

**Wormlike micellar solutions formed by an anionic surfactant and a cationic surfactant  
with two head groups**

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**1. Synthesis of C18-DQA**

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

**(1) Synthesis of methyl stearate.** Octadecanoic acid (250.0 g, 0.88 mol) and anhydrous methanol (168.7 g, 5.27 mol) were added into a 1000 mL three-necked flask. The concentrated sulfuric acid was added as a catalyst. And the reaction was carried out at 85 °C for 24 h. After the reaction was stopped, the raw product was washed three times with deionized water. After drying, the purified methyl stearate was obtained by vacuum distillation. Yield: 72.6 %.

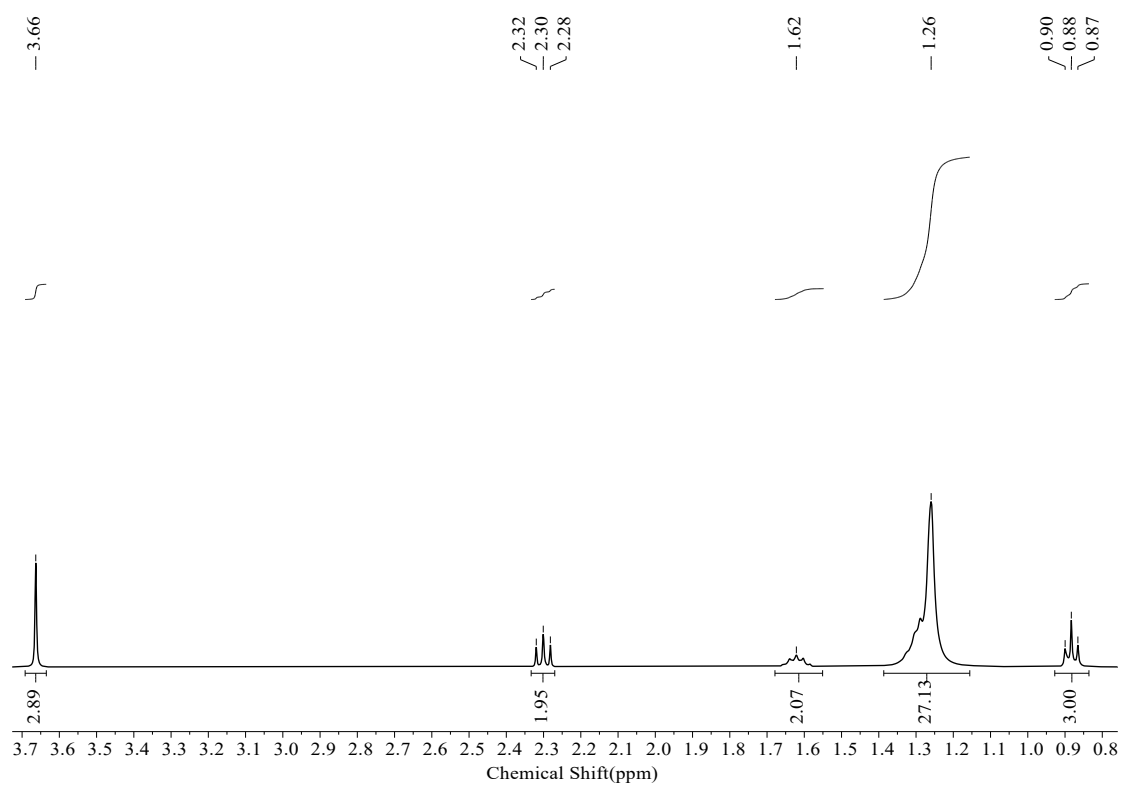
**(2) Synthesis of N-3-(dimethylamino)propyloctadecanamide.** The methyl stearate (189.3 g, 0.63 mol) and 3-dimethylaminopropylamine (194.3 g, 1.90 mol) were added to a 1000 mL single-necked flask. KOH was added as a catalyst. The reaction was carried out at 100°C for 48 h. The mixture was recrystallized three times in a mixed solvent of acetone and ethanol. N-3-(dimethylamino)propyloctadecanamide was obtained after drying under vacuum at 60 °C. Yield: 87.4 %.

**(3) Synthesis of C18-DQA.** N-3-(dimethylamino)propyloctadecanamide (89.0 g, 0.24 mol) and (3-bromopropyl)trimethylnitrogen bromide (56.9 g, 0.22 mol) were placed in a 500 mL single-necked flask. An appropriate amount of ethanol was added as a solvent, and the reaction was carried out at

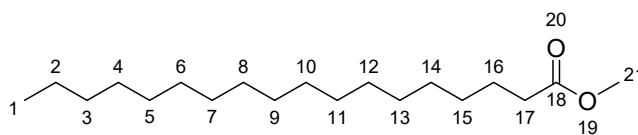
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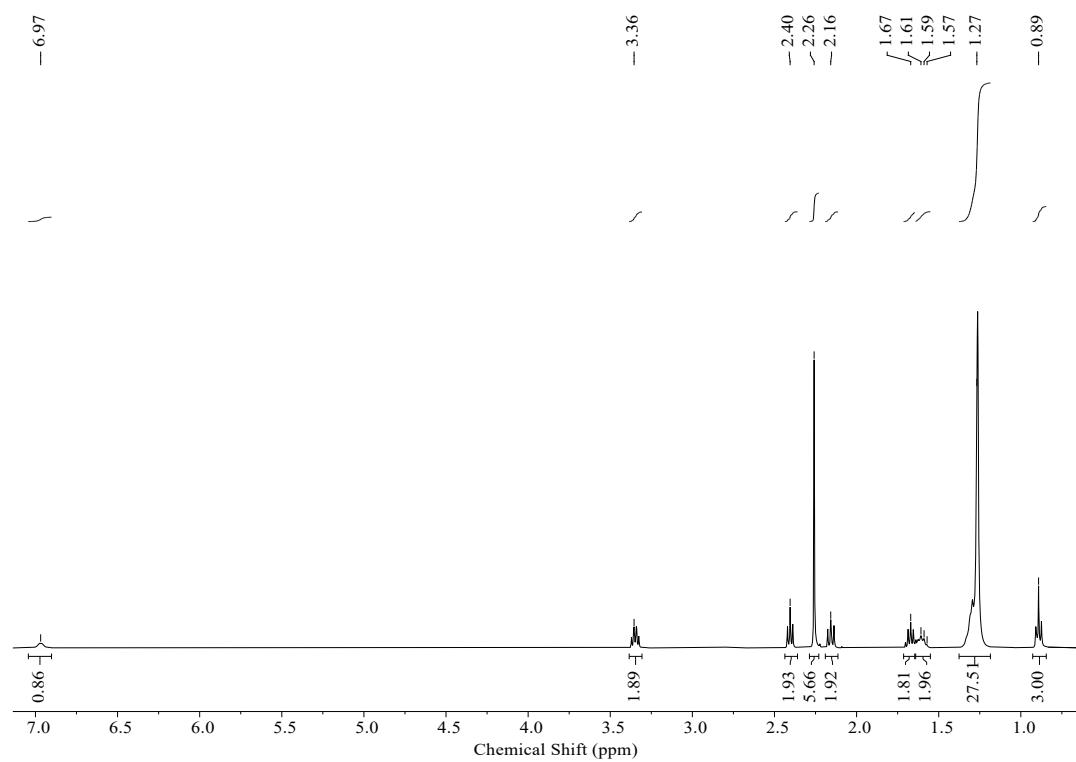
90 °C for 48 h. After the reaction, the solvent was removed under reduced pressure. The residue was washed three times with a mixed solvent of ethyl acetate and ethanol (ethyl acetate: ethanol = 50:1). The final product C18-DQA was obtained as a white power after dried under vacuum at 60 °C. Yield: 86.3 %.



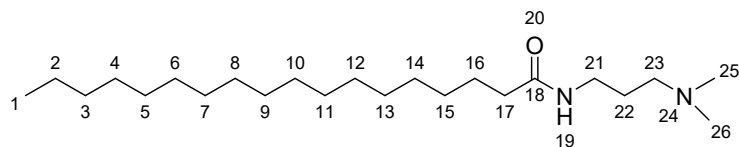
**Fig S1**  $^1\text{H}$  NMR spectrum of methyl stearate (25°C)



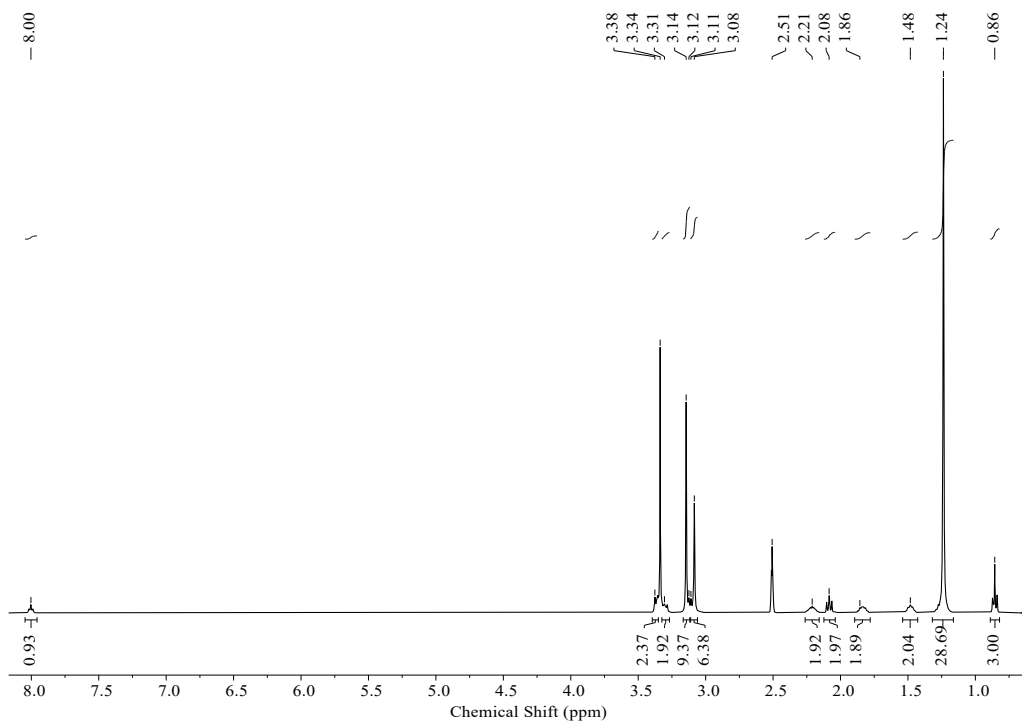
$^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$  3.66 (s, 3H), 2.30 (t, 2H), 1.62 (m, 2H), 1.26 (m, 28H), 0.88 (t, 3H).



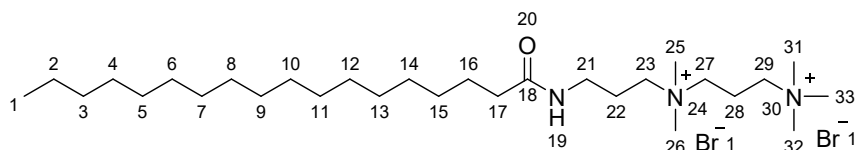
**Fig S2**  $^1\text{H}$  NMR spectrum of N-3-(dimethylamino)propyloctadecanamide (25°C)



$^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$  6.97 (s, 1H), 3.36 (m, 2H), 2.40 (t, 2H), 2.26 (m, 6H), 2.16 (t, 2H), 1.67 (m, 2H), 1.59 (m, 2H), 1.27 (m, 28H), 0.89 (t, 3H).

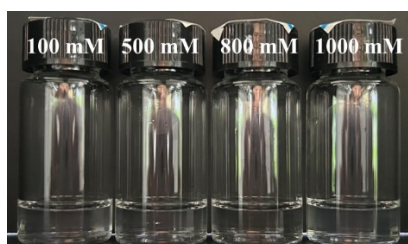


**Fig S3**  $^1\text{H}$  NMR spectrum of C18-DQA (25°C)



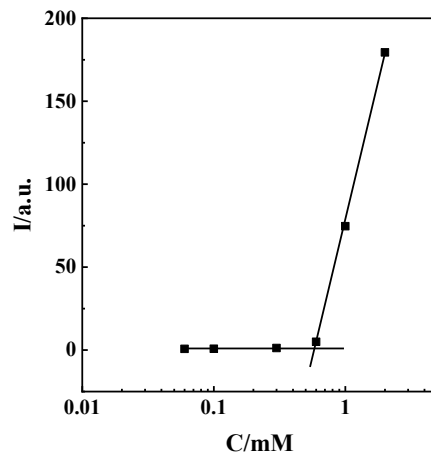
$^1\text{H}$  NMR (400 MHz, DMSO- $d_6$ )  $\delta$  8.00 (t, 1H), 3.39 – 3.31 (m, 8H), 3.14 (s, 9H), 3.08 (s, 6H), 2.21 (m, 2H), 2.08 (t, 2H), 1.86 (m, 2H), 1.48 (m, 2H), 1.24 (m, 28H), 0.86 (t, 3H).

## 2. Physical appearances of C18-DQA solutions with different concentrations



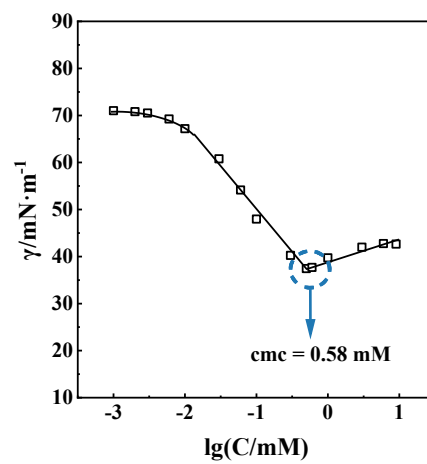
**Fig S4** Digital photos of aqueous solutions of C18-DQA with different concentrations

## 3. Fluorescence measurement



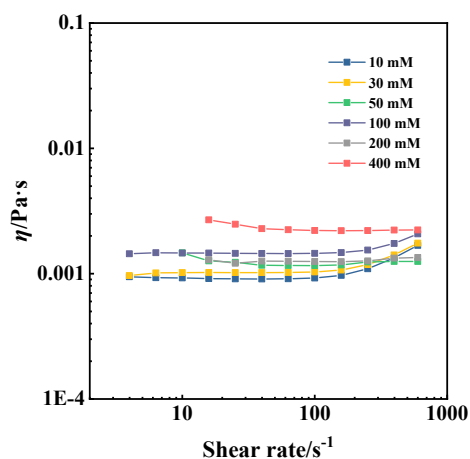
**Fig S5.** Variation of Nile red fluorescence intensity with concentrations of C18-DQA solutions.(25 °C)

#### 4. Surface tension measurement



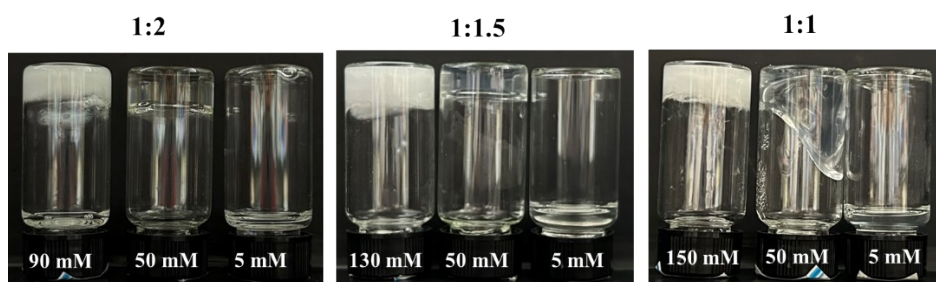
**Fig S6.** The equilibrium surface tension with concentrations of C18-DQA aqueous solutions. (25 °C)

#### 5. Steady rheology of C18-DQA solutions



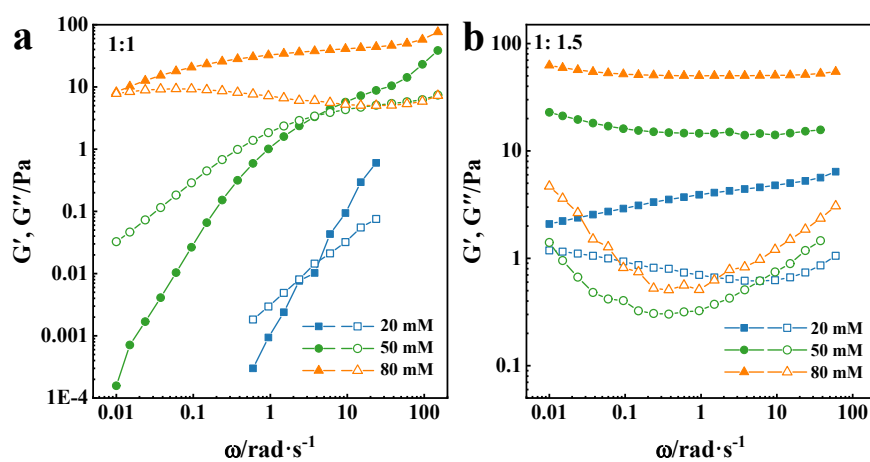
**Fig S7** Viscosity ( $\eta$ ) of C18-DQA solutions of different concentrations as a function of shear rate at 25 °C.

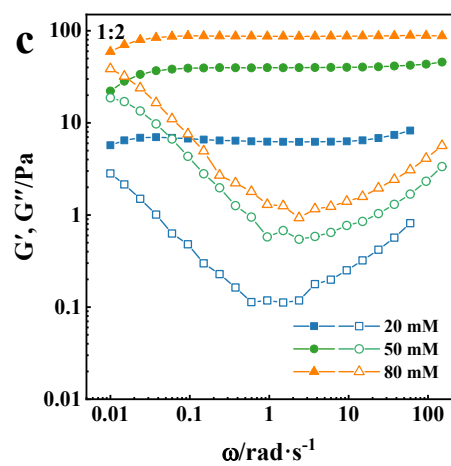
### 6. Physical appearance of C18-DQA and sodium laurate at different ratios



**Fig S8** Digital photos of mixed solutions of C18-DQA and sodium laurate at different ratios (denoted with the concentration of C18-DQA)

### 7. Oscillatory rheological measurement of C18-DQA/SL mixed solutions





**Fig S9** Storage modulus,  $G'$ , (black blocks) and loss modulus,  $G''$ , (white blocks) as a function of

the oscillatory frequency of C18-DQA/SL mixed solutions at molar ratio of (a) 1:1, (b)

1:1.5 and (c) 1:2 at 25 °C, the concentration of C18-DQA is shown in the figure.