Electronic Supplementary Information for:

Integrated asymmetric superwetting Janus membrane for the efficient separation of various surfactant-stabilized oil-water emulsions

Xiuping Chen,[a] Yushuang Yang,[a] Yiming Li,[a,*] Mutai Bao,[a] Dan Zhang,[a] Zhining Wang [b,*]

[a] Key Laboratory of Marine Chemistry Theory and Technology, Ministry of Education, Ocean University of China, Qingdao 266100, P.R. China

[b] Shandong Key Laboratory of Water Pollution Control and Resource Reuse, School of Environmental Science and Engineering, Shandong University, Jinan 250100, P.R. China

* Corresponding author.
E-mail address: liym@ouc.edu.cn (Y. M. Li); wangzhn@sdu.edu.cn (Z.N. Wang).

Text S1 Effects of MHCH nanowires and SiO_2 microspheres concentration on the membrane separation performance

The separation performance of hydrophobic MHCH&SiO_2 membrane for W/O emulsion was influenced by the amounts of MHCH and SiO_2. All the hydrophobic MHCH&SiO_2 membrane is about 6 cm in diameter. As can be seen from **Fig. S5**, when the amount of MHCH nanowires is kept constant, separation flux gradually increases with the amount of SiO_2 microspheres. More SiO_2 microspheres increases the porosity of the membrane, thereby increasing membrane separation flux. However, the separation flux declines progressively with the amount of MHCH nanowires due to the formation of a much denser membrane. The decreased porosity reduces the membrane separation flux. **Fig. S6** shows that when $m_{\mathrm{MHCH}} < 0.08$ g and $m_{\mathrm{SiO_2}} < 0.27$ g, the resulting filtrates were relatively turbid. When 0.12 g < m_{MHCH} < 0.15 g and 0.13 g < $m_{\mathrm{SiO_2}}$ < 1.00 g, clear filtrates were obtained. So, taking into account the separation performance and synthesis cost, 0.12 g MHCH and 0.40 g SiO_2 were used to prepare hydrophobic MHCH&SiO_2 membrane in the following studies. Under this condition, the freestanding MHCH&SiO_2 membrane individually shows a high separation flux of about 2600 L m^{-2} h^{-1} for W-H emulsion and better emulsion separation performance (**Fig. S6 MS9**).

Fig. S1 Digital photo of the filter facility and the vacuum pump.

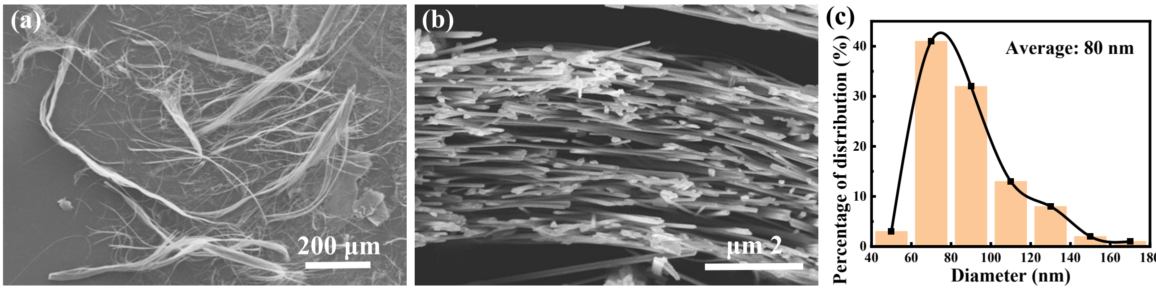

Fig. S2 SEM images (a, b) and diameter distribution (c) of MHCH nanowires. The diameter distribution data is obtained by the Nano Measurer software statistical typical section SEM image (b).

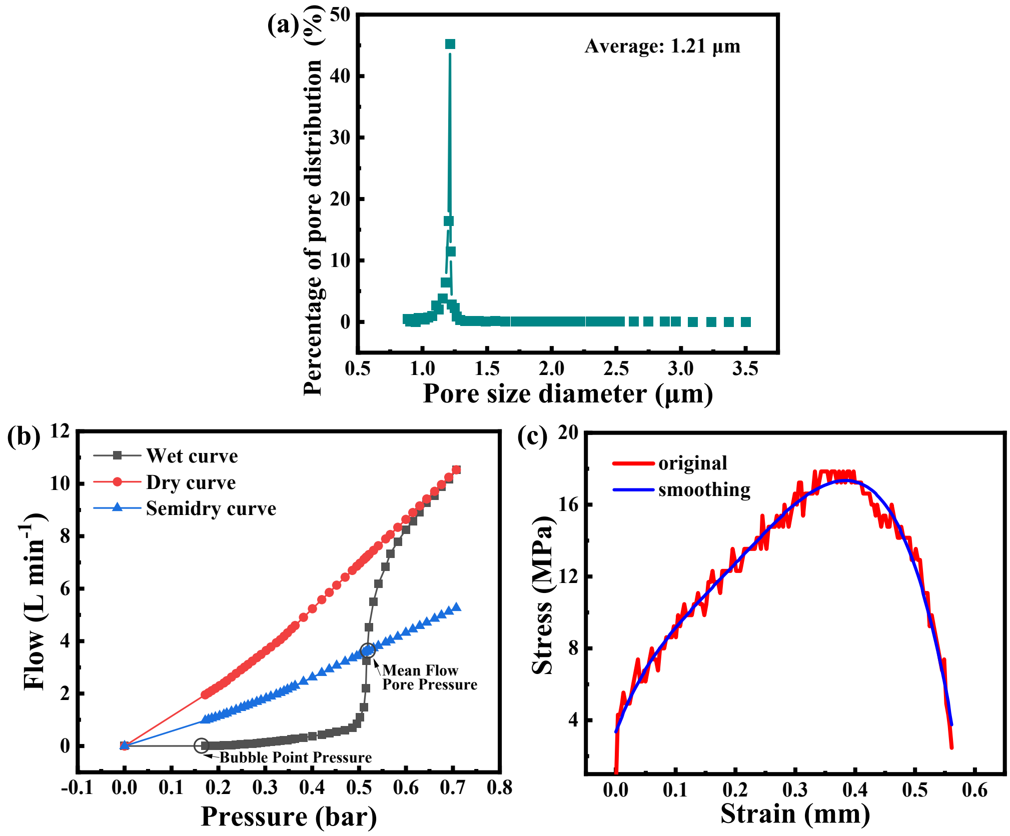

Fig. S3 Pore diameter distributions of the hydrophobic (PDMS&TPE)@(MHCH&SiO_2) membrane (a). The curve of the N_2 flow-pressure of the wet, dry and semidry hydrophobic (PDMS&TPE)@(MHCH&SiO_2) membrane (b). The mechanical properties (c) of the hydrophobic (PDMS&TPE)@(MHCH&SiO_2) membrane.

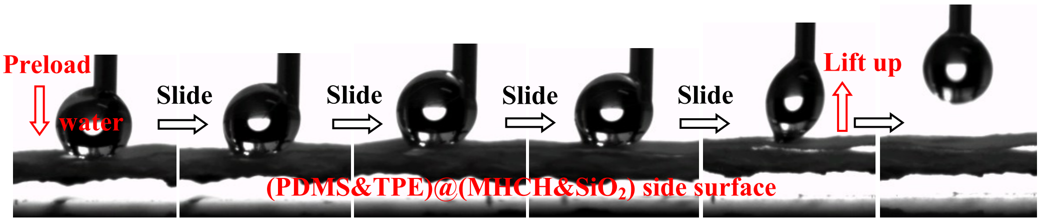

Fig. S4 Photographs of the dynamic water repelling on the hydrophobic (PDMS&TPE)@(MHCH&SiO_2) side in air.

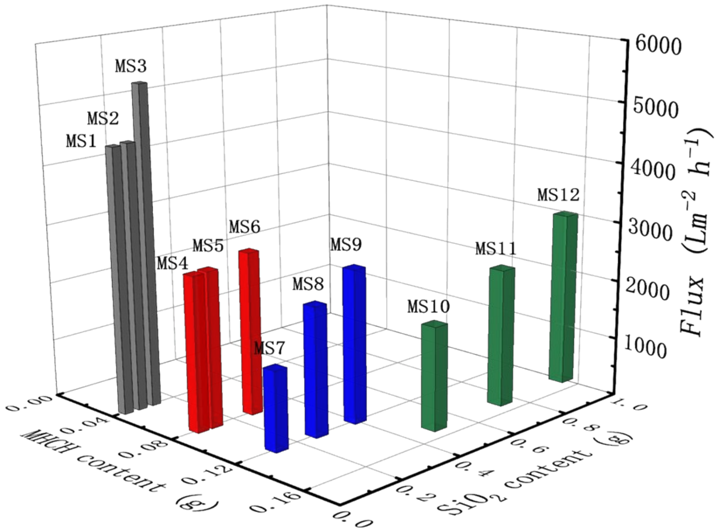

Fig. S5 Summary of W-H emulsion separation flux by hydrophobic MHCH&SiO_2 membrane prepared from different amounts of MHCH nanowires and SiO_2 microspheres. All the hydrophobic MHCH&SiO_2 membrane is about 6 cm in diameter. (C_{MHCH} = 1.0 mg mL^{-1}).

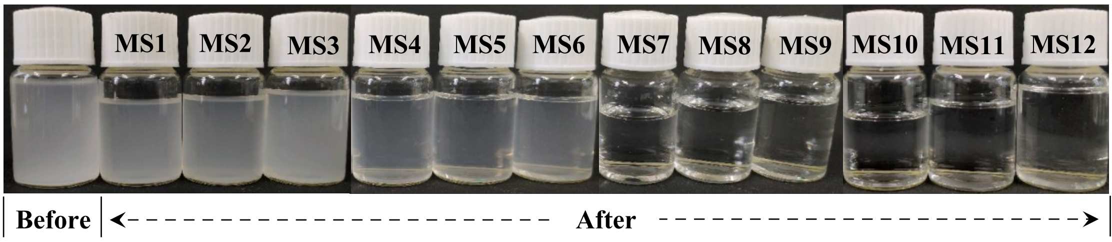

Fig. S6 Digital photos of W-H emulsion before and after separation by hydrophobic MHCH&SiO_2 membrane prepared from different amounts of MHCH and SiO_2 (MS1-MS12). Original W-H emulsion (C_{Span80} = 1.5 mg mL^{-1}, $V_{\mathrm{n\text{-}hexane}}$:$V_{\mathrm{water}}$ = 99:1).

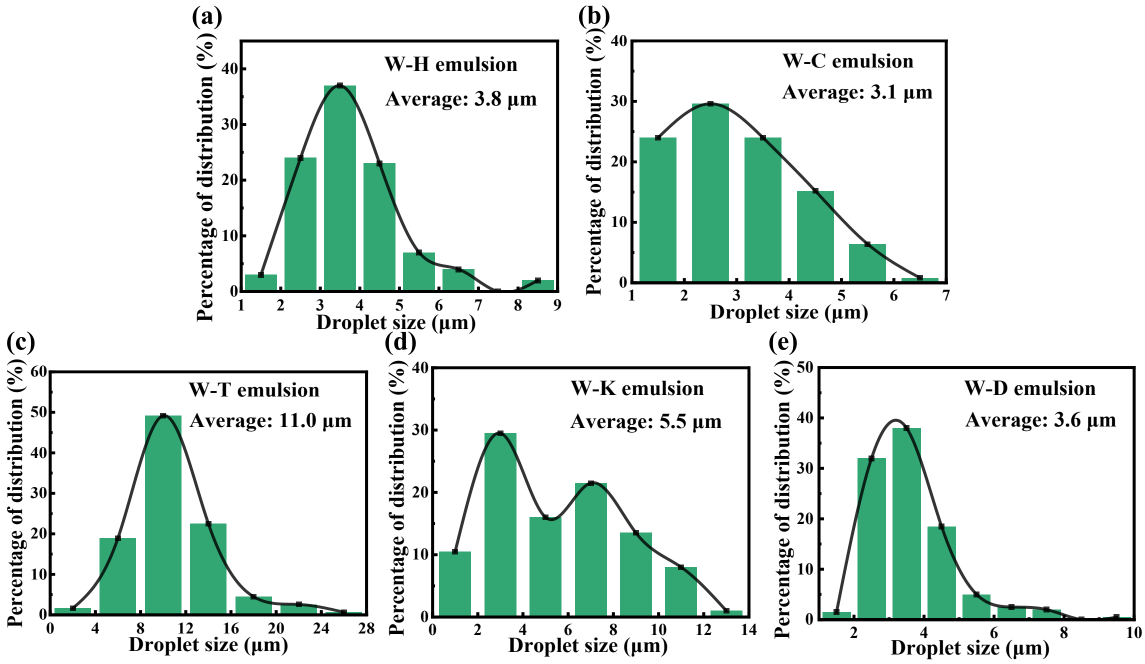

Fig. S7 Corresponding water droplet size distribution of (a) W-H, (b) W-C, (c) W-T, (d) W-K, (e) W-D emulsions stabilized by Span 80 before separation.

Fig. S8 Digital photo of the large water droplets collected on the hydrophobic MHCH&SiO_2 side of Janus membrane.

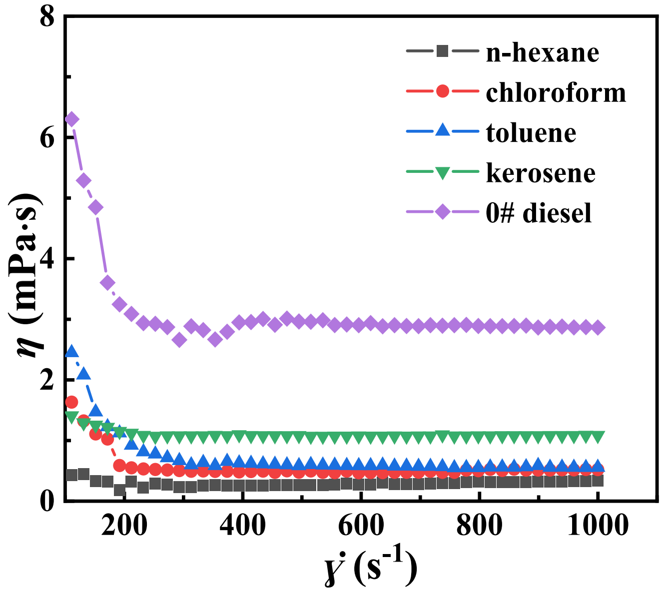

Fig. S9 Shear viscosity of five oils at different shear rates.

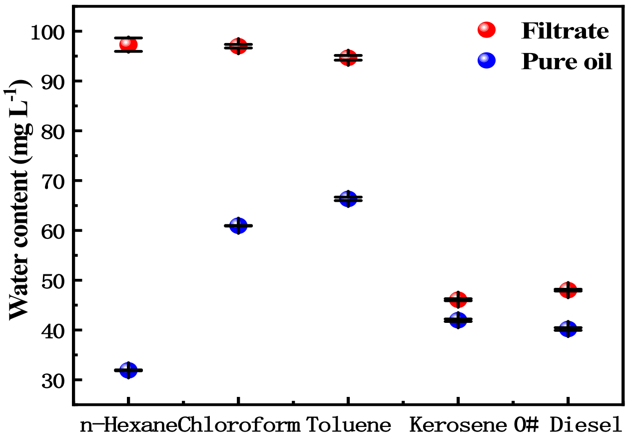

Fig. S10 Water content of the water-in-oil emulsions filtrate and the corresponding pure oil.

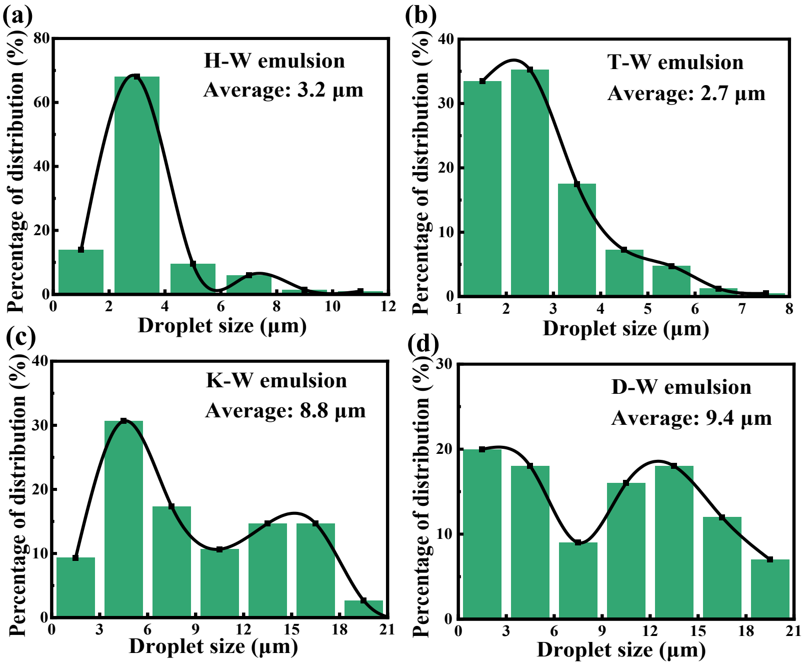

Fig. S11 Corresponding water droplets distribution of the emulsifier SDS stabilized (a) H-W, (b) T-W, (c) K-W, (d) D-W emulsions before separation.

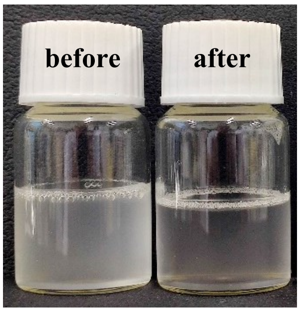

Fig. S12 Digital photo of the H-W emulsions before and after separation by the individual Co_3O_4 nanoneedles@SSM membrane.

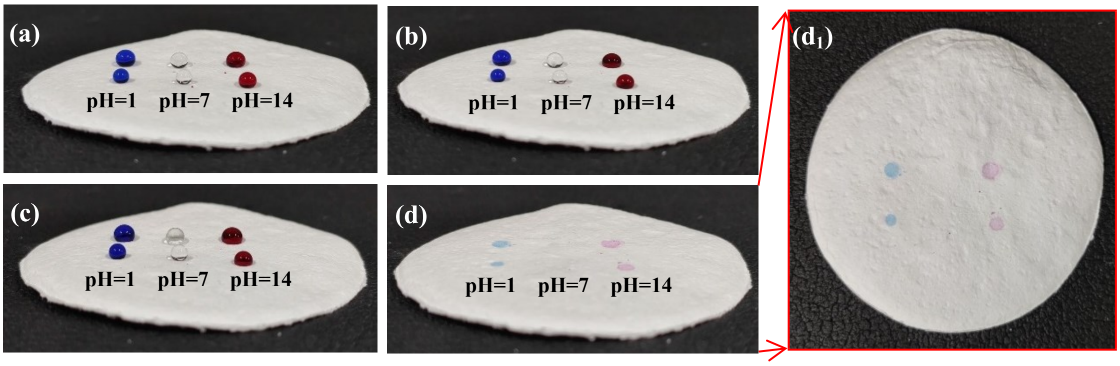

Fig. S13 Digital photos of water droplets toward different pH values on the hydrophobic MHCH&SiO_2 membrane surface after a while (a) 1 h, (b) 3 h, (c) 6 h. Digital photos after the different pH water droplets were removed from the membrane surface after 6 h (d, d_1). The color is used here for better recognition.

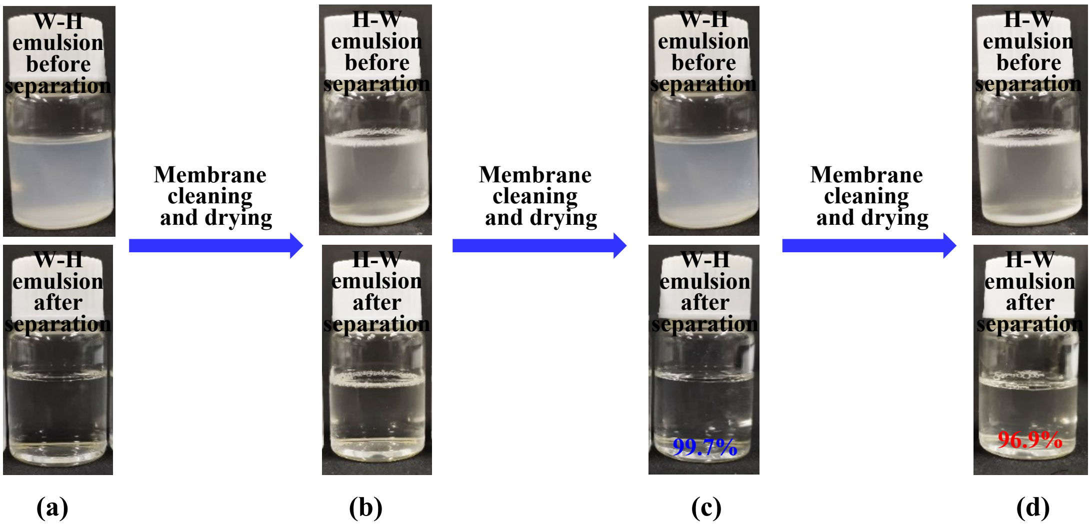

Fig. S14 Alternately used performance of the integrated Janus membrane for W-H and H-W emulsion separation.

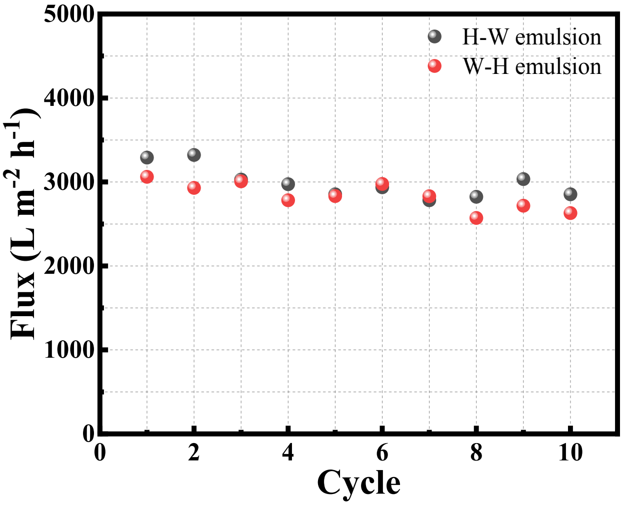

Fig. S15 The recyclability of the integrated Janus membrane for Span 80 stabilized W-H (a) and SDS-stabilized H-W (b) emulsions separation.

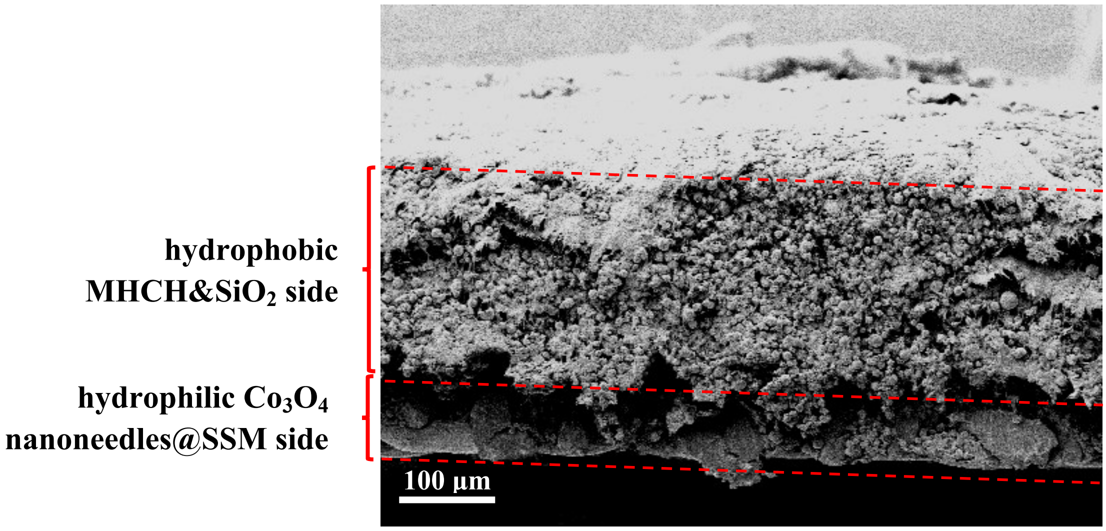

Fig. S16 Cross-section view of the integrated Janus membrane.

Table S1 Pore properties of the hydrophobic (PDMS&TPE)@(MHCH&SiO_2) membrane.

Pore characters	Value
Average Pore Diameter (μm)	1.21
Bubble Point Diameter (μm)	3.64
Minimum Diameter (μm)	0.89
Mean Flow Pore Pressure (bar)	0.52
Bubble Point Pressure (bar)	0.17

Table S2. Comparison of W/O emulsions separation performance with filtration materials reported recently.

Filtration materials	Oil	V_{oil}:V_{water}	Droplet size (μm)	C_{Span80} (mg mL^{-1})	Flux (L m^{-2} h^{-1})	TMP (bar)	Separation efficiency (%)	Ref
PNIPAAm coated nylon membrane	n-hexane	100:1	6.59	1.0	~1300	1.0	>97.8	41
	toluene		3.99		~1050		>97.8	
PANI-SiNPs Janus membrane	n-hexane	100:1	6.84	1.5	~1400	<1.0	>99.0	21
	toluene		9.01		~1500		>99.0	
Janus F-TiO_2@PPS Porous Membrane	n-hexane	99:1	~2	0.5	3500	0.9	98.2	47
	chloroform		~2		2400		98.1	
	toluene		~2		2900		98.4	
Janus CNTs@PAN_{EN} membrane	chloroform	9:1	~1	0.5	~8400	0.7	~99.2	36
Coprinus comatus-coated PVDF membrane	n-hexane	50:1	0.050-1	0.4	78–112	0.85	~99.0	42
	kerosene		0.050-1				~99.4	
	diesel		0.050-1				~98.85	
Egg shells powders coated PVDF membrane	kerosene	50:1	0.1-0.5	2.0	~200	0.5	~99.7	45
Candle soot coated PVDF membrane	kerosene	50:1	0.1-0.9	0.1-0.2wt%	~75	0.85	>99.99	43
PDVB Modified PVDF Membrane	diesel	100:1	0.06-0.09	2.5	~60	0.8	~99.98	44
Janus ZnO-cellulose/MnO_2 hybrid membranes	n-hexane	96:4	-	3.0	~1035	0.3	>99.4	23
	toluene		0.5-3.5		~2589		>99.4	
	chloroform		-		~2652		>99.4	
	diesel		-		~477		>99.4	
Integrated Janus membrane	**n-hexane**	**99:1**	**3.8**	**1.5**	**3349**	**0.6**	**99.03**	**This work**
	chloroform		**3.1**		**2321**		**99.03**	
	toluene		**11.0**		**1757**		**99.06**	
	kerosene		**5.5**		**1817**		**99.54**	
	0# diesel		**3.6**		**1132**		**99.52**	

Table S3. Comparison of O/W emulsions separation performance with some related filtration materials reported previously.

Filtration materials	Oil	V_{water}:V_{oil}	Droplet size (μm)	$C_{Surfactant}$ (mg mL^{-1})	Flux (L m^{-2} h^{-1})	TMP (bar)	separation efficiency (%)	Ref
PNIPAAm coated nylon membrane	n-hexane	100:1	11.34	C_{SDS}=1.0	~2500	gravity	>99	41
	toluene		8.71		~2000		>99	
PANI-SiNPs Janus membrane	n-hexane	100:1	7.98	C_{SDS}=1.5	~1900	<0.3	>99.7	21
	toluene		9.39		~1600		>99.7	
Janus F-TiO_2@PPS Porous Membrane	n-hexane	99:1	0-8	C_{SDS}=0.5	4400	0.9	98.4	47
	toluene		0-8		4700		98.5	
Silica-decorated polypropylene membranes	diesel	99:1	0.1-7	C_{SDS}=0.2	~1300	0.4	>99	48
TiO_2 decorated Superhydrophilic PVDF membranes	diesel	100:1	1-20	C_{SDS}=0.2	382	0.9	~99.52	49
Coprinus comatus-coated PVDF membrane	n-hexane	50:1	0.050-1	C_{Span80}=0.4	70-80	0.85	~98.8	42
	kerosene		0.050-1				~99.2	
	diesel		0.050-1				~99.15	
PVDF/PDA/PMEN membrane	kerosene	99:1	0.12	C_{Span80}=2.5	~1600	1.0	>99.9	46
	n-hexane		0.43		~1700		>99.9	
	toluene		0.31		~1550		>99.9	
Loess-coated PVDF membranes	diesel	50:1	0.1-1	$C_{Tween80}$=2.0	~510	0.85	>99.2	50
Candle soot coated PVDF membrane	kerosene	50:1	0.10-0.50	$C_{Tween80}$ =0.1-0.2 wt%	~75	0.85	~99.3	43
Ultrathin 2D $Ti_3C_2T_x$ MXene membrane	kerosene	100:2	0.10-0.50	$C_{Tween80}$=2.0	~425	0.85	>99.4	51
Janus ZnO-cellulose/MnO_2 hybrid membranes	n-hexane	96:4	-	$C_{Tween80}$=3.0	~4122	0.3	>99.6	23
	toluene		0.5-2.5		~3465		>99.6	
	diesel		-		~2514		>99.6	
Integrated Janus Membrane	**n-hexane**	**99:1**	**3.2**	**C_{SDS}=1.5**	**3121**	**0.8**	**99.6**	**This work**
	toluene		**2.7**		**2863**		**99.9**	
	kerosene		**8.8**		**1975**		**99.6**	
	0# diesel		**9.4**		**2324**		**99.8**	

Table S4 Theoretical calculation of the pressure-related value of the liquid.

Pressure	W/O emulsion separation	O/W emulsion separation

	(MPa)	(MPa)
$P_{L\ (n\text{-}hexane)}$	0.305	-0.269
$P_{L\ (water)}$	-1.041	1.190
$P_{H\ (n\text{-}hexane)}$	0-0.067	−
$P_{H\ (water)}$	−	0.101
P_P	0.060	0.08
$P_{Total\ (n\text{-}hexane)}$	0.365-0.432	-0.189
$P_{Total\ (water)}$	-0.981	1.270-1.371