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Microfluidic flows of wormlike micellar solutions

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ABSTRACT

The widespread use of wormlike micellar solutions is commonly found in household items such as cosmetic products, industrial fluids used in enhanced oil recovery and as drag reducing agents, and in biological applications such as drug delivery and biosensors. Despite their extensive use, there are still many details about the microscopic micellar structure and the mechanisms by which wormlike micelles form under flow that are not clearly understood. Microfluidic devices provide a versatile platform to study wormlike micellar solutions under various flow conditions and confined geometries. A review of recent investigations using microfluidics to study the flow of wormlike micelles is presented here with an emphasis on three different flow types: shear, elongation, and complex flow fields. In particular, we focus on the use of shear flows to study shear banding, elastic instabilities of wormlike micellar solutions in extensional flow (including stagnation and contraction flow field), and the use of contraction geometries to measure the elongational viscosity of wormlike micellar solutions. Finally, we showcase the use of complex flow fields in microfluidics to generate a stable and nanoporous flow-induced structured phase (FISP) from wormlike micellar solutions. This review shows that the influence of spatial confinement and moderate hydrodynamic forces present in the microfluidic device can give rise to a host of possibilities of microstructural rearrangements and interesting flow phenomena.

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Contents

1. Introduction

Surfactants are amphiphilic molecules that consist of a bulk hydrophilic head group, which can be neutrally, positively, or negatively charged, and a hydrophobic tail. When the surfactant concentration is increased above a critical micelle concentration (CMC), surfactant monomers will spontaneously self-assemble into large aggregates known as micelles (in aqueous solutions) to shield the hydrophobic tails from water [1]. The morphological variations of these micelles are dependent on the temperature, pH, concentration, salinity, surfactant packing parameter, and flow conditions. Wormlike micelles are characterized as cylindrically elongated and semi-flexible aggregates. For example, cationic surfactants such as hexadecyltrimethylammonium bromide (CTAB) or cetylpyridinium chloride (CPyCl) in aqueous solutions can form wormlike micelles under specific temperatures and

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concentrations. The addition of inorganic salts, such as sodium chloride (NaCl) and sodium nitrate ($NaNO_3$), or strongly binding counterions such as organic salt sodium salicylate (NaSal), have shown to considerably reduce the CMC. These salts serve to screen electrostatic repulsion of neighboring charged hydrophilic headgroups in the aqueous solution and result in a steep increase of the shear viscosity, which can be correlated with the transition from spherical to wormlike micelles in the surfactant solution [2–4]. The characteristic length scale, flexibility, and interaction between wormlike micelles largely depend on the chemical nature and concentrations of surfactants and salts [1,5–9].

Wormlike micelles are commonly found as additives in consumer hair products, drag reduction agents, and fracture fluids in oil and gas productions [10–13] for their viscoelastic and gel-like properties [14–16, 2,4,3,17,18]. The size and shape of wormlike micelles bear striking similarities to those of polymer chains but with larger diameters (1–10 nm) and longer persistence lengths (10–100 nm) [19–21]. Thus, wormlike micelles have also been referred to as living polymers because they resemble micron sized polymeric chains weakly held together by physical interactions that constantly undergo scission and recombination. Cates model [22], which assumes that a wormlike micellar chain can break and recombine with equal probability, predicts that the stress relief in the wormlike chains can be accomplished by Brownian motion driven reptation, and reforming new end caps if the micelles break and reform faster than their reptation time scale. The linear viscoelastic response of these wormlike micellar solutions can be described by the Maxwell model that characterizes fluid viscosity and elasticity with a single relaxation time. Shikata et al. [23–25] characterized wormlike micellar solutions as either linear and partly entangled or highly entangled and branched micellar networks. They observed that the weakly interacting and short wormlike micellar system could be described by the Cates model (with reversible chain recombination and scission) [22]. However, when a considerably stable and longer micellar network is formed (i.e., an equal molar CTAB:NaSal system), the micelles become too long and stable to reptate. Instead, the micelles exhibit scission and recombination at their entanglement points, with Sal^- ions serving as catalysts [23,25]. By using both traditional bulk rheometry and microfluidic approaches [26–39], the study of wormlike micellar solutions under flow has been ongoing for the past few decades with increasing understanding of their linear and non-linear rheological behavior, microstructural transitions, and higher-ordered structural formations under various external conditions (see reviews [10,40–47]).

Microfluidics has emerged in recent years as a versatile method of handling fluids at small length-scales, in particular, for generating and manipulating complex fluids with controllable size, tailored structure and functionality [48,49]. Microfluidics has been employed to study effects of confined geometry on fiber deformations [50], dynamics of DNA molecules [51,52], and bacteria motion [53–55] at low-Reynolds numbers (see reviews on microfluidics and their applications in [56–58]). More recently, microfluidic systems have been exploited as microfluidic-rheometers [28–32,36–39,59] or as tools to study and generate new microstructures and new flow phenomena [28–32,36–39, 60–65]. Traditional shear rheometry, with typical rheometer gap sizes at least one order of magnitude greater than the intrinsic length scale of the material microstructure, is generally used for measuring fluid responses to shear with gap sizes of O (1 mm) and deformation rates in the range of O (10^2–10^3 s^{-1}) [32,38,39]. Although rheometers with parallel plate geometries and gap sizes in the range of 10–500 μm can be used to access high deformation rates (up to 10^2 s^{-1}), it requires careful alignment of the plates and identifying zero gap error [26,27]. Additionally, when the deformation rate reaches 10^4 s^{-1} or higher, complications with edge fracture, secondary flow, and viscous heating become significant for complex fluids [32,38,39,59]. By taking advantage of the micron length scales present in microfluidics, higher deformation rates ($\geq 10^5$ s^{-1}) can be easily achieved with negligible inertial or viscous heating effects [39]. A comprehensive review of microfluidic rheometry on capillary, stagnation, and contraction flows has been reported by Pipe and Mckinley [38,39]. A complementary review on extensional flow fields within the context of microfluidic rheometry can be found in Galindo et al. [66]. In addition, microfluidics have recently been used to probe the influence of spatial confinement on wormlike micelles [28–32,36–39,61–65] and to provide a robust and efficient path to study the flow of wormlike micelles in geometries similar to the ones found in porous media flow, oil recovery, and drug delivery applications [63–65,67–71].

In this article, we first review and discuss recent experimental and theoretical work of wormlike micelles under shear and elongational flow fields by using microfluidics with integrated flow interrogation techniques (i.e., micro particle image velocimetry (μ-PIV) [29,31,60,72,73] and flow induced birefringence (FIB) [28,36,37,73]). We then highlight the behavior of wormlike micellar solutions under complex flow fields and illustrate how spatial confinement and high strain rates provided by microfluidics offer unique opportunities to create new microstructures. We conclude with outlook for future research on microfluidic flows of wormlike micellar solutions.

2. Wormlike micelles under shear flow in microfluidics

2.1. Shear banding of wormlike micellar solutions

2.1.1. Background

Under simple shear flow, such as in a Couette cell shown in Fig. 1A, homogeneous flow of wormlike micellar solutions can become unstable and separate into coexisting shear bands with different local viscosities and internal structures [20,75,76], above a critical shear rate or shear stress γ_1. Rehage and Hoffmann [20] first reported the rheological behavior of the stress plateau in reversible giant wormlike micellar solutions experimentally with the micelles being slightly oriented and maintained homogeneous when $\dot{\gamma} < \dot{\gamma}_1$. When $\dot{\gamma}_1 < \dot{\gamma} < \dot{\gamma}_2$, the flow becomes unstable with two stable branches containing different viscosities coexisting at the constant stress σ^* regime. When $\dot{\gamma} > \dot{\gamma}_2$, a homogeneous fluid phase is recovered and the micelles are again strongly aligned in the flow direction. Macroscopic rheology coupled with the local measurements such as nuclear magnetic resonance [77, 78], dynamic light scattering [79], PIV [80], high frequency ultrasonic velocimetry [81,82], rheo-optics [47] and other techniques [41,72,74, 76,83–95] have been widely used to explore the complex dynamics of the internal structure of the fluid and flow at the shear plateau regime. For example, employing a shear rheometer coupled with flow birefringence and small angle neutron scattering (SANS), Cappelaere et al. [76] showed an isotropic to nematic phase transition of a CTAB based wormlike micellar system under shear flow, by spatially resolving the shear bands in flow-vorticity plane and convoluting across the structures of both bands. Liberatore et al. [90,96] probed the microstructure evolution of similar CTAB based shear banding solutions in a Couette cell, by using a combination of dynamic rheology, rheo-optics, and small angle neutron scattering (SANS). Compared with the results from Cappelaere et al. [76], spatially-resolved SANS measurements in the flow-gradient plane provided more insights towards both segment orientation and degree of alignment of micelles in low and high shear bands. Subsequently, Helgeson et al. [97,98] used the same shear cell SANS setup to study two CTAB solutions with similar values of λ_0, G_0, and ξ, one shear thinning (15.6 wt.% CTAB in D_2O) and one shear banding (16.7 wt.% CTAB in D_2O). In contrast to a weak distortion of scattering patterns in the non-shear banding system, a high degree of scattering anisotropy was observed in the shear banding solution after reaching a critical shear rate. By quantitatively coupling anisotropy data with Giesekus constitutive model with stress diffusion, Helgeson et al. proposed a critical anisotropic parameter α that corresponded to the critical level of segment orientation and alignment required for shear banding. Berret et al. [83–85] also observed the isotropic to nematic phase transition induced by shear flow in CPyCl/NaSal in 0.5 M NaCl aqueous solution with the surfactant CPyCl concentration ranging from 1% to 30% by

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