Supporting Information

Triazine porous organic polymer thin film by nanoparticle-polymer reticulation for high-efficient molecule/ion separation

Yan-Li Ji,[a]* Kai Zhang,[a] Jin-Lin Wang,[a] Zhuang-Zhuang Liu,[a] Xin Wen[b]* and Cong-Jie Gao[a]

a) *Center for Membrane and Water Science & Technology, Zhejiang University of Technology, Hangzhou 310014, China. E-mail: yanliji@zjut.edu.cn*

b) *Key Laboratory of Chemical Biology of Hebei Province, College of Chemistry and Environmental Science, Hebei University, Baoding 071002, China. E-mail address: wenxin767@hotmail.com*

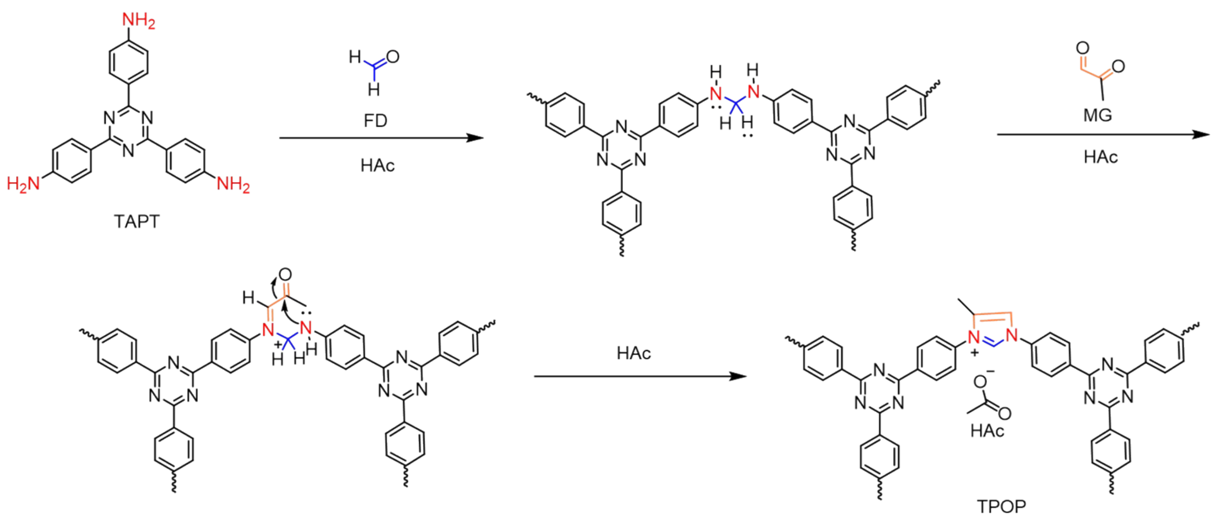

Fig. S1 The synthetic procedure of TPOP nanoparticles.

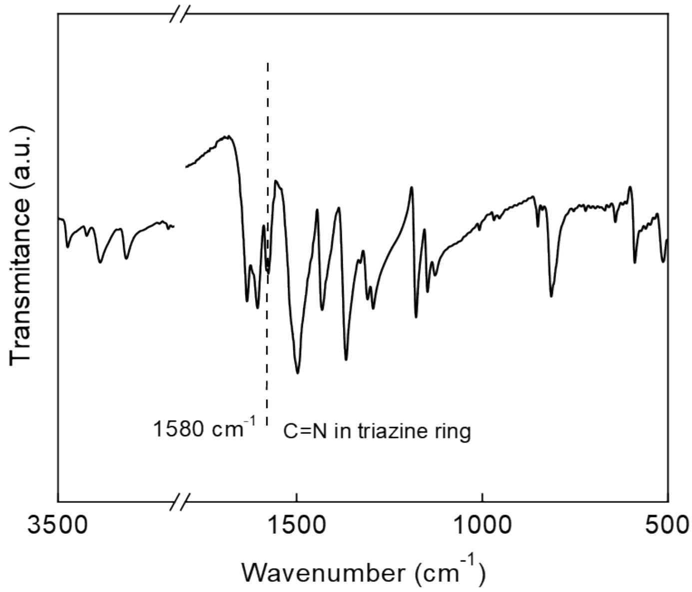

Fig. S2 FT-IR spectrum of TAPT monomer.

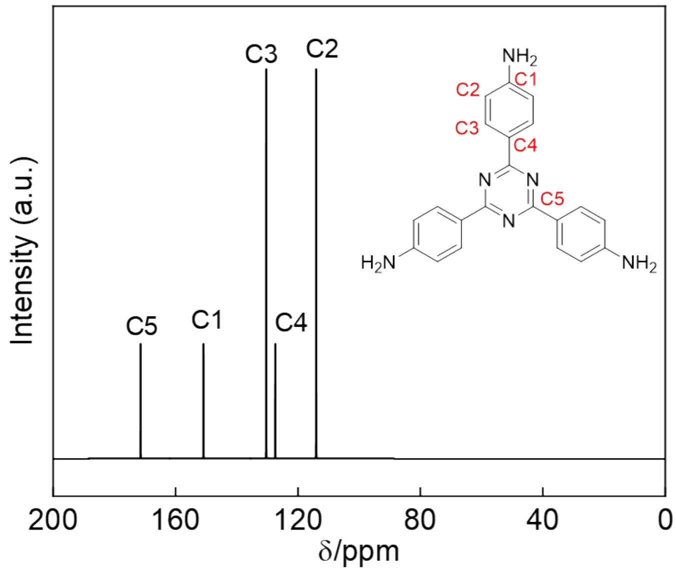

Fig. S3 The simulated ^{13}C NMR spectrum of TAPT monomer.

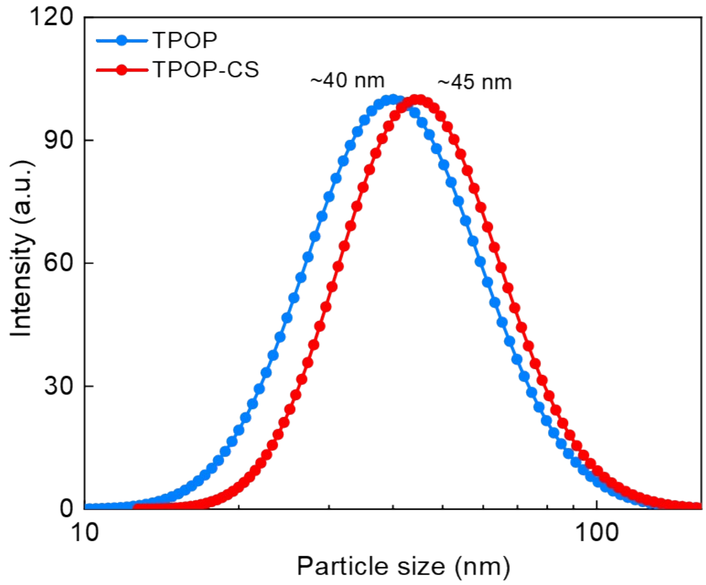

Fig. S4 DLS particle size distribution of TPOP and TPOP-CS nanoparticles in 0.001 wt% aqueous solution.

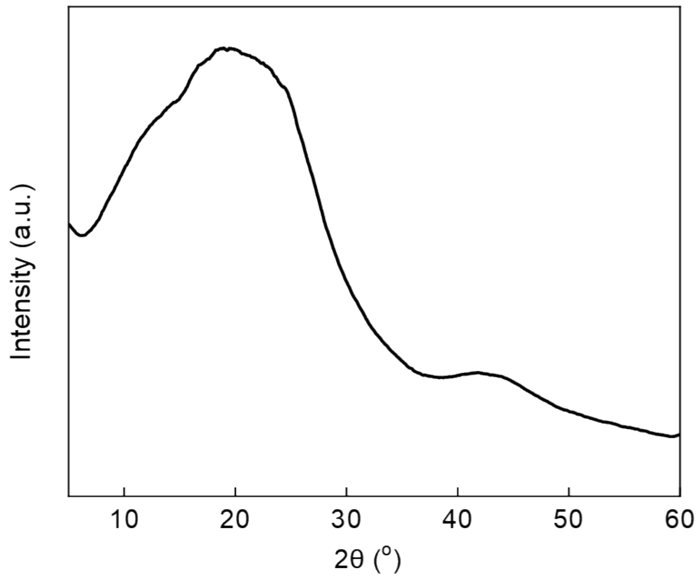

Fig. S5 XRD pattern of TPOP powders.

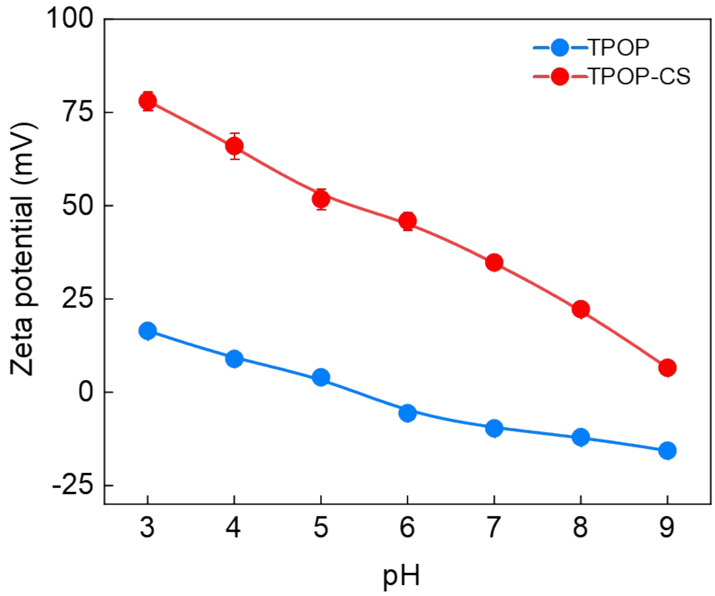

Fig. S6 Zeta potential of TPOP and TPOP-CS nanoparticles varies with the pH value of aqueous solution (0.001 wt%).

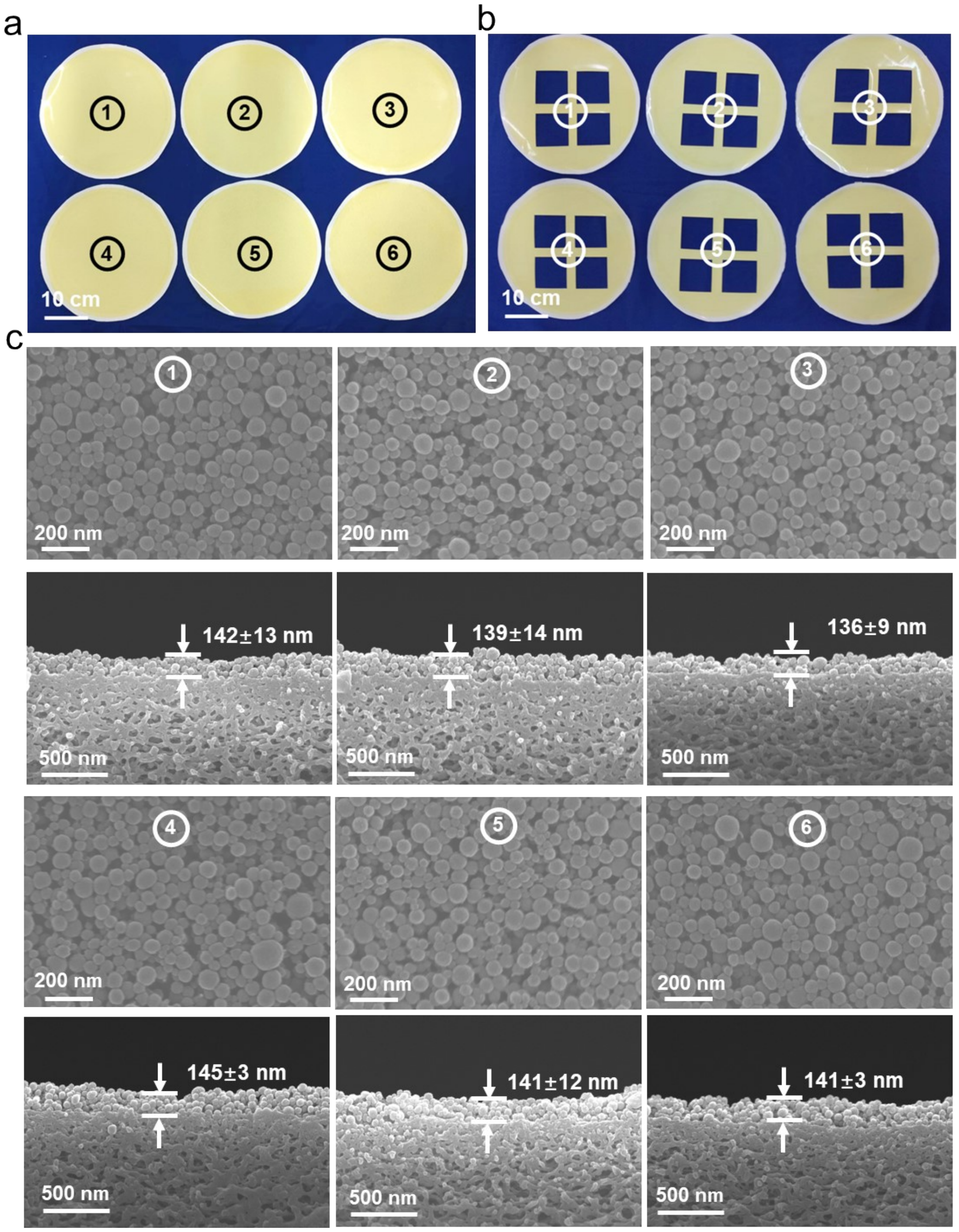

Fig. S7 Optical photographs of (a) six pieces of 707 cm^2 TPOP-CSM membranes and (b) the samples were cut off from the membranes. (c) SEM surface/cross-section images of six pieces of 707 cm^2 TPOP-CSM membranes.

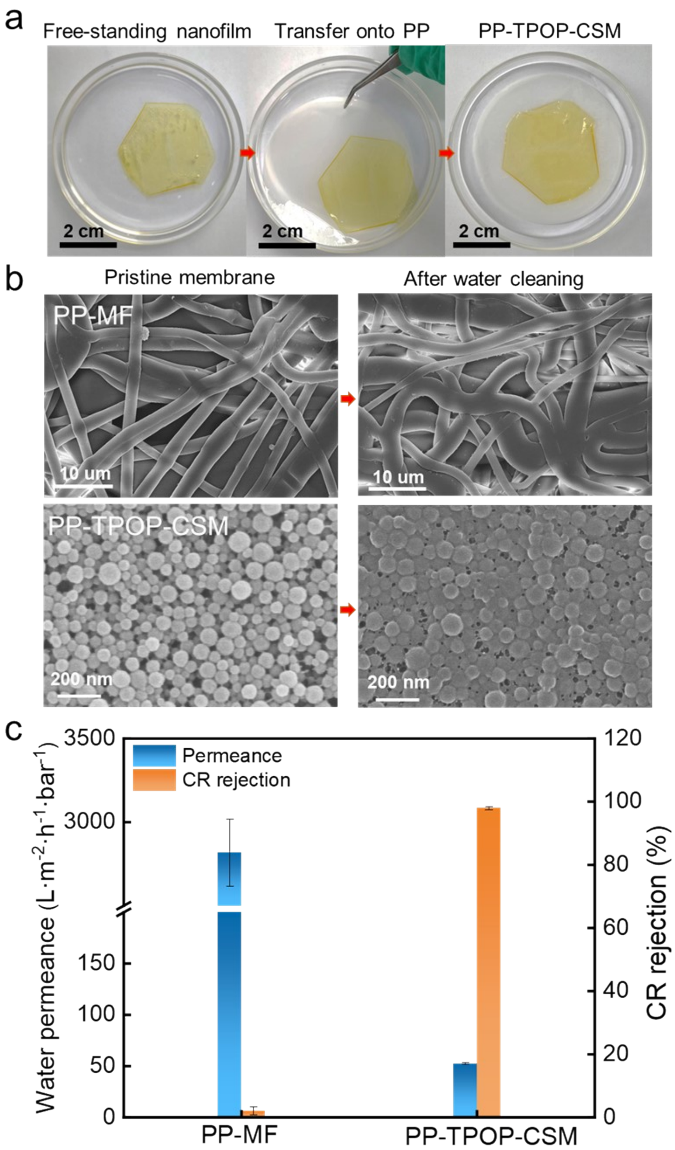

Fig. S8 (a) Optical photographs of a free-standing TPOP-CSM nanofilm was transferred onto a polypropylene microfiltration (PP-MF) supporting membrane. (b) FESEM surface images of PP-MF and PP-TPOP-CSM membranes (left: pristine membrane, right: after water cleaning membrane). (c) Separation performance of PP-MF and PP-TPOP-CSM membranes (test with 0.1 g·L^{-1} CR aqueous solution at 25 °C and 2.0 bar.).

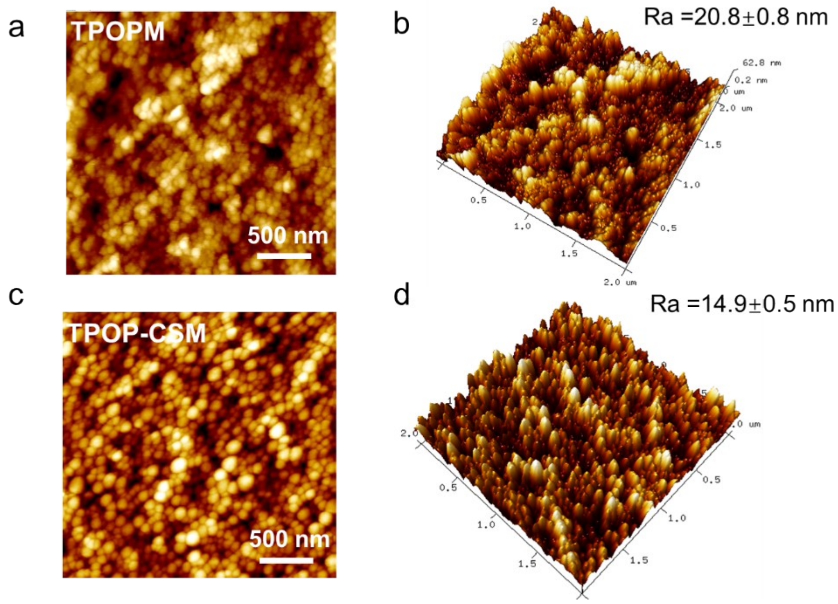

Fig. S9 AFM surface images of (a, b) TPOPM and (c, d) TPOP-CSM membranes with a scan area of 2 um×2 um and the corresponding arithmetic average roughness (Ra).

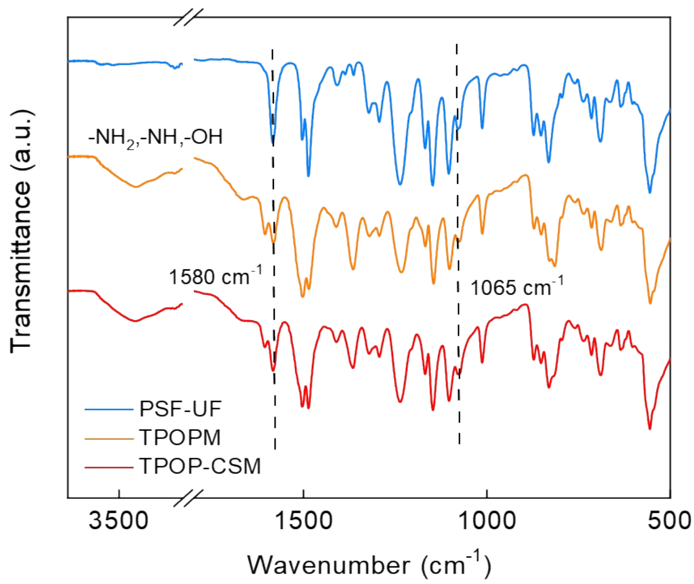

Fig. S10 ATR-FTIR spectra of PSF-UF, TPOPM and TPOP-CSM membranes.

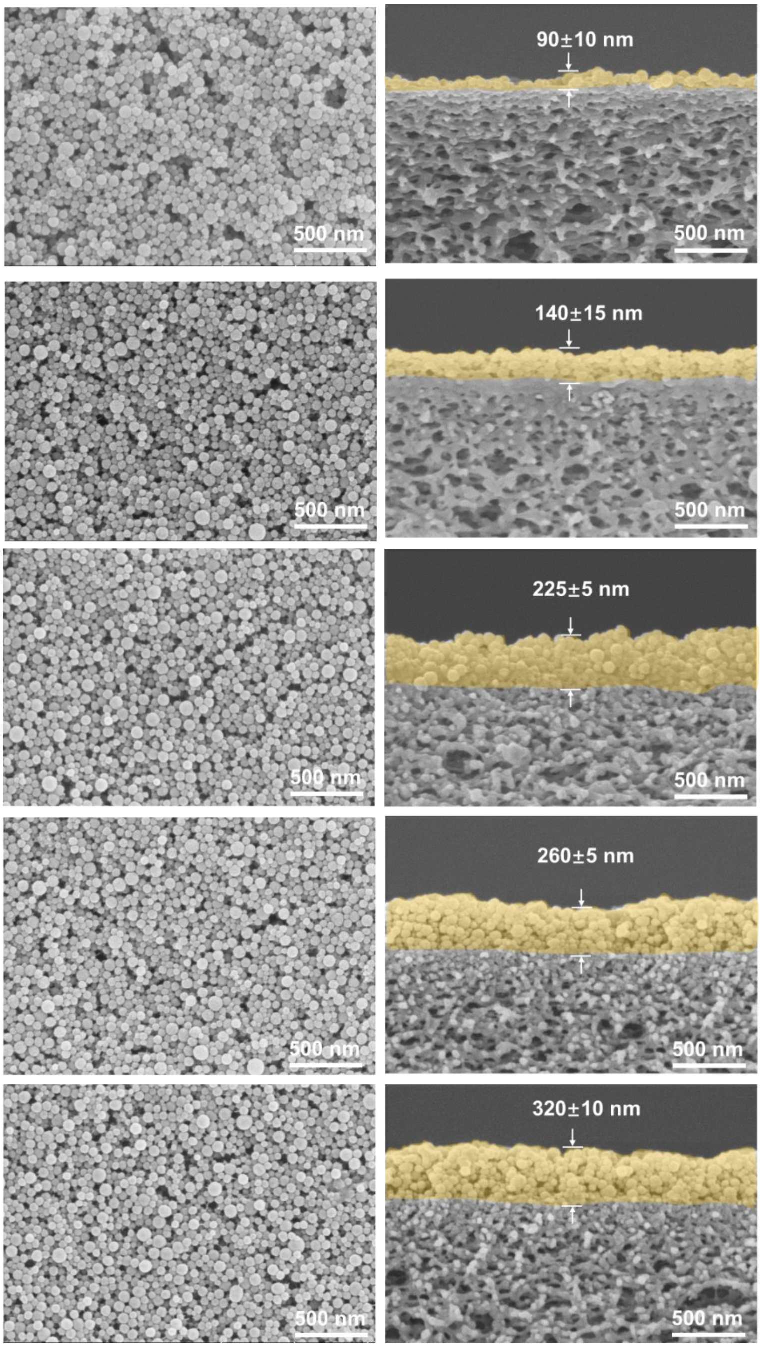

Fig. S11 FESEM surface and cross-section images of TPOP-CSM membranes preparing with 200 mL, 300 mL, 400 mL, 500 mL and 600 mL TPOP-CS dispersion solution (the mass ratio of TPOP and CS is 10:1).

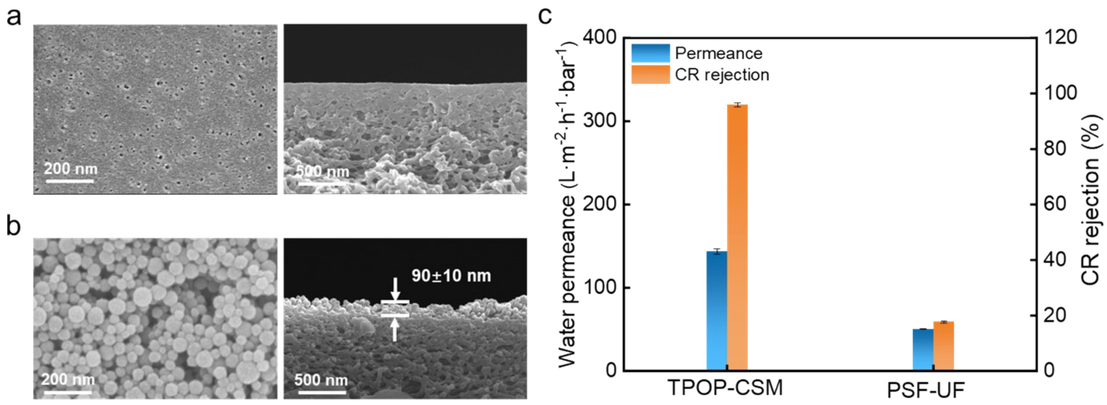

Fig. S12 FESEM surface and cross-section images of (a) PSF-UF supporting membrane and (b) TPOP-CSM membranes preparing with 200 mL TPOP-CS dispersion solution, (c) Separation performance of PSF-UF supporting membrane (the separation performance was obtained after the CR molecules adsorption saturation) and TPOP-CSM membrane preparing with 200 mL TPOP-CS dispersion solution. Test with 0.1 g·L^{-1} CR aqueous solution at 25 °C under 2.0 bar.

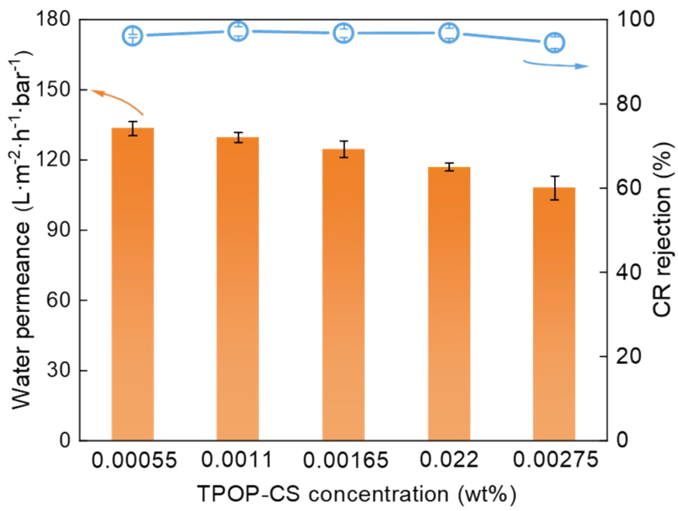

Fig. S13 Separation performance of TPOP-CSM membranes preparing with different concentration of TPOP-CS dispersion solution (300 mL, the mass ratio of TPOP and CS is 10:1). Test with 0.1 g·L^{-1} CR aqueous solution at 25 °C under 2.0 bar.

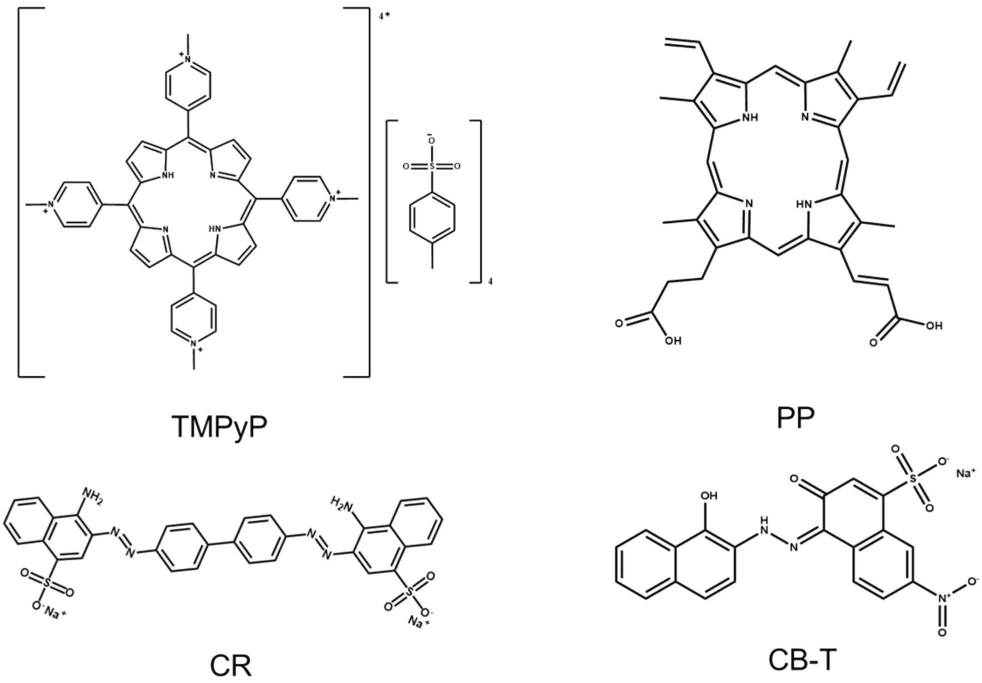

Fig. S14 Chemical structure of dye molecules.

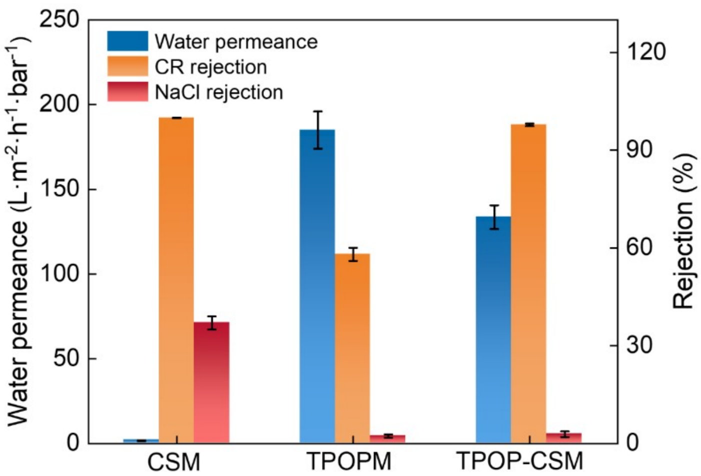

Fig. S15 Water permeance, CR and NaCl rejection of TPOP-CSM, TPOPM and CSM membranes. Test with 1.0 g·L^{-1} NaCl and 0.1 g·L^{-1} CR mixture aqueous solution at 25 °C and 2.0 bar.

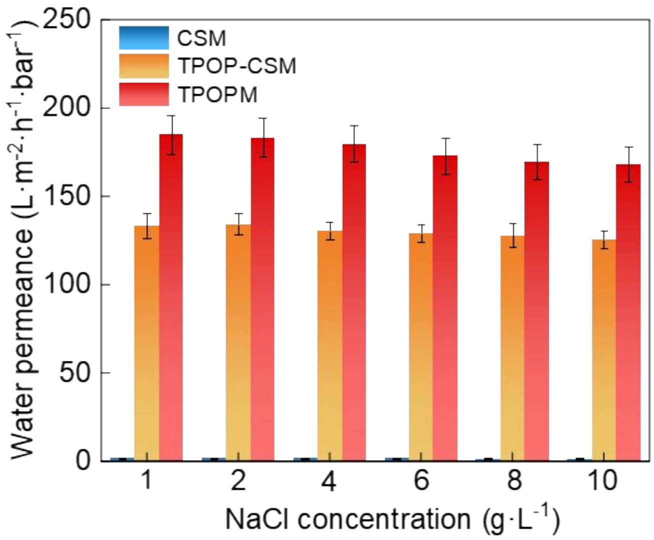

Fig. S16 Effect of NaCl concentration on the water permeance of TPOP-CSM, TPOPM and CSM membranes. Test with 0.1 g·L^{-1} CR and NaCl mixture aqueous solution at 25 °C and 2.0 bar.

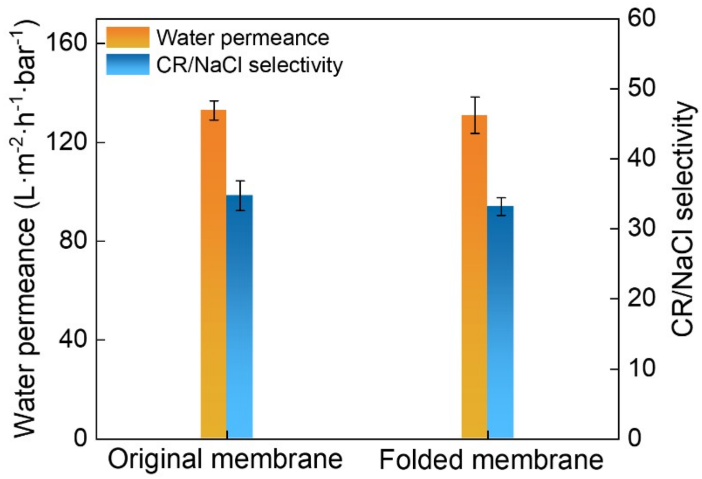

Fig. S17 Water permeance, CR/NaCl selectivity of original TPOP-CSM, and folded TPOP-CSM membranes. Test with 1.0 g·L^{-1} NaCl and 0.1 g·L^{-1} CR mixture aqueous solution at 25 °C and 2.0 bar.

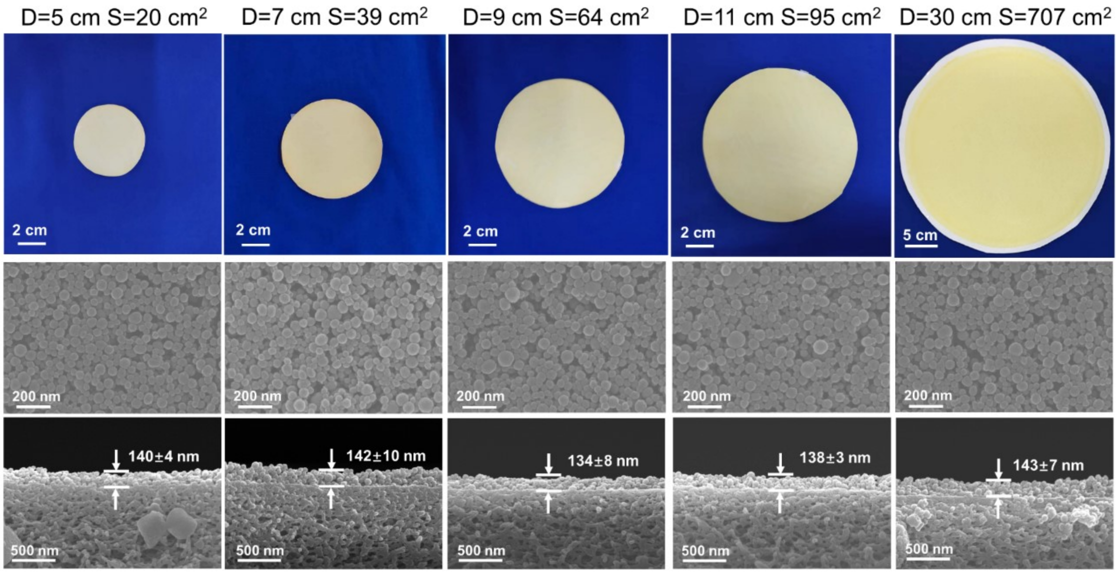

Fig. S18 Optical photographs, SEM surface and cross-section images of different sizes of TPOP-CSM membranes.

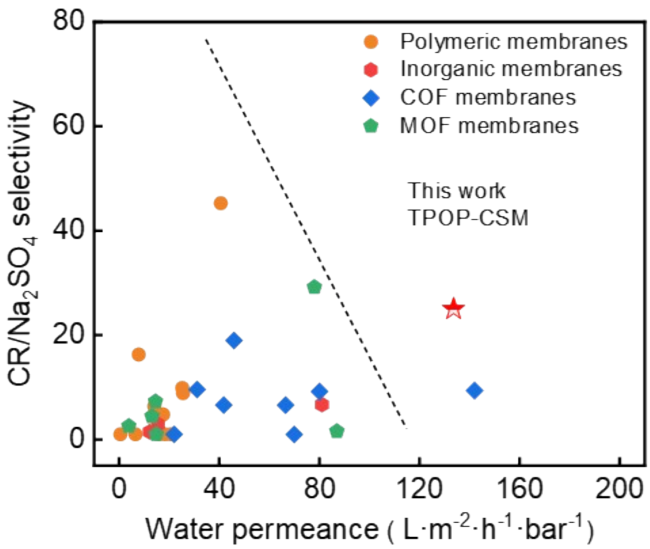

Fig. S19 Comparison TPOP-CSM separation performance with polymeric/inorganic nanofiltration membranes, COFs and MOFs nano-based membranes reported in the literature in consideration of water permeance and CR/Na_2SO_4 selectivity.

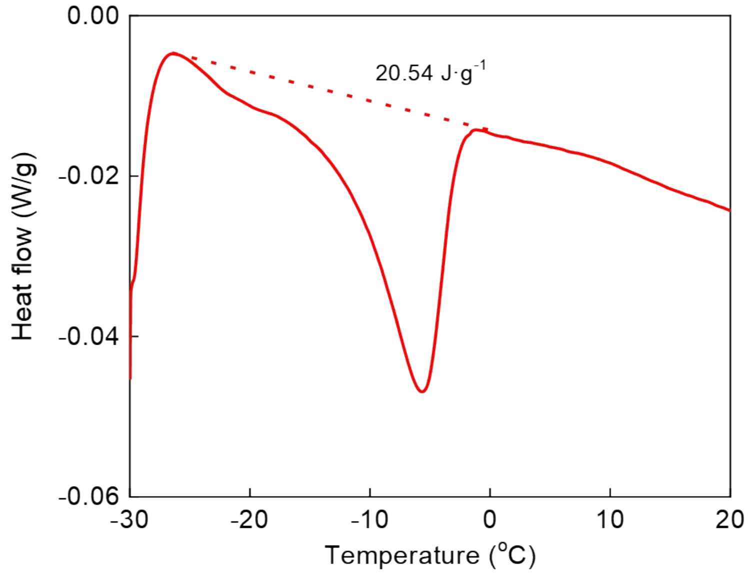

Fig. S20 DSC thermogram of TPOP-CSM freestanding film.

Note: The membrane surface porosity was determined by the DSC method. It is seen that the shape endothermic peak is at -5.6 °C due to the melting of frozen water. According to the variation of enthalpy, the water content of TPOP-CSM freestanding film is estimated to be 6.03 %.

The simulated water flux of TPOP-CSM membrane could be calculated based on the Hagen-Poiseuille equation: J=$\varepsilon\pi r_p{}^2\Delta P/8\mu L$ (S1)

Where the water viscosity μ is 8.9×10^{-4} Pa s, the surface porosity ε is 6.03 %, the membrane pore radius r_p (0.65 nm), the pressure drop ΔP (100 kPa), and the film thickness L is ~140 nm. Thus, the predicted water permeance for the TPOP-CSM membrane was calculated to be 28.9 L·m^{-2}·h^{-1}·bar^{-1}.

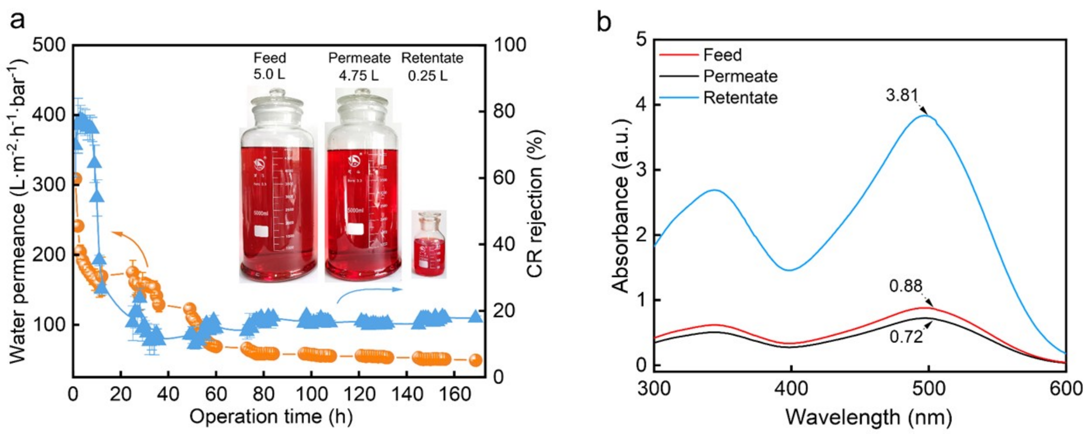

Fig. S21 (a) Water permeance and CR rejection of PSF-UF as a function of operation time (test with 0.1 g·L^{-1} CR aqueous solution for 12 h at 25 °C and 2.0 bar (solid symbol), and then clean with water for 12 h (only line), the operation was repeated for 7-time cycles). and the optical photographs of feed, permeate and retentate solution for the last seventh cycle test. (b) The UV-vis spectra of feed, permeate and retentate solution for the last seventh cycle test.

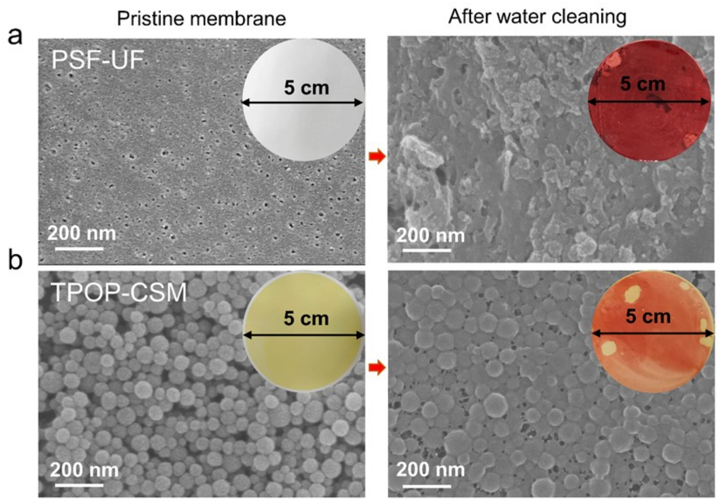

Fig. S22 The optical photographs and FESEM surface images of (a) PSF-UF and (b) TPOP-CSM membranes (left: pristine membrane, right: after water cleaning membrane).

Table. S1 BET and Langmuir specific surface area (SSA), and pore size of TPOP and TPOP-CS nanoparticles.

Sample name	SSA BET ($m^2 \cdot g^{-1}$)	SSA Langmuir ($m^2 \cdot g^{-1}$)	Pore size (nm)
TPOP	241.4	825.7	~1.4
TPOP-CS	182.0	786.7	~1.3

Table. S2 Size of dye molecules and diameter of hydrated ions.

Dye molecules and salt ions	Molecules/ions size (nm)
TMPyP	1.52×1.59
PP	1.45×1.54
CR	1.1×2.2
CB-T	0.88×1.55
Mg^{2+}	0.86
Li^{+}	0.76
Na^{+}	0.72
K^{+}	0.66

Note: the size of dye molecules and salt ions was measured by Materials Studio 8.0, and cited from references.[1,2]

Table S3 The preparation method, thickness, surface area and separation applications of nano-assembled membranes reported in the literature.

Membrane	Preparation method	Thickness (um)	Surface area (cm^2)	Separation	Ref
(1)TpEBr@TpPa-SO_3Na	LBL assembly	0.04	3	Gas molecules	3
(2) ICOM@TCBP	Self-assembly	100	3	QDs	4
(3) Mxene	Self-assembly	0.37	3	Dye molecules	5
(4) MOF	Self-assembly	1.6	4	Oil/water	6
(5) EB-COF:Br	Self-assembly	0.16	5	Dye molecules	7
(6) GO	Self-assembly	5	5	Salts	8
(7) FS-COM-2	Self-assembly	14	7	Salts	9
(8)COF@rGO	Self-assembly	0.2	8	Dye molecules	10
(9) MXene	Self-assembly	0.7	9	Dye molecules	11
(10) MXene	Self-assembly	2	10	Gas molecules	12
(11) GO@PAA	Self-assembly	1	10	Dye molecules	13
(12) COF-1	Self-assembly	0.1	11	Gas molecules	14
(13) GO/gC_3N_4@TiO_2	Self-assembly	0.25	13	Oil/water	15
(14) COF-CNF	Self-assembly	/	16	Salts	16
(15)2D COF	Self-assembly	0.03	20	Water/alcohol	17
(16) PAMAM/GO	Self-assembly	0.18	20	Water/n-butanol	18
(17) PDA/PEI	Self-assembly	0.2	24	Oil/water	19
(18) POP–GO	Self-assembly	45	33	Gas molecules	20
(19) GO	Self-assembly	0.2	88	Salts	21
TPOP-CS	Self-assembly	0.14	707	Dye molecules/salts	This work

Table S4. Comparison of the raw material price, nanomaterial synthesis/membrane preparation method, and the resultant membrane diameter, thickness and price in literatures and this work.

Membrane	Raw material Price	Nanomaterial synthesis method	Membrane preparation method	Membrane diameter (cm)	Membrane thickness (um)	Membrane price ($ m^{-2})	Ref.
MOF-NS	$Zn(NO_3)_2 \cdot 6H_2O$ ~$4.1 g^{-1} mIM ~$0.04 g^{-1} HCOONa ~$0.03 g^{-1}	Vigorous stirring. Vacuum oven	Controlled growth followed by a surface coating	6	6	~8000	22
$Zn_2(bim)_4$ MSN	$ZnCl_2$ ~$0.14 g^{-1} bIm ~$0.06 g^{-1} $Zn(NO_3)_2 \cdot 6H_2O$ ~$4.1 g^{-1}	React at 130 °C for 42 h in high pressure reactor	Vacuum filtration	0.5	10	~1300	23
FS-COM-2	TFP ~$86 g^{-1} Sa ~$6 g^{-1} TAPA ~$18 g^{-1}	React at 120 °C for 72 h.	Self-assembly	3	12	~500	9
COFDT-*Ex*	TAPB ~$49 g^{-1} $Sc(CF_3SO_3)_3$ ~$7.8 g^{-1} DOBDA~$1 g^{-1}	React for 96 h	Interfacial polymerization	2	0.2	~100000	24
GO	~$5.5 g^{-1}	Ultrasonic stripping	Ion cross-linking	2	0.28	~10	25
GO	~$5.5 g^{-1}	Ultrasonic stripping	Epoxy cross-linking	3	100	~1300	26
MoS_2	MoS_2 ~$1.4 g^{-1} C_4H_9Li ~$1 ml^{-1}	Ultrasonic stripping	Self-assembly	2	1	~40	27
MoS_2	MoS_2 ~$0.2 g^{-1} Peptide ~$136.8 g^{-1}	Ultrasonic stripping	Peptide-decoration	2.5	1	~1300	28
CNT	~$3.7 g^{-1}	Ultrasonic	Interfacial self-assembly	20	~0.35	~7	29
CNT	~$4.1 g^{-1}	Ultrasonic	Vacuum filtration	4	1.2	~8	30
Mxene	~$8.2 g^{-1}	Etching Ultrasonication	Ion intercalated	2	0.37	~8	5
Mxene	~$8.2 g^{-1}	Etching Ultrasonication	Ion cross-linking	2	0.2	~7	31
MXene	~$8.2 g^{-1}	Etching Ultrasonication	Self-assembly	3.5	2	~55	12
TPOP-CSM	TAPT ~$1.4 g^{-1} MG ~$0.12 g^{-1} FD ~$0.014 g^{-1} CS ~$0.14 g^{-1}	Debus-Radziszewski reaction at 80 °C for 12 h	Nanoparticle-Polymer Reticulation	30	0.14	~6	This work

Table. S5 Comparison of dye/salt separation performance in literatures and this work.

Membrane sample	Water permeance ($L \cdot m^{-2} \cdot h^{-1} \cdot bar^{-1}$)	Separation factor	Testing condition	Ref.
PEI/ALG/PSf	15.5	1.1	CR, NaCl, 0.2 MPa, 25 °C	32
PAN/Boltorn	11.0	8.0	CR, NaCl, 0.5 MPa, 25 °C	33
PEI/TA/PES	40.6	16.4	CR, NaCl, 0.4 MPa, 25 °C	34

PEI/TA/PES	40.6	45.3	CR, Na_2SO_4, 0.4 MPa, 25 ^oC	34
PAA/PVA/GA	4.2	32.0	CR, NaCl, 0.6 MPa, 25 ^oC	35
CMCNa/PP	0.8	49.8	CR, NaCl, 0.8 MPa, 25 ^oC	36
DEA-Modified PA-TFC	15.7	1.0	CR, Na_2SO_4, 0.5 MPa, 25 ^oC	37
DEA-Modified PA-TFC	15.7	2.0	CR, NaCl, 0.5 MPa, 25 ^oC	37
Sericin–TMC	6.5	2.4	CR, NaCl, 0.5 MPa, 25 ^oC	38
Sericin–TMC	6.5	1.0	CR, Na_2SO_4, 0.5 MPa, 25 ^oC	38
PEI-GA/PAN	25.5	4.9	CR, NaCl, 0.2 MPa, 25 ^oC	39
PEI-GA/PAN	25.5	8.9	CR, Na_2SO_4, 0.2 MPa, 25 ^oC	39
Arg-PIP-TMC	17.6	4.8	CR, Na_2SO_4, 0.2 MPa, 25 ^oC	40
PEI/CMCNa/PP	14.0	2.7	CR, NaCl, 0.3 MPa, 25 ^oC	41
PEI/CMCNa/PP	14.0	6.4	CR, Na_2SO_4, 0.3 MPa, 25 ^oC	41
PAEK-COOH	25.2	9.9	CR, Na_2SO_4, 0.4 MPa, 25 ^oC	42
ePDA-10	7.8	49	CR, NaCl, 1.5 MPa, 25 ^oC	43
ePDA-10	7.8	16.3	CR, Na_2SO_4, 1.5 MPa, 25 ^oC	43
CNCs/PIP/TMC	20.5	2.7	CR, NaCl, 0.6 MPa, 25 ^oC	44
CNCs/PIP/TMC	20.5	1.0	CR, Na_2SO_4, 0.6 MPa, 25 ^oC	44
Co-NF-2	18.2	1.2	CR, NaCl, 0.8 MPa, 25 ^oC	45
Co-NF-2	18.2	1.0	CR, Na_2SO_4, 0.8 MPa, 25 ^oC	45
(PEI/GO)/PAA/PVA/GA	0.8	2.7	CR, NaCl, 0.5 MPa, 25 ^oC	46
GO-TETA/PA	12.2	3.0	CR, NaCl, 0.6 MPa, 25 ^oC	47
GO-TETA/PA	12.2	1.5	CR, Na_2SO_4, 0.6 MPa, 25 ^oC	47
GO/NH_2-Fe_3O_4	15.6	6.2	CR, NaCl, 0.5 MPa, 25 ^oC	48
GO/NH_2-Fe_3O_4	15.6	2.9	CR, Na_2SO_4, 0.5 MPa, 25 ^oC	48
MXene/PES	81.0	7.0	CR, NaCl, 0.1 MPa, 25 ^oC	49
MXene/PES	81.0	6.7	CR, Na_2SO_4, 0.1 MPa, 25 ^oC	49
BUT-7(A)/PEI-HPAN	68.3	10.6	CR, NaCl, 0.5MPa, 25 ^oC	50
PEM/UiO-66	14.8	1.0	CR, $MgSO_4$, 0.8 MPa, 25 ^oC	51
BUT-203/PEI-HPAN	87.0	1.6	CR, Na_2SO_4, 0.1 MPa, 25 ^oC	52
MIL-101/MPD/TMC	3.9	2.6	CR, Na_2SO_4, 1.0 MPa, 25 ^oC	53
PVA-PEI- Zn(Ⅱ)TFC	14.5	7.3	CR, Na_2SO_4, 5 Mpa, 25 ^oC	54
PVP-UiO-66/HPAN	13.1	19.2	CR, NaCl, 0.4 Mpa, 25 ^oC	55
PVP-UiO-66/HPAN	13.1	4.47	CR, Na_2SO_4, 0.4 Mpa, 25 ^oC	55
ZIF-8/PEI-HPAN	78.0	29.2	CR, Na_2SO_4, 0.1 MPa, 25 ^oC	56
Cellulose acetate	0.5	1.5	CR, NaCl,0.5 MPa, 25 ^oC	57
Cellulose acetate	0.5	1.0	CR, Na_2SO_4,0.5 MPa, 25 ^oC	57
COF- LZU1/Al_2O_3	53.4	21.9	CR, NaCl, 0.5 MPa, 25 ^oC	58
TpTGCl@CNFs-4/PAN	70.0	1.0	CR, Na_2SO_4, 0.2 MPa, 25 ^oC	16
TpPa-1/HPAN	41.8	6.6	CR, Na_2SO_4, 0.1 MPa, 25 ^oC	59

GO/COF-1	31.1	9.6	CR, Na_2SO_4, 0.4 MPa, 25 °C	60
COF-LZU1/PES	80	16.6	CR, NaCl, 0.2 MPa, 25°C	61
COF-LZU1/PES	80	9.2	CR, Na_2SO_4, 0.2 MPa, 25°C	61
PA/TpPa/PAN	22	1.0	CR, Na_2SO_4, 0.5MPa, 25 °C	62
PA/TpPa/PAN	22	4.6	CR, NaCl, 0.5MPa, 25 °C	62
COF-TpPa1/PVDF	45.9	30.9	CR, NaCl, 0.2 MPa, 25°C	63
COF-TpPa1/PVDF	45.9	19	CR, Na_2SO_4, 0.2 MPa, 25°C	63
POP/TFC	66.5	6.6	CR, Na_2SO_4, 0.4 MPa, 25 °C	64
ACOF-1/HPAN	142.0	9.4	CR, Na_2SO_4, 0.4 MPa, 25 °C	65
ZIF-8@ZNPM	192.4	30.9	CR, NaCl, 0.1 MPa, 25 °C	66
GO-TA-APTES	111.7	12	CR, NaCl, 0.1 MPa, 25 °C	67
TPOP-CSM	133.6	35	CR, NaCl, 0.2 MPa, 25 °C	This work
TPOP-CSM	132.5	25	CR, Na_2SO_4, 0.2 MPa, 25 °C	This work

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