

Electronic Supplementary Information for

## **Preparation of freestanding graphene-based laminar membrane for clean-water intaking via forward osmosis process**

*Feng Yan,<sup>a,b, †</sup> Chuhong Yu,<sup>a, †</sup> Bowu Zhang,<sup>a, \*</sup> Tao Zou,<sup>a</sup> Hongwei Zhao<sup>a</sup> and Jingye Li<sup>a, \*</sup>*

<sup>a</sup> Shanghai Institute of Applied Physics, Chinese Academy of Sciences, No. 2019, Jialuo Rd., Shanghai 201800, China

<sup>b</sup> University of Chinese Academy of Sciences, No. 19A, Yuquan Rd., Beijing, 100049, China

<sup>†</sup>These authors contributed equally to this work.

Correspondence and requests for materials should be addressed to Dr. B. Zhang ([zhangbowu@foxmail.com](mailto:zhangbowu@foxmail.com)) and Prof. J. Li ([jingyeli@sinap.ac.cn](mailto:jingyeli@sinap.ac.cn)).

## **Experimental**

### **Material and reagents**

Natural graphite powder (average powder size, 500  $\mu\text{m}$ ) was purchased from Sigma-Aldrich Co. LLC. The concentrated sulfuric acid ( $\text{H}_2\text{SO}_4$ , 98%), concentrated nitric acid ( $\text{HNO}_3$ ), phosphorus pentoxide ( $\text{P}_2\text{O}_5$ ), potassium persulfate ( $\text{K}_2\text{S}_2\text{O}_8$ ), potassium permanganate ( $\text{KMnO}_4$ ), hydrogen peroxide solution ( $\text{H}_2\text{O}_2$ , 30%), hydrochloric acid ( $\text{HCl}$ , 36-38%), propandioic acid (PA), sodium chloride ( $\text{NaCl}$ ), methyl orange (MO, Mw 327.3), rhodamine B (RB, Mw 479.0) were all purchased from Sinopharm Chemical Reagent Co., Ltd. All reagents were used as received without any purification. Ultrapure water used for all the experiment in this paper was made from Milli-Q purification system (Millipore, USA). Cellulose acetate (CA) membranes (pore size, 0.22  $\mu\text{m}$ ) were provided by Shanghai Xingya Purifying Material Factory.

### **Preparation of GO Dispersion**

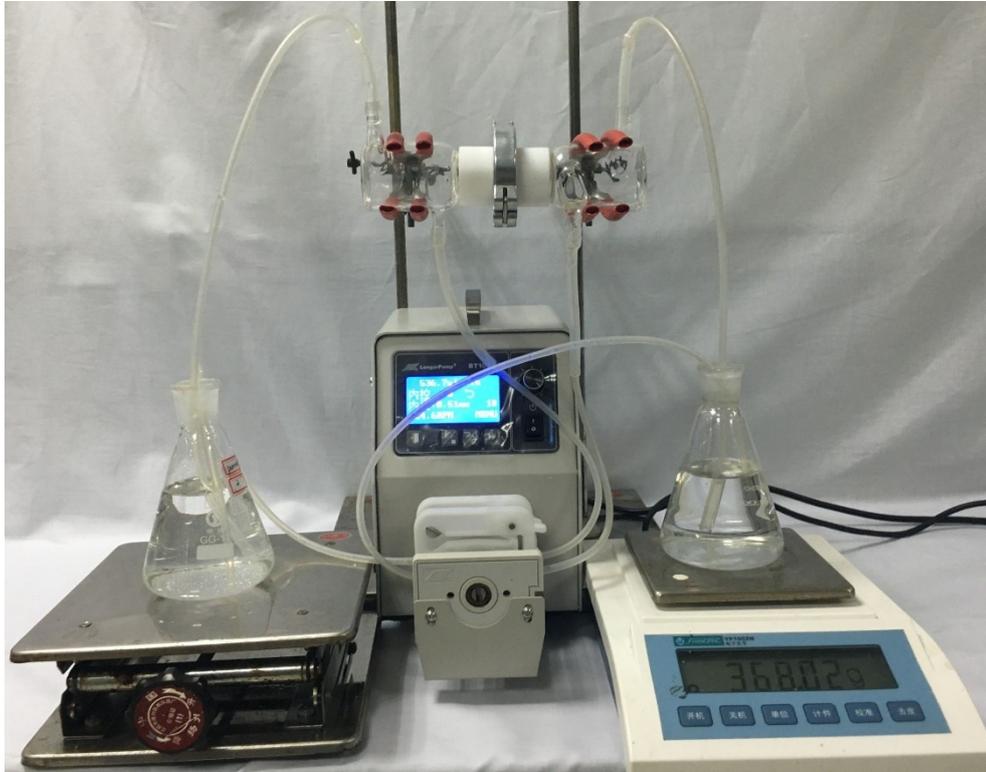
GO was prepared from graphite powder with lateral size of 10  $\mu\text{m}$  with by modified *Hummers'* method, which has been reported in our previous works.<sup>1-3</sup> The productive GO suspension was filtration and put into dialysis bag then dialysis in diluted HCl solution several times for 3 days to remove the metal ions, followed by deionized water dialyzing to remove acid until the dialysis water close to neutral. Successively, we try to use vacuum filtration to remove the unoxidized or incompletely oxidized graphite powder with qualitative filter paper in Berlese funnel. Finally it purified by dialysis for about 1 week to remove the residuals. In order to get the large-size GO sheets, we use strong magnetic stirring instead of sonication for the whole process.

## **Characterization and methods**

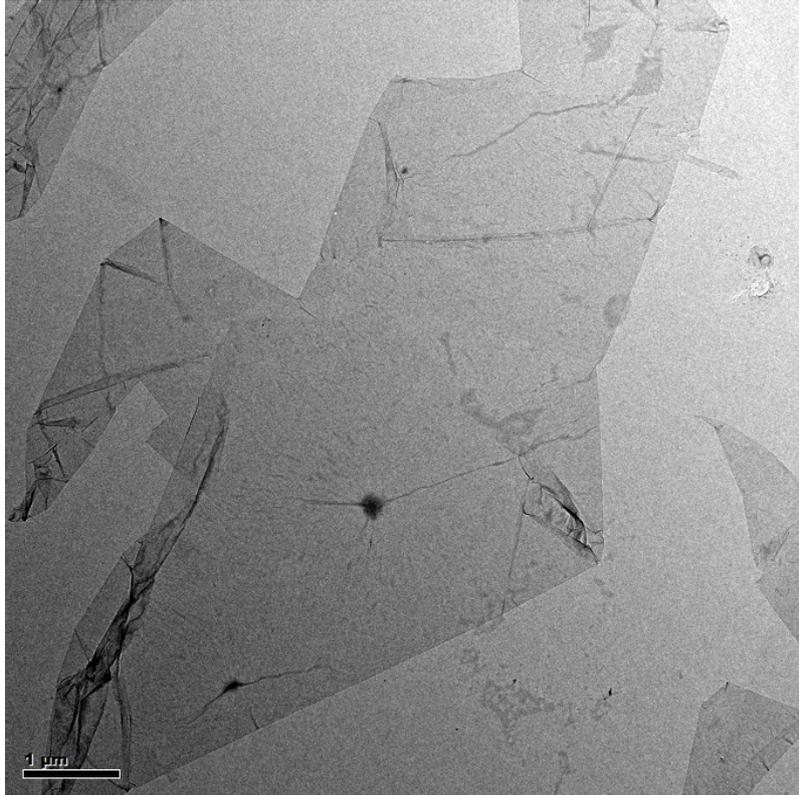
The morphological images of GO membranes surface and cross-section were examined on a field-emission scanning electron microscope (FE-SEM, JEOL, JSM-6700F) at 10 kV. TEM images were obtained using a Tecnai F20 transmission electron microscope with an acceleration voltage of 200 kV. Atomic force microscope (AFM) images were taken on a SPM 9600 microscope (Shimadzu, Japan) to check the thickness and morphology of GO sheets. Fourier transform infrared spectra (FT-IR) were characterized using GO membranes directly on the transmission module of Thermo Nicolet Avatar 370 FT-IR spectrometer. The interlayer spacing of GO membranes was characterized with X-ray diffraction which was performed on a D8Advance X-ray diffractometer with Cu K $\alpha$  radiation ( $\lambda=0.15418$  nm, Bruker, Germany). Raman spectra were obtained by the use of a Lab RAM HR Evolution (HORIBA Jobin Yvon, France). X-ray photoelectron spectroscopy of GO membranes was taken out on a SHIMADZU Kratos AXIS Ultra DLD XPS instrument. THz absorption spectra were recorded on a TAS7400TS THz-TDS system with frequency range from 0.5 to 3.5 THz (Advantest Corporation, Japan). The laser wavelength is 1560 nm, pulse width is 300 fs and frequency resolution is 1.9 GHz. The sample chamber was purged continuously with dry air to keep its moisture below 1.5%. The GO membranes in different wet states were tested subsequently, including dry state (stored in 25 °C desiccator overnight), wet state (picked out from ultrapure water after immersing 1 h and blotted with paper) and half-wet state (leaving in sample chamber of THz-TDS system for 10 min). UV-vis absorption spectra of RB and MO in FO test were recorded on UV-2600

spectrophotometer (Shimadzu, Japan). The concentrations of Na<sup>+</sup> and Cu<sup>2+</sup> were tested with ICP-AES (Optima 8000, Perkin Elmer) instrument.

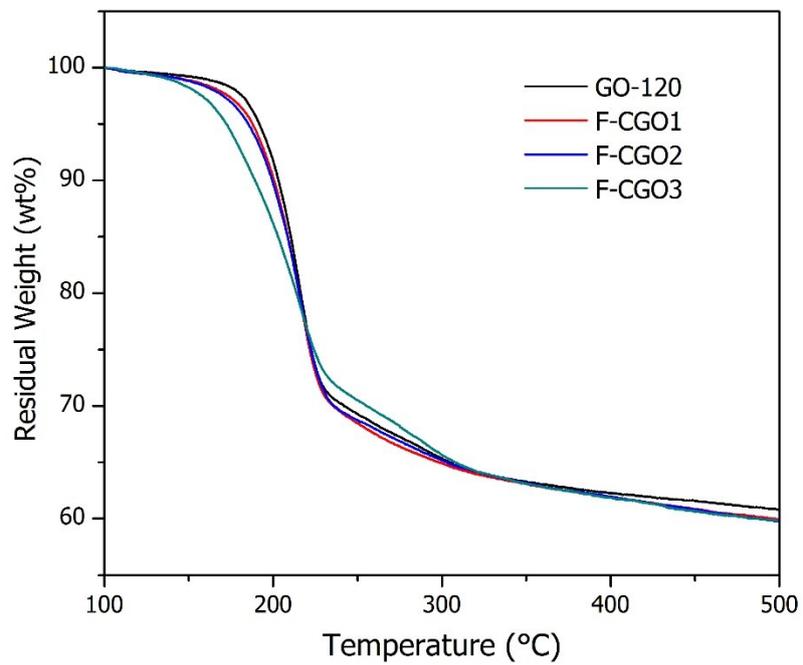
## Supplementary Figures



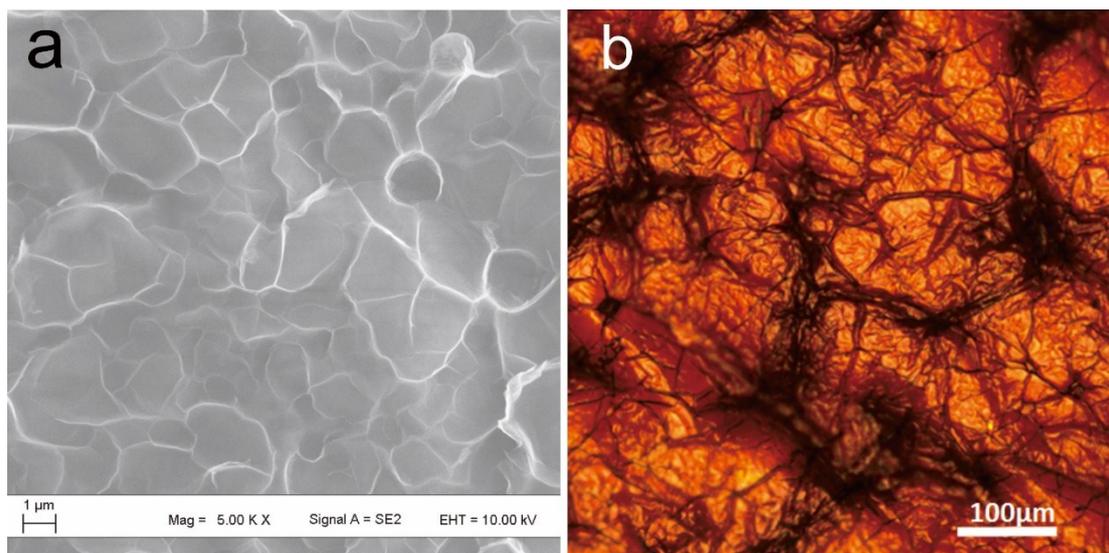
**Figure S1.** The setup of water permeation and reverse salt diffusion testing.



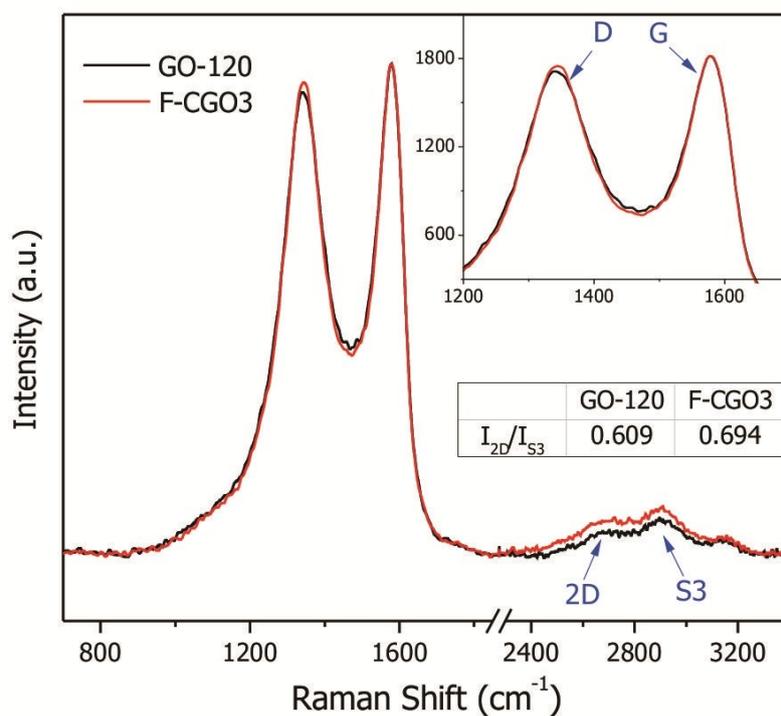
**Figure S2.** The TEM image of GO sheets. The scale bar is 1  $\mu\text{m}$ .



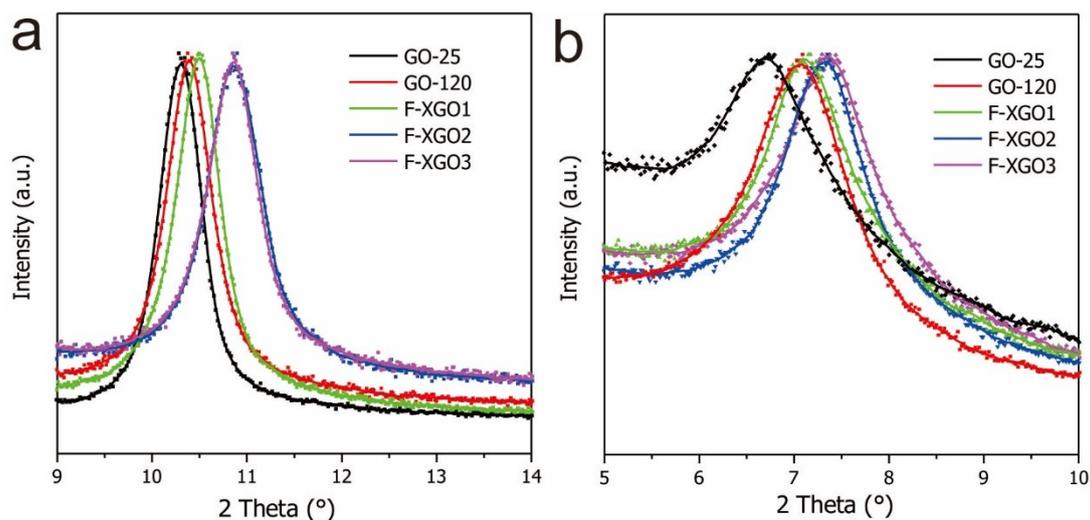
**Figure S3.** TGA curves of different GO membranes.



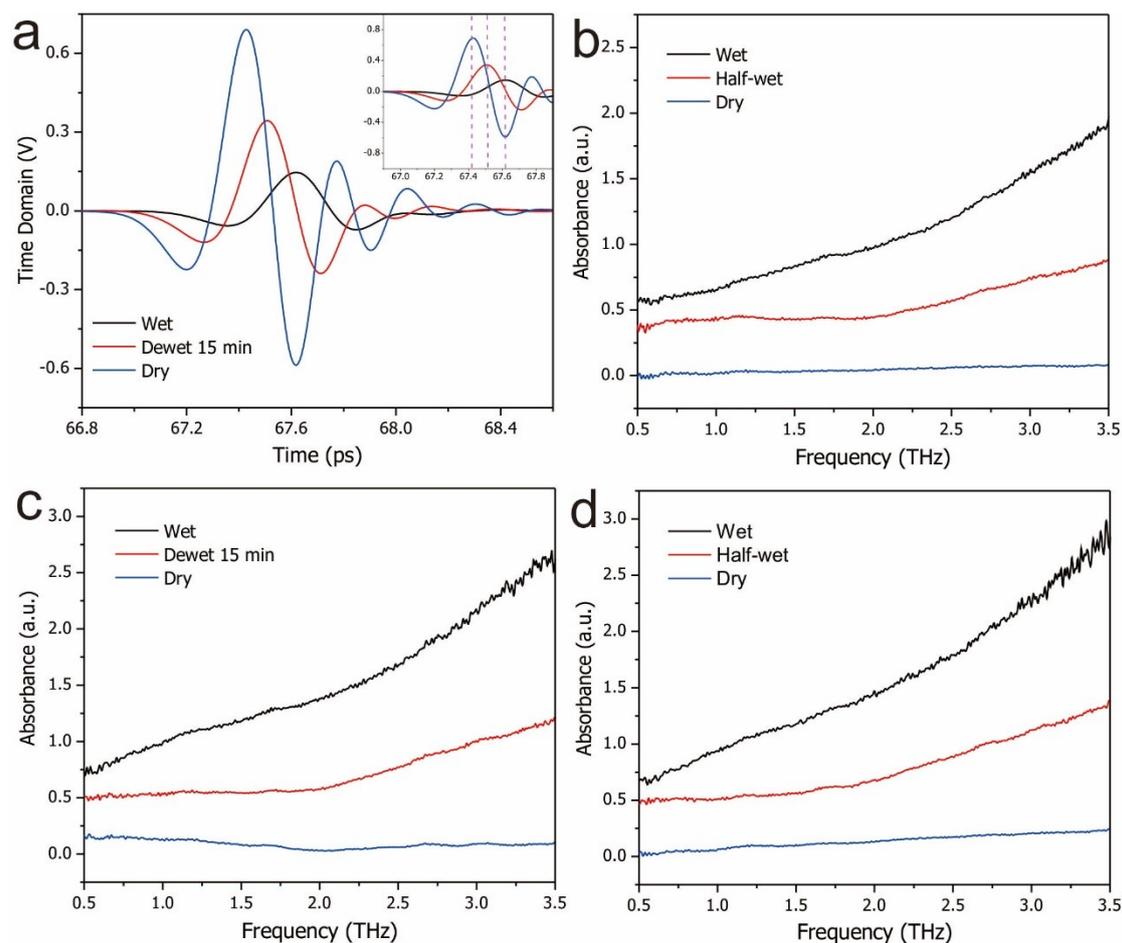
**Figure S4.** The (a) SEM image of reverse surface micro-morphology and (b) optical microscope image of F-CGO3 membrane. The scale bar in (a) is 1 μm.



**Figure S5.** Raman spectra of the GO-120 and F-CGO3 membranes.



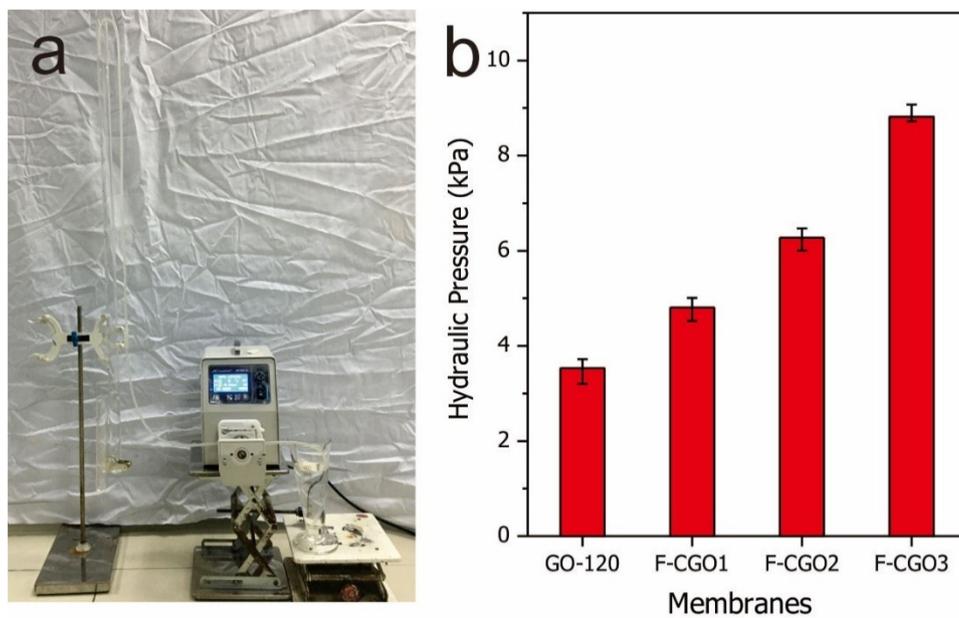
**Figure S6.** The XRD patterns of different GO membranes at (a) dry state and (b) wet state (after immersed in water for 1 h).



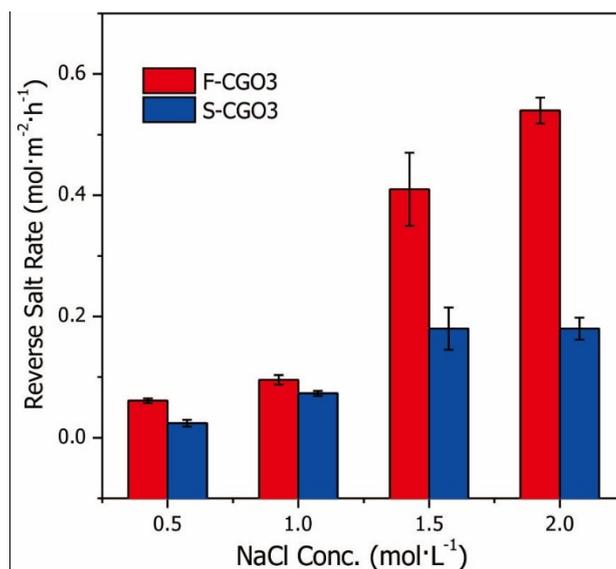
**Figure S7.** (a) THz-TDS spectra of GO-120 membrane at different wet states. THz absorption spectra of the (b) GO-25, (c) GO-120 and (d) F-CGO3 membranes in different wet states.



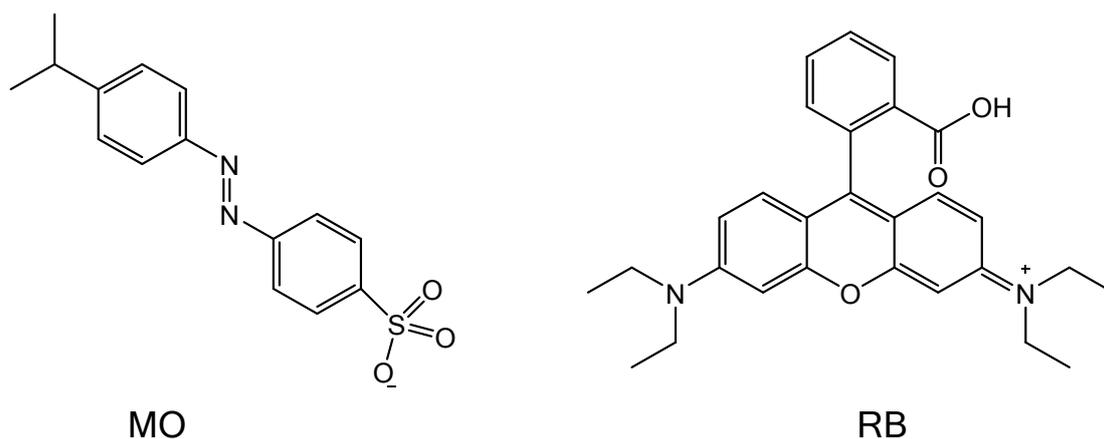
**Figure S8.** Digital pictures of the mechanical testing of the F-CGO3 membrane by overhanging weight.



**Figure S9.** (a) The hydraulic pressure tolerance testing setup, and (b) the maximum hydraulic pressure tolerance of different GO membranes.



**Figure S10.** Comparison of the reverse salt diffusion of the F-CGO3 and S-CGO3 membranes.



**Figure S11.** The chemical structures of MO and RB.

### References:

1. C. Yu, B. Zhang, F. Yan, J. Zhao, J. Li, L. Li and J. Li, *Carbon*, 2016, **105**, 291-296.
2. B. Zhang, L. Li, Z. Wang, S. Xie, Y. Zhang, Y. Shen, M. Yu, B. Deng, Q. Huang, C. Fan and J. Li, *Journal of Materials Chemistry*, 2012, **22**, 7775-7781.
3. B. Zhang, Y. Zhang, C. Peng, M. Yu, L. Li, B. Deng, P. Hu, C. Fan, J. Li and Q. Huang, *Nanoscale*, 2012, **4**, 1742-1748.