Electronic Supplementary Information

Dual functional Janus membrane combined superwettability with electrostatic force for controllable anionic/cationic emulsion separation and *in-situ* surfactant removal

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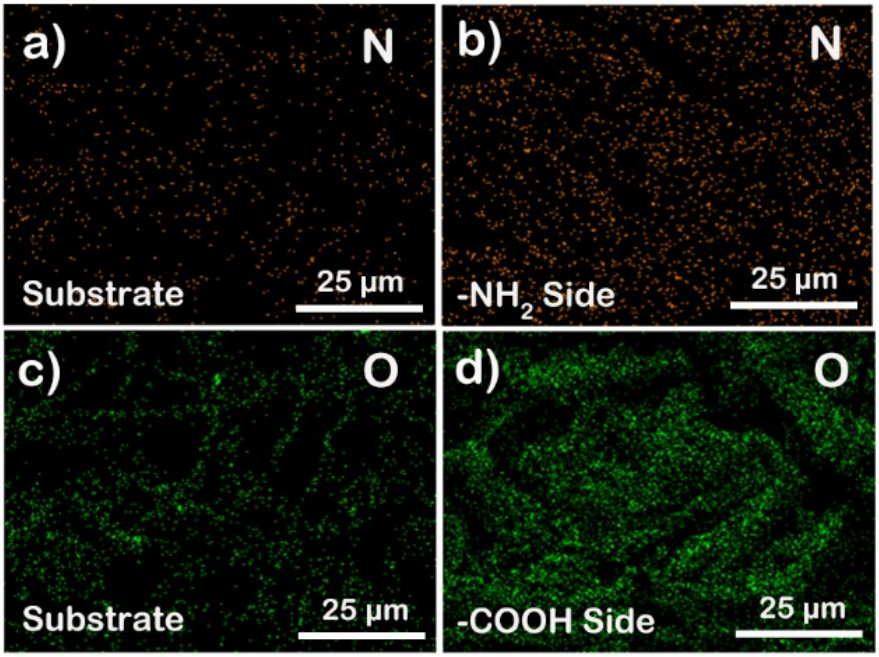

Fig. S1 a) and b) N element EDX mapping images of the substrate and $–NH_2$ side surface, respectively. c) and d) O element EDX mapping images of the substrate and –COOH side surface, respectively.

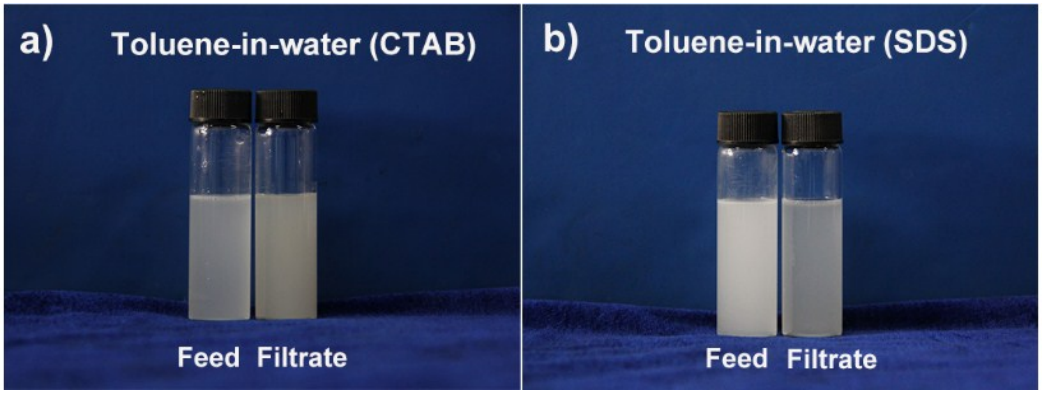

Fig. S2 a) and b) SDS toluene in water and CTAB toluene in water emulsion separation performance of substrate.

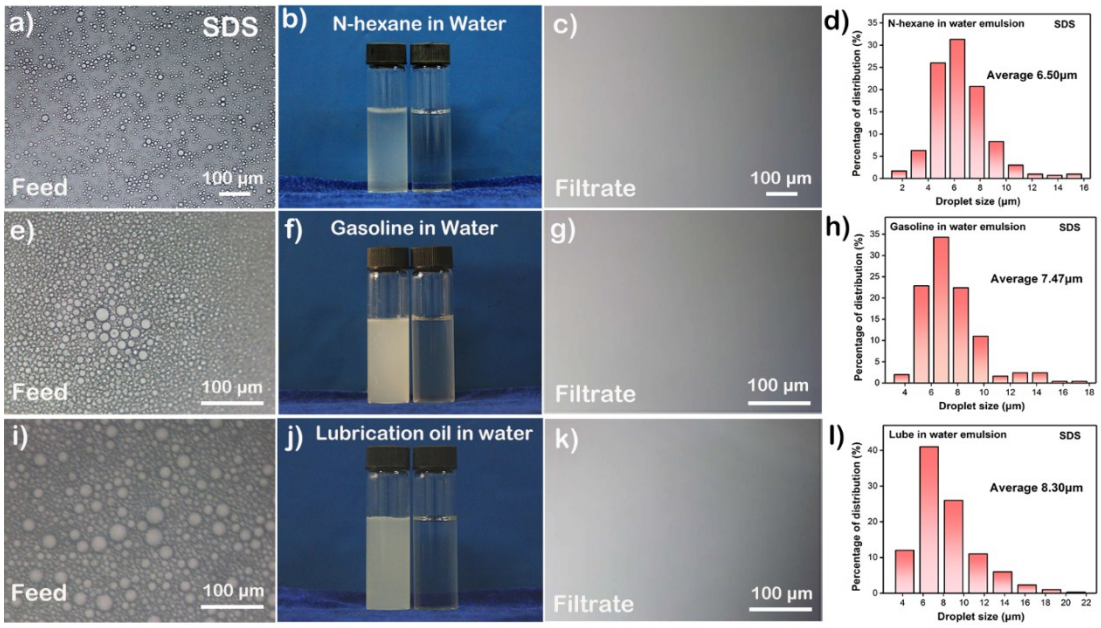

Fig. S3 SDS oil-in-water emulsion separation performance of $-NH_2$ side and the oil droplet distribution.

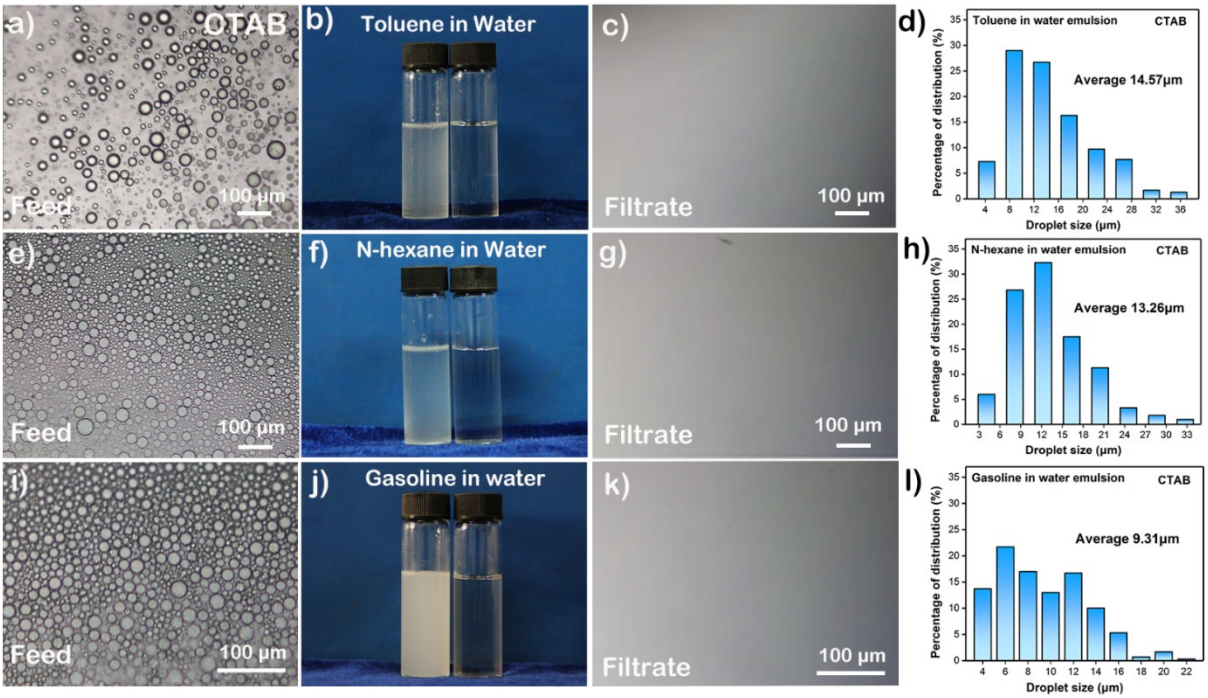

Fig. S4. CTAB oil-in-water emulsion separation performance of –COOH side and the oil droplet distribution.

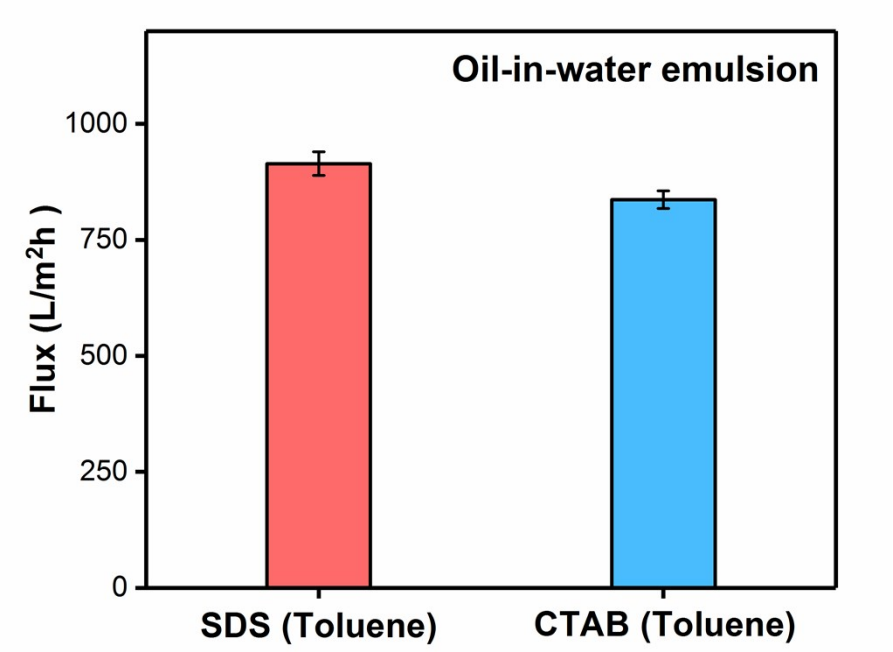

Fig. S5 The flux of anionic toluene in water emulsion with SDS surfactant and cationic toluene in water emulsion with CTAB surfactant.

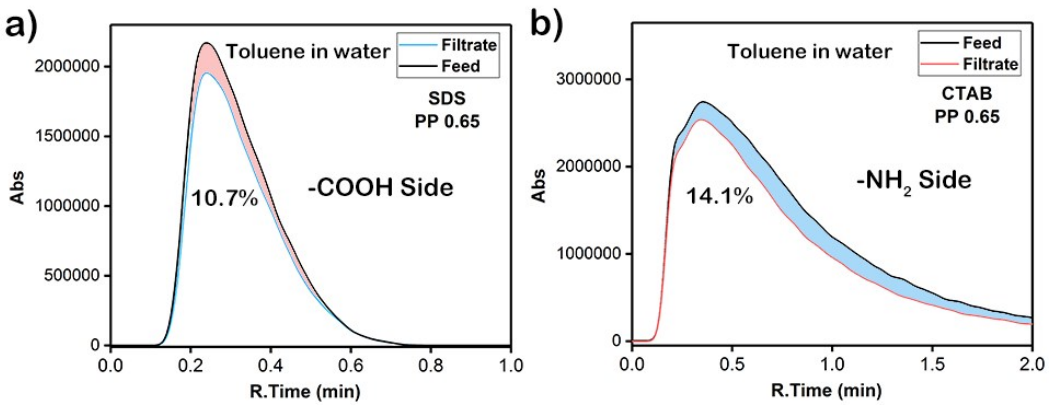

Fig. S6 a) Anionic SDS surfactant removal performance of –COOH side. b) Cationic CTAB surfactant removal of $–NH_2$ side.

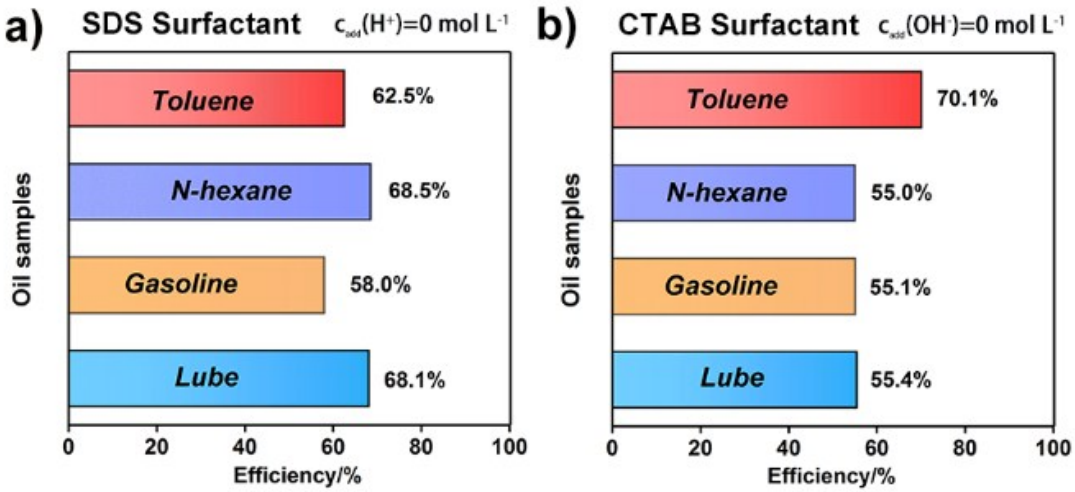

Fig. S7 a) *In-situ* SDS surfactant removal performance and b) *In-situ* CTAB surfactant removal performance without the adding of H^+ or OH^-.

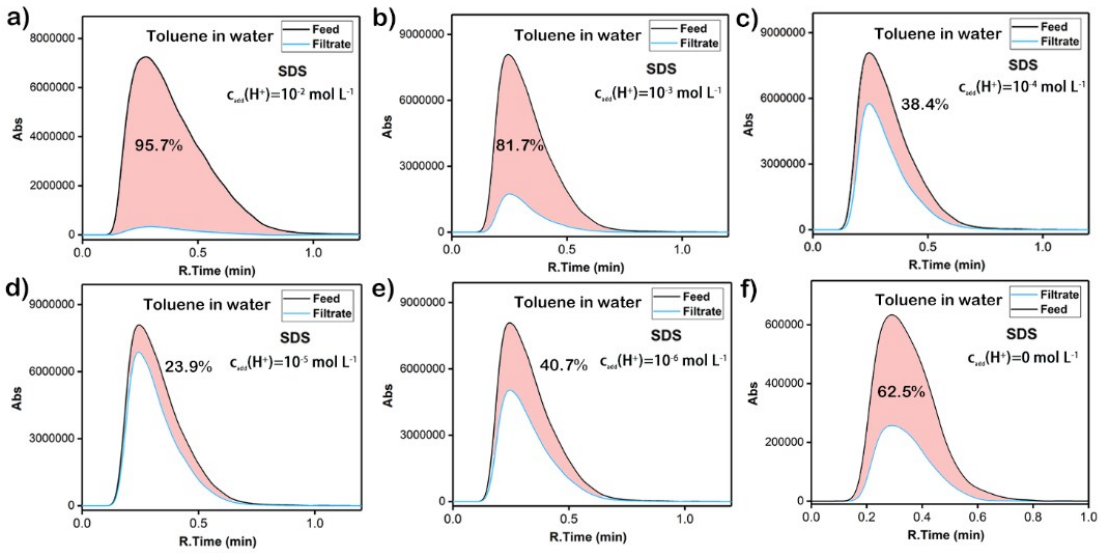

Fig. S8 The influence of the adding of H^+ on the SDS surfactant removal performance. a)-f) The EIC images of the emulsion and filtrates with the $C_{add}(H^+)$ at 10^{-2} mol L^{-1}, 10^{-3} mol L^{-1}, 10^{-4} mol L^{-1}, 10^{-5} mol L^{-1}, 10^{-6} mol L^{-1}, and 0 mol L^{-1}, respectively.

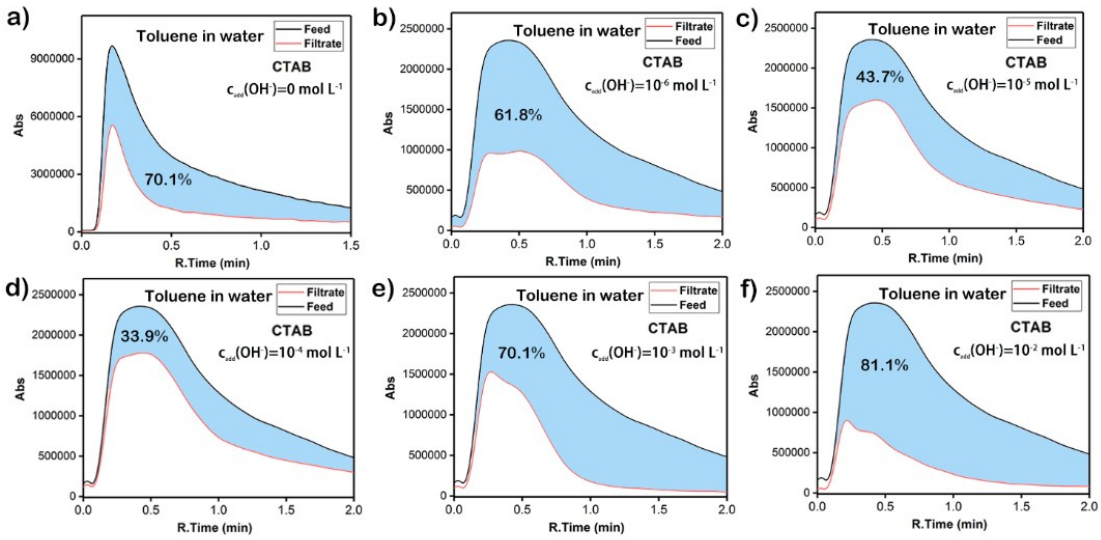

Fig. S9 The influence of the adding of OH^- on the CTAB surfactant removal performance. a)-f) The EIC images of the emulsion and filtrates with the $C_{add}(OH^-)$ at 0 mol L^{-1}, 10^{-6} mol L^{-1}, 10^{-5} mol L^{-1}, 10^{-4} mol L^{-1}, 10^{-3} mol L^{-1} and 10^{-2} mol L^{-1}, respectively.

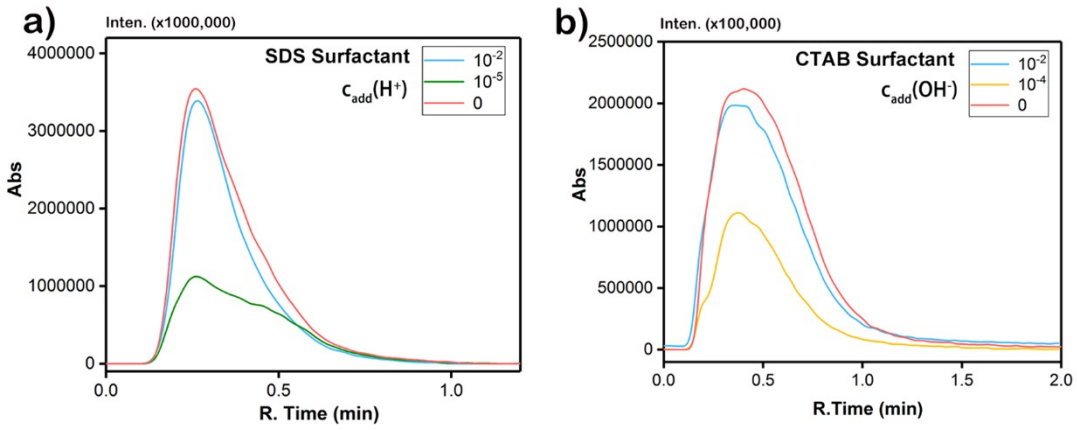

Fig. S10 a) The EIC spectra of the SDS solution with different concentrations of H^+. b) The EIC spectra of the CTAB solution with different concentrations of OH^-.

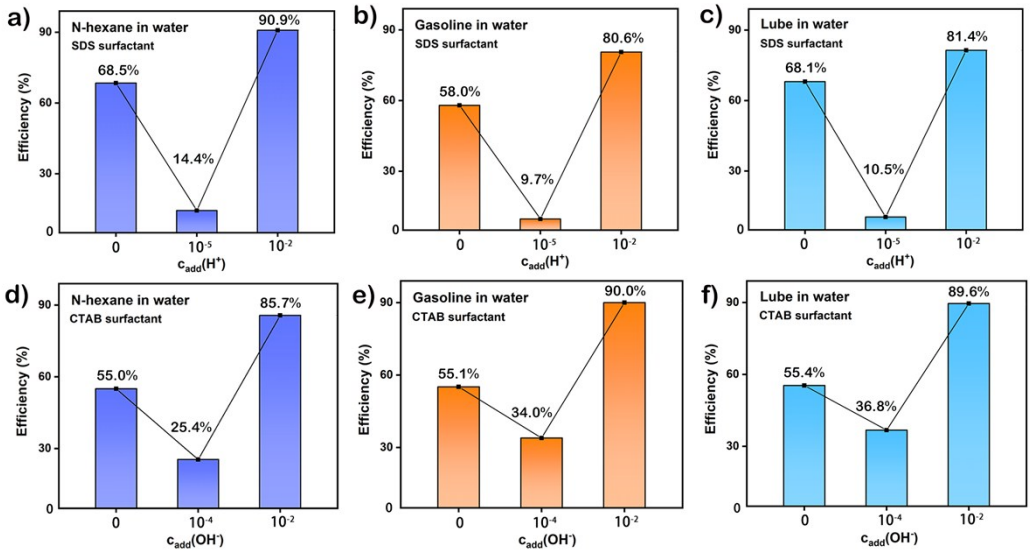

Fig. S11 a)-c) *In-situ* SDS surfactant removal performance of n-hexane in water emulsion, gasoline in water emulsion and lubrication oil in water emulsion, respectively (with the $C_{add}(H^+)$ at 0 mol L^{-1}, 10^{-5} mol L^{-1} and 10^{-2} mol L^{-1}). d)-f) *In-situ* CTAB surfactant removal performance of n-hexane in water emulsion, gasoline in water emulsion and lubrication oil in water emulsion, respectively (with the $C_{add}(OH^-)$ at 0 mol L^{-1}, 10^{-4} mol L^{-1} and 10^{-2} mol L^{-1}). It is clear that with the concentration of H^+ or OH^- increasing, the removal efficiency decreases at first and then rises to a relatively high piont (above 80%) for all kinds of emulsions used in this work. This trend is in concordance with that of toluene in water emulsion.

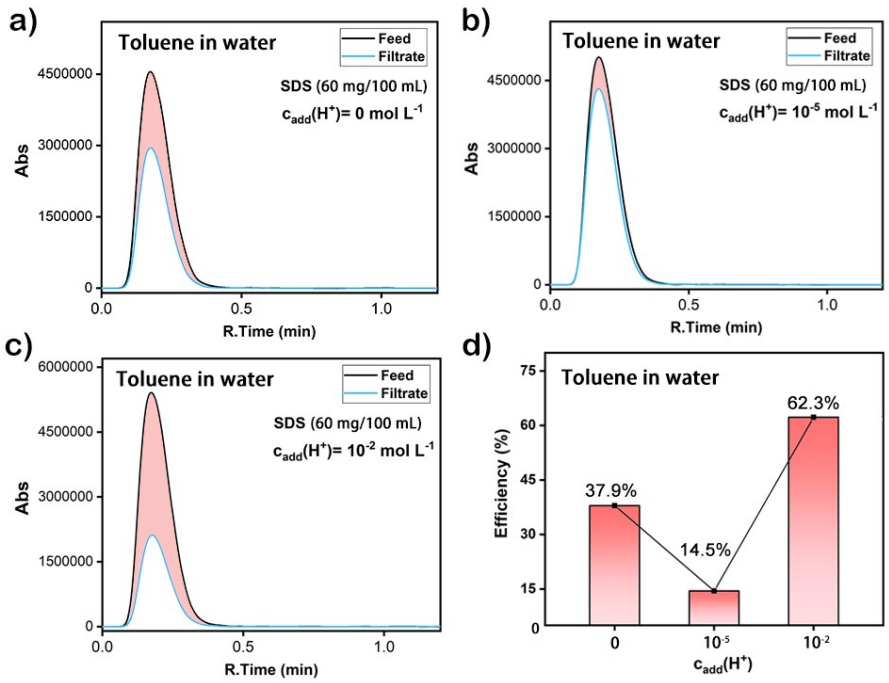

Fig. S12 The influence of the adding of H^+ on SDS surfactant removal performance (the concentration is 60 mg/100 mL). a)-c) The EIC images of the feed emulsions and filtrates with the $C_{add}(H^+)$ at 0 mol L^{-1}, 10^{-5} mol L^{-1} and 10^{-2} mol L^{-1}, respectively. d) The correspinding surfactant removal efficiencies of the $–NH_2$ side under the aforementioned concentrations. Although the removal efficiency reduces with the concentration of SDS surfactant increasing, the removal efficiency still decreases at first and then rises to a relatively high piont.

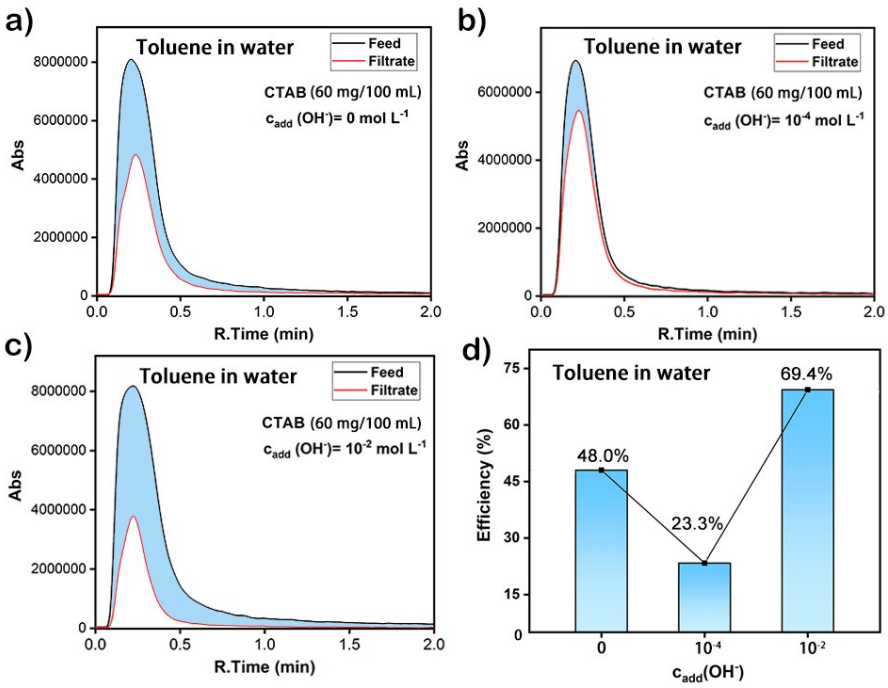

Fig. S13 The influence of the adding of OH^- on CTAB surfactant removal performance (the concentration is 60 mg/100 mL). a)-c) The EIC images of the feed emulsions and filtrates with the $C_{add}(OH^-)$ at 0 mol L^{-1}, 10^{-4} mol L^{-1} and 10^{-2} mol L^{-1}, respectively. d) The correspinding surfactant removal efficiencies of the –COOH side under the aforementioned concentrations. Although the removal efficiency reduces with the concentration of CTAB surfactant increasing, the removal efficiency still decreases at first and then rises to a relatively high piont.

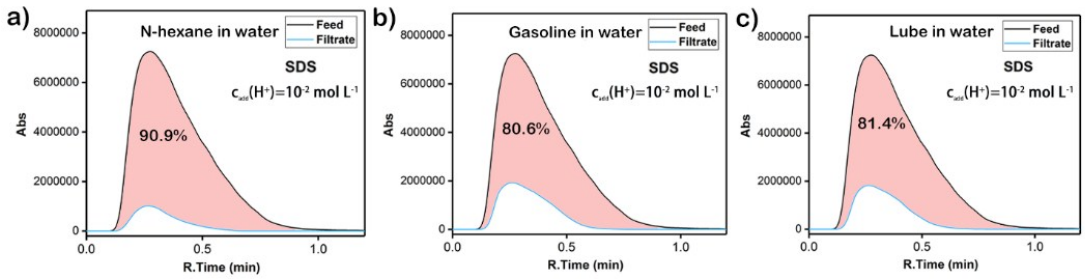

Fig. S14 *In-situ* SDS surfantant removal performance of n-hexane in a) water emulsion separation, b) gasoline in water emulsion separation and c) lubrication oil in water emulsion separation with the $C_{add}(H^+)$ at 10^{-2} mol L^{-1}.

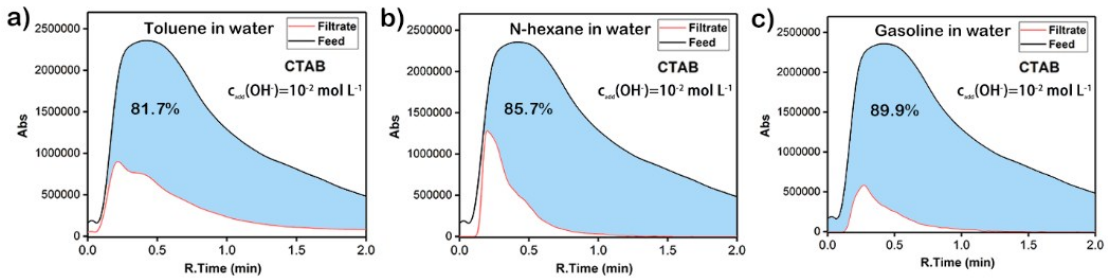

Fig. S15 *In-situ* CTAB surfantant removal performance of a) toluene in water emulsion separation, b) n-hexane in water emulsion separation and c) gasoline in water emulsion separation with the $C_{add}(OH^-)$ at 10^{-2} mol L^{-1}.

Discussion: The classification of Janus membrane and the advantages of triple layered "A and B" Janus material fabricated in this work

With the continuous deepening of the research on Janus membrane for oil/water separation, this kind of material has been divided in to two categories which have their own characteristics and applications. The first type is "A on B" or "A on B" Janus membrane with complementary pattern which the referee has mentioned aforementioned. In this situation, two layers of the membrane work together to achieve the whole separation process. Based on capillary effect and thickness modulation (one layer is relatively thin and the other is relatively thick), the membrane is able to realize directional transport, which is the main function of this kind of material. Up to date, there are numerous excellent works about synergistic patterned Janus membrane and most of them utilize two layers to realize one purpose (such as immiscible oil/water mixture separation or oil collection from oil/water emulsion).

The second type is "A and B" Janus membrane with individual pattern, in which the two layers are relatively thick. So they can maintain their own property and work independently (the membrane fabricated in this work belongs to this category). This kind of material has also been studied in detail since it can achieve switchable permeation by just altering the sides, thus accomplishing different objectives in separation field. It is always of great significance to design multi-functional superwetting material and realize the separation of different oil/water mixtures in one material (especially in emulsion separation area where various kinds of complex emulsions need to be disposed), because it can save cost for fabricating different types of materials, reduce energy use and simplify the whole separation device. These are also the advantages of multi-functional membranes over materials with single function. Based on the above discussion, two types of Janus membranes have their own application fields and are worth of studying.

When it comes to this work, the dual functional Janus membrane has its own advantages. The first combination of functional groups with superhydrophilic wettability makes the material achieve *in-situ* oil in water emulsion separation and

surfactant removal. While the second combination of two different functional layers ensures that the Janus membrane can dispose both anionic and cationic stabilized emulsions. In addition, if the whole device is placed horizontally and the Janus membrane is placed vertically, different emulsions are able to pass through the membrane from different functional sides. In this situation, there is no need to take out the material, thus simplifying the operating process and reducing the replacing time for other membranes. Moreover, different from traditional Janus membranes with different wettability, since this Janus material exhibits homogeneous superhydrophilic wettability, the separation efficiency and flux are not affected by hydrophobic layers, leading to good emulsion separation performance. All of the aforementioned strengths increase the potential of the material utilized in industrial field.