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Effect of EG and low temperature on solution behaviors of wormlike micelles

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ABSTRACT

Wormlike micelles (WLMs) are prepared in ethylene glycol (EG)-aqueous mixed solution. Gemini surfactant, 2-hydroxyl-propanediyl- α,ω -bis-(dimethyldodecylammonium bromide) and sodium benzoate were used, referred as 12-3(OH)-12 and NaBen respectively, to construct WLMs. The effect of EG on WLMs was investigated by rheological measurements. Steady rheological responses of mixed solution slide down like a terrace with the displacement of water by EG. Dynamic rheology spectra shift towards high angular frequency and the mixture transformed from viscoelastic to less viscous fluid. The addition of NaBen into 12-3(OH)-12 solution causes similar salt effect within different EG content. However, the peaks of viscosity and relaxation time both shifts towards high NaBen concentration and the maximum values decreases one order of magnitude when EG content varying from 0% to 20%. All rheological behaviors were investigated below room temperature. The results show that the WLMs system still keeps desirable rheological properties in EG-water system. The evolution of sample viscosity upon temperature conforms to Arrhenius law. The study provides deep comprehension on the self-assembly behavior of WLMs in low-temperature region.

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1. Introduction

Due to unfavorable contact with water, the surfactants can spontaneously self-assemble into kinds of aggregates, such as spherical micelles, rodlike micelles, wormlike micelle (WLMs), vesicle, bilayer structure etc., among which WLMs have attracted plenty of research interests [1,2]. The formation of WLMs is a consequence of unidimensional micellar growth [3]. Above a critical overlapping concentration, WLMs entangle with each other into transient network and exhibit viscoelastic properties, analogous to flexible polymer solutions [4,5]. Unlike polymer susceptible to mechanical shearing, WLMs can be deformed and reformed reversibly when exposed to shear destroying. Consequently the rheological properties can survive under some harsh conditions [4]. The unique properties makes WLMs find extensive applications in industry fields, such as drag reduction, enhanced oil recovery, clean fracturing fluid, etc. [6,7]. (See Scheme 1.)

Hydrophobic interaction or solvophobic interaction between surfactants is the primary drive force for micelle growth. The solvent properties play a decisive role in controlling the micellization and aggregation behaviors [8]. The majority of theoretical studies and practical applications are centered on the aqueous media. The studies on non-aqueous or alcohol-water media have been seldom reported. But for some industrial applications involving alcohol medium, such as cosmetics, paints and detergents, also need rheology controller. The introduction of co-solvent, like glycerol and ethylene glycol (EG), are unfavorable for the aggregation of micelles due to reduced cohesive energy [8]. Therefore the surfactant molecules tend to form spherical aggregates in these solvents, and most of researches are focused on aggregation behavior including aggregation number, micelle size and counterions, etc. [9–13]. Rodrguez et al. studied the effect of EG content on the critical Gemini concentration at sphere-to-rod transition [14]. The increase of EG weight percentage is unfavorable for Gemini to form cylindrical micelles. The emphasis of the study is also put on the aggregation behavior of spherical micelles. Hoffmann and Abdel-Rahem have successfully prepared WLMs using conventional cationic surfactant cetyltrimethylammonium bromide (CTAB) and sodium salicylate (NaSal) with 1,3-butanediol and glycerol as co-solvent [15,16].

EG are important and even indispensible ingredients in antifreezing fluid or anti/deicing fluids for cars and aircraft which also demand good rheological reagent. The commonly used rheology controller in anti-icing fluid is polymer solution. The macramolecular structure of polymer also introduces adverse limitations except rheology modification for the present composition, such as gelation and stratification. The small molecule self-assembled structure of wormlike micelles makes it a potential candidate to apply to anti-icing fluid. Considering its weaker polarity than water, it is challengeable to prepare viscoelastic WLMs in EG-rich solvent. Gemini surfactants have received extensive attentions in both pure and applied sciences [17,18]. They show superiority and high efficiency in self-assembling into WLMs, including strong hydrophobic contribution, tightly bonded headgroups, and tailored configuration fit for molecule pack [19].

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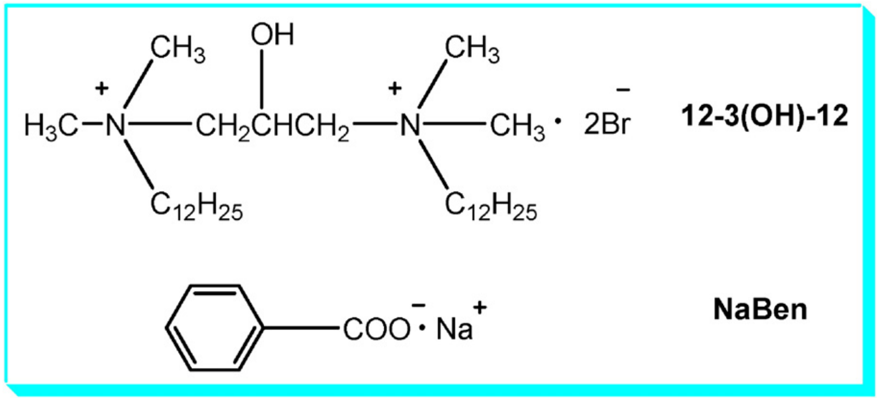

Scheme 1. Chemical structure of 12-3(OH)-12 and NaBen.

In this paper, Gemini surfactant, 2-hydroxyl-propanediyl- α,ω -bis-(dimethyldodecylammonium bromide), referred as 12-3(OH)-12 is employed to construct WLMs under the inducement of sodium benzoate, referred as NaBen. As water is replaced by EG gradually, the rheological behavior of WLMs solution was studied in detail to understand the influence of EG. In addition, the effect of temperature on apparent viscosity was further investigated in the range from 0 to 25 °C.

2. Materials and methods

Synthesis of gemini surfactants 12-3(OH)-12 was performed as described in refs. [20,21]. NaBen, (supported by Sinopharm Chemical Reagent Co.) analytical grade reagent, was used as received. EG was supported from Tianjin Kermel Chemical Reagent Company and used without further purification. The samples were prepared by adding appropriate surfactants and NaBen into accurately mixed solvents which are composed by EG and water in weight percentage. Water was purified by the Millipore Milli-Q system.

Rheolgical measurements were performed on a stress controlled rheometer (TA Instruments, AR2000ex) with cone-plate sensor (diameters of 40 mm and a cone angle of 1°). A sample cover provided with the instrument was used to minimize the change of sample composition by evaporation during the measurement. Dynamic rheological experiments were performed in the linear viscoelastic regime, which was examined from dynamic strain sweep measurement. The stress during oscillation frequency measurement was fixed at 0.1 Pa. The temperature was kept at 10 °C throughout the experiments unless otherwise specified. Samples were equilibrated at the temperature of interest for at least 20 min prior to experiment.

3. Results and discussion

3.1. Effect of EG content on steady rheology

The effect of NaBen concentration on rheological properties is investigated at fixed 12-3(OH)-12 concentration, 70 mM. The solvent composition is designed by displacing water with EG in weight percentage. The steady rheology for samples with varied NaBen concentration and solvent composition are comparatively exhibited in Fig.1. We take the pure water system free of EG as an example to describe. The high viscosity at low shear rate manifests the formation of giant tridimensional network. The viscosity keeps constant irrespective of shear rate in the regime, suggesting that the network receives negligible influence from mild shear. In the regime of high shear rate the shear-thinning phenomenon is observed, which is ascribed to the rearrangement of micelles along the direction of shear [22,23]. Samples with different NaBen concentration exhibit similar steady rheo-grams, while the plateau viscosity fluctuates with NaBen content. The plateau viscosity enhances with NaBen and then decreases. Zero-shear viscosity (η_0) was obtained by extrapolating the viscosity plateau to zero shear rates. The data of η_0 under varied NaBen and EG content is shown in Fig.4A.

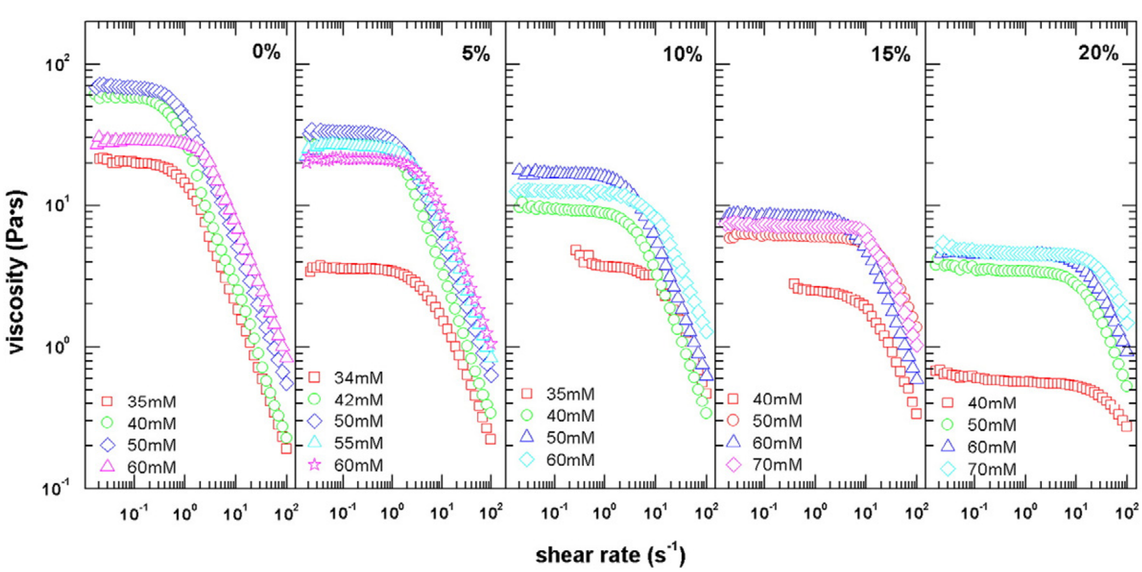

Fig. 1. The effect of NaBen concentration and EG weight percentage on the steady-shear rheology of 70 mM 12-3(OH)-12 samples at 10 °C.

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