

**Graphene oxide (GO) interlayered thin film nanocomposite (TFN) membranes  
with high solvent resistance for organic solvent nanofiltration (OSN)**

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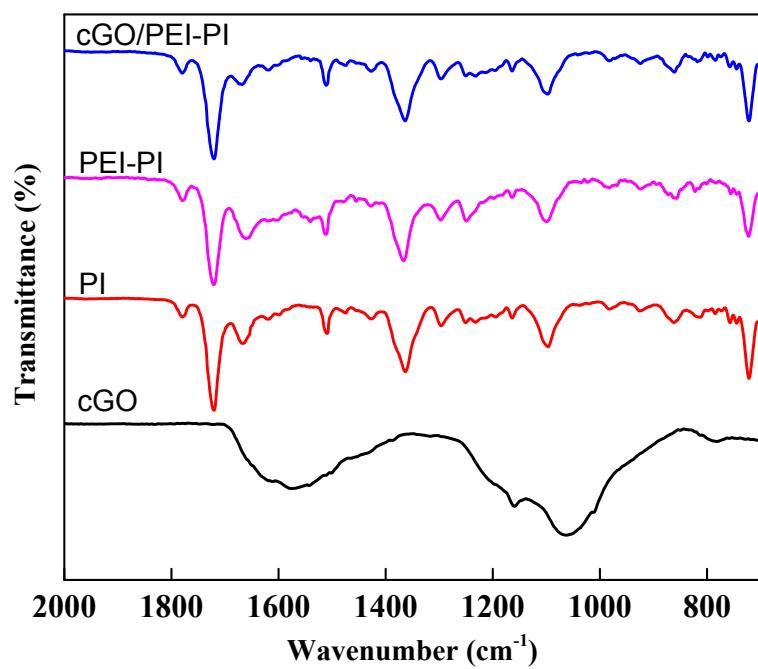


Fig. S1 The FTIR spectra of the cGO nanosheets, PI substrate, PEI-PI substrate, and cGO/PEI-PI substrate. (PEI: the concentration is  $5 \text{ mg L}^{-1}$ , the reaction time and temperature were 30 s and  $25^\circ\text{C}$ ; cGO nanosheets: the concentration is  $2 \text{ mg L}^{-1}$ , the deposition time is 60 s)

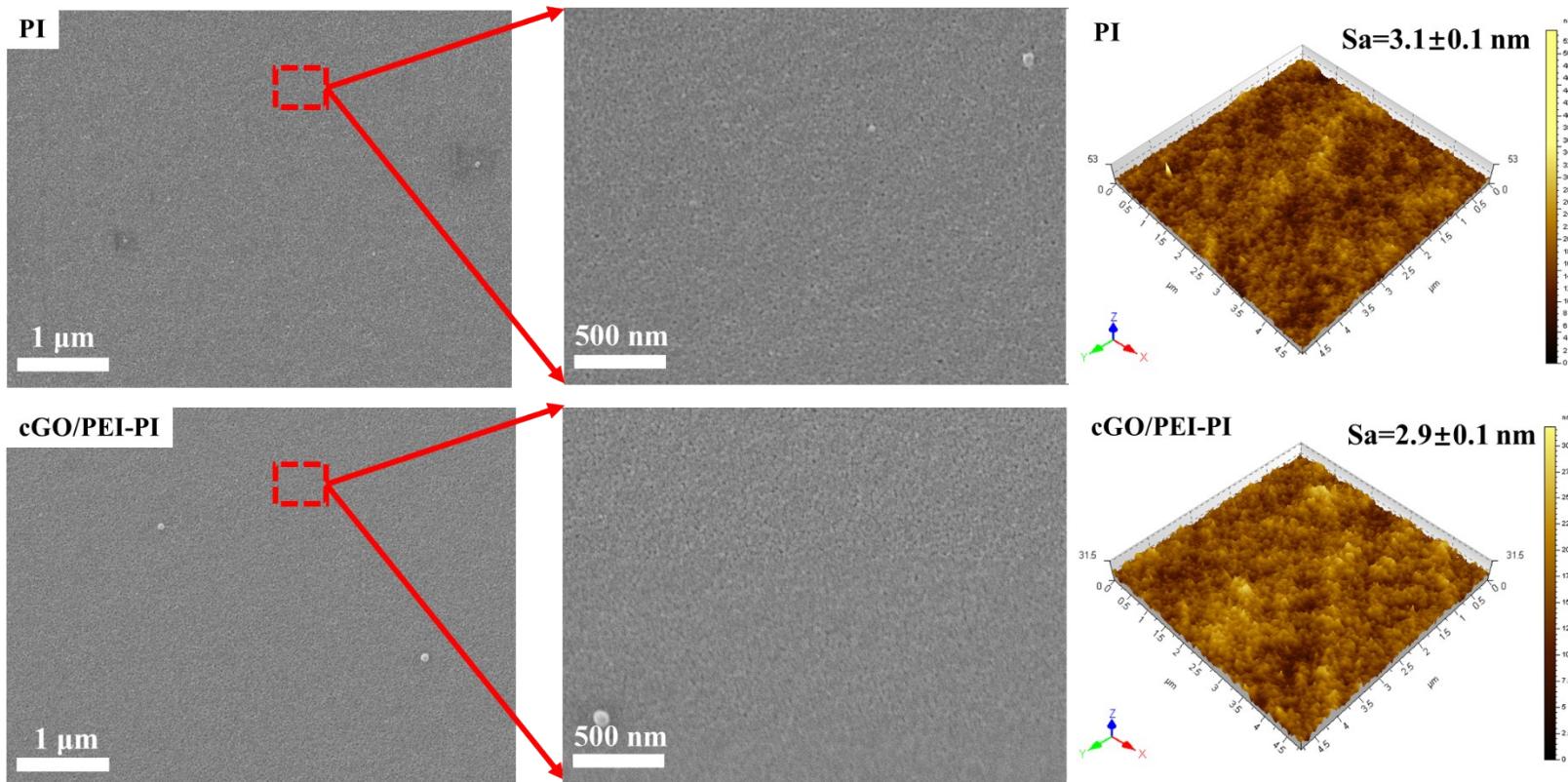


Fig. S2 Surface SEM and AFM images of the PI and cGO/PEI-PI substrate.

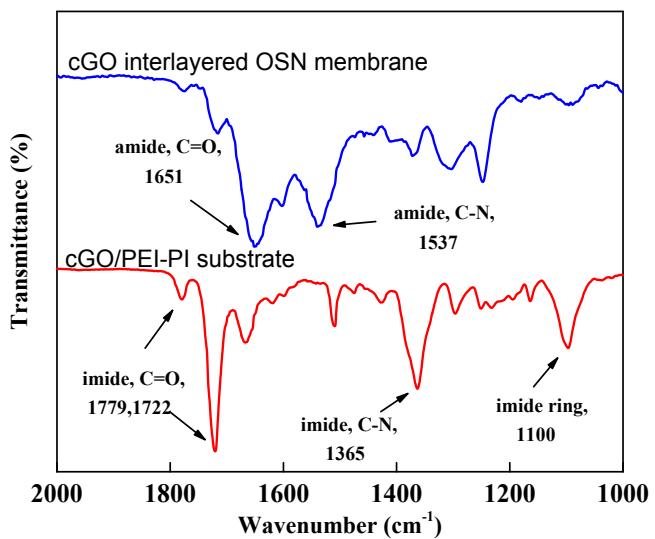


Fig. S3 The FTIR spectra of the interlayer OSN membrane and PI substrate. (PEI: the concentration is 5 mg L<sup>-1</sup>, the reaction time and temperature were 30 s and 25 °C; cGO nanosheets: the concentration is 2 mg L<sup>-1</sup>, the deposition time is 60 s)

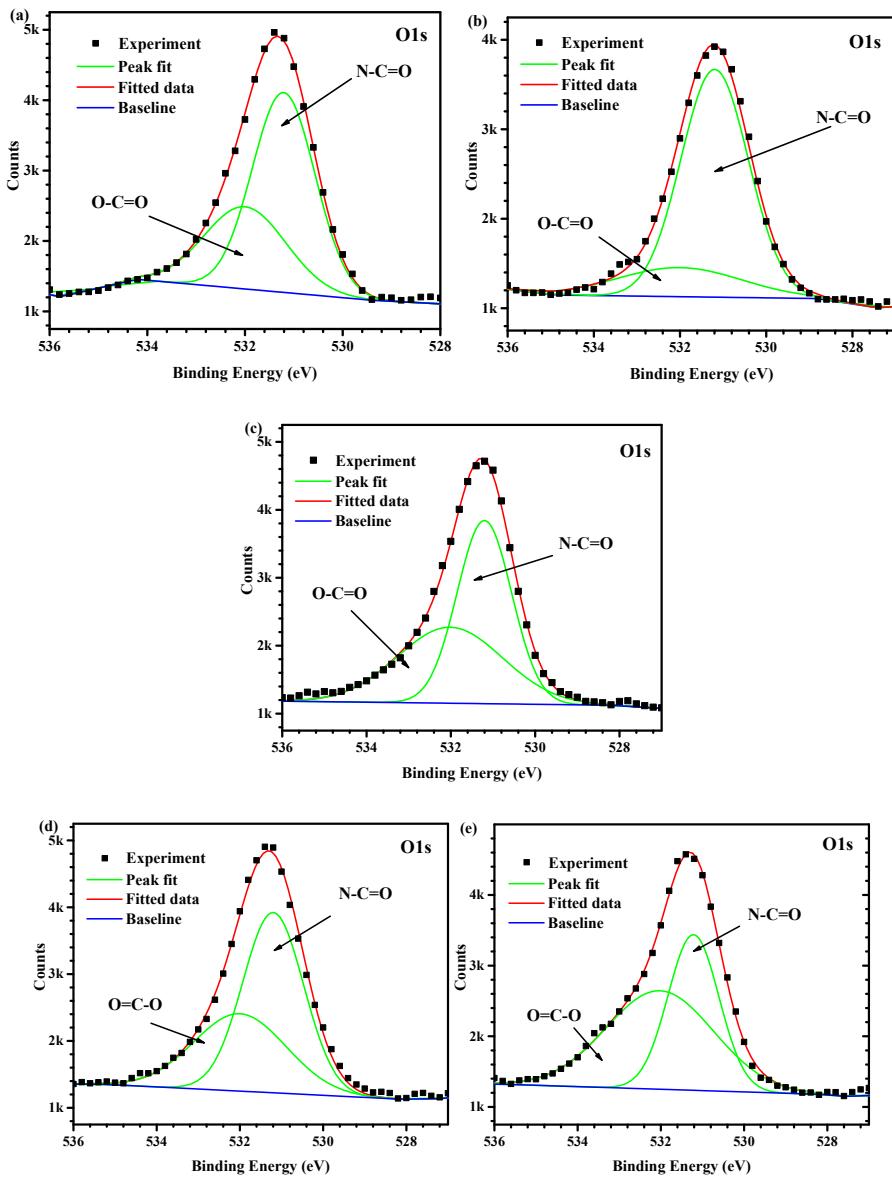


Fig. S4 High-resolution O1s spectra of the cGO-interlayered OSN membranes with different PEI concentration. (a) 1 mg L<sup>-1</sup>; (b) 5 mg L<sup>-1</sup>; (c) 10 mg L<sup>-1</sup>; (d) 100 mg L<sup>-1</sup>; (e) 1000 mg L<sup>-1</sup>.

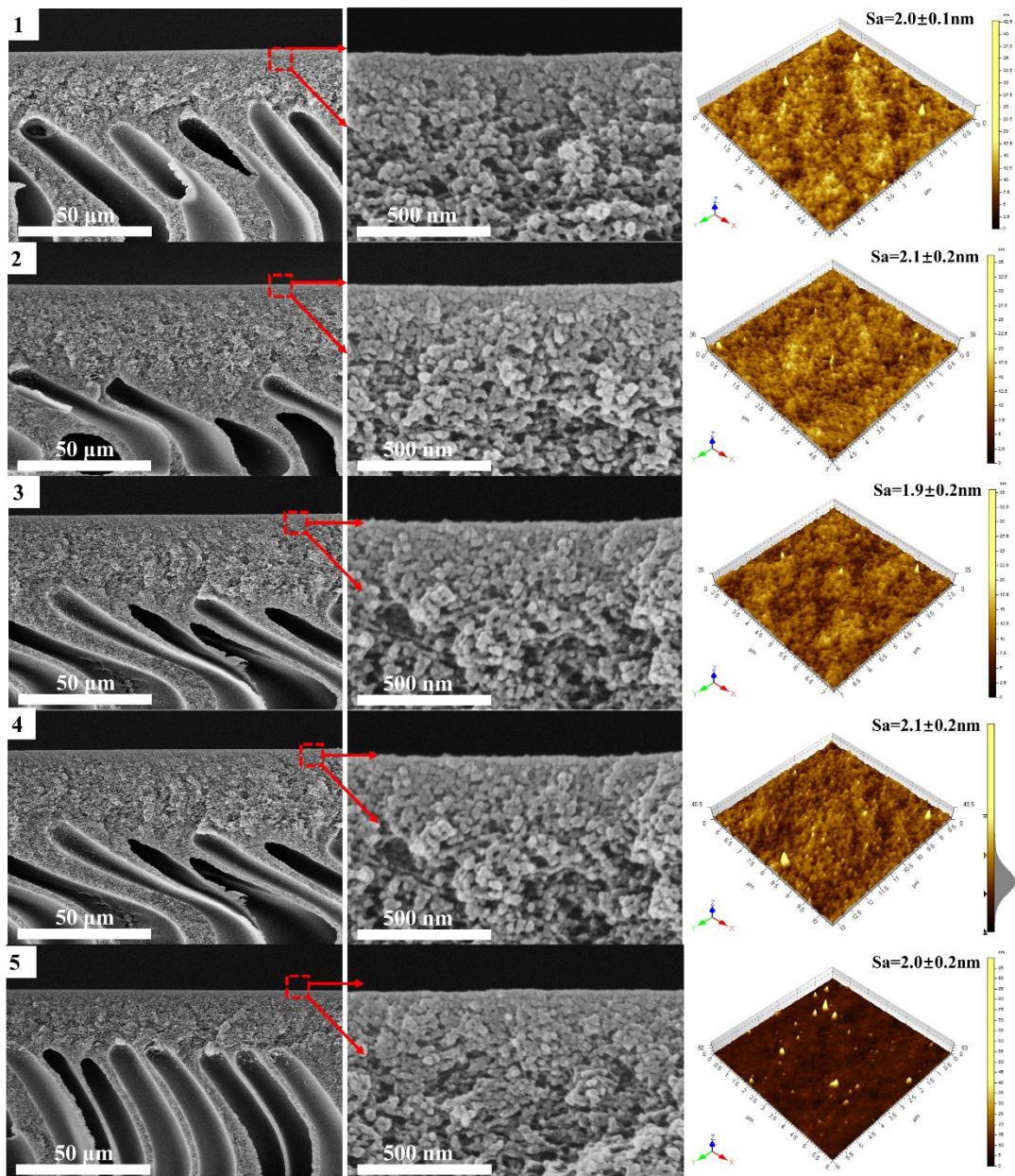


Fig. S5 Cross-sectional SEM images and surface AFM morphologies of the cGO-interlayered OSN membranes with different PEI concentration. (1) 1 mg L<sup>-1</sup>; (2) 5 mg L<sup>-1</sup>; (3) 10 mg L<sup>-1</sup>; (4) 100 mg L<sup>-1</sup>; (5) 1000 mg L<sup>-1</sup>.

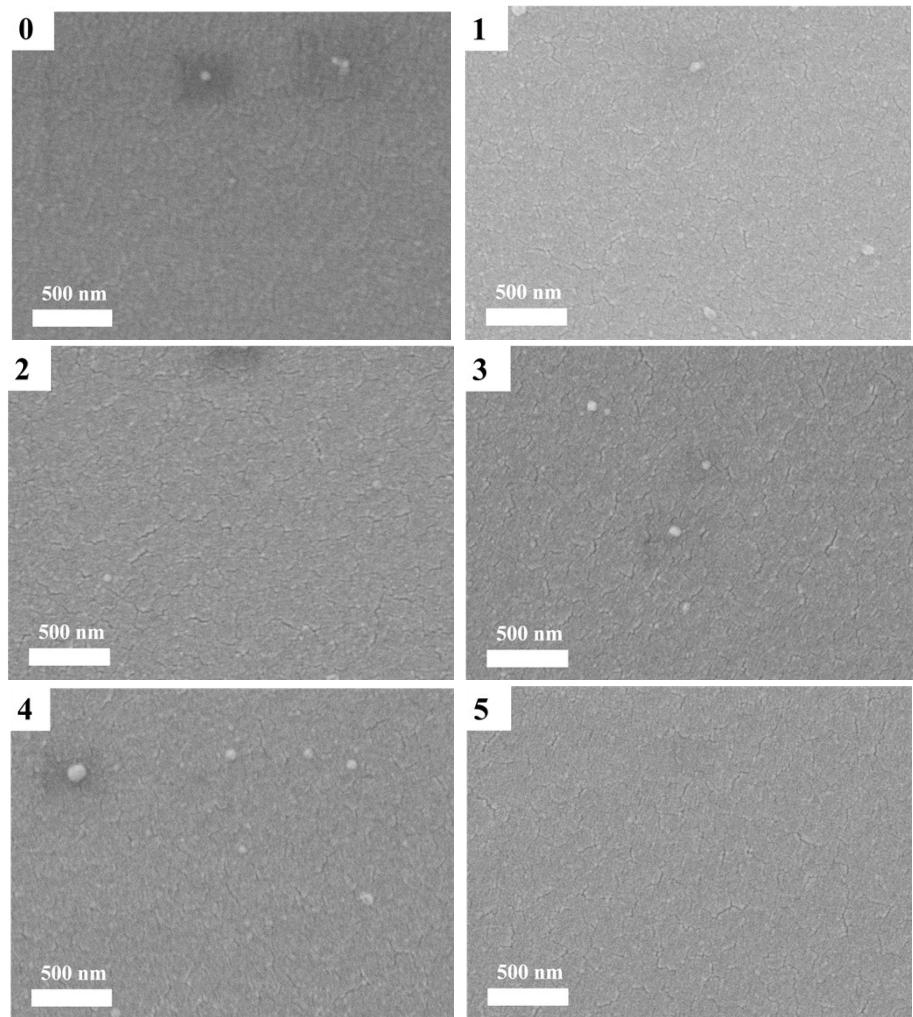


Fig. S6 SEM images of surface morphologies of the cGO-interlayered OSN membranes with different PEI concentration. (0)  $0 \text{ mg L}^{-1}$ ; (1)  $1 \text{ mg L}^{-1}$ ; (2)  $5 \text{ mg L}^{-1}$ ; (3)  $10 \text{ mg L}^{-1}$ ; (4)  $100 \text{ mg L}^{-1}$ ; (5)  $1000 \text{ mg L}^{-1}$ .

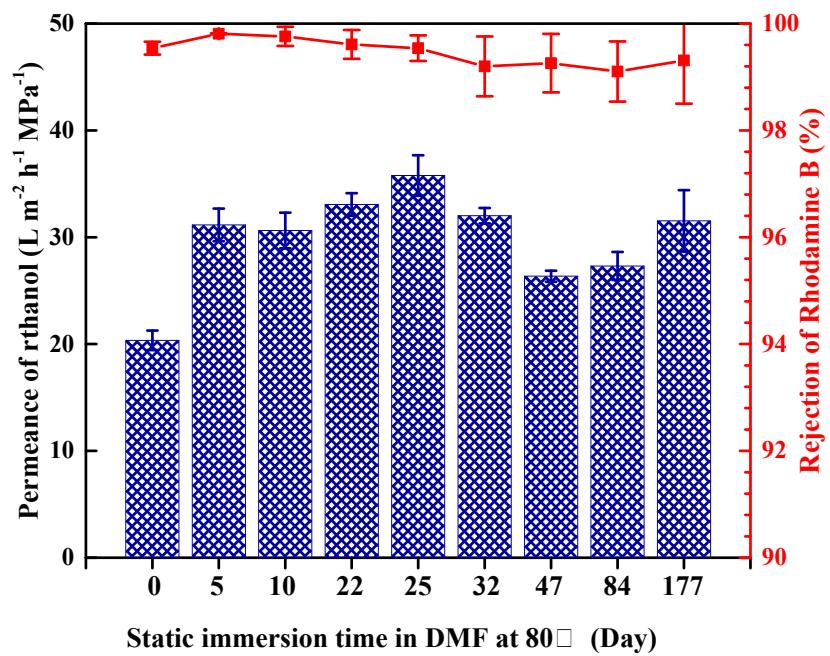
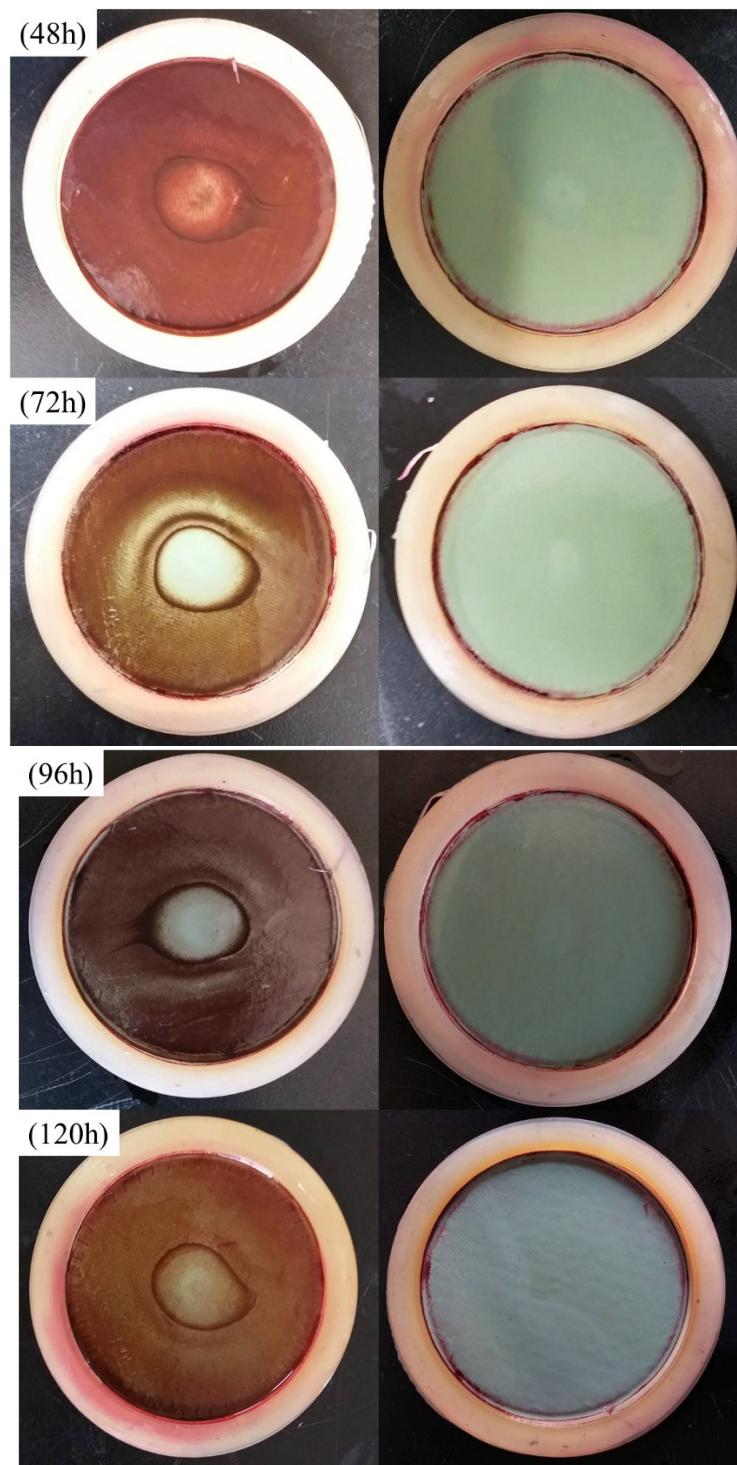


Fig. S7 Long-term static immersion in DMF at 80 °C for the cGO-interlayered OSN membrane. (The other conditions were the same as the optimized cGO-interlayered OSN membrane expect that the PEI concentration was 100 mg L<sup>-1</sup>)

Before cleaning

After cleaning



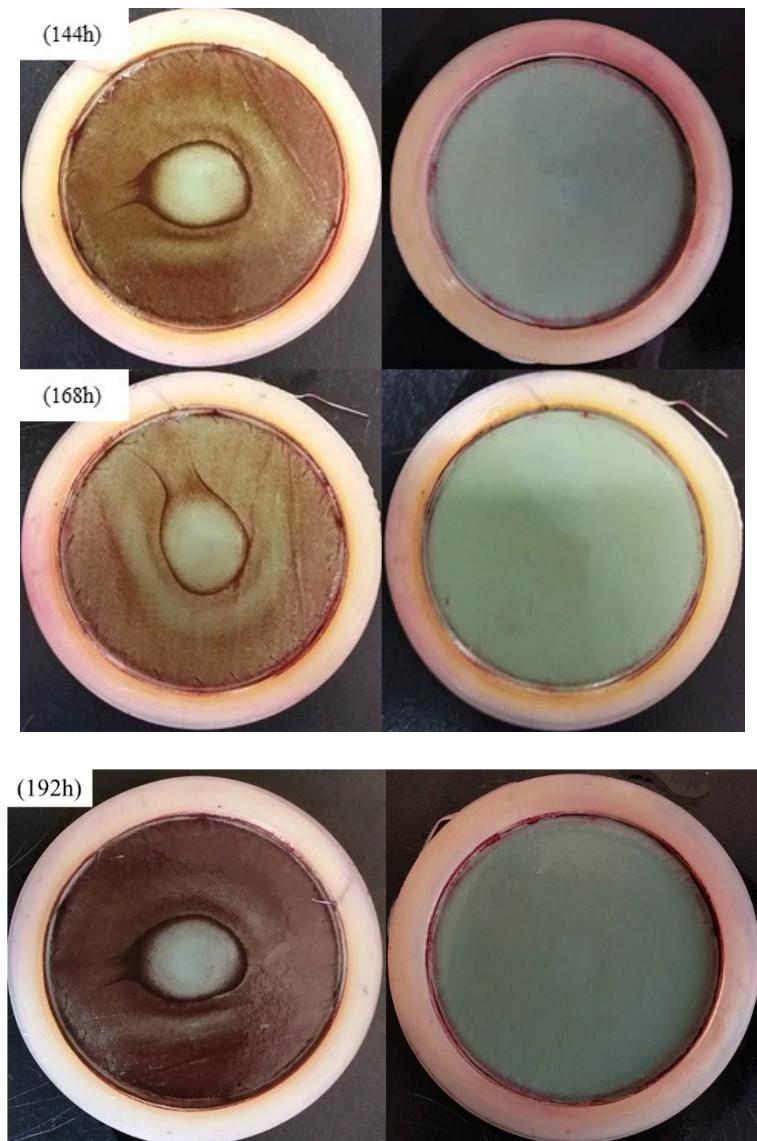


Fig. S8 the images of the cGO-interlayered OSN membrane after the continuous cross-flow filtration of  $100 \text{ mg L}^{-1}$  RB/DMF solution at room temperature for different time.  
(before and after cleaning with DMF)

Table S1 The percentage of the cGO-interlayered membranes with different PEI concentration from XPS.

PEI concentration (mg L <sup>-1</sup> )	C1s			N1s			O1s		
	Binding			Binding			Binding		
	Energy (eV)	Species	(%)	Energy (eV)	Species	(%)	Energy (eV)	Species	(%)
1	284.7	C-H/C-C	66.27	399.9	N-C=O	91.20	531.2	N-C=O	65.15
	286.0	C-N	20.35	401.7	R-N <sup>+</sup> H <sub>3</sub>	8.80	532.0	O-C=O	34.85
	288.0	N-C=O	13.38						
5	284.7	C-H/C-C	74.98	399.0	R-NH <sub>2</sub>	18.20	531.2	N-C=O	81.00
	286.0	C-N	15.86	399.9	N-C=O	71.78	532.0	O-C=O	19.00
	288.0	N-C=O	9.16	401.7	R-N <sup>+</sup> H <sub>3</sub>	10.02			

	284.7	C-H/C-C	74.90		399.9	N-C=O	83.63	531.2	N-C=O	55.55
10	286.0	C-N	11.27		401.7	R-N <sup>+</sup> H <sub>3</sub>	16.37	532.0	O-C=O	44.45
	288.0	N-C=O	13.83							
100	284.7	C-H/C-C	67.55		399.9	N-C=O	84.60	531.2	N-C=O	60.60
	286.0	C-N	20.00		401.7	R-N <sup>+</sup> H <sub>3</sub>	15.40	532.0	O-C=O	39.40
	288.0	N-C=O	12.45							
1000	284.7	C-H/C-C	77.56		399.9	N-C=O	82.17	531.2	N-C=O	42.77
	286.0	C-N	9.36		401.7	R-N <sup>+</sup> H <sub>3</sub>	17.83	532.0	O-C=O	57.23
	288.0	N-C=O	13.08							

Table S2 Contact angle of the PI substrates with different PEI concentration with/without cGO nanosheets

PEI concentration (mg L <sup>-1</sup> )	Contact angle (°)	
	PEI-PI	cGO/PEI-PI
1	70.6 ± 0.8	71.3 ± 0.4
5	70.1 ± 1.0	55.1 ± 0.8
10	70.0 ± 0.3	51.2 ± 0.1
100	69.3 ± 0.4	49.2 ± 0.2
1000	63.9 ± 0.6	48.3 ± 0.7

Table S3 Physical properties of different solvents.<sup>1-6</sup>

Solvent	Polarity Index	Viscosity, (cP)	Surface tension, (dyne/cm)	Hansen solubility parameter, (MPa <sup>-1/2</sup> )	Hansen solubility parameter for polarity (MPa <sup>-1/2</sup> )	Kinetic diameter, (nm)	Molar volume, (cm <sup>3</sup> /mol)
DMF	6.4	0.920	37.1	24.7	13.7 <sup>3</sup>	0.55	77.11
Acetone	5.1	0.312	24.6	19.3	10.4	0.46	74.1
THF	4.0	0.480	26.4	19.4	5.7 <sup>3</sup>	0.47	82.1
EtOH	4.3	1.018	22.34	25.0	8.8	0.44	58.2
IPA	3.9	2.092	23.34	21.3	7.2	0.47	77.2
n-Hexane	0.1	0.307	18.17	14.9	0	0.43	1304

## Reference

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