Janus-like Pickering Emulsions and Their Controllable Destabilization

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Experimental section

Materials. Dopamine hydrochloride, hexadecane, NaOH, NaCl and HCl were purchased from Sigma-Aldrich and used without purification. Milli-Q water with resistance of 18.2 M Ω cm⁻¹ was used as solvent.

Preparation of emulsifier PDA particles. PDA particles were synthesized by selfpolymerization of dopamine in basic solutions according the literature.¹ In Brief, 5 mg of dopamine was dissolved in 10 ml of NaOH (0.06 M) solution. The solution was kept shaking for 18 h. The colour of the solution turned from colourless into black. The PDA particles were collected by centrifugation, and washed by water for several times. The obtained particles were re-dispersed in 1 ml of water.

Preparation of Janus-like Pickering emulsions. 1 ml of above PDA suspension with different pH values (pH value was adjusted via NaOH and HCl solutions) was mixed with 1 ml of hexadecane. The mixture was then shaken by vortex for 30 s, shaking frequency is 2400/min.

Drop test. The obtained Janus-like emulsion droplets was placed into the water and hexadencane separately. The emulsion droplets could disperse in water, but sediment in hexadencane, demonstating that the Janus-like emulsions were of oil-in-water type.

Coalescence of Janus-like Pickering emulsions.

pH induced coalsescence of Janus-like emulsion droplets: The as-pareapred Janus-like emulsion (pH 4.5) was extracted and transferred to 5 mL of H_2O with pH value of 3.2 and 2.0.

Salt induced coalescence of Janus-like emulsion droplets: The as-parepared Janus-like emulsion (pH 4.5) was extracted and transferred to 5 mL of NaCl solution with concentration of 50 mM, the pH value of the NaCl solution is 4.5.

Characterization. UV-vis adsorption spectra were recorded with a Shimadzu UV-2600 UV-vis spectrophotometer. Zeta potential of the particles and oil droplets were measured on

Malvern zetasizer nano ZS. Transmission electron microscopy (TEM) imaging was implemented with FEI Tecnai G2 Sprit TEM at an acceleration voltage of 100 kV. Optical microscopy images were recorded on Nikon Eclipse Ti-U fluorescent microscope.



Figure S1. (a) illustration of self-polymerization of dopamine. (b) TEM image of PDA particles obtained via self-polymerization of dopamine in NaOH solution. (c) Histogram of the particle size distribution reveals an average particle diameter of 192±7.5 nm.



Figure S2. (a) UV-vis spectra of water phase before and after emulsification, and (b) plot of PDA remaining in water phase after emulsification as a function of pH. C is the concentration of PDA particles remaining in water phase after emulsification. C_0 is the initial concentration of PDA particles in water phase.



Figure S3. Optical microscopy images of the emulsion prepared with PDA particles at pH of (a) 5.1, (b) 8.4. Scale bar, 100 μ m.



Figure S4. Digital photographic illustration of the coalescence of Janus-like emulsion droplets via lowering the pH of the solution from 4.5 to 2.0.



Figure S5. Photographic illustration of coalescence of Janus-like emulsions via increasing the NaCl concentration to 50 mM.

Reference:

1. H. L. Xu, X. K. Liu, G. Su, B. Zhang and D. Y. Wang, *Langmuir*, 2012, **28**, 13060