Mechanistic constitutive model for wormlike micelle solutions with flow-induced structure formation

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Abstract

We present a tensor constitutive model for predicting stress and flow-induced structure formation in dilute wormlike micellar solutions. The micellar solution is treated as a dilute suspension of rigid Brownian rods whose length varies dynamically. Consistent with the mechanism presented by Turner and Cates [J. Phys.: Condens. Matter 4, 3719 (1992)], flow-induced alignment of the rods is assumed to promote increase of rod length that corresponds to the formation of flow-induced structures observed in experiments. At very high deformation rate, hydrodynamic stresses causes the rod length to decrease. These mechanisms are implemented in a phenomenological equation governing the evolution of rod length, with the number density of rods appropriately modified to ensure conservation of surfactant mass. The model leads first to an increase in both shear and extensional viscosity as deformation rate increases and then to a decrease at higher rates. If the rate constant for flow-induced rod growth is sufficiently large, the model predicts a multivalued relation between stress and deformation rate in both shear and uniaxial extension. Predictions for shear and extensional flow at steady state are in reasonable agreement with experimental results. By design, the model is simple enough to serve as a tractable constitutive relation for computational fluid dynamics studies.

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I. INTRODUCTION

Surfactant solutions are used in a wide range applications from fracking fluids in the oil and gas industry to heat-transfer fluids in the chemical process industries to various personal care and cleaning products [1]. Specifically, it has long been known that dilute surfactant solutions can behave very differently in turbulent flow than do simple fluids, leading to dramatic reductions in energy consumption [2]. Particularly in Japan, use of surfactants as drag reducing additives in water is becoming increasingly widespread in closed-loop district heating and cooling systems for large buildings and other facilities, resulting in substantial energy savings [3, 4]. Surfactants that form wormlike micelles are found to have drag reduction characteristics very similar to those of dilute polymer solutions [5–8].

At sufficiently high concentration, surfactants self-assemble into micelles that can take on a variety of forms including cylindrical, or wormlike. Wormlike micellar surfactant solutions (WMS) display extremely complex behavior when subjected to flow, and in particular can display dramatic increases in viscosity that arise due to a flow-induced transition from an isotropic liquid to a nematic-like gel phase [9–12]. In many fluids this transition is transient [12] while in others it can be irreversible [13]. The basic phenomenology underlying this transition is reasonably well-understood (see [14] for a comprehensive review). However, this understanding has not yet been translated into a mathematical model (constitutive equation) that can be used to predict the stresses and rheological state of surfactant solutions in complex flow systems characteristic of many engineering applications such as turbulent drag reduction. Here we present a simple mechanistic model for dilute wormlike micellar solutions that can serve as a tractable constitutive relation in computational fluid dynamics simulations. We emphasize from the outset that, in the interest of generating a tractable constitutive model, our approach dramatically oversimplifies many aspects of the flow-induced structure formation problem – we are proposing an engineering model, not an ab initio one.

Over the past three decades, a large body of experimental studies involving surfactants of different chemical compositions have unambiguously demonstrated the existence of dramatic shear-thickening in dilute WMS at shear rates higher than a critical value [9, 11, 12, 15–18]. Analogous observations have been made in extensional flow [19], but with a critical extension rate that is smaller than the critical rate in shear. Imaging studies by Berret and coworkers [12, 20] and Pine and coworkers [10, 11, 16, 21] show that shear-thickening is accompanied
by formation of a highly elastic and birefringent gel-like structure – the so-called flow-
induced structure (FIS). For many cationic systems, substantial evidence exists that at the
high deformation rates characteristic of turbulent flow, transient FIS form, leading to drag
reduction levels that are substantially higher than found in dilute polymer systems [22].
Despite its increasingly frequent application, this phenomenon remains poorly understood
at a fundamental level.

PIV measurements in circular Couette flow [11] (in which the inner cylinder is rotating)
indicate that the FIS grows from the inner wall of the Couette cell, eventually filling the
gap and resulting in plug flow in the core with lubricating layers near the wall. Smaller gaps
shift the critical shear rate to higher values – Pine and coworkers [11] interpret this as a
consequence of the slipping of the FIS at the walls [14]. However, the growth of FIS can also
be viewed in terms of development of vorticity bands, which we further describe below [23].
The Weissenberg numbers based on the relaxation time of the FIS phase can be extremely
large ($10^1 - 10^3$) at the critical shear rate, suggesting that elasticity-driven flow instabilities
will be a distinct possibility [14]. At even higher shear rates, shear-thinning is ultimately
observed.

Experiments by the Pine group [11, 21] in both shear rate controlled and shear stress
controlled settings have demonstrated the existence of a multivalued or reentrant flow curve
(plot of shear stress vs. shear rate). Note that this behavior is quite distinct from the flow
curve multiplicity found in models of entangled WMS and polymer melts. For dilute solu-
tions, multiple stresses can be found for the same shear rate, while for entangled solutions,
multiple shear rates can display the same shear stress. The latter multiplicity underpins
the shear-banding phenomenon [24, 25], while the former, which is the topic of the present
work, can in principle lead to so-called vorticity bands [23]. In a shear flow experiment the
shear stress is constant in the gradient direction while the nominal shear rate is constant
along with vorticity direction. Thus constitutive behavior that exhibits multiple stable shear
stresses for the same shear rate can display bands with different behavior along the vorticity
axis, i.e. vorticity bands.

In dilute WMS, the multivalued stress is observed only in a stress-controlled experiment;
the corresponding behavior in a shear rate controlled experiment manifests as a discontinuous
jump in shear stress at a particular shear rate. Reentrant behavior was also reported by
Dehmoune et al [18], whereas a discontinuous jump in shear rate controlled experiments was
reported in studies by several groups [5, 6, 26]. Note that shear-thickening without reentrant behavior has also been observed in several studies [12, 17, 27, 28].

Based on experimental observations, the rheological behavior of dilute WMS has been classified into three regimes – (i) at low shear rates, the solution remains essentially Newtonian, (ii) at intermediate shear rates shear-thickening occurs, accompanied by the formation of FIS, and (iii) at very high shear rates shear thinning is observed, along with breakdown of FIS [14]. Some studies [21, 26] indicate the existence of four regimes, where the second regime in the former classification is divided into two regimes – the first corresponding to inhomogeneous nucleation of FIS, and the second to homogeneous nucleation of FIS.

In most experiments with dilute WMC solutions, the FIS is transient and the fluid will eventually revert to an isotropic low viscosity state with negligible birefringence [14]. Recently, Shen and coworkers [13] performed an experiment where dilute WMS was driven through a microfluidic device containing a packed bed of glass beads to create a microfluidic packed bed (porous medium). In such a geometry, the flow has a very large extensional component whereas most prior experiments have been performed in shear rheometers. Under these conditions, they found that the solution formed an irreversible rather than a reversible gel. In this paper, we will be concerned solely with reversible FIS.

Only a small number of studies have attempted to put forth physically motivated yet mathematically tractable constitutive models for surfactant solutions. In the context of thixotropic fluids, Bautista, Manero, Puig, and coworkers [29] introduced a model (now known as the BMP model) whose variants have been used in predictions of shear-thickening in dilute WMS [30, 31] as well as shear-banding in entangled WMS [30, 32, 33]. The generalized BMP model, which can capture shear-thickening as well as reentrant behavior [31], couples a standard upper convected Maxwell equation for the micellar contribution to the stress with an evolution equation for the fluidity (inverse viscosity) of the solution. The viscosity and modulus that appear in the Maxwell equation are functions of fluidity. The evolution equation for fluidity consists of a term that captures spontaneous breakdown of micellar structure, and a second term that accounts for the buildup of structure due to work done on the fluid. Moreover, the rate constants for structure buildup and breakdown are made to depend on the stress. However, the use of the Maxwell equation in the BMP model is problematic in extensional flow, as the steady state extensional viscosity will diverge at a finite extension rate.
A phenomenological model that qualitatively captures shear-thickening and reentrant behavior was proposed by Goveas and Pine [34]. Without appealing to any microstructural mechanism, their model considers an insoluble gel phase to form when the local stress in the solution exceeds a critical value. Beyond this critical stress, the evolution of the gel phase is governed by the rate of gel formation that depends on the local stress, and a constant rate of gel destruction.

Shear-thickening due to onset of instability above a critical shear rate was investigated by Barentin and Liu [35]. According to their model, this instability arises from electrostatic attractions between individual micelles, which fosters the growth of micellar bundles that eventually form networks, giving rise to shear-thickening.

An important class of models is based on population dynamics, where the evolution of sub-populations of micelles of different lengths are tracked, and the material properties are calculated based on the interactions within each sub-population as well as between different sub-populations. A particularly successful model in this class was developed by Vasquez, McKinley, and Cook [36], albeit for explaining the rheology of entangled WMS. While their model considered only two sub-populations, very good qualitative agreement with some rheological data for entangled micellar solutions have been obtained, while other experimental observations, such as second-normal stresses and complex transients are still not accessible [37]. The model does display shear-banding [38, 39] as well as the rupture phenomenon experimentally observed in uniaxial extensional flow [40]. Models based on three sub-populations – long micelles, short micelles, and a gel phase have also been proposed [41]. Depending on appropriate parameter values, this model predicts shear-thickening as well as shear-banding and spatial inhomogeneity. However, the presence of a large number of empirical constants in the model poses significant difficulty for experimental validation.

A relatively simple theoretical model of a dilute solution of rodlike micelles that qualitatively captures the key experimental observation of a flow-induced gelation transition has been proposed by Cates and Turner (CT) [42, 43]. In their model, rods (i) align with the flow and diffuse rotationally, (ii) can react end-to-end to form longer rods, and (iii) can spontaneously break up into shorter rods. The reaction to form longer rods only happens when rods are collinear. The model takes the form of a master equation for the evolution of the probability distribution function for rod orientation and length; in the absence of reactions, each rod would evolve the same way that a rigid rod evolves in dilute solution and
the model reduces to the standard Fokker-Planck equation for that case. In elongational flow, which strongly aligns the rods, CT find that the reaction to form long rods dominates so that eventually the rod length diverges leading to a gelation transition. In shear, a sharp transition is absent but the mean micellar length still becomes much larger than in the absence of flow. This is extremely important work, as it clearly indicates the dominant physics of structure formation in these systems. Nevertheless, it leaves unanswered many questions that are important if we are to be able to quantitatively reproduce experimental observations or make predictions for flow systems in practical applications. In particular, CT do not write an evolution equation for the stress tensor – a constitutive equation. While micelles have been modeled as rigid rods in several past theoretical approaches, notably by Cates and Turner [42, 43], we are not aware of any model that provides a constitutive relation predicting shear-thickening with a reentrant phase diagram.

To perform direct numerical simulations for dilute surfactant solutions, a constitutive equation to relate the local stress with the local deformation gradient is required. To our knowledge, no closed form continuum-level constitutive model for FIS-exhibiting dilute surfactant solutions has been developed that uniformly treats both shear and extensional flow. It is important to note that the inherent complexity of the numerical algorithms required for solving the coupled nonlinear partial differential equations resulting from discretization of the Navier-Stokes equations in a general domain restricts the complexity of the constitutive relation. This is so because only the simplest models are tractable with the current level of computational capability, within reasonable bounds of time and resources. For example, in case of turbulent flow in dilute polymer solutions, the most widely used constitutive model is FENE-P [44], although numerous models exist that are much more detailed. Indeed, studies have shown that the FENE-P model works remarkably well in capturing key experimental observations in viscoelastic turbulence [45]. In this paper, we present a constitutive model for dilute WMS that is of similar complexity as FENE-P for dilute polymers and evaluate its predictions in simple shear and uniaxial extensional flows.

II. MODEL DESCRIPTION

We begin by providing a qualitative description of our model, which we call the reactive rod model (RRM). A schematic of the mechanisms involved is shown in Fig. 1. We consider
a dilute solution of wormlike micelles as a suspension of rigid Brownian rods. In the absence of external flow, the orientation of the rods remains isotropic. Since the solution is dilute, the rods do not have steric or hydrodynamic interactions with each other.

Physically, a micellar solution is polydisperse with the length following an exponential distribution [1], but in order to make analytical progress we will assume that a single length is sufficient to characterize the solution at all times. Such a description has been used in prior studies of coagulation of spherical particles to describe particle size [46]. We emphasize from the outset that our approach dramatically oversimplifies many aspects of the flow-induced structure formation problem in the interest of generating a constitutive model that is tractable for computational fluid dynamics applications.
When an external flow field is imposed, individual rods preferentially align in the flow direction – the degree of alignment being governed by a balance between rotational diffusion and convection. We further assume that the rods can react, such that the reaction rate increases with alignment to generate longer rods. This mechanism is based on experimental observations that micelles are found to grow in length in presence of flow. As rotational diffusion of the longer rods is slower compared to shorter rods, they align more strongly in flow and keep growing. Eventually this process generates a collection of very long rods – a flow-induced structure that exhibits shear-thickening. However, when the rods grow sufficiently long, they can no longer withstand hydrodynamic stresses; they break down, leading to shear-thinning. Note that we do not specify the microscopic mechanisms of rod coalescence or dissociation – they are simply accounted for by introducing phenomenological expressions that capture key experimental observations.

A. Theory of rigid rods

Before presenting the RRM in detail, we review some basic results for the rheology of dilute suspension of rigid Brownian rods. Consider a dilute suspension of rigid rods with number density $n_0$. Let each rod be of length $L_0$ and radius $b$. The orientation of a rod is described by a unit vector $u$. The solvent is assumed to be Newtonian with viscosity $\eta_s$. The suspension is subject to a homogeneous flow $\mathbf{v}$ with transpose velocity gradient $K = \nabla \mathbf{v}^\top$. (In Cartesian tensor notation $K_{ij} = \partial v_i / \partial x_j$.)

In homogeneous flow, the stress in a suspension depends solely on rotational motion. The rotational diffusion coefficient of a rod is [47]

$$D_{r,0} = \frac{3k_B T}{\pi \eta_s L_0^3} \ln \left( \frac{L_0}{2b} \right), \quad (1)$$

where $k_B$ is the Boltzmann constant and $T$ is the temperature. The average orientation is represented by the orientation tensor $S$, defined as the second moment of $u$, i.e. $S = \langle uu \rangle$, where angle brackets represent ensemble average. Starting from a Smoluchowski equation for rotational motion, the time evolution of the orientation tensor $S$ can be written as [47]

$$\frac{dS}{dt} = -6D_{r,0} \left( S - \frac{1}{3} I \right) + K \cdot S^\top + S \cdot K^\top - 2K : \langle uu uu \rangle. \quad (2)$$

The double dot product is defined as follows: $A : B = \text{Tr} \left( A \cdot B^\top \right)$. 


The stress $\tau$ in a dilute suspension of rods can be written as a sum of the stress due to the solvent and an extra stress tensor $\tau^p$ that accounts for the contribution due to the presence of the rods. Thus

$$\tau = 2\eta_s D + \tau^p, \quad (3)$$

where

$$\tau^p = 3n_0 k_B T \left( S - \frac{1}{3} I \right) + \frac{n_0 k_B T}{2D_{r,0}} K : \langle uuuu \rangle \quad (4)$$

and the rate of deformation tensor $D = \left( K + K^T \right) / 2$.

To proceed analytically, a closure approximation for the fourth moment $\langle uuuu \rangle$ is necessary. We use an expression due to Dhont and Briels [48] that reproduces physically reasonable rheological behavior over a very large range of shear and extension rates. The approximation is as follows:

$$K : \langle uuuu \rangle \approx \frac{1}{5} \left[ S \cdot D + D \cdot S - S \cdot D \cdot S + 2S \cdot D \cdot S + 3 (S : D) S \right]. \quad (5)$$

Using Eq. (2) – Eq. (5), the stress of the suspension can be calculated for an arbitrary flow. The results for steady state cases are obtained by setting the time derivatives to zero. It is known that such a suspension of rods exhibits shear-thinning and extension-thickening [44].

**B. Reactive rod model (RRM)**

Now we consider the case where the rods can change length in response to flow. In addition to the notations introduced earlier, let $n$ and $L$ represent the number density and rod length at any time $t$ after the initiation of flow. In contrast to $n$ and $L$, we take the radius of the rods $b$ to remain constant at all times. In accordance with the picture presented by Turner and Cates [43], we assume that growth of rods increases with alignment. From conservation of surfactant mass (total micelle length) we have $n = n_0 L_0 / L$ at all times. Since $D_{r,0}$ is the diffusion coefficient based on rod length $L_0$ and $D_r$ is that based on rod length $L$, from Eq. (1) we have

$$\frac{D_r}{D_{r,0}} = \frac{1}{L^3} \left( \frac{\ln L^* + m}{m} \right), \quad (6)$$

where $L^* = L / L_0$ is the rod length normalized with the initial length and $m = \ln \left[ L_0 / (2b) \right]$ is a constant that serves as a measure of the initial aspect ratio of the rods.
Accounting for the variation of $n$, $L$ and $D_r$, Eq. (2) for the time evolution of the orientation tensor becomes
\[
\frac{dS}{dt} = -6D_r \left( S - \frac{1}{3}I \right) + K \cdot S^\top + S \cdot K^\top - 2K : \langle uuuu \rangle. \tag{7}
\]
and Eq. (4) for the stress becomes
\[
\tau^p = 3nk_BT \left( S - \frac{1}{3}I \right) + \frac{nk_BT}{2D_r} K : \langle uuuu \rangle. \tag{8}
\]

Further, we define a nondimensional time $t^* = D_{r,0} t$ and a Péclet number $Pe = \dot{\gamma}/D_{r,0}$ in shear flow and $Pe = \dot{\varepsilon}/D_{r,0}$ in extensional flow. In addition, we introduce a scalar orientational order parameter
\[
\hat{S} = \sqrt{\frac{3}{2} (\hat{S} : \hat{S})}, \tag{9}
\]
where $\hat{S} = S - \frac{1}{3}I$ is the traceless part of $S$. When the rods are completely aligned, $\hat{S} = 1$ whereas for isotropic conditions (e.g. at equilibrium) $\hat{S} = 0$.

Now we address the evolution of the rod length. In general we will write
\[
\frac{dL^*}{dt^*} = R_a + R_s, \tag{10}
\]
where $R_a$ represents the rate of alignment-induced growth and $R_s$ represents the rate of spontaneous growth and breakdown of micelles. We will take the alignment-induced growth term to increase linearly with the degree of alignment of the rods, i.e.
\[
R_a = k\hat{S}, \tag{11}
\]
where $k$ is a constant. The spontaneous growth and breakage rate $R_s$ is taken to be proportional to the deviation of the current length from its equilibrium value, with a constant of proportionality $\lambda$. However, if the rods become long enough, hydrodynamic stresses will break them apart. Thus at a given deformation rate, there must be a maximum rod length that can be sustained without rupture. We capture this idea by introducing the maximum rod length $L^*_{\text{max}}$. As $L^*$ approaches $L^*_{\text{max}}$ the breakage rate increases without bound, thus forcing $L^*$ to decrease at high deformation rates. Since the hydrodynamic stresses increase with deformation rate, the maximum rod length must decrease with $Pe$. We choose a very simple functional form of this decay as:
\[
L^*_{\text{max}} = \alpha + \frac{\beta}{Pe}, \tag{12}
\]
where $\alpha$ and $\beta$ are model parameters. Thus we take

$$R_s = \frac{\lambda}{1 - \left(\frac{L^*}{\alpha + \frac{\beta}{P_e}}\right)^2} (1 - L^*)$$  \hspace{1cm} (13)$$

Substituting Eq. (11) and Eq. (13) into Eq. (10) we have

$$\frac{dL^*}{dt^*} = \frac{\lambda}{1 - \left(\frac{L^*}{\alpha + \frac{\beta}{P_e}}\right)^2} (1 - L^*) + k\hat{S}.$$  \hspace{1cm} (14)$$

Eq. (14) combined with Eq. (9), Eq. (1), and Eq. (7) results in a system of ODEs that governs the time evolution of $S$ and $L^*$. The corresponding stress can be obtained by further substitution into Eq. (8). We provide the specific equations for shear and uniaxial extension below.

C. Shear flow

For simple shear flow, $\mathbf{v} = (\dot{\gamma}y, 0, 0)^\top$. Recall that the rotational Péclet number in shear flow is defined as $Pe = \dot{\gamma}/D_{r,0}$. The scalar orientation parameter in this case is

$$\hat{S} = \left[\frac{3}{2} \left( \left(S_{xx} - \frac{1}{3}\right)^2 + \left(S_{yy} - \frac{1}{3}\right)^2 + \left(S_{zz} - \frac{1}{3}\right)^2 + 2S_{xy}^2 \right) \right]^{1/2},$$  \hspace{1cm} (15)$$

where $S_{xx} + S_{yy} + S_{zz} = 1$. Substituting the closure relation Eq. (5) into Eq. (7) and after rearrangement we have

$$\frac{\partial S_{xx}}{\partial t^*} = -\frac{6}{L^*^3} \left(\ln L^* + m\right) \left(S_{xx} - \frac{1}{3}\right) + 2PeS_{xy} - \frac{2}{5}PeS_{xy}(1 + 4S_{xx} - S_{yy})$$

$$\frac{\partial S_{yy}}{\partial t^*} = -\frac{6}{L^*^3} \left(\ln L^* + m\right) \left(S_{yy} - \frac{1}{3}\right) - \frac{2}{5}PeS_{xy}(1 + 4S_{yy} - S_{xx})$$

$$\frac{\partial S_{zz}}{\partial t^*} = -\frac{6}{L^*^3} \left(\ln L^* + m\right) \left(S_{zz} - \frac{1}{3}\right) - \frac{6}{5}PeS_{yy}S_{zz}$$

$$\frac{\partial S_{xy}}{\partial t^*} = -\frac{6}{L^*^3} \left(\ln L^* + m\right) S_{xy} + PeS_{yy} - \frac{Pe}{5} \left[6S_{xy}^2 + S_{xx} + S_{yy} - (S_{xx} - S_{yy})^2 \right].$$  \hspace{1cm} (16)$$
Similar steps lead to the expressions for the stress tensor:

\[
\frac{\tau_{xx}}{3n_0k_B T} = \frac{1}{L^*} \left( S_{xx} - \frac{1}{3} \right) + \frac{m Pe L^*^2}{30 (\ln L^* + m)} S_{xy} (1 + 4S_{xx} - S_{yy})
\]

\[
\frac{\tau_{yy}}{3n_0k_B T} = \frac{1}{L^*} \left( S_{yy} - \frac{1}{3} \right) + \frac{m Pe L^*^2}{30 (\ln L^* + m)} S_{xy} (1 + 4S_{yy} - S_{xx})
\]

\[
\frac{\tau_{zz}}{3n_0k_B T} = \frac{1}{L^*} \left( S_{zz} - \frac{1}{3} \right) + \frac{m Pe L^*^2}{10 (\ln L^* + m)} S_{xy} S_{zz}
\]

\[
\frac{\tau_{xy}}{3n_0k_B T} = \frac{S_{xy}}{L^*} + \frac{m Pe L^*^2}{60 (\ln L^* + m)} \left[ 6S_{xy}^2 + S_{xx} + S_{yy} - (S_{xx} - S_{yy})^2 \right]
\]

D. Uniaxial extensional flow

For uniaxial extensional flow, the velocity field \( \mathbf{v} = (\dot{\varepsilon}^2, -\dot{\varepsilon}^2, \dot{\varepsilon})^T \) and the scalar orientation parameter

\[
\hat{S} = \left[ \frac{3}{2} \left\{ \left( S_{xx} - \frac{1}{3} \right)^2 + \left( S_{yy} - \frac{1}{3} \right)^2 + \left( S_{zz} - \frac{1}{3} \right)^2 \right\} \right]^{\frac{1}{2}},
\]

where symmetry and the unit trace condition dictates that \( S_{xx} = S_{yy} = \frac{1 - S_{zz}}{2} \). Recall that in extensional flow, the rotational Péclet number is defined as \( Pe = \dot{\varepsilon}/D_{r,0} \). Substituting the expression for the velocity field into the closure relation Eq. (5), and from Eq. (7) we have

\[
\frac{\partial S_{xx}}{\partial t^*} = -\frac{6}{L^*^{\frac{5}{2}}} \left( \frac{\ln L^* + m}{m} \right) \left( S_{xx} - \frac{1}{3} \right) - Pe S_{xx} - \frac{Pe S_{xx}}{5} (9S_{zz} - 5)
\]

\[
\frac{\partial S_{zz}}{\partial t^*} = -\frac{6}{L^*^{\frac{5}{2}}} \left( \frac{\ln L^* + m}{m} \right) \left( S_{zz} - \frac{1}{3} \right) + 2Pe S_{zz} - \frac{Pe}{5} (9S_{zz}^2 + S_{zz}).
\]

Similar manipulations lead to the component-wise expressions for the stress tensor:

\[
\frac{\tau_{xx}}{3n_0k_B T} = \frac{1}{L^*} \left( S_{xx} - \frac{1}{3} \right) + \frac{m Pe L^*^2}{60 (\ln L^* + m)} S_{xx} (9S_{zz} - 5)
\]

\[
\frac{\tau_{yy}}{3n_0k_B T} = \frac{1}{L^*} \left( S_{yy} - \frac{1}{3} \right) + \frac{m Pe L^*^2}{60 (\ln L^* + m)} (9S_{zz}^2 + S_{zz})
\]

where \( \tau_{xx} = \tau_{yy} = \tau_{rr} \) from symmetry, where \( r = \sqrt{x^2 + y^2} \) is the radial coordinate.

III. RESULTS AND DISCUSSION

We present results for both shear and extensional flow at steady state over a wide range of Pe (dimensionless shear or extension rate). Solutions are found using the multivariate
FIG. 2. (a) Shear stress vs. dimensionless shear rate for different values of growth rate $k$. (b) Normalized rod length in shear flow vs. dimensionless shear rate for different values of growth rate $k$, with model parameters $\alpha = 5$ and $\beta = 500$. The thick lines represent values accessible under shear rate control; the entire curves are accessible under shear stress control. The black dashed lines ($k = 0$) show the values in the absence of any rod growth: i.e. for a dilute suspension of rigid rods with constant length.

root finding algorithm hybr as implemented in the SciPy package [49]. For all the results shown in the following, we take $\lambda = 1$ and $m = 7$, the latter corresponding to an initial aspect ratio of 1000. At steady state, $\lambda$ can be combined with $k$ to form the ratio $k/\lambda$, which is equivalent to setting $\lambda = 1$ and varying $k$. Varying $m$ does not have a strong effect on the qualitative nature of the results presented. Before proceeding we note that the linear viscoelastic behavior of this model is identical to that for rigid rods of constant length $L_0$ (see e.g. [47]): coupling between the change of length and stress enters only at $O(\text{Pe}^2)$.

A. Shear flow

Fig. 2 (a) shows the shear stress as a function of $\text{Pe}$ for different values of $k$ and Fig. 2 (b) shows the corresponding results for rod length $L$. Note that if $k = 0$ there is no growth of rods (as shown by the black dashed line in Fig. 2 (b)), and RRM reduces to a suspension of monodisperse rigid rods. For low values of $k$ (see graph for $k = 2.5$), the shear stress increases monotonically with shear rate. The results are identical if shear stress is increased
FIG. 3. Micelle contribution to shear viscosity normalized by that at vanishing shear rate ($\eta_s$ is the solvent viscosity and $\eta_0$ is the zero-shear viscosity of the solution) vs. dimensionless shear rate for different values of growth rate $k$, corresponding to the data presented in Fig. 2. The thick lines represent values accessible under shear rate control; the entire curves are accessible under shear stress control. The black dashed line ($k = 0$) shows the well-known shear-thinning result for rigid rod suspensions without reaction.

and the shear rate obtained as an output. From Fig. 2 (b) we see the increase in rod length at $Pe \approx 1$: i.e. the formation of FIS. This leads to an increase in shear viscosity, i.e. shear-thickening behavior, as shown in Fig. 3. Note that at such low values of $k$, the viscosity is still continuous. At sufficiently high $Pe$, the rods break down, resulting in shear-thinning.

At higher values of the growth rate $k$, the curve of $\tau_{xy}^p$ vs. $Pe$ displays a region where it is multivalued: three values of shear stress are possible for the same shear rate. Thus in a shear rate controlled experiment, Fig. 2 (a) should show a jump discontinuity in stress, the height of the jump increasing with increase in $k$. At constant $Pe$ the intermediate branch of the $\tau_{xy}^p$ vs. $Pe$ is unstable. The discontinuity is more prominent in viscosity data, as can be seen in Fig. 3. Only in a stress-controlled experiment would the intermediate solution branch be observed. Note that the multivalued nature of the flow curves presented here is in contrast to that observed in shear banding of entangled WMS, where the shear rate becomes multivalued over a certain range of shear stress.

At higher values of $Pe$, the multiplicity disappears – we see from Fig. 3 that this cor-
FIG. 4. First (a) and second (b) normal stress differences in shear flow vs. dimensionless shear rate for different values of growth rate $k$, with model parameters $\alpha = 5$ and $\beta = 500$. The thick lines represent values accessible under shear rate control; the entire curves are accessible under shear stress control. The black dashed lines ($k = 0$) show the values in the absence of any rod growth.

responds to shear-thinning behavior. The multiplicity is also seen in the first and second normal stress differences (Fig. 4), though the second normal stress difference is numerically much smaller. The shear-thinning region is characterised by a decrease in rod length (Fig. 2 (b)) as $\text{Pe}$ increases.

The presence of multiple values of stress at steady state for a given shear rate obviously raises the question as to which specific value is attained under transient conditions. To investigate this issue, we calculated the transient shear stress for two cases – (i) The shear rate is slowly ramped up from zero through the multivalued region, and (ii) The shear rate is slowly ramped down from beyond the multivalued region back to zero. To access the entire flow curve would require a quasisteady ramp in shear stress rather than shear rate. For the ramp calculations we define $\text{Pe}(t) = 10^{-4}t$ on the upward ramp and $\text{Pe}(t) = 0.5 - 10^{-4}t$ on the downward, the ramps being deliberately low so as to emulate a quasi-static process. The results are shown in Fig. 5, along with the steady state data from Fig. 2 (a) corresponding to $k = 10$. We find that on ramping up, the transient stress follows the lower branch till the first turning point, when it suddenly jumps up to the upper branch. The reverse is seen in case of ramping down from beyond the multivalued region.

The region of multiplicity for both shear and extensional flow as a function of $k$ and $\text{Pe}$
FIG. 5. Transient shear stress vs. Pe on ramping up (down) the shear rate through the region exhibiting multiplicity in stress. Model parameters are $k = 10$, $\alpha = 5$ and $\beta = 500$. Note the jumps between the branches on ramp up and ramp down indicating which specific value of stress is attained under transient conditions.

is shown in Fig. 6. Note that this region is larger for extension than for shear; in particular it extends to lower $k$. Recalling that $k$ is the rate constant for alignment-induced growth, this result makes sense physically because extension increases $\hat{S}$ more rapidly with Pe than does shear.

The effect of the parameters $\alpha$ and $\beta$ are shown in Fig. 7. Recall that $\alpha$ and $\beta$ control the maximum rod length at a given $Pe$ (see Eq. (12)). As expected, we see that $\alpha$ determines the rod length at very high $Pe$, whereas $\beta$ determines the shear rate where hydrodynamic stresses kick in to break down the rods. Note that $\alpha$ and $\beta$ have minimal effect on the multivalued nature of the flow curves, their primary role is to enforce shear-thinning behavior at high $Pe$.

We compare the model predictions with experimental data for shear viscosity in Fig. 8. The datasets labeled $(L_1)$ and $(L_2)$ represent shear-rate controlled data from Liu and Pine [10] for an equimolar aqueous solution of cetyltrimethylammonium bromide (CTAB) and sodium salicylate (NaSal) at 500 ppm and 1000 ppm, respectively. Dataset $(D)$ shows stress-controlled data from Dehmoune et al [18] for octadecyltrimethylammonium bromide ($C_{18}TAB$)/NaSal at 3 mM concentration. For normalizing the viscosity we have used the
data reported at the lowest shear rate as the zero-shear viscosity; the solvent viscosity (taking \( \eta_s \approx 0.001 \) Pas for water) is negligible for all cases compared to the viscosity of the solution. All the three datasets were obtained from experiments performed in a Couette rheometer. The model parameters in Fig. 8 were generated not through any rigorous algorithm, but via simple trial-and-error and checked visually. Insofar as our present aim is to demonstrate that our model generates reasonable agreement with experimental observations, we consider this to be sufficient.

While our model is able to capture the shear-thickening and the subsequent shear-thinning behavior reasonably, the experimental data exhibits a small amount of initial shear-thinning that is absent from the model predictions. Note that a suspension of rigid rods is shear-thinning in the absence of any reaction, but this is observed at a higher Pe compared to the onset of the multivalued region (see Fig. 3 for \( k = 0 \)). The problem here is primarily that polydispersity of rod lengths is neglected – i.e. the distribution is a \( \delta \)-function, and so is the associated time scale. Such a distribution cannot be expected to capture features possibly arising due to a broad distribution, as in an experimental system. An analogous difficulty arises in using the FENE-P model for describing polymer solutions. We speculate that a certain amount of flow-induced alignment is required before the rods start reacting. Indeed, a small amount of initial shear-thinning can be observed if the growth equation Eq. (11)
is made to depend on a higher power of \( \dot{S} \), say the third power (a second power produces negligible amount of shear-thinning). However, since our expression for the growth rate is purely phenomenological and there is no specific reason for choosing a particular power of \( \dot{S} \), we have refrained altogether from introducing yet another parameter for the power law.

Another issue worth pointing out is that we have used the rotational diffusion coefficient \( D_{r,0} \) as a fitting parameter. We find the fitted values to be significantly more than their approximate physical values. As our model stands, the effect of changing \( D_{r,0} \) is to shift and stretch the viscosity curves along the \( \dot{\gamma} \)-axis. Nevertheless, we reiterate that our model predicts the correct qualitative dependence of the shear-viscosity on the shear rate.
FIG. 8. Comparison of steady shear viscosity predicted by our model with experimental measurements. $(L_1)$ and $(L_2)$ show data from Liu and Pine [10], and $(D)$ shows data from Dehmoune et al [18]. Solid lines show results from our model fitted to the experimental measurements. The values of the fitting parameters are $(L_1)$: $D_{r,0} = 20s^{-1}$, $k = 7.5$, $\alpha = 4$, $\beta = 10$; $(L_2)$: $D_{r,0} = 8s^{-1}$, $k = 3.5$, $\alpha = 2$, $\beta = 50$; $(D)$: $D_{r,0} = 11.8s^{-1}$, $k = 6$, $\alpha = 2$, $\beta = 6$.

B. Extensional flow

Fig. 9 (a) shows the extensional stress difference $\tau_{zz}^p - \tau_{rr}^p$ as a function of Pe for different values of the growth rate $k$ and Fig. 9 (b) the corresponding plot for $L$. Analogous to the situation in shear flow, there is flow-induced structure formation and multiplicity in the stress response at higher values of $k$. The second normal stress difference is identically zero in uniaxial extension. The dependence of extensional viscosity $\eta_E = (\tau_{zz} - \tau_{rr})/\dot{\varepsilon}$ is shown in Fig. 10. Compared to shear flow, the increase in rod length resulting in extension-thickening is much higher. Correspondingly, the region in parameter space exhibiting multiplicity is substantially larger for extension than for shear as illustrated in Fig. 6.

While we are not aware of experimental data reporting a multivalued extensional viscosity, viscosity data showing dramatic extension-thickening has been reported by Prud’homme and Warr [19] for equimolar solutions of tetramethyltri ammonium bromide (TTABr) and NaSal using an opposing jet rheometer. We compare our model predictions with their viscosity data in Fig. 11. The datasets labeled $(P_1)$ and $(P_2)$ show experimental results for a 25.2 mM
FIG. 9. (a) Extensional stress difference and (b) rod length vs. dimensionless strain rate at steady state in uniaxial extensional flow for different values of growth rate $k$, with model parameters $\alpha = 5$ and $\beta = 500$. The thick lines represent values accessible under extension rate control; the entire curves are accessible under stress control. The black dashed lines ($k = 0$) show the values in the absence of any rod growth.

solution for nozzle diameters (of the opposing jet rheometer) 2 mm/2 mm and 4 mm/4 mm, respectively; whereas dataset ($P_3$) is for a 69.2 mM solution for diameters 2 mm/2 mm. Even though there is significant scatter in the experimental data, the qualitative trend in terms of shear-thickening matches reasonably well with the model predictions. However, the issue with the large fitted values of $D_{r,0}$ alluded to for shear flow holds here as well. Note that these experiments did not report a discontinuous jump in viscosity, so the fits with the model are only for lower values of $k$.

IV. CONCLUSION

We presented a closed-form mechanistic model that can capture shear (extension)-thickening and subsequent shear (extension)-thinning in dilute wormlike micellar solutions. One of our goals in constructing the model was to ensure that it is expressed in tensorial form, allowing it to be applied for arbitrary flow fields, and that it is simple enough so that it can be incorporated as a constitutive relation in existing solvers for numerical simulation of complex flows, for example in investigating turbulent drag reduction of micellar solutions.
FIG. 10. Extensional viscosity vs. dimensionless strain rate at steady state in uniaxial extensional flow for different values of growth rate $k$, corresponding to the data presented in Fig. 9. The black dashed lines ($k = 0$) show the values in the absence of any rod growth.

Our model assumes micelles as a collection of rigid rods that can grow in length in response to flow-induced alignment. There is a maximum length, depending on shear (extension)-rate to which the rods may grow before they break down into smaller rods. This mechanism is intended to capture rupture of long micelles due to hydrodynamic stresses. We do not specify the microscopic details of the growth or rupture processes, nor is such a specification necessary for the intended level of detail. The model couples the equations for time evolution of the orientation tensor with a kinetic equation for rod length. The equation for rod length has a growth term that is proportional to orientation and a destruction term for capturing breakdown of rods. For small shear (extension) rates, our model predicts Newtonian behavior, whereas at relatively higher values shear (extension)-thickening is observed. Furthermore, at sufficiently high deformation rates, shear (extension)-thinning is observed. The shear (extension)-thickening predicted corresponds to flow-induced structure formation observed in experiments. If the growth rate is increased beyond a certain value, reentrant behavior (multiplicity of stress for a given deformation rate) is observed. Our model predictions agree reasonably well with experimental data for both stress-controlled and shear-rate controlled cases.

We believe that the work presented here takes an important step toward understanding
FIG. 11. Comparison of steady extensional viscosity with experimental data of Prud’homme and Warr [19]. The solid lines show model predictions fitted to the experimental data. The parameter values are

- \((P_1)\): \(D_r,0 = 5.4s^{-1}, k = 1.25, \alpha = 0.5, \beta = 10\);
- \((P_2)\): \(D_r,0 = 8.5s^{-1}, k = 3.2, \alpha = 1, \beta = 1\);
- \((P_3)\): \(D_r,0 = 14.5s^{-1}, k = 1.15, \alpha = 0.5, \beta = 10\).

how to construct models for an important class of self-assembling fluids, dilute wormlike micelle solutions. Such models will have a broad impact on our capability to make reliable predictions about the wide range of flow processes that involve such materials. Natural applications for the present model include inhomogeneous flows, including especially circular Couette flow, where many experiments have been performed, as well as turbulent drag reduction, one of the most important applications of these fluids. Finally, many improvements to the current model can be envisioned. For example, more refined and fundamentally-based models for the growth and breakage rates should be developed. Additionally, the model assumes diluteness of the solution, which requires that \(nL^3 \ll 1\). In terms of our model \(nL^3 = n_0L_0L^2\) which does not remain small when \(L\) is sufficiently large. Thus we overpredict the rotational diffusivity at high \(L\), where the solution effectively becomes semidilute. It would also be desirable to overcome the assumption of a delta-function length distribution without adding undue complexity.
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