

Electronic Supplementary Information for

**Are vacuum-filtrated reduced graphene
oxide membranes symmetric?**

Bo Tang^{a,†}, Lianbin Zhang^{a,†}, Renyuan Li^a, Jinbo Wu^a, Mohamed Nejjib Hedhili^b, and Peng Wang^{*a}

^a. Water Desalination and Reuse Center, Division of Biological and Environmental Science and Engineering, King Abdullah University of Science and Technology, Thuwal, Saudi Arabia.

^b Imaging and Characterization Laboratory, King Abdullah University of Science and Technology, Thuwal, Saudi Arabia.

† These authors contributed equally to this work

*Correspondence: Professor P Wang, Water Desalination and Reuse Center, Division of Biological and Environmental Science and Engineering, King Abdullah University of Science and Technology, Thuwal 23955-6900, Saudi Arabia.

E-mail: peng.wang@kaust.edu.sa

Preparation of silicon membrane

The Si wafer with patterned micropores used for the imprinting experiment was prepared using standard lithography etching by deep reactive ion etching (DRIE).¹ The pre-designed pattern was ‘KAUST’ which was made of properly spaced micropores with a uniform diameter of $\sim 5.0\ \mu\text{m}$. The Si wafer was then used in the vacuum-assisted GO suspension filtration using similar procedure. The mass of the GO in suspension was 10 mg and the obtained GO membrane was then reduced by HI.

Due to very poor mechanical strength of thin rGO membranes (< 250 nm), self-standing membrane can be hardly obtained, and thus it is impossible to accurately investigate its surface wetting and morphology, etc. Instead, we made effort in examining the top surface of some thinner rGO membranes. More specifically, rGO membranes with thicknesses of 20, 35, 50, and 100 nm were prepared on the PVDF filter membrane. The surface wetting behaviors of these membranes are shown in Figure S1a. For the rGO membrane with thickness at ~ 20 nm, the receding angle of its top surface is close to that of the bottom surface of the thicker ones. The surface morphology of the top surface of the 20 nm rGO membrane on the PVDF filter membrane was shown in Figure S1b. Due to the flexibility of graphene sheets, the top surface of those thin rGO membranes inherits and exhibits the macroscopic texture of the underneath filtration membranes (Fig. 4e), resulting in their low receding angle. With the rGO membrane thickness increasing, the morphological feature of the underlying filtration membrane disappears from the top surfaces and the surfaces are smoother with an increased receding angle. With the increase in the membrane thickness, the receding angle increases gradually.

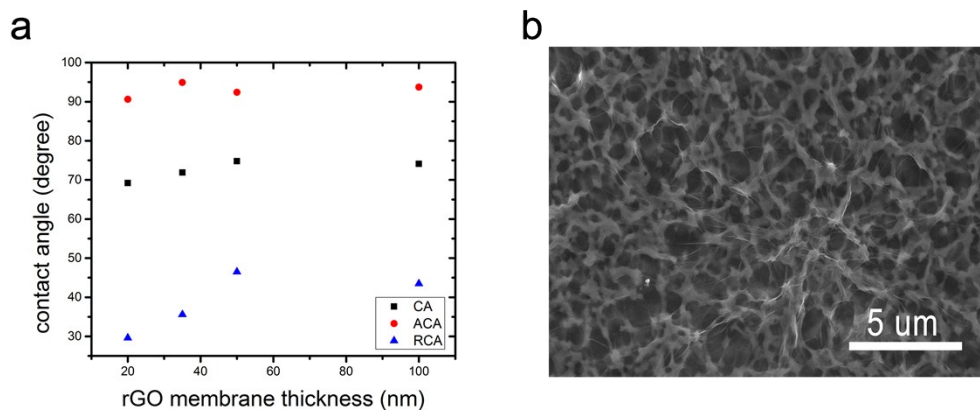


Figure S1. a) The contact angle (CA), advancing contact angle (ACA) and receding contact angle (RCA) of the top surfaces of the rGO membranes with different thickness from 20 to 100 nm. b) SEM image of the top surface of ~ 20 nm rGO membrane prepared on the PVDF filter membrane.

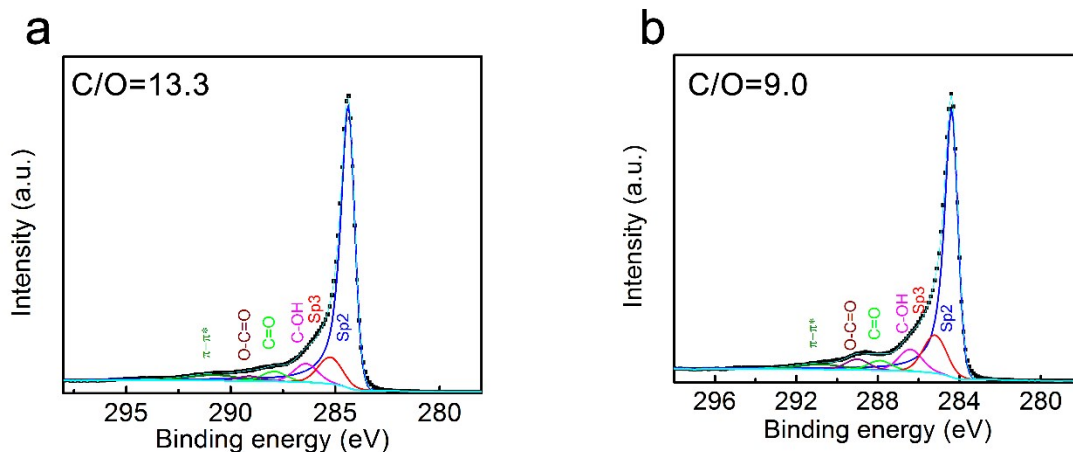


Figure S2. C1s XPS spectra and C/O ratio of rGO membrane with 4 hour HI vapor treatment. (a) The top surface and (b) the bottom surface of the rGO membrane reduced by 4 hour HI vapor treatment. The atomic ratio of C/O between top and bottom surfaces is 13.3 and 9.0, respectively.

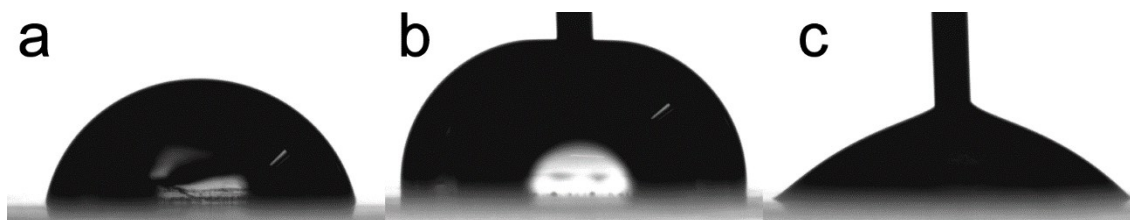


Figure S3. Water wettability on the top surface of the partially reduced GO membrane by 10 minutes HI treatment. (a) Static water contact angle of (79°). (b) Advancing water contact angle (92°). (c) Receding water contact angle (42°).

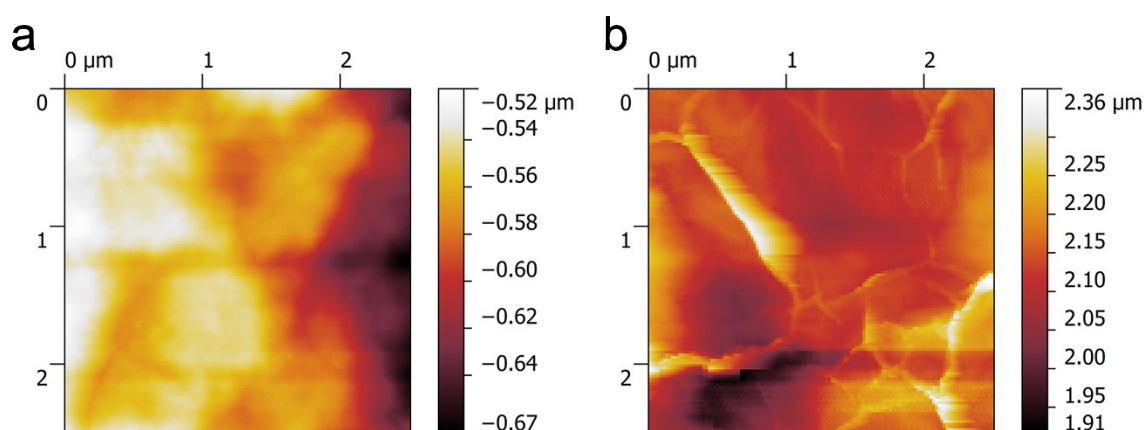


Figure S4. AFM height images of rGO membrane surfaces. (a) top surface and (b) bottom surface of the rGO membrane prepared on the PVDF membrane filter. The calculated R_q values of the top and bottom surfaces are 31.4 nm and 63.4 nm, respectively.

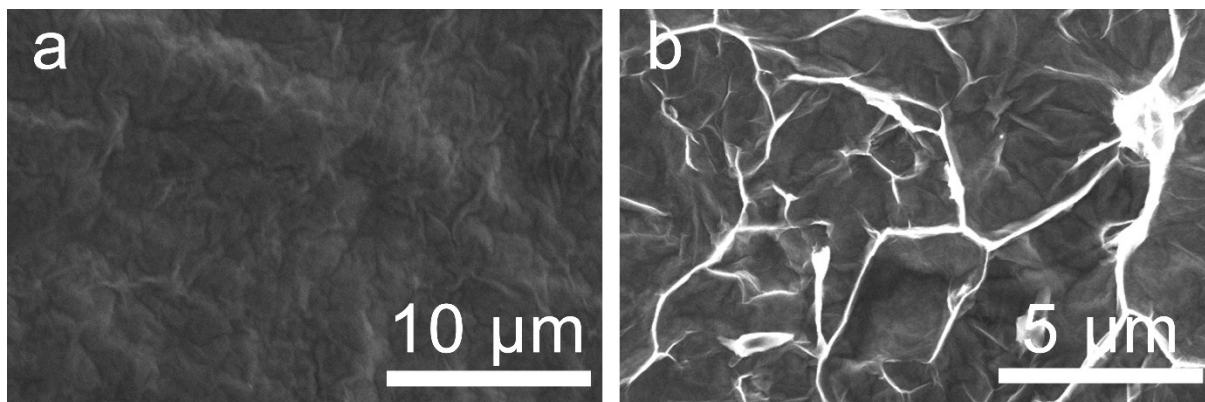


Figure S5. SEM image of GO membrane prepared on PVDF membrane filter. (a) the top surface and (b) bottom surface of the GO membrane prepared on the PVDF filter membrane (with a stated pore size of 0.22 μm). Due to the high content of the oxygen containing groups of GO, the receding contact angles of the two sides are the same (0°). Therefore, from the wetting aspect, the GO membranes are symmetric.

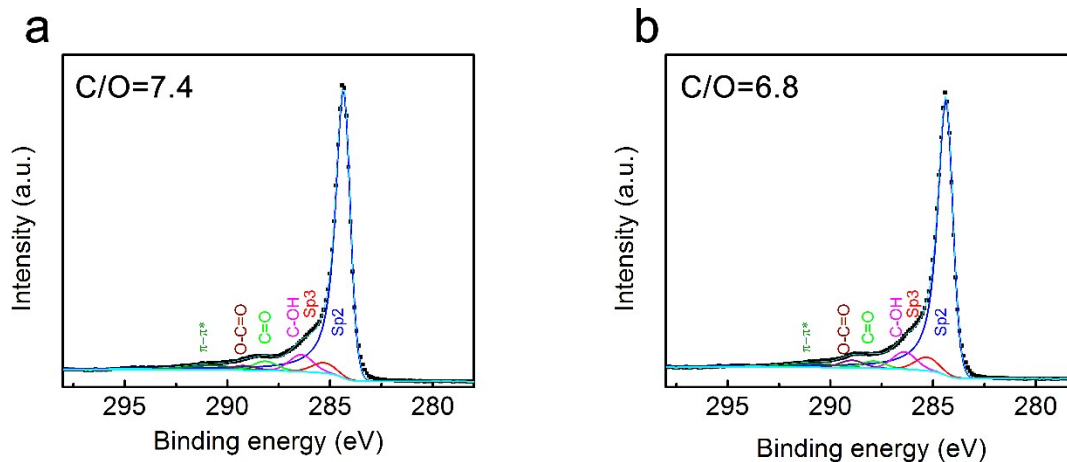


Figure S6. C1s XPS spectra of rGO membrane prepared on silicon wafer. (a) the top surface and (b) the bottom surface of the rGO membrane prepared on the silicon wafer. The atomic ratios of C/O of the top and bottom surfaces are 7.4 and 6.8 respectively.

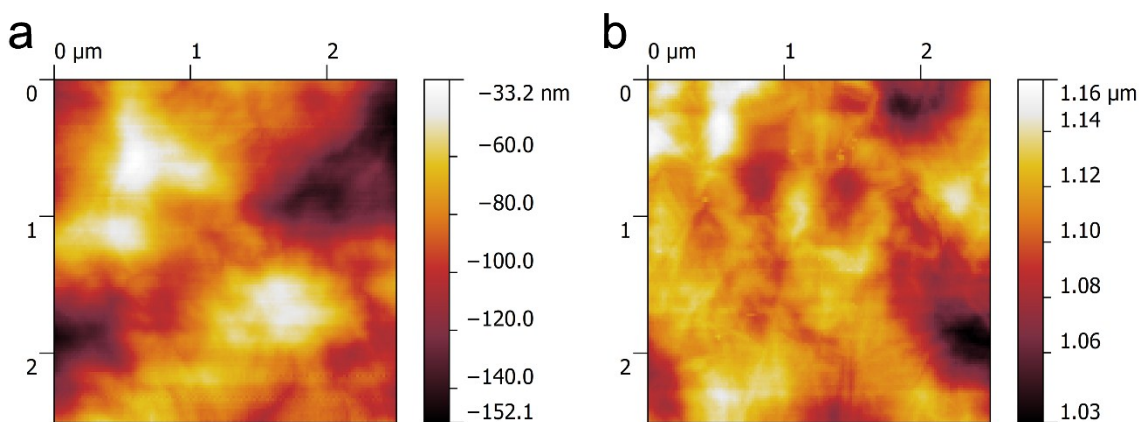


Figure S7. AFM height images of rGO membrane prepared on silicon wafer. (a) the top surface and (b) the bottom surface of the rGO membrane prepared on the silicon wafer. The calculated Rq values of the top and bottom surfaces are 20.1 nm and 23.6 nm, respectively.

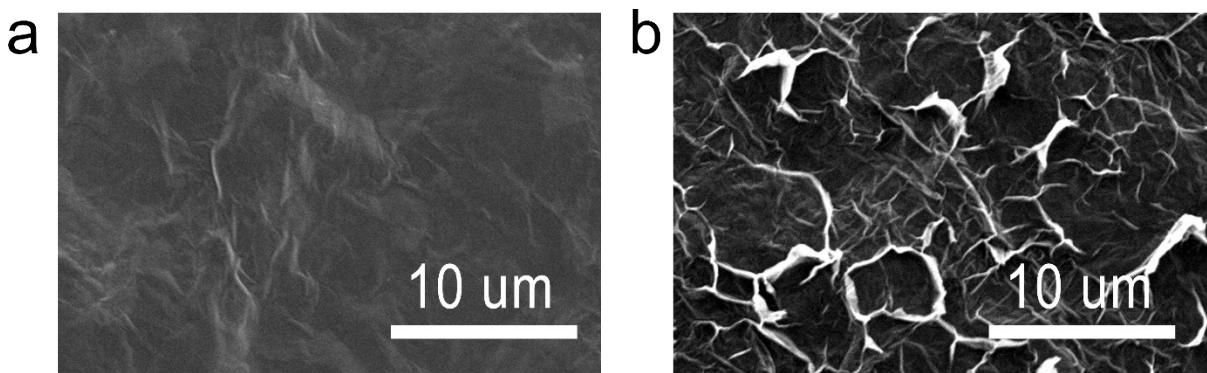


Figure S8. Top viewed SEM images of rGO membrane prepared on nylon filter membrane. (a) the top surface and (b) the bottom surface of the rGO membrane prepared on the nylon filter membrane (with a stated pore size at 0.45 μm).

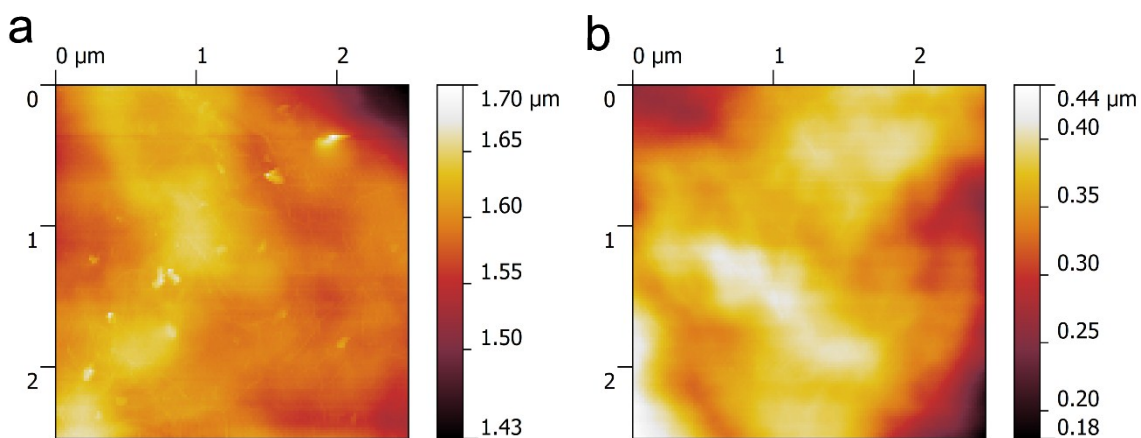


Figure S9. AFM height images of rGO membrane prepared on AAO membrane filter. (a) the top surface and (b) the bottom surface of the rGO membrane prepared on the AAO membrane filter. The calculated R_q values of the top and bottom surfaces are 30.3 nm and 37.8 nm, respectively.

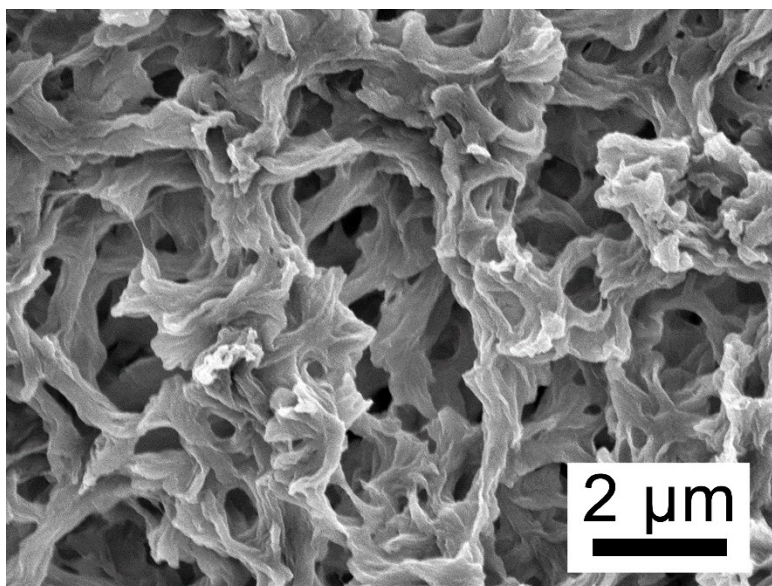


Figure S10. SEM image of the original nylon membrane filter (with a stated pore size $\sim 0.45\ \mu\text{m}$). The actual surface pore size ranges from 0.5 to 4.0 μm .

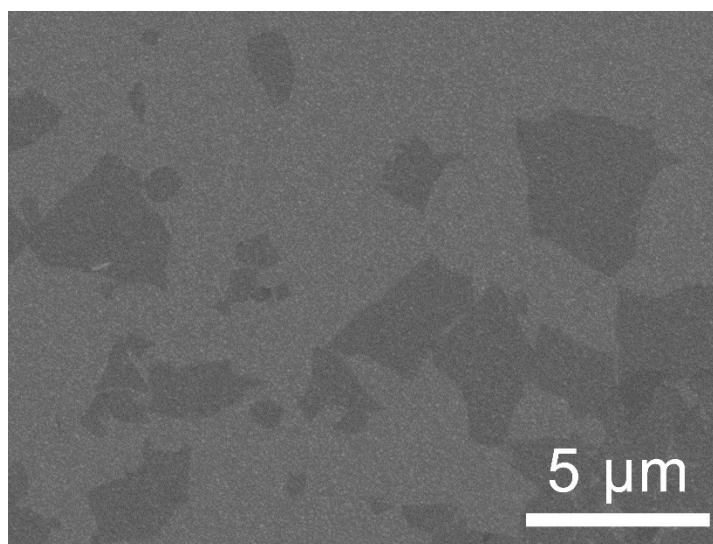


Figure S11. SEM image of the GO nanosheets on the silicon wafer. The size of individual nanosheet ranges from 0.5 μm up to 5.0 μm .

Supplementary references:

1. Wu, J. Zhang, M., Chen, L., Yu, V., Wong, J. T.-Y., Zhang, X., Qin, J. and Wen, W. *RSC Adv.* **1**, 746-750 (2011)

Legends for Supplementary videos:

Video S1. Water wetting behaviors on the top and bottom surfaces of the rGO membrane prepared on a PVDF filter membrane.

Video S2. Capture and the movement of the water droplets on the bottom surface of the rGO membrane prepared on a PVDF filter membrane.

Video S3. Capture and the movement of the water droplets on the top surface of the rGO membrane prepared on a PVDF filter membrane.