Supporting Information

Atomistic insights into the separation mechanism of multilayer graphene membranes for water desalination

Jing Zhang [a], chen Chen [a], Jianuan Pan [a], Li Zhang*[a], Lijun Liang[b], Zhe Kong[c], Jia-Wei Shen[d]*, Xinping Wang[a], Wei Zhang[a]

[a] Department of Chemistry, Zhejiang Sci-Tech University, Hangzhou, 310018, People's Republic of China. Fax: +86-571-8684-3600; Tel: +86-571-8684-3691; E-mail addresses: lizhang@zstu.edu.cn.

[b] College of Life Information Science and Instrument Engineering, Hangzhou Dianzi University, Hangzhou, 310018, People's Republic of China.

[c] College of Material & Environmental Engineering Science Hangzhou Dianzi University, Hangzhou, 310018, People's Republic of China.

[d] School of Medicine, Hangzhou Normal University, Hangzhou 310016, People's Republic of China. E-mail addresses: shen.jiawei@hotmail.com.

Fig.S1 shows the number of transferred water molecules through MGM as a function of simulation time at 200MPa, the number of water molecules increases at the beginning of simulation, then the permeation rate of water molecules would decrease gradually due to the increase of osmotic pressure in the right box. When the osmotic pressure is equal to the applied pressure, the transmission process would be always underway, while the number of transferred water molecules would keep constant.

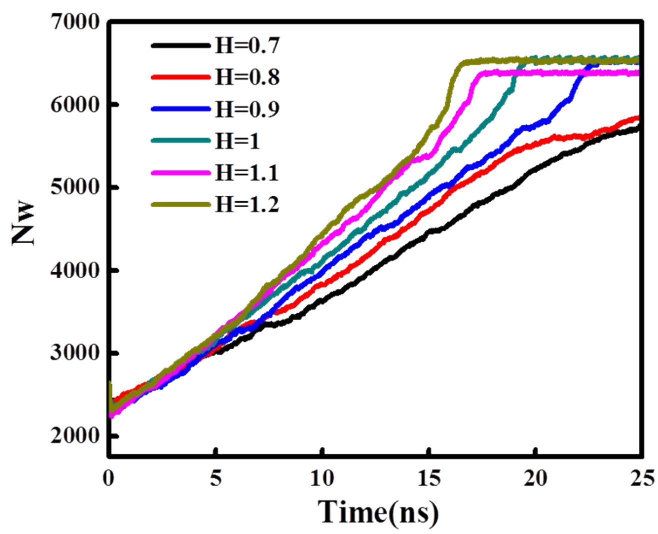

Figure S1. The Number of transferred water molecules through MGM at different layer spacing from NaCl solution to pure water boxes as a function of simulation time at 200MPa.

With the aim of investigating the effect of offset on water flux and salt rejection rate, in this manuscript, the offset (*O*) was set as 0.4 nm, 0.8 nm, 1.26 nm, 1.6nm, respectively. The distance of interlayer spacing (*H*) was set as 0.8nm and 1.0nm, and the width of gap (*dG*) was set as 1.0nm. The relationship between water flux/salt rejection and the offset was calculated and shown in Fig S2.Water flux decrease with offset increasing, and it decrease slightly when the offset is larger than 1.26nm. As to salt rejection rate, ions could pass through the fully aligned membranes even at a low separation distance, it increases with the increase of offset, and the salt rejection rate is higher to 100% when the distance of interlayer spacing is small (*H*=8Å). The values of salt rejection keep constant when the offset is larger than 1.26nm, this result is consistent with Dahanayaka *et.al*[1] work, they reported that ions could pass through the fully aligned membranes even at a low separation distance, while the stacked graphene membrane with large offset shows a complete salt rejection when the distance of interlayer distance is small. Therefore, the offset between different graphene sheets was fixed at 1.26nm in this manuscript.

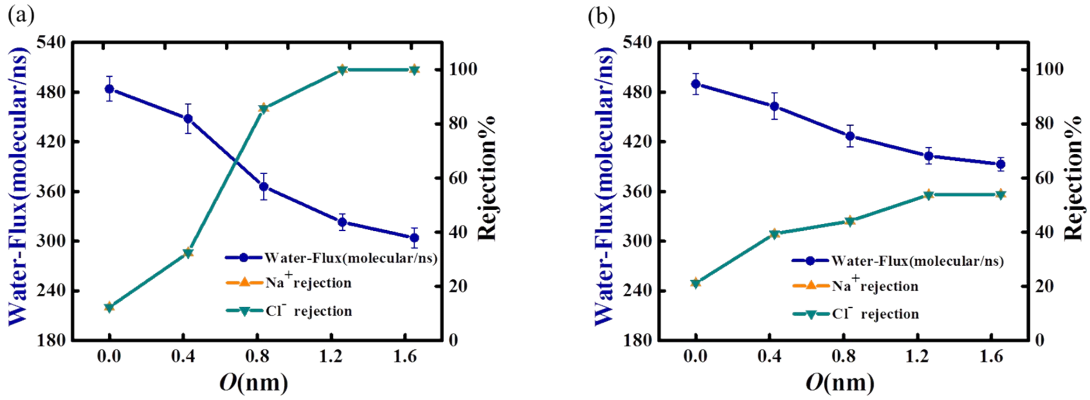

Figure S2. Water flux and salt rejection in MGM membranes with different offset (*O*) under 400MPa when the distance of of interlayer spacing was equal to 0.8nm(a) and 1.0nm(b); water flux (navy), Na^+ rejection (orange), Cl^- rejection (dark cyan).

Figure S3 presents the snapshot of Na^+(in blue) and water molecules in the interlayer spacing of MGM membrane.

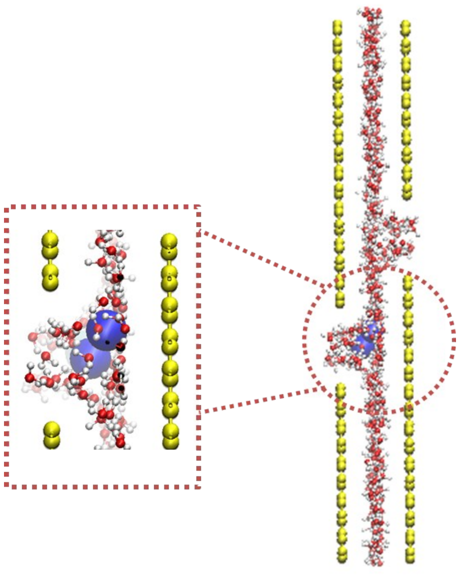

Figure S3. Snapshot of Na^+(in blue) and water molecules in the interlayer spacing of

MGM membrane.

The number density profiles for ions along z direction in MGMs with different graphene sheets were calculated shown in Fig.S4.

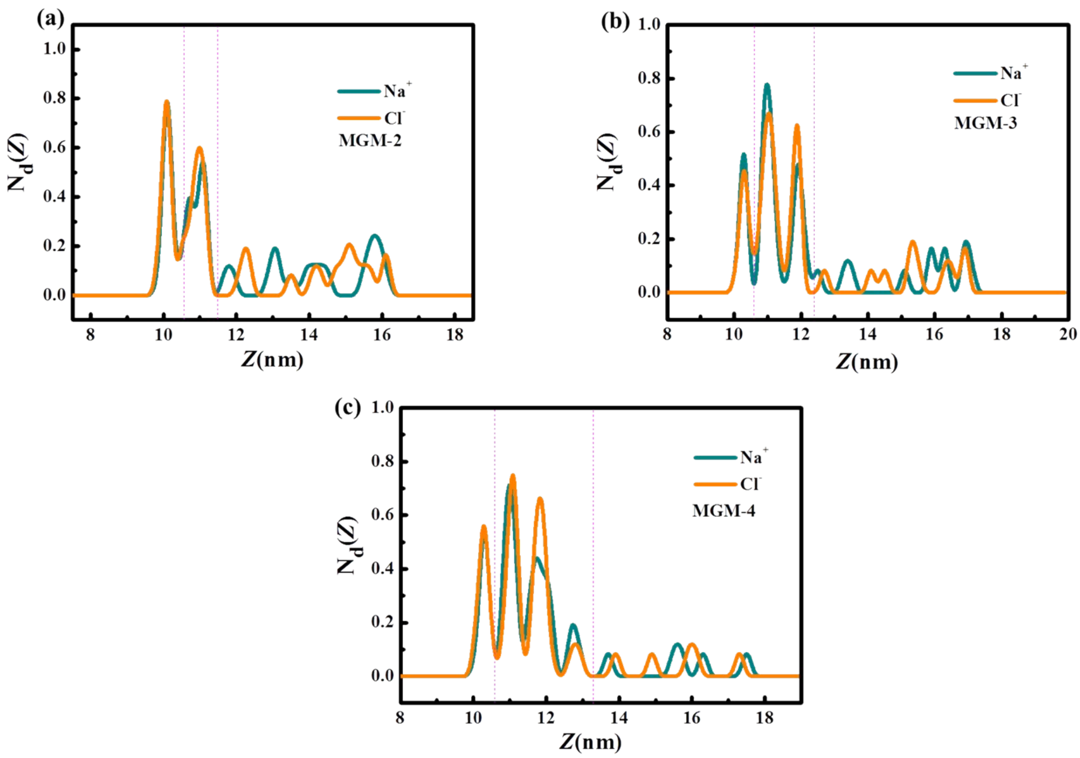

Figure S4. Number density profiles for ions along the *z*-axis in the interlayer of MGM membrane at 10ns. Two ends of the MGM were represented by dashed lines. (a) MGM-2, *z*=10.6nm and 11.5nm; (b) MGM-3, *z*=10.6nm and 12.4nm; (c) MGM-4, *z*=10.6nm and 13.3nm.

In order to investigate the effect of gap number on desalination, the sketch map for MGM-2 systems with different gaps were shown in Fig.S5.

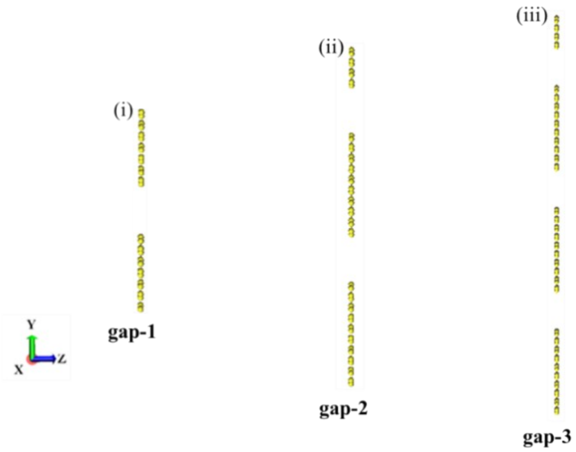

Figure S5. Diagrammatic sketch of the MGM system with one, two and three graphene gaps were named as gap-1(i), gap-2(ii), and gap-3(iii).

Water flux of different solutions in MGM-2 under 200MPa and 200MPa were calculated and illustrated in FigureS6, and the number density profiles for ions along the *z*-axis at 10ns under 400MP were shown in Figure S6

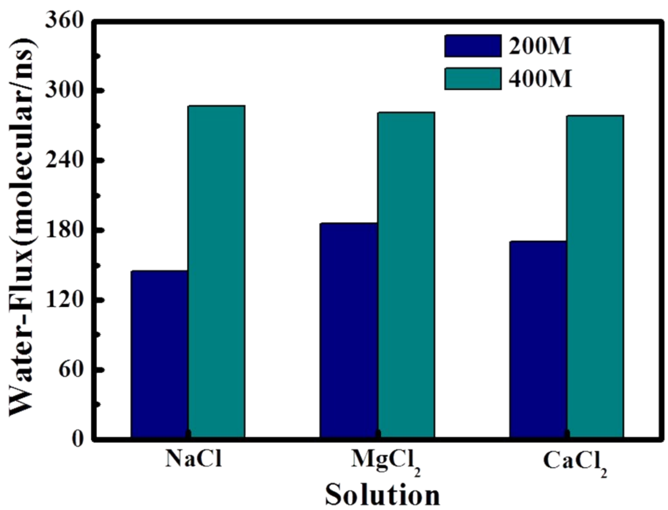

Figure S6. Water flux of different solutions was calculated (NaCl solution, $MgCl_2$

solution and $CaCl_2$ solution) through the MGM under 200MPa (navy column) and 400MPa (dark cyan column).

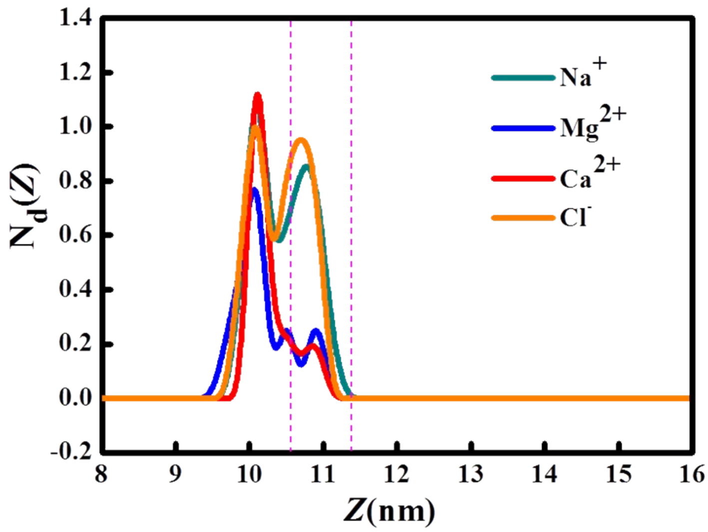

Figure S7. Number density profiles for ions along the *z*-axis at 10ns under 400MPa. Two ends of the MGM were represented by dashed lines (z=10.6nm and 11.4nm). Na^+(in dark cyan), Mg^{2+} (in blue), Ca^{2+} (in red), Cl^- (in orange).

References

1. M. Dahanayaka, B. Liu, Z. Hu, Q.-X. Pei, Z. Chen, A. W.-K. Law and K. Zhou, *Phys. Chem. Chem. Phys.*, 2017, **19**, 30551-30561.