Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.


# Supplementary Materials for Understanding Water Slippage Through Carbon Nanotubes

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## 1. Simulation setup

Table S1: The geometries of CNTs and graphene with the single water model

| | D (Å) | supercell |
|---|---|---|
| (5,5) | 6.78 | a=b= D + ∼100 Å<br>c=7.38, 4.92 and 2.46 Å<br>for 1×1×3, 1×1×2 and 1×1×1 supercell |
| (6,6) | 8.14 | |
| (7,7) | 9.49 | |
| (8,8) | 10.85 | |
| (10,10) | 13.56 | |
| (12,12) | 16.27 | |
| (52,52) | 70.51 | |
| (100,100) | 135.60 | |
| graphene | — | a=7.38, 4.92 and 2.46 Å<br>for 3×24×1, 2×24×1 and 1×24×1 orthorhombic supercell<br>b=102.26 Å<br>c=100 Å |

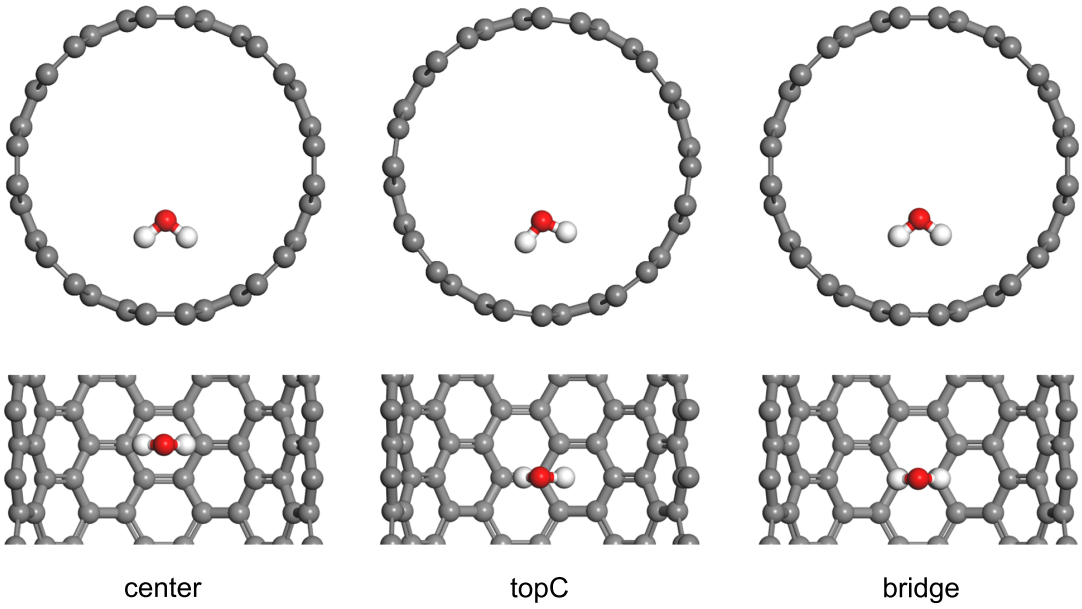


Figure S1: Taking (8,8) CNT with 1×1×3 supercell, the most stable configurations at each high-symmetric site in the single water model

# 2: The two friction models

## 2.1: Phononic friction model

The reason that the friction coefficeint $\eta_v$ in the viscous friction law ($F_f = m\eta_v v_{wall}$) can be parameterized with the phononic friction model ($\eta_v \approx aU_o^2$) is as follows. Pictorially speaking, when particles adsorb on surfaces, they vibrate both parallel and perpendicular to the surface, the parallel component of which probes the potential corrugation of the adsorbatesubstrate interaction. It was later found the dissipation due to the parallel vibration of particles is correlated with sliding friction, i.e., the dissipation energy resulting from the vibration of particles was approximately equivalent to the frictional energy resulting from the sliding of particles along a surface. The details are referred to Ref.[1]

### 2.2: Einsteins diffusion model

Einstein's diffusion model employed in our study can date back to Ref.:[1] Simply put, Einstein noted that the fluctuation of particles has the same origin as dissipation since the same irregular force which causes the diffusion of particles in Brownian motion could also cause friction especially when the particles are moving through the fluid. Using statistical mechanics, Einstein links the particles self-diffusion coefficient $D$ with the viscous friction coefficient $\eta$: $D = \frac{k_B T}{\eta}$. The diffusion coefficient $D$ originating from Ref.[2] corresponds to self-diffusion and can be frequently parameterized as an Arrhenius-type equation.

## 3: Calculation details of Slip lengths

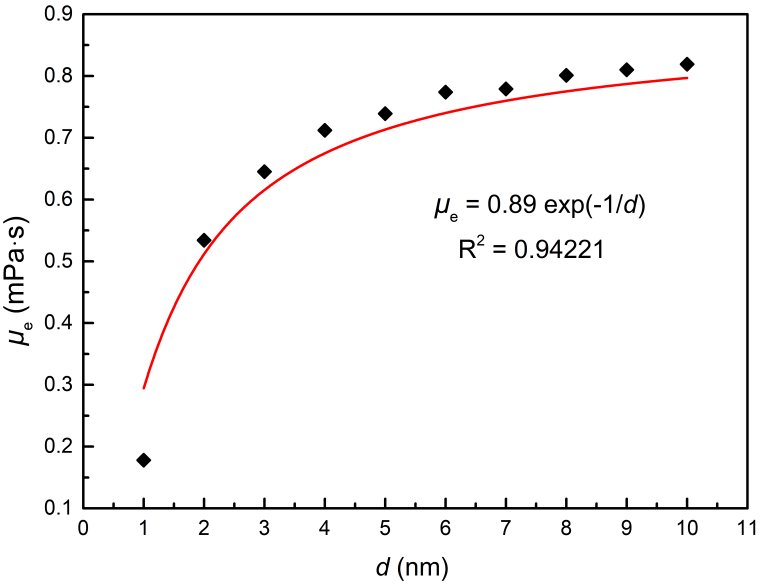


Figure S2: The size-dependent effective viscosity obtained from fitting.

## 4: Mechanism

We refers to the conventional definition of hydrogen bond: the O-O distance is less than 3.5 Å, and the angle of $\angle H - O \cdots O$ is less than 30°. For simplicity, we only refer to the

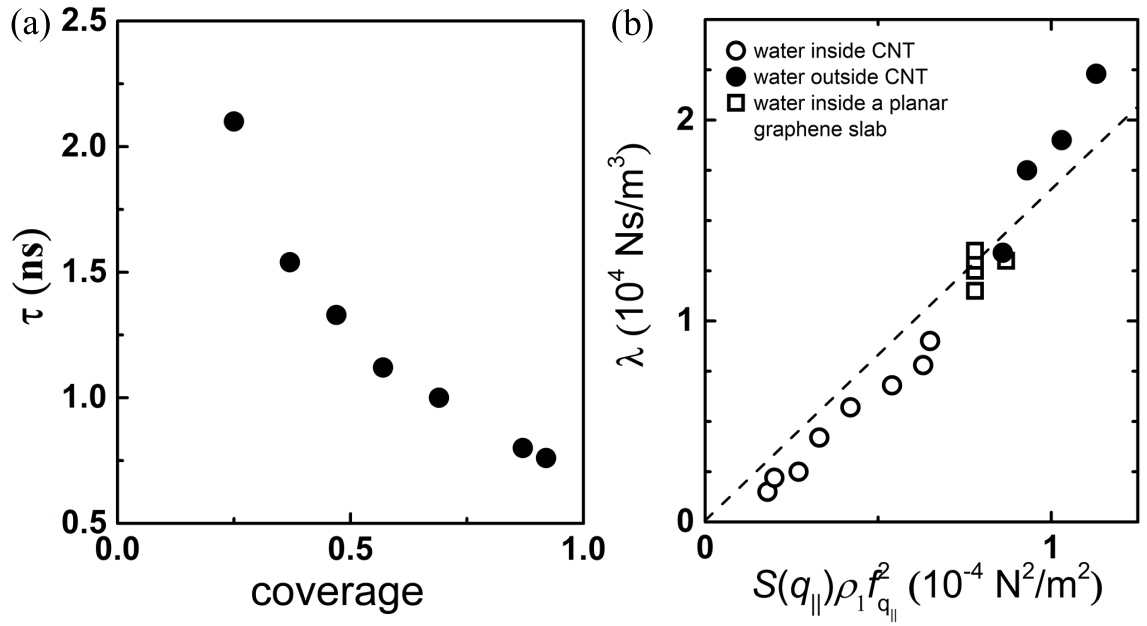


Figure S3: (a) The variation of slip time $\tau$ with the coverage, for Xe on Ag(111).[3,4] (b) The friction coefficient $\lambda$ as a function of its contributions —structural factor $S(q_{||})$, contact density $\rho_1$ and potential corrugation $f_{q_{||}}$.[5] This plot gathers results for all confinements and geometries.

intermolecular distance (i.e., the O-O distance) in the main text. The hydrogen bond in the confined conditions is not totally similar to the conventionally defined one. A more detailed analysis of hydrogen bonds is shown in Figs. S7∼S9.

As shown in Figure S6, one of the H atoms in water molecule points from toward tube wall to along tube axis, with the introduction of hydrogen bond between water molecules. For $d_{w-w}$ = 7.38 Å [Figure S7], the configuration, with both H atoms of water molecule pointing toward the wall, doesn't change with the adsorption site and CNT's diameter. For $d_{w-w}$ = 4.92 Å [Figure S8], the water configuration at the topC site change from with both H atoms pointing to the wall to one of H pointing to O of the next periodic water molecule, but with the $\angle H-O\cdots O$ larger than 30°. Notably, the change of water orientation takes place at all three adsorption sites for $d_{w-w}$ = 2.46 Å [Figure S9] with $\angle H-O\cdots O$ less than 30°.

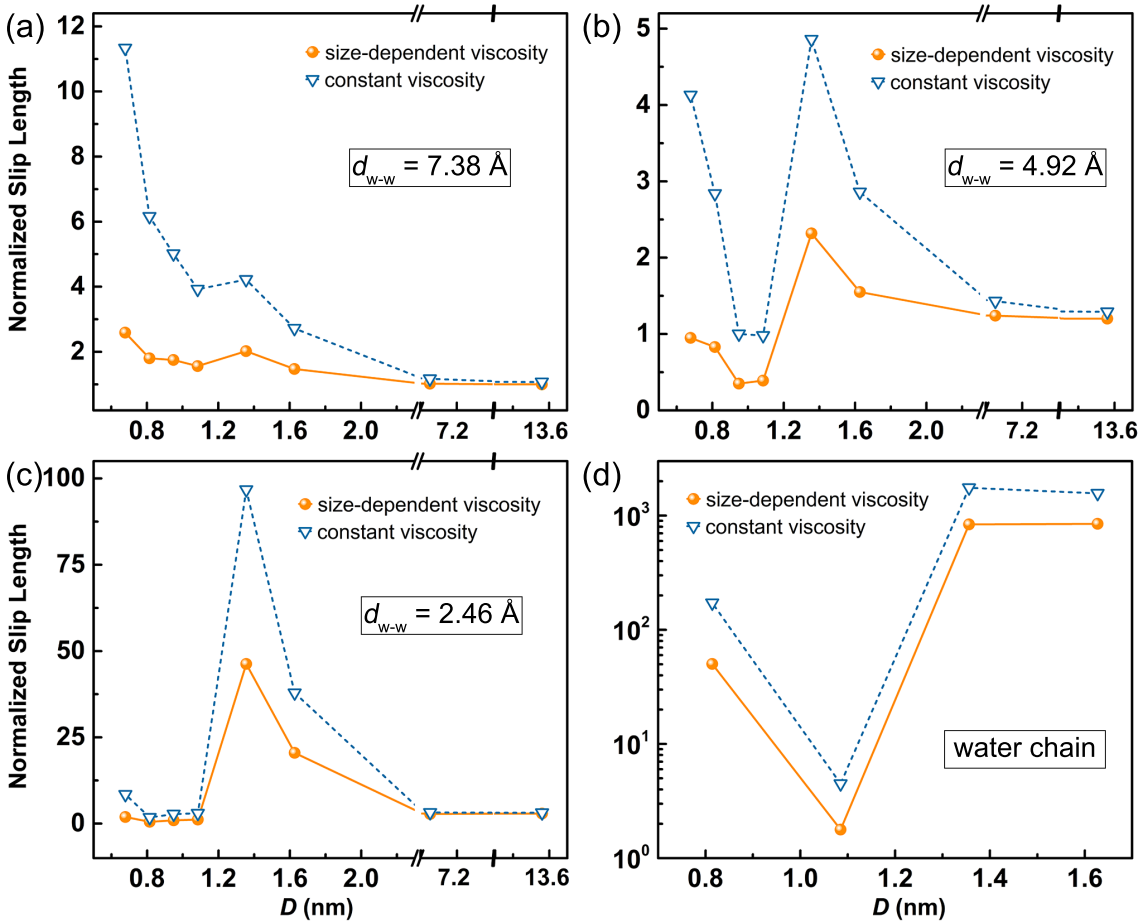


Figure S4: With the phononic friction model, the variation of normalized slip length with the tube size using both the size-dependent viscosity (see Figure 1(c) in the main text) and a constant viscosity, for both the (a-c) single water and (d) water chain model.

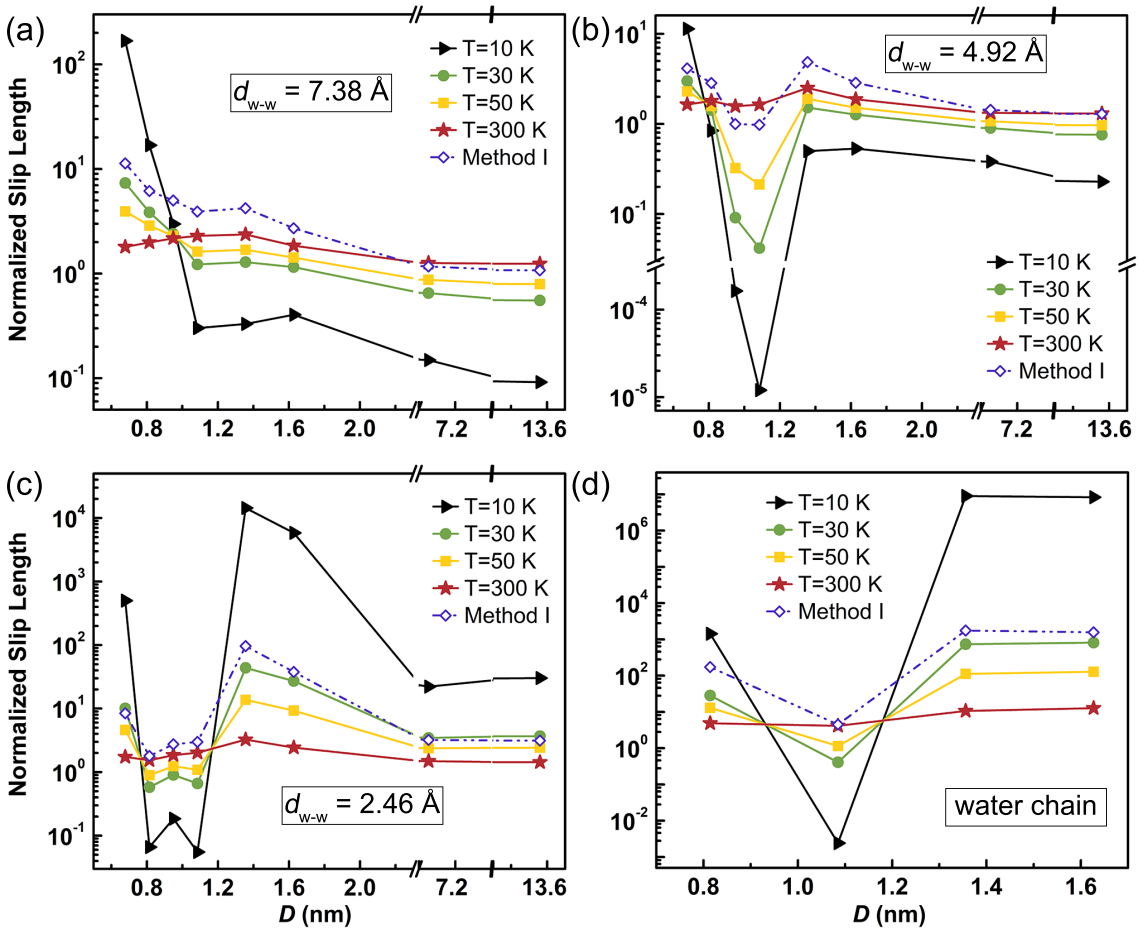


Figure S5: The slip lengths derived from Einsteins diffusion model with a constant viscosity as a comparison with Figure 2 in the main text.

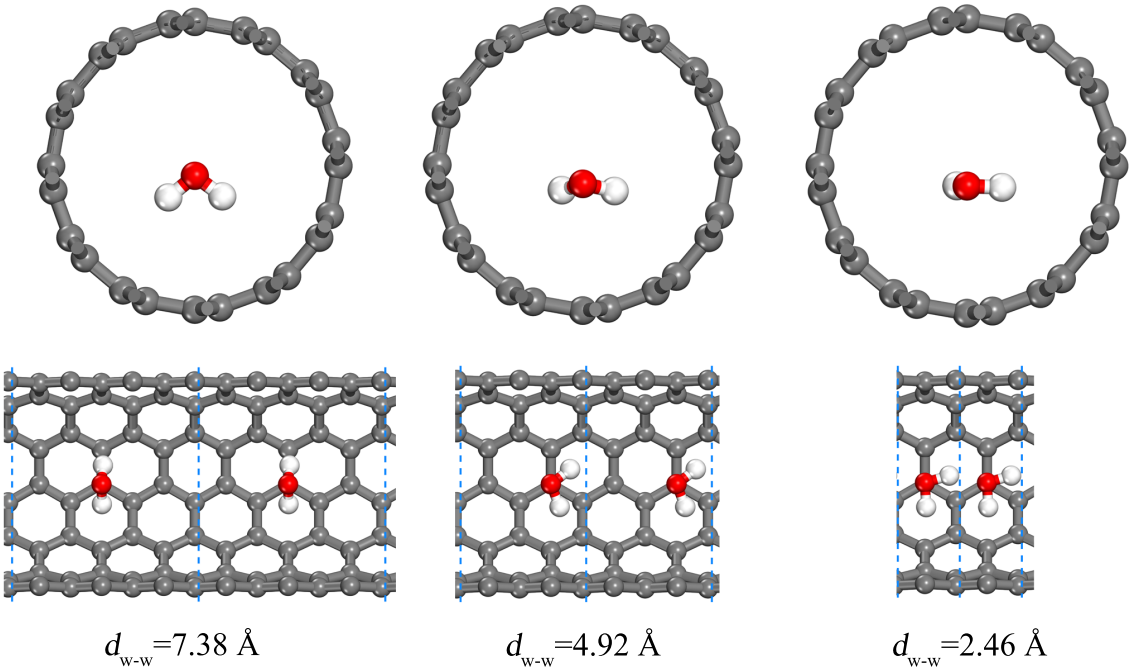


Figure S6: Take the topC site of (6,6) CNT, the change of dipole orientation of water molecule with the introduction of hydrogen bond. The blue dashed line indicates the unit cell along the tube axis.

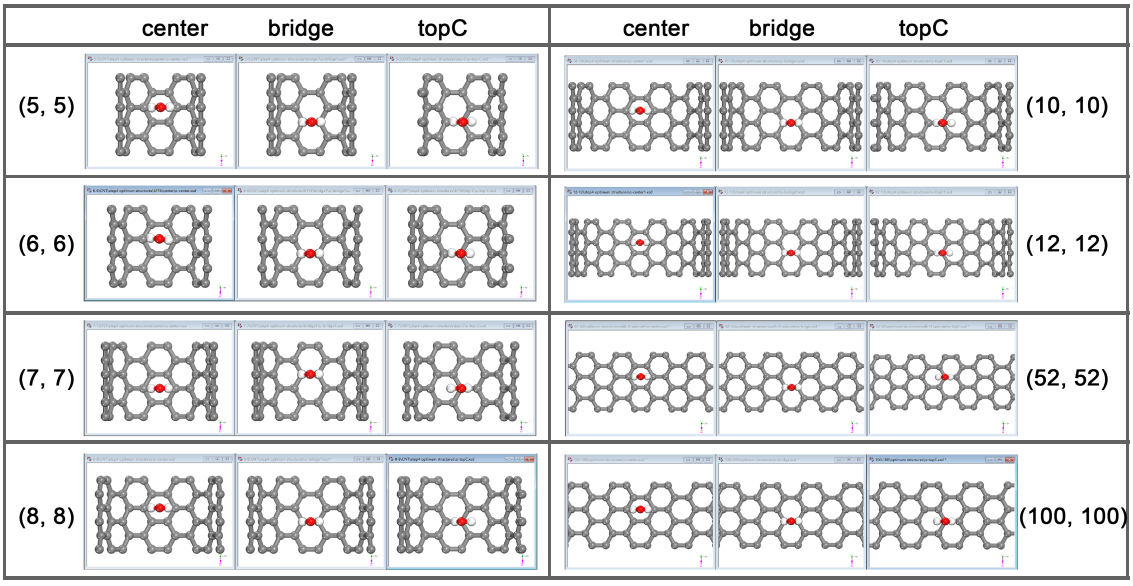


Figure S7: For $d_{w-w}$ = 7.38 Å, water configurations at the above three high-symmetric sites in the varied CNTs.

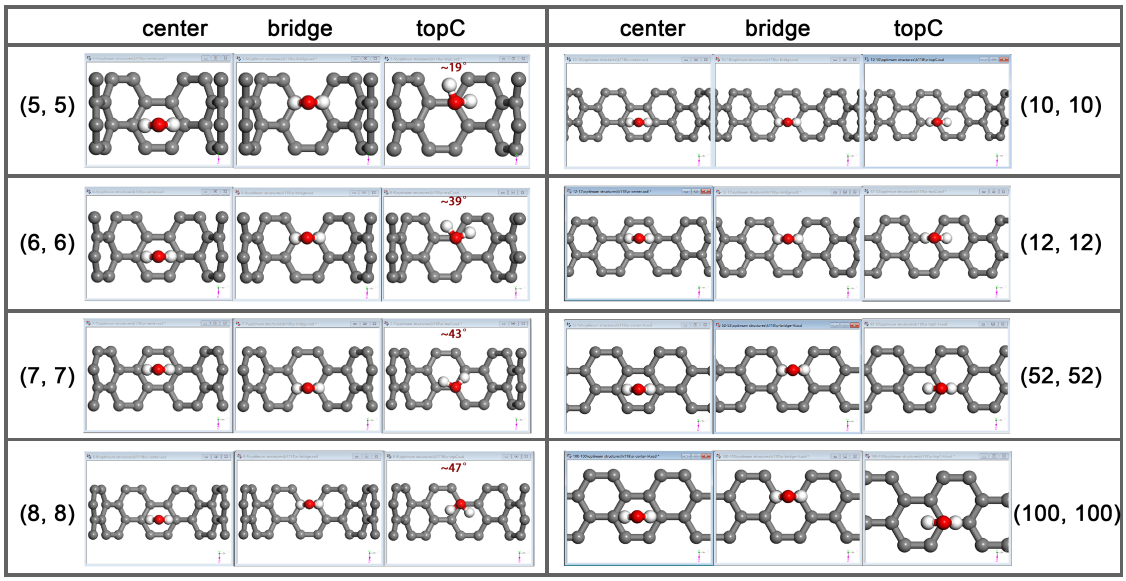


Figure S8: For $d_{w-w} = 4.92$ Å, water configurations at the above three high-symmetric sites in the varied CNTs. The angle of $H - O \cdots O$ is marked for the cases with one of H atom pointing to the next periodic water molecule

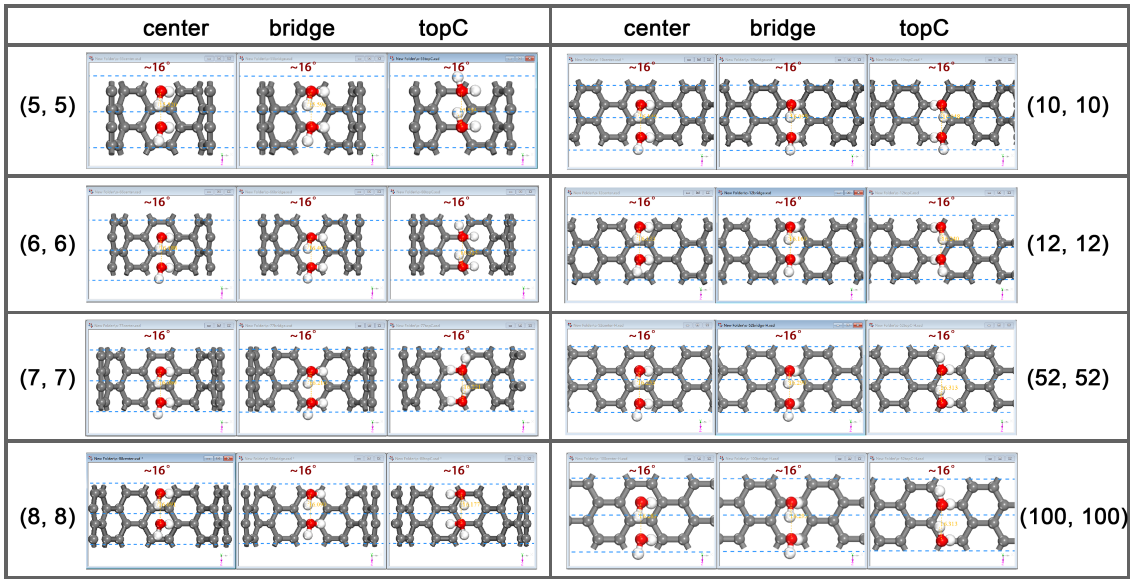


Figure S9: For $d_{w-w} = 2.46$ Å, water configurations at the above three high-symmetric sites in the varied CNTs. The angle of $H - O \cdots O$ is $\sim 16°$, marked correspondingly in the figure. The blue dashed line indicates the unit cell along the tube axis.

# References


(1) Krim, J. Friction and energy dissipation mechanisms in adsorbed molecules and molecularly thin films. *Adv. Phys.* **2012**, *61*, 155–323.

(2) Pisov, S.; Tosatti, E.; Tartaglino, U.; Vanossi, A. Gold clusters sliding on graphite: a possible quartz crystal microbalance experiment? *J. Phys. Condens. Matter* **2007**, *19*, 305015.

(3) Daly, C.; Krim, J. Sliding Friction of Solid Xenon Monolayers and Bilayers on Ag(111). *Phys. Rev. Lett.* **1996**, *76*, 803–806.

(4) Liebsch, A.; Goncalves, S.; Kiwi, M. Electronic versus phononic friction of xenon on silver. *Phys. Rev. B* **1999**, *60*, 5034.

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