I. (2000) *Engineering rheology* 2nd Edition. Oxford University Press. [A comprehensive text book on the subject.]

Tanner R.I. and Walters K. (1998) *Rheology: An historical perspective*. Elsevier. [A useful text on the history of the field.]

Tomé M.F., Grossi L., Castelo A., Cuminato J.A., McKee, S. and Walters K. (2007) *Die-swell, splashing drop and a numerical technique for solving the Oldroyd B model for axisymmetric free surface flows*. J. non-Newtonian fluid mechanics **141**, 148-166. [An encouraging recent development in the field of computational rheology.]

Truesdell C. and Noll W. (1965) *The non-linear field theories of mechanics*. Springer. [An influential book on continuum mechanics.]

Walters K. (1979) *Developments in non-Newtonian fluid mechanics – a personal view*. J. non-Newtonian fluid mechanics **5**, 113-124. [Contains a critical assessment of work in continuum non-Newtonian fluid mechanics which existed in the late 1970s.]

Walters K. (2006) *A rheological "ocean of truth"*. Proc. SASOR 2006, Cape Town, 24-27 September 2006, 13-22. [Discusses some of the recent successes and failures in the field.]

Walters K. and Rawlinson D.M (1982) *On some contraction flows for Boger fluids.* Rheologica Acta **21**, 547-552. [Shows the lack of vortex enhancement when Boger fluids flow in planar contractions.]

Walters K. and Webster M.F. (2003) *The distinctive CFD challenges of computational rheology*. Int. J. for Numerical Methods in Fluid **43**, 577-596. [A review article on the state of the field in 2003.]

Biographical Sketch

Professor Ken Walters was appointed Professor at the University of Wales Aberystwyth (now Aberystwyth University) in 1973. He was awarded a DSc degree in 1985. He is a former President of the British Society of Rheology and received their gold medal in 1984. He was elected a Fellow of the Royal Society in 1991 and is a Foreign Associate of the National Academy of Engineering of the United States. In 1998, he was awarded an Honorary Doctorate by the Université Joseph Fourier in Grenoble, France and received the Weissenberg Award of the European Society of Rheology in 2002.

Professor Walters is the author of several books on rheology, rheometry and non-Newtonian fluid flow. He was Executive Editor of the Journal of Non-Newtonian Fluid Mechanics from its launch in 1976 until the publication of Volume 100 in 2002. From 1996-2000, Professor Walters was the first President of the European Society of Rheology, and from 2000-2004, he was Chairman of the International Committee on Rheology.