## **Supporting Information:**

## Water Transport and Desalination through Double-Layer Graphyne Membranes

Mojdeh Akhavan<sup>a</sup>, Jeremy Schofield<sup>b</sup> and Seifollah Jalili<sup>\*a,b,c</sup>

<sup>a</sup>School of Nano-Science, Institute for Research in Fundamental Sciences (IPM), P.O. Box 19395-5531, Tehran, Iran
<sup>b</sup>Chemical Physics Theory Group, Department of Chemistry, University of Toronto, 80 Saint George Street, Toronto, Ontario, Canada, M5S 3H6
<sup>c</sup>Department of Chemistry, K. N. Toosi University of Technology, P. O. Box 15875-4416, Tehran, Iran

<sup>\*</sup> Corresponding author. Tel: +98 21 22853649, E-mail: sjalili@kntu.ac.ir (Seifollah Jalili)



**Figure S1** Molecular structure of graphyne-3 (a) and graphyne-4 (b) sheets together with an AB-stacked graphyne-4 bilayer (c).



**Figure S2** (a) Variation of the number of water molecules permeated with simulation time. (b) Linear dependence of the water flux across a bilayer on the applied pressure.



**Figure S3.** Density profiles of water molecules along the z-axis for single-layer G3 and G4 and for double layer G3 membranes with different layer spacings. The dashed lines show the positions of graphyne membrane sheets.



**Figure S4.** Hydrogen bonding profiles as the average number of hydrogen bonds per water molecule at small intervals along z-axis for single layer graphyne-3 and graphyne-4 membranes with AA stacking configuration. The membrane is located at z = 15 nm. A donor-acceptor distance of 0.35 nm and donor-hydrogen-acceptor angle of 30° have been taken as the geometrical criteria for hydrogen bonding. The feed values being slightly lower due to the gradual depletion of the feed solution during the time interval of averaging (i.e. from 0 to  $t_{\rm f}$ ).



**Figure S5.** Variation of the salt rejection for graphyne-4 membranes (a) with layer spacing at the applied pressure of 200 MPa; (b) with the applied pressure (single layer).