



## NON-NEWTONIAN SOLVENT EFFECTS ON THE RHEOLOGICAL RESPONSES OF WORMLIKE MICELLAR SOLUTIONS

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**Abstract.** *Surfactant molecules in selective solvents can self-assemble into long flexible structures of cylindrical shape, known as wormlike micelles, and have been extensively exploited in different industrial and technological fields. However, the structures and the mechanisms that influence the formation of the wormlike micelles under the flow action are not well understood and the description of their rheological behavior remains a challenge. Above a threshold concentration, wormlike micelles can entangle, forming polymer-like microstructures that continuously break and reform, and exhibit a rich variety of viscoelastic behavior. Under imposed shear stress flow conditions, the relaxation mechanism can become highly concentrated leading to the formation of shear-banding structures. In this work, we examine the effects of a power law solvent model on the rheological response of wormlike micellar solutions in a steady Poiseuille flow through a planar channel. The VCM based model is used, which represents a network scission description for concentrated wormlike micellar solutions. The resulting equations are numerically solved using finite elements method. The simulations show that, beyond a critical shear rate, the micellar fluid exhibits a dramatic change on the velocity profiles and on the stress field as a result of the addition of a non-Newtonian solvent. A detailed analysis of the influence of the various fluid parameters on the flow field will also be made, showing that the non-Newtonian solvent exerts a deep influence on the entanglement process on the polymer chains like formation structures.*

**Keywords:** *Rheology, Non-Newtonian fluids, Wormlike micelles, Constitutive modeling*

### 1. INTRODUCTION

Surfactant molecules is a very interesting topic not only from the scientific point of view but also has many engineering and applied applications. If a selective solvent is used the surfactant molecules can self-assemble into long flexible structures of cylindrical shape known as wormlike micelles, or “living polymer”, and have been extensively exploited in different industrial and technological fields (see for example Ref. Dreiss (2007)). For example, micellar solutions are used as drag reducing agents in pipe flow, due to their ability to self-repair after mechanical breaking, and fracture fluids to stimulate the production from reservoir by taking advantage of their viscoelastic nature to transport solids, as well as their micelle forming abilities when they mix with the produced oil during enhanced oil recovery.

The adequate rheological description of the so called wormlike micelles is still a challenge, and has become an very intensive topic of scientific research (see for example Cates and Fielding (2006)). One of the models that attempts to describe the behavior of such substances is the VCM model (Vasquez, Cook and McKinley), cf. Vasquez *et al.* (2007). This model represents a network scission/reforming description for concentrated wormlike micellar solutions. The original VCM model assumes a newtonian solvent and considers that all of the non-newtonian behavior of the solution is due to the surfactant molecules. It is well known however, that it is not true for many industrial situations where now newtonian solvents are largely used (see Ezrahi *et al.* (2006), Cates and Fielding (2006) and reference therein).

In this work, the influence of a non newtonian solvent on the rheological response of wormlike micellar solutions in steady Poiseuille flow through a planar channel is analysed, and a power law model is used to describe the rheological behavior of the solvent. The resulting equations were numerically solved in the finite element software package COMSOL Multiphysics. The simulations show that, beyond a critical shear rate, the micellar fluid exhibits a dramatic change on the velocity profiles and on the stress field as a result of the addition of a non-Newtonian solvent. It is demonstrated that the solvent stress has a major influence on the break down of the long chains into short chains. This fact has a great influence on the generation or not of a centerline plug flow region due to the shear banding. The influence of the power law index is discussed, for shear thinning fluids the long chain shear stress peak is near of the channel centerline causing a low long chain concentration at the wall region. For shear thickening fluids the opposite behavior is observed and the long chain molecules can reach the wall region; that is, the contribution to the total shear stress of the long chain component is less

evident to the shear banding generation.

In section 2 of this paper, we describe our model and present the model equations. As will be discussed, the model based on an existing network scission model for wormlike micellar solutions (the VCM model). We include a power law model to describe the solvent. In section 3, we detail the numerical procedure used to solve the system of coupled, nonlinear partial differential equations in the finite element software package COMSOL Multiphysics. Then in section 4, we present results and some discussion from the model and show that a non-newtonian solvent can strongly alter the rheological behaviour of wormlike micellar solutions. Finally, we end in section 5 with concluding remarks.

## 2. NON-NEWTONIAN MODEL DESCRIPTION

The non-newtonian constitutive model used in this work is based on a two-species network scission-reforming model for wormlike micellar solutions, originally developed by Vasquez, McKinley, and Cook (The VCM model); cf. Vasquez *et al.* (2007). This model is itself based on a discrete version of “living polymer” theory (Cates, 1996) for wormlike micelles. It describes an entangled fluid in which micelles species  $A$ , which are long chains of equilibrium length  $L$ , can break apart in half to form short chains species  $B$  of equilibrium  $L/2$ , and any short two chains can recombine to form a long chain. Species  $A$  represents the average of the long chains involved in the entanglements, with an relaxation via reptation. Similarly, species  $B$  represents the average of the short micellar chains and their relaxation is via a Rouse-like mechanism. The break rate of the long species  $A$  is composed of an equilibrium breakage rate, the rate of breakage in the absence of flow, plus an additional term that depends on the local stress and the strain rate and represents the stress-induced breakage (Cromer *et al.*, 2011).

The equations governing the evolution of the number density,  $n_A$  and  $n_B$ , and the conformational stress tensors,  $\mathbf{A}$  and  $\mathbf{B}$ , associated with each species  $A$  and  $B$  are derived from the number of density distribution equations formulated in configuration space; see the Ref. Vasquez *et al.* (2007) for the detailed derivations. The VCM constitutive model in dimensionless variables is described by the evolution equations:

$$\mu \frac{Dn_A}{Dt} + \text{div}(-\delta_A(2 \text{grad } n_A - \text{div } \mathbf{A})) = \frac{1}{2} c_B n_B^2 - c_A n_A \quad (1)$$

$$\mu \frac{Dn_B}{Dt} + \text{div}(-\delta_B(2 \text{grad } n_B - 2 \text{div } \mathbf{B})) = -c_B n_B^2 + 2c_A n_A \quad (2)$$

$$\mu \mathbf{A}^\nabla + \mathbf{A} - n_A \mathbf{I} + \text{div}(-\delta_A \text{grad } \mathbf{A}) = c_B n_B \mathbf{B} - c_A \mathbf{A} \quad (3)$$

$$\epsilon \mu \mathbf{B}^\nabla + \mathbf{B} - \frac{1}{2} n_B \mathbf{I} + \text{div}(-\epsilon \delta_B \text{grad } \mathbf{B}) = 2\epsilon(-c_B n_B \mathbf{B} + c_A \mathbf{A}) \quad (4)$$

where  $D(\cdot)/Dt = \partial(\cdot)/\partial t + \mathbf{u} \cdot \text{grad}(\cdot)$  is the material derivative and  $(\cdot)^\nabla = D(\cdot)/Dt - \mathbf{L}^\top(\cdot) - (\cdot)\mathbf{L}$  is the upper convected time derivative (with velocity  $\mathbf{u}$  and  $\mathbf{L} = \text{grad } \mathbf{u}$ ),  $c_A$  is the breakage rate of the long species  $A$  and  $c_B$  is the recombination rate of the short species  $B$ , without any additional contribution due to stress. The reforming rate of the short chains is constant,  $c_B = c_{Beq}$ , and the breakage rate depends on the local strength rate of the flow and the extension of the long chains, and is given by  $c_A = c_{Aeq} + \mu(\xi/3)[2\mathbf{D} : (\mathbf{A}/n_A)]$ , where  $c_{Aeq}$  is the equilibrium breakage rate and  $\xi$  is a parameter controlling stress-induced network breakage; that is, a parameter that represents the degree of partial retraction of the micellar chains upon breakage.  $\mathbf{D} = \frac{1}{2}(\text{grad } \mathbf{u} + (\text{grad } \mathbf{u})^\top)$  is the strain tensor rate, where  $\mathbf{u}$  represents the velocity.

For a characteristic microscopic length scale we use  $\sqrt{n_A^0 k_B' T'/H_A'}$ , and the following nondimensionalization of the result equations is employed (cf. Vasquez *et al.* (2007), Pipe *et al.* (2010) and Cromer *et al.* (2011)):

$$\mathbf{x} = \frac{\mathbf{x}'}{h}, \quad t = \frac{t'}{\lambda}, \quad \mathbf{u} = \frac{\mathbf{u}'}{U_0}, \quad (\mathbf{A}, \mathbf{B}) = \frac{H_A}{n_A^0 k_B' T'} (\mathbf{A}', \mathbf{B}'), \quad (n_A, n_B) = \frac{1}{n_A^0} (n_A', n_B'), \quad c_A = \lambda c_A', \quad c_B = \lambda c_B', \quad (5)$$

where  $\mathbf{x}'$  is the spatial coordinate,  $h$  is a macroscopic characteristic length scale,  $U_0$  is a macroscopic reference velocity,  $H_A$  is the spring constant of species  $A$ ,  $k_B'$  is the Boltzmann constant,  $T'$  is the temperature,  $n_A^0$  is the dimensional value of the equilibrium number density of the long species  $A$ , and  $\lambda$  is the effective relaxation time of the network. The dimensionless variables in the above nondimensional equations include two ratios of time constants,

$$\mu = \frac{\lambda_A'}{\lambda}, \quad \epsilon = \frac{\lambda_B'}{\lambda}, \quad (6)$$

and two diffusivity parameters,

$$\delta_A = \frac{\lambda_A' D_A'}{h^2}, \quad \delta_B = \frac{\lambda_B' D_B'}{h^2} \quad (7)$$

where where  $D'_A$  and  $D'_B$  are the diffusivities of the long wormlike chains,  $A$ , and the short chains  $B$ . In the this work, dimensional quantities are denoted with a prime “'”, and dimensionless quantities without.

These evolution equations are coupled to the flow equations of conservation of momentum and mass (in dimensional variables):

$$\mathbf{0} = \text{div} \left( -p'\mathbf{I} + 2\eta\mathbf{D}' + \mathbf{S}' \right), \quad (8)$$

$$\text{div} \mathbf{u}' = 0, \quad (9)$$

where  $p'$  is the dimensional pressure field and  $\eta$  is the dynamic viscosity of the solvent. The micellar contribution to the total stress of the system is given by  $\mathbf{S}' = H_A\mathbf{A}' + H_B\mathbf{B}'$ ; when nondimensionalized using (5) a factor  $H_B/H_A = 2$  appear multiplying the conformation tensor  $\mathbf{B}$  (details of which are given later). Here, we assume that the inertial effects are negligible throughout the flow and that the flow is incompressible. For the complete spatially inhomogeneous equations the reader is referred to Ref. Vasquez *et al.* (2007).

In this work the VCM model predictions considering a power law solvent in Poiseuille flow is investigated. We consider a straight, two-dimensional microchannel of length  $L$  and height  $H$ , so that  $-H/2 \leq y \leq H/2$  and  $L \gg H$ . The forcing is a constant pressure gradient in the  $x$ -direction and the velocity field has the form  $\mathbf{u} = u(y)\mathbf{e}_x$ , satisfying the conservation of mass  $\text{div} \mathbf{u} = 0$ ; cf. Cromer *et al.* (2011). Thus, the Poiseuille flow through a straight microchannel of the VCM model the dimensionless system of equations are written as:

$$0 = 1 - \frac{d}{dy} \left( \beta \frac{du}{dy} + G(A_{12} + 2B_{12}) \right), \quad (10)$$

$$\mu \frac{\partial n_A}{\partial t} = \frac{d}{dy} \left( 2\delta_A \frac{dn_A}{dy} - \delta_A \frac{dA_{22}}{dy} \right) + \frac{1}{2} c_{B_{eq}} n_B^2 - c_{A_{eq}} n_A - \left( \frac{2\xi\mu}{3} \frac{du}{dy} A_{12} \right), \quad (11)$$

$$\mu \frac{\partial n_B}{\partial t} = \frac{d}{dy} \left( 2\delta_B \frac{dn_B}{dy} - 2\delta_B \frac{dB_{22}}{dy} \right) - c_{B_{eq}} n_B^2 + 2c_{A_{eq}} n_A + 2 \left( \frac{2\xi\mu}{3} \frac{du}{dy} A_{12} \right), \quad (12)$$

$$\mu \left( \frac{\partial A_{11}}{\partial t} - 2 \frac{du}{dy} A_{12} \right) + A_{11} - n_A = \delta_A \frac{d^2 A_{11}}{dy^2} + c_{B_{eq}} n_B B_{11} - c_{A_{eq}} A_{11} - \left( \frac{2\xi\mu}{3} \frac{du}{dy} A_{12} \right) \frac{A_{11}}{n_A}, \quad (13)$$

$$\mu \left( \frac{\partial A_{12}}{\partial t} - \frac{du}{dy} A_{22} \right) + A_{12} = \delta_A \frac{d^2 A_{12}}{dy^2} + c_{B_{eq}} n_B B_{12} - c_{A_{eq}} A_{12} - \left( \frac{2\xi\mu}{3} \frac{du}{dy} A_{12} \right) \frac{A_{12}}{n_A}, \quad (14)$$

$$\mu \frac{\partial A_{22}}{\partial t} + A_{22} - n_A = \delta_A \frac{d^2 A_{22}}{dy^2} + c_{B_{eq}} n_B B_{22} - c_{A_{eq}} A_{22} - \left( \frac{2\xi\mu}{3} \frac{du}{dy} A_{12} \right) \frac{A_{22}}{n_A}, \quad (15)$$

$$\epsilon\mu \left( \frac{\partial B_{11}}{\partial t} - 2 \frac{du}{dy} B_{12} \right) + B_{11} - \frac{n_B}{2} = 2\epsilon \left( \frac{\delta_B}{2} \frac{d^2 B_{11}}{dy^2} - c_{B_{eq}} n_B B_{11} + c_{A_{eq}} A_{11} + \left( \frac{2\xi\mu}{3} \frac{du}{dy} A_{12} \right) \frac{A_{11}}{n_A} \right), \quad (16)$$

$$\epsilon\mu \left( \frac{\partial B_{12}}{\partial t} - \frac{du}{dy} B_{22} \right) + B_{12} = 2\epsilon \left( \frac{\delta_B}{2} \frac{d^2 B_{12}}{dy^2} - c_{B_{eq}} n_B B_{12} + c_{A_{eq}} A_{12} + \left( \frac{2\xi\mu}{3} \frac{du}{dy} A_{12} \right) \frac{A_{12}}{n_A} \right), \quad (17)$$

$$\epsilon\mu \frac{\partial B_{22}}{\partial t} + B_{22} - \frac{n_B}{2} = 2\epsilon \left( \frac{\delta_B}{2} \frac{d^2 B_{22}}{dy^2} - c_{B_{eq}} n_B B_{22} + c_{A_{eq}} A_{22} + \left( \frac{2\xi\mu}{3} \frac{du}{dy} A_{12} \right) \frac{A_{22}}{n_A} \right), \quad (18)$$

where the nonlinear term  $(2\xi\mu/3) \frac{du}{dy} A_{12}$  on the right-hand side of the each equations represent the shear stress induced breakage of the long chains. We set  $U_0 = \left( -\frac{dp}{dx} \frac{h}{\rho} \right)^{1/2}$  as macroscopic reference velocity, where  $\rho$  is the density of de fluid and  $dp/dx$  is the constant pressure gradient in the  $x$ -direction. With this scaling, and (5), the dimensionless parameters in Eq. (10) include  $G = n_A^0 k_B' T' / \rho U_0^2$  which relates the microscopic elastic force for an segment of wormlike chains  $A$  and the inertial forces. The solvent contribution to the total viscosity is given by  $\beta = \eta_s / \eta_0'$ , where  $\eta_s$  is the solvent viscosity and  $\eta_0$  is the dimensional zero shear rate micellar viscosity; see Cromer *et al.* (2011).

As we examine the effects of a non-newtonian model solvent on the rheological response of wormlike micellar solutions, we generalized the solvent by allowing the dynamic viscosity in Eq. (10) to depend on the shear rate  $\dot{\gamma}$  such that  $\eta_s = \eta_s(\dot{\gamma})$ , where  $\dot{\gamma} = \sqrt{2(\mathbf{D} : \mathbf{D})} = du/dy$ . For the non-newtonian solvent we use the power law model

$$\eta_s(\dot{\gamma}) = \frac{1}{Re} \dot{\gamma}^{n-1}, \quad (19)$$

with  $Re$  the Reynolds number defined as

$$Re = \frac{U_0^{2-n} h^n}{K/\rho} \quad (20)$$

where  $n$  is the index of the power law and  $K$  is the consistence index. It is well known that fluids with  $n < 1$  are pseudo-plastic fluids; those with  $n > 1$  are dilating fluids (see for example Deville and Gatski (2012)).

For the nonlinear coupled system of partial differential equations formed by Eqs. (10)-(18), we employed the well known no slip boundary condition at the wall:

$$u = 0 \quad \text{on} \quad y = \pm 1/2, \quad (21)$$

no flux of conformation stress tensors through the wall:

$$\frac{\partial \mathbf{A}}{\partial y} = 0 \quad \text{and} \quad \frac{\partial \mathbf{B}}{\partial y} = 0 \quad \text{on} \quad y = \pm 1/2, \quad (22)$$

and no flux of the number density of species through the wall:

$$\frac{\partial n_A}{\partial y} = 0 \quad \text{and} \quad \frac{\partial n_B}{\partial y} = 0 \quad \text{on} \quad y = \pm 1/2. \quad (23)$$

Finally, the initial conditions for the number densities and conformation stress tensors write at equilibrium as:

$$n_A = n_A^0 = 1, \quad n_B = n_B^0 = \sqrt{2c_{Aeq}/c_{Beq}}, \quad \mathbf{A} = n_A^0 \mathbf{I} \quad \text{and} \quad \mathbf{B} = \frac{1}{2} n_B^0 \mathbf{I} \quad (24)$$

It is worth to noting that the diffusive coupling between the evolution equations for the stress tensor and microstructure imposes boundaries conditions for both the micellar conformation tensor and number density of each long and short chains. For the complete discussion concerning the use of these boundary conditions the reader is referred to Ref. Cromer *et al.* (2011) and reference therein.

### 3. NUMERICAL IMPLEMENTATION

The VCM model presented in the previous section has been implemented and solved numerically in the finite element software package COMSOL Multiphysics. In the implementation in COMSOL Multiphysics, the evolution equations and boundary conditions were written in general form, or strong form, which is a formulation suited for nonlinear partial differential equations (see for example Zimmerman (2006)).

We applied the PARDISO solver with a sufficiently small tolerance. All computations were carried out under the  $P^2$  (piecewise polynomial of degree two) finite element space for the velocity  $u$ , the number densities variables  $n_A$  and  $n_B$ , and the conformation stress tensors  $\mathbf{A}$  and  $\mathbf{B}$  variables. The VCM numerical predictions in Poiseuille flow have been originally given in Cromer *et al.* (2011) but without considering the effects of a non-newtonian solvent in their results. Here, the values for the VCM model parameters were fitted to data from Pipe *et al.* (2010), the same used by Cromer *et al.* (2011) in their simulations.

### 4. RESULTS AND DISCUSSION

It is well known that high concentration of large micelles can generate an evident plug flow region in some specific flow situations. It will be demonstrate that the solvent contribution can dramatically change that situation.

The influence of the solvent on the velocity profiles can be observed on Figs. 1a and 1b, for shear thinning fluids,  $n < 1$ , the central plug flow region becomes more evident as the exponent index  $n$  becomes smaller. The opposite behaviour is observed for the shear thickening situation. This fact can be explained by observing Fig.2, for shear thinning fluids the  $\dot{\gamma}$ , which is responsible for the break down of the long micellar chains, is much larger at the near wall region.

Figures 3 and 4 show the influence of the non-newtonian solvent characteristics on the long and short chains tensor. Note that the individual shear stress contribution for the total shear stress vary in a nonlinear way. For low values of  $n$ , the shear stress contribution due to the long chain is very low near the wall. The behaviour of number densities  $n_A$  and  $n_B$ , see Fig. p6, are in agreement with the conformation stress tensor  $\mathbf{A}$  and  $\mathbf{B}$  (see Figs. 3 and 4). The spatial variations in the number density for long "A" and short "B" chains is shown for several values of power law index. It is clear that the shear stress for it component is related to the respective concentration levels. At low power law index values, the majority of the micellar species in the flow exist as long chains in the center of the gap channel and  $n_A$  is almost constant zero at the near the wall region. The opposite behaviour is observed for the short chains and  $n_B$  is almost constant at the near wall region.

The viscosity behaviour is described in Fig. 5. It is clear that a two power law plateaus are present for high and low values of  $\dot{\gamma}$ . This behaviour can be described as a power law model with a Carreau like consistent index as suggested by Cruz *et al.* (2013).

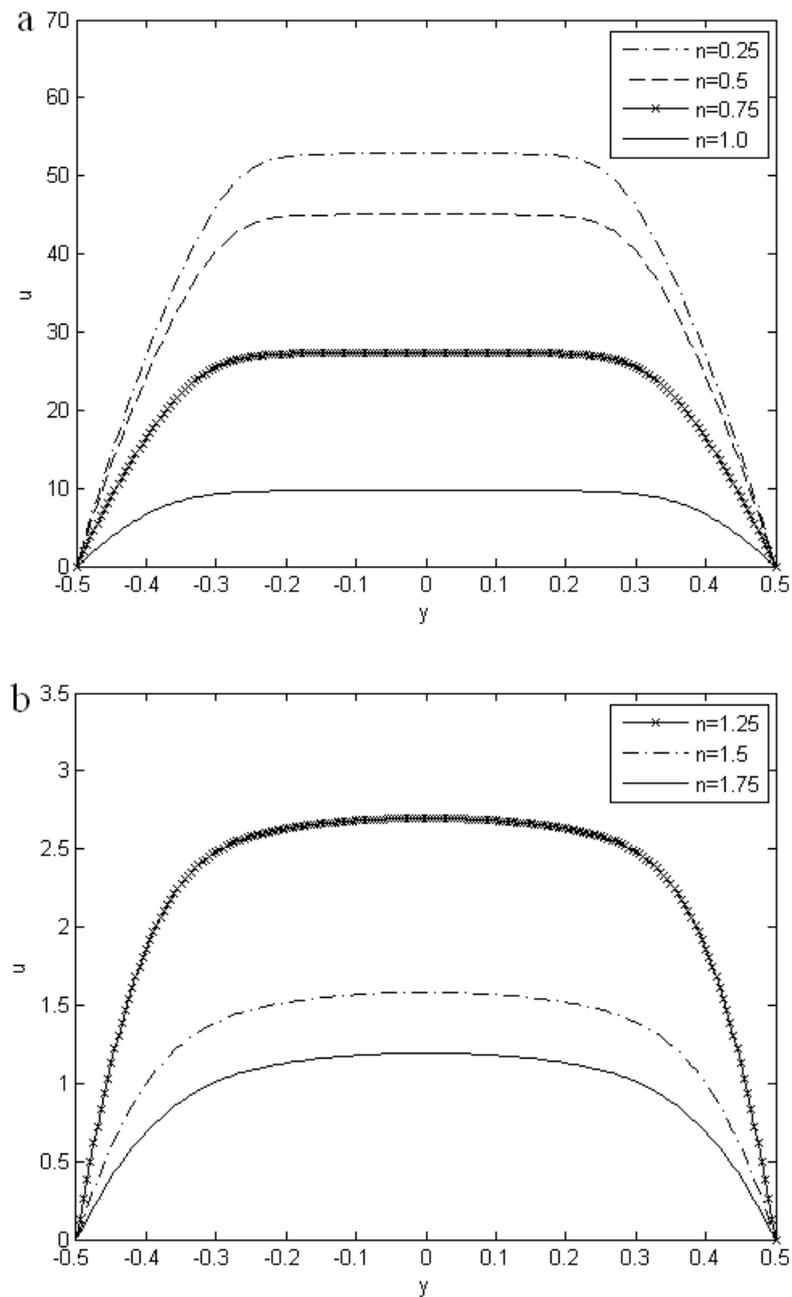


Figure 1. Shear banding velocity profiles for the VCM model considering a non-newtonian solvent for Reynolds number  $Re = 300$ , dimensionless diffusion coefficient  $\delta = 10^{-1}$ ,  $G = 0.4$ , and various values of the power law index. For (a)  $n = 1, 0.75, 0.5$ , and  $0.25$ , (b)  $n = 1.25, 1.5, 1.75$ . It is clear that the the non-newtonian solvent have an important influence on the sharpness of the transition between the high and low shear rate bands as well as the magnitude of the centerline plug-flow velocity depends on the value of the power law index  $n$ .

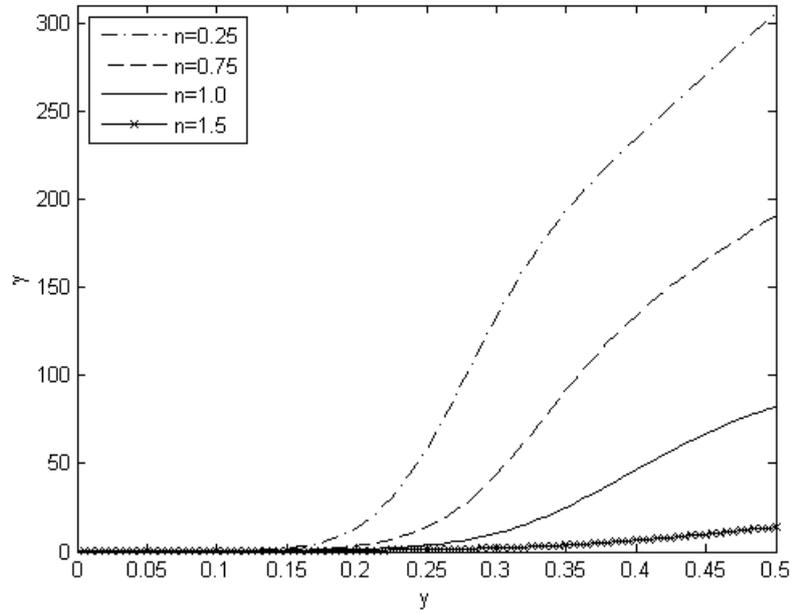


Figure 2. Variation in the shear rate across the the gap for  $n = 0.25, 0.75, 1,$  and  $1.5$ . It is clear that the magnitude of the shear rate near the wall region depends on the value of the power law index  $n$ .

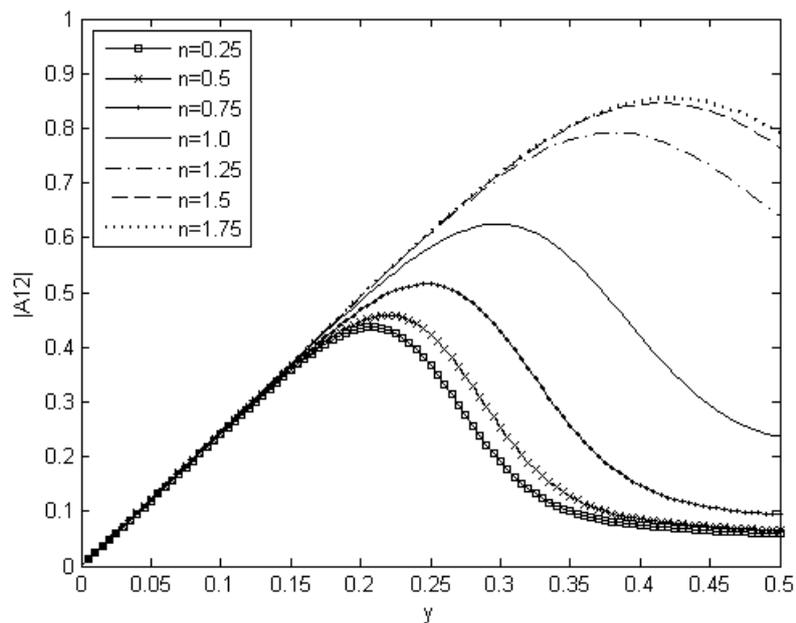


Figure 3. Variation in the conformation tensor  $A_{12}$  of the long chain (“A” species) to the shear stress across the gap channel for Reynolds number  $Re = 300$ , dimensionless diffusion coefficient  $\delta = 10^{-1}$ ,  $G = 0.4$ , and various values of the power law indexes:  $n = 0.25, 0.5, 0.75, 1.0, n = 1.25, 1.5, 1.75$ . The shear stress contribution due to the long chain is very low near the wall for low values of the power law index.

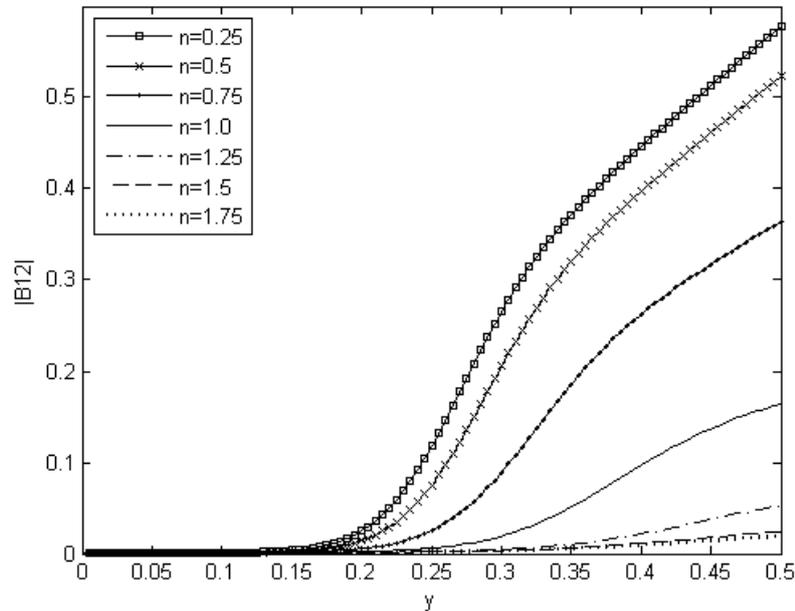


Figure 4. Variation in the conformation tensor  $B_{12}$  of the short chain (“B” species) to the shear stress across the gap channel for Reynolds number  $Re = 300$ , dimensionless diffusion coefficient  $\delta = 10^{-1}$ ,  $G = 0.4$ , and various values of the power law indexes:  $n = 0.25, 0.5, 0.75, 1.0, n = 1.25, 1.5, 1.75$ . The shear stress contribution due to the short chain is very high near the wall for low values of the power law index.

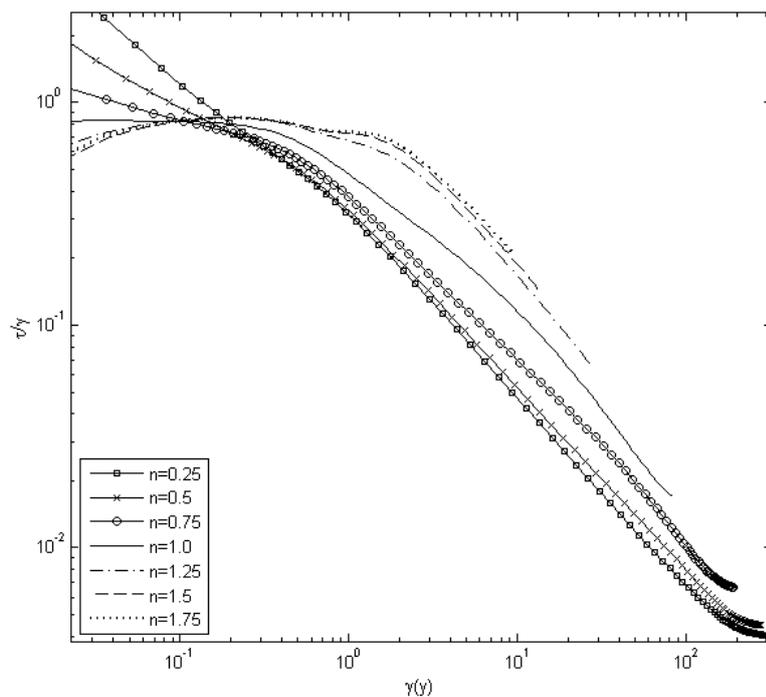


Figure 5. The steady-state shear viscosity,  $\tau/\dot{\gamma}$ , as a function of the shear rate,  $\dot{\gamma}$ . The model results are for  $Re = 300$ ,  $\delta = 10^{-1}$ ,  $G = 0.4$ , and different values of the power law index:  $n = 0.25, 0.5, 0.75, 1.0, n = 1.25, 1.5, \text{ and } 1.75$ . It is clear that a two power law plateaus are present for high and low values of  $\dot{\gamma}$ .

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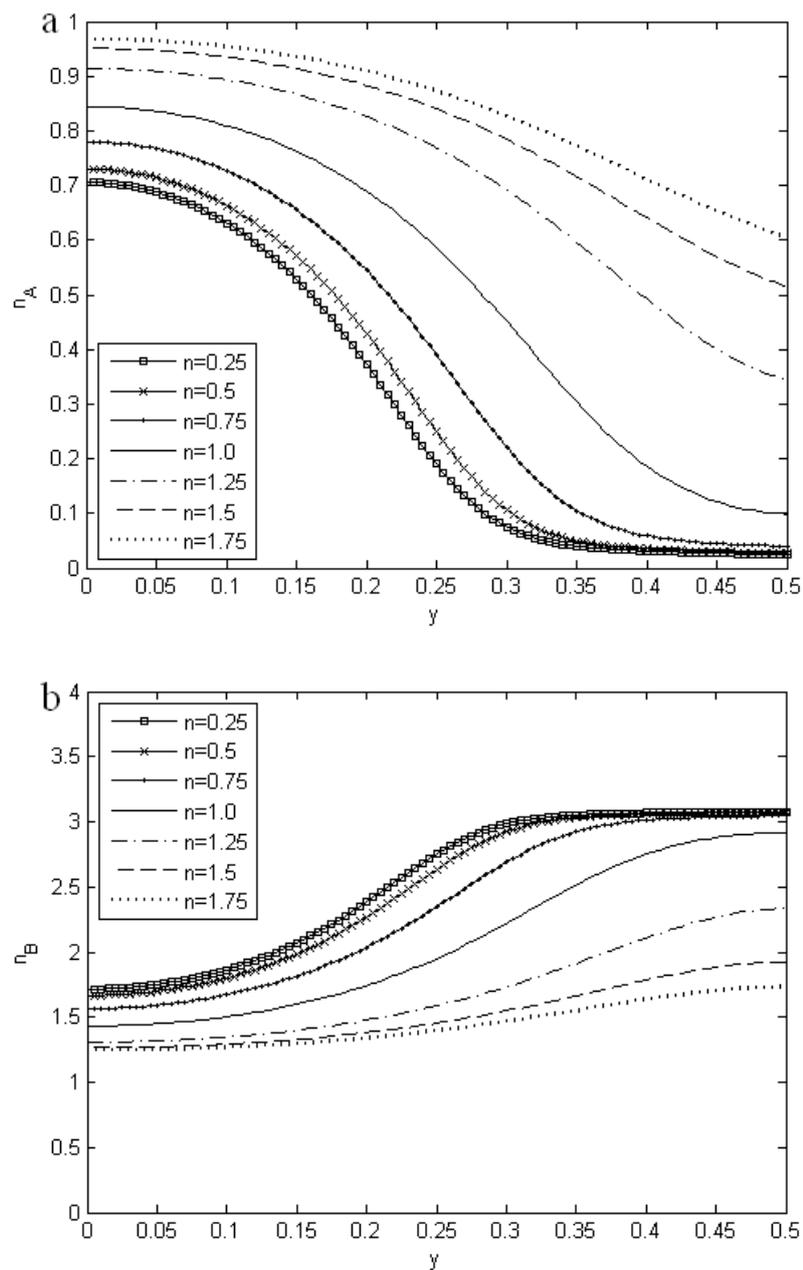


Figure 6. Variation in the number density across the gap channel for Reynolds number  $Re = 300$ , dimensionless diffusion coefficient  $\delta = 10^{-1}$ ,  $G = 0.4$ , and various values of the power law index:  $n = 0.25, 0.5, 0.75, 1.0, n = 1.25, 1.5, 1.75$ .  
 (a) Number density of long chains ("A" species), (b) Number density of short chains ("B" species).

## 5. CONCLUDING REMARKS

We have studied the effects of a non-newtonian solvent on rheological response of wormlike micellar solutions in steady Poiseuille flow through a planar channel. By modifying the existing VCM model for concentrated wormlike micellar solutions through the addition of a power law model to describe the solvent, we have demonstrated the influence of a selective non-newtonian solvent on the scission and reforming of micelles. As a consequence, a dramatic change on the stress field and on the velocity profiles is observed with a high shear band developing near the walls and a low shear rate, or plug-like flow, near the centerline of the channel. The predicted variations in the local stress viscosity versus shear rate are similar with those observed in Ref. Cruz *et al.* (2013). This suggests that a non-Newtonian solvent can exert a deep influence on the entanglement process of “living polymer” chains. In closing, we believe that the effects of selective solvents should be considered in the modeling process of wormlike micellar solutions. However, more investigations are needed (which include other non-newtonian solvent models, micellar systems, “living polymers” solutions, etc.) for new insight on rheological response of micellar solutions.

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