Electronic Supplementary Information

Salt Concentration, pH and Pressure Controlled Separation of Small Molecules through Lamellar Graphene Oxide Membranes

Hubiao Huang,[a] Yiyin Mao,[a] Yulong Ying,[a] Yu Liu,[a] Luwei Sun,[a] Xinsheng Peng[a,b]*

[a]State Key Laboratory of Silicon Materials, Department of Materials Science and Enginnering, Zhejiang University, Hangzhou 310027, China; [b]Cyrus Tang Center for Sensor Materials and Applications, Zhejiang University, Hangzhou 310027, China.

E-mail: pengxinsheng@zju.edu.cn

1. Experimental Section

Preparation of GO: The preparation of graphene oxide (GO) is typically synthesized by a modified Hummer Method.[1] As-prepared GO was centrifuged at 8000 r. p. m for ten times at least in order to remove residual acids and salts with de-ionized water. Then, the brown GO dispersion was dried in a vacuum chamber at 60 °C for three days. The obtained GO was redispersed in ultrapure water with a concentration of 0.2 mg L^{-1}. The pH of GO aqueous solution was tuned at 7 by sodium hydroxide and hydrochloride solution.

Characterization: GO membranes were fabricated by vacuum filtration using a SIBATA filtration system and the support is the polycarbonate (PC) membrane (Whatman, 25 mm diameter, 0.2 μm pore size). UV-Vis absorption spectra of the permeates and retentates were obtained by a UV-Vis spectroscopy (SHIMAZU 3600). The morphologies and structures of GO sheets were characterized by transmission electronic microscope (Tecnai G2 F20 S-TEIN) and field-emission scanning electronic microscopy (Hitachi SU-70). For AFM observation (Tapping Mode, Bruke Dimension Icon), the samples were prepared by depositing one droplet of GO suspension on single crystal Si substrate and dried in ambient temperature. The GO membrane was placed on a filter holder (Millipore P158, 25 mm) separation for

pressure testing with effective area of 2.83 cm^2.

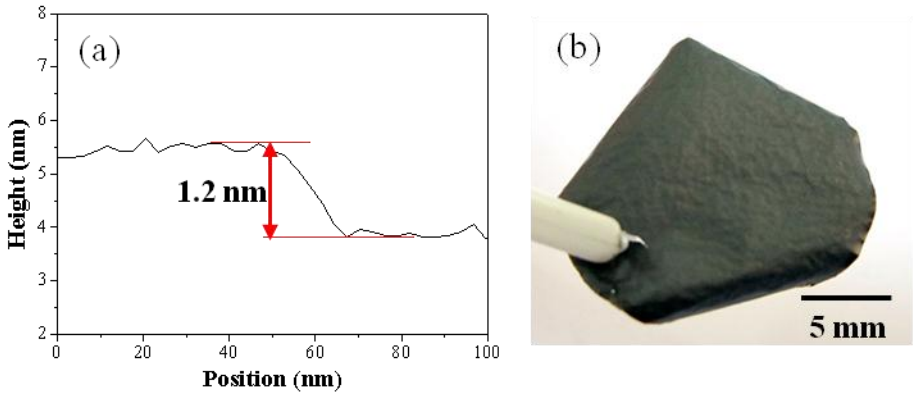


Figure S1 (a) the thickness profile from the AFM image as marked in main text Figure 1a. (b) Photo image of the free-standing GO membrane prepared from 3 ml 0.02% GO dispersion.

2. Separation performance of GO membranes with different thickness

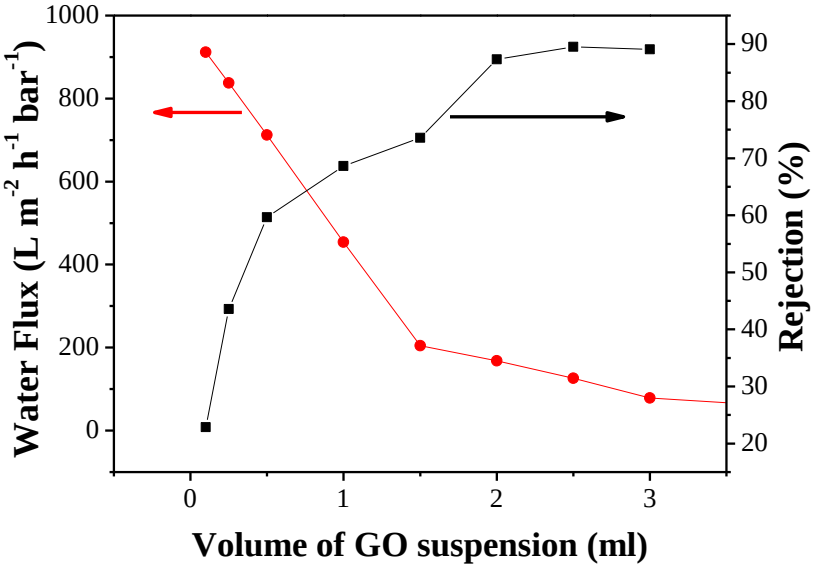


Fig. S2. The water flux and rejection rate of EB molecules of GO membrane prepared from different volume of 0.02 mg ml^{-1} GO suspension.

3. The water flux and rejection rate of EB molecules of LGO membrane depending on pH

Table S1. The separation performances of GO membranes at different pH

pH Value	3	4	5	6	7	8	9	10	11	12
Water Flux ($L/h \cdot m^2 \cdot bar$)	12.19	15.68	37.00	71	63.43	59.64	39.21	31.72	23.41	18.88
EB rejection (%)	100	96.98	90.07	85	87.24	83.69	79.29	81.99	94.24	96.63

4. Zeta potential of GO suspension depends on pH

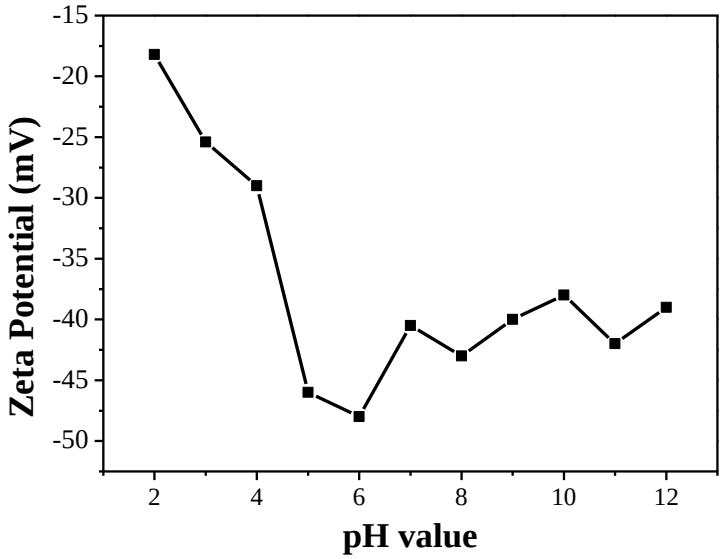
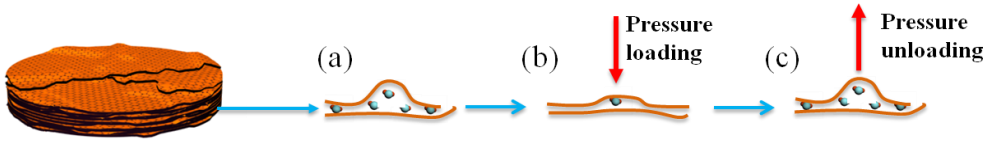


Fig. S3. Zeta potential of 0.02 mg ml^{-1}GO suspension depends on pH.

5. Schematic illustration of the pressure loading and releasing process:



Scheme S1. Schematic illustration of the pressure loading and releasing process: (a) initial state; (b) pressure loading and shrinking of the channel; (c) pressure releasing and recovery of the channel.

Reference:

1 Y. X. Xu, H. Bai, G. W. Lu, C. Li and G. Q. Shi, *J. Am. Chem. Soc.*, 2008, **130**, 5856–5857.