Supplementary Information (SI)

High-flux nanofiltration membranes tailored by bio-inspired co-deposition of hydrophilic g-C_3N_4 nanosheets for enhanced selectivity towards organics and salts

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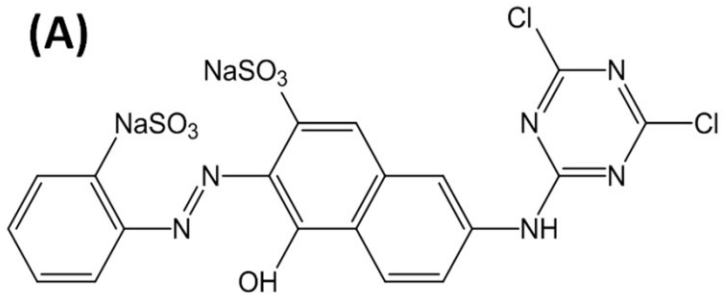
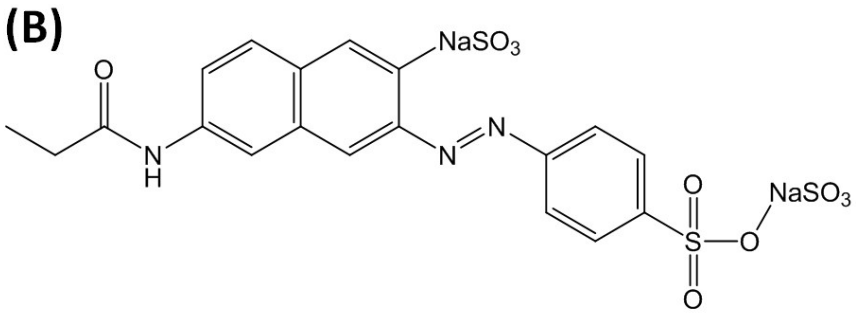


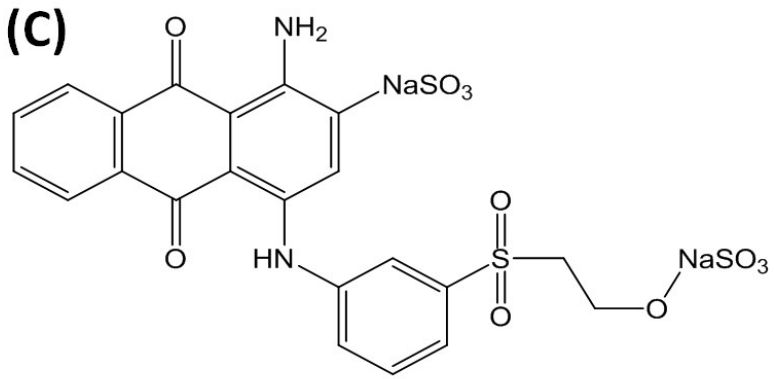
Reactive orange 1 (RO1)

($C_{19}H_{10}Cl_2N_6Na_2O_7S_2$, 615.3 Da)



Reactive orange 16 (RO16)

($C_{20}H_{17}N_3Na_2O_{11}S_3$, 617.5 Da)



Reactive blue 19 (RB19)

($C_{22}H_{16}N_2Na_2O_{11}S_3$, 626.6 Da)

Fig. S1 Chemical structures of the reactive dyes tested in the study. (A): Reactive orange 1; (B): reactive orange 16; (C): reactive blue 19.

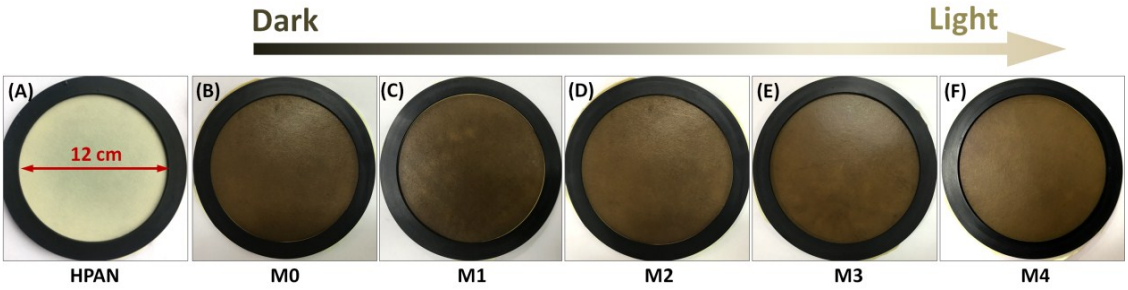


Fig. S2 Digital images of the pristine HPAN substrate and the modified membranes through bio-inspired co-deposition of the hydrophilic g-C_3N_4 nanosheets. (A): Pristine HPAN; (B): M0; (C): M1; (D): M2; (E): M3; (F): M4.

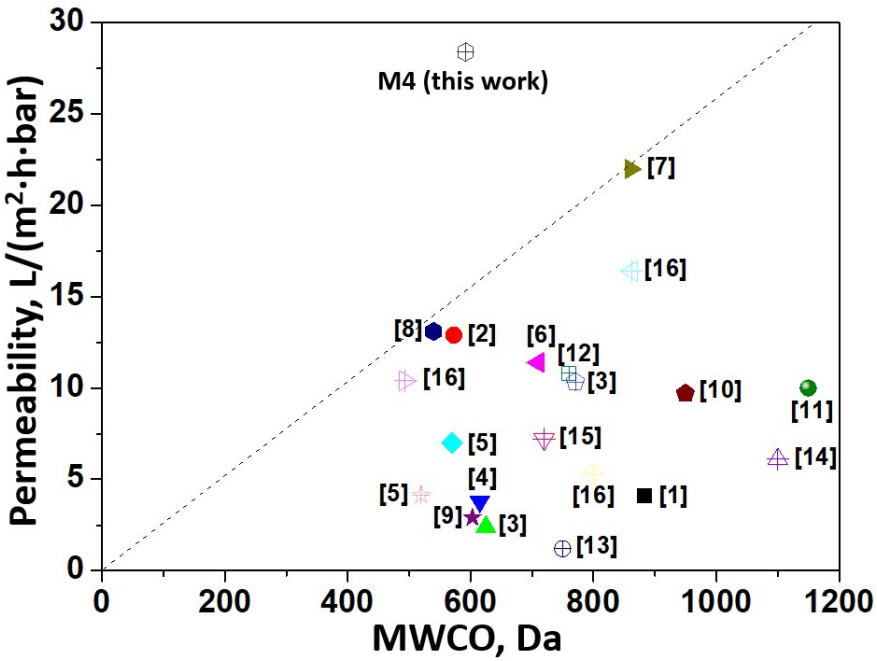


Fig. S3 Summary of the filtration performance of the state-of-the-art NF membranes reported in literature in consideration of permeability and MWCO ([1] refers to di-quaternized poly(sulfone-co-ethernitrile) random copolymer NF membrane[1]; [2] refers to cellulose nanocrystals-based thin-film composite (TFC) NF membrane[2]; [3] refers to graphene oxide incorporated TFC NF membrane[3]; [4] refers to phosphorylated chitosan NF membrane[4]; [5] refers to asymmetric cellulose acetate and composite polyamide NF5; [6] refers to TFC NF membrane mediated by polydopamine[6]; [7] refers to amphiphilic NF membrane grafted by triethylenetetramine and 2,2,3,4,4,4-hexafluorobutyl methacrylate[7]; [8] refers to alginate coated NF membrane[8]; [9] refers to polyethyleneimine based NF membrane[9]; [10] refers to penta-block copolymer based NF membrane[10]; [11] refers to zirconia-based NF membrane[11]; [12] refers to sodium carboxymethyl cellulose based hollow fiber NF membrane[12]; [13] refers to PEEK NF membrane[13]; [14] refers to PAEK-COOH-PEI NF membrane[14]; [15] refers to catechin-modified chitosan NF membrane[15]; [16] refers to commercial NF 6, NF 2A and NTR-7450 NF membranes[16]).

Table S1 Chemical composition of g-C_3N_4 nanosheets before and after oxygen plasma treatment

g-C_3N_4 sample	Chemical composition			
	C (%)	N (%)	O (%)	O/C
Before treatment	39.6	49.1	11.3	0.29
After treatment	32.1	42.2	25.7	0.80

Table S2 Performance comparisons between as-prepared membranes in this work and previously reported NF membranes in water permeability, dye retention, and salt permeation

Membrane	Permeability ($LMH \cdot bar^{-1}$)	Dye species	Dye rejection	Salt rejection		Ref.
				NaCl	Na_2SO_4	
TFN-mZIF2 (-)	14.90	Reactive blue 2	99.2%	12.0%	90.0%	17
TMC-PEI (511 Da)	9.5	Chromotrope FB	98.8%	49.0%	75.9%	18
BHAC-PIP (570 Da)	15.3	Methyl blue	98.9%	59.6%	23.4%	19
PA-PP (570 Da)	7.0	Reactive black 5	99.6%	65.0%	98.5%	20
VES/AgCl-PEI (681 Da)	10.6	Crystal violet	99.2%	8.3%	12.8%	21
SiO_2-PSS/PES (655 Da)	23.3	Reactive black 5	92.0%	3.0%	10.5%	22
TMC-Sericin (880 Da)	11.9	Methyl blue	99.5%	40.8%	95.4%	23
PEI-g-SBMA/TMC (-)	13.2	Orange GII	90.6%	7.1%	50.4%	24
M4 (592 Da)	28.4±1.2	Reactive blue 19	99.8%	2.9%	7.6%	This work

References

1 C. Liu, Y. Sun, Z. Chen, and S. Zhang, *Sep. Purif. Technol.,* 2019, **212**, 465-473.

2 L. Bai, Y. Liu, N. Bossa, A. Ding, N. Ren, G. Li, H. Liang and M.R. Wiesner, *Environ. Sci. Technol.,* 2018, **52**, 11178-11187.

3 G.S. Lai, W.J. Lau, P.S. Goh, A.F. Ismail, N. Yusof and Y.H. Tan, *Desalination*, 2016, **387**, 14-24.

4 Y. Song, Q. Hu, T. Li, Y. Sun, X. Chen and J. Fan, *Chem. Eng. J.,* 2018, **352**, 163-172.

5 S. Yu, M. Liu, M. Ma, M. Qi, Z. Lü and C. Gao, J. Membr. Sci., 2010, **350**, 83-91.

6 Y. Li, Y. Su, J. Li, X. Zhao, R. Zhang, X. Fan, J. Zhu, Y. Ma, Y. Liu and Z. Jiang, *J. Membr. Sci.,* 2015, **476**, 10-19.

7 R. Zhang, Y. Li, Y. Su, X. Zhao, Y. Liu, X. Fan, T. Ma and Z. Jiang, *J. Mater. Chem. A,* 2016, **4**, 7892-7902.

8 Y. Du, C. Zhang, Q. Zhong, X. Yang, J. Wu and Z. Xu, *ChemSusChem*, 2017, **10**, 2788-2795.

9 X. Wei, J. Hong, S. Zhu, J. Chen and B. Lv, *J. Mater. Sci.,* 2017, **52**, 11701-11714.

10 R. Muppalla, S.K. Jewrajka and A.V.R. Reddy, *Sep. Purif. Technol.,* 2015, **143**, 125-134.

11 Y. Lv, H. Yang, H. Liang, L. Wan and Z. Xu, *J. Membr. Sci.,* 2016, **500**, 265-271.

12 S. Yu, Y. Zheng, Q. Zhou, S. Shuai, Z. Lü and C. Gao, *Desalination*, 2012, **298**, 49-58.

13 J. Da Silva Burgal, L. Peeva, P. Marchetti and A. Livingston, *J. Membr. Sci.,* 2015, **493**, 524-538.

14 C. Liu, W. Bi, D. Chen, S. Zhang and H. Mao, *Chin. J. Chem. Eng.,* 2017, **25**, 1685-1694.

15 S. Liu, Z. Wang and P. Song, *ACS Sustain. Chem. Eng.,* 2018, **6**, 4253-4263.

16 J. Lin, C.Y. Tang, C. Huang, Y.P. Tang, W. Ye, J. Li, J. Shen, R. Van den Broeck, J. Van Impe, A. Volodin, C. Van Haesendonck, A. Sotto, P. Luis and B. Van der Bruggen, *J. Membr. Sci.,* 2016, **501**, 1-14.

17 J. Zhu, L. Qin, A. Uliana, J. Hou, J. Wang, Y. Zhang, X. Li, S. Yuan, J. Li, M. Tian, J. Lin and B. Van der Bruggen, *ACS Appl. Mater. Interfaces* 2017, **9**, 1975-1986.

18 X. Wei, S. Wang, Y. Shi, H. Xiang and J. Chen, *Ind. Eng. Chem. Res.,* 2014, 53, 14036-14045.

19 T. Wang, Y. Yang, J. Zheng, Q. Zhang and S. Zhang, *J. Membr. Sci.,* 2014, **448**, 180-189.

20 S. Yu, M. Liu, M. Ma, M. Qi, Z. Lü and C. Gao, *J. Membr. Sci.,* 2010, **350**, 83-91.

21 S. Liu, Z. Wang, M. Ban, P. Song, X. Song and B. Khan, *J. Membr. Sci.,* 2018, **566**, 168-180.

22 L. Xing, N. Guo, Y. Zhang, H. Zhang and J. Liu, *Sep. Purif. Technol.,* 2015, **146**, 50-59.

23 C. Zhou, Y. Shi, C. Sun, S. Yu, M. Liu and C. Gao, *J. Membr. Sci.,* 2014, **471**, 381-391.

24 T. Ma, Y. Su, Y. Li, R. Zhang, Y. Liu, M. He, Y. Li, N. Dong, H. Wu, Z. Jiang, *J. Membr. Sci.,* 2016, **503**, 101-109.