Electronic Supplementary Information (ESI) for

Study on the mechanism of organic acid structure on the rheological behavior and aggregate transformation of pH-responsive wormlike micelles system

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Additional Results

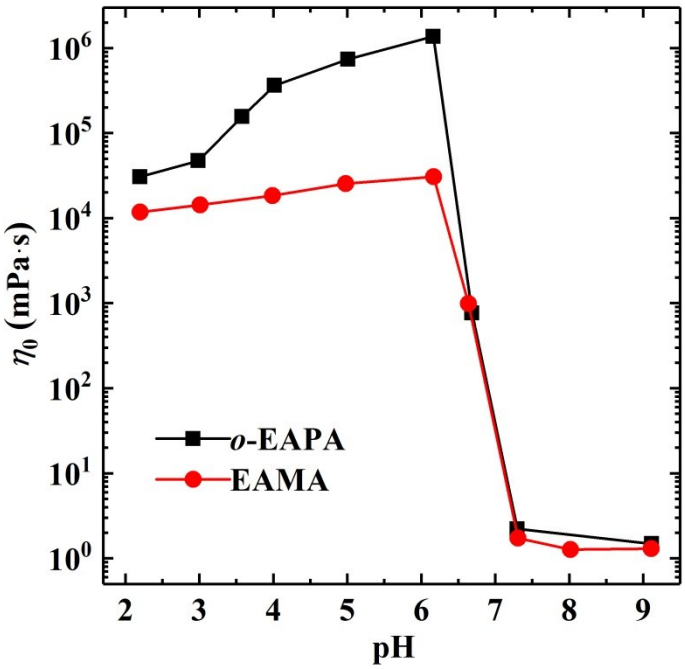


Fig.S1 The curve of zero-shear viscosity (η_0) at different pH values for 30 mM EAMA and 30 mM *o*-EAPA solutions at 25 ˚C

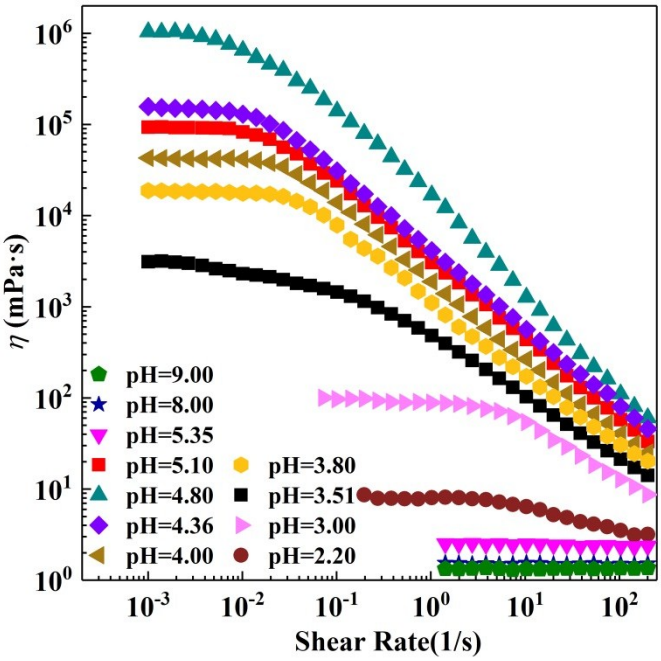
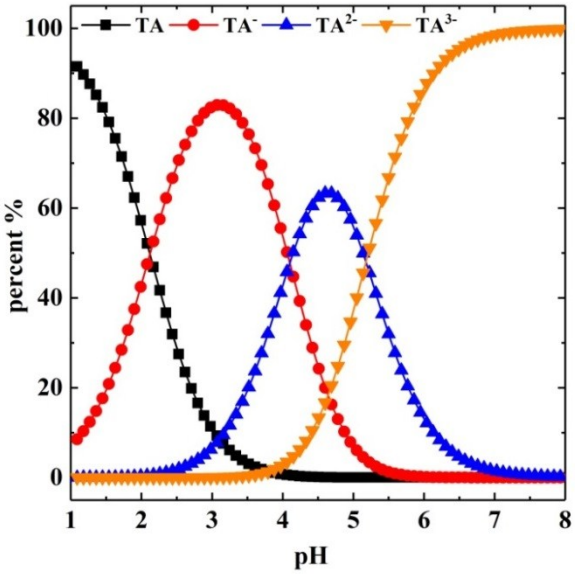


Fig.S2 Steady rheological curves of 30 mM EATA solution at different pH values and 25 ˚C



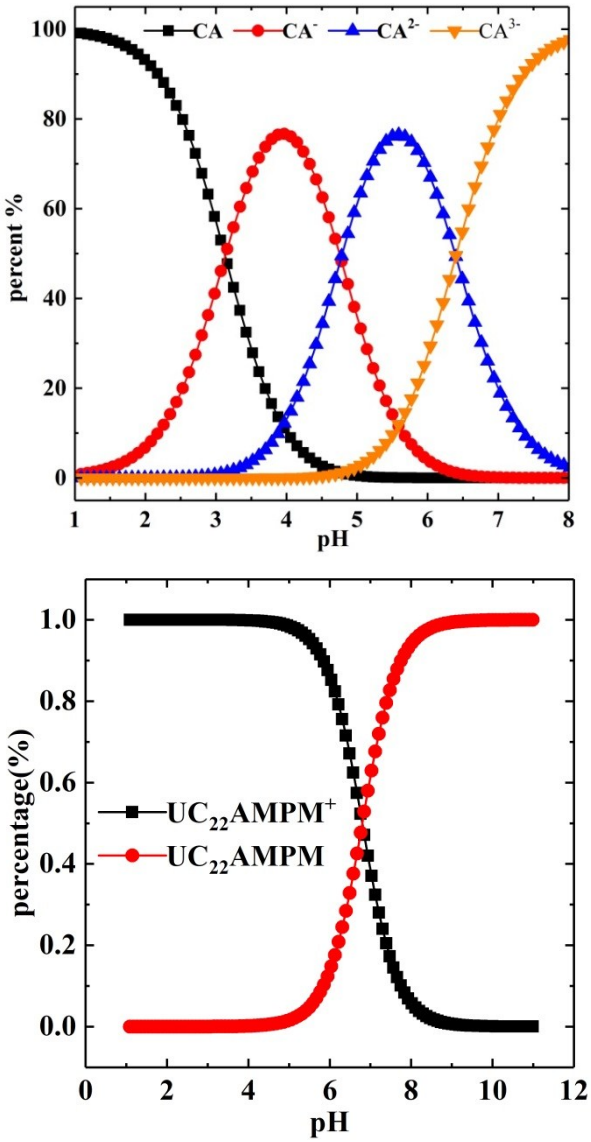


Fig.S3 Species distribution resulting from an aqueous solution of TA, CA and $UC_{22}AMPM$ at 25 ˚C

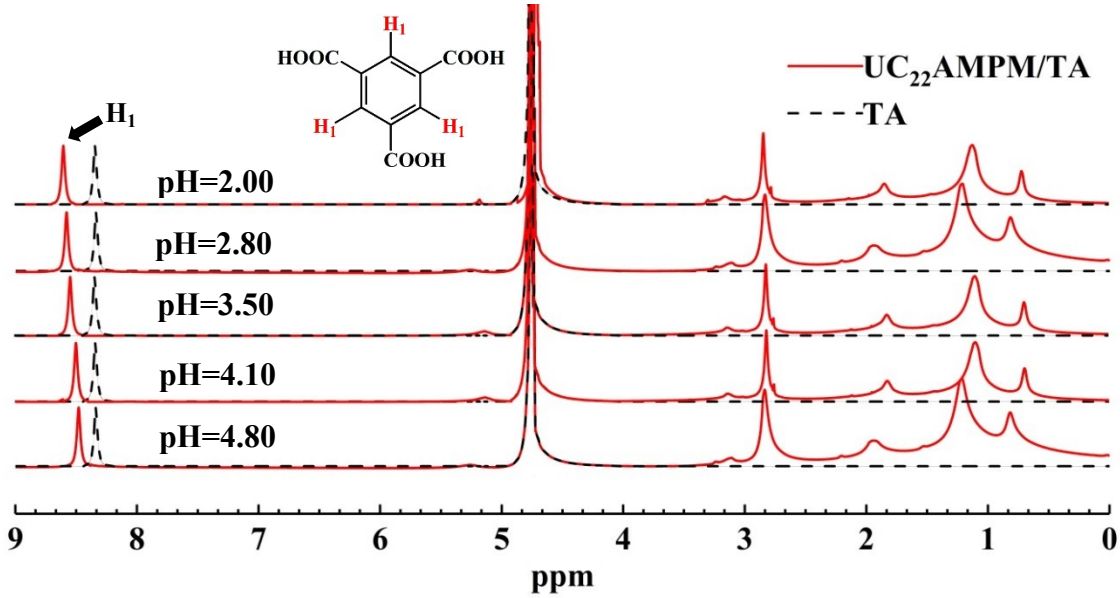


Fig.S4 Proton resonances for 30 mM $UC_{22}AMPM$/10 mM TA system at different pH values

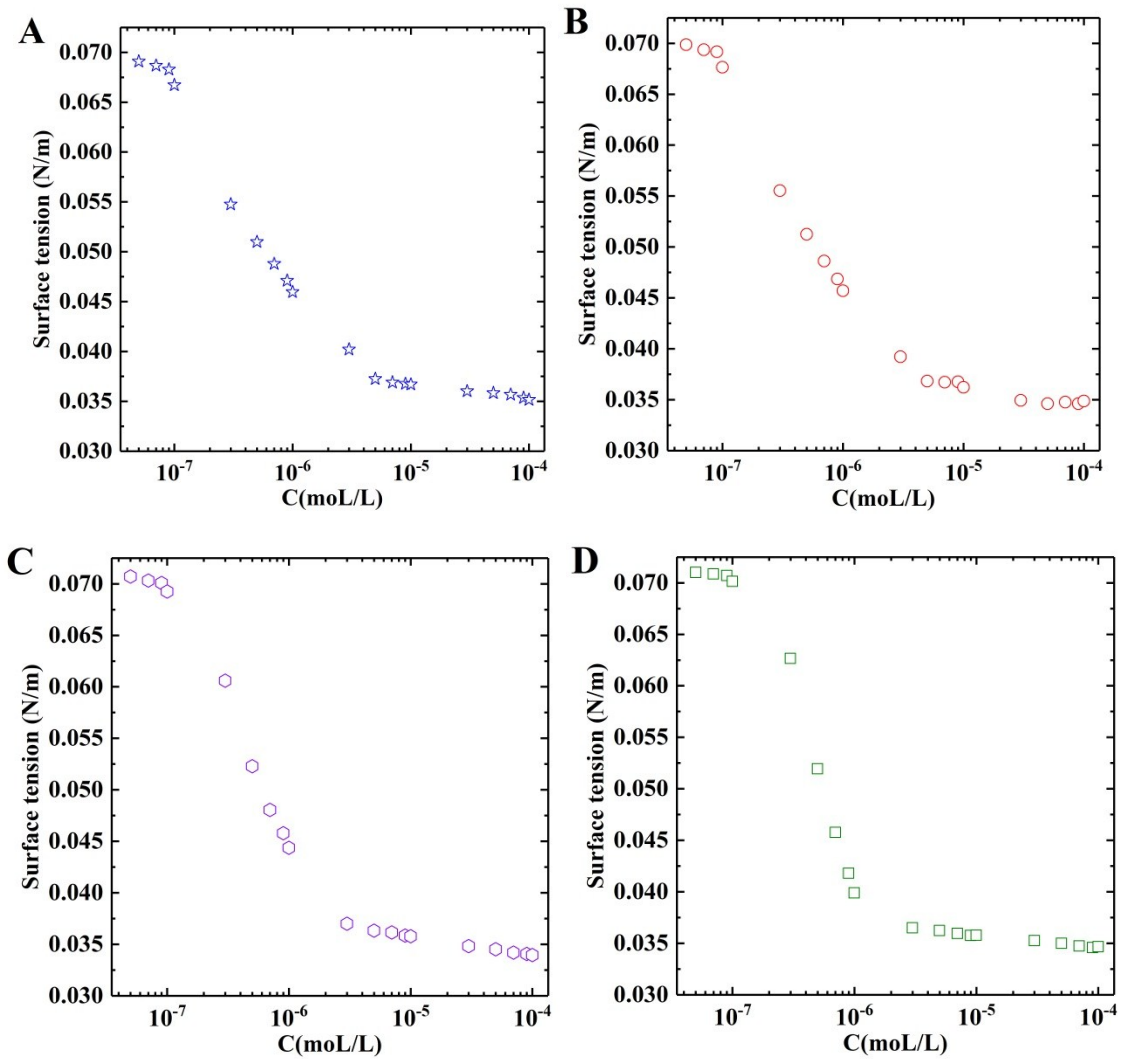


Fig.S5 Variation in surface tension with concentration of EATA at 25 ˚C (A: pH=2.20, B: pH=3.00, C: pH=3.80, D: pH=4.80)

The minimum average area per surfactant molecule was calculated by Gibbs adsorption equation:

$$\Gamma_{\max} = -\frac{1}{nRT}\left(\frac{\partial \gamma}{\partial \ln c}\right)_T \quad (1)$$

$$A_{\min} = \frac{1}{\Gamma_{\max} N_A} \quad (2)$$

Where, $(\partial\gamma/\partial \ln c)$ is the slope of the surface tension curve, R = 8.31 J / (mol·K), T = 298.15 K, N_A = 6.02 × 10^{23}, n is a constant which depends on the number of species constituting the surfactant and which are adsorbed at the interface.[49] The value of n can be determined by comparing the value of Γ, which respectively obtained from neutron reflectivity and Gibbs adsorption equation.[50-52] And n takes the value 2 for an ionic surfactant where the surfactant ion and the counterion are univalent, while n takes 3 for Gemini surfactants.[49, 51, 53] And the agreement between the two measurements is now seen to be excellent using

the normal prefactor n of 2 and 3 in the Gibbs equation.[54] According to the species distribution of TA, TA gradually transform into TA^- and TA^{2-} as increasing the pH, which means that the oligomeric surfactant in EATA solution will switch from one protonated $UC_{22}AMPM$ and TA^- to Gemini surfactant made up by two protonated $UC_{22}AMPM$ and TA^{2-}. Therefore, n takes 2 at pH 2.20 and 3.00, while takes 3 at pH 3.80 and 4.80 in this research.

The length l_c (cm) and volume v (cm^3) of hydrophobic chain of surfactants was obtained by characteristic parameters of surfactants:

$$l_c = (1.50 + 1.265n_c) \times 10^{-8} \tag{3}$$

$$v = (27.4 + 26.9n_c) \times 10^{-24} \tag{4}$$

Where n_c is the number of carbon atoms in hydrophobic chain of surfactants and takes 21 according to the structure of $UC_{22}AMPM$.

According to the A_{min}, l_c and v, the packing parameter p can be calculated by $p=v/al_c$. And the calculated results were listed in the Table.S1.

Table.S1 The Surface parameters of EATA system at different pH values and 25 ˚C

pH	l_c(nm)	V(nm^3)	A_{min} (nm^2)	p
2.20	2.8065	0.5923	0.708	0.298
3.00			0.651	0.324
3.80			0.563	0.375
4.80			0.450	0.469

The p of EACA system was calculated by using the same method, and the data of A_{min} was provided by Wang (Wang *et al.* "A pH-responsive wormlike micellar system of a noncovalent interaction-based surfactant with a tunable molecular structure", *Soft Matter*, 2017, 13, 1182-1189). The calculated results were listed in the Table.S2.

Table.S2 The Surface parameters of EACA system at different pH values and 25 ˚C

pH	l_c(nm)	V(nm^3)	A_{min} (nm^2)	p
2.20	2.8065	0.5923	0.611	0.345
3.00			0.580	0.364
4.00			0.560	0.377
5.00			0.481	0.439
6.17			0.449	0.470