COMPUTATIONAL RHEOLGY

M.F. Webster, H.R. Tamaddon-Jahromi, and **F. Belblidia**
Institute of non-Newtonian Fluid Mechanics - Wales, School of Engineering, Swansea University, Singleton Park, SA2 8PP, Swansea, UK

Keywords: Rheology, Viscoelasticity, Inelastic, Constitutive models, Kinetic theory, Contraction flows, Finite elements/volume, Weissenberg number, Compressibility, Transient flows, Vortex activity, Pressure-drop, Extensional flow, Numerical predictions, Experiments

Contents

Summary

A general overview of the present state of the art in the field of computational rheology is reviewed, emphasizing the various numerical challenges that have emerged through the material modeling and discretization approaches adopted. There is brief coverage of the alternative levels of modeling involved, from macroscopic to mesoscopic, alongside the different discrete techniques utilized and the challenges posed by prescribed benchmark problems. Progress in the field is described historically through consideration of a particular benchmark problem, contraction flow, detailing advances in experimental, numerical and analytical developments that illustrate the many branches of computational rheology. Several case studies are also presented that illustrate some selected applications, covering such interests areas as transient flow solutions, kinetic-based models, pressure drop analysis and compressible-viscoelastic computations.

1. Introduction

Computational rheology involves the use of numerical simulation for non-Newtonian fluid flow in complex geometries, and in particular introduces viscoelastic effects in polymeric liquids. For more than thirty years, the subject has been under intense development alongside the tremendous increase in computer power, and has now reached a state of relative maturity with new horizons ahead. To date, many reviews have appeared by an evolving international research community; see for example

general reviews by Tanner & Walters, Crochet et al., Keunings, Baaijens, and Walters & Webster.

Industrial application remains a principal driving force behind the development and advance in computational rheology. Fluids of interest include molten plastics and polymer solutions, as well as industrial suspensions, multigrade oils, liquid detergents, printing inks and various food and confectionary products. The processes of relevance include injection molding, extrusion, coating, mixing and lubrication. The research field is strongly motivated, broad in its scope and challenging. This introduces some fascinating numerical challenges to computational rheology. Across the broad range of application this article is purposely restricted to viscoelastic liquids and contraction flow geometries.

The challenge for computational rheologists is to develop accurate, stable and robust numerical schemes based on realistic physical-mathematical models. This advancing field offers opportunities at different levels. Theoreticians, and material modelers, are required to reduce the gap between the several levels of description employed to predict polymer dynamics, including the choice of appropriate macroscopic constitutive equation, or through molecular modeling. This aids in improved understanding of some of the macroscopic flow phenomena observed in non-Newtonian fluid mechanics, such as vortex activity and flow instability. Experimentalists wish to improve the design of their equipment and experimental procedures through better interpretation of their data. This may be advanced by identifying and eliminating experimental deficiencies, such as flow heterogeneities. It is the combination of flow experiments and numerical predictions that is sought to characterize rheological behavior in complex flows. With industrial application and production in mind, improved properties of end-of-line products are desired through the combination of computational rheology and computer aided design studies.

Polymeric liquids are considered as viscoelastic materials, with their particular properties responsible for some extravagant flow phenomena. There is need to analyze and predict their flow behavior, through a combination of suitable physical models and numerical techniques. The challenge posed by polymeric materials is the broad range of time and length-scales separating the relevant atomistic and macroscopic processes involved, supplemented by the large number of microstructural entities. Analysis is made possible by appealing to a coupling of theoretical models, spanning from quantum mechanics to continuum mechanics.

At the micro-structural level, the atomistic approach remains restricted to coarse models for polymeric liquid flow in geometries of molecular dimensions, such as considered in the wall-slip problem. This is mainly due to the significant computer resources required in such simulations. The ultimate aim, here is to solve such flow problems on the macroscopic level, yet based on this atomistic approach.

The kinetic theory level is the next level of description of polymeric liquids, where stochastic simulation or Brownian dynamics methods are employed. For example, a dilute solution of linear polymers in a Newtonian solvent may be described at a macroscopic level, by a freely-jointed bead-rod Kramers chain of a number of beads

connected linearly by rigid segments or springs. Increasing the number of beads provides model resolution at this level of representation. Kinetic theory models provide a coarse-grained description of molecular configurations, wherein atomistic processes are ignored. Such a model tackles important features that govern the flow-induced evolution of configurations. In recent years, kinetic theory has advanced well beyond the classical reptation tube-model proposed by de Gennes (1971, J. Chem. Phys.) and further developed by Doi & Edwards (1986, *The theory of polymer dynamics*, Clarendon). An impressive number of recent Brownian dynamics studies have emerged based on Kramers chains, bead-spring chains and dumbbells. The interest in such theory continues to grow, through which numerical studies have significantly advanced the understanding of polymer dynamics in general.

Under the widely utilized continuum mechanics approach, the fluid microstructure is not explicitly taken into account. The combination is forged between a suitable constitutive equation (differential or integral) which reflects material response under deformation, with the governing equations of motion for viscoelastic fluids; typically for isothermal incompressible flow, those of continuity and momentum equations. The resulting system is solved commonly through space-time partial differential equations, in the form of non-linear, mixed-type parabolic-hyperbolic equation systems. Some constitutive equations applied in continuum modeling have originated from molecular models and kinetic theory. This is typified through the pom-pom constitutive equation, which was developed for branched polymer architectures.

It is the coupling of the coarse-grained molecular scale of kinetic theory to the macroscopic scale of continuum mechanics that constitutes the micro-macro approach to computational rheology. The micro-macro approach is much more demanding upon computer resources than more conventional continuum simulations, allowing for the direct exploitation of kinetic theory models, and hence, avoiding potentially harmful closure approximations. Under the micro-macro methodology, appropriate distributions of configuration spaces for molecular orientation must be sought, while under the continuum assumption, an integration of a constitutive equation is applied to evaluate the viscoelastic contribution to the stress tensor.

Recently, alternative approaches have begun to appear in the computer modeling of polymeric liquids covering a pore-scale description of flow. The attraction of such approaches is their inherent particle-based construction, resulting in meshless schemes. Here, only two formulations are cited: dissipative particle dynamics and Lattice Boltzmann modeling. Both such methods are increasing in popularity and beginning to tackle the more complex problems of serious interest to the field.

In the rapidly evolving research area of computational rheology, there is an overarching requirement to quality assess any numerical method proposed. This has led the field to establish a set of benchmark problems, each exposing particular features of interest, and against which the properties of a numerical scheme may be judged. Some of these problems are introduced and discussed in Tanner (Engineering Rheology, 2000). The selected benchmarks outlined with their features are: flow around a sphere in a tube, flow past a cylinder, flow near sharp corners and separation points, entry flow such as contraction flow, and the extrusion problem. The flow around a motionless sphere in a

tube is a geometrically smooth problem, yet numerical failure occurs for this problem at relatively low levels of elasticity. This is caused by the emergence of a thin stress boundary layer in the wake region beyond the sphere and the drag exerted on the sphere itself. Some experimental observation (as in Walters & Tanner, 1992) has suggested that drag reduction is caused by shear thinning, while fluid elasticity triggers drag enhancement. In this regard, drag calculation has become accepted as a useful means to assess numerical precision and to distinguish between the performance of a wide variety of predictive techniques. In a similar manner, drag characterization has been utilized in the alternative configuration of flow around a cylinder in a channel. Alternatively, flow near corners and separation points introduces additional numerical difficulties, such as typified in the stick-slip flow problem. This problem exhibits steep stress gradients that necessitate rigorous mesh refinement procedures. Similarly, entry flow problems such as contraction flows, present yet further numerical complexity, where the contraction ratio is a parameter of choice, popularly selected as four. In addition to the presence of the stress singularity at the re-entrant corner for abrupt contractions, this problem offers features of vortex enhancement/inhibition and pressure loss at the entry as further means to evaluate predictive precision. Moreover, the availability of asymptotic analytical solution for velocity and stress near the singularity for certain models, such as Upper Convective Maxwell, Oldroyd-B and Phan-Thien/Tanner models, validates mesh convergence findings. Stress singularity is also observed in the extrusion problem, alongside a free-surface to the extrudate. Here, the singularity is related to the change in the boundary condition at the exit of the die (transition from stick to slip), and further complication arises through the discrete treatment of the free boundary condition on the jet. This necessitates tracking of the free-surface, which is a challenging task even for relatively simplistic material properties.

In this coverage of the wide subject matter on computational rheology, attention is purposely restricted to focus upon applications of interest and features that arise through one of these benchmark problems, that of contraction flow. The merits and features of this problem are drawn out historically, through the many branches of computational techniques that have developed. In order to introduce the discrete treatment of such problems, there is brief coverage of various aspects involved in material and flow modeling. Several case studies on this benchmark problem illustrate some novel and current topics of interest, including: the prediction of some transient flows, solutions with pom-pom models, pressure drop analysis and viscoelastic-compressibility computations.

2. Flow and Governing Equations

The formulation of constitutive equations is essential in order to predict the behavior of viscoelastic materials in many complex and industrial flows including those involving polymers, colloids, foams, and gel processes. There is need here to provide a brief overview of this wide topic, so that motivation and background introduces the computational aspects to follow. Being a rapidly growing and industrially important field, rheology plays a significant role in polymer processing, food processing, coating and printing, and many other manufacturing processes. In the early days, the so-called Upper-Convected Maxwell (UCM) and Oldroyd-B models (implicit in stress) were strongly favored. This was partly due to the fact that they assumed the ‘bottom-line’ of

acceptable mathematical simplicity, whilst also being able to mimic the complex rheometrical behavior for a class of polymer solutions, known as Boger fluids, which became popular in the late 1970s.

To establish a rheological equation of state, the principle of material objectivity states that such an equation remains unchanged under a change of frame, even if the frame is time dependent. Then, if one assumes that the present response of a material is dependent upon its past history of deformation, the constitutive equation assumes a relationship between stress and history of response of the material for all times in the subsequent past. Based upon continuum modeling, the Cauchy stress tensor σ_{ik} may be represented as

$$\sigma_{ik} = -p\delta_{ik} + T_{ik} \ , \ T_{ik} = T_{ik}^{(\mathrm{s})} + T_{ik}^{(\mathrm{p})}, \qquad (1)$$

where p is an arbitrary isotropic pressure, δ_{ik} the Kronecker delta, and (T_{ik}) is an extra-stress tensor that may be split into viscous $(T_{ik}^{(\mathrm{s})})$ and polymeric $(T_{ik}^{(\mathrm{p})})$ contributions. $T_{ik}^{(\mathrm{s})}$ may be described by a generalized Newtonian model, with a viscosity (η) dependent on second and third invariants, $\mathbf{I}_2$ and $\mathbf{I}_3$, of the deformation-rate tensor $\mathrm{d} = (\nabla\mathbf{u} + \nabla\mathbf{u}^{\dagger})/2$, as

$$T_{ik}^{(\mathrm{s})} = 2\eta\left(\mathbf{\Gamma},\mathbf{\Sigma}\right) d_{ik}, \quad \Gamma = 2\sqrt{\mathbf{I}_2}, \quad \mathbf{I}_2 = \frac{1}{2}\mathrm{tr}\left(\mathrm{d}^2\right), \quad \mathbf{I}_3 = \det\left(\mathrm{d}\right). \qquad (2)$$

The generalized variable $\mathbf{\Gamma}$ can be identified as a shear-rate $(\dot{\gamma})$ in a steady simple shear flow. Similarly, $\mathbf{\Sigma} = 3\mathbf{I}_3/\mathbf{I}_2$ can be identified as an extension-rate $(\dot{\varepsilon})$ in a steady uniaxial extensional flow.

The specific form of $\eta(\mathbf{\Gamma},\mathbf{\Sigma})$ is model dependent. Various forms are commended from constant, to shear dependent, to extensional dependent, and blends of both.

To appreciate some of the properties of non-Newtonian fluids under flow, it is useful to consider classical viscometric flow conditions. So, for example, the velocity field $\mathbf{u} = (u, v, w)$ and deformation-rate tensor $\mathbf{d}$ under simple shear deformation are given as:

$$\begin{aligned} u(y) &= \dot{\gamma} y \\ v &= 0 \\ w &= 0 \end{aligned} \qquad \text{and} \qquad \mathbf{d} = \frac{1}{2}\begin{bmatrix} 0 & \dot{\gamma} & 0 \\ \dot{\gamma} & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix}. \qquad (3)$$

Here, other properties that vary between Newtonian and non-Newtonian fluids are the first and second normal stress difference, N_1 and N_2, with respective coefficients, ψ_1 and ψ_2. Their magnitudes are zero for Newtonian fluids, whilst for non-Newtonian

fluids, their definitions are:

$$N_1(\dot{\gamma}) = \sigma_{xx} - \sigma_{yy} = \psi_1(\dot{\gamma})\dot{\gamma}^2,$$
$$N_2(\dot{\gamma}) = \sigma_{yy} - \sigma_{zz} = \psi_2(\dot{\gamma})\dot{\gamma}^2. \quad (4)$$

This normally leads to relations, $N_1 > 0$ and $N_1 >> |N_2|$.

Similarly, the velocity field $\mathbf{u} = (u, v, w)$ and deformation-rate tensor $\mathbf{d}$ in uniaxial extensional flow can be expressed as a function of extension-rate $(\dot{\varepsilon})$ as:

$$u(\mathrm{x}) = \dot{\varepsilon}\,\mathrm{x}$$
$$v(\mathrm{y}) = -\tfrac{1}{2}\dot{\varepsilon}\,\mathrm{y} \quad \text{and} \quad \mathbf{d} = \begin{bmatrix} \dot{\varepsilon} & 0 & 0 \\ 0 & -\frac{1}{2}\dot{\varepsilon} & 0 \\ 0 & 0 & -\frac{1}{2}\dot{\varepsilon} \end{bmatrix}. \quad (5)$$
$$w(\mathrm{z}) = -\tfrac{1}{2}\dot{\varepsilon}\,\mathrm{z}$$

Here, the resistance to flow, or extensional viscosity (η_E), is then expressed through the relationship,

$$\sigma_{xx} - \sigma_{yy} = \sigma_{xx} - \sigma_{zz} = \eta_{\mathrm{E}}(\dot{\varepsilon})\dot{\varepsilon}. \quad (6)$$

For a Newtonian fluid, η_{E} is constant under all strain-rates, and is simply three times the shear viscosity (η_{S}):

$$\eta_{\mathrm{E}} = 3\eta_{\mathrm{S}}. \quad (7)$$

Additionally, all fluids, including those with viscoelastic properties, satisfy relation (7) at low deformation-rates, that is, $\eta_{\mathrm{E}}(\dot{\varepsilon} \to 0) = 3\eta_{\mathrm{S}}(\dot{\gamma} \to 0)$. A further related quantity of interest is the so-called Trouton ratio (Tr), defined as the quotient of extensional and shear viscosities. In order to relate $\dot{\gamma}$ and $\dot{\varepsilon}$ to evaluate shear and extensional viscosities, one may define the Trouton ratio in the form:

$$Tr = \frac{\eta_{\mathrm{E}}(\dot{\varepsilon})}{\eta_{\mathrm{S}}(\dot{\gamma} = \sqrt{3}\,\dot{\varepsilon})}. \quad (8)$$

For inelastic fluids with shear-dependent viscosity, Tr is three for all values of $\dot{\varepsilon}$, and for viscoelastic fluids this ratio is anticipated to satisfy: $Tr(\dot{\varepsilon} \to 0) = 3$. In contraction flows, pure extension takes place along the centerline. In axisymmetric configurations this elongation is uniaxial, whilst in 2D planar flows, such deformation is that of *planar extension*, where the fluid sample is stretched in one direction and compressed in another, leaving one dimension unchanged. Expressions for extensional viscosity and Trouton ratio in planar extension are similar to those for uniaxial flow,

$$
\begin{aligned}
&\eta_{\mathrm{E}} = 4\eta_{\mathrm{S}} \quad (\textit{Newtonian}),\\
&\eta_{\mathrm{E}}\left(\dot{\varepsilon}\to 0\right) = 4\eta_{\mathrm{S}}\left(\dot{\gamma}\to 0\right) (\textit{Viscoelastic}),\\
&Tr = \frac{\eta_{\mathrm{E}}\left(\dot{\varepsilon}\right)}{\eta_{\mathrm{S}}\left(\dot{\gamma}=2\dot{\varepsilon}\right)},\\
&Tr\left(\dot{\varepsilon}\to 0\right) = 4 \qquad (\textit{Viscoelastic}).
\end{aligned}
\tag{9}
$$

Assuming isothermal condition for simplicity, the governing differential equations for viscoelastic flow may be represented through those for mass-conservation and momentum-transport, in conjunction with equations of state for stress and density (compressible flow). As such, the flow equations governing continuity and momentum balance may be expressed as:

$$
\begin{aligned}
&\frac{\partial\rho}{\partial t} + \nabla\cdot\left(\rho\mathbf{u}\right) = 0,\\
&\rho\frac{\partial\mathbf{u}}{\partial t} = \nabla\cdot\left(2\eta_{\mathrm{s}}\,\mathrm{d} + \eta_{\mathrm{s}}\left(\frac{\kappa}{\eta_{\mathrm{s}}} - \frac{2}{3}\right)\left(\nabla\cdot\mathbf{u}\right)\mathbf{I} + \boldsymbol{\tau}\right) - \nabla p
\end{aligned}
\tag{10}
$$

where, ρ, $\mathbf{u}$, and $\boldsymbol{\tau}$ represent density, velocity, and extra-stress, respectively. Viscosity material parameters of η, η_{p} and η_{s}, represent factors of total, polymeric-fraction and solvent-fraction, respectively, where $\eta = \eta_{\mathrm{p}} + \eta_{\mathrm{s}}$. κ is a generalized factor that mimics the role of bulk viscosity. Bulk viscosity arises as a consequence of active rotational and vibrational modes at the polyatomic molecular level, relevant in compressible gas or granular matter flow. For convenience, $\bar{\tau}^{(s)}$ can be introduced as being the augmented solvent stress referenced within the momentum equation,

$$
\bar{\boldsymbol{\tau}}^{(s)} = \left(2\eta_s\,\mathbf{d} + \eta_s\left(\frac{\kappa}{\eta_s} - \frac{2}{3}\right)\left(\nabla\cdot\mathbf{u}\right)\mathbf{I}\right).
\tag{11}
$$

Clearly, considerable simplification results under incompressible flow conditions (constant density). To complete the set of governing equations for compressible flow, it is necessary to introduce an equation of state to relate density to pressure. Here, for example, one may employ amongst others, the modified Tait equation of state a well-established formulation for liquids,

$$
\frac{\tilde{p}+B}{\tilde{p}_0+B} = \left(\frac{\rho}{\rho_0}\right)^m, \text{ with augmented pressure } \tilde{p} = p - \frac{1}{3}\mathrm{tr}\left(\tau + 2\frac{\eta_{\mathrm{s}}}{\eta}\mathrm{d}\right).
\tag{12}
$$

Parameters B and m are constants, and $\tilde{p}_0$, ρ_0 denote reference scales for pressure and density. Note that, this state law is often approximated to a linear form with index m set to unity.

To extend incompressible algorithms to deal with compressible flows, it is important to appreciate the key role that pressure plays in a compressible flow. In the low Mach number (Ma) limit, where density is almost constant, the role of pressure is to influence velocity through the continuity equation, so that conservation of mass is satisfied. Indeed, in this instance, density and pressure are only weakly-linked variables. To recast an incompressible scheme into one appropriate for weakly-compressible highly-viscous flow, one may replace the temporal derivative of density in the continuity equation with its equivalent in pressure, appealing to an equation of state. Assuming isentropic conditions, and employing the differential chain rule, one gathers,

$$\frac{\partial \rho}{\partial t} = \frac{\partial \rho}{\partial p}\frac{\partial p}{\partial t} = \frac{1}{c^2_{(X,t)}}\frac{\partial p}{\partial t},$$
$$\frac{\partial p}{\partial \rho} = \frac{m(\tilde{p}+B)}{\partial \rho} = c^2_{(X,t)}, \qquad (13)$$

where, $c_{(X,t)}$ introduces the speed of sound, a field variable distributed in space and time, often interpreted as a ratio formed with fluid velocity and expressed through the Mach number $Ma = u/c$. This switch of representation between density and pressure time derivatives has a major impact in pressure-correction algorithms governing the continuity-pressure equation (see discussion below).

Using general physical principles, constitutive equations have been derived to relate the state-of-stress to the history-of-deformation in a material. Yet, in order to obtain quantitative agreement with experimental data, realistic models are required that are capable of well representing the rheometrical response of actual fluids. Typical examples would include: Phan-Thien/Tanner (PTT), Giesekus, and the Kaye-Bernstein-Kearsley-Zapas (K-BKZ) models**,** which are phenomenological constitutive equations commonly used to model polymer melt behavior. Characteristically, the Giesekus model in extensional deformation sustains hardening with increase in strain-rates, yet ultimately, reaches a plateau. A modified multi-mode K-BKZ model has also been developed that successfully predicts contraction flows of LDPE melts. Moreover, PTT models can reproduce a variety of rheological responses in both planar and uniaxial extension, though the parameter controlling the degree of extension, ε_{PTT}, also affects shear-viscosity properties. In addition, eXtended Pom-Pom (XPP) models are capable of reproducing the response of polymeric systems in rheometrical flows. These versions are derived from the kinetic-based pom-pom model introduced by McLeish & Larson, based on reptation dynamics of an idealized linear molecule with an equal number of branched arms at both ends.

Here to supply a concrete example, one may employ a generalized formulation to express the relevant constitutive law. The notation encompasses the Single eXtended Pom-Pom (SXPP) model, which requires a set of additional parameters $(\varepsilon, q, \lambda, \alpha)$, selected to represent system entanglement (ε), number of side-branch arms to the backbone segment (q), stretch of the backbone segment (λ), and anisotropy of the

polymeric network (α). With $\beta_s = \eta_s / \eta = \dfrac{\eta_s}{\eta_s + \eta_p}$, such a generalized constitutive equation statement covers a series of models, taking the form:

$$f(\lambda,\boldsymbol{\tau})\boldsymbol{\tau} + Wi\overset{\nabla}{\boldsymbol{\tau}} + \frac{(1-\beta_s)}{Wi}\left[g(\lambda,\boldsymbol{\tau})\right]\mathbf{I} + \frac{\alpha Wi}{(1-\beta_s)}\boldsymbol{\tau}\cdot\boldsymbol{\tau} = 2(1-\beta_s)\mathbf{d}. \qquad (14)$$

Here, **I** is the unit tensor, $\overset{\nabla}{\boldsymbol{\tau}}$ represents the upper-convected material derivative of $\boldsymbol{\tau}$, and the convected derivative is expressed as

$$\overset{\nabla}{\boldsymbol{\tau}} = \frac{\partial \boldsymbol{\tau}}{\partial t} + \mathbf{u}.\nabla\boldsymbol{\tau} - (\nabla\mathbf{u})^{\dagger}.\boldsymbol{\tau} + \boldsymbol{\tau}.(\nabla\mathbf{u}). \qquad (15)$$

The generalized functions $f(\lambda,\boldsymbol{\tau})$ and $g(\lambda,\boldsymbol{\tau})$ categorizing the choice over models. Table 1 provides the classification through functions $\{f,g\}(\lambda,\boldsymbol{\tau})$ and setting of (α)-parameter to distinguish between Oldroyd-B, PTT and SXPP fluids.

Model	$f(\lambda,\tau)$	$g(\lambda,\tau)$	α
Oldroyd $-$ B(τ)	1	0	0
LPTT(τ)	$1+\dfrac{\varepsilon_{PTT} Wi}{(1-\beta_s)}\mathrm{tr}(\tau)$	0	0
EPTT$(\boldsymbol{\tau})$	$\exp[\dfrac{\varepsilon_{PTT} Wi}{(1-\beta_s)}\mathrm{tr}(\tau)]$	0	0
SXPP(τ)	$f(\lambda,\tau)$	$f(\lambda,\tau)-1$	α

Table 1. Classification of functions; $f(\lambda,\boldsymbol{\tau})$, $g(\lambda,\boldsymbol{\tau})$

The definition of function $f(\lambda,\boldsymbol{\tau})$ is all important to this class of models, being

$$f(\lambda,\boldsymbol{\tau}) = \frac{2}{\varepsilon}\left(1-\frac{1}{\lambda}\right)e^{\nu(\lambda-1)} + \frac{1}{\lambda^2}\left(1-\left\{\frac{Wi}{(1-\beta_s)}\right\}^2\frac{\alpha}{3}\mathrm{tr}(\boldsymbol{\tau}\cdot\boldsymbol{\tau})\right). \qquad (16)$$

In the expression for $f(\lambda,\boldsymbol{\tau})$, the free parameter ν is estimated by data-fitting and found to be inversely proportional to the number of side-branch arms to the molecular chain-segment $(\nu = 2/q)$. With the single-equation form of the pom-pom model, the backbone stretch parameter λ is identified through an algebraic expression,

$$\lambda = \sqrt{1+\frac{1}{3}\frac{Wi}{(1-\beta_s)}\left|\mathrm{tr}(\boldsymbol{\tau})\right|}. \qquad (17)$$

Essentially, this formulation collapses the double-equation version of the pom-pom model (DXPP), where a differential equation dictates the change in $\lambda(t)$. The pom-pom model has been derived to characterize the rheological behavior of polymer melts with long side-branches, such as applicable for low density polyethylenes. In addition, this model possesses features of the Giesekus model since a non-zero second normal stress-difference is predicted when anisotropy parameter $\alpha \neq 0$. The Oldroyd-B model corresponds to setting $f(\lambda, \boldsymbol{\tau}) = 1$ and, $\alpha = 0$. Then, the conventional definitions of group numbers, in Reynolds (*Re*) and Weissenberg (*Wi*) numbers, with pom-pom parameters β_s and ε, are defined as:

$$Re = \frac{\rho U L_{\text{scale}}}{\eta}, \quad Wi = \frac{\tau_b U}{L_{\text{scale}}}, \quad \beta_s = \frac{\eta_s}{\eta_s + G_0 \tau_b}, \quad \varepsilon = \frac{\tau_s}{\tau_b}. \tag{18}$$

In this notational form, τ_b and τ_s represent the backbone orientation and stretch relaxation time-scales, respectively. G_0 is the linear relaxation modulus and the parameter ε is the ratio of stretch to orientation relaxation times. Typically, re choice of scales and for contraction flows in particular, the characteristic velocity (*U*) and length (L_{scale}) scales are frequently taken as the downstream mean velocity and channel half-height, respectively. For simplicity in the case of pom-pom models, η_p may be defined as $\eta_p = G_0 \tau_b$ to preserve similarity between the versions of Oldroyd-B and SXPP models.

In contrast to the above instantaneous differential view, integral constitutive models provide an alternative description of non-Newtonian elastico-viscous response, that incorporate the past history of deformation. In this formulation, the Cauchy stress σ_{ik} may be represented as,

$$\sigma_{ik} = -p\delta_{ik} + \int_{-\infty}^{t} h(t - t')\, g(d_{ik}(t', t))\, dt' \tag{19}$$

where t and t' are present and past times of the fluid element flowing along its trajectory, h is the memory function of linear viscoelsticity, and *g* is a model-dependent non-linear strain measure. Such an approach gives rise to integro-differential systems to solve, necessitating integral evaluation along fluid element trajectories throughout the past history of deformation. Each of these approaches has its merits and drawbacks – preference is given henceforth to the differential view, that is pursued below.

-
-
-

Bibliography

Aguayo J. P. (2006). Prediction of viscoelastic fluid flow in contractions, PhD thesis, Swansea University, Computer Science Department, http://137.44.2.180/21.FiniteVolume/Thesis_Aguayo/ HTML+Thesis/ AguayoThesismain.html. [Work covers excess pressure-drop, pom-pom modeling, and Bautista-Manero models in contraction flows].

Baaijens F.P.T. (1998). Mixed finite element methods for viscoelastic flow analysis: a review, *J. Non-Newtonian Fluid Mech.* **79**, 361-385. [Research review paper describes the algorithmic developments for the classical mixed formulation, the elastic viscous stress splitting (EVSS) method as well as the related discrete variants].

Binding D.M., Phillips P.M., and Phillips T.N. (2006). Contraction-expansion flows: The pressure drop and related issues, *J. Non-Newtonian Fluid Mech.* **137**, 31-38. [Paper concerned primarily with the numerical prediction of the pressure-drop associated with the flow of an Oldroyd-B fluid through 4:1 contractions, 1:4 expansions and combined 4:1:4 contraction/expansions].

Crochet M.J., Davies A.R. and Walters K. (1984). *Numerical Simulation of non-Newtonian Flow*, Elsevier, Amsterdam. [First book to appear on numerical methods for viscoelastic flow].

Crochet M.J. and Walters K. (1993). Computational rheology: a new science, *Endeavour* **17**, 64-77. [Review of progress relating to experimental and theoretical advances in the mechanics of non-Newtonian fluids, with particular reference to extrusion processes].

Denn M.M. (2004). Fifty years of non-Newtonian fluid dynamics, *AIChE J.* **50**, 2335-2345. [Overview on the classes of flows in which inherent nonlinearity causes non-Newtonian fluids to exhibit qualitatively different behavior from Newtonian fluids].

Keunings R. (1990). Progress and challenges in computational rheology, *Rheol. Acta* **29**, 556-570. [Illustration of the basic issues associated with the numerical prediction of viscoelastic complex flows. Numerical challenges are highlighted in flow behavior close to walls and singularities].

Keunings R. (2000). A survey of computational rheology, *Edited by Binding et al., 13th International Congress on Rheology, BSR*, 7-14. [Survey outlines the modeling approaches adopted in numerical simulation and reviews developments in macroscopic and micro-macro simulation strategies].

Keunings R. (2003). Finite element methods for integral viscoelastic fluids, *Edited by Binding et al., BSR*, 167-195. [Mathematical formulation and numerical approaches employed for solving complex flows of viscoelastic fluids described by a constitutive model of the integral type. A brief review of non-trivial flow problems is included].

Keunings R. (2004). Micro-Macro Methods for the Multiscale Simulation of Viscoelastic Flow using Molecular Models of Kinetic Theory. In: Binding et al. (Ed.), Rheology Review, 67-98. [A survey of the field of micro-macro numerical techniques for predicting complex flows of viscoelastic fluids. The focus is mainly on mathematical formulation and numerical approaches with a review of applications to polymer solutions and melts, liquid crystalline polymers and fiber suspensions].

Larson R.G. (1988). *Constitutive Equations for Polymer Melts and Solutions*, Series in Chemical Engineering, Butterworths, Boston. [An introduction and guide to constitutive equations, and to molecular and phenomenological theories of polymer flow].

McLeish T.C.B. and Larson R.G. (1998). Molecular constitutive equations for a class of branched polymers: The pom-pom polymer, *J. Rheology* **42**, 81-110. [Paper that proposed the differential/integral

molecular constitutive equation for polymers melts with long-chain side branches (pom-pom). The model predicts both shear and extension qualitatively more accurately than K-BKZ model].

Nigen S. and Walters K. (2002). Viscoelastic contraction flows: comparison of axisymmetric and planar configurations, *J. Non-Newtonian Fluid Mech.* **102** 343-359. [An experimental study, which investigates the behavior of two constant-viscosity polymer solutions (Boger fluids) in both axisymmetric and planar contractions].

Rodd L.E., Scott T.P., Boger D.V., Cooper-White J.J. and McKinley G.H. (2005). The inertio-elastic planar entry flow of low-viscosity elastic fluids in micro-fabricated geometries, *J. Non-Newtonian Fluid Mech.* **129**, 1-22. [A study of length-scale effects in micro-fabricated geometries - strong viscoelastic effects, inertia versus elasticity. The paper includes a brief review of viscoelastic entry flow phenomena based on macro-scale geometries].

Rothstein J.P. and McKinley G.H. (2001). The axisymmetric contraction-expansion: the role of extensional rheology on vortex growth dynamics and the enhanced pressure drop, *J. Non-Newtonian Fluid Mech.* **98** 33-63. [Investigates experimentally the creeping flow of Boger fluids in axisymmetric contraction-expansion flows for various contraction ratios covering a large range of *De* and varying degrees of re-entrant corner curvature].

Szabo P., Rallison J.M. and Hinch E.J. (1997). Start-up of flow of a FENE-fluid through a 4:1:4 constriction in a tube, *J. Non-Newtonian Fluid Mech.* **72** 73-86. [FENE-fluid flow through a tube is computed by a split Lagrangian-Eulerian finite element method, with a proposed model for upstream vortex growth dependent upon elasticity].

Tanner R.I. (2000). *Engineering Rheology*, Oxford University Press. [Study on the behavior of non-Newtonian materials in engineering, with a review on the rapid growth and change in the subject of rheology].

Tanner R.I. and Walters K. (1998). *Rheology: An Historical Perspective*, Elsevier, Oxford. [This book contains an in-depth study of the history of rheology, including an overview of various constitutive equations, some distinctive rheological concepts and phenomena, and a background for computational rheology].

Walters K., Tanner R.I. (1992). The motion of a sphere through an elastic fluid. In: Chhabra RP, DeKee D (eds) Transport processes in bubbles, drops and particles, Hemisphere, New York, pp 73–86. [A benchmark problem for computational rheology and the evaluation of numerical codes, the sedimentation of a sphere in an elastic fluid].

Walters K. and Webster M.F. (2003). The distinctive CFD challenges of computational rheology, *Int. J. Num. Meth. Fluids* **43**, 577-596. [Research outlines major historical developments and numerical challenges in the field of computational rheology with the emphasis on contraction flow problems].

White S.A., Gotsis A.D. and Baird D.G. (1987). Review of the entry flow problem: Experimental and numerical, *J. Non-Newtonian Fluid Mech.* **24** 121-160. [Review for numerical and experimental studies of axisymmetric and planar entry flows; analyzes prediction of vortices and accuracy of numerical predictions].

Biographical Sketches

Professor Webster's formative education was conducted through a BSc. degree in mathematics (1974) at University of Manchester, followed by an MSc. degree in numerical analysis (1976). His early research career was developed under Professors Walters (FRS) and Davies at University of Wales Aberystwyth, culminating in a Doctorate (1979) in applied mathematics on the numerical simulation of rheological flows, followed by a postdoctoral appointment. In 1982, he travelled to Australia to work with Professor Tanner as a research fellow at the University of Sydney. On returning to the UK (1983), he took a senior research officer appointment in the Institute of Computational Fluid Dynamics at University of Reading, where he worked with Professor Morton and Dr Baines. In 1986, Professor Webster took a full-time lectureship at University of Wales Swansea, collaborating with Professor Townsend on the numerical simulation of non-Newtonian flows and forming the Computational Rheology and Flow Visualisation research group in the Department of Computer Science. At Swansea, Professor Webster was promoted to

Senior Lecturer (1991) and to Reader (1995).

Professor MF Webster, AFIMA, currently holds a personal chair in School of Engineering at Swansea University (from 1999), ranked grade5, RAE 2001. Professor Webster has built up over thirty years experience in the fields of computational rheology, numerical analysis and computational fluid dynamics, which has been extended more recently into the domains of parallel processing and multimedia studies. He has published over 140 articles in leading international journals in these areas, leading to key advances in predictive algorithms for viscoelastic flows – industrial and otherwise. He has received funding from a wide range of sources including the funding Councils (EPSRC, BBSRC, HEFCW), the European Union and industry. Industrial collaboration has been undertaken with companies such as Philips, DSM, BICC, EA Technology, Alcoa, Unilever, Schlumberger, RHM Research, United Biscuits, Pillsbury, Mono Equipment and SASIB Bakeries.

Professor Webster is the current Treasurer on the European Society of Rheology executive committee (ESR, 1 of 5, EU elected officers), executive member of the British Society of Rheology (BSR, nationally elected, previous Treasurer/Membership Secretary), and a current Director of Institute of non-Newtonian Fluid Mechanics (INNFM Wales). Through conference organisation (INNFM annual symposia, AERC and IRC), research contracts, joint publications, and doctoral supervision (Thailand, Iran, Iraq, Pakistan, Mexico), Professor Webster has international collaborative links with many fellow academics/industrialists worldwide (EU, USA, Australia, Mexico and Korea).

Hamid Reza Tamaddon Jahromi graduated with a BSc in Planning from School of Planning and Computer Applications, Iran and a MSc in Computer Science from California Polytechnic State University, USA. He received his PhD degree in Computer Science from Swansea University (1992), UK. He took a position as an assistant professor at Iran University of Science and Technology from 1992 to 2000. He then joined the Computational Rheology and Flow Visualization research group in Swansea University. His research interests lie in areas of: algorithms, computer graphics, parallel processing, scientific computation, and science of rheology.

Fawzi Belblidia graduated with a BSc (1987) and a MSc (1990) in Mechanical Engineering department from Polytechnic National School (ENP) – Algiers (Algeria). He held a position of assistant lecturer, and then, lecturer in ENP (from 1987 to 1997), teaching numerical methods in engineering and applications of the finite element method. He received his PhD degree in Civil Engineering department from Swansea University (SU) in 1999 – UK on structural optimization of plates and shells, under the supervision of late Prof. E. Hinton (1946-1999), founder of ASMO UK. He joined the research group of Prof. Hinton FIDO-ADOPT as a research officer from 1999 to 2000. Then, he joined the Polysim research group (2000-2002) to work primarily on advanced injection molding technologies for plastics: computer simulation with experimental validation. From 2002 to date, he joined Prof. M.F. Webster and the Computational Rheology and Flow Visualization research group, working extensively and gaining first-hand knowledge of the *fe/fv*-Swansea algorithms, particularly concerning viscoelastic-compressible computations, algorithmic variations, thermal considerations and problem singularities.