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NUMERICAL METHODS FOR MEMORY FLUIDS: A CRITICAL SURVEY

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ABSTRACT

We summarize our recent review of the state of the art in the numerical analysis of viscoelastic flows [1]. After a description of the mathematical models used in computer simulations, we organize the spectrum of available numerical techniques on the basis of the two approaches of handling the memory of viscoelastic fluids: the *coupled approach,* where the constitutive model and the conservation laws are solved simultaneously, and the *decoupled approach,* where the computation of the viscoelastic stresses is done separately from that of the flow kinematics. These two methodologies have been used with both differential and single-integral constitutive models, and in conjunction with various discretization methods. We refer to a number of successful simulations where significant viscoelastic effects have been predicted. Finally, we briefly review the outstanding numerical, mathematical, and modeling problems in the field of viscoelastic flow computations.

1. INTRODUCTION

It is well established that the range of validity of the Newtonian constitutive equation is limited to low molecular weight liquids. The provocative flow phenomena observed with polymeric fluids cannot be predicted by the Navier-Stokes equations. Non-Newtonian behavior has many facets. Among them are the shear-rate dependence of the shear viscosity, the presence of normal stresses in viscometric flows, high resistance to elongational deformation, and memory effects associated with the elasticity of the material. Viscoelastic effects, i.e. flow phenomena that cannot be explained on the basis of linear or nonlinear purely-viscous behavior, can be important in polymer processing applications. Flow instabilities, for example, limit the rate

of production in many processing operations. The instabilities often occur at very low Reynolds numbers, where corresponding flows of low molecular weight liquids are stable. Viscoelastic behavior is also responsible for complex flow patterns, such as large recirculation regions in confined geometries. These flow patterns can have a significant impact on product quality. An obvious example is that of the processing of fiber-reinforced polymer materials, where the flow-induced distribution of fiber orientations determines the mechanical properties of the final product.

Over the last ten years, much research activity has been focused on the development of numerical techniques for predicting the flow of viscoelastic fluids in complex geometries. Even though the flow of polymers often occurs at very low Reynolds numbers, the numerical prediction of viscoelastic effects in complex geometries has proven very difficult in view of nonlinearities related to the fluid memory. The convergence of numerical algorithms has long been restricted to small values of the Weissenberg number, a dimensionless group which quantifies the elastic character of the flow. As a result, significant viscoelastic effects seen in laboratory experiments or processing applications could not possibly be predicted. The situation has improved over the last three years or so. Numerical solutions are now available in the range of Weissenberg numbers covered in actual flow experiments. Some of these solutions do predict observed viscoelastic effects, at least qualitatively. Others do not agree with experimental observations, which indicates that uncertainties remain in the mathematical description of the physics of polymer flows. In addition, much progress has been made in the identification of the underlying causes of numerical difficulties and in the development of more accurate discretization techniques. The reader will find a comprehensive review of the field of viscoelastic flow computations in the recent work by Keunings [1], of which the present paper is a brief summary.

2. MATHEMATICAL MODELS

2.1 Preliminaries

In the context of a continuum mechanical approach, the flow of memory fluids can be formulated as a set of conservation and constitutive equations augmented with suitable boundary and initial conditions. The selection of a constitutive equation obviously constitutes a critical step in the modeling of viscoelastic flows. A large number of constitutive models have been developed (and indeed are still being developed) to describe the rheological behavior of polymeric fluids. It is however essential to be aware that none of these models leads to realistic predictions in all types of deformation of any

particular polymeric fluid. This is in marked contrast to Newtonian fluid mechanics, where the mathematical description of the flow is well established. The constitutive models that are used in numerical simulations are classified as *differential* and *single-integral* models. Their predictive abilities in standard rheometrical flows range from very poor to excellent, depending of the type of motion and/or the class of materials. Another integral part of the modeling process is the selection of appropriate boundary conditions. This step is a complex one with viscoelastic fluids, for at least two reasons. First, the fluid memory requires that the pre-history of the fluid motion be specified in the analysis of flow problems with inlet boundaries. The motion pre-history, if at all known, can be as complex as the flow problem under investigation. The second difficulty is related to the behavior of polymeric liquids near solid boundaries. In the analysis of highly-viscous Newtonian flows, it is generally appropriate to assume that the fluid sticks to solid boundaries. Such is not always the case in polymer processing applications. Actually, flow phenomena associated with viscoelastic fluids (including low Reynolds number instabilities) may well find their origin not only in the non-Newtonian character of the bulk flow, but also in slip mechanisms at solid boundaries. To summarize, current mathematical formulations of the flow of polymeric fluids, both in the bulk and near solid surfaces, are likely to be altered as our understanding of the physics of polymer flow increases.

2.2 Conservation Equations

The vast majority of viscoelastic simulations have been for incompressible, isothermal flows. The Cauchy stress tensor σ is thus given by $-P\delta + \tau$, where P is the pressure, δ is the unit tensor, and τ is the extra-stress tensor. The set of conservation laws reduces to the continuity equation

$$\nabla \cdot \mathbf{v} = 0 \quad , \tag{1}$$

and the momentum equation

$$\nabla \cdot \sigma + \rho\, \mathbf{f} = \rho \frac{D\mathbf{v}}{Dt} \quad . \tag{2}$$

Here, $\mathbf{v}$ is the velocity vector, $\mathbf{f}$ is the body force per unit mass of fluid, and ρ is the fluid density. The operator D/Dt is the material time derivative $\partial/\partial t + \mathbf{v} \cdot \nabla$. The set of governing equations (1-2) is closed with a constitutive model that relates the extra-stress τ to the deformation experienced by the fluid. It is important at this stage to

distinguish between constitutive models which have or do not have a Newtonian viscosity. In general, we can decompose the extra-stress as a sum of a viscoelastic component τ_V and a Newtonian component τ_N, i.e. $\tau = \tau_V + \tau_N$. The Newtonian component is given by $\mu_N \dot{\gamma}$, where $\dot{\gamma}$ is the rate of strain tensor $(\nabla \mathbf{v} + \nabla \mathbf{v}^\dagger)$ and μ_N is a constant viscosity coefficient. Viscoelastic fluid models without Newtonian viscosity $(\mu_N = 0)$ can exhibit a variety of hyperbolic phenomena, including mathematical change of type and propagation of waves.

2.3 Differential Constitutive Models

Differential models used currently in numerical simulations can be written in the general form

$$\mathbf{A}(\tau_V) \cdot \tau_V + \lambda \frac{\delta \tau_V}{\delta t} = \mu_V \dot{\gamma} \quad . \tag{3}$$

Here, λ is a relaxation time and μ_V is a viscosity coefficient. They are usually taken as constants, but can also be made functions of the magnitude of the rate of strain tensor, if desired. The symbol $\mathbf{A}$ denotes a model-dependent tensor function. Finally, the operator $\delta/\delta t$ is an objective time derivative defined as a linear combination of the lower and upper-convected derivatives of τ_V given respectively by

$$\begin{aligned} \tau_V{}^{(1)} &= \frac{D\tau_V}{Dt} + \tau_V \cdot \nabla \mathbf{v}^\dagger + \nabla \mathbf{v} \cdot \tau_V \quad , \\ \tau_{V\,(1)} &= \frac{D\tau_V}{Dt} - \tau_V \cdot \nabla \mathbf{v} - \nabla \mathbf{v}^\dagger \cdot \tau_V \quad . \end{aligned} \tag{4}$$

The generic constitutive equation (3) is readily extended to the case of a spectrum of relaxation times by writing τ_V as a finite sum of partial extra-stresses $\tau_{V,k}$ obeying (3) with material coefficients λ_k and $\mu_{V,k}$.

The simplest differential constitutive equations capable of predicting memory effects are the Maxwell models $(\mathbf{A} = \delta)$. More complex constitutive equations of the type (3) include the models of Phan Thien and Tanner, and Giesekus. It should be noted that the addition of a Newtonian component τ_N is equivalent to introducing a retardation time. For example, the upper-convected Maxwell fluid plus a Newtonian viscosity yields the Oldroyd-B model.

A number of important remarks should be made at this point. First, the differential models (3) are *implicit* in the extra-stress τ_V. As a result, it is impossible to substitute the extra-stress out of the momentum equation (2), as one does with a Newtonian fluid to obtain

the classical Navier-Stokes equations. Second, we note that any fluid mechanical problem involving a constitutive equation of the type (3) is inherently nonlinear, even in the absence of inertia terms in the momentum equation. This is due in part to the nonlinear coupling between extra-stresses and velocities embedded in the definition of the convected derivatives. Finally, in the particular case of steady-state flows, equation (3) constitutes a set of first-order hyperbolic equations with the streamlines as characteristic curves. When the flow domain contains an inlet boundary, one must thus specify values of the extra-stress along a line crossing the incoming streamlines in order to obtain a well-posed problem. These *initial stresses* contain in a disguised form the necessary information on the flow pre-history.

2.4 Single-Integral Constitutive Models

Let us consider a fluid particle whose position at present time t is given by $\mathbf{x}(t)$. The fluid motion can be described by the vector relation $\mathbf{x}(t') = \chi(\mathbf{x}(t),t,t')$ which gives the particle position $\mathbf{x}(t')$ at historical time t' ranging between $-\infty$ and t. The relative deformation gradient $\mathbf{F}_t$ and the right Cauchy-Green strain tensor $\mathbf{C}_t$ are given respectively by $\mathbf{F}_t(t') = \partial\chi/\partial\mathbf{x}$ and $\mathbf{C}_t(t') = \mathbf{F}_t^{\dagger}(t')\cdot\mathbf{F}_t(t')$.

Single-integral constitutive equations give the viscoelastic extra-stress τ_V at a fluid particle through a time integral of the deformation history. In numerical studies, researchers have used integral models of the form

$$\tau_V(t) = \int_{-\infty}^{t} m(t-t')\,\mathbf{S}_t(t')\,dt' \quad . \tag{5}$$

Here, the operator $\int \cdot\, dt'$ is a time integral taken along the particle path parameterized by the historical time t'. The kernel $\mathbf{S}_t$ is a deformation-dependent tensor of the form

$$\mathbf{S}_t(t') = \phi_1(I_1,I_2)\,[\mathbf{C}_t^{-1}(t') - \delta\,] + \phi_2(I_1,I_2)\,[\mathbf{C}_t(t') - \delta\,] \quad , \tag{6}$$

where $\mathbf{C}_t^{-1}$, the inverse of $\mathbf{C}_t$, is the Finger strain tensor. The functions ϕ_1 and ϕ_2 are dimensionless scalar functions that depend on the invariants $I_1 = tr(\mathbf{C}_t^{-1})$ and $I_2 = tr(\mathbf{C}_t)$. Finally, the function $m(t-t')$ appearing in (5) is the time-dependent memory function of linear viscoelasticity. It is usually expressed as a sum of exponential functions involving the relaxation times λ_k and the viscosity coefficients $\mu_{V,k}$:

$$m(t-t') = \sum_{k=1}^{n} \frac{\mu_{V,k}}{\lambda_k^2} \, exp[\frac{-(t-t')}{\lambda_k}] \quad . \tag{7}$$

This definition illustrates the notion of *fading memory,* i.e. the deformations experienced by a fluid element in the recent past contribute more to the current stress in that element than those deformations which took place in the distant past.

One of the simplest integral constitutive equations is the so-called rubberlike liquid model developed by Lodge. It is obtained by setting $\phi_1 = 1$ and $\phi_2 = 0$ in (6). When used with a memory function of the form (7), Lodge's equation is equivalent to the upper-convected differential Maxwell model. A more realistic model of the type (5) is the factorized BKZ model in which the kernel functions ϕ_1 and ϕ_2 derive from a potential. The BKZ equation includes the model of Doi and Edwards as a particular case.

Single-integral constitutive equations present interesting computational challenges. First, the particle paths needed for the computation of the memory integral are unknown a priori. This particularity leads to flow problems which are inherently nonlinear, and in a sense akin to free surface flows. The second challenge is quite new in computational fluid dynamics: integral models are formulated in a *Lagrangian form* which does not involve the Eulerian velocity field explicitly. For this reason only, one would expect that numerical schemes aimed at computing the flow of integral fluids differ drastically from classical techniques for Newtonian fluids.

3. BASIC COMPUTATIONAL APPROACHES

The solution of viscoelastic flow problems presents different numerical challenges whith differential and integral constitutive models. A common feature, however, is the nonlinear character of the governing equations brought about by the fluid memory. Two basic approaches have been adopted to handle this nonlinearity. Herafter, we shall refer to them as the *coupled* and *decoupled* approaches. In the coupled approach, the discretized governing equations are solved simultaneously for the whole set of primary variables, usually by means of Newton's iterative scheme. In the decoupled approach, the computation of the viscoelastic extra-stress is performed separately from that of the flow kinematics. From known kinematics, one calculates the viscoelastic extra-stress by integrating the constitutive equation. The kinematics are then updated by solving the conservation equations, and the procedure is iterated upon. The update scheme is usually akin to Picard's iterative algorithm.

The vast majority of coupled techniques have been developed for differential models. Actually, it is quite difficult to use a coupled

approach with integral models. The main advantage of coupled techniques lies in the iterative procedure itself. Newton's scheme has well-known convergence properties, and it usually converges in a few iterations when initial estimates are computed by a continuation method. Furthermore, there is a well-established methodology based on Newton's scheme for investigating the temporal stability of the numerical solutions, tracking irregular points in solution families (e.g. turning points), and computing bifurcating solution branches. This methodology can also be used to explain possible convergence difficulties with Newton's method. The main disadvantage of coupled techniques is related to simulation cost. Coupled techniques are generally very demanding in terms of computer storage and execution time. As a result, coupled techniques for differential models have not been used with a spectrum of relaxation times.

Decoupled techniques have been developed for both differential and integral models. Some of them are much cheaper than coupled techniques, so much so that they can be used on micro-computers. In general, the use of a spectrum of relaxation times does not increase simulation costs significantly. Another advantage of the decoupled approach is the breakup of the governing equations into a Newtonian problem (i.e. the conservation equations with known viscoelastic extra-stresses), and the constitutive equations. One can thus use well-established methods to discretize the conservation equations. Viscoelasticity only enters through a model-dependent stress integrator. The main disadvantage of decoupled techniques lies in the iterative procedure. Picard-type schemes (or improvements thereof) are often slow to converge, and their convergence is never guaranteed. Furthermore, they do not provide any information on the qualitative behavior of the numerical solutions (i.e. stability, multiplicity, etc.).

In conjunction with the coupled and decoupled approaches, researchers have used a broad spectrum of *discretization techniques* that include finite element, boundary element, finite difference, and spectral methods. It should be mentioned, however, that the majority of published simulations have been carried out with finite element techniques.

4. COUPLED TECHNIQUES

4.1 Differential Models

Almost all coupled techniques for differential models are based on a mixed Galerkin/finite element solution of (1-3). In one formulation, which we call MFE1, approximations of the finite element type are defined for the viscoelastic extra-stress, the velocity, and the pressure. Discretization of the governing equations is performed by means of the Galerkin method. The appropriate selection of shape functions is a

delicate task which remains the subject of active investigation. A popular choice is the use of C^0–P^2 interpolations for velocity and stress, with C^0–P^1 elements for pressure. The mixed formulation MFE1 has been extended to the case of transient free surface flows.

Two alternative Galerkin/finite element formulations of (1-3) are available, which we call MFE2 and MFE3. It should be pointed out that no argument has yet been offered that would conclusively dictate which of the three formulations is best in terms of numerical accuracy. A spectral/Galerkin/finite element technique has also been developed recently; it shows a remarkable increase in accuracy relative to current mixed finite element methods for flows endowed with smooth velocity and stress fields. In all cases, Newton's iterative technique is the method of choice for the solution of the discrete equations.

4.2 Integral Models

The development of coupled methods for integral models is difficult for at least two reasons: the constitutive equation (5) is given in a Lagrangian form and the particle paths are unknown a priori. To date, only two techniques have been proposed in the literature. One is based on an Eulerian formulation of (5) valid for flows without recirculation regions; the streamlines are treated as primary unknowns together with velocities and pressures. The other method is more general and uses the Lagrangian formulation of the conservation laws; the primary kinematical variables are the particle positions $\mathbf{x}(t)$, and the domain of integration is a time-dependent *material volume*. Both techniques are based on the Galerkin/finite element method.

5. DECOUPLED TECHNIQUES

5.1 Basic Procedure

The idea behind a decoupled technique is to separate the computation of the viscoelastic extra-stress from that of the kinematics. Of course, an iterative procedure is needed to arrive at a solution of the full set of governing equations. Most decoupled methods are based on the following iterative scheme:

step 1: solve the constitutive equation for the viscoelastic extra-stress using the kinematics calculated at the previous iteration,

step 2: update the kinematics by solving the conservation laws with the viscoelastic extra-stress computed in step 1,

step 3: check for convergence; if needed, return to step 1.

Step 2 amounts to solving a Newtonian flow problem with a known, fictitious body force. We refer to this step as the *perturbed Newtonian problem*. Well-established procedures have been used to solve the

perturbed Newtonian problem, including the velocity-pressure Galerkin/finite element technique, the boundary element method, and the stream function/vorticity finite difference technique. Let us now discuss step 1, i.e. the computation of viscoelastic extra-stresses at integration or grid points from a known velocity field.

5.2 Differential Models

In steady flow, the differential model (3) is a set of first-order hyperbolic equations with the streamlines as characteristic curves. First-order hyperbolic equations are most naturally solved by the method of characteristics, whereby the original set of partial differential equations is transformed into a set of ordinary differential equations to be solved along the characteristic curves. We can re-write (3) in the form

$$\lambda \mid \mathbf{v} \mid \frac{d\tau_V}{dl} = \mathbf{B}(\tau_V, \mathbf{v}, \nabla\mathbf{v}) \ , \tag{8}$$

where $\mathbf{B}$ is a model-dependent tensor function, l is the arc length along a streamline and $\mathbf{v}$ is the velocity field computed at the previous iteration. Equation (8) defines an initial-value problem for τ_V which can be integrated accurately by means of standard procedures (e.g. fourth-order Runge-Kutta's method). In the case of non-closed streamlines, the initial values correspond to the inlet extra-stress boundary conditions discussed in Section 2.3. Closed streamlines are difficult to handle with this technique, for they would require a shooting method. Note that the use of a spectrum of relaxation times involves the integration of (8) for each partial extra-stress $\tau_{V,k}$; this task can be fulfilled without a significant increase of storage. This is not true with the mixed methods of Section 4.

5.3 Integral Models

Integral models have been used in conjunction with both finite difference and finite element solutions of the perturbed Newtonian problem. Available techniques are based on the Lagrangian description (5). Schematically, the computation of viscoelastic extra-stresses at integration or grid points is performed in three steps:

(1) tracking: on the basis of a known steady-state velocity field, compute the upstream trajectory and the travel time of each integration or grid point,

(2) strain evaluation: at selected past times, compute the deformation gradient $\mathbf{F}_t$ and from it the integrand of (5),

(3) stress evaluation: compute the integral (5) numerically, using the results of step 2.

Steps 1 and 3 are readily carried out using, respectively, the velocity field known from the previous iteration, and a Gauss-Laguerre quadrature rule. The main difficulty lies in the computation of the strain history. The latter can be evaluated through the streamline integration of

$$\frac{D}{Dt'}\mathbf{F}_t(t') = \nabla\mathbf{v}^\dagger(t')\cdot\mathbf{F}_t(t') \,, \tag{9}$$

with the initial condition $\mathbf{F}_t(t) = \delta$. Accurate procedures have been proposed recently in the context of the finite element solution of the perturbed Newtonian problem. One is based on a low-order finite element interpolation of the velocity field which makes it possible to perform the tracking procedure as well as the strain computation *analytically* within each element. The other consists in the numerical solution of (9) along the particle paths.

6. COMPLETED WORK

The numerical techniques described in this paper have been applied to a number of steady-state flow problems, including entry flows, extrusion flows, flows over a transverse slot, flows past submerged objects such as spheres and cylinders, and flows in a journal bearing. A few time-dependent flows have been computed, such as the squeeze film problem and the breakup of jets. In view of the significant computer resources involved in viscoelastic computations, available simulations are for two-dimensional or three-dimensional axisymmetric flows. No fully three-dimensional results have been reported yet.

Viscoelastic simulations have long been plagued by a fundamental difficulty, i.e. the divergence of *all* numerical iterative schemes beyond some critical value of the Weissenberg number *We*, the dimensionless group which quantifies the elastic character of the flow. One usually defines *We* as the product of a characteristic relaxation time of the fluid and a characteristic velocity gradient of the flow. The critical value of *We* usually depends on the flow problem, the numerical method, the iterative scheme, the grid, and the constitutive equation; it has long been so low that the provocative flow patterns observed with polymeric fluids could not possibly be predicted. This frustrating state of affairs has improved recently, in the sense that several simulations have now been performed successfully over a range of Weissenberg numbers which covers the experimental range. In isolated cases, the simulations were interrupted not by lack of convergence of the iterative process, but by lack of interest in pursuing expensive computations to higher values of *We*. Also, a few numerical simulations have been reported which predict dramatic viscoelastic

effects on both stress and velocity fields (e.g. die swell computations, flows through sudden contractions, and the jet breakup problem).

7. THE HIGH WEISSENBERG NUMBER PROBLEM

Despite the recent progress, most numerical simulations can be carried out only up to a critical value of the Weissenberg number, beyond which the iterative scheme ceases to converge. Our understanding of the so-called high Weissenberg number problem (HWNP) has increased dramatically over the last three years. Furthermore, important mathematical results have been obtained which shed considerable light on the numerical difficulties associated with viscoelastic simulations.

The existence of turning points in discrete solution families has clearly been identified as the cause for the HWNP in various simulations with *coupled techniques*. In some cases, but not all, these turning points are numerical artifacts generated by excessive discretization errors. It is more difficult to identify the actual cause for the divergence of *decoupled iterations*. Turning points (or other irregular points which may cause the divergence of decoupled iterations, such as bifurcation points) may well be present in the discrete solution families, but they cannot be tracked unambiguously with Picard-type schemes.

Researchers have identified two classes of basic causes for discretization problems in viscoelastic computations: (i) the existence of flow regions endowed with high stress or velocity gradients (including possibly non-integrable singularities), and (ii) the hyperbolic character of viscoelastic problems, which can, in particular, induce local changes of type of the governing equations as well as loss of evolution. These difficult topics cannot be treated adequately in the present summary; a detailed discussion can be found in [1].

8. CONCLUSIONS

The field of large-scale viscoelastic simulations has progressed significantly in recent years. It is no longer true that all numerical techniques fail to provide solutions at high Weissenberg numbers. Actually, a few simulations have been reported which predict observed viscoelastic effects either quantitatively or qualitatively. The range of discretization methods applied to viscoelastic problems has also been considerably enlarged. Further progress is likely to occur with the development of more accurate discretization methods and the use of more realistic, and thus more complex, constitutive equations.

Despite the evident progress, the numerical prediction of complex viscoelastic flows remains a difficult task whose success is not guaranteed. Recent mathematical and numerical results have identified a number of difficult challenges for the numericist.

Boundary layers, stress singularities, bifurcations, turning points, changes of type, and loss of evolution are potential features of current formulations of viscoelastic flows. Some of these features may reflect the actual physics of polymer flows, while others may only signal the inadequacy of the mathematical model. Uncertainties regarding the mathematical description of polymer flows constitute in our opinion the major difficulty facing those involved in the prediction of viscoelastic effects. Much research is needed at both experimental, theoretical, and numerical levels before the numerical simulation of viscoelastic flows realize its full potential and become a routine design tool in the polymer processing industry.

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