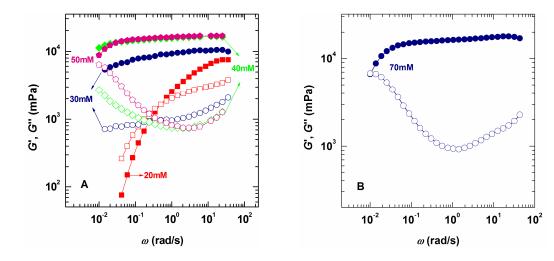
## Wormlike Micelles Formed by Sodium Erucate in the Presence of a Tetraalkylammonium Hydrotrope

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## 4 S3.2. Additional Figures for the effect of hydrotrope concentration on rheological 5 behaviors





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Figure S1. Dynamic rheology for the NaOEr-BTAB system at different BTAB concentrations,
 (A) 20, 30, 40 and 50 mM; (B) 70 mM. The NaOEr concentration is held constant at 70 mM.
 The lines are guide for eyes.

4 The supplementary dynamic rheology of NaOEr-BTAB system is plotted against frequencies 5 showing in Figures S1. The data of G' and G'' at 25 mM BTAB are characteristic of a 6 viscoelastic behavior (Figure 2A). However, G" increases within the investigated frequencies 7 monotonously rather than decrease at some frequency like reptative behavior, and G' shows no 8 obvious plateau at high frequencies, which deviates from the Maxwellian model. G' and G'' of 9 samples containing 30 mM, 40 mM and 50 mM BTAB show no crossovers within accessible 10 frequencies range. At 70 mM TBAB, the frequency spectrum shows a crossover of G' and G''. 11 With the increase of salt content, the dynamic spectra show gradual approximation to 12 Maxwellian model.

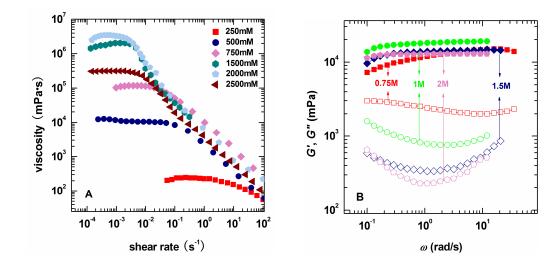
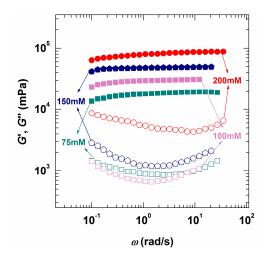




Figure S2. Steady (A) and dynamic (B) rheology for the NaOEr-TMAB system at different
TMAB concentrations. The NaOEr concentration is held constant at 70 mM.

16 The steady and dynamic rheological behaviors of NaOEr-TMAB system are shown in Figures 17 S2.  $\eta_0$  shows initial increase within the salt content from 250 mM to 2 M. Thereafter further

addition of salt causes a decrease of viscosity. It is normally believed the origin of reduced viscosity is resulted from formation of branched micelles,<sup>1</sup> here probably from the coexistence of vesicles and sponge phases. Within the accessible frequencies, no crossovers of G' and G'' are observed for all the investigated samples.



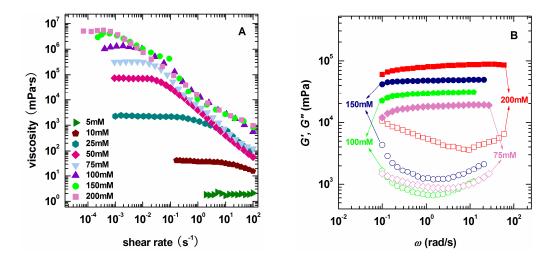
## 5 S3.3. Additional results for effect of surfactant concentration on rheological behaviors.

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7 Figure S3. Dynamic rheology for the NaOEr-BTAB system at different NaOEr concentrations.

8 The BTAB concentration is held constant at 50 mM.

9 The dynamic responses of the NaOEr-BTAB samples with increasing surfactant concentration 10 are shown in Fig S3. G' and G'' have no intersections within more than two decade of 11 frequencies. The crossovers may exist at smaller frequencies. This denotes that micellar solutions 12 possess very long relaxation time. The plateau moduli increase steadily with increasing 13 surfactant concentration.



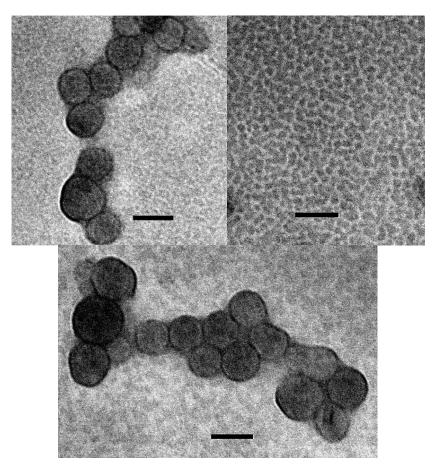
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Figure S4. Steady (A) and dynamic (B) rheology for the NaOEr-TMAB system at different
NaOEr concentrations. The TMAB concentration is held constant at 50 mM.

4 The steady and dynamic rheological properties for the NaOEr-TMAB system at different 5 NaOEr concentrations are shown in Figure S4. The rheological responses present much 6 similarity with those of NaOEr-BTAB samples.

In order to further verify the presence of vesicle phases and sponge phases in the sample system "150 mM NaOEr+1000BTAB" ( $C_S/C_D=14.3$ ), two other typical graphs are adopted and inserted in Figure S5. They are selected from different regions of the same sample solution. Several vesicles are clearly exhibited in Figure S5A and sponge phases are definitely present in Figure S5B.

The properties of ionic surfactant systems are strongly dependent on the salt content. At low salt concentration, the surface charges on headgroups are not sufficiently screened. The flexibility of the cylindrical aggregates is restricted.<sup>2</sup> The rheological experiment deviates significantly from the Cates model, which is founded on the base of neutral or completely screened micelle. With increasing salt concentration, the charges on the micelles are further screened and the responses approach theoretical model. Such viscoelastic behavior complying with the Maxwell behavior is indicative of flexible wormlike micelles,<sup>3</sup> which is further
confirmed by Cryo-TEM observations (Figure 4A in the main text).



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5 Figure S5. Different Cryo-TEM images of the sample 150 mM NaOEr+1000BTAB 6  $(C_S/C_D=14.3)$ . The bars are 100 nm.

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## 8 **References**

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