Supplementary Information (SI) for Soft Matter.

Viscoelastic behavior of the wormlike micellar solutions formed by an aspartame-based bicephalous anionic surfactant

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1. Synthesis of C18-AP-2Na

The detailed synthetic process, which is shown in **Figure 1**, is described as follows.

(1) Synthesis of **Intermediate 1**. Stearic acid (28.5 g, 0.1 mol) was added in a 500 mL three-necked flask. Thionyl chloride (14.3 g, 0.12 mol) was slowly added at 65 ℃, and the acidic gas produced was absorbed by NaOH aqueous solution. The reaction was carried out for 3 h until the system became clear and transparent, and no bubbles were produced in the NaOH solution. Subsequently, $SOCl_2$ was removed under reduced pressure to obtain a transparent yellow viscous liquid, which is stearoyl chloride. Aspartame (35.3 g, 0.12 mol) and triethylamine (25.5 g, 0.25 mol) were dissolved in a 1000 mL three-necked flask containing dichloromethane, and a dichloromethane solution of stearoyl chloride (30.3 g, 0.1 mol) was slowly added at 25 ℃. After the addition was complete, the reaction was continued for 2 h. The reaction mixture was then transferred to a 1000 mL separatory funnel and extracted with weakly acidic water 2 to 3 times, followed by extraction with deionized water 3 times until the pH was between 3 and 4. Anhydrous magnesium sulfate was added to dry the dichloromethane phase, and the solid magnesium sulfate was removed by suction filtration. The filtrate was then rotary evaporated under reduced pressure to obtain Intermediate 1. Yield: 89 %.

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(2) Synthesis of **C18-AP-2Na**. Intermediate 1 (30 g, 0.053 mol) was dissolved in ethanol in a 1000 mL flask at 70 °C. Then, NaOH ethanol solution (4.5 g, 0.11 mol) was added, and the reaction was carried out at 70 °C for 2 h. After the reaction was completed, the product was filtered. After vacuum drying, the final product was obtained as white solid. Yield: 93 %.

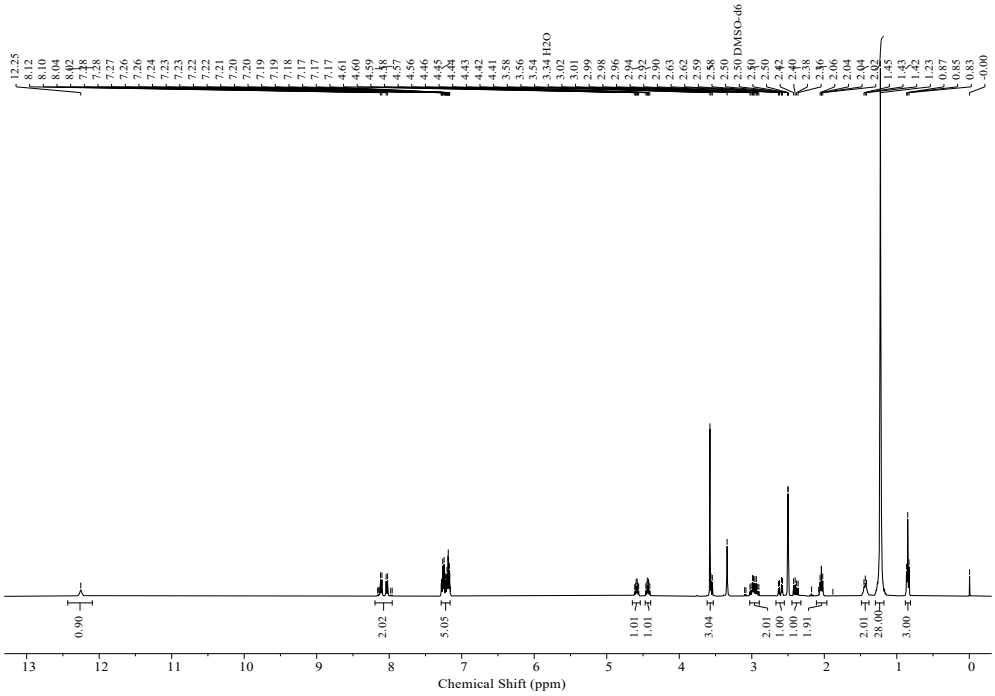
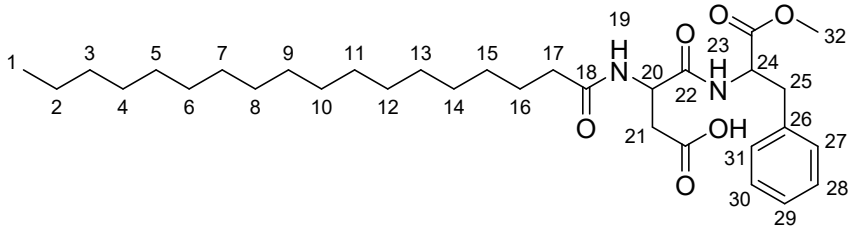


Fig. S1 [1]H NMR spectrum of Intermediate 1 (DMSO)



[1]H NMR（400 MHz, DMSO）δ 12.52 (s, 1H, COOH), 8.16-7.96 (t, 2H, NH), 7.18-7.28 (m, 5H, C27-1H ,C28-1H ,C29-1H ,C30-1H, C31-1H), 4.58 (q, 1H, C20-1H), 4.44 (q, 1H, C24-1H), 3.58 (s, 3H, C32-H), 3.10-2.90 (dd, 2H, C21-1H, C25-1H), 2.58 (dd, 1H, C21-1H), 2.42 (dd, 1H, C25-1H), 2.04 (t, 2H, C17-2H), 1.43 (t, 2H, C16-2H),1.29-1.17(m,28H,C2-2H, C3-2H, C4-2H, C5-2H, C6-2H, C7-2H, C8-2H, C9-2H, C10-2H, C11-2H, C12-2H, C13-2H, C14-2H, C15-2H) ,0.85 (t, 3H, C1-3H).

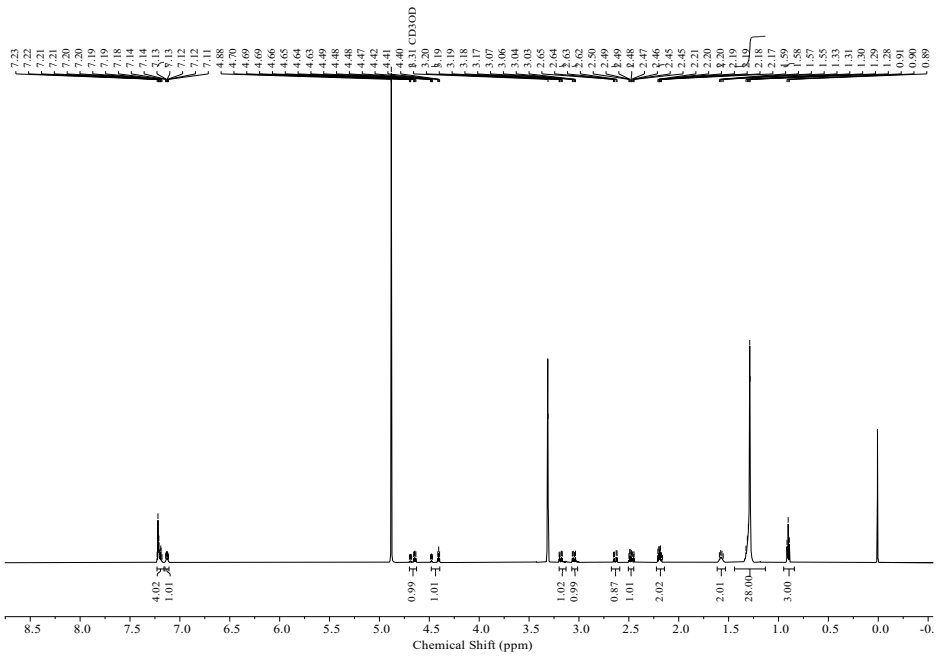
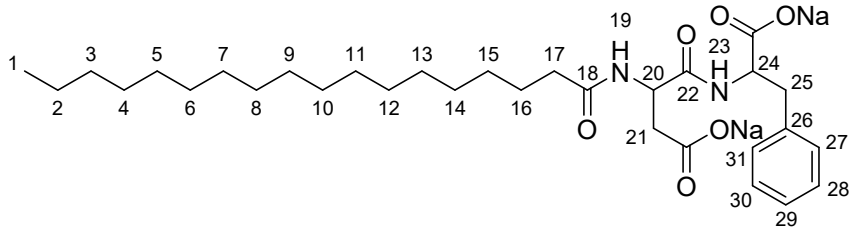


Fig. S2 ^{1}H NMR spectrum of C18-AP-2Na (CD_3OD)



^{1}H NMR（400 MHz, CD_3OD）δ 7.12-7.22 (m, 5H, C27-1H ,C28-1H ,C29-1H ,C30-1H, C31-1H), 4.63 (q, 1H, C20-1H), 4.41 (q, 1H, C24-1H), 3.18(dd, 1H, C21-1H), 3.03(dd, 1H, C25-1H)，2.63 (dd, 1H, C21-1H), 2.48 (dd, 1H, C25-1H), 2.18 (t, 2H, C17-2H), 1.57 (t, 2H, C16-2H),1.34-1.21(m,28H,C2-2H, C3-2H, C4-2H, C5-2H, C6-2H, C7-2H, C8-2H, C9-2H, C10-2H, C11-2H, C12-2H, C13-2H, C14-2H, C15-2H) ,0.85 (t, 3H, C1-3H).

2. Physical appearances of C18-AP-2Na solutions with different concentrations

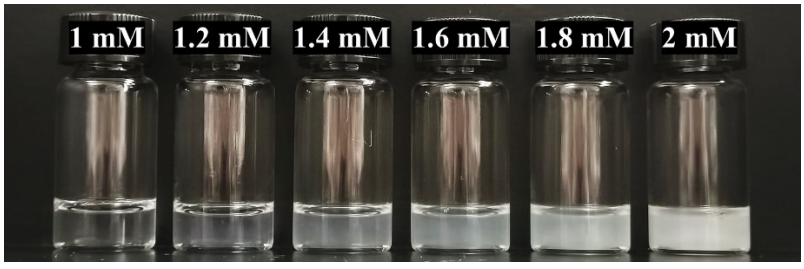


Fig. S3 Macroscopic photographs of C18-AP-2Na with different concentrations at 25 °C.

3. Surface tension and Fluorescence measurement

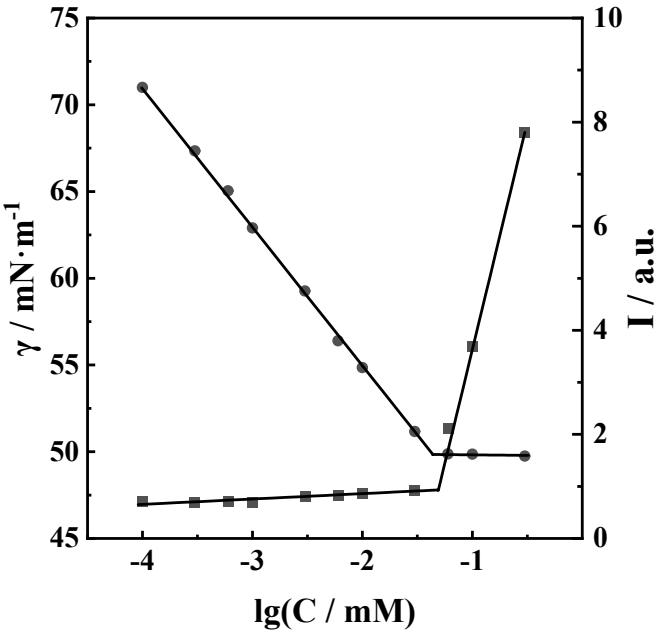


Fig. S4 Variations of the equilibrium surface tensions and Nile Red fluorescence intensity with C18-AP-2Na concentrations (pH = 12) at 25 °C.

4. Physical appearances of C18-AP-2Na/CTAB mixed solutions with different molar ratio

Fig. S5 Macroscopic photographs of different molar ratios of C18-AP-2Na to CTAB at 25 °C.

5. The Cole-Cole plots of C18-AP-2Na / CTAB mixed system.

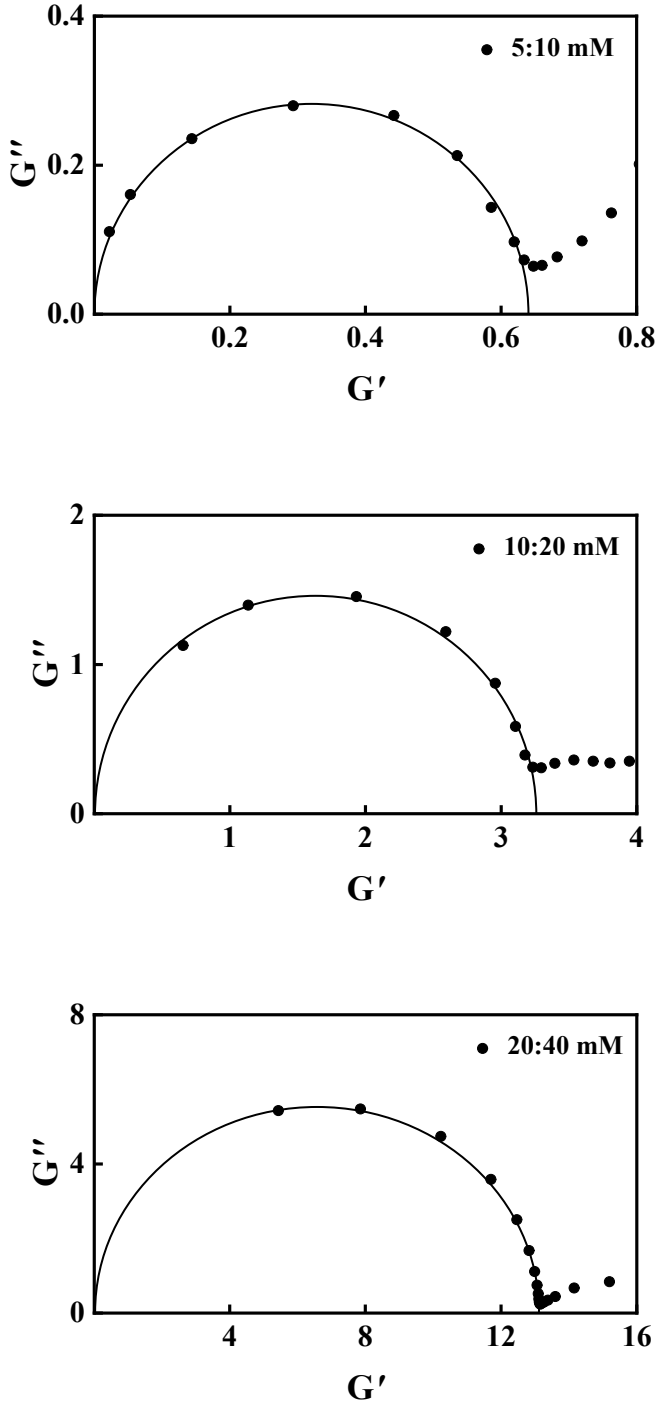


Fig. S6 The Cole-Cole plots of a 1:2 molar ratio solution of C18-AP-2Na / CTAB at 25 °C, with the concentration indicated for C18-AP-2Na.