Supporting Information

Adsorption of a Polyaromatic Compound on Silica Surfaces from Organic Solvents Studied by Molecular Dynamics Simulation and AFM Imaging

Yong Xiong,[†] Tiantian Cao,[†] Qian Chen,[†] Zhen Li,[†] Yue Yang,[†] Shengming Xu,[*,†] Shiling Yuan,[§] Johan Sjoblom,[‖] and Zhenghe Xu[*, †, ‡]

[†]Institute of Nuclear and New Energy Technology, Tsinghua University, Beijing 100084, China

[‡]Department of Chemical and Materials Engineering and Department of Mechanical Engineering, University of Alberta, Edmonton, Alberta T6G 1H9, Canada

[§]Key Laboratory of Colloid and Interface Chemistry, Shandong University, Jinan 250100, China

[‖]Department of Chemical Engineering, Ugelstad Laboratory, Norwegian University of Science and Technology (NTNU), Trondheim NO 7491, Norway

* Corresponding author: Zhenghe.xu@ualberta.ca
smxu@tsinghua.edu.cn
Phone: +1-780-492-7667
Fax: +1-780-492-2881

Table S1 United atom force field parameters for simulations

Atom type	σ/nm	ϵ/kJ·mol^{-1}	q/e
C5Pe			
H (-COOH)	0	0	Figure S1.1
O (-OH)	0.295	0.850	
C (-COOH)	0.358	0.277	
O (-C=O)	0.276	1.279	
CH2	0.407	0.411	
CH3	0.375	0.867	
N	0.357	0.293	
H	0.237	0.118	
Heptane			
CH2	0.407	0.411	Figure S1.2
CH3	0.375	0.867	
Toluene			
C	0.358	0.277	Figure S1.3
H	0.237	0.118	
CH3	0.375	0.867	
SiO_2 (surface atoms)			
H (Si-OH)	0	0	0.40
O (Si-OH)	0.295	0.850	-0.71
Si (Si-OH)	0.339	2.447	0.31

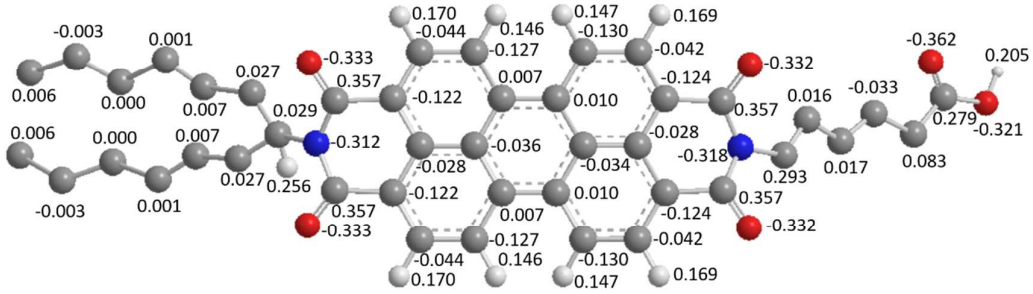

Figure S1.1 Charge distribution of C5Pe

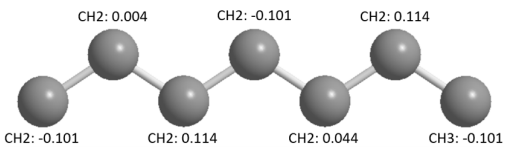

Figure S1.2 Charge distribution of heptane

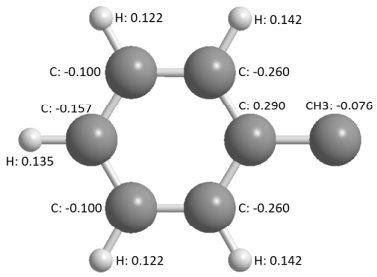

Figure S1.3 Charge distribution of toluene

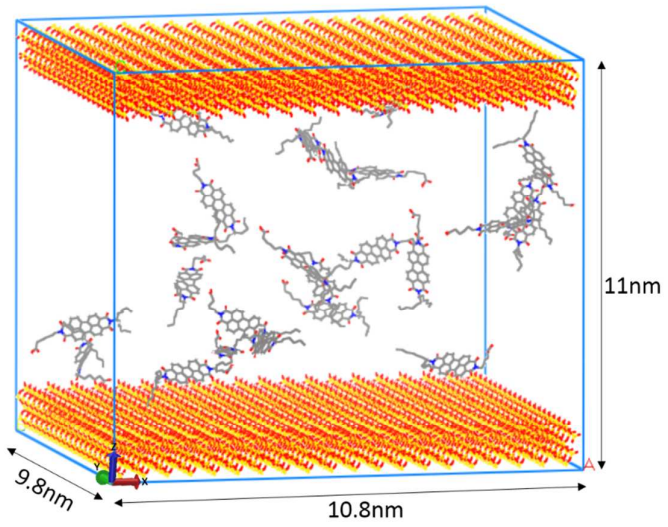

Figure S2. Initial box of simulation systems

All 24 C5Pe molecules were inserted in the middle of two SiO_2 slabs randomly with a simulation box size of 9.8x10.8x11 nm.

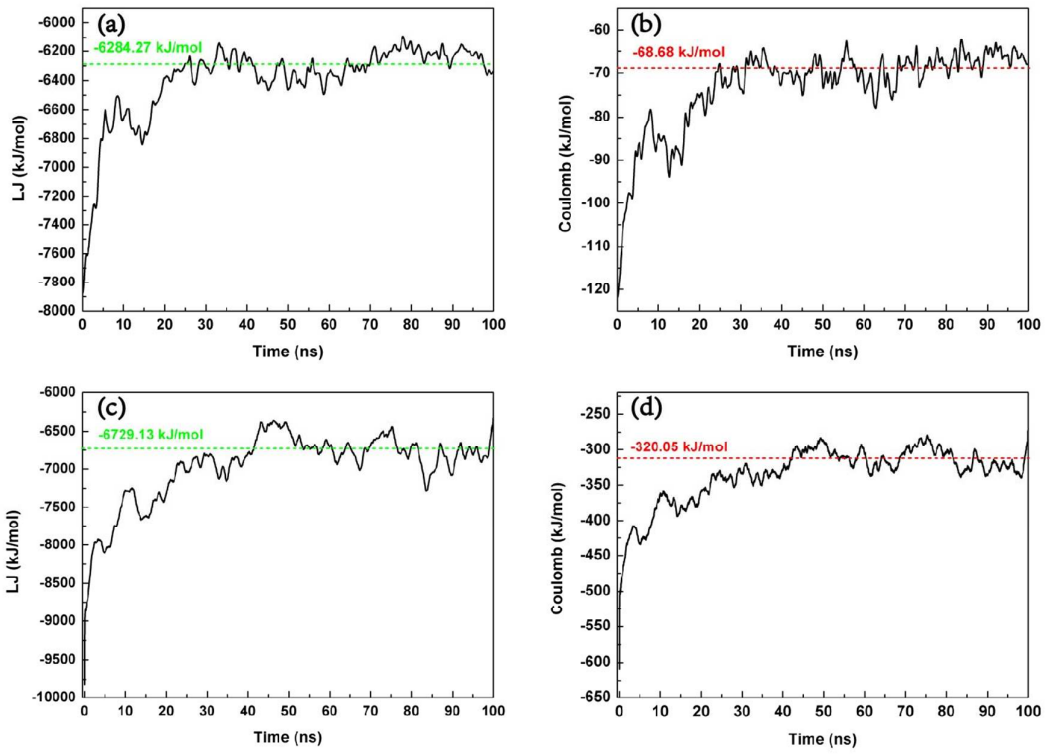

Figure S3. Interaction energy between C5Pe and silica surface (a) LJ interaction in heptane; (b) coulomb interaction in heptane; (c) LJ interaction in toluene; (d) coulomb interaction in toluene.

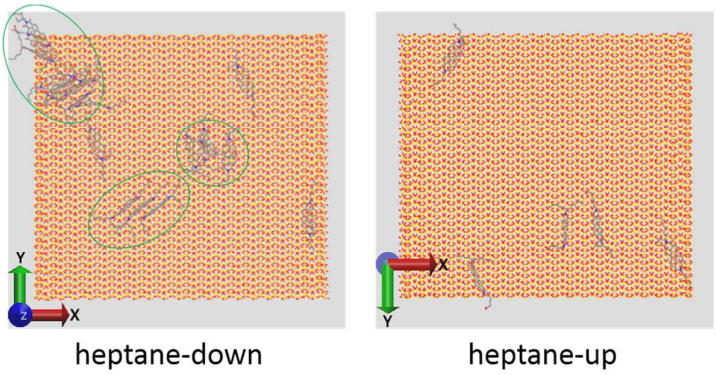

Figure S4. Partition states of systems in heptane

The simulation box was separated from the middle of Z axis. The top view (heptane-down) and upward view (heptane-up) of heptane system are shown separately. Three aggregates adsorbed were highlighted by green ellipse, while the others (8) as single molecules adsorbed in heptane on silica surface.

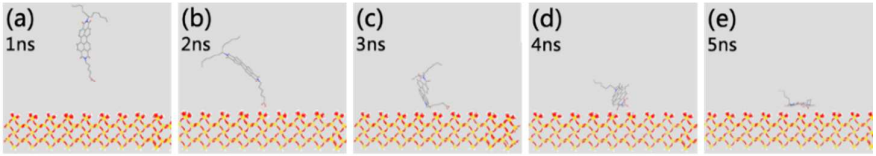

Figure S5. Molecular orientation transformed from slant to flat state on silica in heptane

The adsorption process of a C5Pe molecular was shown in picture from 1 ns to 5 ns. It is evident that the polar terminate carboxylic acid group adsorbed on silica surface first, gradually transferred to the slant state and finally to lay flat on silica surface with entire adsorption process being rapid within 5 ns.

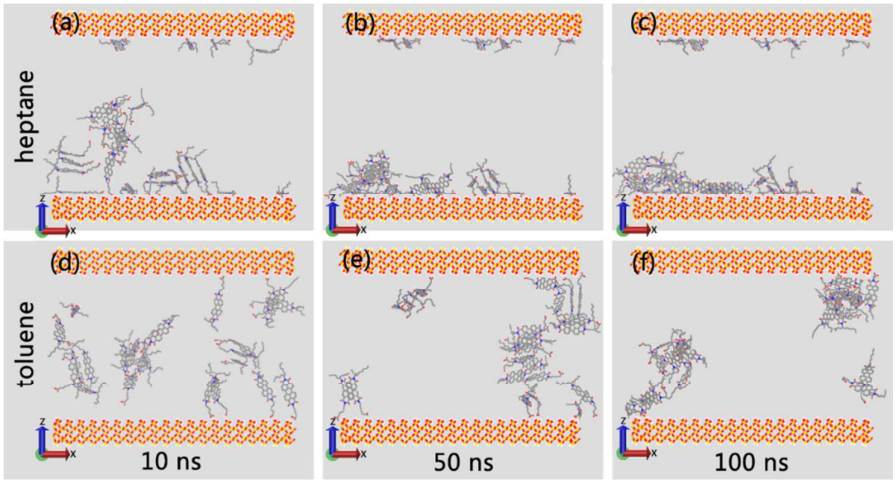

Figure S6. Snapshots of model asphaltenes in heptane-silica and toluene-silica systems

Snapshots of C5Pe adsorption on silica in heptane and toluene at 10 ns, 50 ns, 100 ns, respectively. A strong and stable adsorption was reached in heptane, while desorption from silica surface occurred in toluene, leading to a decrease in the number of hydrogen bonds.

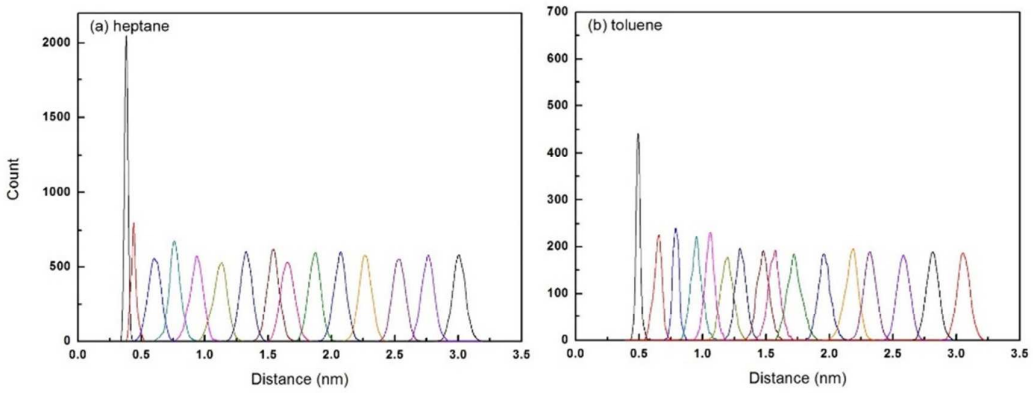

Figure S7. The histograms of the configurations with the umbrella sampling windows of (a) heptane, (b) toluene

It is evident that the histogram shows reasonable overlap between COM of C5Pe and silica surface in Z direction between the windows from about 0.3-3.2 nm.

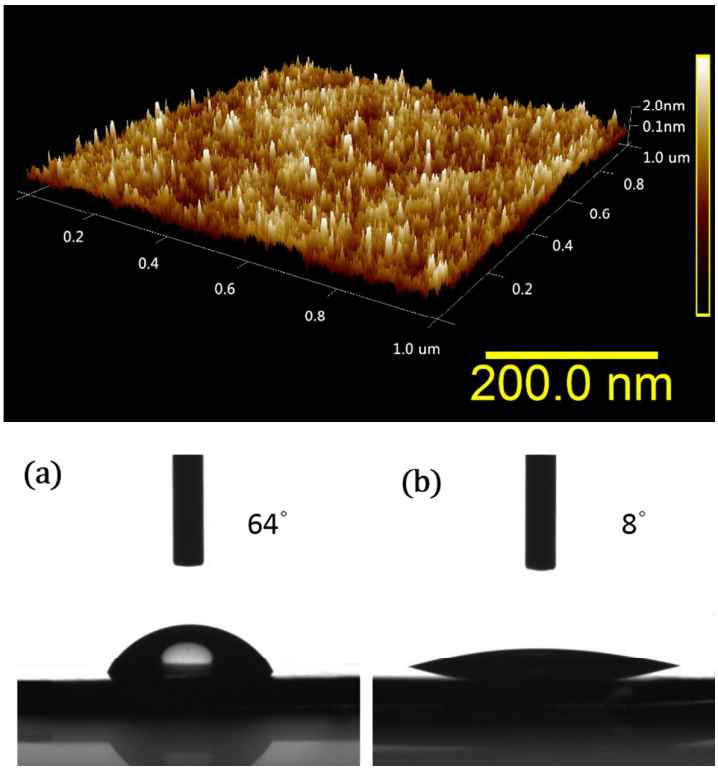

Figure S8. AFM image of silica wafer treated by NaOH (pH=11) and contact angle (a) before and (b) after treated with NaOH.

The silica wafer was flat with an average roughness of 0.416nm. Contact angle of silica before being treated with NaOH is 64˚, and decreased to 8˚ after treatment with NaOH, which indicated the extremely hydrophilic nature of silica surface.