

Supporting Information for

## Aqueous Two-Phase Interfacial Assembly of COF Membranes for Water Desalination

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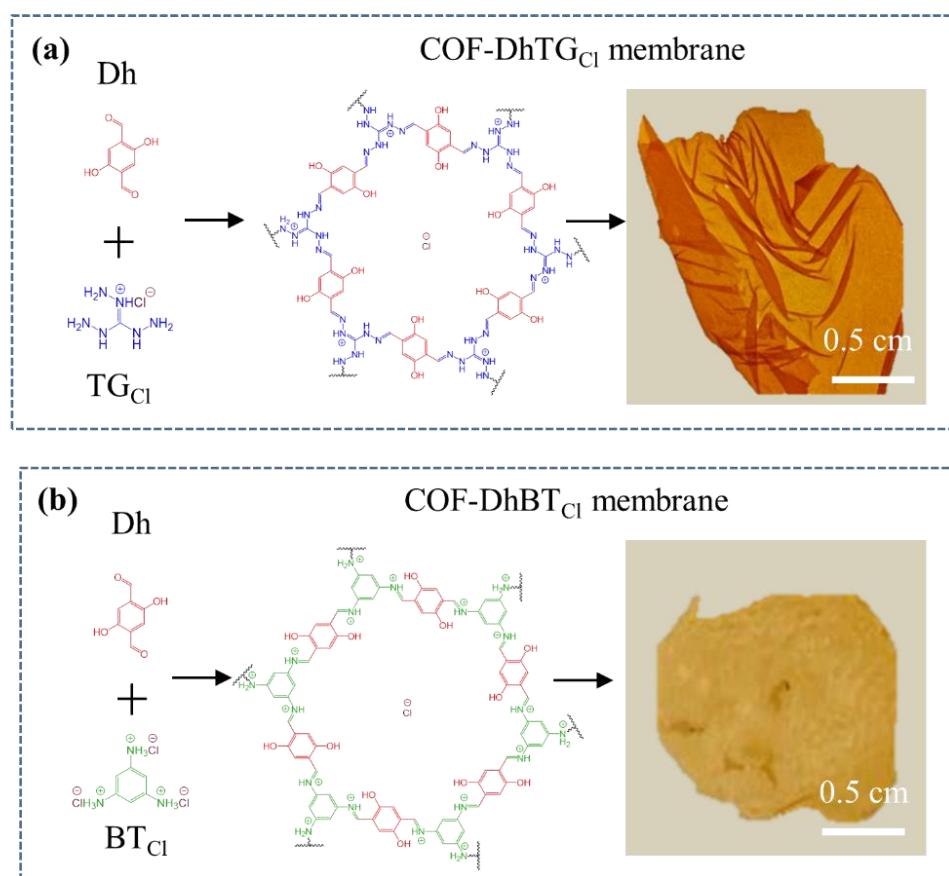
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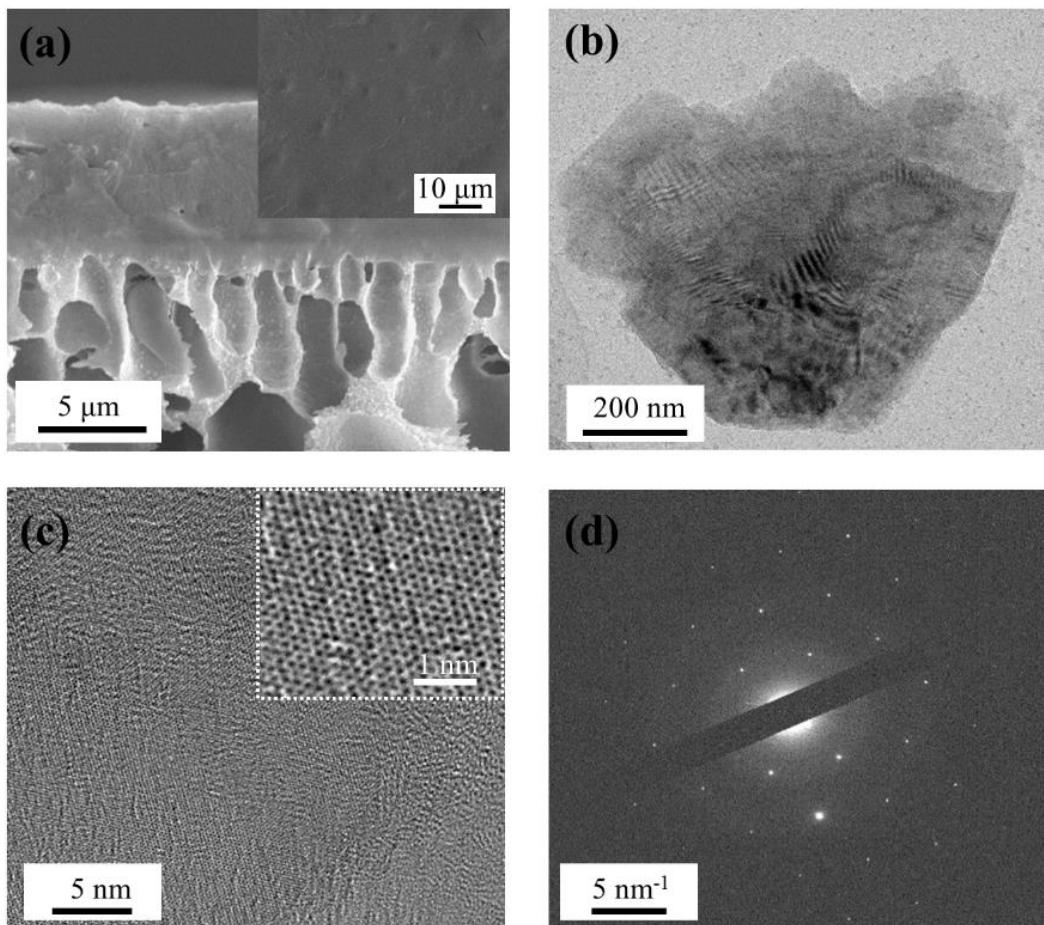
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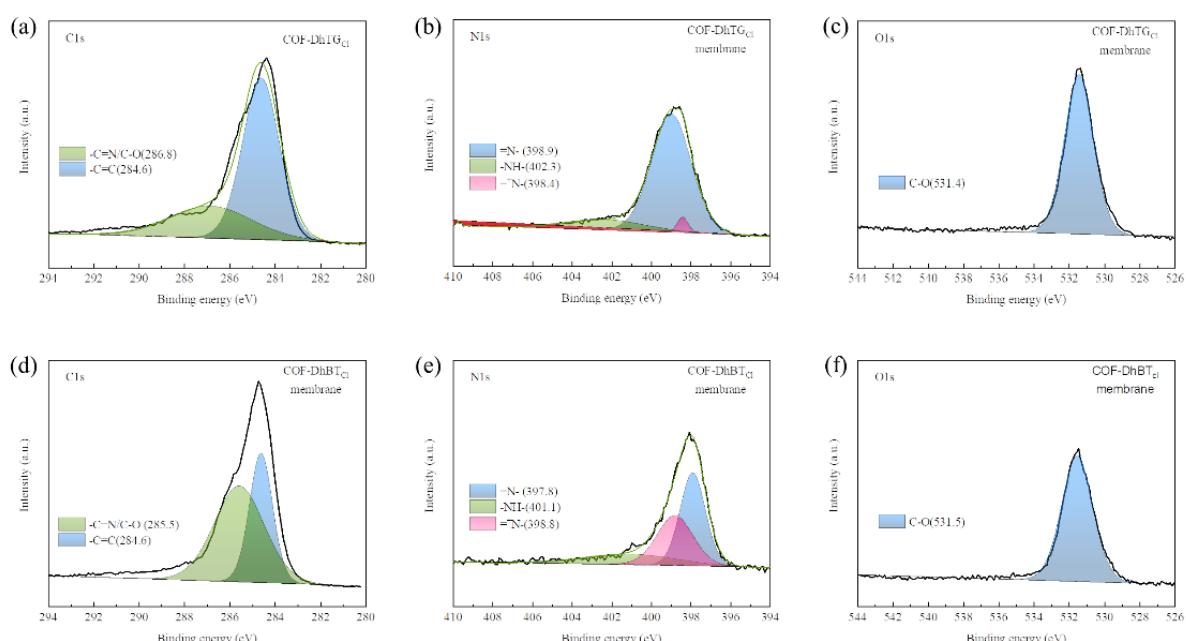
### Supplementary Figures and Tables



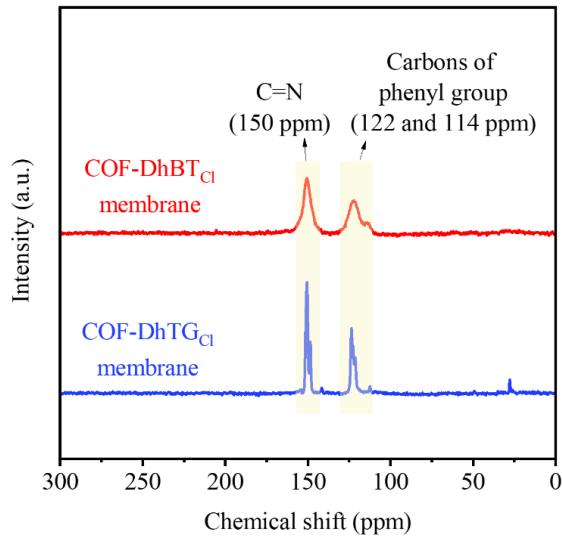
**Fig. S1** Schematic illustration and digital photos of **a** COF-DhTG<sub>Cl</sub> and **b** COF-DhBT<sub>Cl</sub> membranes



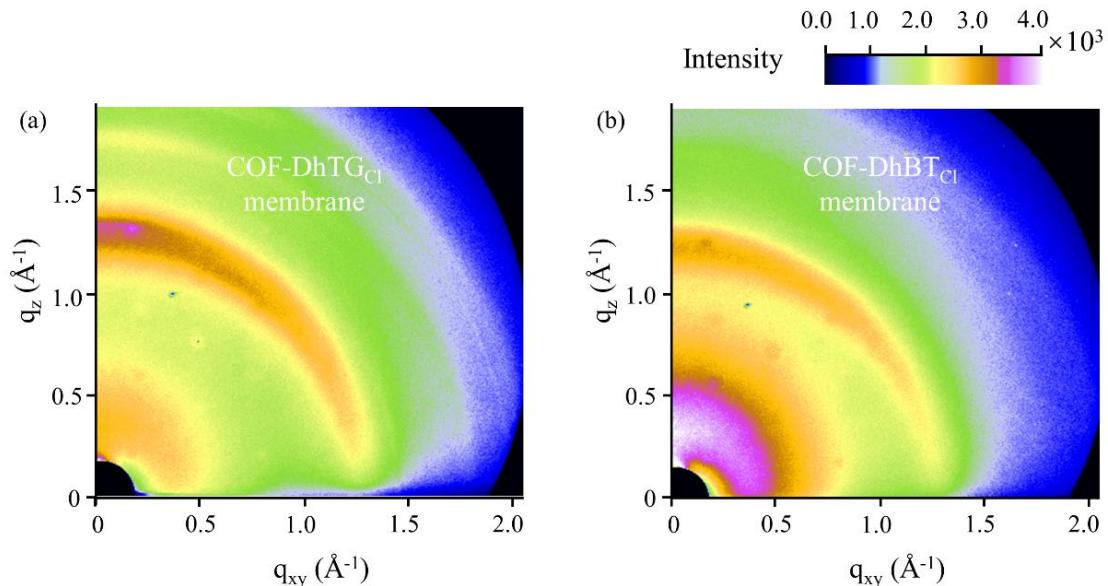
**Fig. S2** **a** SEM, **b-c** TEM and **d** SAED measurements on COF-DhBT<sub>Cl</sub> membrane



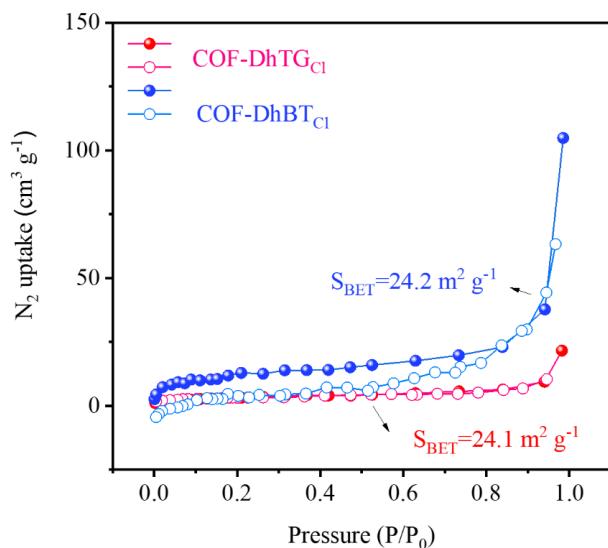
**Fig. S3** XPS spectra of COF membranes. **a** C 1s, **b** N 1s and **c** O 1s spectra of COF-DhTG<sub>Cl</sub> membrane. **d** C 1s, **e** N 1s and **f** O 1s spectra of COF-DhBT<sub>Cl</sub> membrane



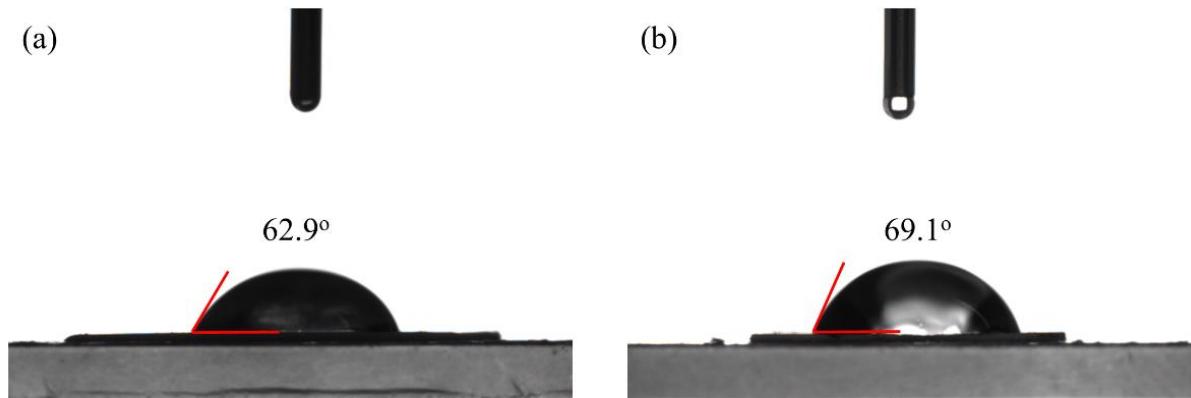
**Fig. S4** ssNMR spectra of COF-DhTG<sub>Cl</sub> and COF-DhBT<sub>Cl</sub> membranes



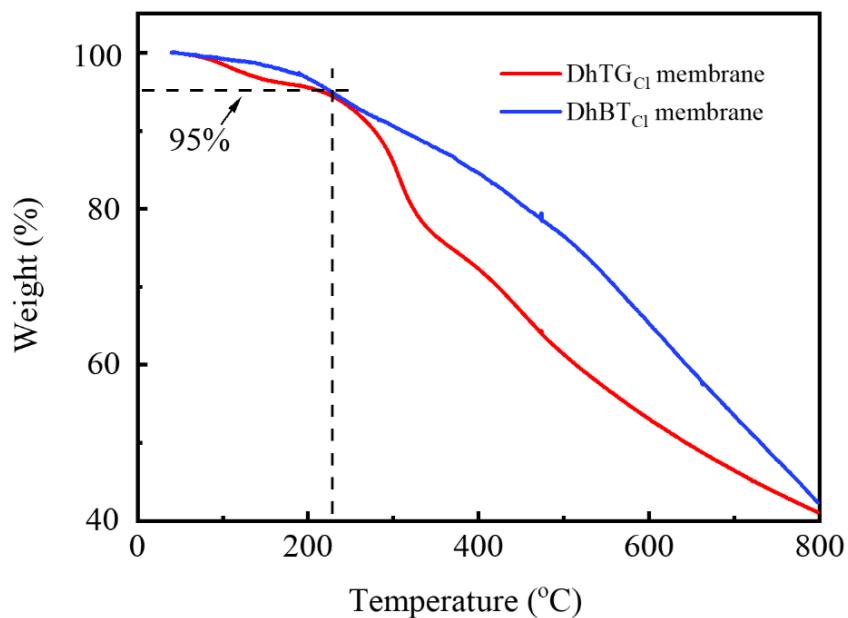
**Fig. S5** 2D-GIWAXS spectra of **a** COF-DhTG<sub>Cl</sub> and **b** COF-DhBT<sub>Cl</sub> membranes



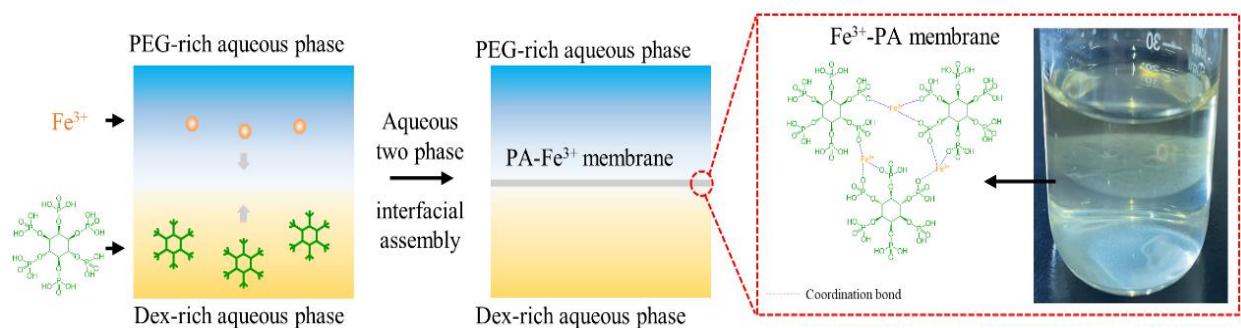
**Fig. S6** N<sub>2</sub> adsorption measurement of COF membranes



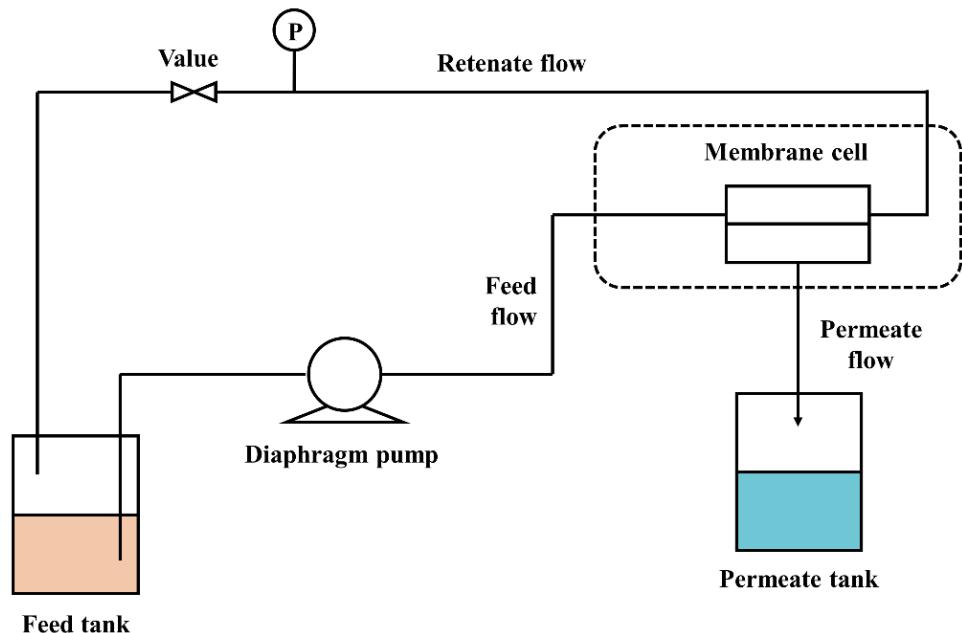
**Fig. S7** WCA measurement of **a** COF-DhTG<sub>Cl</sub> and **b** COF-DhBT<sub>Cl</sub> membranes, respectively



**Fig. S8** TGA measurement of COF membranes



**Fig. S9** Scheme illustration of fabricating metal organic polymer (metal-organophosphate) membranes using aqueous two-phase interfacial assembly



**Fig. S10** Apparatus of nanofiltration desalination with cross-flow configuration

**Table S1** Zeta potential of COF membranes

Membrane	Zeta potential ( $\xi$ , mV)
COF-DhTG <sub>Cl</sub>	20.1 ± 1.2
COF-DhBT <sub>Cl</sub>	9.8 ± 1.8

**Table S2** Monomer concentration in the recipe for COF-DhTG<sub>Cl</sub> membrane fabrication

Name	Dh concentration ( $\mu\text{mol mL}^{-1}$ )	TG <sub>Cl</sub> concentration ( $\mu\text{mol mL}^{-1}$ )
M1	0.4	0.25
M2	0.6	0.4
M3	0.8	0.5
M4	1.0	0.7

**Table S3** Phase composition and interfacial properties [S1, S2] of ATPS in this study

Name	PEG fraction in solution A (wt%)	DEx fraction insolution B (wt%)	Total polymer fraction (wt%)	Interfacial tension ( $\text{mN m}^{-1}$ )
1	2.1	2.6	2.30	0.001
2	5.0	6.4	5.62	0.012
3	10	16	12.6	0.103
4	15	19.2	16.3	0.209
5	20	25.6	21.4	0.381

**Table S4** Nanofiltration/reverse osmosis/forward osmosis desalination (NaCl rejection) performances of membranes in literatures

Membrane	Filtration method	Pressure (bar)	Water permeation ( $\text{L m}^2 \text{ h}^{-1} \text{ bar}^{-1}$ )	NaCl rejection (%)	Refs.
Graphene/GO	Cross flow	50	0.34 ( $\pm 0.1$ )	85 ( $\pm 2$ )	[S3]
Graphene/GO	Cross flow	50	0.22 ( $\pm 0.1$ )	54 ( $\pm 5$ )	[S3]
Graphene/GO	Cross flow	50	0.46 ( $\pm 0.2$ )	85 ( $\pm 1$ )	[S3]
Graphene/GO	Cross flow	50	0.38 ( $\pm 0.1$ )	85 ( $\pm 7$ )	[S3]
Graphene/GO	Cross flow	50	0.44 ( $\pm 0.02$ )	83 ( $\pm 5$ )	[S3]
Graphene/GO	Cross flow	50	0.67 ( $\pm 0.02$ )	79 ( $\pm 8$ )	[S3]
Laminated GO	Dead end	2	4	25	[S4]
GO/TMC	Dead end	3.4	~50	19	[S5]
GO/CNT	Dead end	5	5.5	59	[S6]
GO/PECs	Cross flow	5	0.80	43	[S7]
GO	Dead end	1.5	16.9	50.1	[S8]
GO	Forward osmosis	0.28	0.068 ( $\pm 0.007$ )	90	[S9]
GO	Forward osmosis	0.28	0.029 ( $\pm 0.005$ )	90	[S9]
GO/graphene	Forward osmosis	1	0.035	~94	[S10]
GO/graphene	Forward osmosis	5	0.007	97	[S10]
GO	Forward osmosis	5	0.0084	60	[S10]
Graphene	Dead end	1	20	40	[S11]
Graphene/CNT	Dead end	5	11.3	51	[S6]
Graphene/CNT	Cross flow	5	12.1	39.6	[S6]
Graphene	Dead end	5	3.26	42	[S11]
MoS <sub>2</sub>	Dead end	9	33.7 ( $\pm 13.5$ )	82.5 ( $\pm 6$ )	[S12]
MoS <sub>2</sub>	Dead end	9	1.6 ( $\pm 0.53$ )	87.6 ( $\pm 5.2$ )	[S12]
MoS <sub>2</sub>	Dead end	9	6.7 ( $\pm 1.35$ )	77.9 ( $\pm 17.4$ )	[S12]

## Supplementary References

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