

## Supplementary information

# Electrospray interfacial polymerization for a loose NF membrane: Super-selective dye separation in saline dye wastewater treatment

Yesol Kang<sup>a</sup>, Jaewon Jang<sup>a</sup>, Yunho Lee<sup>a</sup>, and In S. Kim<sup>a,\*</sup>

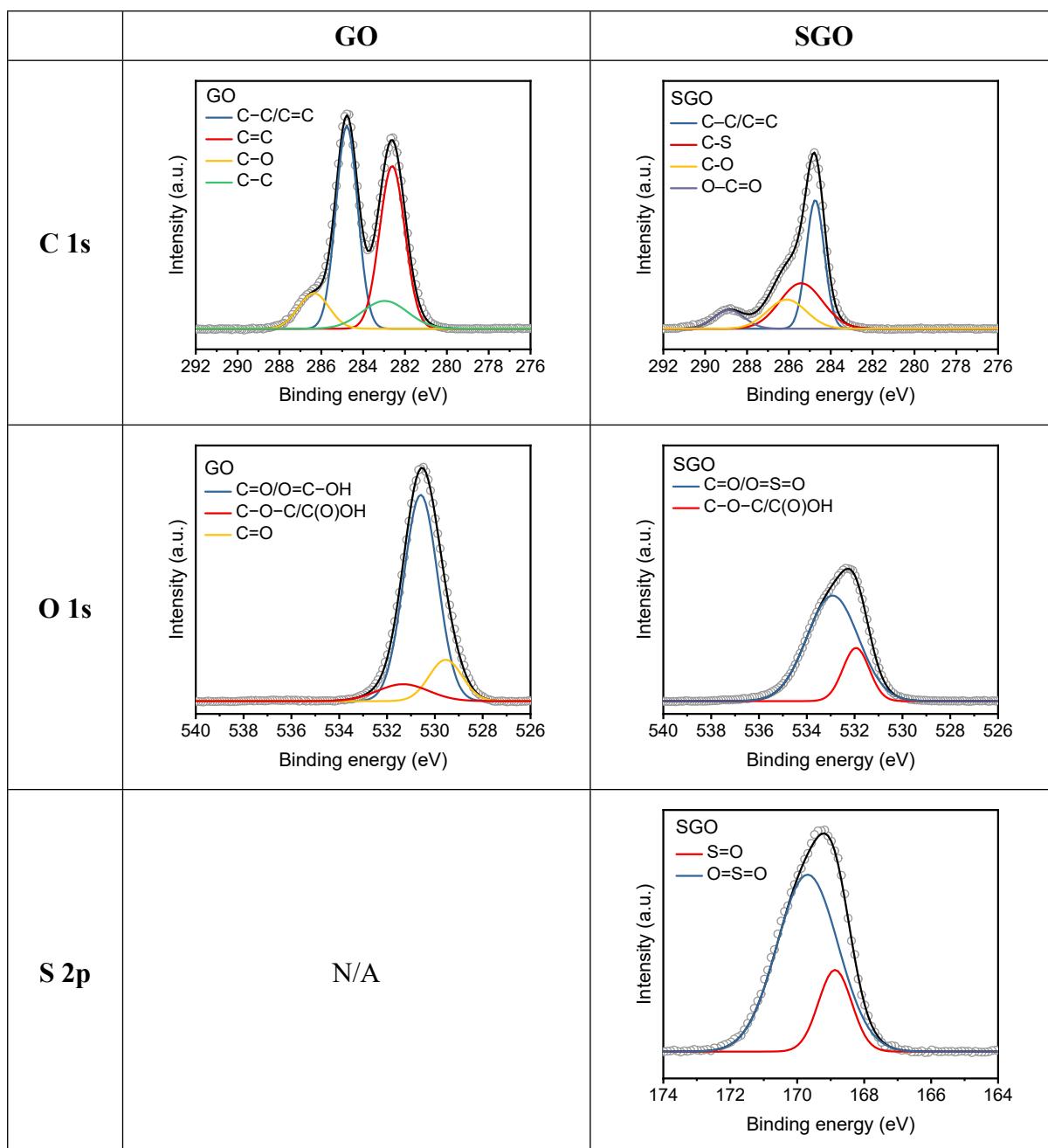
<sup>a</sup>School of Earth Sciences and Environmental Engineering (SESE), Gwangju Institute of Science and Technology (GIST), 123 Cheomdangwagi-ro, Buk-gu, Gwangju 61005, Republic of Korea

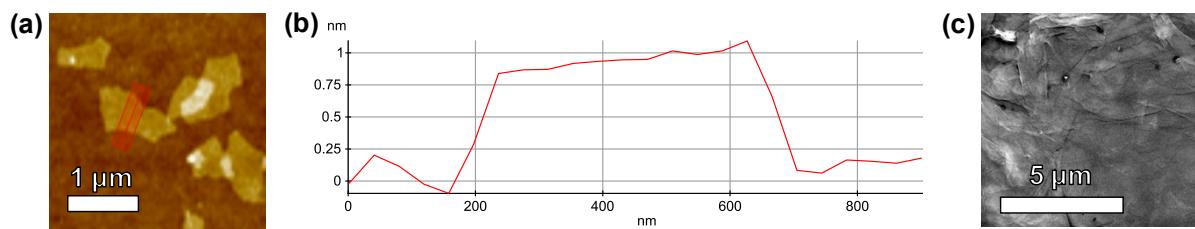
\* Corresponding author: Tel: +82-62-715-2436, e-mail: iskim@gist.ac.kr

**Table S1.** Characteristics of the dye molecules used in this study

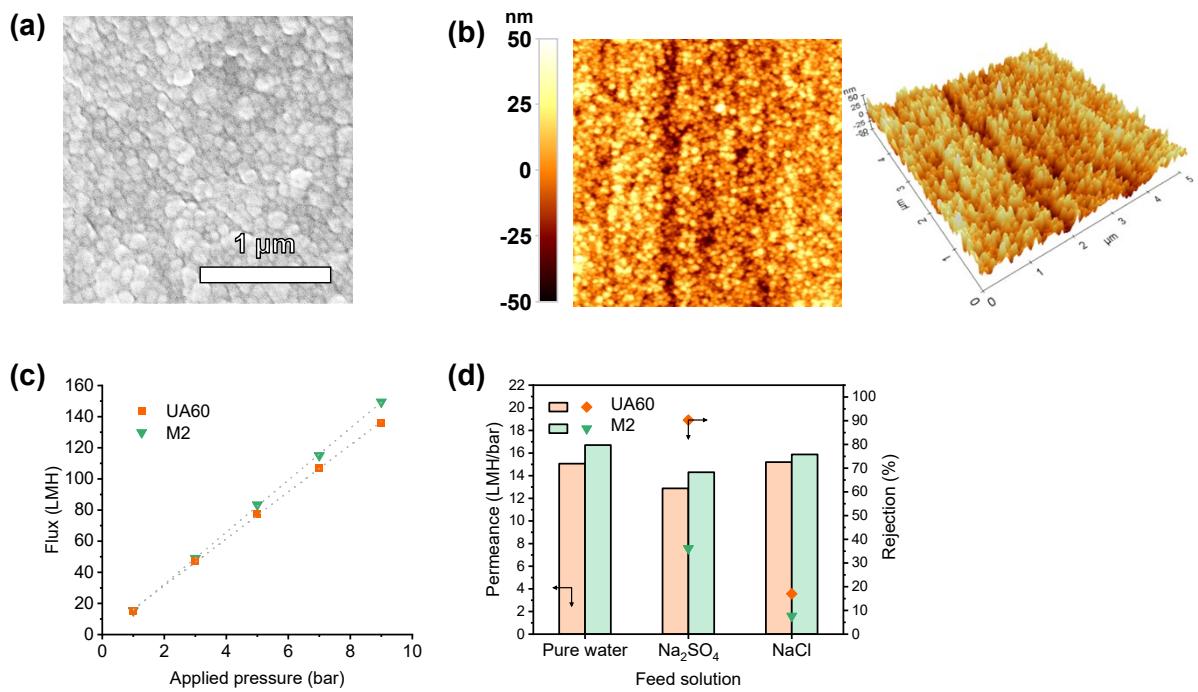
Dye name	Molecular formula	Molecular structure	Molecular weight (g mol <sup>-1</sup> )	Nature	UV adsorption (nm)
Evans blue	C <sub>34</sub> H <sub>24</sub> N <sub>6</sub> Na <sub>4</sub> O <sub>14</sub> S <sub>4</sub>		960.8	Negative	606
Congo red	C <sub>32</sub> H <sub>22</sub> N <sub>6</sub> Na <sub>2</sub> O <sub>6</sub> S <sub>2</sub>		696.7	Negative	495
Methyl orange	C <sub>14</sub> H <sub>14</sub> N <sub>3</sub> NaO <sub>3</sub> S		327.3	Negative	461

**Table S2.** C 1s, O 1s, and S 2p core-level XPS spectra of GO, and SGO.

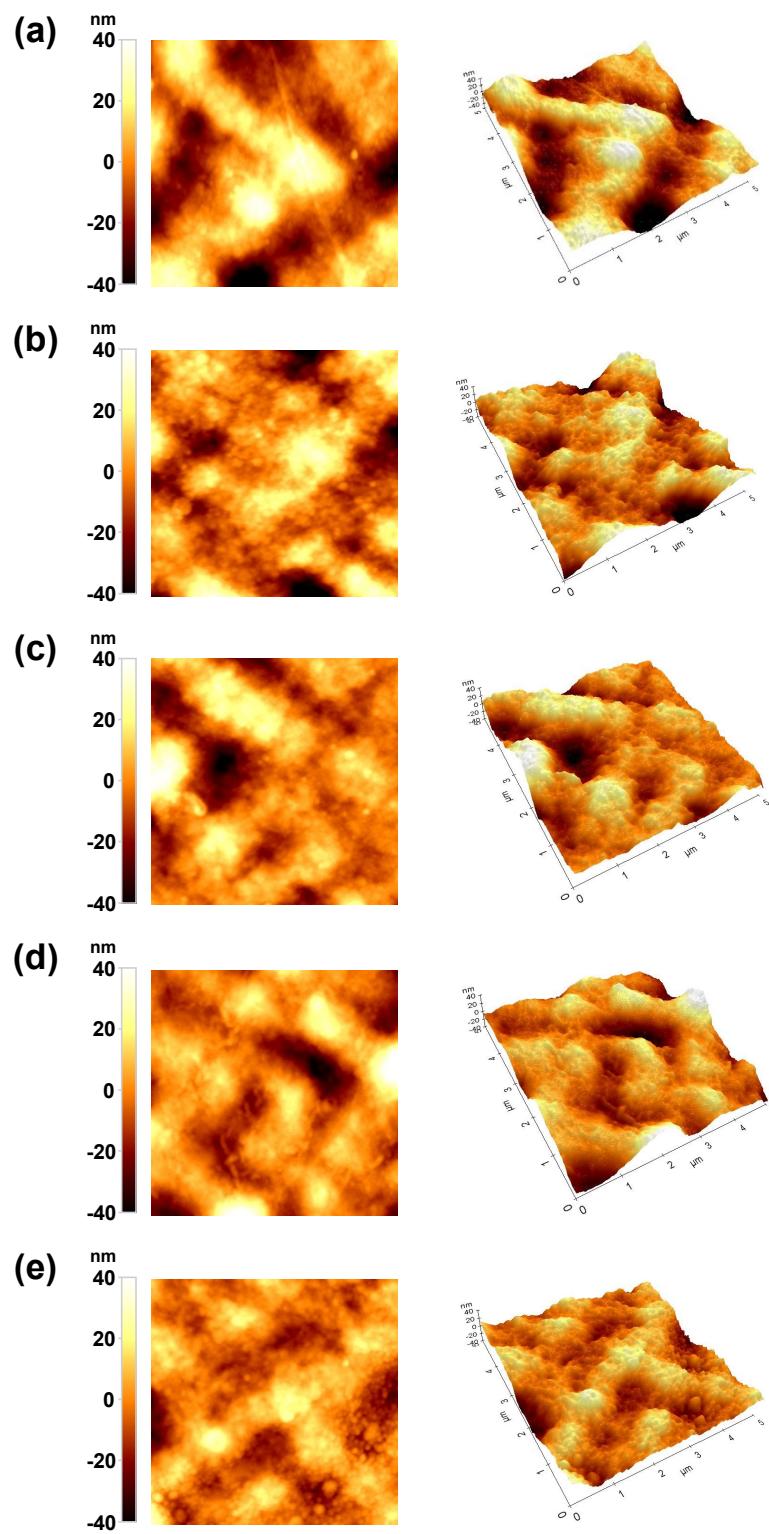




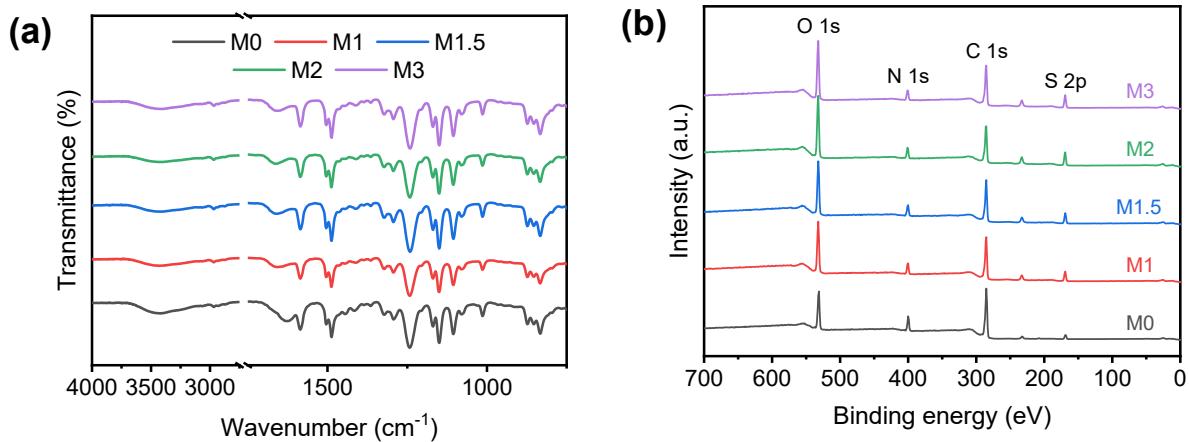
**Fig. S1.** (a) A tapping mode AFM image of SGO sheets and (b) the height profile of the AFM image. (c) The SEM image of SGO sheet at a magnification of  $\times 10\text{k}$ .



**Fig. S2.** (a) SEM surface images, (b) AFM surface scanning images of UA60 membrane, (c) pure water flux depending on the applied pressure (UA60 to  $15.1 \text{ LMH bar}^{-1}$ ), and (d) permeance and salt rejection comparison of UA60 and M2 under operating pressure at 5 bar.



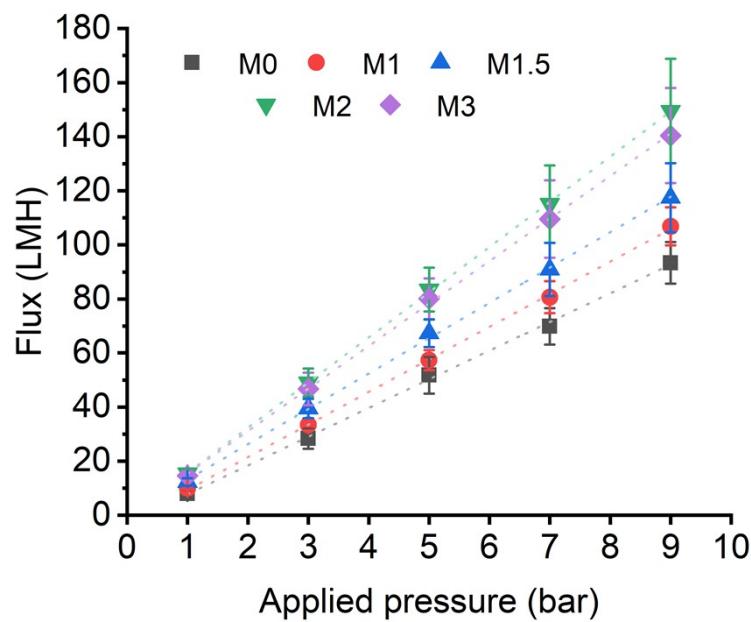
**Fig. S3.** Surface AFM images of SGO contained membranes: (a) M0, (b) M1, (c) M1.5, (d) M2, and (e) M3.



**Fig. S4.** (a) ATR-FTIR and (b) XPS spectra of the prepared membranes.

**Table S3.** The atomic percentage of element (%) obtained from XPS of membranes

Membrane	Elemental ratio (%)			
	C 1s	N 1s	O 1s	S 2p
M0	66.4	10.9	20.1	2.6
M1	58.8	8.4	27.2	5.6
M1.5	56.1	8.9	28.8	6.2
M2	52.1	7.9	32.2	7.8
M3	54.7	7.7	30.3	7.3



**Fig. S5.** Membrane flux differences depending on the applied pressure and membrane; PWP values of membranes are  $11.0 \pm 0.8$ ,  $12.1 \pm 0.9$ ,  $13.1 \pm 1.5$ ,  $16.7 \pm 2.2$ , and  $15.7 \pm 2.0$  LMH bar $^{-1}$ , corresponding to M0, M1, M1.5, M2, and M3.

**Table S4.** Comparison of the permeance, dye rejection, and salt rejection.

Membrane	Permeance (LMH bar <sup>-1</sup> )	Dye	Dye rejection (%)	Salt	Salt rejection (%)	Ref.
GO-PCS	13.2	Direct black 38	99.7	NaCl Na <sub>2</sub> SO <sub>4</sub>	56.8 87.8	<sup>1</sup>
(PEI-modified GO)/PAA/PV A/GA	0.84	Congo red Methyl blue	99.5 99.3	NaCl	37.8	<sup>2</sup>
GOQDs-1	11.7	Congo red Methyl blue	99.8 97.6	NaCl	17.2	<sup>3</sup>
GO(120) NFM	11.1	Methyl blue	98.88	NaCl	27.86	<sup>4</sup>
GO@PAN	5	Congo red	99.9	NaCl Na <sub>2</sub> SO <sub>4</sub>	9.8 56.7	<sup>5</sup>
PDA-rGOC2	16.8	Congo red	99.6	NaCl	43.2	<sup>6</sup>
PAN/GO	5.5	Acid red 18 Methyl blue	99.8 100	NaCl MgCl <sub>2</sub>	10.9 11.3	<sup>7</sup>
PES/Metformin/GO/Fe <sub>3</sub> O <sub>4</sub>	9.02	Direct red 16	99	NaCl	15	<sup>8</sup>
HNTs-poly(NASS)/PES	10.8	Reactive black 5	85.5	NaCl Na <sub>2</sub> SO <sub>4</sub>	6.9 12	<sup>9</sup>
Polypiperazine-amide NF	7.0	Reactive black 5	99.3	NaCl Na <sub>2</sub> SO <sub>4</sub>	66.7 98.9	<sup>10</sup>
UA60	15.1	EB CR	99.8 99.5	NaCl Na <sub>2</sub> SO <sub>4</sub>	17.0 90.3	
M0	11.0	EB CR	99.5 99.9	NaCl Na <sub>2</sub> SO <sub>4</sub>	24.5 52.8	This work
M2	16.7	EB CR	99.6 99.8	NaCl Na <sub>2</sub> SO <sub>4</sub>	7.7 36.1	

**References**

- 1 Y. Song, Y. Sun, M. Chen, P. Huang, T. Li, X. Zhang and K. Jiang, Efficient removal and fouling-resistant of anionic dyes by nanofiltration membrane with phosphorylated chitosan modified graphene oxide nanosheets incorporated selective layer, *J. Water Process Eng.*, 2020, **34**, 101086.
- 2 N. Wang, S. Ji, G. Zhang, J. Li and L. Wang, Self-assembly of graphene oxide and polyelectrolyte complex nanohybrid membranes for nanofiltration and pervaporation, *Chem. Eng. J.*, 2012, **213**, 318–329.
- 3 C. Zhang, K. Wei, W. Zhang, Y. Bai, Y. Sun and J. Gu, Graphene Oxide Quantum Dots Incorporated into a Thin Film Nanocomposite Membrane with High Flux and Antifouling Properties for Low-Pressure Nanofiltration, *ACS Appl. Mater. Interfaces*, 2017, **9**, 11082–11094.
- 4 L. Chen, J. H. Moon, X. Ma, L. Zhang, Q. Chen, L. Chen, R. Peng, P. Si, J. Feng, Y. Li, J. Lou and L. Ci, High performance graphene oxide nanofiltration membrane prepared by electrospraying for wastewater purification, *Carbon*, 2018, **130**, 487–494.
- 5 J. Wang, P. Zhang, B. Liang, Y. Liu, T. Xu, L. Wang, B. Cao and K. Pan, Graphene Oxide as an Effective Barrier on a Porous Nanofibrous Membrane for Water Treatment, *ACS Appl. Mater. Interfaces*, 2016, **8**, 6211–6218.
- 6 J. Zhu, J. Wang, A. A. Uliana, M. Tian, Y. Zhang, Y. Zhang, A. Volodin, K. Simoens, S. Yuan, J. Li, J. Lin, K. Bernaerts and B. Van Der Bruggen, Mussel-Inspired Architecture of High-Flux Loose Nanofiltration Membrane Functionalized with Antibacterial Reduced Graphene Oxide-Copper Nanocomposites, *ACS Appl. Mater. Interfaces*, 2017, **9**, 28990–29001.
- 7 Z. Qiu, X. Ji and C. He, Fabrication of a loose nanofiltration candidate from Polyacrylonitrile/Graphene oxide hybrid membrane via thermally induced phase separation, *J. Hazard. Mater.*, 2018, **360**, 122–131.
- 8 G. Abdi, A. Alizadeh, S. Zinadini and G. Moradi, Removal of dye and heavy metal ion using a novel synthetic polyethersulfone nanofiltration membrane modified by magnetic graphene oxide/metformin hybrid, *J. Membr. Sci.*, 2018, **552**, 326–335.
- 9 J. Zhu, N. Guo, Y. Zhang, L. Yu and J. Liu, Preparation and characterization of negatively charged PES nanofiltration membrane by blending with halloysite nanotubes grafted with poly (sodium 4-styrenesulfonate) via surface-initiated ATRP, *J. Membr. Sci.*, 2014, **465**, 91–99.
- 10 S. Yu, M. Liu, M. Ma, M. Qi, Z. Lü and C. Gao, Impacts of membrane properties on reactive dye removal from dye/salt mixtures by asymmetric cellulose acetate and composite polyamide nanofiltration membranes, *J. Membr. Sci.*, 2010, **350**, 83–91.

**Table S5.** BSA adsorption capability, and FRR of the fabricated membranes

Membrane	BSA adsorption capability ( $\mu\text{g cm}^{-2}$ )	FRR (%)	
		2 <sup>nd</sup> cycle	3 <sup>rd</sup> cycle
M0	33.9 $\pm$ 3.3	88.5	87.9
M2	10.5 $\pm$ 6.1	96.4	97.9