

Electronic Supplementary information

High water permeance and ions rejection through F-graphene oxide membrane

Wuerkaixi Maimuli^{1#}, Rujie Yang^{2#}, Shuai Wang², Junfan Liu¹, Fangfang Dai^{3,4}, Jun Wang², Lu Li⁵, Liang Chen^{6,1*} and Shanshan Liang^{2*}

¹Department of Environment and Resources, Zhejiang A&F University, Hangzhou 311300, China

²School of Physics, East China University of Science and Technology, Shanghai, 200237, China

³Wenzhou Institute, University of Chinese Academy of Sciences, Wenzhou, 325000, China

⁴Oujiang Laboratory (Zhejiang Lab for Regenerative Medicine, Vision and Brain Health), Wenzhou, 325001, China

⁵College of Chemistry and Materials Engineering, Zhejiang A&F University, Hangzhou 311300, China

⁶School of Physical Science and Technology, Ningbo University, Ningbo 315211, China

Corresponding Email: liangshanshan@ecust.edu.cn and chenliang2@nbu.edu.cn

[#] *These authors contributed equally to this work.*

Contents

PS 1 AFM results of pristine GO nanosheets.....	1
PS2. The molecule structure of 2,4-diamino-6-(4-fluorophenyl) pyrimidine.....	2
PS3. Property of GO-based suspensions.....	3
PS4 SEM image of F-GO membrane.....	4
PS5. XPS spectra of the GO and F-GO membranes.....	5
PS6. FT-IR spectra of the GO and F-GO membranes.....	7
PS7. Ion adsorption performances of the F-GO membranes.	8
PS8. Comparisons of water permeance and rejection for high valence ions of F-GO membranes in this work with other NF membranes in literatures.	10
PS9. Water permeances and rejection for the F-GO membranes with different anions.	12
PS10. The rejections and water permeances of F-GO membrane for different concentrations of FeCl ₃ solutions.	13
PS11. The long-term cross flow stability of the F-GO membrane.	14
References.....	15

PS 1 AFM results of pristine GO nanosheets

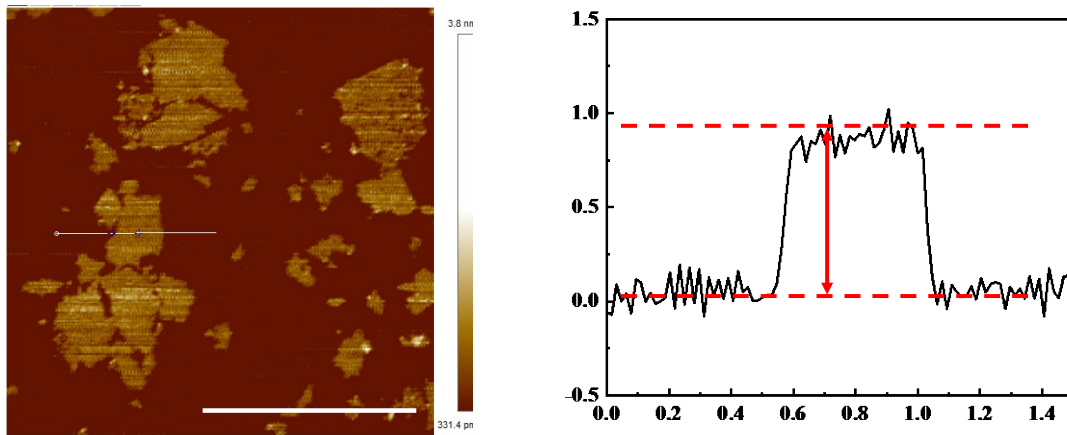


Fig. S1 (a) AFM image of pristine GO nanosheets. (b) The corresponding height profile of pieces from(a).

PS2. The molecule structure of 2,4-diamino-6-(4-fluorophenyl) pyrimidine.

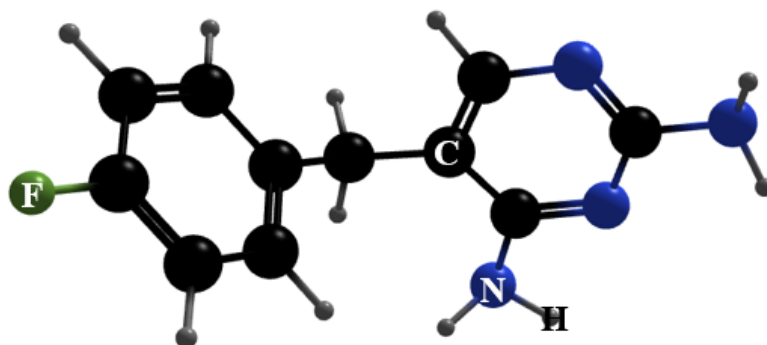


Fig. S2 The molecule structure of 2,4-diamino-6-(4-fluorophenyl) pyrimidine.

PS3. Property of GO-based suspensions

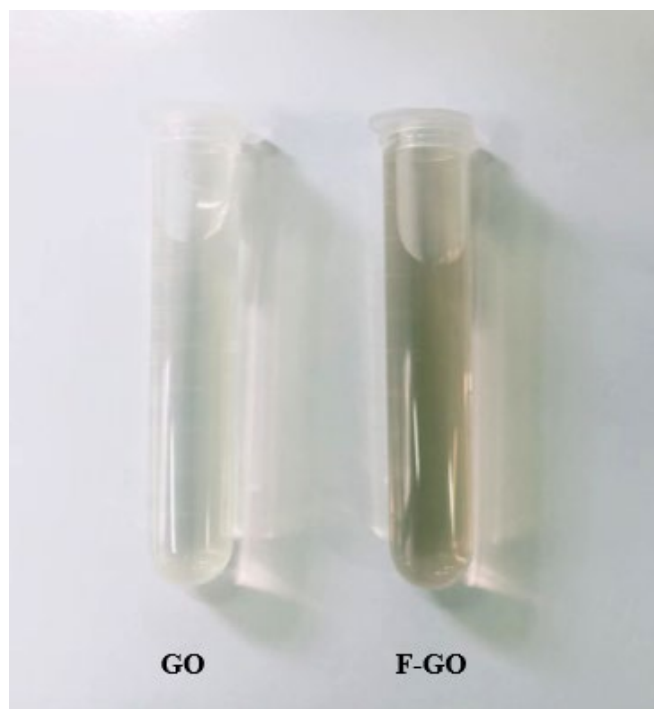


Fig. S3. Optical photo of GO and F-GO suspensions.

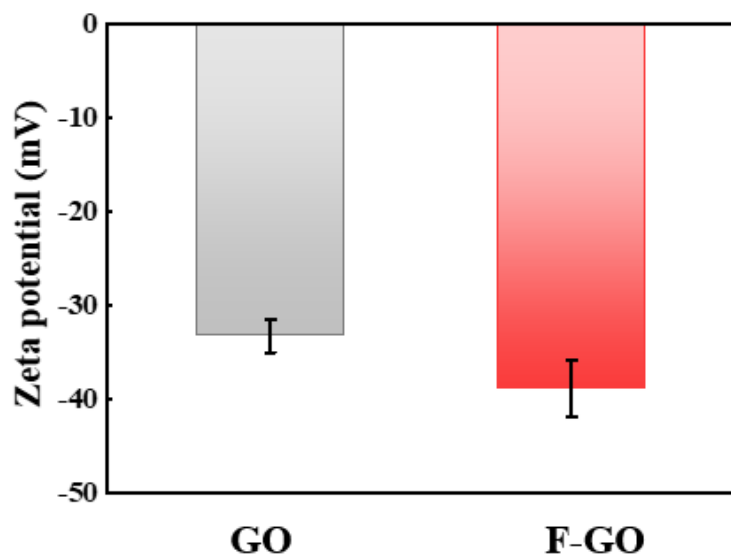


Fig. S4 Zeta potential of GO and F-GO suspensions.

PS4 SEM image of F-GO membrane.

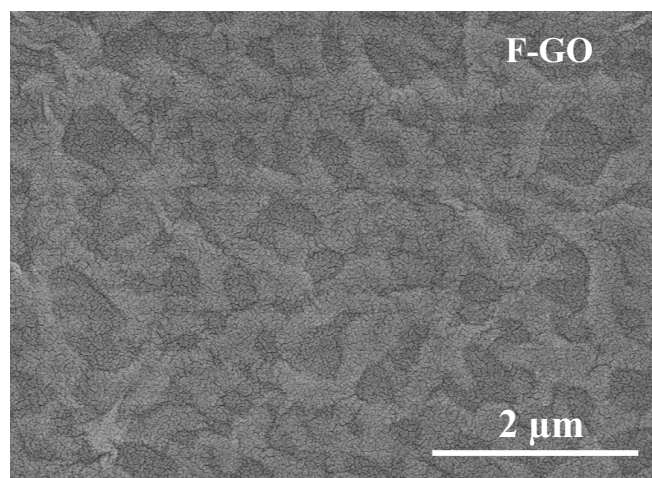


Fig. S5 SEM surface image of F-GO membrane.

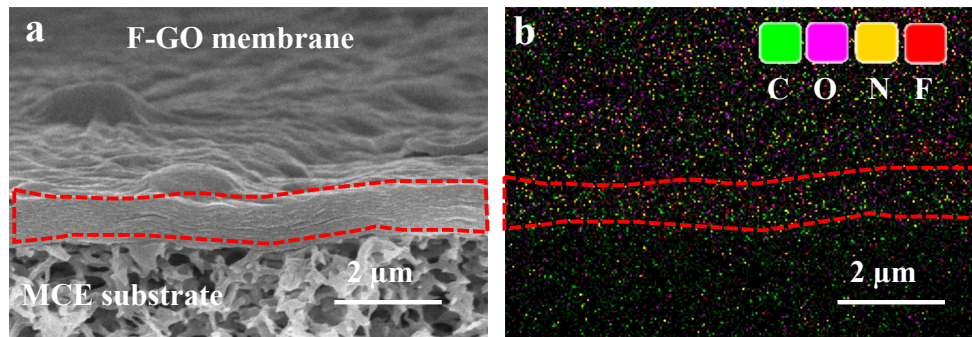


Fig. S6 (a) SEM/EDX images of cross-section of the F-GO membrane, where the enclosed image is mapped for the distribution of atoms; (b) EDX mapping of carbon atoms, oxygen atoms, nitrogen atoms, fluorine atoms on the cross-section of the F-GO membrane.

PS5. XPS spectra of the GO and F-GO membranes

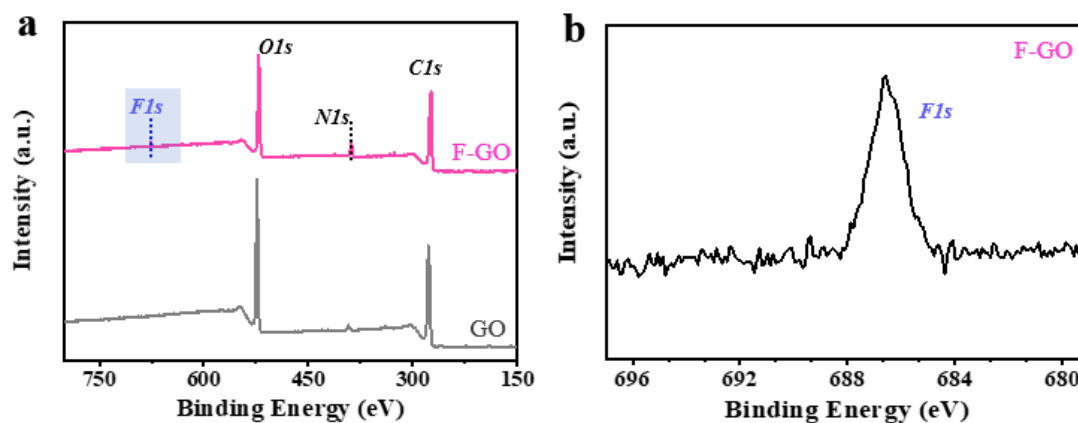


Fig. S7 (a) XPS wide scan of the prepared GO and F-GO membranes. (b) XPS spectra of the GO and F-GO membranes for F1s core level.

Table S1 Element composition of GO and F-GO membrane.

Membranes	Element composition				
	C	O	N	F	C/O
	(at.%)	(at.%)	(at.%)	(at.%)	
GO	67.67	32.33	-	-	2.09
F-GO	66.86	25.80	6.47	0.87	2.59

Table S2 XPS Element composition with different etch depth of F-GO membrane.

Etch depth (nm)	Element composition			
	C (at.%)	O (at.%)	N (at.%)	F (at.%)
0	66.8	25.27	6.25	1.58
10	84.81	7.19	6.21	1.7
20	85.92	6.83	5.67	1.58
30	85.49	6.83	5.97	1.71

PS6. FT-IR spectra of the GO and F-GO membranes.

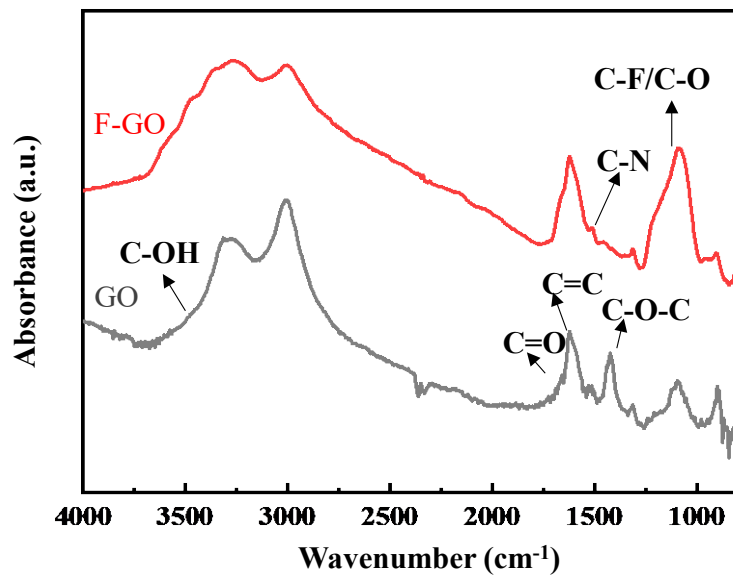


Fig. S8 FT-IR spectra of the GO and F-GO membranes.

PS7. Ion adsorption performances of the F-GO membranes.

After F-GO membranes were prepared using vacuum filtration, 300 mL 50 mg L⁻¹ FeCl₃ solutions were added to the feed side. The salt solution was stirred with a blender at ~180 rpm without filtration. Afterwards, the samples solutions were collected at pre-determined time intervals in 180 min, and analyzed for residual FeCl₃ ion concentrations. Due to the FeCl₃ solutions own gravity, the filtrates were collected only 2.5 mL after 180 min. A small amount of filtrates solution has negligible effect on the concentration of the salt concentration during the adsorption experiments.

The adsorption efficiency (AE) was calculated by using the following expression S(1) :

$$AE = \left(1 - \frac{C_a}{C_0}\right) * 100\% \quad S(1)$$

where C₀ and C_a (mg L⁻¹) are the initial solution concentrations and final solution concentrations after adsorption equilibrium, respectively.

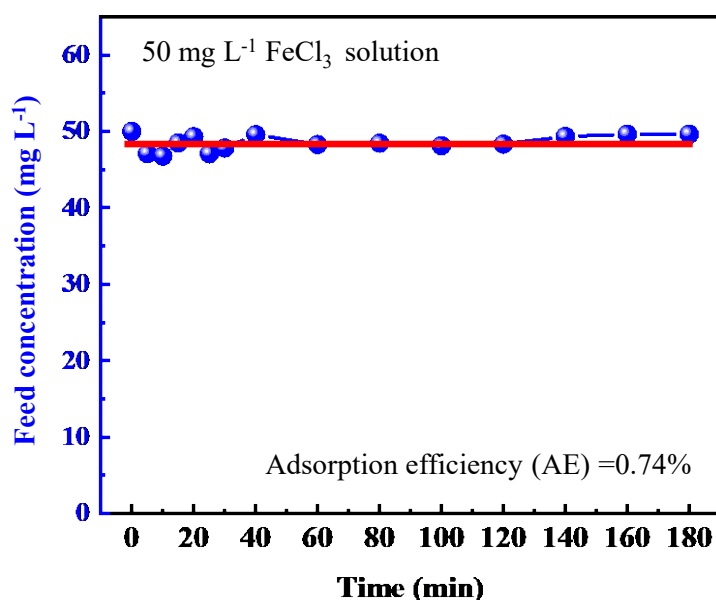


Fig. S9 The adsorption experiment for F-GO membrane in 50 mg L⁻¹ FeCl₃ solution.

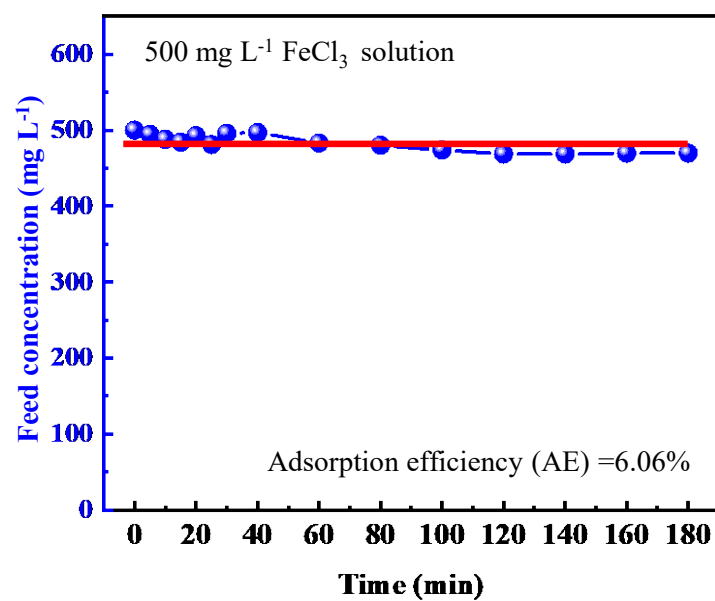


Fig. S10 The adsorption experiment for F-GO membrane in high concentration 500 mg L⁻¹ FeCl₃ solution.

PS8. Comparisons of water permeance and rejection for high valence ions of F-GO membranes in this work with other NF membranes in literatures.

Table S3. Comparisons of water permeance and rejection for high valence ions of F-GO membranes in this work with other NF membranes in literature.

Membrane	Test condition	Salts or ions	Rejection	Permeance (L h ⁻¹ m ⁻² bar ⁻¹)	References
GO-IPDI membrane	0.1 mM ,1bar	Cr(NO ₃) ₃	71.1%	100	S 1
PDA/TFC composite membrane	500 mg L ⁻¹ , 4 bar	Pb(NO ₃) ₂	91.1%	3.5	S 2
GO/Torlons composite membrane	1000 mg L ⁻¹ , 3bar	Pb(NO ₃) ₂	95%	4.7	S 3
PSE-GO-DMF	50 mg L ⁻¹ , 414 kPa	Cr ³⁺	91.2%	13.5	S 4
trGOM	0.1 mM, 2 bar	Cu ²⁺	65.4%	0.27	S 5
HNTs decorated commercial NF membranes	5 mg L ⁻¹ , 3 bar	Cu ²⁺	74.3%	13.9	S 6
γ-Al ₂ O ₃ /α-Al ₂ O ₃ hollow fiber	2000 mg L ⁻¹ , 5.0 bar	FeCl ₃	97.1%	17.4	S 7
PDA functionalized nanotube PEI	1000 mg L ⁻¹ , 3 bar	Pb(NO ₃) ₂	79.0%	50.0	S 8

membranes					
Commercial NF	2000 mg L ⁻¹ , 6-15bar	MgSO ₄	40%-98%	5-14	S 9
DK	50 mg L ⁻¹ , 2bar	MgSO ₄	56.5%	7.3	S 9
DL	50 mg L ⁻¹ ,2bar	MgSO ₄	60.7%	6.2	S 9
PA-1/MCE	1000 mg L ⁻¹ , 2bar	MgSO ₄	98%	60	S10
rGO-K ⁺	100 mg L ⁻¹ , 1bar	FeCl ₃	99.8%	86.1	S11
GO	50 mg L ⁻¹ , 1 bar	FeCl ₃	99.9	75	S12
AH-rGO	50 mg L ⁻¹ , 1bar	FeCl ₃	91.6%	142.5	S13
hrGO-Trp	50 mg L ⁻¹ , 1bar	FeCl ₃	98.2%	191.0	S14
	50 mg L ⁻¹ , 1bar	FeCl ₃	99.9%	219.7	
F-GO	50 mg L ⁻¹ , 1bar	CrCl ₃	99.7%	198.4	This Work
	50 mg L ⁻¹ , 1bar	CuSO ₄	99.4%	199.1	
F-GO (50 nm)	50 mg L ⁻¹ , 1bar	FeCl ₃	99.0%	305.0	

PS9. Water permeances and rejection for the F-GO membranes with different anions.

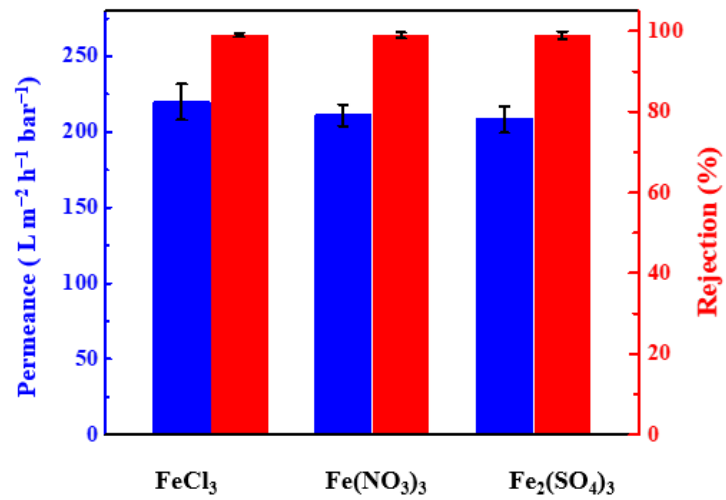


Fig. S11. Water permeances and rejection for the F-GO membranes with different anions.

PS10. The rejections and water permeances of F-GO membrane for different concentrations of FeCl₃ solutions.

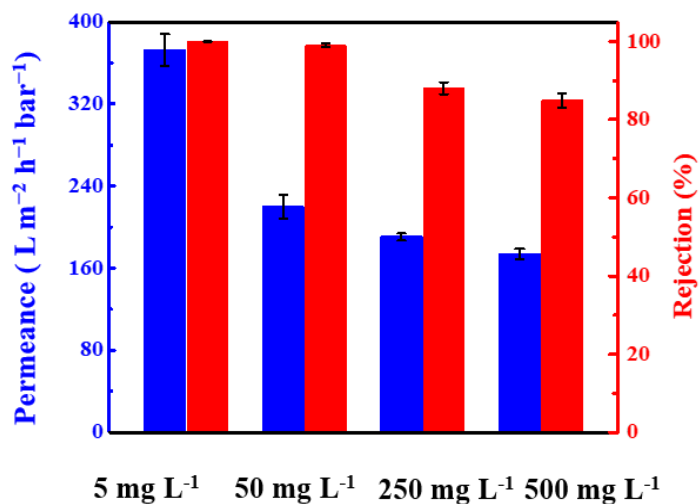


Fig. S12 The rejections and water permeances of F-GO membrane for different concentrations of FeCl_3 solutions.

PS11. The long-term cross flow stability of the F-GO membrane.

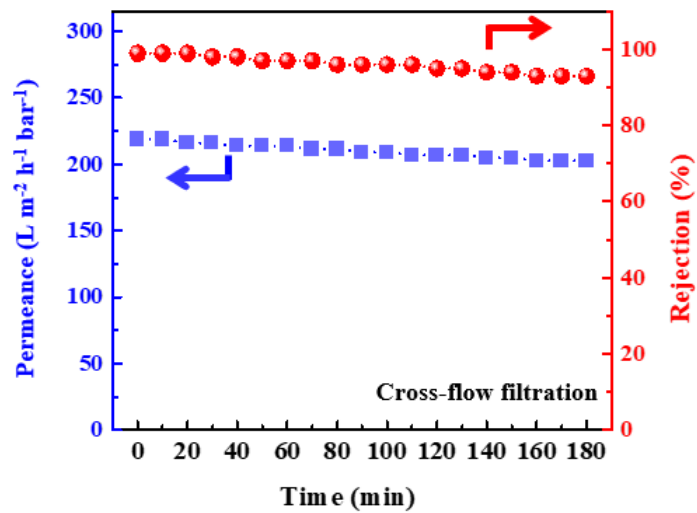


Fig. S13. The long-term cross flow stability of the F-GO membrane for separation of 50 mg L⁻¹ FeCl₃ solution.

References

- S 1 P. Zhang, J.-L. Gong, G.-M. Zeng, C.-H. Deng, H.-C. Yang, H.-Y. Liu and S.-Y. Huan, *Chem. Eng. J.*, 2017, **322**, 657-666.
- S 2 T. Wang, H. Qiblawey, E. Sivaniah and A. Mohammadian, *J. Membr. Sci.*, 2016, **511**, 65-75.
- S 3 Y. Zhang, S. Zhang, J. Gao and T.-S. Chung, *J. Membr. Sci.*, 2016, **515**, 230-237.
- S 4 R. Mukherjee, P. Bhunia and S. De, *Chem. Eng. J.*, 2016, **292**, 284-297.
- S 5 J.-H. Jang, J. Y. Woo, J. Lee and C.-S. Han, *Environ. Sci. Technol.*, 2016, **50**, 10024-10030.
- S 6 X. Liu, P. Feng, L. Zhang and Y. Chen, *Polym. Adv. Technol.*, 2020, **31**, 665-674.
- S 7 *J. Membr. Sci.*, 2016, **503**, 69-80.
- S 8 R. S. Hebbar, A. M. Isloor, K. Ananda and A. F. Ismail, *J. Mater. Chem. A*, 2016, **4**, 764-774.
- S 9 M. Zhang, K. Guan, Y. Ji, G. Liu, W. Jin and N. Xu, *Nat. Commun.*, 2019, **10**, 1253.
- S10 L. Gui, J. Dong, W. Fang, S. Zhang, K. Zhou, Y. Zhu, Y. Zhang and J. Jin, *Nano Lett.*, 2020, **20**, 5821-5829.
- S11 R. Yang, Y. Fan, R. Yu, F. Dai, J. Lan, Z. Wang, J. Chen and L. Chen, *J. Membr. Sci.*, 2021, **635**, 119437.
- S12 F. Dai, F. Zhou, J. Chen, S. Liang, L. Chen and H. Fang, *J. Mater. Chem. A*, 2021, **9**, 10672-10677.
- S13 F. Dai, R. Yu, R. Yi, J. Lan, R. Yang, Z. Wang, J. Chen and L. Chen, *Chem. Commun.*, 2020, **56**, 15068-15071.
- S14 J. Liu, S. Wang, R. Yang, L. Li, S. Liang and L. Chen, *J. Membr. Sci.*, 2022, 120745.