

Enhancing Surface/Interface Activity and Wettability *via* Trimeric Surfactant-Containing Mixtures

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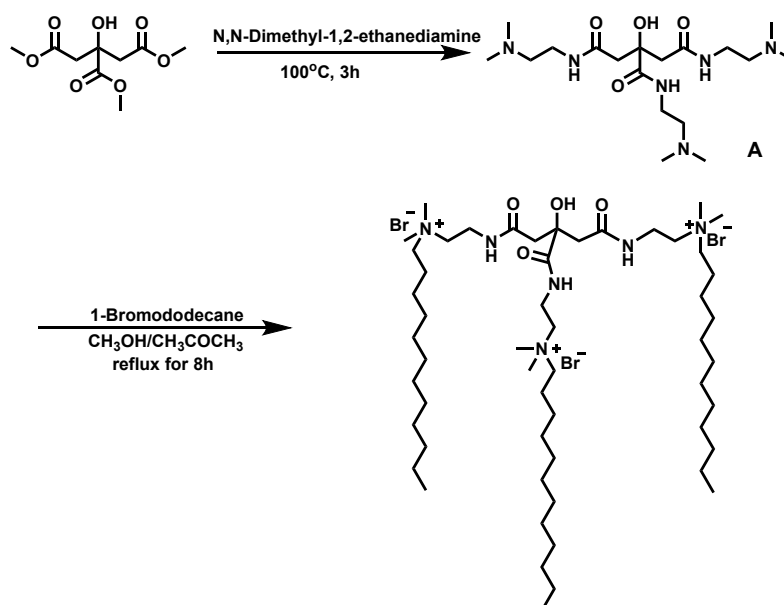


Figure S1. Synthetic procedure of **Citric-3C12**.

Material.

N,N-Dimethyl-1,2-ethanediamine, trimethyl citrate and 1-bromododecane were obtained from Innochem and used without further purification. CH₃OH, CH₃COCH₃, and all organic solvents were purchased from Beijing Chemical Co. Milli-Q (18.2 MΩ MΩ·cm) water was used in all experiments.

Synthesis.

Compound **Citric-3C12** was synthesized as outlined in **Figure S1** and was characterized by ¹H NMR, mass spectrum. 2.34 g trimethyl citrate was dispersed in 20 mL *N,N*-Dimethyl-1,2-ethanediamine and stirred at 100°C for 4 h. The *N,N*-

Dimethyl-1,2-ethanediamine was then removed under reduced pressure, and the residue was recrystallized three times from methanol/ethyl acetate to afford intermediate **A**.

Intermediate **A** (4 g) and 1-bromododecane (8.9 g) were added to 100 mL of CH₃OH/CH₃COCH₃ (v: v = 1:1) and heated at 45 °C for 72 h. The solvent was removed under reduced pressure, and the residue was recrystallized three times from methanol/ethyl acetate, yielding **Citric-3C12** as white solid in 41% yield. ¹H NMR (CD₃OD, 400 MHz): δ = 0.88 (m, 9H), 1.12–1.39 (m, 46H), 1.80 (s, 8H), 2.52-2.64 (m, 4H), 2.71–2.82(m, 4H), 3.00–3.06 (m, 5H), 3.06–3.17 (m, 12H), 3.17–3.29 (m, 4H), 3.28–3.50 (m, 12H), 3.51–3.69 (m, 8H). MS-ESI (m/z): calculated: 1146.64; found: 495.40 ([M-2Br]²⁺/2). FTIR (KBr, cm⁻¹): 3225, 2907, 2827, 1648, 1458, 1349, 1228,1073, 1015, 969, 906, 739.

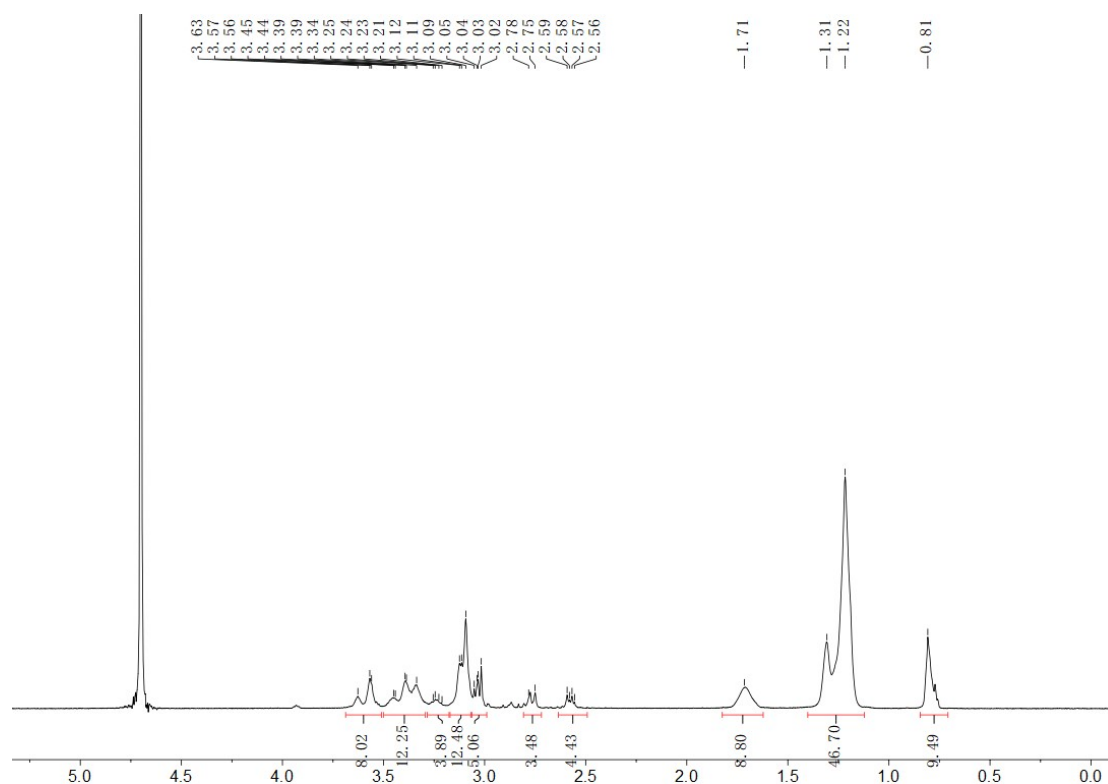


Figure S2. ¹H NMR spectra of **Citric-3C12** in D₂O.

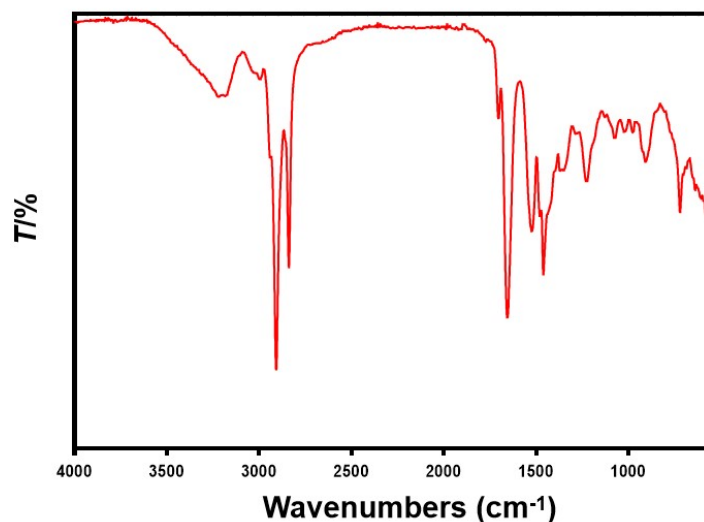


Figure S3. FT-IR spectra of **Citric-3C12**.

Table S1. Values of the CMC, interaction parameters (β^σ , β^m), interfacial composition (X^σ), micellar composition (X^m) Surface Excess (Γ_{\max}), and Area Per Molecule (A_{\min}) for the **Citric-3C12/9-EOS** mixture in aqueous solution at 25.0 °C

$X_{\text{Citric-3C12}}$	CMC (mM)	pC20	χ^m	β^m	χ^σ	β^σ	$\Gamma \times 10^{-6}$ (mol m ⁻²)	A (Å ² /mole cule)	A_{ideal} (Å ² /mole cule)
1	0.25	1.1	-	-	-	-	1.699	97.763	-
0.9	0.052	1.9	0.491	-3.721	0.42	-4.012	1.835	90.526	97.847
0.8	0.038	2.2	0.419	-3.665	0.365	-4.431	2.015	82.418	97.855
0.7	0.022	2.4	0.398	-5.148	0.383	-7.605	2.303	72.104	97.889
0.6	0.018	2.5	0.38	-5.656	0.362	-7.678	2.031	81.778	97.855
0.5	0.00455	2.6	0.334	-8.123	0.323	-6.773	1.855	89.543	97.861
0.4	0.018	2.8	0.309	-4.969	0.404	-11.050	1.683	98.679	97.849
0.3	0.018	3.1	0.279	-4.922	0.334	-9.764	1.208	137.563	97.859
0.2	0.017	2.8	0.256	-5.302	0.292	-8.746	1.713	96.9632	97.866
0.1	0.012	2.8	0.275	-7.702	0.277	-9.694	1.574	105.547	97.868
0	0.022	2.4	-	-	-	-	1.697	97.914	-

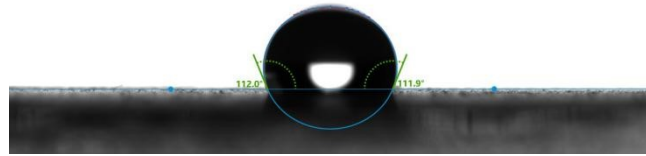


Figure S4. Contact angle photographs of water on oil-wet silica gel surfaces

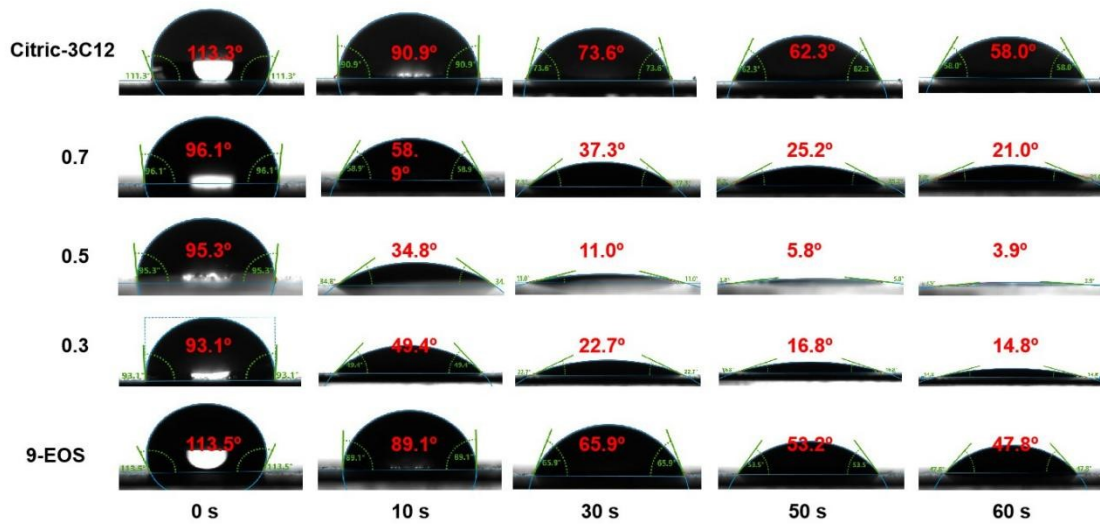


Figure S5. Contact angle photographs of oil-wet silica gel surfaces treated with **Citric-3C12/9-EOS** at $X_{\text{Citric-3C12}} = 1.0, 0.7, 0.5, 0.3$ and 0 .