

**Electronic Supplementary Information (ESI):**

**Ultralight Free-Standing Reduced Graphene Oxide Membranes for Oil-in-Water Emulsions Separation**

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## **Experiments in details:**

### **Materials**

Dopamine-hydrochloride (Sangon, Biotech Co. Ltd., Shanghai, China) was used as purchased. Tween 20, cetyl trimethyl ammonium bromide (CTAB) and sodium dodecyl sulfate (SDS) were of analytical grade from Sinopharm Chemical Reagents. All other chemicals were used without further purification.

### **Fabrication of Free-Standing RGO Membranes**

GO was synthesized by the modified Hummers Method.<sup>47</sup> The initial GO suspension was diluted with 5 mL of water (1: 10). PDA coating was performed by mixing the GO suspension with buffer solution (10 mg of dopamine per mL of 1M Tris buffer, pH 8.5) at room temperature. After 24 h of magnetically stirring, 4 mL of dispersion was filtered on a MCE filter membrane with pore sizes of 0.22  $\mu\text{m}$  average diameter under vacuum filtration. Free-standing RGO membranes were subsequently obtained by dried at ambient condition for several minutes.

### **Preparation of Oil-in-Water Emulsions**

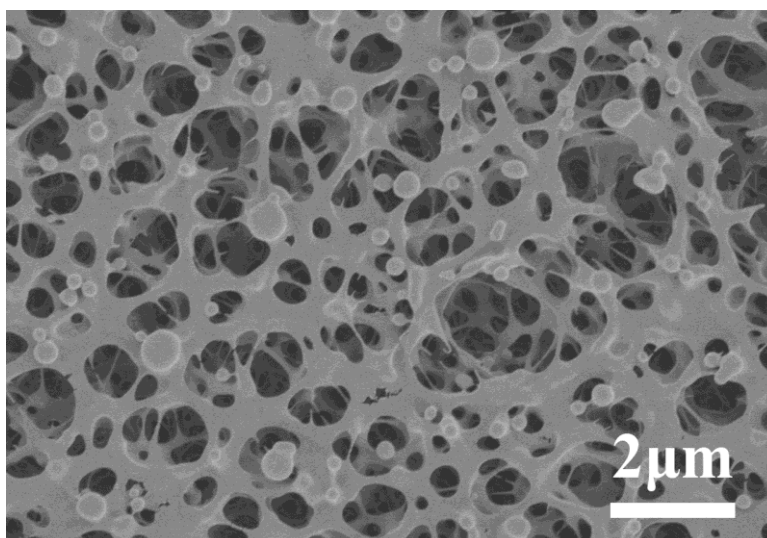
Surfactant-stabilized oil-in-water emulsions were prepared by mixing water and oil (toluene, n-hexane and diesel, respectively) in 100 : 1 (v/v) with addition of 0.5 mg of surfactant (Tween 20, CTAB and SDS, respectively) under high stirring for 24h.

### **Instruments and Characterization**

FESEM images were obtained on a field emission scanning electron microscope (SU-8010, Hitachi Limited, Japan). The AFM image was measured on atomic force microscope (SPM-9600 series, shimadzu, Japan). X-ray photoelectron spectroscopy (XPS) measurements were carried out on a Thermo escalab 250Xi spectrometer using an Al K $\alpha$  X-ray source (1486.6 eV). FTIR spectra were recorded using a Fourier Infrared Spectrometer (NICOLET6700, Thermo Co-operation, USA). Contact angles were measured on a contact angle measurement machine (OCA 15 machine, Data-Physics, Germany). Optical microscopy images were taken on a polarizing microscope (Nikon ECLIPSE LV100POL, Japan). Dynamic light scattering (DLS) measurements were performed on a Zeta Plus apparatus (Zeta Plus, Brookhaven Instruments, Holtsville, NY). The oil content in the filtrate was measured by the infrared spectrometer oil content analyzer (Oil480, Beijing Chinainvent Instrument Tech. Co. Ltd., China).

### **Emulsion Separation Tests**

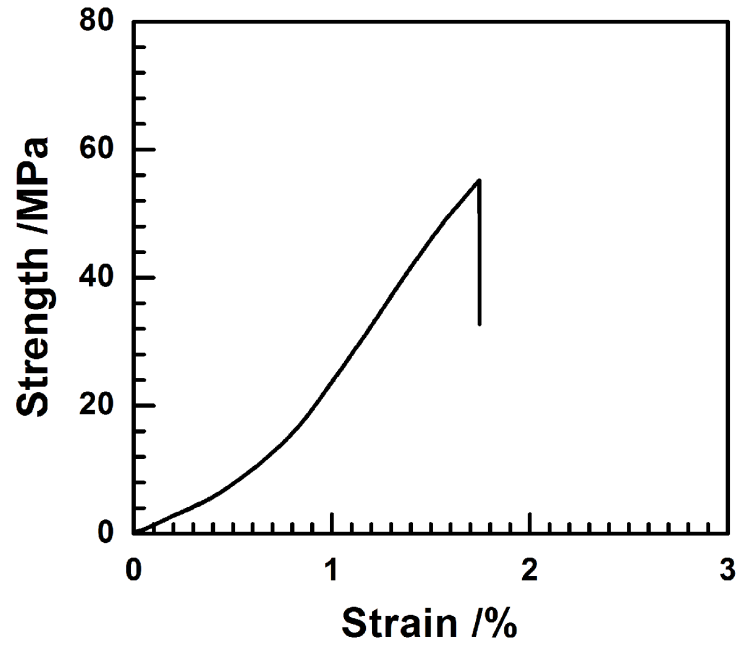
The filtered RGO membrane was placed on the filtration apparatus. A certain volume of oil-in-water emulsion was added into the apparatus. The separation processes were carried out under a pressure of  $\sim 0.1$  MPa.



**Fig. S1** FESEM image of the MCE support exhibiting an abundant porous structure consisting of a polymer framework with a fairly smooth surface.

Atomic /%	C	N	O
Before	37.03	/	62.97
After	74.67	4.56	20.77

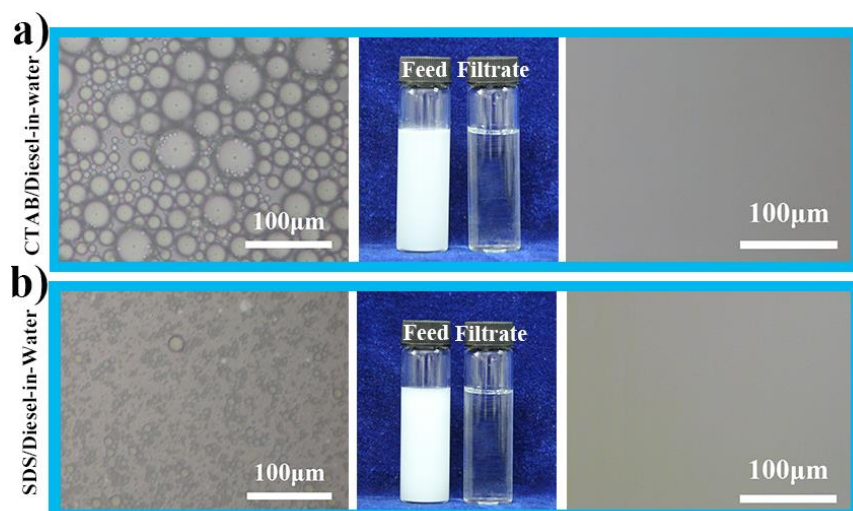
**Tab. S1** The element change comparison of GO before and after reaction with dopamine.



**Fig. S2** Mechanical performance of the free-standing RGO membrane.



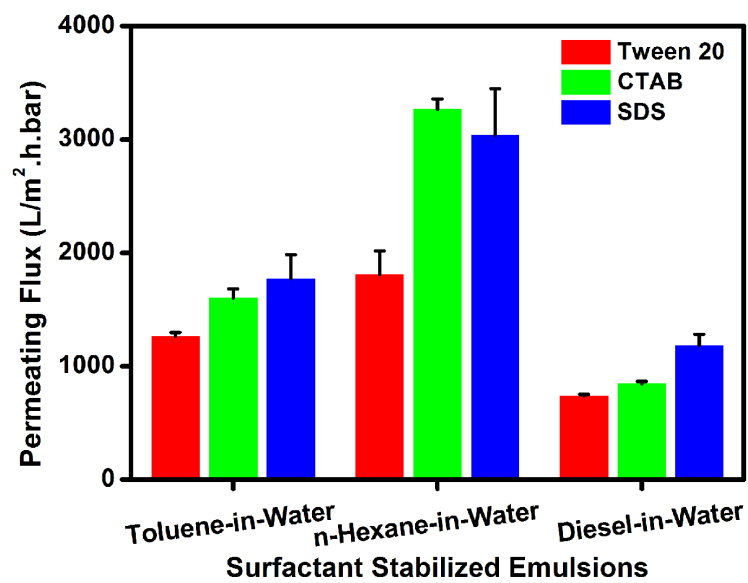
**Fig. S3** The vacuum driven filtration system used in this work.



**Fig. S4** Photographs of CTAB-stabilized and SDS-stabilized diesel-in-water emulsions before and after filtration.



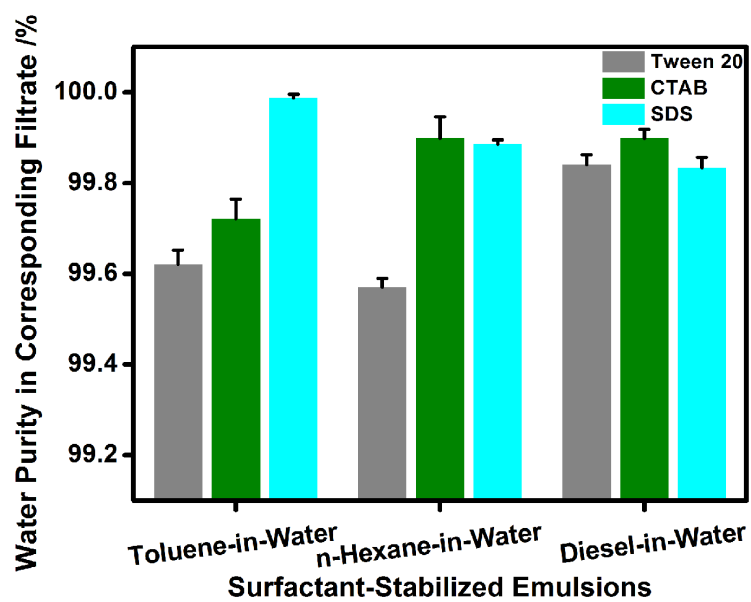
**Fig. S5** Tween 20-stabilized diesel-in-water emulsion separation processes of two layers of MCE filter membranes (in the middle) and the MCE-supported RGO membrane (in the right).



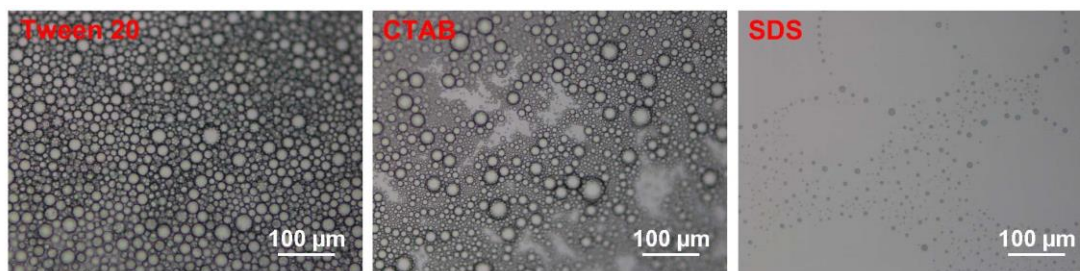
**Fig. S6** Permeation flux of the PDA-RGO membranes for toluene-in-water, n-hexane-in-water and diesel-in-water emulsions stabilized with different surfactants.

Oil	Concentration /mg.L <sup>-1</sup>
Toluene	~ 8670
n-Hexane	~ 6614
Diesel	~ 8246

**Tab. S2** The initial oil concentration in the feed before separation.



**Fig. S7** Water purity in corresponding filtrate for toluene-in-water, n-hexane-in-water and diesel-in-water emulsions stabilized with different surfactants.



**Fig. S8** Optical microscopy images of three types of surfactants stabilized diesel-in-water emulsions after maintained for 10 d without any disturbance.



**Fig. S9** Wetting behavior toward water in air and 1, 2-dichloroethane under water as well as microstructure of the RGO membrane after cycles of emulsion separation tests.