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Supplementary Information

Mechanistic Studies on the Anomalous Transport Behaviors of Water Molecules in Nanochannels of Multilayer Graphynes

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Table S1. Simulation model details for studied simulation systems

Substrate	Dimensions in x and y (Å)	Number of Water Molecules
Graphyne-3	42.32×48.86	942
Graphyne-4	51.38×44.50	1044
Graphyne-5	60.45×52.35	1940

Table S2. The Lennard-Jones parameters and atomic charge information

$$E_{ij} = 4\varepsilon_{ij}\left[\left(\frac{\sigma_{ij}}{r_{ij}}\right)^{12} - \left(\frac{\sigma_{ij}}{r_{ij}}\right)^{6}\right] + \frac{q_i q_j}{r_{ij}}$$

atom	ε (kcal·mol^{-1})	σ (Å)	q(e)
C	0.08633	3.400	0.000
O	0.1554	3.166	-0.8476
H	0.0000	0.000	0.4238

Table S3. HB lifetime τ_S^{HB} of water molecules inside the nanochannels of different graphyne membrane systems.

System	τ_S^{HB} (300 K) (ps)	τ_S^{HB} (350 K)
Graphyne-3	25.91	12.38
Graphyne-4	4.97	3.10
Graphyne-5	3.96	2.11

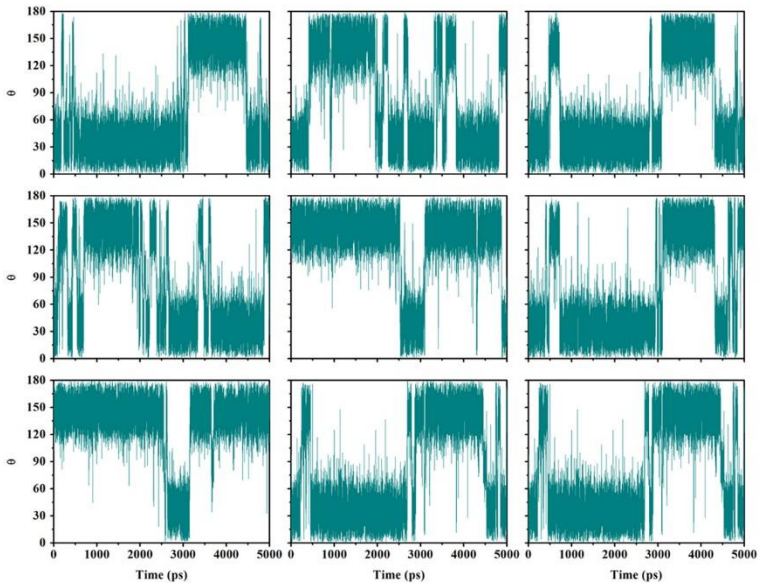

Fig. S1. The orientation variation of angle θ, formed between water dipole and unit vector in z-direction, with respect to the simulation time for typical water molecules between first and second nanosheets in graphyne-4 system (see the nanosheet label in Fig. S6).

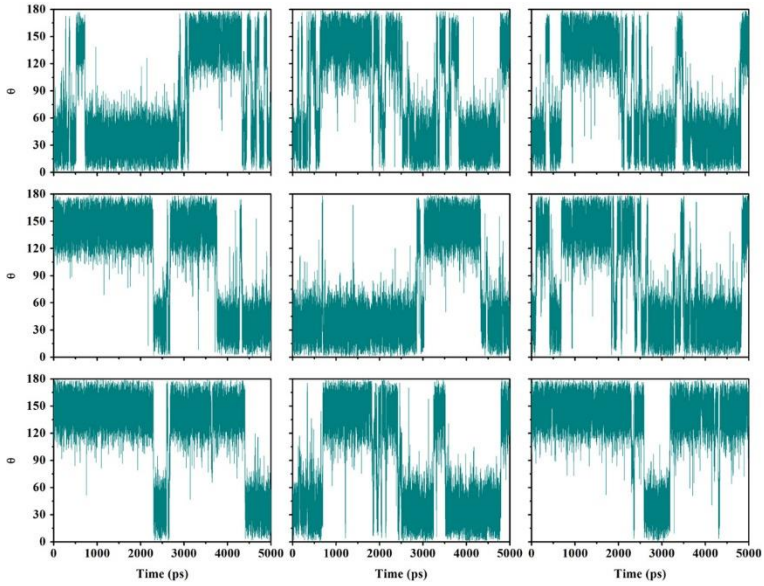

Fig. S2. The orientation variation of angle θ, formed between water dipole and unit vector in z-direction, with respect to the simulation time for typical water molecules between second and third nanosheets in graphyne-4 system (see the nanosheet label in Fig. S6).

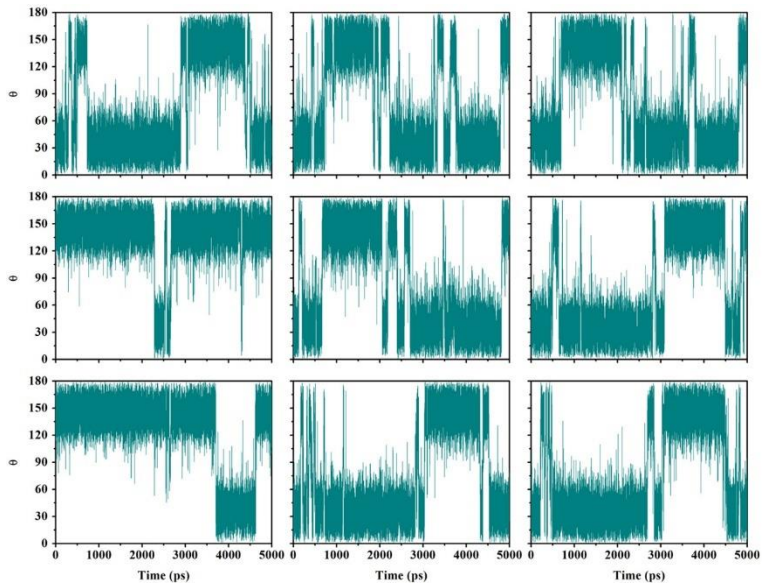

Fig. S3. The orientation variation of angle θ, formed between water dipole and unit vector in z-direction, with respect to the simulation time for typical water molecules between third and fourth nanosheets in graphyne-4 system (see the nanosheet label in Fig. S6).

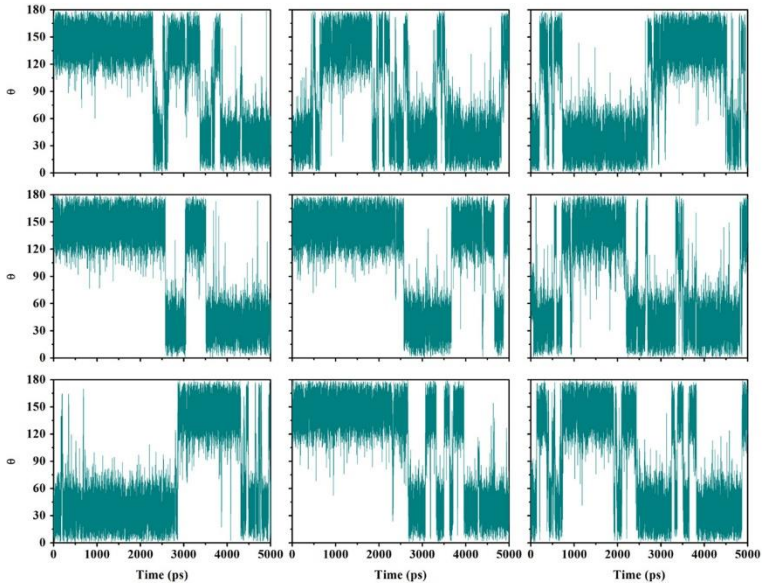

Fig. S4. The orientation variation of angle θ, formed between water dipole and unit vector in z-direction, with respect to the simulation time for typical water molecules between fourth and fifth nanosheets in graphyne-4 system (see the nanosheet label in Fig. S6).

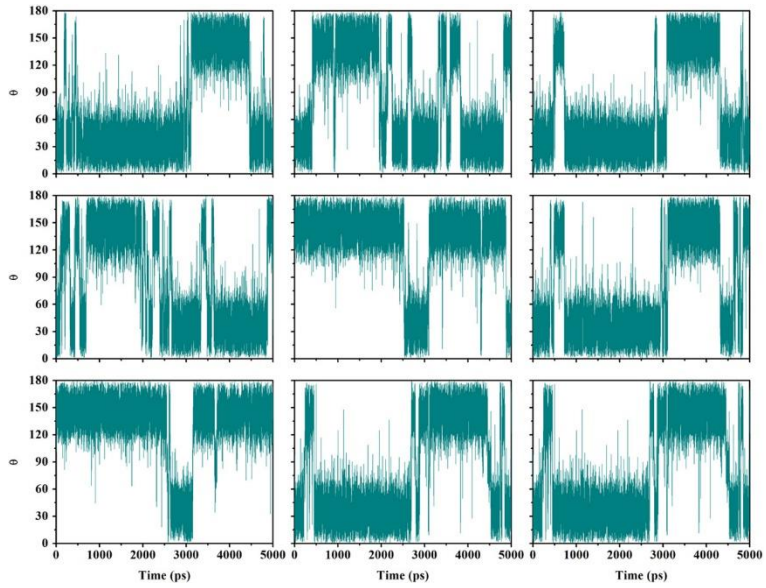

Fig. S5. The orientation variation of angle θ, formed between water dipole and unit vector in z-direction, with respect to the simulation time for typical water molecules between fifth and sixth nanosheets in graphyne-4 system (see the nanosheet label in Fig. S6).

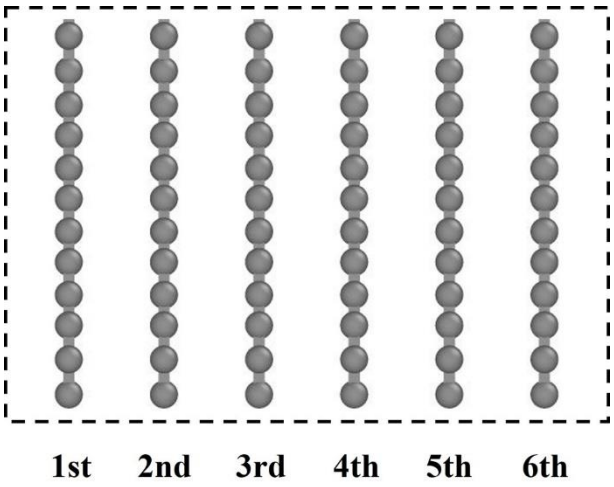

Fig. S6. Schematic illustration of nanosheet label in graphyne-4 system.

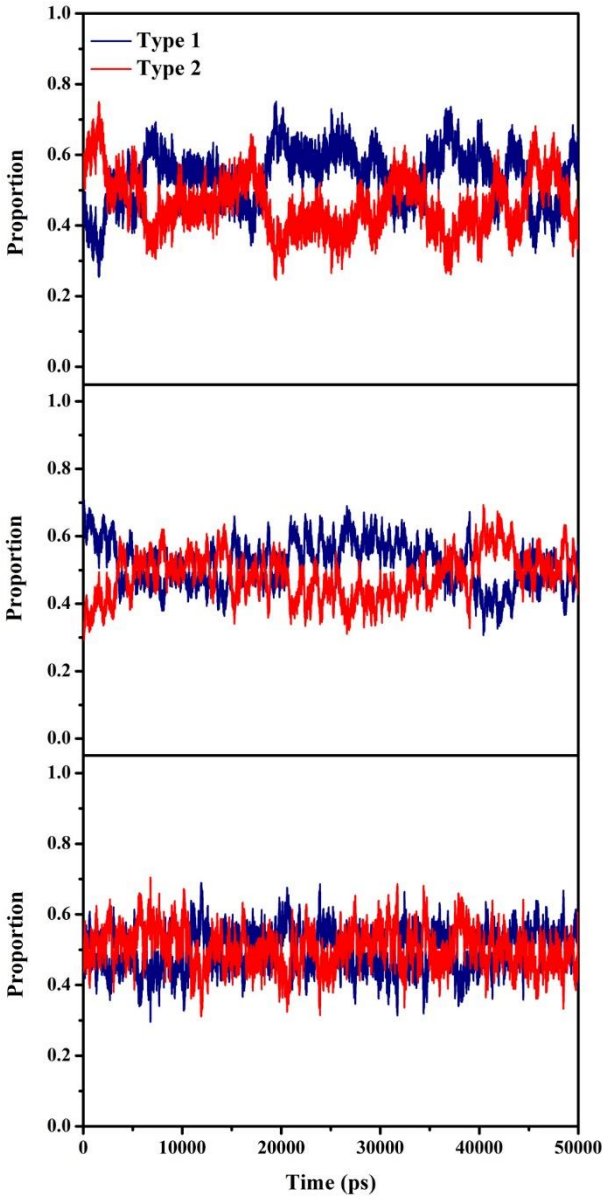

Fig. S7. The variations of proportion for both Type1 and Type2 orientations with respect to the simulation time in the systems of (a) graphyne-3, (b) graphyne-4, and (c) graphyne-5 membranes.

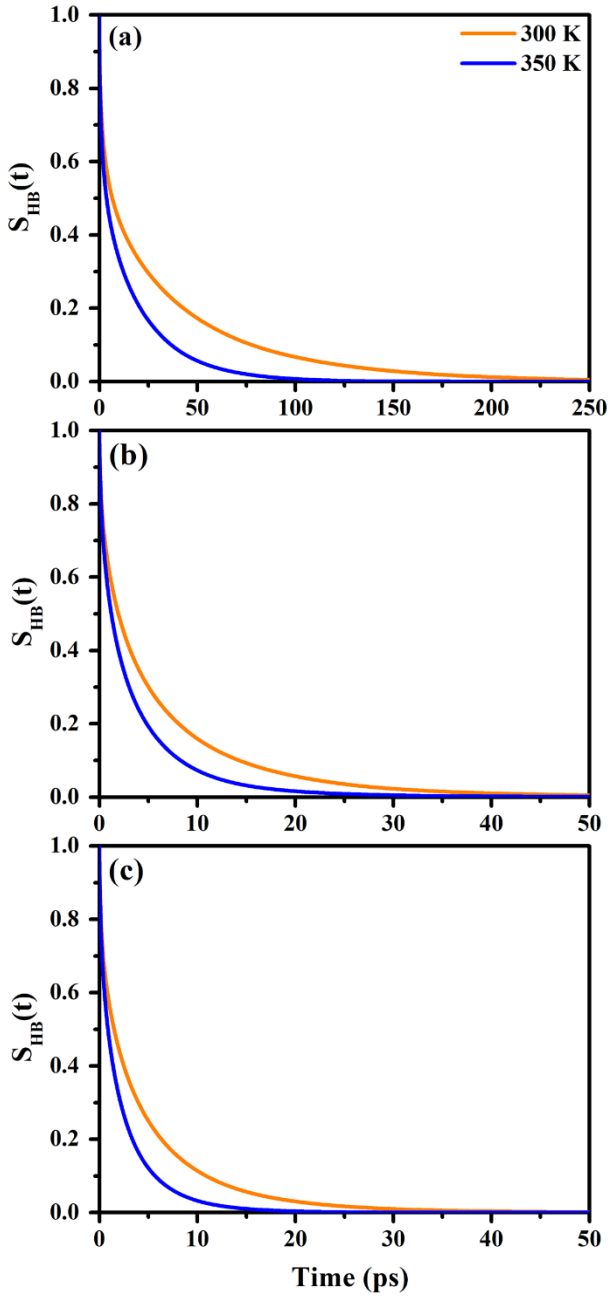

Fig. S8. Continuous TCF $S_{HB}(t)$ of HB for water molecules within the triangular nanochannels of (a) graphyne-3, (b) graphyne-4, and (c) graphyne-5 membranes at the temperature of 300 and 350 K.

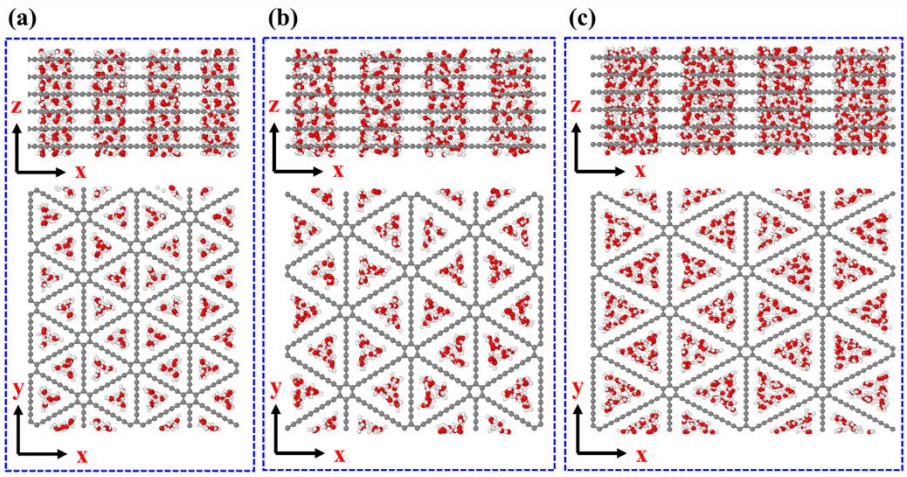

Fig. S9. Top and side views of equilibrium snapshots for (a) graphyne-3, (b) graphyne-4, and (c) graphyne-5 systems.

Fig. S9 shows the equilibrium snapshots of water molecules inside the triangular nanochannels of three graphyne systems and it is clear that water molecules are preferential to distribute in the space between two graphyne nanosheets. To better understand such distributions, we analyzed the density distributions of water molecules along the axial direction (z-direction). The simulation box was divided into 408 (based on the box length in the z-direction) rectangular shells along the axial direction and then the water density was calculated by using the following equation:[S1]

$$\rho(z) = \frac{\langle \sum_i^N \delta(z - z_i) \rangle}{L_x \cdot L_y \cdot d}$$

where Lx and Ly are the simulation dimensions in x and y directions, respectively. d is the thickness of the rectangular shell and a value of 0.1 Å was used in the present study. N is the number of water molecules in each shell and z_i is the z-component coordinates of water molecule i. Here, it is worthwhile noting that the O atoms are used to represent the water molecules. First, we counted the number of water molecules in each shell (N) according to the z-coordinates of oxygen atoms. Then, the number density in each shell can be obtained via the equation $\rho(z) =$

$\frac{N(z)}{V}$, wherein the accessible volume V equal to the product of L_x, L_y, and d. Finally, the number density was converted it into the mass density following the relation: $\rho_m(z) = \frac{\rho(z)\cdot M}{N_A}$ (M refers to the molecular weight of water molecule and N_A is the Avogadro constant). The equation and parameters used to calculate density distribution are listed in Table S4 below. The obtained density distribution profiles are shown in Fig. S10 and it can be found that the maximum densities appear at the central region between two graphyne nanosheets for all three systems. Since the distance of adjacent graphyne nanosheets remains a constant of 3.4 Å during the simulations, this leads to the difference between two peaks around 3.4 Å.

Table S4. The equation and parameters used to calculate density distribution.

Equation for density calculation	$\rho(z) = \frac{\langle \sum_i^N \delta(z - z_i) \rangle}{L_x \cdot L_y \cdot d}$
Thickness of rectangular shell (d)	0.1 Å
Accessible volume (V)	$V = L_x \cdot L_y \cdot d$
Number density to mass density	$\rho_m(z) = \frac{\rho(z) \cdot M}{N_A}$
Bulk density line (ρ_0)	1.0 g/cm^3

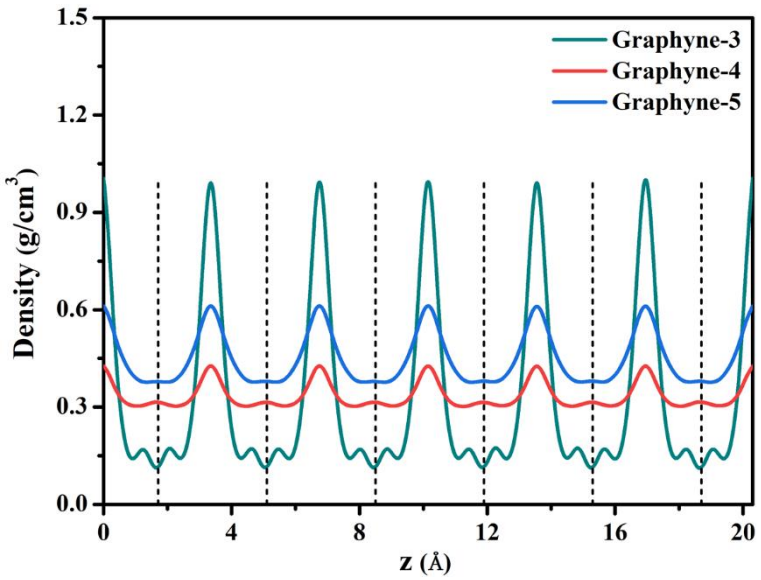

Fig. S10. The density distributions of water molecules along the axial direction for different graphyne membranes. Note: the dash line represents the position of graphyne nanosheet.

References

S1. G. C. Zuo, R. Shen, S. J. Ma and W. L. Guo, *ACS Nano*, 2010, **4**, 205-210.