Supporting Information for the Effects of Electrostatic Interaction and Chirality on the Friction Coefficient of Water Flow Inside Single-Walled Carbon Nanotubes and Boron Nitride Nanotubes

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1. Standard Deviation of the Friction Coefficient Calculated from the Green-Kubo Formula

In order to help the convergence of the friction coefficient in the Green-Kubo (GK) calculation in equilibrium molecular dynamic (EMD) simulations, different number of ensembles (from 10 to 50) were studied. The final results in the main article are reported with 50 ensembles for the one water models, and 30 ensembles for the multiple water models. Figure S1 shows the standard deviation (STD) of the calculated friction coefficient of one water molecule transport in the zigzag (14, 0) carbon nanotube (CNT) with artificial partial charges of +/- 1.0 *e*. The STD exponentially decays with the number of independent runs, and the exponential index is ~ - 0.6. Meanwhile, for the zigzag (14, 0) boron nitride nanotube (BNNT) case, the exponential decay index is ~ - 0.5. These observations imply that the friction coefficient data calculated from GK roughly follows a normal distribution, which should have STD reducing with the number of samples at an exponential of - 0.5. [1] The standard errors used in the figures are calculated by STD/(number of ensembles)$^{1/2}$.

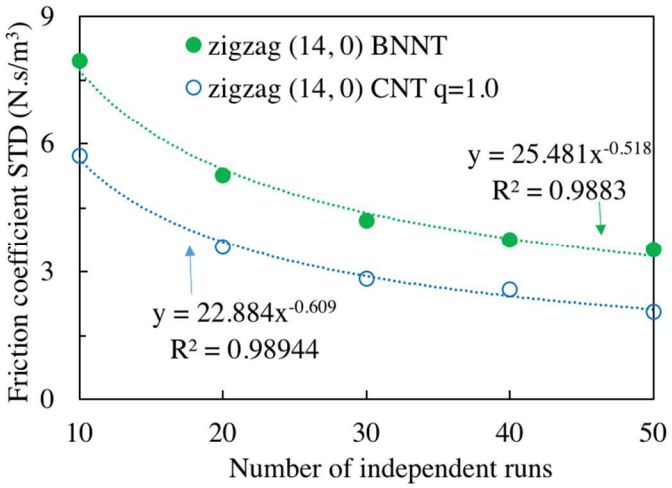

Figure S1. The STD of the friction coefficient is plotted against the number of independent EMD simulations. The green closed circles represent the STD of the friction coefficients of one water molecule transporting in a zigzag (14, 0) BNNT, and the blue open circles represent that in a zigzag (14, 0) CNT with partial changes of +/- 1.0 *e*. The dashed green and blue lines are the exponential fitting curves.

2. Raw Friction Coefficient Data

The friction coefficient is calculated in EMD with the Green-Kubo formula (Eq. 1 in the main text), which integrates the autocorrelation function of the forces experienced by water in the axial direction. When the integration time of the autocorrelation is large enough, the friction coefficient should converge as the autocorrelation decays to zero. For each simulation, we have the production simulation for 1 ns, and the autocorrelation as a function of time is calculated from 0 to 500 ps. Figure S2(a) shows the raw data from the cases where one water molecule transports in different types of nanotubes after 50 ensembles average. Figure S2(b) shows that applying the smooth function in MATLAB with a step size of 5000 the friction coefficient curves are more distinguishable. It is obvious that the zigzag (14, 0) CNT with partial charges of

+/- 1.0 *e* in Fig. S2 (dark blue curve) and the zigzag (14, 0) BNNT (green curve) have higher friction coefficient than the other four types of nanotubes.

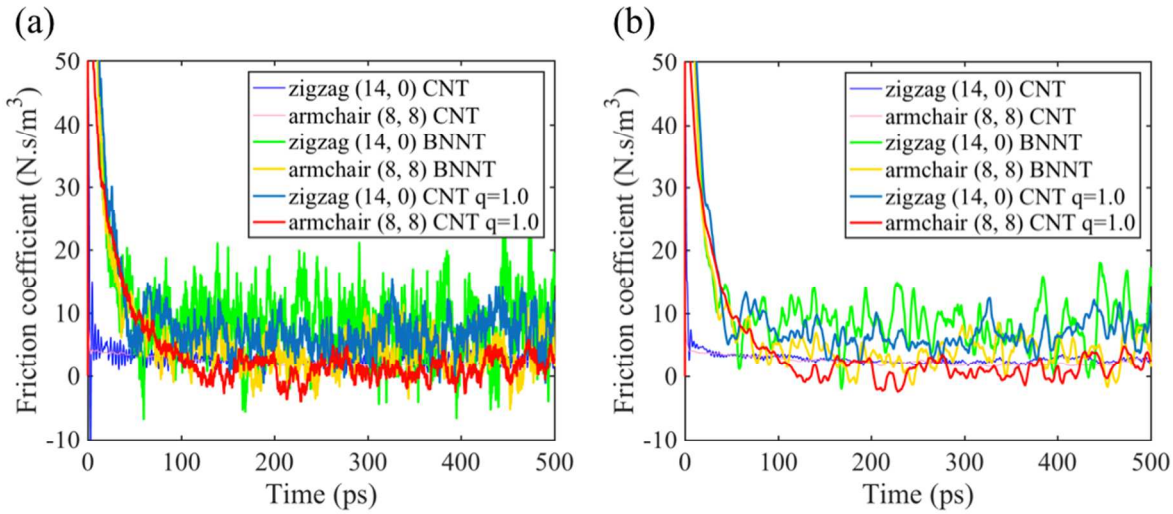

Figure S2. Friction coefficients of one water molecular transport inside different CNTs and BNNTs. (a) The raw friction coefficient data as a function of integration time; (b) the smoothed data from (a) by a running average of 5000 time steps.

Figure S3 shows that with multiple water molecules filled inside the nanotube, the friction coefficients with respect to the integration time show a trend similar to that of the single water molecule cases. The zigzag (14, 0) CNT with partial charges of +/- 1.0 *e* (dark blue curve) and the zigzag (14, 0) BNNT (light green curve) have much higher friction coefficients than the other four types of nanotubes, Fig. S3.

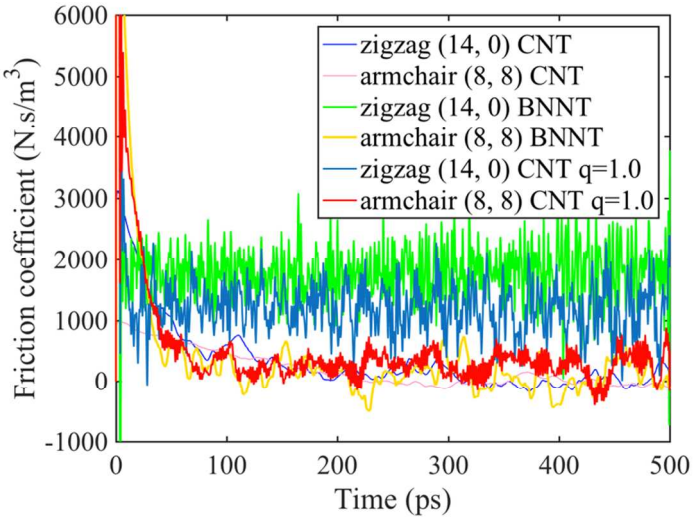

Figure S3. Friction coefficients of multiple water molecules transport inside different types of CNTs and BNNTs. The data shown are from 30 ensemble average and smoothed by a running average over 5000 time steps.

3. Convergence of Friction Coefficient

From Figs. S2 and S3, the convergences of the friction coefficients are evident in the integration time range of 200-500 ps. Our reported data are based on averaging the converged friction coefficients over this time range. However, a previous study used much shorter integration time (2 ps in their supporting information) to obtain the converged friction coefficient in CNTs. [2] For CNTs the friction coefficients at 2 ps at the the peak value, which will decay in the following a few hundreds of picoseconds. We do not agree with such a treatment. However, even if we use the short integration time to obtain the converged values and taking the first peaks (0.5 ps) of the integration curves (Fig. S4(a)), the trend in friction coefficients (Fig. S4(b)) is surprisingly the same as that shown in Fig. 2 in the main text. Figure S5 shows a clearer comparison of the convergences in the short and long integration time for CNTs, which can be compared with reference 2.

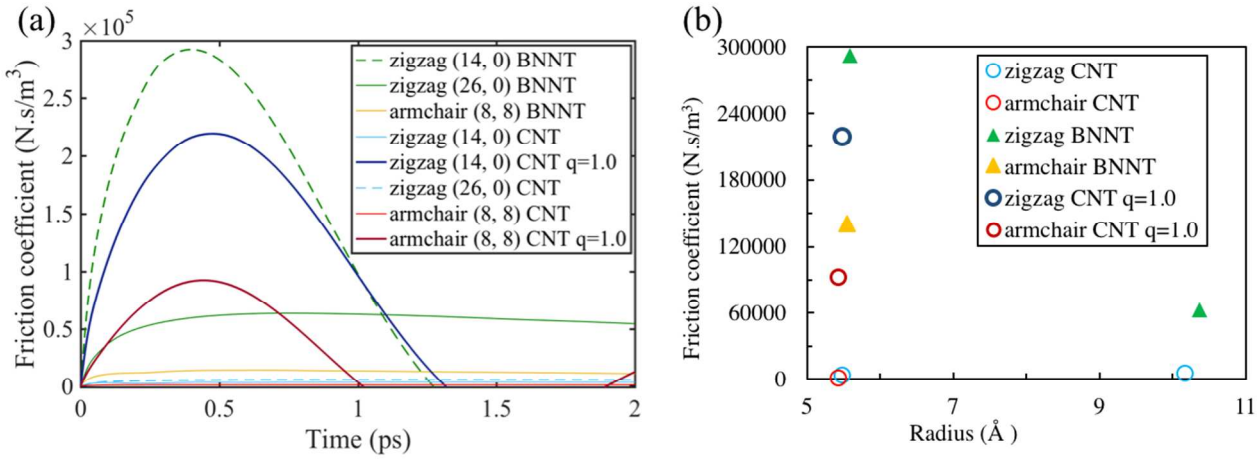

Figure S4. Friction coefficients of multiple water molecules transporting in CNTs and BNNTs. (a) Raw data from 0 to 2 ps of integration time; (b) Friction coefficient values obtained as the peaks in panel (a).

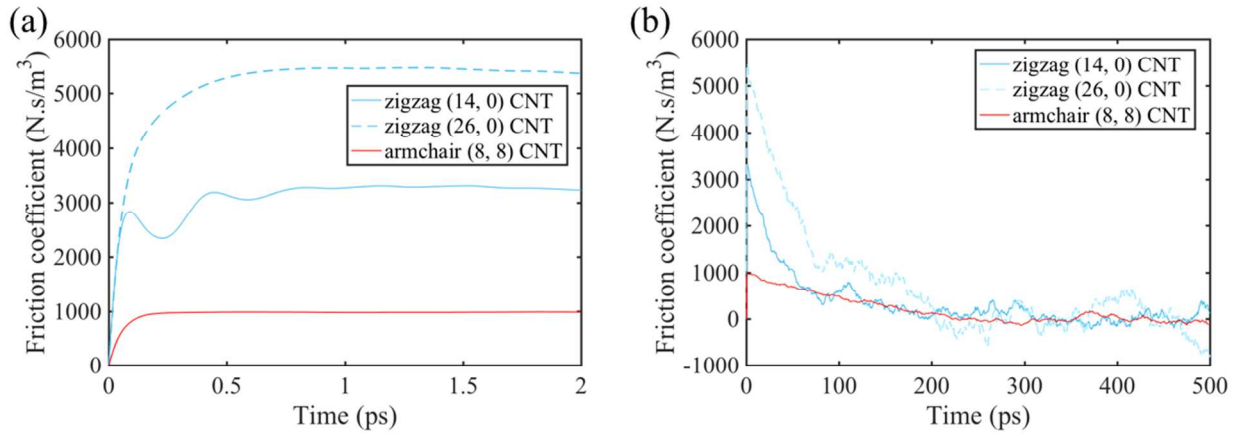

Figure S5. Friction coefficients of multiple water molecules transporting in CNTs. Comparing integration time (a) from 0 to 2 ps and (b) from 0 to 500 ps.

4. Friction Coefficient of Water in Nanotubes with Different Sizes

The friction coefficients of a single water in a zigzag (26, 0) CNT with a radius of 10.17 Å and a zigzag (26, 0) BNNT with a radius of 10.37 Å are shown in Fig. S6(a). The friction coefficients of multiple water in the zigzag (26, 0) CNT and the zigzag (26, 0) BNNT are shown in Fig. S6(b). In both cases, it is obvious that the zigzag (26, 0) BNNT has a much larger friction coefficient than that of the zigzag (26, 0) CNT.

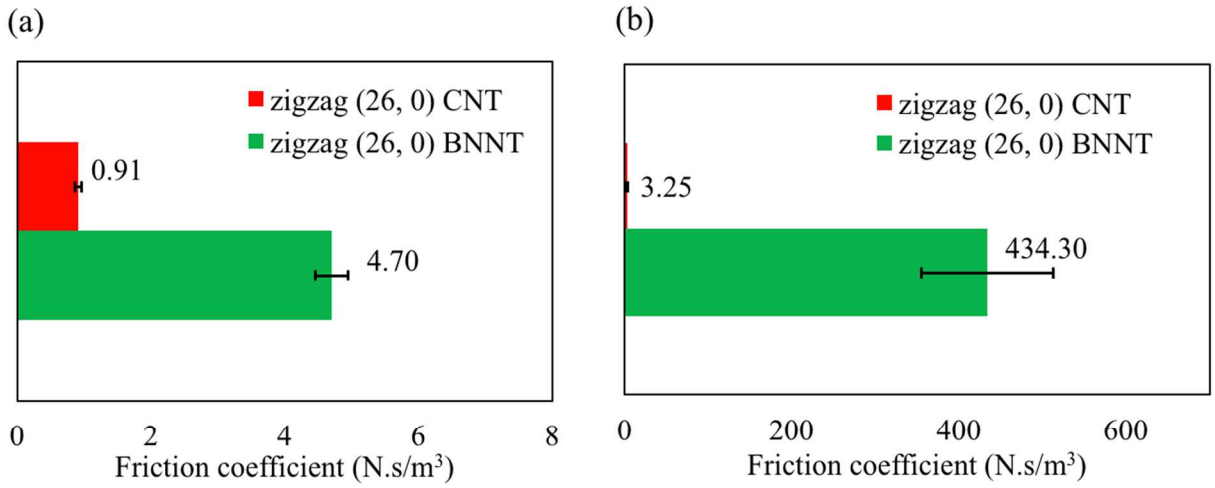

Figure S6. Friction coefficients of: (a) a single water molecule and (b) multiple water molecules in a zigzag (26, 0) CNT and a zigzag (26, 0) BNNT. For the multiple water molecules model the zigzag (26, 0) CNT has 176 water molecules, and the zigzag (26, 0) BNNT has 209 water molecules. The number of water molecules are determined from GCMC simulations. The error bar is calculated by mean value/(number of ensembles)$^{1/2}$, when STD is larger than the mean value.

5. Friction Coefficient of Water in the Zigzag (14, 0) CNT with Different Partial Charges

Different values of partial charges are artificially assigned to the zigzag (14, 0) CNT. Figure S8 that adding partial charges of +/-0.1 *e* will lead to a slight increase in the friction coefficient

(green bar). When the partial charges are +/-1.0 *e* or +/-1.5 *e* (blue and brown bars), the friction coefficients show more significant increases (Fig. R4). These observations are intuitive as larger partial charges induce stronger electrostatic interactions and thus larger friction.

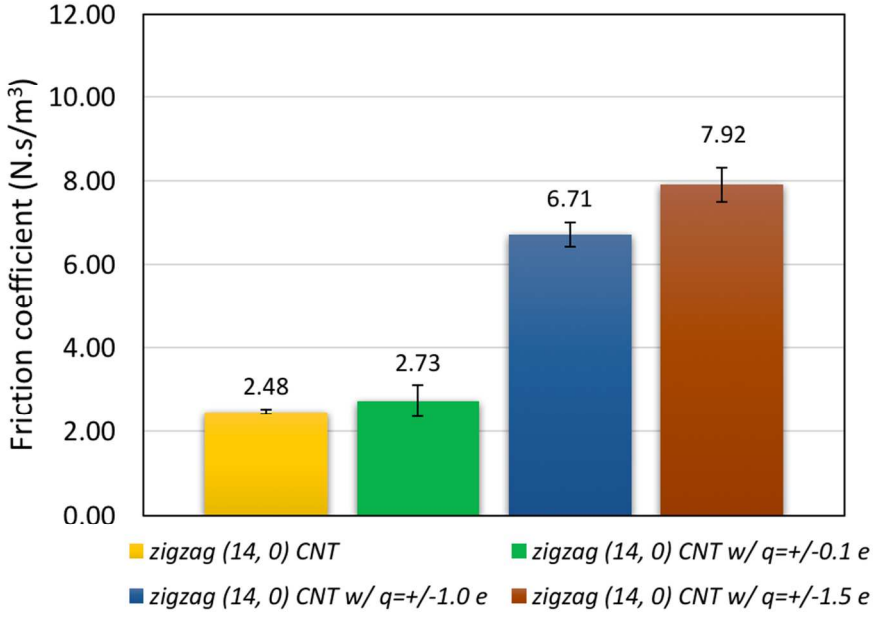

Figure S7. Friction coefficients of one water molecule transport inside zigzag (14, 0) CNT with different artificial partial charges.

6. The Distributions of Water Molecules in Different Nanotubes In the Axial Direction

In order to understand the distributions of water molecules (mainly the oxygen atoms) inside nanotubes during MD and map them back onto the potential energy landscape, we analyzed 150-500 trajectories during the NVE ensembles for different types of nanotubes with multiple water molecules. Figure S7 shows the numbers of water molecules at the center of the nanotubes around 15.5 Å, where Fig. 3 in the main manuscript shows a low energy region, and around 16.0 Å, which is a high energy region, for the zigzag (14, 0) BNNT and the zigzag (14, 0) CNT with artificial partial charges of +/- 1.0 *e*.

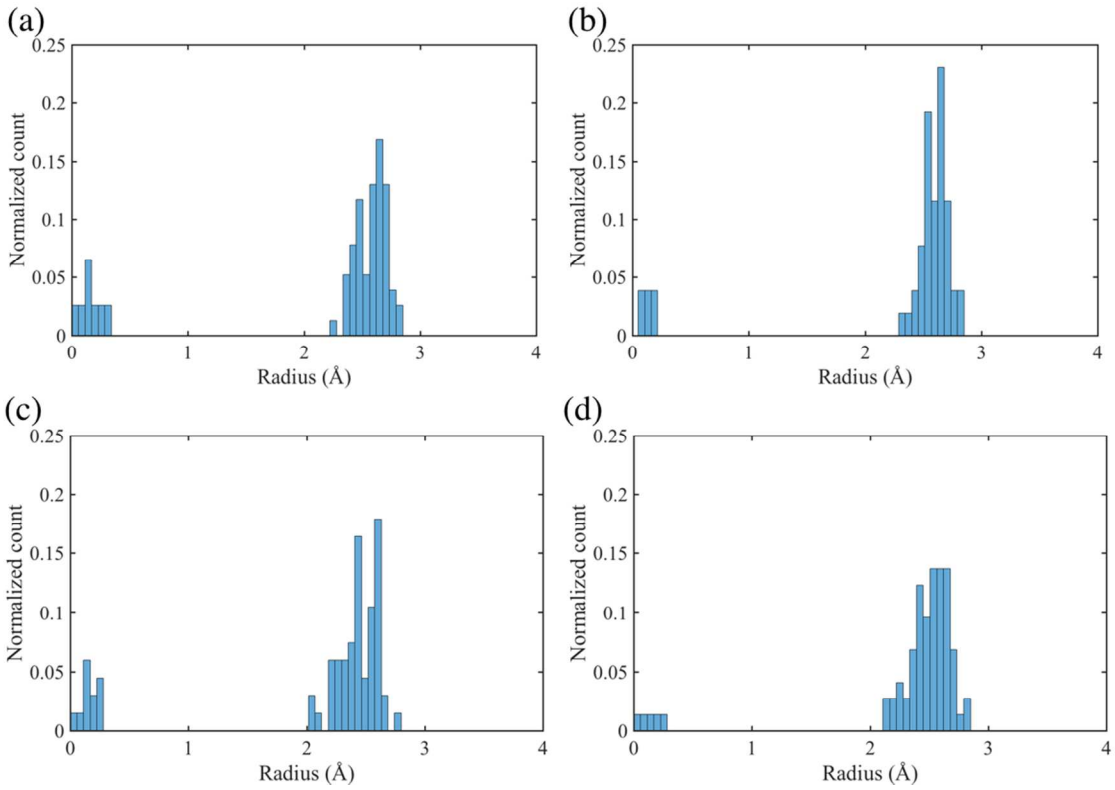

Figure S8. The normalized distribution of oxygen atoms: along the axial direction in the zigzag (14, 0) BNNT counted over the nanotube length between (a) 15.4 to 15.6 Å, and (b) 15.9 to 16.1 Å; along the radial direction in the zigzag (14, 0) CNT with artificial partial charges of +/- 1.0 *e* counted over the nanotube length between (c) 15.4 to 15.6 Å, and (d) 15.9 to 16.1 Å.

7. Friction Coefficient Data Summarize

All the reported friction coefficient data are summarized in Table S1, which can give reader more information of our simulation result.

Table S1. Friction coefficient values from Figs. 2, 7, 8, S6 and S8.

Fig. 2	Friction coefficient	mean	error bar
Single TIP3P water	zigzag (14,0) CNT	2.48	0.05
	zigzag (14,0) BNNT	8.26	0.50
	armchair (8,8) CNT	2.05	0.04
	armchair (8,8) BNNT	3.72	0.31
	zigzag (14,0) CNT w/ q=+/-1.0 e	6.71	0.29
	armchair (8,8) CNT w/ q=+/-1.0 e	1.08	0.22
Fig. 7	Friction coefficient	mean	error bar
Multiple TIP3P water	zigzag (14,0) CNT	60.51	11.05
	zigzag (14,0) BNNT	1803.12	79.10
	armchair (8,8) CNT	1.49	0.27
	armchair (8,8) BNNT	77.83	14.21
	zigzag (14,0) CNT w/ q=+/-1.0 e	1171.08	73.89
	armchair (8,8) CNT w/ q=+/-1.0 e	275.49	37.71
Fig. 8a	Friction coefficient	mean	error bar
Multiple TIP4P/2005 water	zigzag (14,0) CNT	89.41	12.61
	zigzag (14,0) BNNT	1630.17	297.63
	armchair (8,8) CNT	73.55	8.09
	armchair (8,8) BNNT	195.79	35.75
Fig. 8b	Friction coefficient	mean	error bar
Tersoff C/B/N	zigzag (14,0) CNT	70.83	12.93

Multiple TIP3P water	zigzag (14,0) BNNT	2205.71	402.71
Fig. S6a	Friction coefficient	mean	error bar
Single TIP3P water	zigzag (26,0) CNT	0.91	0.05
	zigzag (26,0) BNNT	4.70	0.25
Fig. S6b	Friction coefficient	mean	error bar
Multiple TIP3P water	zigzag (26,0) CNT	3.25	0.59
	zigzag (26,0) BNNT	434.30	79.29
Fig. S8	Friction coefficient	mean	error bar
Single TIP3P water	zigzag (14,0) CNT	2.48	0.05
	zigzag (14,0) CNT w/ q=+/-0.1 e	2.73	0.38
	zigzag (14,0) CNT w/ q=+/-1.0 e	6.71	0.29
	zigzag (14,0) CNT w/ q=+/-1.5 e	7.92	0.41

Reference

1. Wang, Z., Safarkhani, S., Lin, G., & Ruan, X. Uncertainty quantification of thermal conductivities from equilibrium molecular dynamics simulations. *International Journal of Heat and Mass Transfer* **2017,** *112*, 267-278.

2. Falk, K.; Sedlmeier, F.; Joly, L.; Netz, R. R.; Bocquet, L. Molecular Origin of Fast Water Transport in Carbon Nanotube Membranes: Superlubricity Versus Curvature Dependent Friction. *Nano letters* **2010**, *10*, 4067-4073.