Supporting Information

for

Polyamide-based membranes with structural homogeneity for ultrafast molecular sieving

Liang Shen\textsuperscript{a,h,1}, Ruihan Cheng\textsuperscript{c,l}, Ming Yi\textsuperscript{a,h,1}, Wei-Song Hung\textsuperscript{d,e}, Susilo Japip\textsuperscript{f}, Lian Tian\textsuperscript{a,b}, Xuan Zhang\textsuperscript{a,b}, Shudong Jiang\textsuperscript{g}, Song Li\textsuperscript{c,*} and Yan Wang\textsuperscript{a,b,*}

\textsuperscript{a} Key Laboratory of Material Chemistry for Energy Conversion and Storage (Huazhong University of Science and Technology), Ministry of Education, Wuhan, 430074, P.R. China

\textsuperscript{b} Hubei Key Laboratory of Material Chemistry and Service Failure, School of Chemistry and Chemical Engineering, Huazhong University of Science & Technology, Wuhan, 430074, P. R. China

\textsuperscript{c} School of Energy and Power Engineering, Huazhong University of Science & Technology, Wuhan, 430074, P. R. China

\textsuperscript{d} Graduate Institute of Applied Science and Technology, National Taiwan University of Science and Technology, Taipei, 10607, Taiwan

\textsuperscript{e} R&D Centre for Membrane Technology, Chung Yuan Christian University, Taoyuan, 32023, Taiwan

\textsuperscript{f} Department of Chemical & Biomolecular Engineering, National University of Singapore, 10 Kent Ridge Crescent, Singapore 119260, Singapore

\textsuperscript{g} College of Chemistry and Chemical Engineering, Anhui University, 111 Jiulong Road, Hefei, Anhui, 230601, P.R. China

\textsuperscript{1} these authors contribute equally to this work

* Corresponding authors: songli@hust.edu.cn (Song Li)
wangyan@hust.edu.cn (Yan Wang)
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1. Supplementary Methods

1.1 PALS characterization of as-fabricated TFC membranes

The detailed experiment procedure for PALS characterization is described below. A variable mono energetic slow positron beam was operated in the range of 0–30 keV positron incident energy (equivalent to a mean depth of 0–10 μm) at room temperature under a vacuum of ~10^-8 torr. The positron source is of a 50 mCi of \(^{22}\)Na radioisotope beam. Each membrane sample for PALS characterization was in area of 1*1 cm\(^2\). The as-fabricated membrane samples were measured by two positron annihilation techniques, including Doppler broadening of energy spectra (DBES) and positron annihilation lifetime (PAL) measurements. The DBES spectra were measured using a solid state HP Ge detector (EG & G Ortec) at a counting rate of approximately 3000 counts per second (cps). The total number of counts for each DBES was 1.0 million. The S parameter data from DBES was fitted by VEPFIT program. The PALS spectra were obtained by taking coincident events between two signals—the start signal detected by a multichannel plate from the secondary electrons and the stop signal discerned by a BaF\(_2\) lifetime detector from the annihilation photons at a counting rate of ~200-300 cps. A PALS spectrum contains 2.0 million counts. The positron lifetimes (t) and intensities (I) were determined by PATFIT program, and the lifetime distribution was obtained by MELT analysis.
2. Supplementary Figures

**Supplementary Figure 1.** NaCl concentration in the washing water of the MPD@NaCl membrane. The error bars represent the standard deviation and were calculated on the basis of at least three data points measured from different samples. It can be seen that NaCl concentration in the washing water sharply decreases after the first day immersion and gradually slows down in the following days. It can be deduced that a large amount of salt removed in the first day is generally the salt trapped in the substrate membrane as well as between the substrate membrane and the PA layer. However, after the first-day washing, the salt trapped in the PA layer can be partially washed out with a longer washing duration (5 days) due to the complexion between the salt ions and the carbonyl groups of the PA network.\textsuperscript{15-17,23,24}
Supplementary Figure 2. EDX characterization results. (a) (top) SEM images and (bottom) corresponding Na mapping images of modified membranes before and after washing; (b) atom percent of Na in the pristine and modified membranes before and after washing as examined by EDX analysis. It can be found that Na atom percentage in modified PA layers sharply decreases after washing.
Supplementary Figure 3. PAS results of modified membranes before and after washing. (a) NaCl@MPD, (b) NaHCO₃@MPD, (c) NaCl@PIP, and (d) NaHCO₃@PIP membranes. $S$ profiles as a function of positron incident energy of salt-modified membranes before and after washing. It can be seen that $S$ values of salt-modified membranes increase after washing, indicating the increased free volume. The increased free volume of NaCl-modified membrane is solely resulted from the removal of trapped NaCl salts. Instead, the increased free volume of NaHCO₃-modified membrane is contributed by both the removal of trapped inorganic salts and generated CO₂ nanobubbles by the reaction between NaHCO₃ and IP byproduct HCl. However, the later factor is believed to outweigh the former factor. NaHCO₃ in the reaction zone would be consumed by the byproduct HCl generating CO₂ as shown in Supplementary Equation 1. It can be seen that 1 mole NaHCO₃
consumed will generate 1 mole NaCl and 1 mole CO$_2$. Supplementary Figures 3b and 3d show that $S$ values of NaHCO$_3$-modified membranes also increase after washing, indicating the increased free volume resulted from the partial removal of generated NaCl. However, the trapped salt in the PA layer can be only partially removed according to the AAS result ($< 3\%$). Meanwhile, all generated CO$_2$ can create nanobubbles in the PA layer. Therefore, the increased free volume of NaHCO$_3$-modified membranes should be predominantly contributed by the nanobubbles of CO$_2$ generated.

$$\text{NaHCO}_3 + \text{HCl} = \text{NaCl} + \text{H}_2\text{O} + \text{CO}_2 \uparrow$$  \hspace{1cm} (1)
Supplementary Figure 4. Rejection test results of neutral organic solutes. (a) Rejection of neutral organic solutes with different molecular weights by the pristine salt-modified TFC membranes formed by PIP/TMC. (b) Pore size distribution of the pristine salt-modified TFC membranes formed by PIP/TMC.
Supplementary Figure 5. XPS characterization results. Wide-scan XPS spectra of the pristine and modified membranes. Membranes formed by (a) MPD and TMC, and (b) PIP and TMC. (c) In-depth profiles of membranes Pristine@MPD and NaCl@MPD. It can be found that peaks representing C, O, N atoms appear in all curves. Additionally, the peak of Na element is found in curves of modified membranes, and the peak of Cl element is noticed in the curve of NaCl-modified membrane. Meanwhile, Na content in NaCl@MPD membrane decreases with the deeper detection depth of the PA layer.
Supplementary Figure 6. PAS results of the pristine and modified membranes. $S$ profiles of the pristine and modified membranes as a function of the positron incident energy for (a) MPD/TMC and (b) PIP/TMC systems. Supplementary Figure 6 shows the $S$ profiles of the pristine and modified membranes as a function of positron incident energy (PIE). It can be observed that all profiles present the similar variation...
trend to PIE. In this study, the PIE ranges from 0.5 to 3 keV representing the PA selective layer. It can be found in Supplementary Figure 6a that $S$ values of modified membranes are lower than that of the pristine membrane, particularly for the NaHCO$_3$@MPD membrane, indicating the formation of a denser PA layer with small free volume. Similarly, the $S$ values of NaCl@PIP and NaHCO$_3$@PIP membranes are also lower than that of Pristine@PIP membrane as shown in Supplementary Figure 6b. The results suggest that the addition of inorganic salts does favor the formation of a dense PA layer.
Supplementary Figure 7. XRD patterns of the pristine and NaCl-modified TFC membranes. It can be seen that the characteristic peak at $2\theta = 17.7^\circ$ representing the polyamide in the curve of Pristine@MPD membrane shifts to $18.5^\circ$ in that of NaCl@MPD membrane, suggesting the smaller $d$-spacing of modified PA layer.
Supplementary Figure 8. MD simulation results. (a) The snapshots describe the diffusion behaviors of MPD molecules in the system with (right) and without (left) NaCl, respectively. It can be clearly found that ions tend to accumulate and surround MPD molecules gradually near the water-oil interface. In contrast, more MPD molecules are inclined to assemble at the interface without NaCl. (b) The number density ($\rho_N$) distribution of MPD (red line), H$_2$O (cyan line) and hexane (black line) along z direction in the system without NaCl. (c) The number density ($\rho_N$) distribution of MPD (red line), H$_2$O (cyan line), hexane (black line) Na$^+$ (blue line), and Cl$^-$ (green line) ions along z direction in the system with NaCl. The water/hexane interface width of the two MD systems was $\sim$70 Å according to the distribution of water and hexane molecules (Supplementary Figures 8b and 8c).
Supplementary Figure 9. Morphology properties of the pristine and modified membranes. (a) Surface SEM images, (b) cross-sectional SEM and (c) TEM images, (d) AFM topology images of NaHCO₃-modified membranes of the pristine and modified membranes.
Supplementary Figure 10. PA layer thickness based on TEM results and surface roughness of the pristine and modified membranes. The error bars represent the standard deviation and were calculated on the basis of at least ten data points measured from different samples. PA layer thickness is measured based on TEM images. It can be seen that both the thickness and roughness of salt-modified membranes sharply decrease compared to those of the pristine membrane.
Supplementary Figure 11. Water contact angle of the pristine and modified membranes. The error bars represent the standard deviation and were calculated on the basis of at least ten data points measured from different samples.
Supplementary Figure 12. Surface zeta potentials of the pristine and modified membranes as a function of pH. (a) MPD/TMC system, and (b) PIP/TMC system. It can be seen that Zeta potentials of modified membranes in both systems are lower than that of the pristine membrane at the same solution pH, indicating more negative charges on the surface of modified PA layers.
**Supplementary Figure 13.** FO performance of the pristine and modified membranes under FO mode (PA selective layer facing the feed solution) using DI water and 2 M NaCl as the feed and draw solutions, respectively. The error bars represent the standard deviation and were calculated on the basis of at least three data points measured from different samples. It can be seen that water fluxes follow the order of NaHCO$_3$@MPD > NaCl@MPD > Pristine@MPD, while the reverse salt fluxes present the opposite order, overcoming the permeability-selectivity tradeoff.
Supplementary Figure 14. Dynamic fouling test results of the pristine and modified membranes under FO model using the synthetic wastewater containing SA and Ca$^{2+}$ ions as the feed solution and 2 M NaCl solution as the draw solution. (a) Normalized water flux against the testing time. (b) Normalized water flux after fouling and after cleaning. The error bars represent the standard deviation and were calculated on the basis of at least three data points measured from different samples. It can be found that the flux of the pristine membrane drops severely and whose flux
recovery after cleaning is only around 70%. Instead, modified membranes exhibit the steadier and less flux decline as well as the higher flux recovery ratios (~91% to 93%), indicating the improved fouling resistance.
Supplementary Figure 15. SEM images of various salts modified membranes. (a) MPD/TMC monomer system; and (b) PIP/TMC monomer system. Supplementary Figure 15a shows that all modified membrane shows the honey-bomb-like Turing surface and thin PA layers. On the other side, Supplementary Figure 15b shows that all modified membranes show the nodular-like surface structures and thin PA layers.
Supplementary Figure 16. Water contact angles (WCA) of various salt-modified membranes. (a) MPD/TMC monomer system, (b) PIP/TMC monomer system, and (c) PEI/TMC monomer system. The error bars represent the standard deviation and were calculated on the basis of at least ten data points measured from different samples. It can be found that the WCA values of modified membranes are all smaller than that of the pristine membrane.
Supplementary Figure 17. FO performance of modified membranes formed by TMC/MPD. (a) Water fluxes and (b) reverse salt fluxes of various salt-modified membranes formed by MPD and TMC under FO and PRO modes using DI water and 2 M NaCl as the feed and draw solutions, respectively. The error bars represent the standard deviation and were calculated on the basis of at least three data points measured from different samples. It can be seen that both the water flux and reverse salt flux are influenced by the salt type added in the amine solution.
Supplementary Figure 18. $S$ profiles of different neutral salts modified membranes formed by PIP and TMC versus the positron incident energy. It can be found that $S$ values in range of 0.5 – 1.5 keV follows the order of LiCl@PIP < NaCl@PIP < KCl@PIP < NH$_4$Cl@PIP.
Supplementary Figure 19. MD simulation and neutral solute rejection test results.

(a) MSD profiles of MPD molecules in systems of pure MPD, MPD+NaCl, and MPD+NH₄Cl. (b) The number density (ρN) distribution of MPD molecules in the two systems of pure MPD, MPD+NaCl, and MPD+NH₄Cl. (c) Solute rejection as a function of solute molecular weight and (d) pore size distribution of membranes Pristine@PIP, NaCl@PIP, NH₂Cl@PIP.
Supplementary Figure 20. Separation performances of the pristine and various salts modified membranes formed by PEI and TMC. The error bars represent the standard deviation and were calculated on the basis of at least three data points measured from different samples. It can be seen that both the water permeance and NaCl rejection are higher than or comparable to those of the pristine membrane.
Supplementary Figure 21. FO performances of TFC membranes formed by MPD and TMC modified with various salt concentrations. (a) NaCl and (b) NaHCO$_3$-modified membranes using DI water and 2 M NaCl solution as the feed and draw solutions, respectively. The error bars represent the standard deviation and were calculated on the basis of at least three data points measured from different samples. Supplementary Figure 21a shows that the water flux increases initially (till 3 wt.%) and then decreases as the increase of NaCl content, while the reverse salt flux decreases and increases as a function of NaCl content. Supplementary Figure 21b shows that the water flux increases with the increasing NaHCO$_3$ content till 2 wt.% and then decreases with further increment of NaHCO$_3$ content, but the reverse salt flux presents a continuous decline with the increase of NaHCO$_3$ content. Results presented here suggest the optimized salt content is about 3 wt.%. Therefore, the salt content is fixed at 3 wt.% for other salts employed to prepare modified membranes.
## 3. Supplementary Tables

**Supplementary Table 1.** Summary of reported salt additives added in aqueous amine solution for fabricating TFC membranes.

| Additive category | Additives | Aqueous monomer | Key findings about salt effects | Refs. |
|-------------------|-----------|-----------------|--------------------------------|-------|
| **Organic salts** | TEBAB/TMBAB/TEBAC | MPD/ PIP | 1) Phase transfer catalysts improved the polymerization efficiency in interfacial polymerization by helping the monomer in the water phase move into the organic layer. | [1] |
| | CSA-TEA | MPD | 1) TEA acted as a catalyst to accelerate the MPD–TMC reaction by neutralizing HCl produced during amide formation.  
2) The addition of CSA protected the microporous skin layer of the support membrane from annealing during curing. | [2] |
| | TEA+SDS+BMIC/OMIC | PIP | 1) OMIC had a large polar head and a long hydrophobic tail in the structure, playing the role of a surfactant.  
2) BMIC as a phase transfer catalyst, not only improved the transfer mechanism across the interface, but also increased the diffusion rate. | [3] |
| | SDS+CTAB | PIP | 1) The porosity of thin layers decreased in the presence of CTAB and SDS.  
2) Surfactant-modified membranes showed a dense and compressed thin layer. | [4] |
| | TEAC/TBAB/ | PIP | 1) Amine salt containing larger steric configuration cationic amine group resulted | [5] |
| Additive                        | Description                                                                 |
|--------------------------------|-----------------------------------------------------------------------------|
| CSA-TEA/BMMIC                  | in a TFC membrane with better performance.                                  |
|                                | 2) The extent of cross-linking was increased with the increasing molecular weight of cationic amine groups. |
| TEAC/TBAB/CSA-TEA/BTMAC/BTEAC/DTMAC| 1) A phase transfer additive should have suitable organic structures and be loose enough to ensure a high capacity to absorb anionic or complex agent and acted as a phase transfer catalyst. |
|                                | 2) The ammonium salt with more and longer lipophilic alkyl groups showed the higher catalytic efficiency. |
|                                | 3) The larger ammonium salt complex would take over larger space in polymer, and result in a larger free volume. |
| Surfactant + amine salt        | Amine                                                                       |
|                                | The salt effect was not stated.                                             |
| Amine salt                     | Amine                                                                       |
|                                | The salt effect was not stated.                                             |
| Surfactant + amine salt        | Amine                                                                       |
|                                | The salt effect was not stated.                                             |
| Tertiary amine salt            | Amine                                                                       |
|                                | 1) Tertiary amine acted as both pore forming agent and catalyst (absorbing the IP reaction by-products). |
| Inorganic salts                | LiBr + SDS                                                                  |
|                                | (Alcohol amine)                                                             |
|                                | 1) The surface roughness and coverage varied with the variation of LiBr concentration. |
|                                | 2) Li⁺ ions induce an increased interaction coefficient between TMC and TEOA, |
|                                | [15]                                                                        |

[6] [7-9] [10] [11] [12-14]
forming a dense skin layer.

3) Complexation between Li+ ion and the carbonyl in TMC causes the hydrolysis of acid chloride groups of TMC, inducing a hydrophilic and loose surface layer.

| **CaCl₂** | **TEPA** | 1) The addition of CaCl₂ enhanced the interfacial tension and reduced the mass transfer of TEPA to the organic phase.  
   2) Higher CaCl₂ content induced an unevenly distributed interfacial polymerization reaction, resulting in the increased surface roughness. |
| **TEA+NaCl** | **PIP** | 1) The addition of salt to the aqueous phase increased the interfacial tension, depressing the mass transfer of PIP and constructing a loose PA layer.  
   2) Further increase NaCl content induced the uneven distribution in nanoscale of the IP reaction, leading to the morphological structure transition from the crisscrossed ridge networks to crowded nodular arrays. |
| **NaHCO₃** | **MPD** | 1) Under different pH conditions, different amounts of CO₂ nanobubbles were generated resulting in different morphological features and separation properties.  
   2) Under different concentration conditions, the resulting membranes showed different morphological features, thus tuning the roughness and separation properties. |
1) The growth of inorganic salt crystals stretched the nascent and flexible PA layer, and the inorganic salt crystals can sacrifice themselves in water, contributing to the formation of a thin and rough PA layer with the crumpled nanostructure.

2) The density and size of NaCl crystals increase with the increasing NaCl concentration, resulting in unidirectional scattering distribution to interconnected dendritic distribution. [20]
**Supplementary Table 2.** Surface elemental compositions of the NaHCO$_3$- and NaCl-modified membranes with and without washing by EDX analysis. It can be found that Na and/or Cl content in the NaHCO$_3$ and NaCl-modified FO membranes sharply decrease after washing.

| Code               | C    | O    | N    | Na   | Cl   |
|--------------------|------|------|------|------|------|
| NaHCO$_3$@MPD-unwashed | 52.88| 27.68| 13.78| 5.66 | /    |
| NaHCO$_3$@MPD-washed | 58.07| 26.92| 13.21| 1.80 | /    |
| NaCl@MPD-unwashed  | 61.32| 21.35| 11.38| 3.40 | 2.54 |
| NaCl@MPD-washed    | 64.64| 23.11| 10.92| 0.94 | 0.39 |
**Supplementary Table 3.** Na content after 1-day and 6-day washing by AAS detection of TFC membranes and free-standing PA films. It can be found that the salt content trapped in the PA layer after 1-day and 6-day washing decreases slightly from 0.0902/0.0888 to 0.0871/0.0856 μg/cm² of membranes NaCl@MPD/NaHCO₃@MPD, respectively. On the other side, the content of trapped salt in the free-standing PA layer (NaCl@MPD) reduces slightly from 161.455 mg/kg to 157.280 mg/kg for 1-day and 6-day washing. It can be deduced that a large amount of salt removed on the first day is generally the salt trapped in the substrate membrane as well as between the substrate membrane and the PA layer. However, after the first-day washing, the salt trapped in the PA layer can be partially washed out with a longer washing duration (6 days) due to the complexion between the salt ions and the carbonyl groups of the PA network\textsuperscript{15-17,23,24}. The ultimate salt content trapped accounts for 0.016% of the weight of the PA layer.

| Membrane code         | Na content (μg cm⁻²) | Film code                  | Na content (mg kg⁻¹) |
|-----------------------|----------------------|----------------------------|----------------------|
| NaCl@MPD-1 day        | 0.0902               | NaCl@MPD-1 day             | 161.455              |
| NaCl@MPD-6 day        | 0.0871               | NaCl@MPD-6 day             | 157.280              |
| NaHCO₃@MPD-1 day      | 0.0888               | /                          | /                    |
| NaHCO₃@MPD-6 day      | 0.0856               | /                          | /                    |
**Supplementary Table 4.** Surface elemental compositions of pristine and modified membranes by XPS analysis. Commonly, the detection depth of XPS technique for polymeric material is less than 10 nm[21], hence the detected O/N ratio actually only refers to the elemental composition of the top skin of the PA layer, which should be the region formed in the diffusion-limited growth regime (relatively loose part). To detect the middle part of the resulting PA layers, the in-depth profile of elemental compositions was therefore further measured by XPS using C$_{24}$H$_{12}^+$ ion source to etch membranes. The results in Supplementary Table 4 reveal that the modified membrane possesses a relatively looser top skin (high O/N ratio) compared to that of the pristine membrane. With a deeper detection depth, the position might reach the middle part of the PA layer in the modified membrane, exhibiting lower O/N ratios (higher crosslinking degree). In comparison with the pristine membrane, the O/N ratio of the modified membrane is even slightly lower than that of the pristine membrane, suggesting the formation of a relatively denser middle layer. Meanwhile, Na content in the modified membrane decreases with the deeper detection depth.

| Membrane code     | C    | O    | N    | Na   | Cl   | O/N  |
|-------------------|------|------|------|------|------|------|
| Pristine@MPD      | 71.84| 16.83| 11.32| /    | /    | 1.49 |
| NaHCO$_3$@MPD     | 69.25| 18.51| 11.26| 0.98 | /    | 1.64 |
| NaCl @MPD         | 71.15| 15.79| 9.93 | 1.66 | 1.47 | 1.59 |
| Pristine@PIP      | 72.92| 15.36| 11.72| /    | /    | 1.31 |
| NaHCO$_3$@PIP     | 71.3 | 16.74| 10.82| 1.14 | /    | 1.55 |
| NaCl@PIP          | 70.42| 17.17| 11.28| 0.37 | 0.76 | 1.52 |
| Pristine@MPD-0nm  | 72.53| 16.59| 10.88| /    | /    | 1.53 |
| Pristine@MPD-20nm | 72.31| 16.16| 11.53| /    | /    | 1.40 |
| Material       | Thickness | Angle 1 | Angle 2 | Angle 3 | Angle 4 | Angle 5 | Angle 6 | Angle 7 |
|---------------|-----------|---------|---------|---------|---------|---------|---------|---------|
| Pristine@MPD  | 40nm      | 74.03   | 14.95   | 11.02   | /       | /       | 1.36    |
| Pristine@MPD  | 60nm      | 73.12   | 15.65   | 11.23   | /       | /       | 1.39    |
| Pristine@MPD  | 80nm      | 73.39   | 15.03   | 11.58   | /       | /       | 1.30    |
| NaCl@MPD      | 0nm       | 70.71   | 15.93   | 10.25   | 1.38    | 1.73    | 1.55    |
| NaCl@MPD      | 20nm      | 72.40   | 14.56   | 11.13   | 0.73    | 1.18    | 1.31    |
| NaCl@MPD      | 40nm      | 72.58   | 14.62   | 11.46   | 0.47    | 0.87    | 1.28    |
| NaCl@MPD      | 60nm      | 72.32   | 14.89   | 11.77   | 0.40    | 0.62    | 1.27    |
| NaCl@MPD      | 80nm      | 72.69   | 14.85   | 11.58   | 0.29    | 0.59    | 1.28    |
**Supplementary Table 5.** Free volume pore properties of the pristine and modified membranes (error bars represent the standard deviation and were calculated on the basis of at least three data points measured from different samples). It summarizes the free volume pore properties of the pristine and modified membranes formed by MPD/TMC or PIP/TMC, including $o$-Ps lifetime ($\tau_3$), free volume pore intensity ($I_3$), free volume pore size ($R$), and fraction free volume ($FFV$). It can be found that $\tau_3$ and $R$ values of modified membranes are all smaller than that of the pristine membrane, indicating the smaller free volume pore size in the modified PA layers. In contrast, $I_3$ values of modified membranes are all larger than that of the pristine membrane, demonstrating more free volume pores in the modified PA layers. Accordingly, modified membranes show the smaller $FFV$ values, suggesting the denser structures of modified PA layers.

| Membrane Code   | $\tau_3$ (ns) | $I_3$ (%) | $R$ (Å) | $FFV$ (%) |
|-----------------|---------------|-----------|---------|-----------|
| Pristine@MPD    | 1.616±0.036   | 13.697±0.563 | 2.473±0.035 | 1.562±0.131 |
| NaCl@MPD       | 1.422±0.049   | 15.353±0.497 | 2.311±0.054 | 1.428±0.146 |
| NaHCO$_3$@MPD  | 1.396±0.042   | 16.874±0.462 | 2.183±0.050 | 1.323±0.127 |
| Pristine@PIP    | 1.922±0.051   | 12.971±0.368 | 2.783±0.041 | 2.107±0.153 |
| NaCl@PIP       | 1.537±0.038   | 16.108±0.417 | 2.386±0.040 | 1.649±0.125 |
| NaHCO$_3$@PIP  | 1.483±0.047   | 14.448±0.483 | 2.324±0.051 | 1.367±0.136 |
**Supplementary Table 6.** Polymer density of free-standing films of Pristine@MPD and NaCl@MPD measured by a pycnometer (error bars represent the standard deviation and were calculated on the basis of at least three data points measured from different samples).

| Film code       | Density (g cm⁻³) |
|-----------------|------------------|
| Pristine@MPD    | 1.183±0.017      |
| NaCl@MPD        | 1.238±0.023      |
**Supplementary Table 7.** Properties of different ions. Supplementary Table 7 summarizes intrinsic properties, such as diffusion coefficient, ionic radius and hydrated radius, of different ions employed in this work, including Li\(^+\), Na\(^+\), K\(^+\), NH\(_4\)^+, Cl\(^-\) and SO\(_4\)^{2-}. It can be found that the water permeance of the modified membrane is positively correlated to the ionic radius of the salt rather than the hydrated radius of the cationic ion.

| Ion Type | Diffusion coefficient \((10^{-9} \text{ m s}^{-1})\) | Ionic radius (nm) | Hydrated radius (nm) |
|----------|----------------------------------|-------------------|---------------------|
| Li\(^+\) | 1.029                            | 0.060             | 0.382               |
| Na\(^+\) | 1.334                            | 0.102             | 0.358               |
| K\(^+\)  | 1.957                            | 0.138             | 0.275               |
| NH\(_4\)^+ | 1.760                            | 0.161             | 0.331               |
| Cl\(^-\) | 2.030                            | 0.181             | 0.332               |
| SO\(_4\)^{2-}\ | 1.060                            | 0.230             | 0.379               |
**Supplementary Table 8.** FO performance comparison with reported lab-made and commercial FO membranes. 2 M NaCl solution and DI water were used as the draw and feed solutions under PRO mode.

| Membrane code         | $J_v$ (L m$^{-2}$ h$^{-1}$) | $J_s$ (g m$^{-2}$ h$^{-1}$) | Ref.    |
|-----------------------|-----------------------------|-----------------------------|---------|
| NaHCO$_3$@MPD        | 62.8                        | 0.4                         | This work |
| PA-2-HMTA             | 50.6                        | 7.5                         | [22]    |
| PA-AMPES/3            | 56.3                        | 9.5                         | [23]    |
| PA-PES/NMP            | 62.7                        | 10.3                        | [24]    |
| PA-PES/SPES           | 42.1                        | 11.1                        | [25]    |
| PA-PSF/HNT            | 54.0                        | 31.0                        | [26]    |
| PA-PEI-DTPMP          | 62.0                        | 23.0                        | [27]    |
| PA-SPPSU              | 48.0                        | 7.6                         | [28]    |
| M-T/CNT-PA            | 45.0                        | 7.0                         | [29]    |
| TFC-D3                | 60.5                        | 16.5                        | [30]    |
| PA-1-TAEA-9           | 57.9                        | 22.4                        | [31]    |
| PESU-co-sPPSU         | 33.0                        | 2.8                         | [32]    |
| TFN0.1                | 36.5                        | 1.5                         | [33]    |
| TFN0.6                | 59.4                        | 31.0                        | [34]    |
| HTI-TFC               | 24.0                        | 10.8                        | [35]    |
| HTI-CTA               | 18.5                        | 20.0                        |         |
**Supplementary Table 9.** Intrinsic separation performance comparison with reported lab-made and commercial FO membranes.

| Membrane code          | Water permeance (L m⁻² h⁻¹ bar⁻¹) | NaCl rejection (%) | Testing condition          | Ref.   |
|------------------------|-----------------------------------|-------------------|---------------------------|--------|
| NaHCO₃@MPD             | 2.9                               | 99.2              | 1000 ppm, 5 bar           | This work |
| M-T/CNT-PA             | 2.0                               | 98.0              | 200 ppm, 5 bar            | [29]   |
| TFC-G₁₀                | 2.1                               | 94.3              | 1000 ppm, 5 bar           | [36]   |
| MT-5                   | 2.3                               | 92.9              | 10 mM, 2 bar              | [37]   |
| TFN-0.25GO             | 1.9                               | 98.7              | 1000 ppm, 10 bar          | [38]   |
| TFC-D3                 | 1.6                               | 97.7              | 2000 ppm, 8.6 bar         | [30]   |
| TFN0.5                 | 2.0                               | 92.7              | 20 mM, 2.5 bar            | [39]   |
| TFC-sPPSU              | 2.0                               | 91.0              | 1000 ppm, 1 bar           | [40]   |
| TFC-D4                 | 1.8                               | 97.9              | 2000 ppm, 8.6 bar         | [41]   |
| TFC-PES                | 1.8                               | 81.5              | 200 ppm, 1 bar            | [24]   |
| Double-skin TFC        | 1.3                               | 88.3              | 2000 ppm, 5 bar           | [42]   |
| TFC PDA@PSf-5h         | 0.5                               | 88.0              | 200 ppm, 2 bar            | [43]   |
| AMTFC-3                | 1.1                               | 96.6              | 1000 ppm, 6 bar           | [23]   |
| MT-75                  | 1.2                               | 93.6              | 10 mM, 3 bar              | [44]   |
| GOT-0.25               | 1.8                               | 98.7              | 1000 ppm, 10 bar          | [38]   |
| TFC-sPPSU              | 0.7                               | 91.5              | 400 ppm, 3.45 bar         | [32]   |
| TFC2                   | 1.8                               | 93.4              |                           |        |
| CTA-HW                 | 1.2                               | 78.5              |                           | [45]   |
| CTA-W                  | 0.3                               | 81.9              | 20 mM, 3.75 bar           | [45]   |
| CTA-NW                 | 0.5                               | 92.4              |                           |        |
| HTI                    | 0.7                               | 89.1              | 2000 ppm, 8.6 bar         | [41]   |
| Aquaporin              | 0.5                               | 97.9              | 2000 ppm, 125 psi         | [46]   |
**Supplementary Table 10.** NF performance comparison with reported lab-made and commercial NF membranes.

| Membrane code   | Water permeance (L m⁻² h⁻¹ bar⁻¹) | NaCl Rejection (%) | Testing condition     | Ref.   |
|-----------------|-------------------------------------|--------------------|-----------------------|--------|
| NaCl@PIP        | 16.6                                | 72.7               | 5 bar, 1000 ppm       | This work |
| NaHCO₃@PIP      | 12.4                                | 75.0               |                       |        |
| Na₂SO₄@PIP      | 15.7                                | 60.3               |                       |        |
| TFC-24          | 2.1                                 | 47.8               |                       |        |
| TFC-15          | 5.8                                 | 35.5               | 10.3 bar, 1000 ppm    | [47]   |
| TFC-12          | 8.3                                 | 44.6               |                       |        |
| M4              | 4.9                                 | 70.0               | 10 bar, 1000 ppm      | [48]   |
| M9              | 8.3                                 | 50.0               |                       |        |
| CSA             | 4.8                                 | 30.4               |                       |        |
| CSA/GO          | 5.5                                 | 33.0               | 10 bar, 2000 ppm      | [49]   |
| CSA/TiO₂        | 5.1                                 | 35.6               |                       |        |
| CSA/rGo-TiO₂    | 6.1                                 | 36.6               |                       |        |
| GO-COCl         | 3.8                                 | 57.2               | 6 bar, 1000 ppm       | [50]   |
| MWCNT-COOH      | 6.2                                 | 34.0               |                       |        |
| MWCNT-OH        | 6.9                                 | 35.3               | 6 bar, 2000 ppm       | [51]   |
| MWCNT-NH        | 5.3                                 | 35.1               |                       |        |
| PMMA-MWCNT      | 7.0                                 | 44.1               | 10 bar, 2000 ppm      | [52]   |
| TS-I            | 13.3                                | 51.2               | 4.8 bar, 2000 ppm     | [53]   |
| PIP             | 12.6                                | 15.0               |                       |        |
| SDS             | 17.1                                | 27.0               |                       |        |
| SDBS            | 14.9                                | 21.3               |                       |        |
| SB3-14          | 20.3                                | 20.9               | 4 bar, 1000 ppm       | [54]   |
| CTAB            | 26.2                                | 9.9                |                       |        |
| TsNA            | 14.9                                | 14.8               |                       |        |
| PIP0.05-SLS     | 23.1                                | 45.0               | 5 bar, 2000 ppm       | [55]   |
| PIP0.1-SLS      | 16.4                                | 42.1               |                       |        |
| Material                  | A (m) | B (m) | Pressure (bar) | Concentration (ppm) |
|---------------------------|-------|-------|----------------|---------------------|
| TFC-2                     | 21.0  | 18.8  | 5              | 2000               |
| NF-0.1%CB-1               | 15.5  | 12.5  | 6              | 1000               |
| EIP                       | 16.6  | 30.0  | 5              | 1000               |
| TFN0.3                    | 2.4   | 59.5  | 8              | 1000               |
| TFC-PDA/PEI               | 10.5  | 50.0  | 6              | 1000               |
| PA/PES/PANI-0.2           | 15.7  | 33.0  | 6              | 2000               |
| CNT0.005                  | 5.3   | 36.7  | 10             | 2000               |
| NFM-3                     | 5.6   | 30.0  | 4              | 1000               |
| TFNMA-GO-ODA              | 8.3   | 38.0  | 6              | 2000               |
| NFM-4                     | 9.8   | 23.5  | 5              | 500                |
| TFC-ZIF-8-PSS             | 9.6   | 38.0  | 5              | 500                |
| PA/PES                    | 12.3  | 48.0  | 10             | 2000               |
| PA-PD/PES                 | 11.0  | 31.0  | 2              | 1000               |
| PA-SiO$_2$/PES            | 9.5   | 26.0  | 6              | 2000               |
| NF270                     | 11.0  | 66.0  | 5              | 2000               |
| NTR-7450                  | 11.0  | 50.0  | 10             | 2000               |
| NTR-7410                  | 18.0  | 10.0  | 10             | 2000               |
| NTR-7470                  | 4.0   | 70.0  | 10             | 2000               |
| NTR-7250                  | 4.0   | 50.0  | 10             | 2000               |
| XN-45                     | 5.0   | 25.0  | 7.6            | 2000               |
| Koch TFC-SR2              | 7.5   | 24.0  | 10             | 585                |
| Koch TFC-SR3              | 2.2   | 38.0  | 10             | 585                |
| GE-Osmonics HL            | 6.9   | 33.0  | 7.6            | 1000               |
**Supplementary Table 11.** NF performance comparison with reported lab-made and commercial NF membranes.

| Membrane code | Water permeance (L m⁻² h⁻¹ bar⁻¹) | Na₂SO₄ Rejection (%) | Testing condition | Ref. |
|---------------|----------------------------------|----------------------|-------------------|------|
| LiCl@PIP      | 12.6                             | 99.2                 |                   |      |
| NaCl@PIP      | 16.6                             | 97.8                 | 5 bar, 1000 ppm   | This work |
| KCl@PIP       | 17.5                             | 97.4                 |                   |      |
| NaHCO₃@PIP    | 12.4                             | 99.2                 |                   |      |
| GO-COCl       | 3.8                              | 97.1                 | 6 bar, 1000 ppm   | [50] |
| MWCNT-COOH    | 6.2                              | 96.6                 |                   |      |
| MWCNT-OH      | 6.9                              | 97.6                 | 6 bar, 2000 ppm   | [51] |
| MWCNT-NH      | 5.3                              | 96.8                 |                   |      |
| PMMA-MWCNT    | 7.0                              | 99.0                 |                   | [52] |
| TS-I          | 12.7                             | 99.1                 | 4.8 bar, 2000 ppm | [53] |
| PIP           | 12.6                             | 97.1                 |                   |      |
| SDBS          | 14.9                             | 98.8                 | 4 bar, 1000 ppm   | [54] |
| TsNA          | 14.9                             | 98.0                 |                   |      |
| NF-0.1%CB-1   | 15.5                             | 98.0                 | 6 bar, 1000 ppm   | [57] |
| EIP           | 16.6                             | 95.5                 | 5 bar, 1000 ppm   | [58] |
| TFC-PDA/PEI   | 10.5                             | 97.0                 | 6 bar, 1000 ppm   | [60] |
| CNT0.005      | 5.3                              | 95.7                 | 10 bar, 2000 ppm  | [62] |
| NFM-3         | 5.6                              | 98.8                 | 4 bar, 1000 ppm   | [63] |
| TFNMA-GO-ODA  | 8.3                              | 98.4                 | 6 bar, 2000 ppm   | [64] |
| NFM-4         | 9.8                              | 95.0                 | 5 bar, 500 ppm    | [65] |
| TFC-ZIF-8-PSS | 9.6                              | 91.0                 | 5 bar, 500 ppm    | [66] |
| PA-PD/PES     | 11.4                             | 93.5                 | 2 bar, 1000 ppm   | [68] |
| PA-SiO₂/PES   | 9.5                              | 97.4                 | 6 bar, 2000 ppm   | [69] |
| PA-TEOS0.5    | 15.9                             | 92.0                 | 5 bar, 500 ppm    | [73] |
| PA-Flower     | 6.4                              | 89.9                 | 10 bar, 1000 ppm  | [74] |
| TFN/UIO-66-NH2| 13.0                             | 98.1                 | 6 bar, 1000 ppm   | [75] |
| Membrane  | TFC NFM | NF90 | NF270 | 6 bar, 1000 ppm | 3.5 bar, 1000 ppm |
|-----------|---------|------|-------|-----------------|------------------|
|           | 17.6    | 7.0  | 11.2  |                 |                  |
|           | 95.0    | 99.1 | 98.5  |                 |                  |
**Supplementary Table 12.** Detailed fabrication parameters of the TFC membranes

| System               | Dope composition (wt.%) | MPD/TMC          | PIP/TMC          | PEI/TMC          |
|----------------------|-------------------------|------------------|------------------|------------------|
| **Substrate membrane** | PSF/PEG-400/NMP =18/16/66 |                  |                  |                  |
|                      |                         | MPD/H$_2$O 3.4/96.6 | PIP/H$_2$O 0.35/99.65 | PEI/H$_2$O 2/98 |
|                      |                         | MPD/salt/H$_2$O 3.4/3/93.6 | MPD/salt/H$_2$O 0.35/3/96.65 | MPD/salt/H$_2$O 2/3/95 |
| **Active layer**     |                         | TMC/Hexane 0.15/100 | TMC/Hexane 0.15/100 | TMC/Hexane 0.15/100 |
|                      |                         | Immersion time in amine solution (min) | IP reaction time (min) |
|                      |                         | 5               | 10               | 30               |
|                      |                         | 1               | 2                | 10               |
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