

## A SURVEY OF COMPUTATIONAL RHEOLOGY

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### ABSTRACT

A survey is presented of the field of computational rheology applied to the analysis of viscoelastic effects in complex flows of polymeric fluids. First, I outline the modelling approaches adopted currently in numerical simulations and discuss the role of computational rheology within the general study of structured liquids. Developments in the macroscopic and micro-macro simulation strategies are then reviewed. Finally, I stress important unsolved problems and offer suggestions for future work.

KEYWORDS: NUMERICAL SIMULATION, COMPLEX FLOWS, VISCOELASTIC EFFECTS, POLYMERIC LIQUIDS, CONSTITUTIVE EQUATIONS, KINETIC THEORY.

### INTRODUCTION

Computational rheology involves the design, implementation, and use of numerical methods for the computer simulation of the flow of non-Newtonian fluids in complex geometries. I shall focus here on the numerical prediction of viscoelastic effects in polymeric liquids. Under development over the last 25 years within a lively and evolving international community of research teams (see [1-6] for reviews), this fascinating but difficult field of investigation has now reached a state of relative maturity. It is thus fitting to take stock of its current condition.

### COMPLEMENTARY THEORETICAL APPROACHES

#### Rheology and structure

Polymer solutions and melts, like other rheologically-complex fluids, exhibit a variety of non-Newtonian flow properties. In particular, polymeric liquids are viscoelastic materials, which implies that the stress endured by a fluid element depends upon the history of the deformation experienced by that element. Viscoelastic properties are responsible for numerous flow phenomena of scientific and industrial relevance [7] that need to be *predicted*, *understood*, and possibly *controlled* by means of a combination of suitable physical models and numerical techniques.

The rheological properties of non-Newtonian fluids are dictated by the flow-induced evolution of their internal *microstructure* [8]. In flowing polymers, the relevant microstructure is the conformation of the macromolecules, namely their orientation and degree of stretch. Each macroscopic fluid element contains a large number of polymers with a *statistical distribution* of conformations. While the flow alters the polymer conformations along the fluid trajectories, the macroscopic stress carried by each material element is itself governed by the distribution of

polymer conformations within that element. Furthermore, the frozen-in microstructure which develops in processing flows governs the physical properties of the final product. Rheologists thus face a challenging *non-linear coupling* between rheological behaviour, flow-induced evolution of the microstructure, flow parameters (such as geometry and boundary conditions), and final product properties. Computational rheology has an important role to play in helping us to elucidate this coupling.

#### Atomistic modelling

Modern research in computational materials science exploits, sometimes in a coupled fashion, a hierarchy of theoretical models ranging from quantum mechanics to continuum mechanics. The difficulty with polymers is of course the gigantic number of microstructural degrees of freedom and the broad range of time and length scales separating the relevant atomistic and macroscopic processes. A modelling approach based on quantum mechanics and related *ab initio* computational techniques must clearly be ruled out at the outset. Indeed, *atomistic modelling* is probably the most detailed level of description that could realistically be applied in rheological studies [9]; techniques of *non-equilibrium molecular dynamics* constitute the associated numerical framework [10-12].

Atomistic flow simulations have been conducted recently [13-15] to study the behaviour of polymers near walls and geometrical singularities such as re-entrant corners. In view of the significant computer resources involved in such calculations, the atomistic approach is currently restricted to very coarse models for the polymer and to flow geometries of molecular dimensions. While I doubt that it may be feasible soon (and even necessary, for that matter!) to solve flow problems of macroscopic size using the atomistic approach, the potential of atomistic simulations is great, however, in helping us resolve difficult issues such as wall slip.

#### Kinetic theory

The next level of description of a polymeric liquid is that of *kinetic theory*. Here, one ignores atomistic processes altogether and focuses rather on the evolution of a more or less coarse-grained model of the polymer conformations [16]. Kinetic theory models for polymer solutions or melts are most naturally exploited numerically by means of *stochastic simulation* or *Brownian dynamics* methods [17].

Like atomistic modelling, kinetic theory offers several levels of description of a given fluid [16]. A dilute solution of linear polymers in a Newtonian solvent, for example, can be described in some detail by a freely jointed, bead-rod *Kramers chain*, which is made of a number of beads (of order 100) connected linearly by rigid segments. A coarser model of the same polymer is the freely jointed

bead-spring chain, made of a smaller number of beads (of order 10) connected linearly by entropic springs. Finally, a coarser model still is the single dumbbell, namely two beads connected by a spring. Clearly, these models of kinetic theory are not meant to describe the chemical structure of the polymer. They do, however, display in a more or less detailed fashion the important features needed to describe the evolution of polymer conformations in a macroscopic flow. Recent Brownian dynamics studies of rheometrical flows of Boger fluids using Kramers chains [18-20], bead-spring chains [21-23], and dumbbells [24-25], have shed new light on the validity of available kinetic theory models for dilute polymer solutions. In combination with the direct experimental observation of the flow-induced conformations of single polymers [26-28], these numerical studies have significantly advanced our understanding of polymer dynamics.

The most successful kinetic theory for concentrated solutions or melts of linear polymers is based on the reptation model proposed by de Gennes and further developed by Doi and Edwards [29]. The Doi-Edwards theory cannot be used as such for simulating complex flows as it predicts a non-monotonic dependence of the shear stress as a function of shear rate. Important modifications have been proposed recently which correct this and other deficiencies of the basic theory [30-34]. Even more detailed reptation models suited for stochastic simulations have become available [35], and significant progress has also been made in extending the theory to branched polymers [36]. One has thus reached the point where reptation-based models can be exploited in the numerical prediction of complex flows.

Use of a model of kinetic theory in the numerical simulation of complex flows, in combination with the macroscopic conservation laws, constitutes the so-called *micro-macro approach* to computational rheology. This is an emerging methodology which I shall discuss later.

#### Continuum mechanics

Finally, besides atomistic modelling and kinetic theory lies the macroscopic approach of *continuum mechanics* [37]. Here, details of the fluid microstructure are not taken into account, at least explicitly. Rather, the stress experienced by the macroscopic fluid elements is related to the deformation history through a suitable *constitutive equation*. Combined with the conservation laws, the constitutive model yields a set of partial differential (or integro-differential) equations that can be solved by means of a suitable grid-based numerical method, such as the finite element technique [38]. A very large percentage of publications in computational rheology have been based on the macroscopic approach. Very much like atomistic modelling and kinetic theory, continuum mechanics offers a hierarchy of constitutive models of diverse predictive ability.

It should be stressed that most constitutive equations used today in numerical work [39] have been *derived* from a molecular model of kinetic theory (a recent addition to the list is the "pom-pom" constitutive equation for

branched polymers [40-41]). Such molecular-based constitutive equations yield quantitative information on the distribution of polymer conformations within a macroscopic fluid element in the form of *averaged quantities* such as the second moment of the distribution of conformations. Unfortunately, closure approximations of a purely mathematical nature are often needed in the derivation of a constitutive model from kinetic theory, and their impact can be significant indeed [42]. In view of the closure problem, the link with the parent molecular model is thus somewhat polluted, and interpretation of the macroscopic results in molecular terms becomes delicate.

Finally, I wish to close this section on modelling by pointing to recent developments in non-equilibrium thermodynamics of complex fluids [43-45]. These should provide guidance in linking the various levels of description of polymeric liquids that I have briefly discussed. They should also help in the development of improved models [46-47].

## RHEOLOGY AND COMPUTATION

### Role of computational rheology

The viscoelastic character of a given flow is often measured by the dimensionless Weissenberg number  $We$ , defined as the product of a characteristic relaxation time of the fluid and a characteristic deformation rate of the flow. While  $We$  vanishes for Newtonian fluids, it is of order 1 or 10 in the polymer flows of interest here. The challenge for computational rheologists is to develop numerical schemes for obtaining *accurate* numerical solutions to the governing equations at values of  $We$  of practical interest, using a *physically-realistic* mathematical model. Let us consider for a while the (ideal) situation in which reliable computational rheology software would be readily available and usable by non-specialists on standard computer hardware, and let us imagine some of the numerous opportunities.

For instance, the theoretical rheologist would have a tool at his or her disposal for the critical evaluation in *complex flows* of his or her new theory, whether it be a constitutive equation, a molecular model, or an improved description of boundary conditions. Complex flows are characterized by *transient* kinematics in the Lagrangian sense (i.e. following the fluid elements) and by a combination of shear and elongational deformations; they indeed constitute a tough testing ground for any theoretical model. The theoretical rheologist could also use tools of computational rheology to bridge the gaps between the several levels of description used to predict the polymer dynamics. One example of such a difficult exercise is the evaluation in complex flows of suitable closure approximations that are invariably needed to derive a macroscopic constitutive equation from a molecular model. Also, non-Newtonian fluid mechanics studies could be performed to understand better the mechanisms responsible for observed macroscopic flow phenomena (such as vortex growth [48-49] and purely-elastic instabilities [50-52]).

The experimentalist would also benefit. For example,

he or she could perform useful *computational rheometry* work wherein the experiment is actually simulated in order to better interpret the data, i.e. to translate better what is measured (such as forces, torques, deformations) into well-defined rheological information. Experimental problems (such as flow inhomogeneities and secondary motions) would thus be identified, possibly eliminated through improved design of the rheometrical equipment, or explicitly taken into account in the data reduction phase. Also, the combination of flow experiments and numerical simulations could be used to characterize rheological behaviour in complex flows, yielding in a more or less automated way the optimal rheological model and material parameters for the fluid under investigation.

Finally, and most importantly in industrial practice, the polymer engineer could perform elaborate Computer Aided Design studies in which the link between the molecular architecture of the raw material and the final properties of the product would be established, at least qualitatively. Production problems (such as extrusion instabilities [53]) would be predicted and partially overcome through improved design. One could also think of using an on-line computational rheology model in concert with appropriate control algorithms to provide for intelligent, physics-based process control techniques.

These are but a few opportunities that a mature field of computational rheology could generate. Not surprisingly, I would argue that we remain far from this ideal situation. Nevertheless, significant collective progress has been made over the years to the extent that the above virtual picture may not be as distant as would appear.

#### Numerical approaches

Since its pioneering days (circa 1975), computational rheology has adopted the macroscopic approach. Recently, computer processing capacity has reached a level such as to make feasible the complementary micro-macro approach, which involves the coupled solution of the conservation laws and a microscopic model of kinetic theory. In the next two sections, I briefly review key developments in these two lines of research. For completeness, I wish to point out that alternative approaches to the computer modelling of polymeric liquids have been advanced very recently, most notably Dissipative particle dynamics [54] and Lattice Boltzmann models [55-56].

### MACROSCOPIC SIMULATIONS

#### Governing equations and numerical challenges

Let us consider for the sake of illustration the case of incompressible isothermal flow in a confined geometry, in the absence of body forces. The Cauchy stress tensor is thus given by

$$\mathbf{s} = -p\mathbf{d} + \mathbf{t} + 2\mathbf{h}\mathbf{D}, \quad (1)$$

where  $p$  is the pressure,  $\mathbf{t}$  is the polymer contribution to the stress, and  $2\mathbf{h}\mathbf{D}$  is an optional purely-viscous component, which involves the rate of strain tensor  $\mathbf{D}$  and a viscosity coefficient  $\mathbf{h}$ . Conservation of linear momentum and mass implies that

$$\tilde{\mathbf{N}} \cdot \mathbf{s} = \rho D\mathbf{v}/Dt, \quad \tilde{\mathbf{N}} \cdot \mathbf{v} = 0, \quad (2)$$

where  $\mathbf{v}$  is the velocity field and  $D\mathbf{v}/Dt$  is its Lagrangian or material derivative. In macroscopic simulations, the set of governing equations is closed with a suitable constitutive equation for the polymer stress.

Over the years, both *differential* and *single-integral models* have been used in numerical work, ranging from the upper-convected Maxwell (UCM) and Lodge models, to more realistic equations like the FENE-CR, Giesekus, Phan-Thien-Tanner, Doi-Edwards, K-BKZ, and "pom-pom" models. Although they differ greatly in complexity as well as in predictive ability, these various models display a (deceptively) simple generic form. Indeed, differential models read

$$Dt/Dt = \mathbf{f}(t, \tilde{\mathbf{N}}\mathbf{v}), \quad (3)$$

meaning that the Lagrangian derivative of the polymer stress is given as a model-dependent function  $\mathbf{f}$  of the local polymer stress and velocity gradient evaluated along the fluid trajectories. For the particular case of steady-state flow (in the Eulerian sense), the left-hand side of (3) reduces to the convective term  $\mathbf{v} \cdot \nabla \mathbf{t}$ . In a complex flow, the fluid trajectories along which (3) applies are of course unknown *a priori*, and one must solve the complete set of equations (1-3). If direct coupling between modes is neglected, a spectrum of relaxation times is readily taken into account by defining the polymer stress as a sum of partial contributions and writing equation (3) for each partial stress. Note that constitutive equations derived recently from kinetic theory of solutions or melts [40-41,57-58] give the polymer stress as an algebraic function of a number of microstructural tensors, which themselves follow an evolution equation similar to (3). In computational work, it has thus become natural to select the microstructural tensors as primary variables, instead of the polymer stress. The basic numerical issues remain identical, however.

Separable single-integral models have the generic form

$$\mathbf{t} = \int_{-\infty}^t m(t-t') \mathbf{S}(t,t') dt', \quad (4)$$

where  $m$  is the memory function of linear viscoelasticity, and  $\mathbf{S}$  is a model-dependent non-linear strain measure relative to the present time  $t$ . Here also, the memory integral is taken along fluid trajectories that are *a priori* unknown. A spectrum of relaxation times is readily introduced through the memory function.

The generic constitutive equations (3) and (4) clearly express the *memory* of polymeric liquids: the polymer stress carried by a fluid element at present (Lagrangian) time  $t$  is a function of the deformation history experienced at past times  $t'$  by the element flowing along its trajectory.

Although they may look rather innocuous, the macroscopic governing equations, supplemented with suitable boundary and initial conditions, actually present formidable mathematical and numerical challenges which certainly were not anticipated 25 years ago by the pioneers of the field. Through a combination of numerical

experiments and mathematical investigations, our collective understanding of the basic reasons for these difficulties progressively developed during the 1980's [1-4]. In a nutshell, we now know the following: the governing equations are of mixed mathematical type (elliptic-hyperbolic), with possible local changes of type [59]; stress boundary layers develop in many flow fields where the corresponding Newtonian fluid mechanical problem is smooth [60-62]; stress singularities (e.g. at re-entrant corners) are much stronger than in the Newtonian case [63-65]; the non-linear qualitative behaviour of the exact solutions can be very rich (e.g. multiplicity of solutions, bifurcations) [66-68]. These features remain quite difficult to handle numerically in an accurate and general way. Whether they reflect the actual physics of polymeric liquids (in which case we have to live with them!) or result from inadequate modelling (governing equations and/or boundary conditions) remains essentially an open issue.

#### Numerical methods and applications

Classical CFD technology offers a wide spectrum of numerical algorithms based on finite element, finite volume, finite difference, boundary integral, spectral methods, and combinations thereof. A similar diversity exists in modern computational rheology, whose fair discussion would require a rather thick monograph indeed. Here, I wish to point to methods and applications that define in my opinion the current state of the art. The reader will not find here my personal list of "best-buys". Indeed, it has been my experience over the years that a particular numerical scheme (very much like a particular constitutive model) enjoys a range of application where it behaves at its best, and perhaps better than other competing techniques, while it may be quite limited or even totally useless in other situations. Practitioners of computational rheology also know that the only way to really evaluate and compare different numerical methods is to implement and test them all for oneself.

This being said, it is fair to state that most of the published work deals with *finite element* methods for solving 2d steady-state flows (in the Eulerian sense) using a differential constitutive equation [5,69-70]. Extensions of these methods have been proposed recently for the temporal stability analysis of complex flows [71-73], and the computation of transient problems [74-80]. Over the last few years, *finite volume* methods have also been actively developed, sometimes in combination with finite elements, to solve 2d and 3d time-dependent problems [81-85].

Methods for integral models have progressed considerably over the last decade, most notably with the introduction of Lagrangian finite element schemes [86] (wherein the mesh deforms with the fluid) and the Deformation field method [87-88] (which uses a fixed Eulerian grid). These techniques open the way for 2d and 3d transient simulations with integral models.

More specific methods for high-Reynolds number viscoelastic flows, based on *spectral* and *finite difference*

schemes and designed for simple geometries, have also been developed recently [89-91], to study in particular the drag-reduction phenomenon.

Finally, efforts have been spent on the algorithmic side, for example with the development of specific iterative solvers [92] and parallel algorithms [93-96].

In view of the progress made in numerical technology, macroscopic simulations have been exploited over the last few years in two important tasks, namely the evaluation of constitutive equations for solutions and melts in benchmark complex flows, usually through a detailed comparison with experimental observations [97-109], and *computational rheometry*, or use of numerical simulation to aid the experimentalist in reducing his or her data [110-113]. It is in such studies that computational rheology has already proven *useful*, in spite of its relatively young age.

I now turn to the emerging and complementary micro-macro approach.

### MICRO-MACRO SIMULATIONS

#### Governing equations

In micro-macro simulations, the conservation equations (2) are solved by means of a grid-based numerical method (e.g. finite elements), while a kinetic theory model is used to evaluate the polymer contribution to the stress. Clearly, this approach is much more demanding in computer resources than macroscopic methods. On the other hand, it allows the direct use of kinetic theory models in complex flows, without having to resort to closure approximations which often are of doubtful validity.

In principle (but not necessarily in practice), the polymer stress is evaluated at each material point by solving the *diffusion* or *Fokker-Planck* equation for the probability density  $\mathbf{y}(\mathbf{X},t)$  of the conformation  $\mathbf{X}$  of the polymer chains [16-17]. Here, the symbol  $\mathbf{X}$  denotes the set of variables defining the coarse-grained microstructure. For example, it reduces to the vector connecting the two Brownian beads in the simple dumbbell model of a dilute polymer solution.

The diffusion equation dictates the evolution of the distribution function. It has the generic form

$$\mathcal{D}\mathbf{y} / \mathcal{D}t = -\tilde{\mathcal{N}}'\cdot\{\mathbf{A}(\mathbf{X},t)\mathbf{y}\} + \tilde{\mathcal{N}}'\tilde{\mathcal{N}}'\cdot\{\mathbf{D}(\mathbf{X},t)\mathbf{y}\}. \quad (5)$$

Here, the symbol  $\tilde{\mathcal{N}}'$  denotes the del operator with respect to the configuration  $\mathbf{X}$  and the factors  $\mathbf{A}$  and  $\mathbf{D}$  define the deterministic and stochastic components of the model, respectively. In particular, the macroscopic velocity gradient  $\tilde{\mathcal{N}}'\mathbf{v}$  enters in the formulation of  $\mathbf{A}$ , while diffusion phenomena associated with Brownian motion are described in  $\mathbf{D}$ . Equation (5), supplemented with suitable initial and boundary conditions in conformation space, allows the computation of the probability density  $\mathbf{y}(\mathbf{X},t)$ . Relevant macroscopic observables (such as the polymer contribution to the stress tensor) are then defined as *statistical averages* of some known function of the polymer conformation  $\mathbf{X}$ . In a complex flow, the time derivative of  $\mathbf{y}$  in (5) becomes a material derivative and one must, in principle again, solve

(5) at each material point of the flow domain.

An early micro-macro method [114] was based on the numerical solution of the Fokker-Planck equation (5). This approach, however, is limited to kinetic theory models with a conformation space of small dimension. Brownian dynamics or *stochastic simulation* techniques provide a powerful alternative [17]. They draw on the mathematical equivalence between the Fokker-Planck equation (5) and the following Itô stochastic differential equation

$$d\mathbf{X} = \mathbf{A}(\mathbf{X}, t) dt + \mathbf{B}(\mathbf{X}, t) \times d\mathbf{W}, \quad (6)$$

where  $2\mathbf{D} = \mathbf{B}\mathbf{B}^T$  and  $\mathbf{W}$  is a Wiener stochastic process, namely an idealization of Brownian motion. Thus, instead of solving the deterministic diffusion equation (5) for the distribution function, one solves the associated stochastic differential equation (6) by means of suitable numerical techniques, which is often a considerably simpler task. Macroscopic fields of interest are then obtained by averaging over a large *ensemble* of realizations of the stochastic process  $\mathbf{X}$ . In a complex flow, the stochastic differential equation (6) applies along the macroscopic flow trajectories, and the time derivative becomes a material derivative.

#### Numerical methods and applications

The basic idea of combining a stochastic simulation of a kinetic theory model with the numerical solution of the conservation equations has been pioneered in the so-called CONNFESSIT method proposed in [115] and further developed in [116-120].

Second-generation micro-macro methods, with much improved numerical properties, have been designed recently for computing 2d transient flows. They are referred to as Brownian configuration field [121-122] and Lagrangian particle [78-80] methods. Being quite new, these techniques have only been implemented to date for rather elementary models of kinetic theory (such as non-linear dumbbell models of a dilute solution), and not much experience has been gained with them yet. Nevertheless, their potential range of applications is very wide indeed.

#### DISCUSSION

##### From numbers to rheology

From a user's viewpoint, any serious exercise in computational rheology should in my opinion follow a *three-step path*: (1) get the numbers, (2) check their numerical accuracy, and (3) check their physical accuracy.

At the outset, step (1) was found to be by no means a trivial matter: obtaining numerical solutions of the discrete, non-linear algebraic equations at significant values of the Weissenberg number  $We$  has long been difficult or even impossible (this is known as the High Weissenberg Number Problem or HWNP, which is discussed in detail in [3]). Though overlooked at times, the importance of step (2) is simply paramount. One must indeed convince oneself and others that the numerical results obtained constitute sufficiently accurate approximations of the exact solutions of the governing equations. Since the mathematical analysis of numerical methods for viscoelastic fluids is quite difficult and of limited applicability to complex flows [123-124], step (2) is usually performed by means of

careful (and often expensive) mesh refinement experiments. Finally, step (3) questions the very validity of the physical model that is implemented in the computer simulation (constitutive equation or molecular model, values of the material parameters, initial and boundary conditions). It is performed by comparing simulation results (such as flow kinematics, overall pressure drop, and birefringence) to available experimental observations.

Since the pioneering days, overall progress along this three-step path has been sometimes chaotic but overall quite steady [1-6]. Difficulties do remain, however, some of which I now wish to discuss briefly.

##### The infamous HWNP and issues of mesh convergence

It is fair to state that the HWNP is now *partially* resolved, in that high- $We$  numerical solutions have been reported over the years for a variety of flow problems. I would argue, however, that viscoelastic flow computations are not yet the routine and safe procedure that more classical sub-fields of computational mechanics (e.g. numerical linear elasticity) have generated, and that their success is not guaranteed even in the very restricted sense of "getting the numbers".

Many, if not most, flow problems of interest are characterised by stress singularities or boundary layers of essentially unknown mathematical structure. In this context, an outside observer should not necessarily be impressed by the high- $We$  solutions reported in the literature. Indeed, *some* have been generated with numerical schemes that either explicitly or implicitly smooth out the difficulties. A similar issue arises in classical CFD of Newtonian fluids, in the quest for numerical solutions at high Reynolds numbers. Getting high- $Re$  or high- $We$  (smooth) solutions using risky, "false-diffusion" numerical schemes often essentially means that the numerics have computed an approximate solution to a *different* problem than the one under investigation.

One could argue that mesh refinement experiments (step (2) alluded to above) would readily settle the matter of numerical accuracy. When feasible, as far as computer resources are concerned, these are unfortunately not always successful in that the range of Weissenberg numbers that can be covered in the simulations may *decrease* as the grid is further refined. This problem often occurs in the presence of a stress singularity, thus preventing the safe interpretation of the rheological results obtained in the vicinity of the singular point. One should also keep in mind that a mesh refinement experiment, while essential in assessing the accuracy of the numerical results, does not constitute a rigorous mathematical *proof* of convergence.

The above discussion does not imply, of course, that all results reported in the literature for high values of  $We$  are necessarily inaccurate. I simply wish to stress that the significant advances made over the last decade in expanding the field to more detailed levels of description of polymeric liquids should not hide the absolute necessity of careful numerical validation of present and future methods, whether they be macroscopic, micro-macro, or atomistic. The role of *benchmark* flow problems is crucial in this regard.

## A LIST OF RESEARCH OPPORTUNITIES

Computational rheology is not Mathematics, and I am no Hilbert to draw the path my present and future colleagues should follow over the next century. And yet, on a modest but hopefully useful note, I wish to close this paper with a list of research opportunities that the field offers. This list is obviously incomplete and probably biased by my own experience and interests. It contains topics that have already received preliminary attention, and others which may appear very far indeed from current practice.

On the purely *numerical side*, much research remains to be performed towards the development of accurate and efficient discretization schemes for 3d, time-dependent macro and micro-macro simulations. The role of flow benchmarks will remain essential for the validation of any new technique. Numerical analysis of the proposed schemes should be developed to the widest possible extent, not only to prove convergence (if at all possible), but also to provide theoretical results that would allow the computation of error estimates and the design of related adaptive discretization schemes. Numerical methods for studying the temporal stability of complex flows will be developed further. Appropriate techniques should also be developed that would allow the automatic identification of rheological models and their parameters on the basis of experimental observations of complex flows. Numerical methods should be faithful to the original mathematical model. I would thus find it crucial that mathematicians continue to explore the properties of the exact (and most probably forever unknown) solutions to the governing equations, a very difficult task indeed!

As far as *algorithmic strategies* are concerned, further work is needed on robust iterative and temporal schemes for the solution of the spatially-discretized governing equations. Robust algorithms for tracing the non-linear qualitative behaviour of the solutions should be further developed as well. The efficient use of massively parallel computers should be encouraged. It will necessarily require the development of parallel algorithms tailored to a particular modelling and numerical approach. Also, the implementation of computational rheology models into control algorithms would be of great help to the process engineer.

On the *modelling side*, computational rheology will continue to be exploited for the evaluation and use in complex flows of theoretical models for the polymer dynamics. It will help identify the actual mathematical structure of boundary layers and stress singularities observed in many circumstances. It will also help the theoretical rheologist assess the relative importance of the particular physical mechanisms put forward in his or her theories. In this context, the study of dissipative phenomena in viscoelastic flows is worth continuing, in order to finally understand why viscoelasticity increases the overall drag in a number of cases. Also worth pursuing is the study of turbulence in dilute polymer solutions, which could be approached with ideas akin to large eddy simulations in Newtonian liquids. Non-isothermal flows are of course essential in industrial applications. Their proper mathematical formulation is currently a matter of

debate, to which computational rheology will most probably contribute. One could also envisage using a modelling approach which would couple, in an automatic and adaptive fashion, different levels of description of the same fluid in different regions of the flow. For example, detailed atomistic models could be used to describe the solid wall and its immediate fluid vicinity, while a series of kinetic theory and continuum mechanics models of decreasing levels of detail could be used at progressively larger distances from the wall. The proper matching between the different modelling domains would be done automatically. Most probably, very different numerical techniques, best suited to the local level of description, would be used in each domain. The corresponding numerical scheme would thus be adaptive both in numerical and modelling terms.

Finally, it is quite obvious that the numerical methodology developed for polymeric liquids should and will be applied to other important classes of rheologically-complex materials as well. Indeed, work in that direction has already begun.

## CONCLUSIONS

This short survey of modelling and numerical approaches in computational rheology applied to polymeric liquids demonstrates that the field has expanded considerably in scope since its inception twenty five years ago.

In the early days, researchers were struggling to compute the flow of memory liquids using constitutive equations of very limited (though not altogether vanishing) value for rheological studies, such as the second-order fluid and the upper-convected Maxwell model. Computing with these "simple" constitutive equations is very difficult indeed, as the community soon discovered with surprise and dismay. The problems met then by all players in the field launched a series of numerical and mathematical investigations, constantly nourished by challenging experimental observations such as those on Boger fluids, which significantly increased our understanding of the basic issues. In particular, that more complex and realistic models are actually easier to exploit in numerical studies, once the proper numerical methodology has been developed, is a welcome and healthy fact that has long been reported and continuously been witnessed since.

Computational rheology has clearly gone a long way since the first successful attempts [125-126] to predict the flow of a memory fluid in a complex geometry. Today, the most sophisticated constitutive equations and molecular models developed by theoretical rheologists can in principle be implemented numerically. Furthermore, our colleagues active in experimental work make an increasing use of simulation software to gain further insight into their observations on real materials. Finally, very detailed molecular dynamics simulations are becoming feasible that should help us to elucidate difficult issues such as wall rheology. These facts are a clear signature of progress.

Difficult issues do remain, however, which point to the ever-present need for the careful evaluation of numerical techniques in benchmark flow problems.

The numerical challenges met by computational

rheologists are intimately linked to mathematical and physical considerations. This makes computational rheology fascinating, difficult, and altogether fun. Further progress will benefit from the continuing interaction between people of diverse background. In that regard, computational rheology is very much akin to its well-established experimental and theoretical relatives.

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#### REFERENCES

1. R.I. Tanner and K. Walters, "*Rheology: An Historical Perspective*", Elsevier, Amsterdam (1998).
2. M.J. Crochet, A.R. Davies, and K. Walters, "*Numerical Simulation of Non-Newtonian Flow*", Elsevier, Amsterdam (1984).
3. R. Keunings, in "*Fundamentals of Computer Modeling for Polymer Processing*", (Ed. C.L. Tucker III), Carl Hanser Verlag, Munich (1989), pp. 402-470.
4. M.J. Crochet, *Rubber Chem. Techn., Rubber Reviews* (1989) 62 426-455.
5. F.P.T. Baaijens, *J. Non-Newtonian Fluid Mech.* (1998) 79 361-385.
6. R. Keunings, "*Advances in the Computer Modelling of the Flow of Polymeric Liquids*", to appear in *Comp. Fluid Dyn. J.* (2000).
7. D.V. Boger and K. Walters, "*Rheological Phenomena in Focus*", Elsevier, New York (1993).
8. R.G. Larson, "*The Structure and Rheology of Complex Fluids*", Oxford University Press, New York (1999).
9. E.A. Colbourn (Ed.), "*Computer Simulation of Polymers*", Longman Scientific and Technical, Harlow (1994).
10. V.A. Harmandaris, V.G. Mavrantzas and D.N. Theodorou, *Macromol.* (1998) 31 7934-7943.
11. V.G. Mavrantzas, T.D. Boone, E. Zervopoulou and D.N. Theodorou, *Macromol.* (1999) 32 5072-5096.
12. C. Aust, M. Kroger and S. Hess, *Macromol.* (1999) 32 5660-5672.
13. J. Koplik and J.R. Banavar, *J. Rheol.* (1997) 41 787-805.
14. J. Koplik and J.R. Banavar, *Phys. Rev. Lett.* (1997) 78 2116-2119.
15. A. Jabbarzadeh, J.D. Atkinson and R.I. Tanner, *Phys. Rev. E* (2000) 61 690-699.
16. R.B. Bird, C.F. Curtiss, R.C. Armstrong and O. Hassager, "*Dynamics of Polymeric Liquids Vol. 2: Kinetic Theory*", 2nd Ed., John Wiley, New York (1987).
17. H.C. Öttinger, "*Stochastic Processes in Polymeric Fluids: Tools and Examples for Developing Simulation Algorithms*", Springer, Berlin (1996).
18. P.S. Doyle and E.S.G. Shaqfeh, *J. Non-Newtonian Fluid Mech.* (1998) 76 43-78.
19. P.S. Doyle, E.S.G. Shaqfeh, G.H. McKinley and S.H. Spiegelberg, *J. Non-Newtonian Fluid Mech.* (1998) 76 79-110.
20. T.C.B. Kwan and E.S.G. Shaqfeh, *J. Non-Newtonian Fluid Mech.* (1999) 82 139-165.
21. R.G. Larson, H. Hu, D.E. Smith and S. Chu, *J. Rheol.* (1999) 43 267-304.
22. L. Li, R.G. Larson and T. Sridhar, *J. Rheol.* (2000) 44 291-322.
23. L. Li and R.G. Larson, *Macromol.* (2000) 33 1411-1415.
24. R. Sizaire, G. Lielens, I. Jaumain, R. Keunings and V. Legat, *J. Non-Newtonian Fluid Mech.* (1999) 82 233-253.
25. M.R.J. Verhoef, B.H.A.A. van den Brule and M.A. Hulsen, *J. Non-Newtonian Fluid Mech.* (1999) 80 155-182.
26. T.T. Perkins, D.E. Smith and S. Chu, *Science* (1997) 276 2016-2021.
27. D.E. Smith and S. Chu, *Science* (1998) 281 1335-1340.
28. D.E. Smith, H.P. Babcock and S. Chu, *Science* (1999) 283 1724-1727.
29. M. Doi and S.F. Edwards, "*The Theory of Polymer Dynamics*", Clarendon Press, Oxford (1986).
30. G. Ianniruberto and G. Marrucci, *J. Non-Newtonian Fluid Mech.* (1996) 65 241-246.
31. D.W. Mead, R.G. Larson and M. Doi, *Macromol.* (1998) 31 7895-7914.
32. G. Marrucci and G. Ianniruberto, *J. Non-Newtonian Fluid Mech.* (1999) 82 275-286.
33. G. Marrucci, F. Greco and G. Ianniruberto, "*Possible Role of Force Balance on Entanglements*", IUPAC Prague Meeting on Rheology of Polymer Systems, *Macromolecular Symposia*, in press (2000).
34. G. Marrucci, F. Greco and G. Ianniruberto, "*Integral and Differential Constitutive Equations for Entangled Polymers with Simple Versions of CCR and Force Balance on Entanglements*", submitted to *Rheol. Acta*, January 2000.
35. C.C. Hua, J.D. Schieber and D.C. Venerus, *J. Rheol.* (1999) 43 701-717.
36. T.C.B. McLeish and S.T. Milner, in "*Advances in Polymer Science*", Vol. 143, Springer verlag, Berlin (1999) pp. 195-255.
37. R.B. Bird, R.C. Armstrong and O. Hassager, "*Dynamics of Polymeric Liquids Vol. 1: Fluid*

- Mechanics*", 2nd Ed., John Wiley, New York (1987).
38. C.L. Tucker (Ed.), *"Fundamentals of Computer Modeling for Polymer Processing"*, Hanser Publishers, Munich (1989).
  39. R.G. Larson, *"Constitutive Equations for Polymer Melts and Solutions"*, Butterworth, Boston (1988). Out of Print; a photocopied version can be ordered from rlarson@engin.umich.edu
  40. T.C.B. McLeish and R.G. Larson, *J. Rheol.* (1998) 42 81-110.
  41. R.J. Blackwell, T.C.B. McLeish and O.G. Harlen, *J. Rheol.* (2000) 44 121-136.
  42. R. Keunings, *J. Non-Newtonian Fluid Mech.* (1997) 68 85-100.
  43. A.N. Beris and B.J. Edwards, *"Thermodynamics of Flowing Systems with Internal Microstructure"*, Oxford University Press, New York (1994).
  44. M. Grmela and H.C. Öttinger, *Phys. Rev. E* (1997) 56 6620-6632.
  45. H.C. Öttinger, *Applied Rheology* (1999) 9 17-26.
  46. H.C. Öttinger and A.N. Beris, *J. Chem. Phys.* (1999) 110 6593-6596.
  47. H.C. Öttinger, *J. Rheol.* (1999) 43 1461-1493.
  48. D.V. Boger, *Ann. Rev. Fluid Mech.* (1987) 19 157-182.
  49. J.P. Rothstein and G.H. McKinley, *J. Non-Newtonian Fluid Mech.* (1999) 86 61-88.
  50. A.M. Grillet, A.G. Lee and E.S.G. Shaqfeh, *J. Fluid Mech.* (1999) 399 49-83.
  51. P. Pakdel and G.H. McKinley, *Phys. Fluids* (1998) 10 1058-1070.
  52. A.M. Grillet, B. Yang, B. Khomami and E.S.G. Shaqfeh, *J. Non-Newtonian Fluid Mech.* (1999) 88 99-131.
  53. J.M. Piau, *Macrom. Symp.* (1999) 143 269-289.
  54. B.I.M. ten Bosch, *J. Non-Newtonian Fluid Mech.* (1999) 83 231-248.
  55. L. Giraud, D. d'Humieres and P. Lallemand, *Europhys. Lett.* (1998) 42 625-630.
  56. A. Malevanets and J.M. Yeomans, *Faraday Discuss.* (1999) 112 237-248.
  57. G. Lielens, R. Keunings and V. Legat, *J. Non-Newtonian Fluid Mech.* (1999) 87 179-196.
  58. G. Lielens, P. Halin, I. Jaumain, R. Keunings and V. Legat, *J. Non-Newtonian Fluid Mech.* (1998) 76 249-279.
  59. J. Baranger, C. Guillopé and J.C. Saut, in *"Rheology for Polymer Melt Processing"*, J.M. Piau and J.F. Agassant (Eds.), Elsevier, Amsterdam (1996) pp. 199-236.
  60. O.G. Harlen, E.J. Hinch and J.M. Rallison, *J. Non-Newtonian Fluid Mech.* (1992) 44 229-265.
  61. T. Hagen and M. Renardy, *J. Non-Newtonian Fluid Mech.* (1997) 73 181-189.
  62. M. Renardy, *J. Non-Newtonian Fluid Mech.* (2000) 90 13-23.
  63. A.R. Davies and J. Devlin, *J. Non-Newtonian Fluid Mech.* (1993) 50 173-191.
  64. E.J. Hinch, *J. Non-Newtonian Fluid Mech.* (1993) 50 161-171.
  65. M. Renardy, *J. Non-Newtonian Fluid Mech.* (1995) 58 83-89.
  66. H.J. Wilson, M. Renardy and Y. Renardy, *J. Non-Newtonian Fluid Mech.* (1999) 80 251-268.
  67. M. Renardy, Y. Renardy, R. Sureshkumar and A.N. Beris, *J. Non-Newtonian Fluid Mech.* (1996) 63 1-31.
  68. H.J. Wilson and J.M. Rallison, *J. Non-Newtonian Fluid Mech.* (1999) 87 75-96.
  69. Y.R. Fan, R.I. Tanner and N. Phan-Thien, *J. Non-Newtonian Fluid Mech.* (1999) 84 233-256.
  70. J. Sun, M.D. Smith, R.C. Armstrong and R.A. Brown, *J. Non-Newtonian Fluid Mech.* (1999) 86 281-307.
  71. E. Brasseur, M.M. Fyrillas, G.C. Georgiou and M.J. Crochet, *J. Rheol.* (1998) 42 549-566.
  72. R. Sureshkumar, M.D. Smith, R.C. Armstrong and R.A. Brown, *J. Non-Newtonian Fluid Mech.* (1999) 82 57-104.
  73. H.K. Rasmussen and O. Hassager, *J. Non-Newtonian Fluid Mech.* (1999) 82 189-202.
  74. P. Szabo, J.M. Rallison and E.J. Hinch, *J. Non-Newtonian Fluid Mech.* (1997) 72 73-86.
  75. P.Y. Huang, H.H. Hu and D.D. Joseph, *J. Fluid Mech.* (1998) 362 297-325.
  76. H. Matallah, P. Townsend and M.F. Webster, *Comm. Numer. Meth. Engng.* (1998) 14 473-491.
  77. G.B. Bishko, O.G. Harlen, T.C.B. McLeish and T.M. Nicholson, *J. Non-Newtonian Fluid Mech.* (1999) 82 255-273.
  78. P. Halin, G. Lielens, R. Keunings and V. Legat, *J. Non-Newtonian Fluid Mech.* (1998) 79 387-403.
  79. X. Gallez, P. Halin, G. Lielens, R. Keunings and V. Legat, *Comp. Meth. Appl. Mech. and Engng.* (1999) 180 345-364.
  80. P. Wapperom, R. Keunings and V. Legat, *J. Non-Newtonian Fluid Mech.* (2000) 91 273-295.
  81. G. Mompean and M. Deville, *J. Non-Newtonian Fluid Mech.* (1997) 72 253-279.
  82. S.C. Xue, R.I. Tanner and N. Phan-Thien, *Comp. Meth. Appl. Mech. Engng.* (1999) 180 305-331.



83. S.C. Xue, N. Phan-Thien and R.I. Tanner, *J. Non-Newtonian Fluid Mech.* (1999) 87 337-367.
84. P. Wapperom and M.F. Webster, *Comp. Meth. Appl. Mech. Engng.* (1999) 180 281-304.
85. T.N. Phillips and A.J. Williams, *J. Non-Newtonian Fluid Mech.* (1999) 87 215-246.
86. H.K. Rasmussen, *J. Non-Newtonian Fluid Mech.* (1999) 84 217-232.
87. A.P.G. van Heel, M.A. Hulsen and B.H.A.A. van den Brule, *J. Rheol.* (1999) 43 1239-1260.
88. E.A.J.F. Peters, M.A. Hulsen and B.H.A.A. van den Brule, *J. Non-Newtonian Fluid Mech.* (2000) 89 209-228.
89. A.N. Beris and C.D. Dimitropoulos, *Comp. Meth. Appl. Mech. Engng.* (1999) 180 365-392.
90. R. Kupferman and M.M. Denn, *J. Non-Newtonian Fluid Mech.* (1999) 84 275-287.
91. S. Kumar and G.M. Homsy, *J. Non-Newtonian Fluid Mech.* (1999) 83 249-276.
92. F.P.T. Baaijens, *J. Non-Newtonian Fluid Mech.* (1998) 75 119-138.
93. R. Aggarwal, R. Keunings and F.-X. Roux, *J. Rheol.* (1994) 38 405-419.
94. P. Henriksen and R. Keunings, *Int. J. Num. Meth. in Fluids* (1994) 18 1167-1183.
95. R. Keunings, *Comput. in Chem. Engng.* (1995) 19 647-669.
96. N. Phan-Thien and H.S. Dou, *J. Non-Newtonian Fluid Mech.* (1998) 77 21-51.
97. R. Ahmed, R.F. Liang and M.R. Mackley, *J. Non-Newtonian Fluid Mech.* (1995) 59 129-153.
98. W.H. Hartt and D.G. Baird, *J. Non-Newtonian Fluid Mech.* (1996) 65 247-268.
99. C. Beraudo, A. Fortin, T. Coupez, Y. Demay, B. Vergnes and J.F. Agassant, *J. Non-Newtonian Fluid Mech.* (1998) 75 1-23.
100. A.C.B. Bogaerds, W.M.H. Verbeeten, G.W.M. Peters and F.P.T. Baaijens, *Comp. Meth. Appl. Mech. Engng.* (1999) 180 413-430.
101. A.W. Liu, D.E. Bornside, R.C. Armstrong and R.A. Brown, *J. Non-Newtonian Fluid Mech.* (1998) 77 153-190.
102. E. Mitsoulis, *J. Non-Newtonian Fluid Mech.* (1998) 78 187-201.
103. J.F.M. Schoonen, F.H.M. Swartjes, G.W.M. Peters, F.P.T. Baaijens and H.E.H. Meijer, *J. Non-Newtonian Fluid Mech.* (1998) 79 529-561.
104. M.A. Zirnsak and D.V. Boger, *J. Non-Newtonian Fluid Mech.* (1998) 79 105-136.
105. J.M. Li, W.R. Burghardt, B. Yang and B. Khomami, *J. Non-Newtonian Fluid Mech.* (1998) 74 151-193.
106. W.R. Burghardt, J.M. Li, B. Khomami and B. Yang, *J. Rheol.* (1999) 43 147-165.
107. G.W.M. Peters, J.F.M. Schoonen, F.P.T. Baaijens and H.E.H. Meijer, *J. Non-Newtonian Fluid Mech.* (1999) 82 387-427.
108. B. Yang and B. Khomami, *J. Non-Newtonian Fluid Mech.* (1999) 82 429-452.
109. J. Remmelgas and L.G. Leal, *J. Non-Newtonian Fluid Mech.* (2000) 89 231-249.
110. M.I. Kolte, H.K. Rasmussen and O. Hassager, *Rheol. Acta* (1997) 36 285-302.
111. R. Sizaire and V. Legat, *J. Non-Newtonian Fluid Mech.* (1997) 71 89-107.
112. M.W. Yao, S.H. Spiegelberg and G.H. McKinley, *J. Non-Newtonian Fluid Mech.* (2000) 89 1-43.
113. D. Rajagopalan, "Computational Analysis of Techniques to Determine Extensional Viscosity from Entrance Flows", submitted to *Rheol. Acta* (Sept. 1999).
114. Fan Xijun, *Acta Mech. Sinica* (1989) 5 216-226.
115. H.C. Öttinger and M. Laso, in "Theoretical and Applied Rheology", Vol. 1, P. Moldenaers and R. Keunings (Eds.), Elsevier, Amsterdam (1992) pp. 286-288.
116. M. Laso and H.C. Öttinger, *J. Non-Newtonian Fluid Mech.* (1993) 47 1-20.
117. K. Feigl, M. Laso, and H.C. Öttinger, *Macromol.* (1995) 28 3261-3274.
118. C.C. Hua and J.D. Schieber, *Chem. Eng. Sci.* (1996) 51 1473-1485.
119. T.W. Bell, G.H. Nyland, J.J. de Pablo and M.D. Graham, *Macromol.* (1997) 30 1806-1812.
120. C.C. Hua and J.D. Schieber, *J. Rheol.* (1998) 42 477-491.
121. M.A. Hulsen, A.P.G. van Heel and B.H.A.A. van den Brule, *J. Non-Newt. Fluid Mech.* (1997) 70 79-101.
122. H.C. Öttinger, B.H.A.A. van den Brule and M.A. Hulsen, *J. Non-Newt. Fluid Mech.* (1997) 70 255-261.
123. M. Fortin, R. Guenette and R. Pierre, *Comp. Meth. Appl. Mech. Engng.* (1997) 143 79-95.
124. J. Baranger and A. Machmoum, *Comp. Meth. Appl. Mech. Engng.* (1997) 148 39-52.
125. M.G.N. Perera and K. Walters, *J. Non-Newtonian Fluid Mech.* (1977) 2 49-81.
126. M. Kawahara and N. Takeuchi, *Comp. Fluids* (1977) 5 33-45.

