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

Electronic Supplementary Information

A Numerical Model for Water Hydration on Nanosurface: From Friction to Hydrophilicity and Hydrophobicity

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Table S1. The forcefield parameters of five native nanostructures.

	□(kJ/mol)	□ (nm)	Charge (e)
C(GRA) [1]	0.360	0.340	0
C(C_3N) [2]	0.360	0.340	0.056
N(C_3N)	0.711	0.325	-0.168
C(C_2N) [3]	0.360	0.340	0.24
N(C_2N)	0.711	0.325	-0.48
B(BC_3) [2]	0.399	0.345	0.378
C(BC_3)	0.360	0.340	-0.126
B(h-BN) [4]	0.399	0.345	0.37
N(h-BN)	0.711	0.325	-0.37

Table S2. Friction coefficient and the maximum corrugation of the free energy $\Delta G_{max\text{-}min}$ of the simulated models.

Model	Friction coefficient ($\times 10^5$ N s/m^3)	$\Delta G_{max\text{-}min}$(kJ/mol)
GRA	0.14 ± 0.01	0.59 ± 0.006
C_3N– 0	0.16 ± 0.01	0.60 ± 0.006
BC_3 – 0	0.26 ± 0.02	0.97 ± 0.009
h-BN– 0	0.30 ± 0.01	0.86 ± 0.008
h-BN	0.35 ± 0.01	0.88 ± 0.010
C_3N	0.47 ± 0.03	0.72 ± 0.009
C_3N– 1.1	0.50 ± 0.01	0.75 ± 0.009
C_3N– 1.2	0.57 ± 0.06	0.81 ± 0.009
C_3N– 1.3	0.65 ± 0.05	0.822 ± 0.009
C_3N– 1.4	0.72 ± 0.03	0.845 ± 0.009
C_3N– 1.5	0.79 ± 0.06	0.877 ± 0.009
C_2N– 0	2.61 ± 0.24	4.23 ± 0.039
BC_3	3.36 ± 0.18	2.78 ± 0.019
BC_3 – 1.1	4.44 ± 0.46	3.23 ± 0.022
BC_3 – 1.2	5.29 ± 0.20	3.77 ± 0.025
BC_3 – 1.3	5.95 ± 0.24	4.31 ± 0.028
BC_3 – 1.4	6.74 ± 0.47	4.92 ± 0.030
BC_3 – 1.5	7.72 ± 0.53	5.50 ± 0.032
C_2N	9.88 ± 0.20	10.16 ± 0.082
C_2N– 1.1	11.50 ± 0.07	10.79 ± 0.082
C_2N– 1.2	13.06 ± 0.12	11.51 ± 0.082
C_2N– 1.3	14.39 ± 0.11	12.47 ± 0.081
C_2N– 1.4	15.89 ± 0.27	13.29 ± 0.080
C_2N– 1.5	17.39 ± 0.32	14.03 ± 0.078

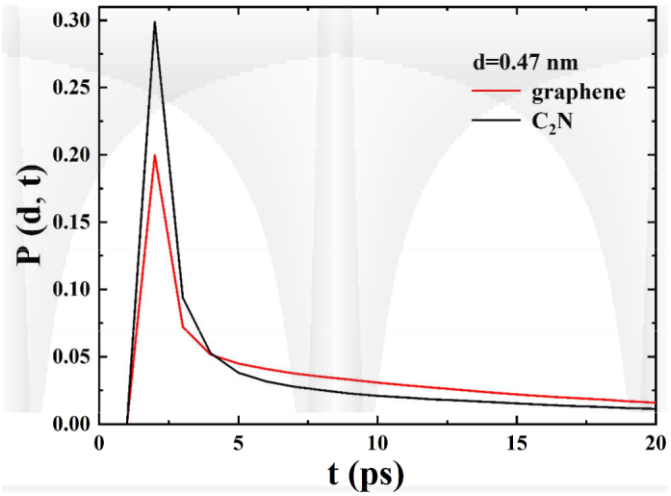

Fig. S1 The $P(d, t)$ (d = 0.47 nm) profiles with respect to t for GRA and C_2N, respectively.

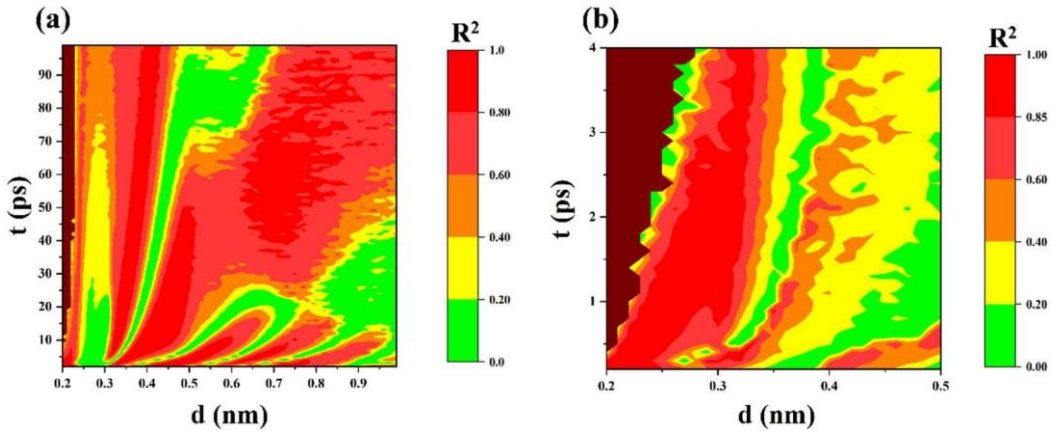

Fig. S2 Coefficient of determination (R^2) between □ and $P(d, t)$ for (a) all the models and (b) 11 models with low friction coefficients.

1. The position-restrained MD simulation

In the real conditions, water molecules interact with the atoms of nanomaterials and may force them to leave their original positions. Thus, the nanomaterials maybe not flat. To test how these structure changes affects the main results, we added simulations where position-restrains were added to the atoms of nanostructures instead of freezing them in the main discussions. The position restraint parameter is set to be $k = 50$ kJ/mol/rad^2. We did not consider fully relaxed nanostructures because in the experiments, the nanomaterials are usually placed on substrate (like Si and Cu) which can maintain a flat surface. The comparison of the friction coefficients for position-frozen and position-restrained MD simulations are summarized in Table S3 and Fig. S3 shown below. It is clear that the movements of the atoms only have a very small effect on the friction coefficients. Therefore, we believe that the results in the main manuscript are reasonable.

Table S3. The comparison of friction coefficients between the position-frozen and position-restrained MD simulations for five native 2D nanostructures.

Model	Friction coefficient ($\times 10^5$ N s/m^3)	
	position frozen	position restrained
GRA	0.14 ± 0.01	0.18 ± 0.01
h-BN	0.35 ± 0.01	0.28 ± 0.03
C_3N	0.47 ± 0.03	0.47 ± 0.03
BC_3	3.36 ± 0.18	3.53 ± 0.23
C_2N	9.88 ± 0.20	10.20 ± 0.24

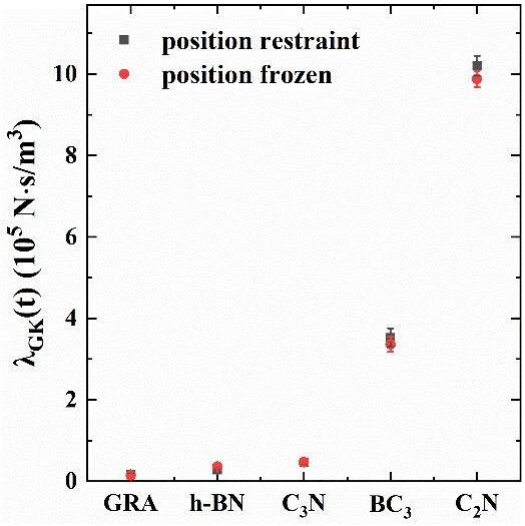

Fig. S3 The comparison of friction coefficients between the position-frozen and position-restrained MD simulations for five native 2D nanostructures.

2. Five more models to estimate the borderline between hydrophobic and hydrophilic surface

In the main manuscript, initially we have used 24 models. From Fig. 3(c)-3(e), the borderline between hydrophobic and hydrophilic surface is between $(0.79 \pm 0.06)\times10^5$ N s/m^3 (C_3N-1.5) and $(2.61 \pm 0.24)\times10^5$ N s/m^3 (C_2N– 0). To enhance the accuracy, we added five more models, say C_3N-1.6, C_3N-1.7, C_3N-1.8, C_3N-1.9 and C_3N-2.0, where the λ is summarized in Table S4. The dependence of λ with $P(d, t)$ are shown in Fig. S4, from which it is clear that these five more models are classified as hydrophobic. Summarizing all the results, the borderline is between $(1.35 \pm 0.08)\times10^5$ N s/m^3 (C_3N-2.0) and $(2.61 \pm 0.24)\times10^5$ N s/m^3 (C_2N- 0), which is estimated to be 2×10^5 N s/m^3.

Table S4. Friction coefficients of five more models.

Model	Friction coefficient ($\times10^5$ N s/m^3)
C_3N-1.6	0.91 ± 0.04
C_3N-1.7	0.97 ± 0.02
C_3N-1.8	1.06 ± 0.05
C_3N-1.9	1.18 ± 0.05
C_3N-2.0	1.35 ± 0.08

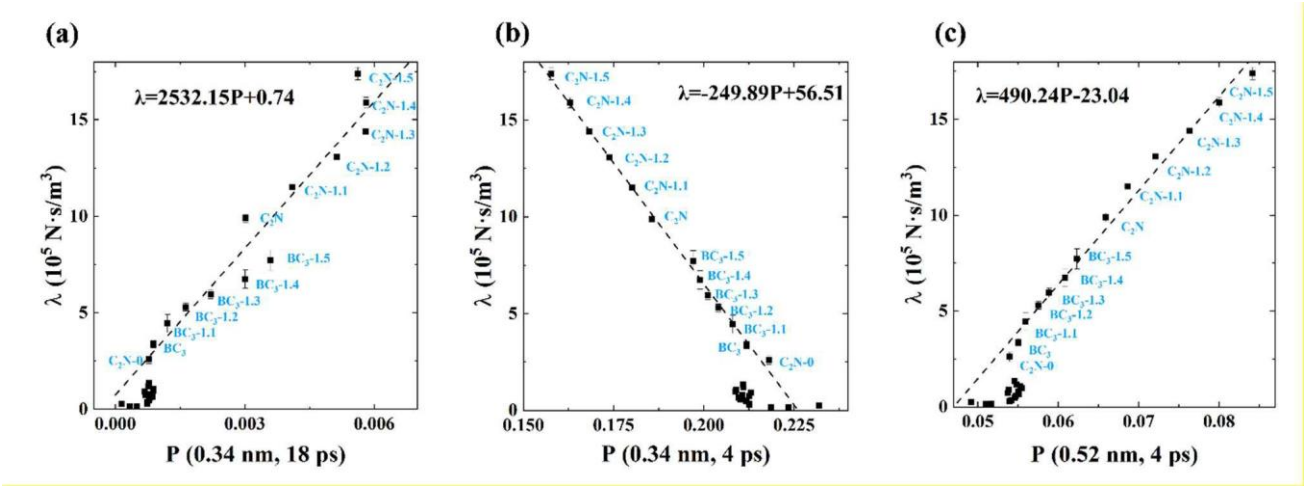

Fig. S4 The relationships between λ and $P(d, t)$ at point c, d and e in Fig. 3(a) of main manuscript for all the 29 systems, respectively. It is clear that the five more models are classified as hydrophobic surfaces.

Reference

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