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Supplementary Information

Few-Layered Ultrathin Covalent Organic Framework Membranes for Gas Separation: A Computational Study

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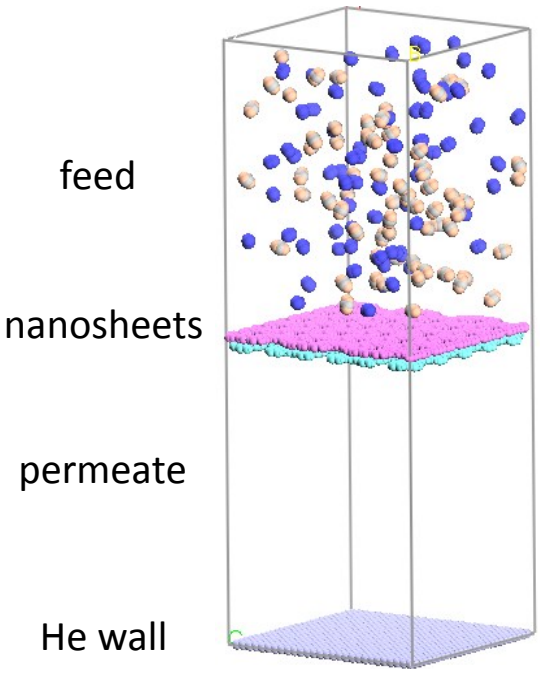

Fig. S1 Illustration of the membrane models used in molecular dynamics simulations. (CO_2: C, silver; O, yolk yellow; N_2, blue)

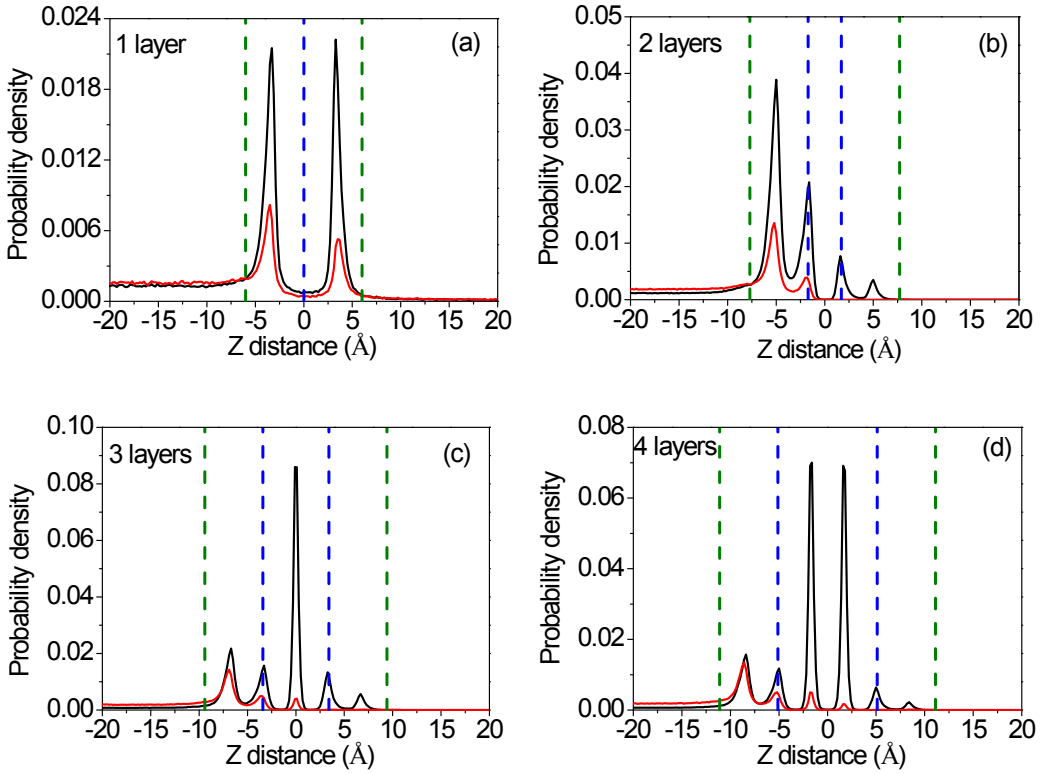

Fig. S2 Probability density distributions of CO_2 and N_2 molecules as a function of the perpendicular distance from the membranes with different number of layers, where the origin of coordinate in each subfigure is located in the membrane center. Black and red curves are the results for CO_2 and N_2, respectively. The region within the two blue (green) dotted lines is the membrane (adsorption-layer) zone.

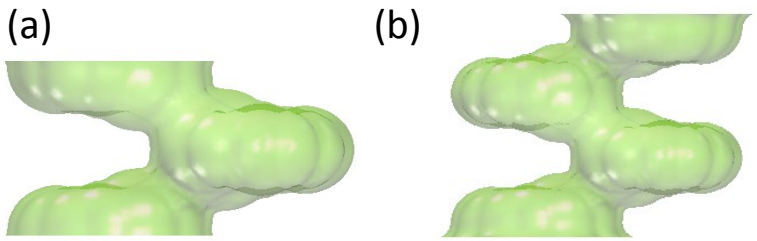

Fig. S3 Channel structures of "periodically" designed (a) three-layered and (b) four-layered membranes

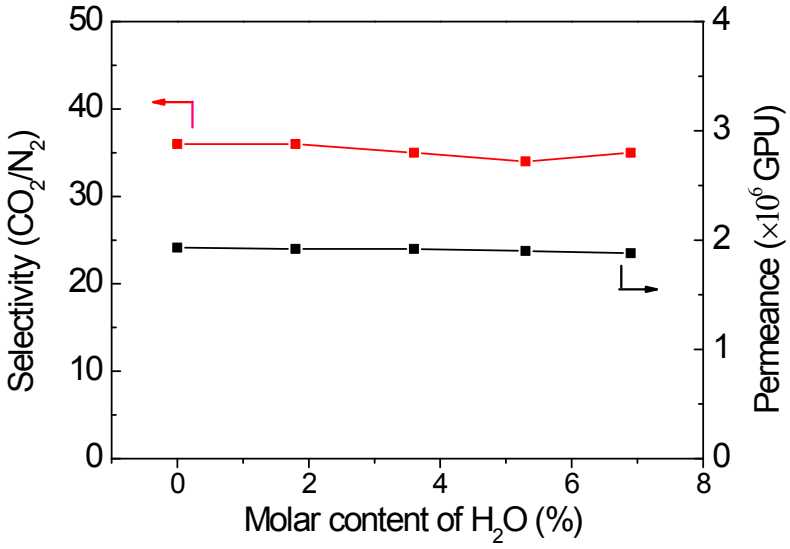

Fig. S4 CO_2/N_2 selectivity and the CO_2 permeance of the four-layered membrane of No.1, as a functional of water content in the feed $CO_2/N_2/H_2O$ gas mixture, CO_2 and N_2 are kept at the equimolar conditions so as to be consistent with the situation considered for the dry mixture.

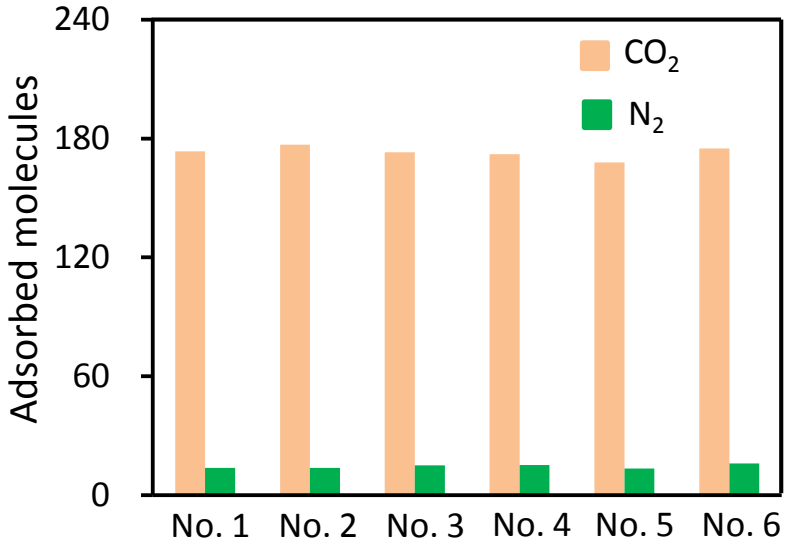

Fig. S5 Statistically-averaged loadings of CO_2 and N_2 adsorbed in the six designed four-layered membranes.

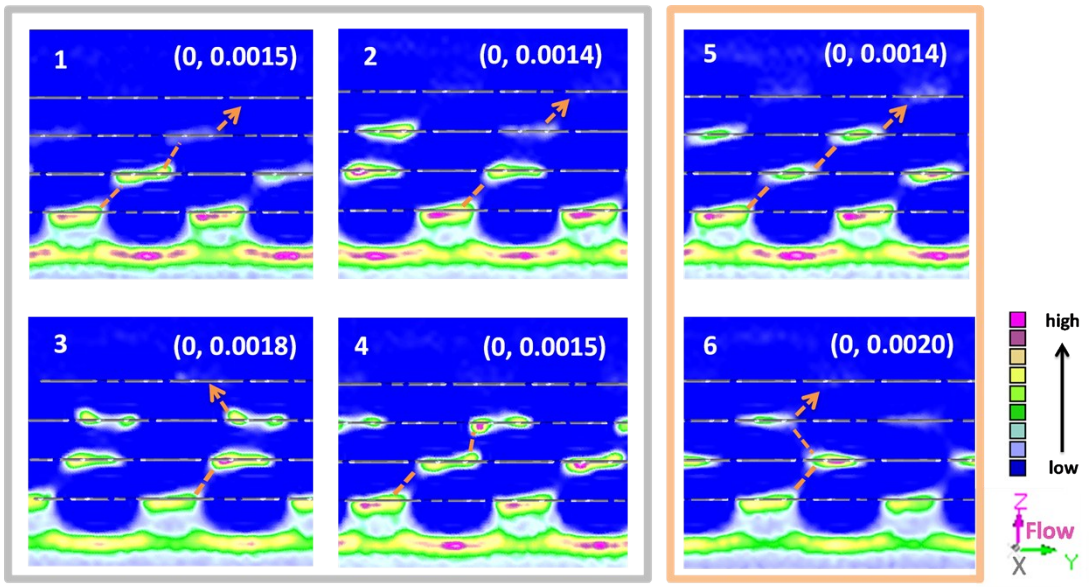

Fig. S6 Contour plots of the COM probability distributions of N_2 in the six designed membranes. One path for the No. 2 to 4 membranes is not shown because of the need to observe from the *y* axis. The two values in the bracket given in each subfigure represent the lowest and the highest number densities in the distributions.

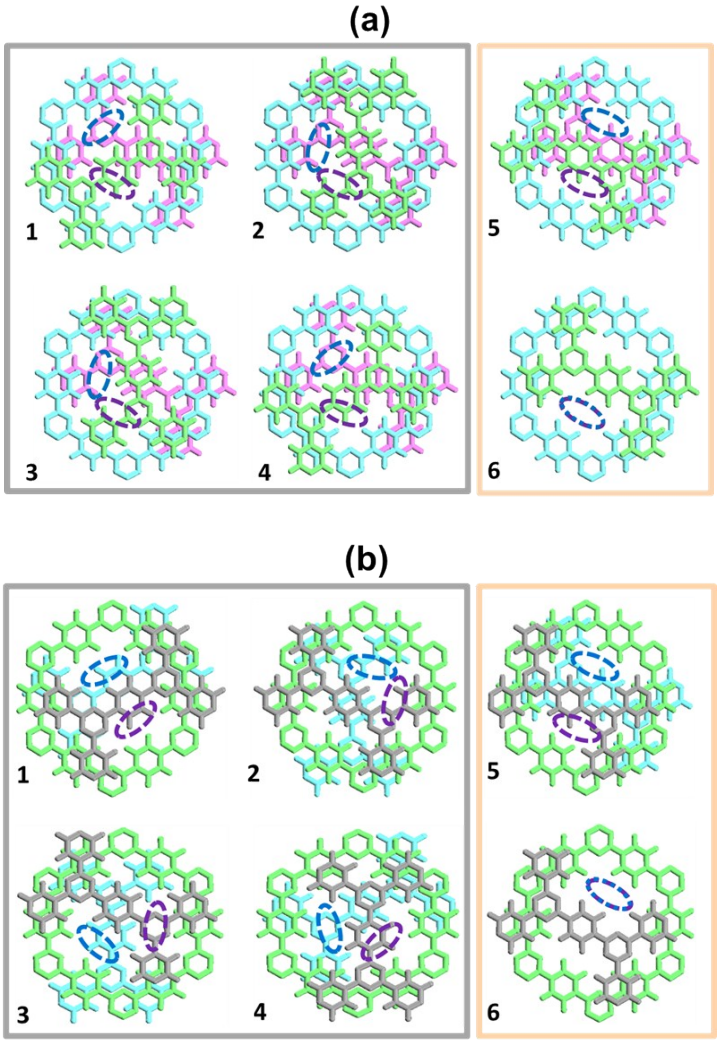

Fig. S7 Top view of a single pore in the (a) second layer and (b) third layer of the six membranes with the up-down stacking environments by the adjacent layers. The areas enclosed by the dashed blue and purple lines are the regions located by the diffusion inlets and outlets, respectively. (First layer, pink; second layer, aquamarine; third layer, green; fourth layer, gray).