

Supplementary Materials

Zwitterionic and hydrophilic polyelectrolyte/metal ion anti-fouling layers via covalent and coordination bonds for reverse osmosis membranes

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The mechanical properties and pressure resistance of membrane

The mechanical properties were performed by a universal tensile testing machine (Instron 5966, Instron Corp., Norwood, USA). Besides, the pressure resistance of obtained membranes is more considered during practical separation.

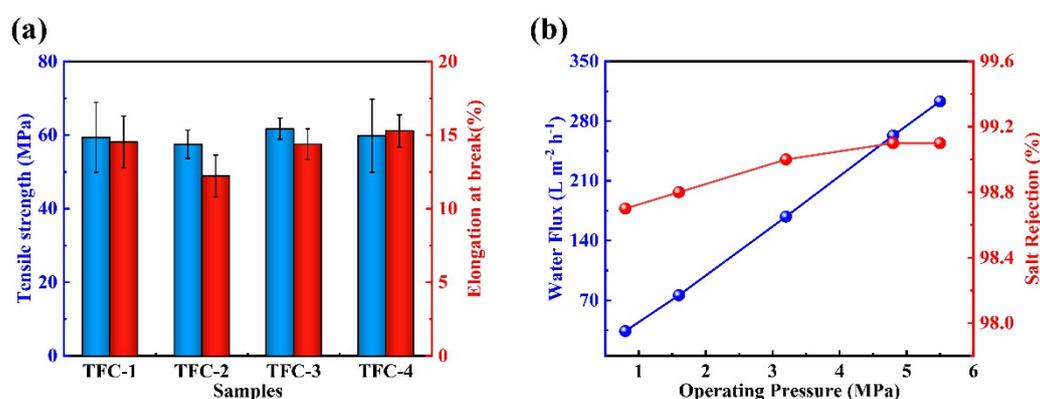


Fig. S1 Mechanical properties of membranes (a) and the separation performance of TFC-4 under different operating pressure (b).

As Fig. S1a shown, modified membranes and the original membrane have similar tensile strength and elongation at break, which is attributed to the surface modification has no obvious effect on membrane thickness and mechanical strength. All membranes represent good

mechanical properties with tensile strength of 57.5-61.7 MPa and elongation at break of 12.2-15.3%. In Fig. S1b, the water flux increases linearly with the improvement of operating pressure, which indicates that the membrane is operated normally at 0-5.5 MPa. Besides, the membrane remains stable salt rejection at a pressure as high as 5.5 MPa. These results indicate PEI/PASP/Fe modified membranes exhibit superior pressure resistance.

Stability of polyelectrolyte/metal ion layer under real water conditions

In order to investigate the stability of the polyelectrolyte/metal ion layer under real water conditions, we designed two different experiments: static and dynamic stability tests. Here, the artificial seawater (with compositions of 26.73 g/L NaCl, 3.25 g/L MgSO₄, 1.15 g/L CaCl₂ and 2.26 g/L MgCl₂, 0.026 g/L AlCl₃, 1.5 mg/L humic acid) was prepared according to Mocledon method. In the static experiment, the PASP/PEI/Fe modified membrane was immersed into the artificial seawater for 10 h. In a dynamic experiment, the membrane was tested with artificial seawater at 5.5 MPa for 1 h. Then, the separation data was collected at 1.6 MPa/2000 ppm NaCl. The separation performance of membranes before and after artificial seawater treatment were recorded. After all tests were finished, the distribution of Fe(III) on membrane surface before and after treatment was observed by energy dispersive X-ray spectroscopy. The membrane separation performance and iron distribution are shown in Fig. S2.

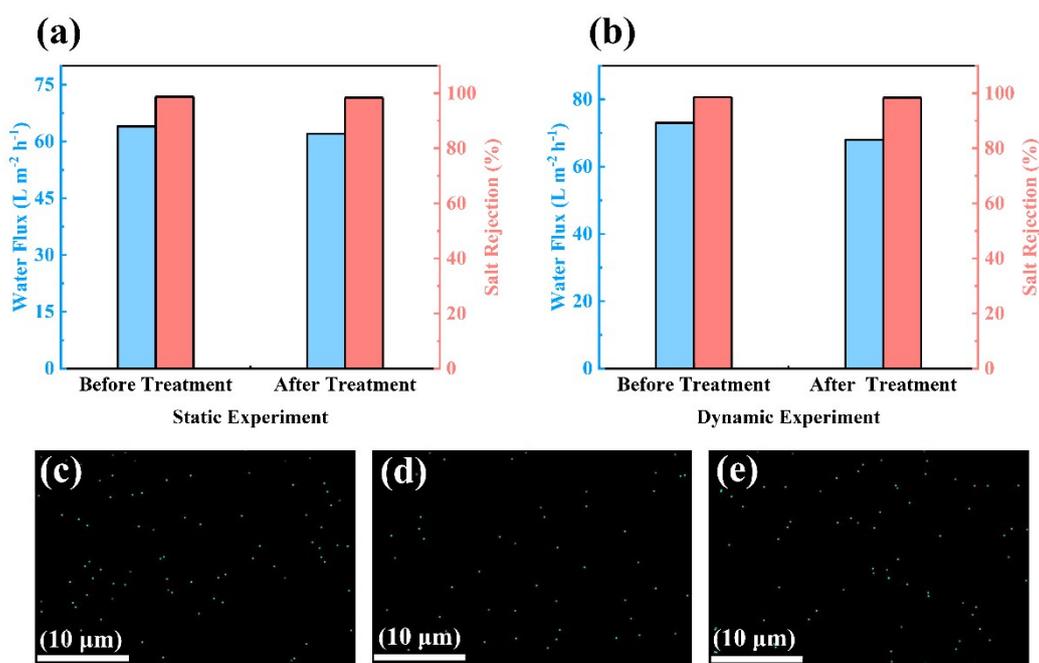


Fig. S2 Membrane performance of TFC-4 before and after artificial seawater static treated for 10 h (a) and dynamic treated for 1 h (b). EDS mapping images of Fe distribution on TFC-4 surface: untreated (c), static treated (d), dynamic treated (e).

Details about the filtration device

The Diagram of filtration device is shown in Fig. S3, the lab-scale cross-flow filtration system with three parallel membrane cells that have an effective testing area of 60 cm² and 1 mm height flow channel. Specifically, the feed solution was transported into the membrane cell by a diaphragm pump. Desired pressure and feed flow rate were achieved by adjusting the flow rate regulator and back-pressure regulator. The permeate flow rate was measured by an analytical balance. Permeate and retentate were recycled back to the feed tank. Besides, feed solution in the tank was maintained at 25 °C by a condensing system and the crossflow rate kept at 35 L/min.

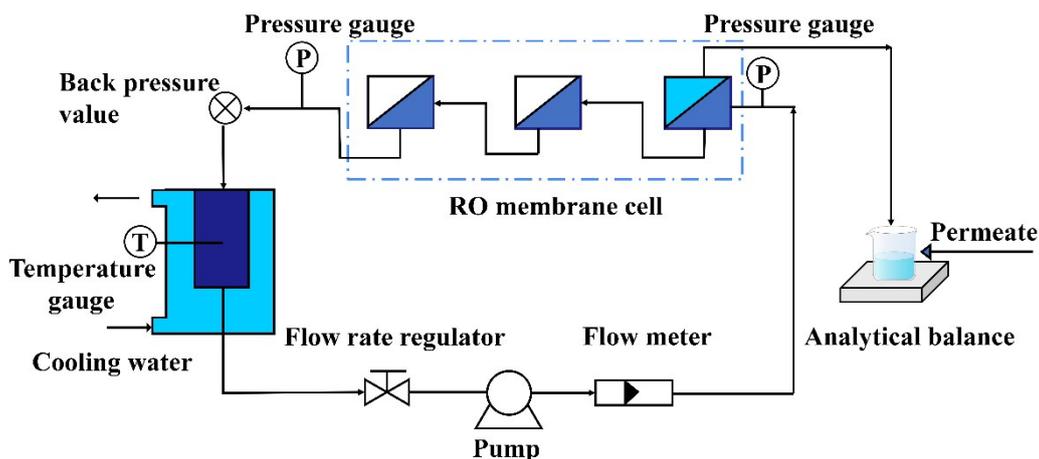


Fig. S3 Diagram of the lab-scale cross-flow filtration system.

Membrane anti-fouling performance against SA solution with Ca^{2+}

Considering the actual water contains some hardness ions such as calcium ions, the SA fouling with 1.5 mmol/L Ca^{2+} was employed to observe the membrane anti-fouling performance, the results are shown in Fig. S4. The water flux drops sharply when adding Ca^{2+} to the SA solution, which is due to the complexation of SA with Ca^{2+} . The decline trend of water flux slows down when the membrane surface was covered by SA-Ca gels completely. The data in Table S1 demonstrates that TFC-4 has a high FRR and a low DRt, which represents the better anti-fouling performance than TFC-3 and TFC-1.

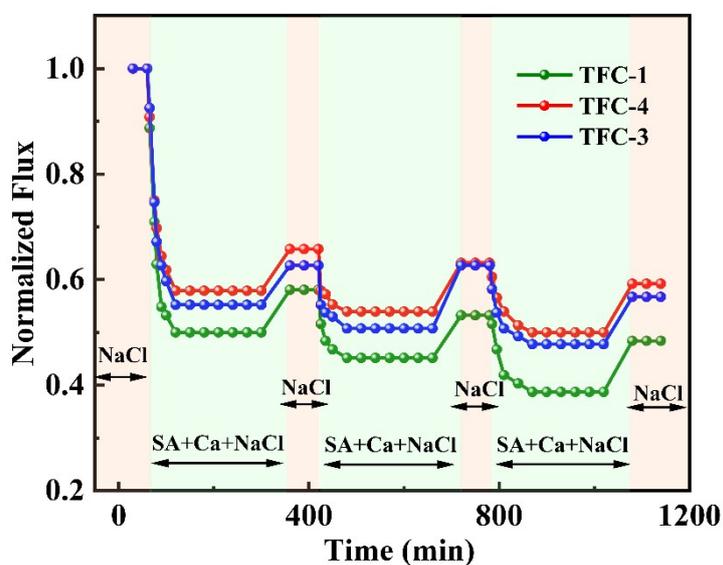


Fig. S4 Normalized water flux of TFC-1, TFC-3 and TFC-4 against SA (the concentration of Ca²⁺ is 1.5 mmol/L) at 25 °C, 1.6 MPa.

Table S1 The DR_t and FRR value for TFC-1, TFC-3 and TFC-4.

	SA + Ca Solution		
	TFC-1	TFC-3	TFC-4
DR _t	61.3%	52.2%	50.0%
FRR	53.2%	56.7%	59.2%