Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Manuscript title: pH-Induced reversible conversion between Non-Pickering and Pickering high internal phase emulsion

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The number of pages: 7

The number of figures:10

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1. The cmc of dodecylammonium.

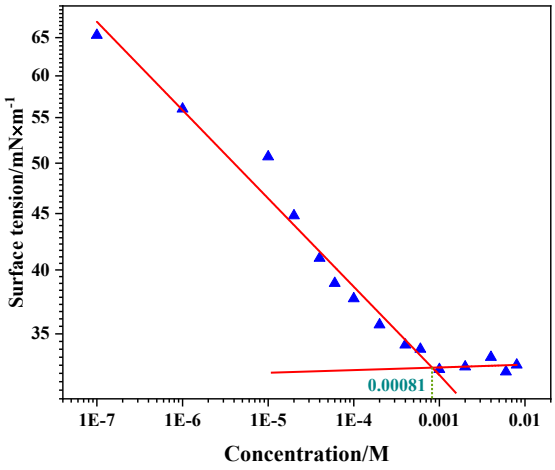

Fig.S1 The surface tension value changes with the concentration of dodecylammonium

2. TEM of SiO_2 nanoparticles.

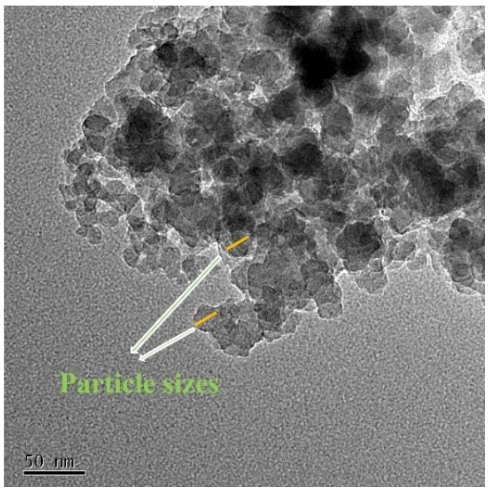

Fig.S2 TEM of nano-SiO_2 particles.

3. The purity of FA-OA.

The purity of the FA-OA was calculated using the following equation (eqn(1)):

$$purity(\%) = \frac{A_4}{A_1 + A_4} \times 100\% \quad \text{(eqn 1)}$$

Where A_1 is the area corresponding to peak '1', A_4 is the area corresponding to peak '4'.

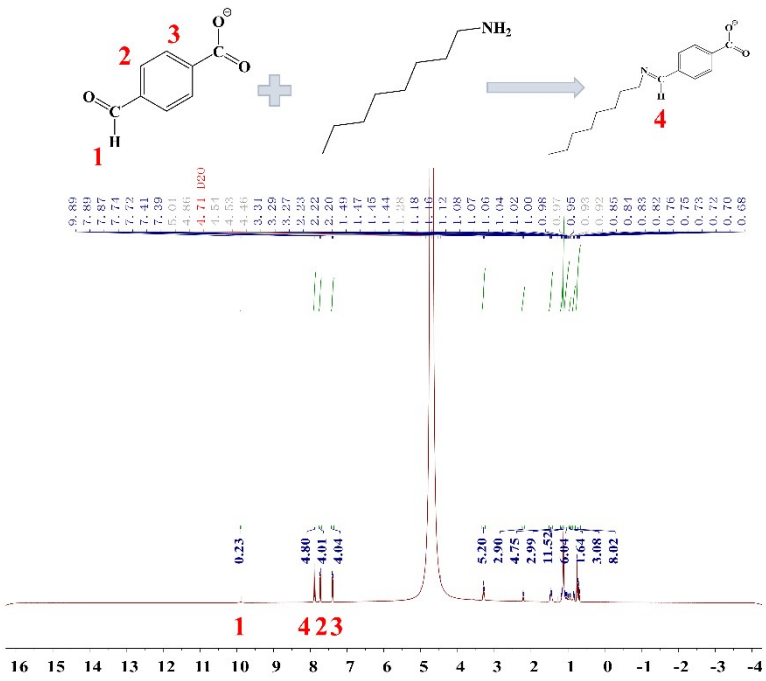

Fig.S3 ^{1}H NMR of FA-OA.

The purity was calculated using the eqn (1). The final purity is 95.43%.

4. FTIR spectra of reactants and FA-OA.

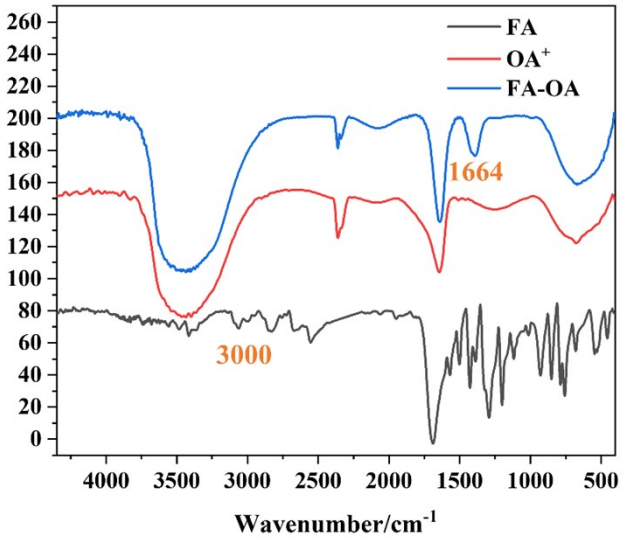

Fig.S4 FTIR spectra of reactants and FA-OA.

As shown in Fig.S4, when comparing the spectra of FA-OA and OA, the -NH_2 stretching vibration doublet at about 3000 cm^{-1} disappeared. The 1664cm^{-1} in the FA-OA spectrum is a stretching vibration peak of -C=N. This shows that FA-OA was successfully synthesized.

5. The cmc of FA-OA.

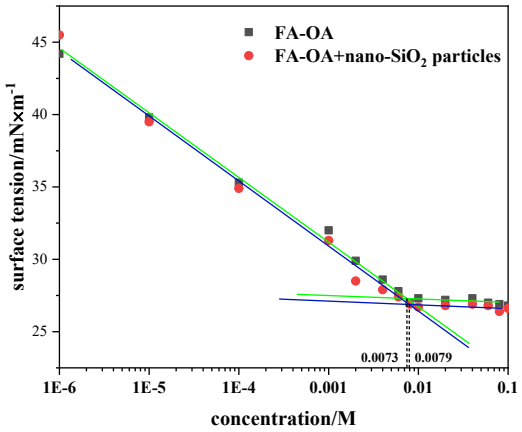

Fig.S5 The surface tension value changes with the concentration of FA-OA.

6. Optical photographs and micrographs of HIPNPEs.

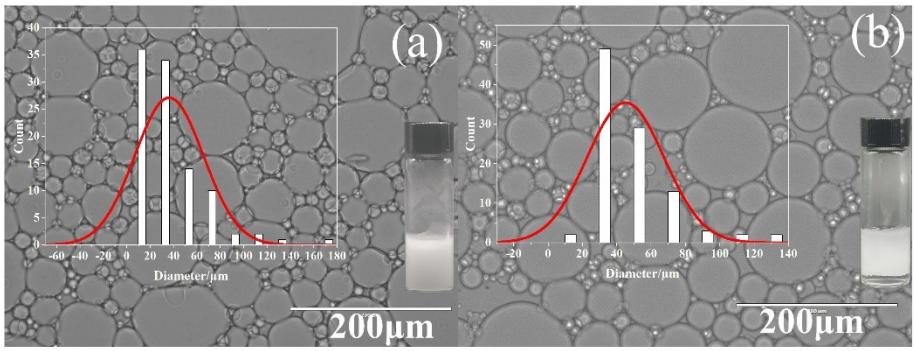

Fig.S6 Optical photographs, micrographs and particle size distribution of HIPNPE just after being prepared (a) and after being placed for30 days (b). The mass ratio of oil-water in the emulsion system was 3: 1, the concentrations of FA-OA and dodecylamine are 1 cmc and 3 cmc, respectively, the concentration of nano-SiO_2 particle was 0.3 wt%. All Optical photographs are magnified 400x.

The micrographs of the emulsions with 1 cmc surfactant and 0.3wt% nanoparticles are shown in Figure S6 after just being prepared and being placed for 30 days. The particle size of the emulsions increased from 41.68 μm to 45.14 μm. It can be seen that the high internal phase Non-Pickering has good stability.

7. SEM image of the dried emulsion.

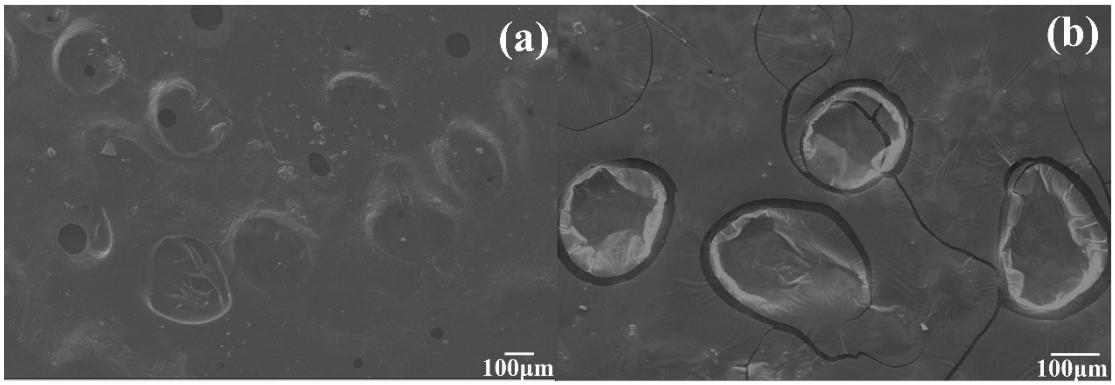

Fig.S7 SEM image of the dried emulsion. (a)HIPNE. (b) after adjusting pH with HCl aqueous solution. Both the micrographs show the emulsion at 4000x magnification.

So as to further study and determine the type of emulsion before and after pH adjustment, we processed the two kinds of emulsions to observe their microstructure. After the treatment, the original dispersed phase of the Non-Pickering emulsion retains its original shape, showing a round shape (Figure S7 (a)). However, because of the particles squeezing the oil-water interface, the Pickering emulsion will cause the interface to wrinkle inward during the drying process (as shown in Figure S7 (b)). This confirmed the transition from HIPNPEs to HIPEs after stimulus-response. However, Since the dispersed phase will flow during the drying process, it will affect the particle size of the emulsion.

8. Effect of unreacted FA and OA on interfacial tension.

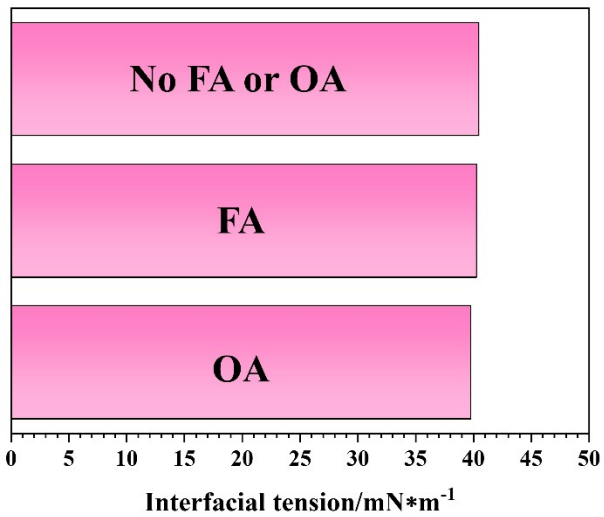

Fig.S8 Interfacial tension between octane and water. From top to bottom: octane

and water ((pH=12.22), $0.33*10^{-3}$ M FA (pH=12.22) and $0.33*10^{-3}$ M OA (pH=12.22).

According to the purity of FA-OA, the molar concentrations of residual FA and OA were calculated. According to the residual molality, oil-water systems containing only FA or OA were prepared respectively, and their interfacial tension was measured, as shown in Figure S8. It can be found that the residual small amount of unreacted FA and OA has little effect on the lowering of oil-water interfacial tension.

9. Optical photographs, micrographs, particle size distribution and pH-responsiveness of emulsion without dodecylamine.

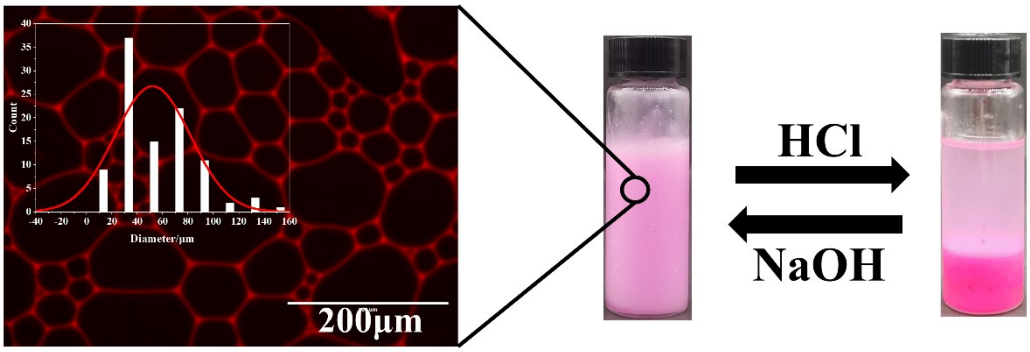

Fig.S9 Optical photographs, micrographs, particle size distribution and pH-responsiveness of emulsion without dodecylamine. The mass ratio of oil-water in the emulsion was 3: 1, the concentration of FA-OA was 1 cmc, and the concentration of nano-SiO_2 particle was 0.3 wt%. The dye is Rhodamine B. Optical photographs show the emulsion at 400x magnification.

10. Surface tension of dodecylammonium aqueous solution.

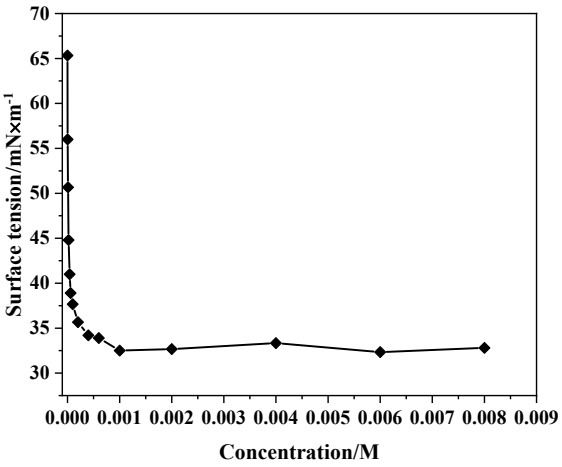

Fig.S10 Calibration curve used to determine the adsorption isotherm of dodecylammonium on SiO_2 nanoparticles, relating the concentration of dodecylammonium to the surface tension of the aqueous solution.