Electronic Supplementary Information (ESI)

## Engineering Defects into Nickel-based Nanosheets for Enhanced Water

Permeability

Huixiang Ang<sup>a</sup> and Liang Hong<sup>\*a</sup>

<sup>a</sup>Department of Chemical and Biomolecular Engineering, National University of Singapore, 4 Engineering Drive 4, Singapore 117585, Singapore

\*Corresponding author. Email: chehongl@nus.edu.sg

Tel: +65 65165029

## **Experimental Section**

**Preparation of Ni-based nanosheets.** In a typical defect-rich nanosheets synthesis, nickel nitrate hexahydrate (24 mmol, Ni(NO<sub>3</sub>)<sub>2</sub>.6H<sub>2</sub>O) and ethanolamine (1 mmol, EA) were dissolved in deionized water (1 mL) with 5 min sonication. Then, the solution was left undisturbed and aged for 48 h under ambient conditions. The final product Ni(OH)<sub>2</sub> was washed with water for several times and dried at 60 °C. Other defect-rich Ni-based nanosheets such NiO, NiS<sub>2</sub>, and Ni<sub>2</sub>P nanosheets were also prepared by the same method. Additionally, NiO synthesis was prepared by annealing the Ni(OH)<sub>2</sub> product in the tube furnace at 400 °C for 16 h under argon atmosphere. For the NiS<sub>2</sub> and Ni<sub>2</sub>P syntheses, S (i.e. pure sulfur, S) and P (i.e. sodium hypophosphite, NaH<sub>2</sub>PO<sub>2</sub>) precursors with their corresponding molar ratio (i.e. Ni:S ratio = 1:2; Ni<sub>2</sub>P ratio = 2:1) were placed at the upstream while the Ni(OH)<sub>2</sub> nanosheets at the downstream followed by annealing at 400 °C for 16 h under argon atmospheres at the downstream followed by annealing at 400 °C for 16 h under argon atmosphere. All the materials were purchased from Sigma-Aldrich and used without further purification. For control experiments, the synthesis of defect-free Ni(OH)<sub>2</sub> nanosheets and the preparation of thicker defect-rich Ni(OH)<sub>2</sub> assemblies, 2 mmol and 0.5 mmol of EA were added into the growth solution containing 24 mmol of Ni(NO<sub>3</sub>)<sub>2</sub>.6H<sub>2</sub>O, respectively, using the same method mentioned above.

**Evaluation of Separation Performances.** For membrane fabrication, the respective diluted 2D Ni-based dispersions (ca. 0.20 mg mL<sup>-1</sup>) were cast on the nylon support through vacuum filtration. Molecular sieving measurements were carried using dead-end filtration device. All the filtration tests were stirred at 500 rpm to minimize concentration polarization and the error bars of permeances and rejection rates were attained based on three sampling tests per membrane. The effective area was estimated to be 1.3 cm<sup>2</sup>. All the filtration tests were performed at an operating pressure of 0.5 bar unless otherwise stated. The tested probes (i.e. Methyl Red (MR), Methyl Orange (MO), Rhodamine B (RB), and Evans Blue (EB)) were all purchased from Sigma-Aldrich and 10 mg L<sup>-1</sup> of each probe solute was dissolved homogeneously in deionized water before filtering through the membrane. Finally, all the concentration of the feed, permeate, and retentate solutions were characterized by using UV-Vis spectrophotometer.

The permeance (J) was calculated using Equation (1):

$$J = \frac{V}{A \times t \times \Delta P} \tag{1}$$

where A is the effective area of the membrane, t is the filtration time,  $\Delta P$  is the pressure drop, and V is the filtrate volume.

The rejection efficiency (*R%*) was calculated using Equation (2):

$$R = \frac{C_o - C_F}{C_o} \times 100\%$$
 (2)

where  $C_o$  is the original feed concentration and  $C_F$  is the final permeate concentration.

**Characterization**. X-ray diffraction (XRD) data were collected on the Shimadzu thin film diffractometer with Cu-K $\alpha$  irradiation ( $\lambda = 1.5406$  Å). The transmission electron microscopy (TEM) was performed on JEOL-2100F field emission electron microscope operating at an acceleration voltage of 200 kV. The field emission scanning electron microscopy (FESEM) images were taken on a JEOL-6700F SEM equipped with energy-dispersive X-ray spectroscopy (EDX) analyzer (Oxford Instruments, model 7426) for elemental mapping. X-ray photoelectron spectroscopy (XPS) analyses were carried out using an AXIS-Hsi, Kratos Analytical X-ray photoelectron spectrometer with an excitation source of Al  $K\alpha = 1486.71$  eV. Carbon-hydrogennitrogen-sulphur (CHNS) elemental analyses were performed using Elementar Vario Micro Cube. Atomic force microscopy (AFM) characterizations were performed on Bruker Multimode 8 equipment. Thermogravimetric analyses were obtained on a TGA Q500 at a heating rate of 10 °C min<sup>-1</sup>. The UV-vis measurements were recorded using Biochrom Libra S32 UV-Vis spectrometer.

**Computational methods.** The molecular solutes were first constructed using Materials Studio 8.0 followed by employing the DND basis set (double numerical basis set with d-type polarization functions) for geometry optimization using the DMol3 program.<sup>S1</sup> Then we estimated the molecular volume and dimensions of the solute by using the Connolly accessible solvent surface method at probe radius of 1.4 Å.<sup>S2</sup>



Fig. S1 (a-b) FESEM images and (c-d) TEM images of the thicker defect-rich nanosheets assemblies at different magnification. The assembly characteristic may be attributed to the aggregation of nanosheets without the protection of enough EA ligand. TEM image shows observable wrinkles containing defects on the nanosheets assemblies.



Fig. S2 TEM images of (a) defect-rich  $Ni(OH)_2$  and (b) defect-free  $Ni(OH)_2$  nanosheets.



Fig. S3 High-resolution XPS spectra of the (a) Ni 2p and (b) O 1s peaks for the defect-rich Ni(OH)<sub>2</sub> nanosheets.



Fig. S4 (a) HAADF-STEM image of defect-rich Ni(OH)<sub>2</sub> nanosheets and its corresponding (b) Ni and (O) elemental mapping images.



Fig. S5 (a) AFM image of defect-rich Ni(OH)<sub>2</sub> nanosheets and its corresponding (b) thickness distribution plot.



Fig. S6 XRD pattern of defect-free  $Ni(OH)_2$  nanosheets.



Fig. S7 (a) FESEM image; (b) TEM image; (c) AFM image and its corresponding (d) thickness distribution plot of the defectfree Ni(OH)<sub>2</sub> nanosheets.



Fig. S8 Quantitative analysis of nitrogen composition in defect-free Ni(OH)<sub>2</sub> versus defect-rich Ni(OH)<sub>2</sub> nanosheets using XPS measurements. The XPS scanning parameters are the same for both samples.



Fig. S9 (a) FESEM image; (b) AFM image and its corresponding height profiles of the defect-free Ni(OH)<sub>2</sub> selective layer.



Fig. S10 The XRD pattern of defect-free Ni(OH)<sub>2</sub> versus defect-rich Ni(OH)<sub>2</sub> selective layers.



Fig. S11 UV-vis absorption spectra of the permeate, feed, and retentate solutions for (a) MR, (b) MO, (c) RB, and EB solutes after filtration through the defect-rich Ni(OH)<sub>2</sub> membrane.



Fig. S12 TEM images of (a) NiO, (b) NiS<sub>2</sub>, and (c) Ni<sub>2</sub>P defect-rich nanosheets.



Fig. S13 XRD patterns of (a) NiO, (b) NiS<sub>2</sub>, (c) Ni<sub>2</sub>P defect-rich materials are in accordance with their JCPDS card No. 47-1049, 11-0099, and 03-0953, respectively.



Fig. S14 Separation performances of defect-rich NiO, NiS<sub>2</sub>, and Ni<sub>2</sub>P membranes.



Fig. S15 (a) FESEM image; (b) TEM image of defect-rich Ni(OH)<sub>2</sub> nanosheets; (c) FESEM image and its corresponding (d) XRD pattern of defect-rich Ni(OH)<sub>2</sub> selective layer after durability test.



Fig. S16 Increasing the pressure loading to evaluate the mechanical properties of the membranes. (a) Water flux and (b) rejection of membranes against various pressure loading.



Fig. S17 Thermal stability evaluation of the 2D materials using thermogravimetric analysis. At temperature between 50  $^{\circ}$ C to 250  $^{\circ}$ C, the Ni(OH)<sub>2</sub> materials undergo surface dehydration followed by decomposition to form NiO at 250  $^{\circ}$ C.<sup>S3</sup>

Solute	Chemical Structure	Dimensions of solute Length (nm) × Width (nm)	Molecular weight (g mol <sup>-1</sup> )	Occupied Volume (nm <sup>3</sup> )
Methyl Red	A A A A A	1.1 × 0.8	269.30	0.76
Methyl Orange	Contraction of the second	1.2 × 1.0	304.34	0.82
Rhodamine B		1.6 × 1.3	443.57	1.18
Evans Blue		2.4 × 0.9	874.88	1.94

Table S1. Chemical structure and molecular size of various solutes

 Table S2. Comparison of the separation performances with various existing membranes.

Membrane	Category	Probe solute	Occupied Volume (nm <sup>3</sup> )	Permeance (L m <sup>-2</sup> h <sup>-1</sup> bar <sup>-1</sup> )	Rejection (%)	Reference
GO	2D Carbon	Evans Blue	1.94	71	85	\$4
HPEI/S-rGO-18		Evans Blue	1.94	85.2 – 86.5	100	S5
uGNMs		Methyl Blue	1.79	21.8	99.2	S6
Nematic GO		Methyl Red	0.76	63-67	90	S7
rGO/MCNT		Rhodamine B	1.18	52.7	100	S8
Nanostrand channeled GO		Evans Blue	1.94	695	84	S9
GO/SWCNT		Rhodamine B	1.18	710	97.4	S10
g-C <sub>3</sub> N <sub>4</sub>		Evans Blue	1.94	29.5	87.2	S11
Ti <sub>3</sub> C <sub>2</sub> T <sub>x</sub>	2D Ceramic	Evans Blue	1.94	1084	90	\$12
WS <sub>2</sub>		Evans Blue	1.94	450	89	S13
Nanostrand channeled WS <sub>2</sub>		Evans Blue	1.94	930	83	S13
MoS <sub>2</sub>		Evans Blue	1.94	245	89	S14
Nickel hydroxide		Methylene Blue	0.85	99	95	S15
ZIF-8 on PES	ZIF-8 based MOF	Rhodamine B	1.18	37.5	98	\$16
Polyamide/ZIF-8		Congo Red	1.53	2.11	99.8	S17
ZIF-8/PSS		Methyl Blue	1.79	26.5	98.6	S18
NF-PES-010	Commercial	Victoria Blue	1.44	40	98	S19
Ni(OH) <sub>2</sub>	2D Defect- rich Ni(OH) <sub>2</sub>	Methyl Red	0.76	1330	96.4	This work
		Methyl Orange	0.82	1323	98.1	
		Rhodamine B	1.18	1307	98.6	
		Evans Blue	1.94	1288	98.2	

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