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MICRO-MACRO METHODS FOR THE MULTISCALE SIMULATION OF VISCOELASTIC FLOW USING MOLECULAR MODELS OF KINETIC THEORY

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ABSTRACT

We survey the field of micro-macro numerical techniques for predicting complex flows of viscoelastic fluids. The micro-macro approach couples the mesoscopic scale of kinetic theory to the macroscopic scale of continuum mechanics. A numerical solution is sought to the coupled non-linear problem involving the conservation laws and a microstructural model of kinetic theory. Although micro-macro techniques are much more demanding in terms of computer resources than conventional continuum computations, they allow the direct use of kinetic theory models in flow simulations, thus avoiding potentially inaccurate closure approximations. The focus of our survey is mainly put on mathematical formulations and numerical approaches. Applications to polymer solutions and melts, liquid crystalline polymers, and fibre suspensions, are briefly reviewed.

KEYWORDS: Viscoelastic fluids, kinetic theory, micro-macro methods.

1. INTRODUCTION

Many natural and synthetic fluids are viscoelastic materials, in the sense that the stress endured by a macroscopic fluid element depends upon the history of the deformation experienced by that element. Notable examples include polymer solutions and melts, liquid crystalline polymers, and fibre suspensions. The remarkable rheological properties of viscoelastic liquids are governed by the flow-induced evolution of molecular configurations. Furthermore, the frozen-in microstructure which develops in processing flows dictates the physical properties of the final product (Larson [1], Tanner [2], Rubinstein and Colby [3]). Rheologists thus face a challenging non-linear coupling between flow-induced evolution of molecular configurations, macroscopic rheological response, flow parameters (such as geometry and boundary conditions), and final product properties. Theoretical modelling and methods of computational rheology have an important role to play in elucidating this coupling.

Atomistic modelling is the most detailed level of description that can be applied today in rheological studies, using techniques of non-equilibrium molecular dynamics. Atomistic flow simulations have been conducted recently to study the behaviour of polymers in the vicinity of solid walls and geometrical singu-

larities such as a re-entrant corner. Wall slip and rupture phenomena can be studied in this fashion (e.g. Koplik and Banavar [4], Cieplak et al [5], Busic et al [6]). Such calculations require enormous computer resources, however, and the atomistic approach is currently limited to flow geometries of molecular dimensions. Consideration of macroscopic flows found in processing applications calls for less detailed mesoscopic models, such as those of kinetic theory.

Models of kinetic theory provide a coarse-grained description of molecular configurations wherein atomistic processes are ignored altogether (Doi and Edwards [7], Bird et al [8]). They are meant to display in a more or less accurate fashion the important features that govern the flow-induced evolution of configurations. For example, a linear polymer chain in a viscous solvent can be described by several models of decreasing complexity: the freely jointed, bead-rod Kramers chain made of a number of beads (of order 100) connected linearly by rigid segments, the bead-spring chain made of a smaller number of beads (of order 10) connected by entropic springs, or the single dumbbell, namely two beads connected by a spring. Over the last few years, these models of dilute polymer solutions have been evaluated in simple flows (shear, extension) by means of stochastic simulation or Brownian dynamics methods (e.g. Liu [9], Rallison [10], Doyle et al [11], Sizaire et al [12], Larson et al [13], Li et al [14], Somasi et al [15], Jendrejack et al [16], Hsieh et al [17]). In combination with the direct experimental observation of flow-induced configurations of single polymers (e.g. Perkins et al [18], Schroeder et al [19]), these studies have significantly increased our understanding of polymer dynamics in dilute solutions. In particular, the contribution of individual polymers on the overall dynamics has been shown to yield remarkable hysteresis effects in flows involving stress growth followed by relaxation.

In recent years, kinetic theory of entangled systems, such as concentrated polymer solutions and polymer melts, has known major developments that go well beyond the classical reptation tube model developed by Edwards, de Gennes, and Doi. The basic Doi-Edwards theory of linear entangled polymers cannot be used as such for simulating complex flows, as it predicts a material instability due to excessive shear-thinning beyond some critical deformation rate (Doi and Edwards [7]). In addition to reptation, other physical mechanisms such as convective constraint release, contour length fluctuations, and tube stretch, have been shown to play an important role. The most recent tube models take account of these effects and correct many of the deficiencies of the basic theory (Mead et al [20], Öttinger [21], Graham et al [22]). Finally, full-chain and temporary network models suited for Brownian dynamics simulations have also been put forward recently (Ianniruberto et al [23], Hua and Schieber [24], Neergaard and Schieber [25], Schieber et al [26], Masubuchi et al [27]). Reviews of modern tube theory are given by McLeish [28, 29] and Marrucci [30].

Kinetic theory models can be very complicated mathematical objects. It is usually not easy to compute their rheological response in rheometrical flows (although these flows have simple, specified kinematics), and their use in numerical simulations of complex flows has long been thought impossible. The traditional approach has been to derive from a particular kinetic theory model

a macroscopic constitutive equation that relates the viscoelastic stress to the deformation history. One then solves the constitutive model together with the conservation laws using a suitable numerical method, to predict velocity and stress fields in complex flows. The majority of constitutive equations used in continuum numerical simulations are indeed derived (or at least very much inspired) from kinetic theory (Bird et al [8]). Promising molecular-based constitutive equations have been proposed recently for dilute polymer solutions (Lielens et al [31], Ghosh et al [32], Zhou and Akhavan [33]), as well as for linear and branched entangled polymers (McLeish and Larson [34], Wagner et al [35], Marrucci and Ianniruberto [36], Likhtman and Graham [37]). Their use in numerical simulations of complex flows is reviewed by Owens and Phillips [38] and Keunings [39]. Clearly, the continuum approach remains an essential component of theoretical and computational rheology. There is however a basic issue in the above scheme which motivates the development of the complementary micro-macro approach.

Indeed, derivation of a constitutive equation from a model of kinetic theory usually involves closure approximations of a purely mathematical nature, such as decoupling or pre-averaging. It is now widely accepted that closure approximations can have a significant impact on rheological predictions for dilute polymer solutions (van den Brule [40], Keunings [41], Lielens et al [42], van Heel et al [43], Zhou and Akhavan [44]). For entangled systems, the impact of pre-averaging assumptions is expected to be less significant, but a careful investigation is lacking (McLeish [29]). The closure problem also arises in other applications of kinetic theory, such as liquid crystalline polymers (Grosso et al [45], Suen et al [46]), reversible networks of associating polymers (Vaccaro and Marrucci [47], Cifre et al [48]), and fibre suspensions (VerWeyst and Tucker [49], Chung and Kwon [50]). Use of closure approximations also raises the important issue of thermodynamic consistency (Edwards [51], Beris [52]).

In this context, micro-macro methods of computational rheology that couple the coarse-grained molecular scale of kinetic theory to the macroscopic scale of continuum mechanics have an important role to play. In a micro-macro simulation, the conservation equations are solved together with a model of kinetic theory. This approach is much more demanding in computer resources than more conventional continuum simulations that integrate a constitutive equation to evaluate the viscoelastic contribution to the stress tensor. On the other hand, micro-macro techniques allow the direct use of kinetic theory models and thus avoid potentially harmful closure approximations. Micro-macro methods have been introduced in the pioneering works by Biller and Petruccione [53, 54] and Fan [55]. Since the early 1990's, the field has developed considerably following the introduction of the CONNFFESSIT method by Öttinger and Laso [56, 57]. Being relatively new, micro-macro techniques have to date been implemented only for models of kinetic theory with few configurational degrees of freedom, such as non-linear dumbbell models of dilute polymer solutions and single-segment tube models of linear entangled polymers. Nevertheless, their potential range of applications is very wide indeed. Successive reviews of the subject matter have been written by Keunings [58], Owens and Phillips [38],

and Suen et al [46]. The goal of the present work is to survey the current state of the art.

2. MULTISCALE FORMULATION OF VISCOELASTIC FLOW

For the sake of illustration, consider a dilute solution of linear flexible polymers in a Newtonian solvent. The configuration of an individual polymer chain is influenced by a number of mechanisms, including Brownian, elastic, and drag forces[1]. Indeed, the solvent molecules undergo thermal agitation and continuously impact the polymer chain; this results in a diffusion process driven by a stochastic, Brownian force. Moreover, an elastic restoring force of entropic nature arises when the chain uncoils. The solvent also exerts viscous drag along the chain which affects the polymer configuration when different parts of the chain feel a different velocity, i.e. when a velocity gradient is imposed on the fluid.

In the framework of kinetic theory, the molecular configuration of an individual polymer chain can be defined in a rather coarse fashion by the vector $\boldsymbol{X}$ connecting the two chain ends. This simple approach provides a measure of both molecular orientation and stretch. The set of possible values for $\boldsymbol{X}$ is called the configuration space C, which has here a dimension N_C equal to 3.

Within each macroscopic fluid element, located at position $\boldsymbol{x}$ in the flow domain Ω, there is a large collection of polymer chains characterised at time t by a probability distribution of configurations $\psi(\boldsymbol{X}, \boldsymbol{x}, t)$. The distribution function ψ is such that $\psi(\boldsymbol{X}, \boldsymbol{x}, t) d\boldsymbol{X}$ gives the probability of finding a polymer with configuration between $\boldsymbol{X}$ and $\boldsymbol{X} + d\boldsymbol{X}$ at time t and position $\boldsymbol{x}$.

Under equilibrium conditions, namely when the macroscopic velocity vector field $\boldsymbol{v}$ vanishes identically, configurations are distributed according to the equilibrium probability function $\psi_{\mathrm{eq}}(\boldsymbol{X})$ that results, in the present illustrative model, from a balance between Brownian and elastic forces. Furthermore, the polymer or viscoelastic contribution to the stress, which we denote by the second-order tensor $\boldsymbol{\tau}_p$, reduces to an isotropic tensor of no rheological importance.

Under non-equilibrium conditions, namely when the polymer solution undergoes a macroscopic flow with velocity field $\boldsymbol{v}(\boldsymbol{x}, t)$, the polymer chains also feel a velocity gradient $\boldsymbol{\nabla v}(\boldsymbol{x}, t)$, and viscous drag exerted by the solvent affects their configuration. The distribution function is no longer equal to the equilibrium value $\psi_{\mathrm{eq}}(\boldsymbol{X})$, and it generally varies both in space and time, namely $\psi = \psi(\boldsymbol{X}, \boldsymbol{x}, t)$. Furthermore, the flow-induced distribution of configurations induces a viscoelastic stress field $\boldsymbol{\tau}_p(\boldsymbol{x}, t)$ resulting from anisotropic orientation and stretch of the polymer chains.

Thus, while the kinematics alter the distribution of molecular configurations along the flow trajectories, the stress experienced by each macroscopic fluid element is itself governed by the distribution of configurations within that element. Furthermore, velocity and stress fields are coupled through the conser-

[1] We ignore in this discussion important mechanisms related to excluded volume and hydrodynamic interactions (Bird et al [8]).

vation principles. Clearly, predicting viscoelastic flow using a molecular model of kinetic theory is a challenging non-linear problem. Let us briefly consider its generic mathematical formulation.

Kinetic theory provides two basic building blocks: the diffusion or Fokker-Planck equation that governs the evolution of the distribution function, and an expression relating the viscoelastic stress to the distribution function (Doi and Edwards [7], Bird et al [8], Öttinger [59]). The Fokker-Planck equation has the general form

$$\frac{D\psi}{Dt} = -\frac{\partial}{\partial \boldsymbol{X}} \cdot \{\boldsymbol{A}\,\psi\} + \frac{1}{2}\frac{\partial}{\partial \boldsymbol{X}}\frac{\partial}{\partial \boldsymbol{X}} : \{\boldsymbol{D}\,\psi\}. \tag{1}$$

Here, the symbol D/Dt denotes the Lagrangian or material derivative $\partial/\partial t + \boldsymbol{v}\cdot\boldsymbol{\nabla}$, where $\boldsymbol{\nabla}$ is the del operator with respect to position $\boldsymbol{x}$ in physical space. Molecules are thus assumed to be convected by the macroscopic velocity field $\boldsymbol{v}$. The vector $\boldsymbol{X}$ defines the coarse-grained configuration and has dimension N_C. The factor $\boldsymbol{A}$ is an N_C-dimensional vector that defines the drift or deterministic component of the molecular model. The macroscopic velocity gradient $\boldsymbol{\nabla v}$ enters in the formulation of $\boldsymbol{A}$, and is assumed constant over the molecular length scale. Finally, $\boldsymbol{D}$ is a symmetric, positive definite $N_C \times N_C$ matrix that embodies the diffusive or stochastic component of the molecular model. In general, both $\boldsymbol{A}$ and $\boldsymbol{D}$ depend upon $(\boldsymbol{X}, \boldsymbol{x}, t)$. Suitable boundary conditions must be specified for ψ, in both configuration and physical spaces.

The second building block of a kinetic theory model is an expression relating stress to molecular configurations. It takes the form of a particular average computed with the distribution function over all possible configurations:

$$\boldsymbol{\tau}_p = \int_C \boldsymbol{g}(\boldsymbol{X})\,\psi\,d\boldsymbol{X} = \langle \boldsymbol{g}(\boldsymbol{X})\rangle. \tag{2}$$

Here, $\boldsymbol{g}$ is a model-dependent tensorial function of configuration. Thus, for a given velocity field, and starting from a specified initial condition for ψ at time t_0, integration of the Fokker-Planck equation (1) yields the distribution function at all time $t \geq t_0$, everywhere in the flow domain Ω. Use of the stress-configuration relation (2) then provides the viscoelastic stress field.

In a complex flow, the velocity field is a priori unknown; velocity and stress fields are coupled through the conservation laws. Consider isothermal, incompressible flows. Conservation of mass and linear momentum[2] is then expressed as

$$\boldsymbol{\nabla}\cdot\boldsymbol{v} = 0, \qquad \varrho\frac{D\boldsymbol{v}}{Dt} = \boldsymbol{\nabla}\cdot\{-p\boldsymbol{I} + \boldsymbol{\tau}_p + \eta_s\dot{\boldsymbol{\gamma}}\}, \tag{3}$$

where ϱ is the fluid density, p is the pressure, $\boldsymbol{I}$ is the unit tensor, and $\eta_s\dot{\boldsymbol{\gamma}}$ is a purely viscous component to the stress which involves the rate of strain tensor $\dot{\boldsymbol{\gamma}} = \boldsymbol{\nabla v} + \boldsymbol{\nabla v}^T$ and a constant viscosity coefficient η_s (Bird et al [60]). The Newtonian stress can be interpreted physically as the solvent contribution

[2]Body forces are ignored.

to the stress in polymer solutions, or as the stress response associated with fast relaxation modes. In other cases, the Newtonian component is added to correct the molecular model itself (e.g., to eliminate excessive shear-thinning). Appropriate boundary conditions supplement the conservation laws (3). One specifies components of either the velocity or the contact force at the boundary $\partial\Omega$ of the flow domain Ω; the pressure is specified at one point of the flow domain if no normal contact forces have been specified anywhere at the boundary. In view of the fluid's memory, additional boundary conditions must be specified when the flow domain Ω contains an inlet boundary. Specification of the flow pre-history is often achieved by assuming fully-developed flow upstream of the inlet boundary. For transient flows, initial conditions are also needed for the distribution function and the velocity.

The set of coupled equations (1) to (3), supplemented with suitable initial and boundary conditions in both physical and configuration spaces, is the generic multiscale formulation of viscoelastic flow that we consider in the present review. Specific examples pertaining to polymer solutions and melts are given in the next section. To date, three basic approaches have been adopted for exploiting the generic multiscale model:

The continuum approach, wherein a constitutive equation of continuum mechanics that relates the viscoelastic stress to the deformation history is derived from, and replaces altogether, the kinetic theory model (1) and (2). The derivation process usually involves closure approximations. The resulting constitutive model takes the form of a differential, integral, or integro-differential equation. It yields molecular information in terms of averaged quantities, such as the second moment $\langle \boldsymbol{X}\boldsymbol{X} \rangle$ of the distribution;

The Fokker-Planck approach, wherein one solves the generic problem (1) to (3) as such, in both configuration and physical spaces. The distribution function is thus computed explicitly as a solution of the Fokker-Planck equation. The viscoelastic stress is merely a by-product obtained from (2);

The stochastic approach, which draws on the mathematical equivalence between the Fokker-Planck equation (1) and the following Itô stochastic differential equation [3]:

$$d\boldsymbol{X} = \boldsymbol{A}\, dt + \boldsymbol{B} \cdot d\boldsymbol{W}, \tag{4}$$

where $\boldsymbol{D} = \boldsymbol{B} \cdot \boldsymbol{B}^T$ and $\boldsymbol{W}$ is a Wiener stochastic process of dimension N_C (Öttinger [59]). In a complex flow, the stochastic differential equation (4) applies along individual flow trajectories; the time derivative is thus a material derivative. Instead of solving the deterministic Fokker-Planck equation (1), one solves the associated stochastic differential equation (4) for a large ensemble of

[3] The Fokker-Planck equation (1) may also contain a source term, as in the tube model proposed by Öttinger [21]. For models with mean-field interactions, such as the Doi-Edwards model without independent alignment and the Doi model for liquid crystalline polymers, the Fokker-Planck equation is non-linear in ψ due to the dependence of $\boldsymbol{A}$ and $\boldsymbol{D}$ on averages computed with ψ (Öttinger [59]). These more complex cases can also be formulated in a stochastic setting.

realisations of the stochastic process $\boldsymbol{X}$ by means of a suitable numerical technique (Kloeden and Platen [61]). The distribution function is not computed explicitly, and the viscoelastic stress (2) is readily obtained as an ensemble average.

As a matter of fact, the continuum approach outlined above has been adopted throughout the development of computational rheology. In particular, the first successful finite element simulations of die swell and laminar jet breakup in viscoelastic liquids were obtained in the early 1980's with the Oldroyd-B constitutive equation (Crochet and Keunings [62], Keunings [63]). The latter is mathematically equivalent to the simplest kinetic theory model of a dilute solution of flexible linear polymers in a Newtonian solvent, known as the Hookean dumbbell model (Bird et al [8]). Two decades later, macroscopic numerical techniques based upon the continuum approach remain under active development. For a detailed account of the state of the art, see the monograph by Owens and Phillips [38] and the review by Keunings [39]. Much of the recent work in continuum simulations has been devoted to the evaluation in complex flows of tube-based constitutive equations for entangled polymers (Peters et al [64], Lee et al [65], Rasmussen [66], Wapperom et al [67], Bent et al [68], Wapperom and Keunings [69, 70]).

The present review focuses on the micro-macro methods needed to implement the Fokker-Planck and stochastic approaches. To the best of our knowledge, the idea of combining in a self-consistent way the conservation laws and a model of kinetic theory was first put forward by Biller and Petruccione [53, 54], using the stochastic approach. The context of their work is somewhat different from the one formulated in this section. Indeed, the authors considered planar Couette [53] and tube flow [54] of dilute polymer solutions in confined geometries with a characteristic size (i.e. the gap between the two plates and the tube radius) that is comparable to the molecular size. In such calculations, the velocity gradient varies on the polymer length scale. The centre of mass of individual polymers (modelled as dumbbells) cannot be assumed to be simply convected by the macroscopic flow, but rather is allowed to migrate across flow trajectories.

Before describing micro-macro techniques in some detail, we consider briefly the FENE dumbbell and Doi-Edwards models. As discussed in the introduction, these two models are not representative of the state-of-the-art in kinetic theory. They do however display enough complexity for our purpose.

3. TWO BASIC EXAMPLES OF KINETIC THEORY MODELS

The FENE dumbbell model

The simplest non-linear kinetic theory model of a dilute polymer solution is known as the Finitely Extensible Non-linear Elastic (FENE) dumbbell model (Bird et al [8]). The polymer solution is viewed as a flowing suspension of dumbbells that do not interact with each other and are convected by the Newtonian solvent. Each dumbbell consists of two identical Brownian beads connected by an entropic spring. Here, the configuration $\boldsymbol{X}$ is the three-dimensional vector

$\boldsymbol{Q}$ connecting the two beads. A FENE dumbbell cannot extend beyond the polymer contour length Q_0. Thus, the configuration space C is the set of three-dimensional vectors of magnitude less than Q_0. The Fokker-Planck equation (1) for the distribution function $\psi(\boldsymbol{Q}, \boldsymbol{x}, t)$ reads

$$\frac{D\psi}{Dt} = -\frac{\partial}{\partial \boldsymbol{Q}} \cdot \left[\left\{ \boldsymbol{\kappa} \cdot \boldsymbol{Q} - \frac{2}{\zeta} \, \boldsymbol{F}^c(\boldsymbol{Q}) \right\} \psi \right] + \frac{2kT}{\zeta} \, \frac{\partial}{\partial \boldsymbol{Q}} \cdot \frac{\partial}{\partial \boldsymbol{Q}} \, \psi, \tag{5}$$

where $\boldsymbol{\kappa} = \boldsymbol{\nabla} \boldsymbol{v}^T$ is the transpose of the velocity gradient, ζ is a friction coefficient, $\boldsymbol{F}^c(\boldsymbol{Q})$ is the entropic connector force, T is the absolute temperature, and k is the Boltzmann constant. The distribution function vanishes at the boundary of the configuration space C, namely for all $\boldsymbol{Q}$ of magnitude Q_0. We consider entropic springs that follow Warner's force law [71],

$$\boldsymbol{F}^c(\boldsymbol{Q}) = \frac{H}{1 - \boldsymbol{Q}^2/Q_0^2} \, \boldsymbol{Q}, \tag{6}$$

where H is a spring constant. The stress-configuration relation (2) is the classical Kramers expression

$$\boldsymbol{\tau}_p = n \, \langle \boldsymbol{Q} \boldsymbol{F}^c(\boldsymbol{Q}) \rangle, \tag{7}$$

where n is the dumbbell number density and the angular brackets $\langle \cdot \rangle$ denote the configuration space average $\int_C \cdot \, \psi \, d\boldsymbol{Q}$. Finally, the Itô stochastic differential equation (4) that is equivalent to (5) reads

$$d\boldsymbol{Q} = [\boldsymbol{\kappa} \cdot \boldsymbol{Q} - \frac{2}{\zeta} \boldsymbol{F}^c(\boldsymbol{Q})] \, dt + \sqrt{\frac{4kT}{\zeta}} \, d\boldsymbol{W}, \tag{8}$$

where $\boldsymbol{W}$ is the three-dimensional Wiener process, namely a Gaussian stochastic process with vanishing mean and covariance $\langle \boldsymbol{W}(t_1) \, \boldsymbol{W}(t_2) \rangle = \min(t_1, t_2) \, \boldsymbol{I}$.

The FENE dumbbell model does not have a mathematically equivalent, closed-form constitutive equation[4]. A closure approximation is thus needed to exploit the model in continuum simulations. The simplest closure, due to Peterlin, replaces the FENE spring force (6) by the pre-averaged FENE-P approximation

$$\boldsymbol{F}^c(\boldsymbol{Q}) \approx \frac{H}{1 - \langle \boldsymbol{Q}^2 \rangle / Q_0^2} \, \boldsymbol{Q}. \tag{9}$$

One thus obtains from (5) and (7) the FENE-P constitutive equation, namely a differential equation for the configuration tensor $\boldsymbol{A} = \langle \boldsymbol{Q} \boldsymbol{Q} \rangle$:

$$\frac{D\boldsymbol{A}}{Dt} - \boldsymbol{\kappa} \cdot \boldsymbol{A} - \boldsymbol{A} \cdot \boldsymbol{\kappa}^T = \frac{4kT}{\zeta} \, \boldsymbol{I} - \frac{4H/\zeta}{1 - \mathrm{tr}(\boldsymbol{A})/Q_0^2} \, \boldsymbol{A}, \tag{10}$$

supplemented by an algebraic expression relating stress to average configuration,

$$\boldsymbol{\tau}_p = n \, \frac{H}{1 - \mathrm{tr}(\boldsymbol{A})/Q_0^2} \, \boldsymbol{A}. \tag{11}$$

[4]The limit of infinitely extensible dumbbells ($Q_0 \to +\infty$) is the Hookean dumbbell model, which is equivalent to the Oldroyd-B constitutive equation.

The Peterlin approximation can be very poor indeed (Keunings [41], Sizaire et al [12]), and much better closure approximations are available (Lielens et al [31, 42]). At any rate, closure-approximated dumbbell models (such as FENE-P) are very useful in the development and evaluation of micro-macro methods, since the micro-macro results can be compared to those obtained with the continuum approach.

The Doi-Edwards model with independent alignment

The simplest tube model of entangled linear polymers is the classical Doi-Edwards model with independent alignment. Reptation is the single relaxation mechanism described by the model, and the dynamics of individual tube segments are decoupled. Constraint release (thermal or convective), tube stretch, and contour length fluctuations are ignored (Doi and Edwards [7]). The configuration $\boldsymbol{X}$ is defined by the unit orientation vector $\boldsymbol{u}$ of a tube segment and the normalized contour label $s \in [0,1]$ of the primitive chain, where $s = 0$ and $s = 1$ correspond to the chain ends. The distribution function $\psi(\boldsymbol{u}, s, \boldsymbol{x}, t)$ is such that $\psi(\boldsymbol{u}, s, \boldsymbol{x}, t) d\boldsymbol{u} ds$ is the joint probability that at time t and position $\boldsymbol{x}$ a tube segment has an orientation in the interval $[\boldsymbol{u}, \boldsymbol{u} + d\boldsymbol{u}]$ and contains the chain segment labelled in the interval $[s, s + ds]$. Thus, the configuration space C is $B(0,1) \times [0,1]$, where $B(0,1)$ is the surface of the unit sphere centered at the origin. The Fokker-Planck equation (1) for the Doi-Edwards model reads

$$\frac{D\psi}{Dt} = -\frac{\partial}{\partial \boldsymbol{u}} \cdot [(\boldsymbol{I} - \boldsymbol{u}\boldsymbol{u}) \cdot \boldsymbol{\kappa} \cdot \boldsymbol{u}\, \psi] + \frac{1}{\pi^2 \tau_d} \frac{\partial^2 \psi}{\partial s^2}, \tag{12}$$

where τ_d is the disengagement time, namely the characteristic time for a chain to escape from its tube by reptation. The boundary conditions in configuration space specify that ψ is isotropic at $s = 0$ and 1, namely

$$\psi(\boldsymbol{u}, 0, \boldsymbol{x}, t) = \psi(\boldsymbol{u}, 1, \boldsymbol{x}, t) = \frac{1}{4\pi} \delta(|\boldsymbol{u}| - 1), \tag{13}$$

where δ is the Dirac delta function. The stress-configuration relation (2) reads

$$\boldsymbol{\tau}_p = G\, \langle \boldsymbol{u}\boldsymbol{u} \rangle, \tag{14}$$

where G is an elastic modulus, and the angular brackets $\langle \cdot \rangle$ denote the configuration space average $\int_0^1 \int_{B(0,1)} \cdot\, \psi\, d\boldsymbol{u} ds$. Finally, the Itô stochastic differential equation (4) that is equivalent to (12) is given by

$$d\boldsymbol{u} = (\boldsymbol{I} - \boldsymbol{u}\boldsymbol{u}) \cdot \boldsymbol{\kappa} \cdot \boldsymbol{u}\, dt,\ ds = \sqrt{\frac{2}{\pi^2 \tau_d}}\, dW, \tag{15}$$

where W is the one-dimensional Wiener process. The evolution equation for $\boldsymbol{u}$ is deterministic; it expresses the assumption that tube segments orient with the macroscopic flow. The equation for s is that of a purely-diffusive process. The coupling between $\boldsymbol{u}$ and s arises through the boundary conditions (13): when

the process s reaches either 0 or 1, it is reflected and $\boldsymbol{u}$ is reset to a randomly oriented unit vector (Öttinger [59]).

In view of its (relative) simplicity, the basic Doi-Edwards model can be cast into an equivalent integral constitutive model. Consider a fluid particle whose position at present time t is given by $\boldsymbol{x}$; its position at some past time t' is $\boldsymbol{x}'$. The particle motion is described by the displacement function $\boldsymbol{x}' = \boldsymbol{x}'(\boldsymbol{x}, t, t')$, while the deformation gradient $\boldsymbol{F}(t, t')$ is defined as $\partial \boldsymbol{x}'/\partial \boldsymbol{x}$. The Doi-Edwards integral constitutive model is formulated in a Lagrangian framework: it gives the viscoelastic stress $\boldsymbol{\tau}_p$ at a moving fluid particle, according to

$$\boldsymbol{\tau}_p(t) = G \int_{-\infty}^{t} m(t, t') \boldsymbol{Q}(t, t')\, dt', \tag{16}$$

where the integral is computed along the past trajectory of the fluid particle, parameterised by the past time t'. Here, m is a memory function given by

$$m(t, t') = \frac{8}{\pi^2 \tau_d} \sum_{k=0}^{\infty} \exp(-\frac{(2k+1)^2 (t - t')}{\tau_d}), \tag{17}$$

and $\boldsymbol{Q}$ is a strain measure defined as

$$\boldsymbol{Q}(t, t') = \langle \boldsymbol{u}\boldsymbol{u} \rangle_{t'} = \langle \frac{(\boldsymbol{F}(t, t') \cdot \boldsymbol{u}(t'))(\boldsymbol{F}(t, t') \cdot \boldsymbol{u}(t'))}{|\boldsymbol{F}(t, t') \cdot \boldsymbol{u}(t')|^2} \rangle_{t'}, \tag{18}$$

where $\langle \cdot \rangle_{t'}$ denotes the average computed with the isotropic distribution (13). Thus, simulations based on the basic Doi-Edwards tube model[5] can be performed either with a micro-macro numerical method, or with a continuum technique for integral constitutive equations such as the method of deformation fields introduced by Peters et al [72]. Again, this is very useful for the validation of numerical developments. More sophisticated tube models that take account of additional physics such as stretch and constraint release generally do not have an equivalent closed-form constitutive equation (Mead et al [20], Öttinger [21], Graham et al [22]) .

4. THE STOCHASTIC APPROACH

CONNFFESSIT

The CONNFFESSIT method introduced in 1992 by Öttinger and Laso [56, 57] is the first implementation of the stochastic approach defined in Section 2. The acronym stands for "**C**alculation **O**f **N**on-**N**ewtonian **F**low: **F**inite **E**lements and **S**tochastic **SI**mulation **T**echnique". Although it can deal with

[5] Such calculations are useful for numerical purposes, but have limited rheological relevance. Indeed, a small Newtonian stress must be added to the viscoelastic stress in the momentum equation (3) to correct the excessive shear-thinning of the basic Doi-Edwards model. Unfortunately, this procedure is not an innocuous fix: the viscous component dominates the fluid's response in the stress boundary layers that are often predicted in the vicinity of no-slip solid walls.

Eulerian steady flows (Feigl et al [73]), CONNFFESSIT is most naturally exploited as a time-marching scheme. A steady-state flow, if it exists, is then obtained as the long-time limit of the calculations (Laso et al [74]). At the start of a simulation, a large number of model molecules (such as dumbbells, bead-spring chains, or any other coarse-grained model) is distributed uniformly over the entire flow domain. Their initial configuration is drawn from the equilibrium distribution. As the simulation proceeds, the molecules are convected along flow trajectories. At each time step, solution of the conservation laws (3) is decoupled from the integration of the stochastic differential equation (4) for each molecule. More precisely, a typical time step is defined by the following algorithm:

S1 Solve the conservation equations (3) by means of a standard finite element method to obtain updated values for the velocity and pressure fields, treating the current approximation to the viscoelastic stress field as a known body force term in the momentum equation;

S2 Using the updated velocity field, compute the path of each model molecule convected by the macroscopic flow;

S3 For each molecule, integrate the stochastic differential equation (4) along the molecule's path to update its configuration;

S4 Update the viscoelastic stress field by means of the average (2) computed over the local ensemble of molecules currently located within each finite element.

The implementation by Laso et al [74] is for two-dimensional flows of FENE-P and FENE dumbbells. A global ensemble of N_g molecules is distributed over the elements of the mesh. Consider a typical time step $t_n \to t_{n+1} = t_n + \Delta t$. S1 is performed using a classical penalty finite element technique, with bi-linear quadrilateral elements for the velocity. Each quadrilateral element is split into two triangles over which the viscoelastic stress has a constant value assigned to the central Gauss integration point. The time-stepping scheme is implicit in the velocity and explicit in the viscoelastic stress. The body force term $\boldsymbol{\nabla} \cdot \boldsymbol{\tau}_p$ in the momentum equation is evaluated with stress values known at time t_n. Thus, S1 yields the approximated velocity field $\boldsymbol{v}^a$ at t_{n+1}. S2 is the classical problem of particle tracking in a given finite element velocity field. The updated location $\boldsymbol{r}^j_{n+1}$ of the centre of mass of the jth molecule ($j = 1, 2, ..., N_g$) at time t_{n+1} is obtained by integrating

$$\frac{d\boldsymbol{r}^j}{dt} = \boldsymbol{v}^a(\boldsymbol{r}^j, t), \tag{19}$$

using the explicit, first-order Euler scheme and the initial condition $\boldsymbol{r}^j_n$ known from the previous time step. The simplest numerical technique for implementing S3 is the Euler-Maruyama scheme,

$$\boldsymbol{X}^j_{n+1} = \boldsymbol{X}^j_n + \boldsymbol{A}(\boldsymbol{X}^j_n, t_n)\Delta t + \boldsymbol{B}(\boldsymbol{X}^j_n, t_n) \cdot \Delta \boldsymbol{W}^j_n. \tag{20}$$

Here, $\boldsymbol{X}_i^j$ denotes the configuration of the jth molecule at time t_i, and the random vector $\Delta \boldsymbol{W}_n^j$ has independent Gaussian components with zero mean and variance Δt. Note that each model molecule feels its own Brownian random force, namely $\Delta \boldsymbol{W}_n^{j_1}$ and $\Delta \boldsymbol{W}_n^{j_2}$ are uncorrelated for $j_1 \neq j_2$. The Euler-Maruyama scheme is of weak order 1. When used with FENE dumbbells, it can lead to difficulties if Δt is too large. Indeed, the length of an individual dumbbell can become larger than the upper bound Q_0, which is unphysical. Use of the predictor-corrector scheme of weak order 2 proposed by Öttinger [59] solves this problem. Finally, S4 amounts to approximating the stress-configuration relation (2) by means of a local ensemble average computed in each element (or possibly sub-element) of the mesh. The updated viscoelastic stress in element k is thus given by

$$\boldsymbol{\tau}_{p,n+1}^k = \frac{1}{N_{\text{loc}}^k} \sum_l \boldsymbol{g}(\boldsymbol{X}_{n+1}^l). \tag{21}$$

Here, the counter l runs over the indices of the N_{loc}^k molecules that are currently present in the kth element. The time step $t_n \rightarrow t_{n+1}$ is thus completed.

Applications of the CONNFFESSIT approach to dumbbell models of dilute polymer solutions, tube models of entangled polymers, colloidal dispersions, and liquid crystalline polymers, have been reported for steady-state and transient two-dimensional flows (Feigl et al [73], Laso et al [74], Laso [75], Öttinger and Laso [76], Hua and Schieber [77, 78]). Extension of CONNFFESSIT to free surface flows is performed by Cormenzana et al [79] and Grande et al [80].

Three challenges and one breakthrough

The original CONNFFESSIT scheme has had a significant impact on the development of micro-macro techniques. It does however present three basic difficulties. First, tracking the motion of the molecules with a simple explicit scheme can be inaccurate in complex flow geometries, especially in the vicinity of impervious walls where numerical errors can result in molecules leaving the flow domain. Tracking all N_g individual molecules is also very expensive. At every time step, one must know the index of the finite element in which each molecule is currently located. Given the large number of molecules and elements, typically $N_g = O(10^5 - 10^7)$ and $N_{\text{elt}} = O(10^3 - 10^4)$ in two-dimensional simulations, one cannot use a brute force search of complexity $O(N_{\text{elt}} \times N_g)$ at each time step. Use of element neighbour lists ordered on the basis of the local direction of flow reduces the complexity of tracking to $O(N_g)$ [74]. The second difficulty is related to the statistical accuracy of the viscoelastic stress. For a given velocity field, the ensemble average (21) carries a statistical error $\sqrt{\Theta / N_{\text{loc}}^k}$, where $\Theta = Var[\boldsymbol{g}(\boldsymbol{X})]$ is the variance of $\boldsymbol{g}(\boldsymbol{X})$. Thus, each element of the mesh must, during the entire simulation, contain enough model molecules for the local ensemble average (21) to be accurate. Ideally, one should have $N_{\text{loc}}^k = O(10^2 - 10^3)$ molecules in each element and at all times. This is not easy (and maybe even impossible) to ensure, especially in flow regions where large spatial gradients develop in

the velocity and stress fields, and where one would precisely wish to use very small elements. A means of reducing the statistical error without increasing the size $N^k_{\rm loc}$ of the ensemble is to apply variance reduction methods, which we discuss shortly. The third difficulty is that CONNFFESSIT stress fields typically show spurious spatial oscillations which affect, via the momentum equation, the numerical accuracy of the velocity field. These basic issues are illustrated by Laso et al [74] and Halin et al [81] who compared continuum and CONNFFESSIT simulations for FENE-P dumbbells. They motivated the development of improved techniques which we review next.

A breakthrough came with the use of correlated local ensembles, first introduced by Hulsen et al [82] in their method of Brownian configuration fields. In this approach, the same local ensemble of model molecules is defined initially within each material element. As time evolves, the configurations of the kth molecule in all local ensembles is determined using the same sequence of random numbers, i.e. the sequence only depends on the index k. Thus, strong spatial correlations develop in the stress fluctuations within neighbouring material elements (which feel a very similar flow history), and evaluation of the divergence of the stress in the momentum equation leads to partial cancellation of the fluctuations. The method of Brownian configuration fields is an Eulerian implementation of correlated local ensembles (Öttinger et al [83]). Alternatively, the so-called Lagrangian particle methods introduced by Halin et al [84] provide a Lagrangian implementation of the same idea. In comparison with CONNFFESSIT, these techniques produce stress and velocity fields that are significantly smoother in space. Moreover, the cost of generating sequences of random numbers is reduced drastically. Of course, the use of correlated local ensembles introduces artificial spatial correlations. It is thus forbidden altogether in problems dominated by physical fluctuations (such as flows on a molecular length scale).

Brownian configuration fields

The method of Brownian configuration fields introduced by Hulsen et al [82] uses correlated ensembles of model molecules and completely avoids the tracking problem. Instead of computing the configuration of discrete molecules along flow trajectories, this method determines the evolution of a finite number of Eulerian configurations fields. Each field feels a random Wiener process that is uniform in space.

Let $\{\boldsymbol{X}_k(\boldsymbol{x},t)\}_{k=1}^{N_f}$ denote an ensemble of N_f configuration fields defined over the entire flow domain Ω. In a typical simulation, N_f is of order 10^3. The field $\boldsymbol{X}_k(\boldsymbol{x},t)$ represents the configuration of the kth model molecule in the local ensemble at position $\boldsymbol{x}$ and time t. At time $t=0$, $\boldsymbol{X}_k$ is set to a spatially uniform value $\boldsymbol{X}_k^0$, drawn from the equilibrium distribution for example; all kth molecules thus have the same initial configuration. The subsequent evolution of the field $\boldsymbol{X}_k$ is governed by

$$d\boldsymbol{X}_k(\boldsymbol{x},t) = \{-\boldsymbol{v}(\boldsymbol{x},t)\cdot\boldsymbol{\nabla}\boldsymbol{X}_k(\boldsymbol{x},t) + \boldsymbol{A}(\boldsymbol{X}_k(\boldsymbol{x},t))\}\,dt + \boldsymbol{B}(\boldsymbol{X}_k(\boldsymbol{x},t))\cdot d\boldsymbol{W}_k(t). \tag{22}$$

This is indeed the Eulerian formulation of the stochastic differential equation (4), with a familiar convection term $\boldsymbol{v}\cdot\boldsymbol{\nabla}\boldsymbol{X}_k$. Note however that the stochastic factor $d\boldsymbol{W}_k$ only depends on time. As a result, the spatial gradient of the configuration field is a well defined function of the spatial coordinates. One can thus discretize the field $\boldsymbol{X}_k$ by means of a standard finite element approximation. Also, in the numerical integration of (22), the same sequence of random numbers is used to determine, everywhere in the flow domain, the configuration of the kth model molecule in the local ensemble.

At any time t, an ensemble average computed over all configuration fields yields the approximated viscoelastic stress,

$$\boldsymbol{\tau}_p^a(\boldsymbol{x},t) = \frac{1}{N_f}\sum_{k=1}^{N_f}\boldsymbol{g}(\boldsymbol{X}_k(\boldsymbol{x},t)). \tag{23}$$

Note that the CONNFFESSIT local ensembles (21) have a size $N_{\rm loc}$ that varies both with time and position in the finite element mesh. In contrast, the ensemble average (23) involves a number N_f of realisations that is fixed and independent of mesh refinement. This provides a much more effective control of the statistical error. In their early implementation for Hookean dumbbells, Hulsen et al [82] solve the N_f evolution equations (22) for the configuration fields by means of a discontinuous Galerkin (DG) finite element method, with explicit Euler-Maruyama time stepping. In the DG approach, the interpolation functions are discontinuous across neighbouring elements, which induces only weak coupling between elements. Thus, at each time step, all configuration and stress calculations can be performed at the element level. Spatial discretization of the conservation laws is achieved by means of the Discrete Elastic-Viscous Split Stress (DEVSS) formulation of Guénette and Fortin [85].

Van Heel et al [86] have applied the method of configuration fields to the basic Doi-Edwards model described in Section 3. They define a set $\{\boldsymbol{u}_k(\boldsymbol{x},t), s_k(t)\}_{k=1}^{N_f}$ of N_f configuration fields $\boldsymbol{u}_k$ and associated random walkers s_k. In view of (15), the field $\boldsymbol{u}_k$ evolves according to

$$\frac{\partial}{\partial t}\boldsymbol{u}_k(\boldsymbol{x},t) + \boldsymbol{v}(\boldsymbol{x},t)\cdot\boldsymbol{\nabla}\boldsymbol{u}_k(\boldsymbol{x},t) = [\boldsymbol{I} - \boldsymbol{u}_k(\boldsymbol{x},t)\boldsymbol{u}_k(\boldsymbol{x},t)]\cdot\boldsymbol{\kappa}(\boldsymbol{x},t)\cdot\boldsymbol{u}_k(\boldsymbol{x},t), \tag{24}$$

while the random walker s_k performs Brownian motion,

$$ds_k = \sqrt{\frac{2}{\pi^2\tau_d}}\,dW_k(t), \tag{25}$$

with a reflecting boundary condition when it reaches 0 or 1. Note that s_k is not a function of position $\boldsymbol{x}$. Whenever s_k is reflected, the associated field $\boldsymbol{u}_k$

is instantaneously reset to a spatially uniform, random field drawn from the isotropic distribution on the surface of the unit sphere; its subsequent evolution is again governed by (24) until the next reflection of s_k. Here again, the method of configuration fields produces much smoother results than CONNFFESSIT. Indeed, in a typical CONNFFESSIT simulation of the Doi-Edwards model, individual tube segments are convected by the flow and have their own associated random walkers. Thus, not only are the tube segmental orientations reset at uncorrelated times (when the associated random walker is reflected), they are also reset to uncorrelated random values. This induces severe spatial oscillations in the numerical results for velocity and stress. Finally, we note that in more complex tube models (e.g. Öttinger [21]), the random walker s_k associated with the field $\boldsymbol{u}_k$ follows a stochastic differential equation that contains a deterministic drift term. As a result, the local velocity gradient has an impact on the evolution of s_k, and reflections of s_k are dependent on both time and space. The method of configuration fields can be modified to handle this complication (Gigras and Khomami [87]).

Other applications and extensions of the method of configuration fields have appeared recently. Simulations of fibre suspensions are reported by Phan-Thien and Fan [88], and Fan et al [89, 90]. The linear stability of simple flows involving FENE dumbbells is studied by Somasi and Khomami [91]. Chauvière and Lozinski [92] have derived from the method of configuration fields a new technique for computing viscoelastic flows with dumbbell models that is devoid of stochastic noise and is competitive with conventional continuum schemes (see also Chauvière [93]). In fact, the basic reason why the authors could derive noise-free expressions for the stress tensor is that they consider molecular models (Hookean and FENE-P dumbbells) which have Gaussian statistics and an equivalent constitutive equation. Their approach would not apply to FENE dumbbells, for example. Finally, Ramirez and Laso [94] have recently extended the method of Brownian configuration fields to handle three-dimensional flows.

Lagrangian particle methods

The Lagrangian particle method (LPM) introduced by Halin et al [84] is another further development of CONNFFESSIT. Here again, the conservation laws are solved at each time step by means of a standard finite element technique, with the viscoelastic stress known from the previous step. In LPM, the stress is computed at a number N_{part} of Lagrangian particles that are convected by the flow. Each Lagrangian particle carries a number N_d of model molecules. Tracking is thus performed for N_{part} particles instead of $N_{\text{part}} \times N_d$ molecules, using a very accurate fourth-order Runge-Kutta scheme within the parent element. Along the path of each particle, LPM solves the stochastic differential equation (4) for each of the N_d molecules. Note that LPM can be used either with uncorrelated or correlated local ensembles of model molecules. In the first case, $N_{\text{part}} \times N_d$ independent Wiener processes drive the stochastic evolution of molecular configurations, as in a CONNFFESSIT simulation. In the second case, the same initial ensemble of molecules is used in each Lagrangian particle,

and the same N_d independent Wiener processes are generated to compute the configuration of corresponding molecules in each particle. In fact, LPM with correlated ensembles of molecules can be viewed as a (Lagrangian) method of characteristics for solving the evolution equation (22) that governs a particular Brownian configuration field; the number N_f of fields then corresponds to the number N_d of molecules carried by each Lagrangian particle. The updated viscoelastic stress is computed at each Lagrangian particle by means of a local ensemble average involving the configurations of N_d molecules. At the end of a typical time step for the solution of the conservation laws, we have at our disposal values of the viscoelastic stress at discrete Lagrangian particles with a known position. These Lagrangian results then feed the Eulerian discretized momentum balance as follows: within each finite element, LPM computes the linear least-squares polynomial that best passes through the available Lagrangian stress data. Clearly, this procedure requires that at least three Lagrangian particles be present in each element at all times. LPM results obtained for non-linear dumbbells [84] are in excellent agreement with those provided by the method of configuration fields.

Use of LPM with highly graded finite element meshes requires a large number of Lagrangian particles. This motivated the development of the adaptive Lagrangian particle method (ALPM) by Gallez et al [95]. At each time step, ALPM ensures that all elements of the mesh have a number of Lagrangian particles in the user-specified interval $[N_{\text{part}}^{\text{elt,min}}, N_{\text{part}}^{\text{elt,max}}]$. This necessitates an elaborate adaptive procedure wherein Lagrangian particles are either created or destroyed. Once a new particle is created, the configuration of the local N_d molecules must be properly initialised in order to allow for the subsequent solution of the stochastic differential equation (4) along the particle's path. Initialisation is performed at the element level, by means of a least-squares approximation based on the current configuration of molecules in neighbouring particles. ALPM is superior to LPM both in terms of cost and numerical accuracy. Unfortunately, it is much more intricate to implement, and can only be used with correlated local ensembles in view of the initialisation step for newly-created particles. Note that Gallez reports in his Ph.D. thesis [96] various ALPM simulation results for FENE bead-spring chains with up to 8 springs. This demonstrates the feasibility of stochastic micro-macro methods for molecular models with configuration spaces of dimension $N_C = O(10)$.

Wapperom et al [97] further developed LPM with their backward-tracking Lagrangian particle method (BLPM). Instead of dropping Lagrangian particles into the flow and following them through the flow domain, the particle positions at which the viscoelastic stress is evaluated are fixed and specified a priori. The stress is computed by tracking the particles backwards in time over a single time step Δt, evaluating suitable initial values of configurations at that point, and then integrating the stochastic differential equation forwards along the obtained trajectories. Let $\boldsymbol{r}_B$ denote an arbitrary fixed position in the mesh. In general, the particle trajectory leading to that position changes from one time step to the next, so that different Lagrangian particles arrive at $\boldsymbol{r}_B$ as time evolves. The position $\boldsymbol{r}_I$ at t_{n-1} of the Lagrangian particle which is located at $\boldsymbol{r}_B$ at

current time t_n is obtained by integrating (19) backwards in time,

$$\boldsymbol{r}_I = \boldsymbol{r}_B - \int_{t_{n-1}}^{t_n} \boldsymbol{v}\, dt. \tag{26}$$

Then, starting from the previously computed values of the N_d molecular configurations $\boldsymbol{X}^j$ at $\boldsymbol{r}_I$ and time t_{n-1}, one obtains the updated configurations at $\boldsymbol{r}_B$ and t_n by integrating the stochastic differential equation (4) along the path joining $\boldsymbol{r}_I$ and $\boldsymbol{r}_B$. The initialisation process requires that an Eulerian field be computed from the Lagrangian data for each realisation $\boldsymbol{X}^j$ and at each time step. As with ALPM, correlated ensembles of molecules must be used for the initialisation process to be meaningful. Wapperom et al [97] have implemented BLPM for two-dimensional transient flows of non-linear dumbbell models. In each quadrilateral element of the mesh, the fixed positions $\boldsymbol{r}_B$ are defined at the nine nodal positions. For all problems which can be solved with correlated ensembles of model molecules, BLPM is the best available Lagrangian particle method, both in terms of numerical behaviour and cost. BLPM is also very efficient in continuum simulations with a constitutive equation of differential type (e.g. Wapperom and Keunings [70]). More generally, Lagrangian particle methods take account in a most natural way of the purely convective nature of the stochastic differential equation (4).

Mathematical issues and variance reduction

The stochastic formulation of viscoelastic flows raises several mathematical issues of fundamental interest. In particular, the non-linear coupling between the conservation laws (3) and the stochastic differential equation (4) has been investigated recently in a series of theoretical papers by Jourdain et al [98, 99], and Lelièvre [100]. The results are valid for the start-up of two-dimensional planar shear flow of Hookean and FENE dumbbells. For this simple but non-trivial flow, all convection terms of the form $\boldsymbol{v} \cdot \boldsymbol{\nabla}$ vanish in the governing equations. The problem for Hookean dumbbells is shown to be well posed in the sense of Cauchy, and convergence of the numerical solution to the exact solution is established for a particular finite element micro-macro scheme (Jourdain et al [98]). An optimal error estimate is derived by Lelièvre [100]. The analysis is more complicated for FENE dumbbells. In view of (6), the drift term in the stochastic differential equation (8) is non-linear and singular. This raises issues of existence and uniqueness of solution of the stochastic differential equation itself, even for a known velocity field; theoretical results are given by Jourdain and Lelièvre [101]. For the coupled problem defined by equations (2) to (4), Jourdain et al [99] present a proof of local-in-time existence and uniqueness of the exact solution.

The Eulerian evolution equation (22) for a Brownian configuration field is in fact a stochastic partial differential equation to which a rigorous meaning is technically difficult to assign, even though the random process $d\boldsymbol{W}$ is uniform in space (Le Bris and Lions [102]). A similar theoretical problem would arise if one were to write down the Eulerian formulation of the stochastic differential

equation (4) in the context of uncorrelated local ensembles of model molecules. In this case, $d\boldsymbol{W}$ would be a function of space labelled by the flow trajectory.

Another important topic is that of the statistical variance of the numerical solution. In the continuous (or exact) formulation of the coupled problem, the velocity and viscoelastic stress fields are deterministic, while only the molecular configurations are random. In the discrete problem, all variables are random and the numerical accuracy of a simulation is dictated by the variance of the discrete variables. In practice, a large variance in the numerical results would imply that independent micro-macro simulations yield vastly different solutions for the same flow problem. We have already discussed a significant beneficial effect of using correlated ensemble of model molecules: at each time step, the approximate velocity and viscoelastic stress fields are much smoother functions of $\boldsymbol{x}$ than those obtained with uncorrelated ensembles. But what is the effect on the variance? The early numerical experiments with correlated ensembles by Öttinger et al [83] showed that the variance of the approximate velocity field is drastically reduced; this led the authors to regard the method of Brownian configuration fields as a very powerful and general variance reduction technique. In fact, the latter statement is only partially correct. Indeed, Halin et al [84] and Bonvin and Picasso [103] have independently shown in their simulations with non-linear dumbbells that use of correlated ensembles reduces the variance of the velocity but increases the variance of the viscoelastic stress. This counter-intuitive experimental finding must be due to the non-linear coupling between the conservation laws and the stochastic differential equations.[6] It has been convincingly established and explained in the recent theoretical work by Jourdain et al [104] for the start-up of shear flow of Hookean dumbbells. These authors also address the question of finding the optimal spatial dependence of the Wiener processes that would yield minimum variance in the approximate stress and velocity. Moreover, using spatially-uniform Wiener processes, Jourdain et al [104] observe by way of numerical experiments that the ensemble average for the stress is a biased estimator[7]. The bias is of order $1/N_f$ (or equivalently $1/N_d$), and is also due to the non-linear coupling between velocity and molecular configurations.

A standard strategy for variance reduction is the use of control variates obtained from parallel process simulations. For an early application to polymer dynamics, see Melchior and Öttinger [105]. The basic idea is quite simple to state: a control variate should have about the same fluctuations as the random process of interest, but a vanishing average. Upon subtraction of the control variate from the variable of interest, the average is unchanged but the fluctuations are reduced. Bonvin and Picasso [103, 106] have implemented control variates within the framework of Brownian configuration fields. In addition to the viscoelastic stress $\boldsymbol{\tau}_p$, the authors consider the auxiliary stress $\bar{\boldsymbol{\tau}}_p$ such that

$$\bar{\boldsymbol{\tau}}_p = \langle \bar{\boldsymbol{g}}(\bar{\boldsymbol{X}}) \rangle, \tag{27}$$

[6] Indeed, for a given deterministic velocity field, the variance of the approximate stress is not affected by the spatial dependence of the Wiener processes.

[7] This also confirms an earlier finding by Halin et al [84].

where $\bar{\boldsymbol{X}}$ evolves according to

$$d\bar{\boldsymbol{X}} = \{-\bar{\boldsymbol{v}} \cdot \boldsymbol{\nabla}\bar{\boldsymbol{X}} + \bar{\boldsymbol{A}}\}\, dt + \bar{\boldsymbol{B}} \cdot d\boldsymbol{W}. \tag{28}$$

The stress equation (2) is written in the equivalent form

$$\boldsymbol{\tau}_p = \langle \boldsymbol{g}(\boldsymbol{X}) - \bar{\boldsymbol{g}}(\bar{\boldsymbol{X}})\rangle + \bar{\boldsymbol{\tau}}_p. \tag{29}$$

Note that the same Wiener process is used in the evolution equations for $\bar{\boldsymbol{X}}$ and $\boldsymbol{X}$. Thus, if $(\bar{\boldsymbol{g}}, \bar{\boldsymbol{v}}, \bar{\boldsymbol{A}}, \bar{\boldsymbol{B}})$ are selected sufficiently close to $(\boldsymbol{g}, \boldsymbol{v}, \boldsymbol{A}, \boldsymbol{B})$, one expects that the difference $\boldsymbol{g}(\boldsymbol{X}) - \bar{\boldsymbol{g}}(\bar{\boldsymbol{X}})$ has a much smaller variance than $\boldsymbol{g}(\boldsymbol{X})$. The average $\langle \boldsymbol{g}(\boldsymbol{X}) - \bar{\boldsymbol{g}}(\bar{\boldsymbol{X}})\rangle$ is computed by means of parallel stochastic simulations for $\boldsymbol{X}$ and $\bar{\boldsymbol{X}}$. In order to apply the decomposition (29), a constitutive equation must be available to compute the auxiliary stress $\bar{\boldsymbol{\tau}}_p$ in a deterministic way, in parallel with the velocity and actual viscoelastic stress. For example, Bonvin and Picasso [103, 106] select $\bar{\boldsymbol{v}} = \boldsymbol{v}$ and use the FENE-P dumbbell model to define control variates for FENE dumbbells. Their numerical results for the flow through a contraction indeed show a significant reduction of variance in the numerical results relative to the method of Brownian configuration fields. The CPU time to compute the viscoelastic stress is more than doubled, however.

Other developments

The micro-macro techniques described above decouple at each time step the solution of the stochastic equations (micro) and conservation laws (macro). Even though both the micro and macro problems can be discretized in time by means of an implicit technique, the decoupled micro-macro algorithm is at best semi-explicit. This may require the use of very small time steps to ensure stability. Laso et al [107] have recently proposed an approach to fully implicit micro-macro stochastic simulations based upon a size reduction technique. The authors show that the very large non-linear set of discrete equations to be solved at each time step can be reduced (by means of the Schur complement) to a system having the same size as a purely macroscopic formulation. The feasibility of implicit micro-macro techniques is illustrated in the start-up of one-dimensional shear flow of Hookean dumbbells.

Bell et al [108] combine Brownian dynamics and a spectral method (Chebyshev collocation) to study the one-dimensional transient problem of recovery after shear flow. Here, an ensemble of model molecules is attached to each collocation point. Results are obtained for the bead-spring FENE chain model of dilute polymer solutions (with up to 8 springs) and the Curtiss-Bird model of entangled systems. Finally, Tran-Canh and Tran-Cong [109] couple neural networks and stochastic simulation to study the start-up of planar Couette flow of non-linear dumbbells.

5. THE FOKKER-PLANCK APPROACH

We have seen in the previous section that control of the statistical noise is a major issue in stochastic micro-macro simulations. This problem does not arise at all in the Fokker-Planck approach, since the viscoelastic stress $\boldsymbol{\tau}_p$ and the primary unknown fields $(\boldsymbol{v}, p, \psi)$ governed by eqs. (1) to (3) are deterministic quantities. The difficulty, however, is that the Fokker-Planck equation (1) must be solved for ψ in both physical and configuration spaces. This necessitates a suitable discretization procedure for all relevant variables, namely position $\boldsymbol{x}$, configuration $\boldsymbol{X}$, and time t (in transient flow). The dimensionality of the problem can be daunting, and consideration of molecular models with many configurational degrees of freedom does not appear feasible. This probably explains why relatively few studies based on the Fokker-Planck approach have appeared in the literature, until very recently at least.

Fokker-Planck simulations of rheometrical flows

It is useful to briefly consider rheometrical flows, wherein the velocity field is specified and the distribution function only depends on configuration $\boldsymbol{X}$ and possibly time t. To date, most of the Fokker-Planck simulations of rheometrical flows have been for models of rod-like polymers (Doi and Edwards [7]), either in dilute solution or in the nematic liquid crystalline phase. Thus, the configuration space is only of dimension $N_C = 2$. Fokker-Planck simulations have also been performed with molecular models having a three-dimensional configuration space, but we are not aware of results published for $N_C > 3$. In contrast, stochastic simulations of rheometrical flows have been reported for molecular models with $N_C = O(10^2)$ (e.g. Somasi et al [15]).

Typically, a numerical approximation is sought for the distribution function ψ by means of the Galerkin spectral technique. This is a standard method of weighted residuals wherein the trial functions are identical to the basis functions. Proper selection of the basis functions is crucial for the accuracy of the numerical approximation. In 1972, spherical harmonics were used by Stewart and Sørensen [110] in their pioneering study of steady shear flow of a dilute suspension of rigid dumbbells. Fifteen years later, Strand et al [111] extended these results to start-up of shear flow. Since spherical harmonics are the eigenfunctions of the Laplace operator on the unit sphere, this particular choice of basis functions is expected to be optimal in the diffusion-dominated limit of small deformation rates. A similar approach was used in the analysis of the Doi model for rod-like polymers in the nematic phase. Here, the Fokker-Planck equation is non-linear in ψ in view of the mean-field interaction potential between rods. Detailed Fokker-Planck simulations of the Doi model in shear flow have uncovered the very rich non-linear dynamical behaviour of nematic rod-like polymers (Marrucci and Maffettone [112], Larson [113], Larson and Öttinger [114], Faraoni et al [115], Grosso et al [116], Suen et al [117]).

To our knowledge, the first Fokker-Planck simulations in a three-dimensional configuration space are due to Warner [71]. The author extended the technique developed by Stewart and Sørensen [110] for rigid polymers to study steady-

state and oscillatory shear flows of FENE dumbbells. Unfortunately, there are problems with this work, one of which being the singularity of the computed distribution for a vanishing dumbbell length. It was only 13 years later that Fan [118] improved Warner's technique, using spherical harmonics for the orientational dependence of ψ, and Jacobi polynomials for the dependence on dumbbell length. Fan [119] applied the same technique to study the steady-state shear response of the Bird and DeAguiar encapsulated FENE dumbbell model of entangled polymers.

When the flow strength increases, the diffusion term no longer dominates in the Fokker-Planck equation, and the distribution function usually becomes highly localized as the flow tends to orient the model molecules in preferred directions. Thus, the number of basis functions required to obtain accurate results grows drastically and spherical harmonics are no longer the most appropriate basis functions. For models with a configuration-dependent diffusivity, namely $\boldsymbol{D} = \boldsymbol{D}(\boldsymbol{X})$, spherical harmonics are not eigenfunctions of any operator present in the Fokker-Planck equation. Moreover, conventional spectral basis functions like spherical harmonics have global support, which leads to discrete systems with dense matrices. These observations led Armstrong et al [120] and Suen et al [117] to select Daubechies wavelets as alternative basis functions, in view of their localization properties and compact support.

Fokker-Planck simulations of complex flows

The first implementation of the Fokker-Planck approach for a complex flow appeared in 1989 and is due to Fan [55]. The author used a boundary element method for the conservation laws and a Galerkin spectral scheme with spherical harmonics for the Fokker-Planck equation. Fan reported results for planar channel flow of the dilute multibead-rod model ($N_C = 2$), assuming that the convection term $\boldsymbol{v}\cdot\boldsymbol{\nabla}\psi$ vanishes identically in the Fokker-Planck equation. Consideration of the convection term is a major challenge, first addressed in 1998 by Nayak [121] for two-dimensional flows of rod-like polymers either in the dilute or nematic phase. She discretized the convection operator by means of the discontinuous Galerkin finite element method, and used Daubechies wavelets for the discretization in configuration space (see also Suen et al [46]). Grosso et al [45] computed the response of the Doi model of rod-like polymers in two-dimensional flow between eccentric cylinders. They combined a Galerkin spectral solution in configuration space and a Lagrangian streamline integration in physical space (as in LPM). In their work, however, the coupling between viscoelastic stress and kinematics is ignored, so that the velocity field is the one computed for a Newtonian fluid. The state of the art in Fokker-Planck simulations has been updated drastically in a series of reports bearing 2003 as publication or submission year (Lozinski et al [122], Chauvière and Lozinski [123, 124], Lozinski and Chauvière [125], Suen et al [126]). Although many important features of these recent techniques largely depend on the particular molecular model selected for the simulations, we attempt in the remainder of this section to identify and describe the common general ideas.

As in the stochastic approach, solution of the conservation and Fokker-Planck equations are decoupled at each time step. For solving the conservation laws, Suen et al [126] use the DEVSS-G finite element technique, while a Galerkin spectral element technique is implemented by Lozinski et al [122], Chauvière and Lozinski [123, 124], and Lozinski and Chauvière [125]. The new velocity field is inserted in the Fokker-Planck equation, which is solved in configuration and physical spaces to update the distribution function ψ. To this end, use of a fully implicit method would be prohibitively expensive. It is imperative to somehow decouple the computation in physical space Ω from that in configuration space. Lozinski et al [122] achieve this by splitting each time step into two consecutive sub-steps,

$$\frac{\tilde{\psi} - \psi_n}{\Delta t} = -\frac{\partial}{\partial \boldsymbol{X}} \cdot \{\boldsymbol{A}\, \psi_n\} + \frac{1}{2} \frac{\partial}{\partial \boldsymbol{X}} \frac{\partial}{\partial \boldsymbol{X}} : \{\boldsymbol{D}\, \psi_n\}, \tag{30}$$

$$\frac{\psi_{n+1} - \tilde{\psi}}{\Delta t} + \boldsymbol{v}_n \cdot \boldsymbol{\nabla} \psi_{n+1} = 0. \tag{31}$$

Equation (30) for the intermediate value $\tilde{\psi}$ is defined at each collocation point of the spectral element mesh covering the flow domain Ω. It is solved by means of a Galerkin spectral method, as discussed in the previous section. The first sub-step is thus an ensemble of explicit, local updates in configuration space. The second sub-step (31) is an implicit, global update of the distribution function in physical space. Lozinski et al [122] solve (31) by means of a streamline-upwind-Petrov-Galerkin spectral element method, namely an element-by-element technique designed for convection problems. Note that the authors actually split the first sub-step into $O(10)$ smaller explicit steps in order to ensure numerical stability. In further developments of the method, Chauvière and Lozinski [123, 124], and Lozinski and Chauvière [125] adopt an implicit scheme, wherein ψ_n is replaced by $\tilde{\psi}$ in the right-hand-side of (30). Thus, the method can be used with a much larger time step Δt. The above ideas have been applied by Lozinski et al [122] to two-dimensional flows of the Öttinger tube model [21], for which $N_C = 3$. Implementations for FENE dumbbells are reported by Chauvière and Lozinski [123, 124], and Lozinski and Chauvière [125]. These authors considered both the (actual) three-dimensional configuration space of FENE dumbbells [124] and the (artificial) two-dimensional case wherein planar dumbbell orientation is assumed [123, 125]. They report an increase in simulation cost of one order of magnitude when going from $N_C = 2$ to $N_C = 3$. This clearly emphasizes the dimensionality problem of the Fokker-Planck approach alluded to previously.

Building on the work of Nayak [121], Suen et al [126] have recently proposed a mixed finite-element/wavelet-Galerkin method for computing two-dimensional flows with the double reptation tube model ($N_C = 3$) of Bird et al [8]. The authors solve the Fokker-Planck equation in physical space by means of the discontinuous Galerkin technique. A semi-implicit time-stepping scheme is used,

wherein orientation diffusion is treated implicitly and all other operators in the Fokker-Planck equation are treated explicitly. Together with a node-by-node decomposition strategy, this allows for an efficient solution within each finite element covering the flow domain in physical space.

Finally, two recent papers consider non-homogeneous flows wherein the centre of mass of the model molecules is not assumed to be simply convected by the flow, but rather is allowed to migrate across trajectories. Thus, the Fokker-Planck equation contains an additional spatial diffusion term. Lozinski et al [127] study the start-up of Poiseuille flow of FENE dumbbells with two-dimensional orientation. The results are in agreement with the early stochastic simulations of the same problem by Biller and Petruccione [54]. Suen et al [128] simulate pressure-driven channel flow of a modified Doi model for liquid crystalline polymers with spatially dependent concentration.

Fokker-Planck versus stochastic simulations

Only few comparisons have been reported to date between the stochastic and Fokker-Planck approaches, in the simulation of the same flow problem and with identical discretization in physical space (Lozinski and Chauvière [125], Chauvière and Lozinski [124], Suen et al [126]). Since these simulations are for molecular models with a configuration space of dimension $N_C \leq 3$, the comparison is bound to be somewhat biased in favour of the deterministic Fokker-Planck methods. The authors have implemented the method of Brownian configuration fields by means of the same discretization technique in physical space as that used in their Fokker-Planck simulations. The results are for the benchmark problem of planar flow past a cylinder confined in a channel. As could be expected, one concludes from these comparisons that the Fokker-Planck approach produces much more accurate results for a given overall cost, namely CPU time and memory requirements. Chauvière and Lozinski [124] also report, however, that the stochastic approach is significantly more stable as far as the maximum attainable Weissenberg number is concerned. Future work will reveal whether the Fokker-Planck approach is feasible at all for molecular models living in dimensions of order 10 or more.

Finally, hybrid techniques based on stochastic simulation and approximate representations of the distribution function have been proposed recently by Jendrejack et al [129] and Ellero and Kröger [130]. Although promising, these schemes remain to be tested in the simulation of complex flows.

6. CONCLUSIONS

The multiscale modelling of viscoelastic flows is an exciting recent development in computational rheology, which nicely complements the efforts made by the community on more conventional continuum simulations. Micro-macro methods allow the analysis of complex flow using a kinetic theory model that does not have an equivalent closed-form constitutive equation. The difficult theoretical problem of obtaining accurate closure approximations is thus bypassed

altogether, and the computational rheologist gains access to a potentially much wider modelling space. Micro-macro techniques are of course more demanding in computer resources than continuum methods, but they are generally easier to implement efficiently on parallel computers. To date, available stochastic and Fokker-Planck micro-macro techniques have been implemented for models of kinetic theory having but few configurational degrees of freedom. This particular context currently favours the deterministic Fokker-Planck methods. For more complex molecular models, however, extension of the stochastic approach seems to be the only feasible route. Creative ideas will perhaps disprove this statement.

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