Supporting Information

Microscopic mechanisms for the dynamic wetting of heavy oil mixture on rough silica surface

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Silica surface model construction:

To construct the rough surfaces with narrow grooves (i.e., D_1H_1, D_1H_2, D_1H_3, D_1H_4, D_1H_5), the unit cell (Fig. S1a) was repeated by 5-, 5-, and 6-time in the x-, y- and z-direction, respectively, to form a small supercell (Fig. S1b). The undesired atoms were manually removed and the exposed oxygen atoms were protonated (Fig. S1c). The resulted structure was then replicated by 5-time in both x and y direction to form the final configuration (Fig. S1d).

To construct the rough surfaces with wider grooves (i.e., D_2H_1, D_2H_2, D_2H_3, D_2H_4, D_2H_5), the unit cell was repeated by 6-, 6-, and 6-time in the x-, y- and z-direction, respectively, to form a small supercell. The rest procedures were similar as that performed for constructing surfaces with narrow grooves.

To construct the rough surfaces with widest grooves (i.e., D_3H_1, D_3H_2, D_3H_3, D_3H_4, D_3H_5), similar procedures were performed as above except that the replication time of the initial unit cell was 7-, 7-, and 6-time in the x-, y- and z-direction, respectively.

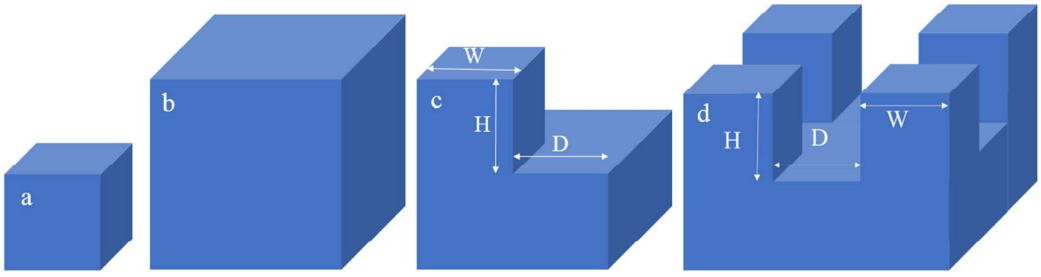

Fig. S1 Schematic for the rough surface model construction

Table S1 Box size for the 15 simulations

	D_1	D_2	D_3
H_1	12.3nm×10.8nm×10.0nm	14.7nm×13.5nm×10.0nm	17.2nm×16.2nm×10.0nm
H_2	12.3nm×10.8nm×10.0nm	14.7nm×13.5nm×10.0nm	17.2nm×16.2nm×10.0nm
H_3	12.3nm×10.8nm×10.0nm	14.7nm×13.5nm×10.0nm	17.2nm×16.2nm×10.0nm
H_4	12.3nm×10.8nm×10.0nm	14.7nm×13.5nm×10.0nm	17.2nm×16.2nm×10.0nm
H_5	12.3nm×10.8nm×10.0nm	14.7nm×13.5nm×10.0nm	17.2nm×16.2nm×10.0nm

Table S2 Force field parameters used in this study

Species	Atom type	Non-bond parameters		
		charge	Sigma (nm)	Epsilon(kJ/mol)
Silica	Tetrahedral silicon	2.100	0.37064	7.70065×10^{-6}
	Bridging oxygen	-1.050	0.35532	0.6495
	Hydroxyl oxygen	-0.950	0.35532	0.6495
	Hydroxyl hydrogen	0.425	0	0
Oil	Carbon in CH_3 group	-0.270	0.36527	0.32635
	Carbon in CH_2 group	-0.180	0.35814	0.23430
	Carbon in CH_1 group	-0.090	0.35636	0.13389
	Hydrogen in CH_3 group	0.090	0.23876	0.10042
	Hydrogen in CH_2 group	0.090	0.23876	0.14644
	Carbon in benzene ring	-0.115	0.34677	0.11069
	Hydrogen in benzene ring	0.115	0.29027	0.28067
	Bridging Sulphur with	-0.220	0.34745	1.88280
	Bridging Nitrogen with	-0.300	0.32963	0.83680
	Bridging Oxygen with	-0.340	0.29399	0.41840
	Carbon in carbon-oxygen	0.400	0.35635	0.09355
	Oxygen in carbon-oxygen	-0.480	0.30290	0.20920

Bonds for clayFF force field		b_0 (nm)	k_b(kJ/mol/nm^2)
Species i	Species j		
Hydroxyl	Hydroxyl hydrogen	0.1	463532.808

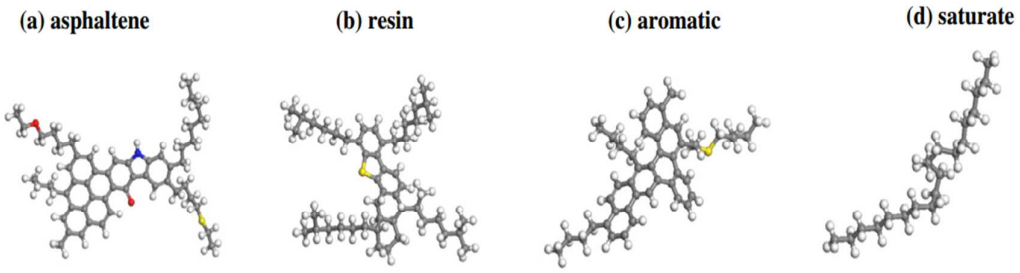

Fig. S2 Three-dimensional structure of the model oil fractions

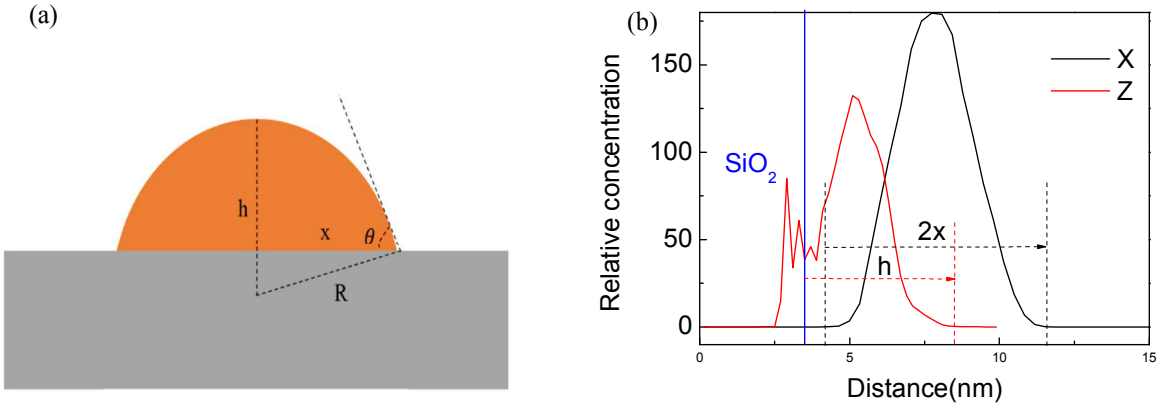

Fig. S3 (a) Schematic for the contact angle calculation. (b) An example of the concentration profiles of oil molecules on the silica surfaces along the X, Z direction.

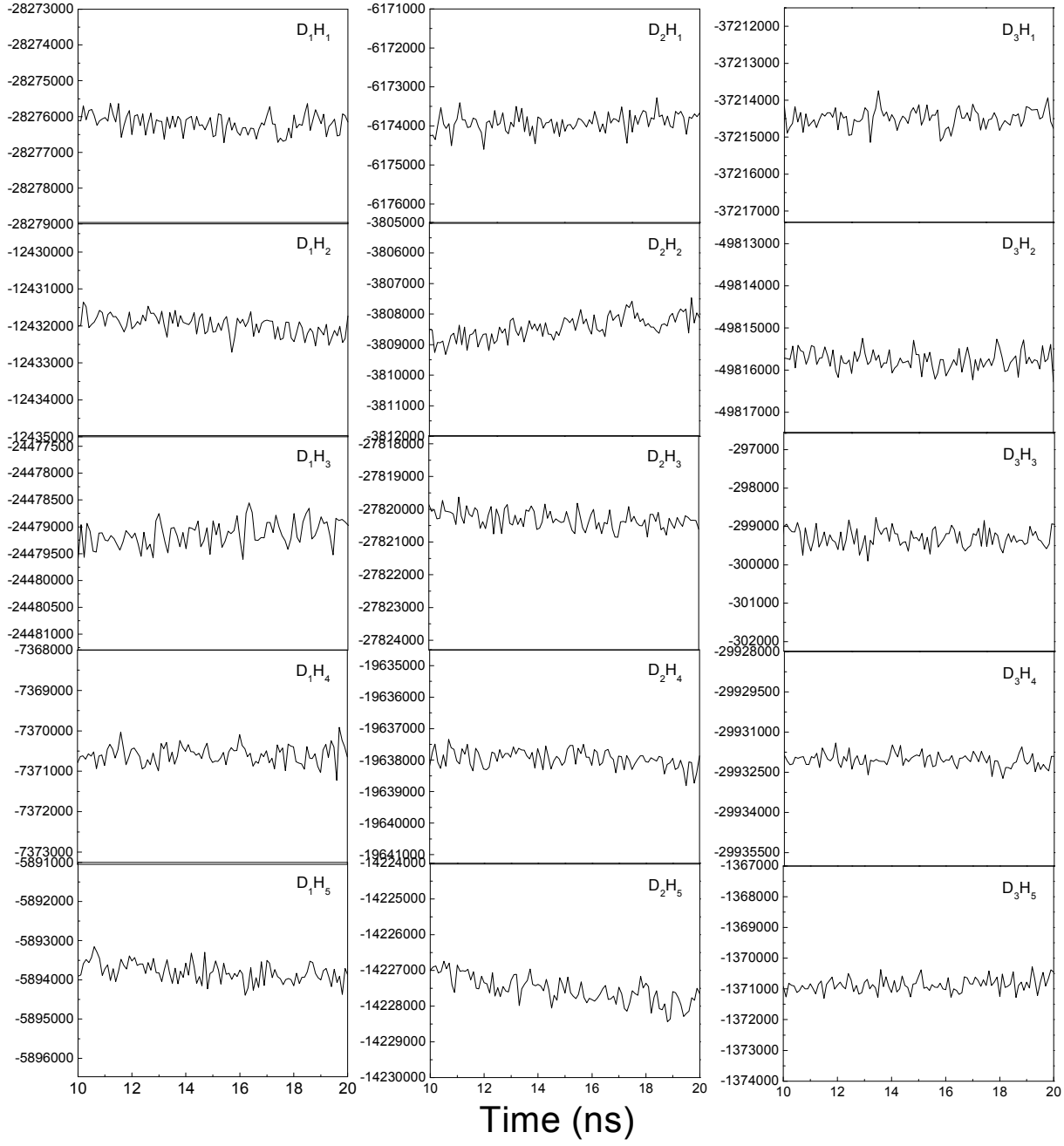

Fig. S4 Energy profile during molecular dynamic simulations

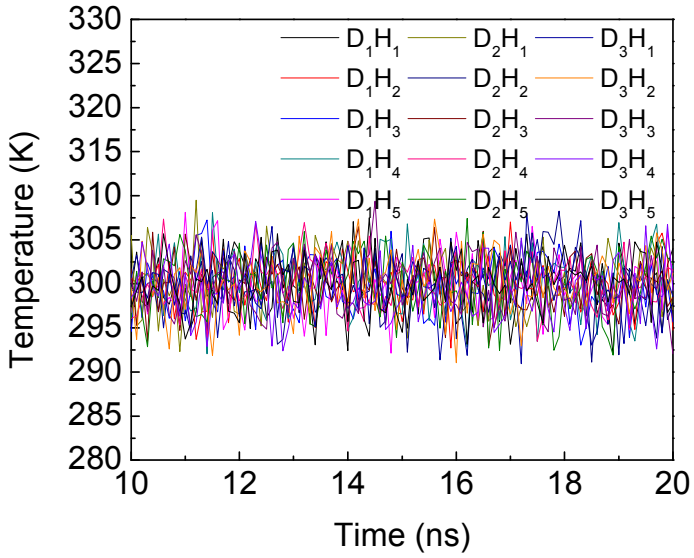

Fig. S5 Temperature profile during molecular dynamic simulations

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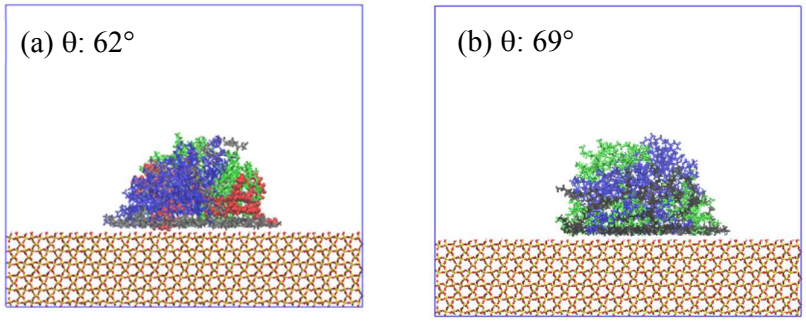

Fig. S6 Configuration of the oil droplet (a) with and (b) without asphaltenes on smooth silica surface at the end of simulation

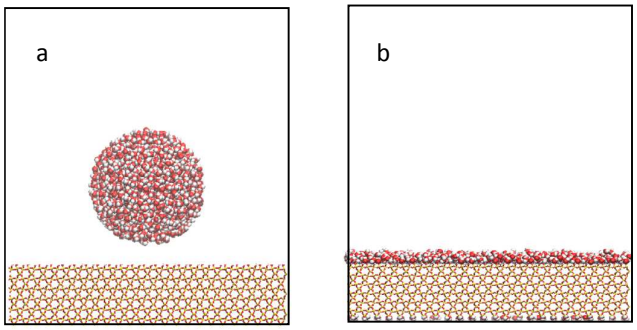

Fig. S7 (a) Initial and (b) final distribution of water on smooth silica surfaces

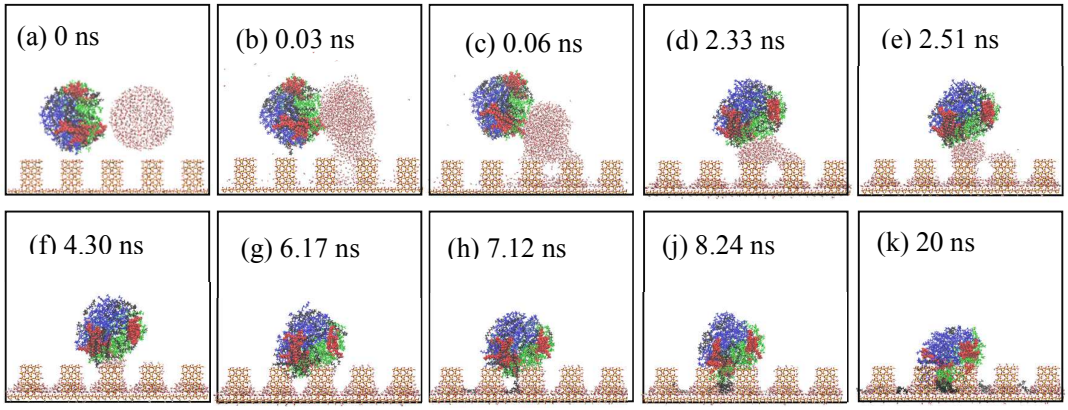

Fig. S8 Snapshots of the adsorption process of oil droplet in the presence of water droplet on D_2H_5 surface (z-axis dimension = 100 nm)

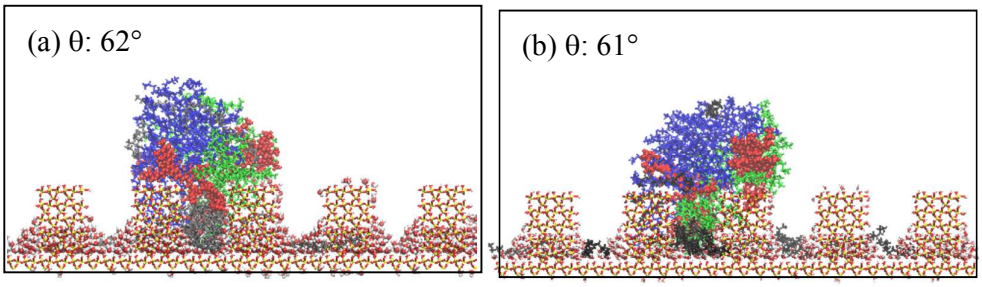

Fig. S9 Oil configuration on the D_2H_5 surface at the end of simulation (a) z-axis dimension = 27 nm; (b) z-axis dimension = 100 nm

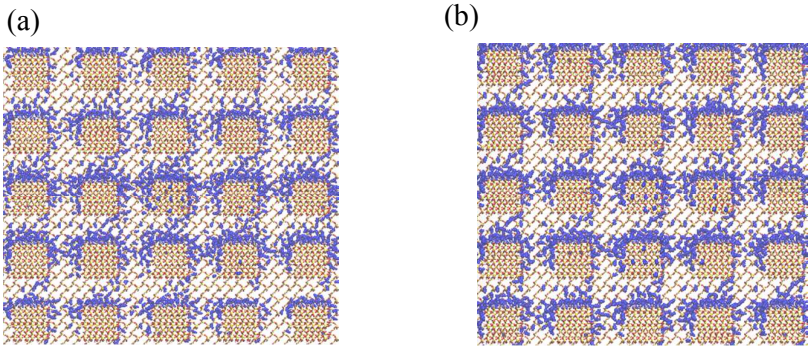

Fig. S10 Water distribution on the D_2H_5 surface at the end of simulation (a) z-axis dimension = 27 nm; (b) z-axis dimension = 100 nm

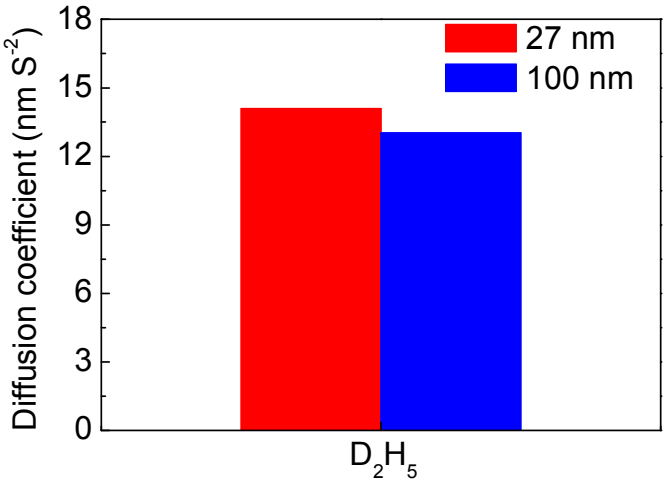

Fig. S11 Diffusion coefficient of saturates on D_2H_5 surface at the end of simulation (red) z-axis dimension = 27 nm; (blue) z-axis dimension = 100 nm

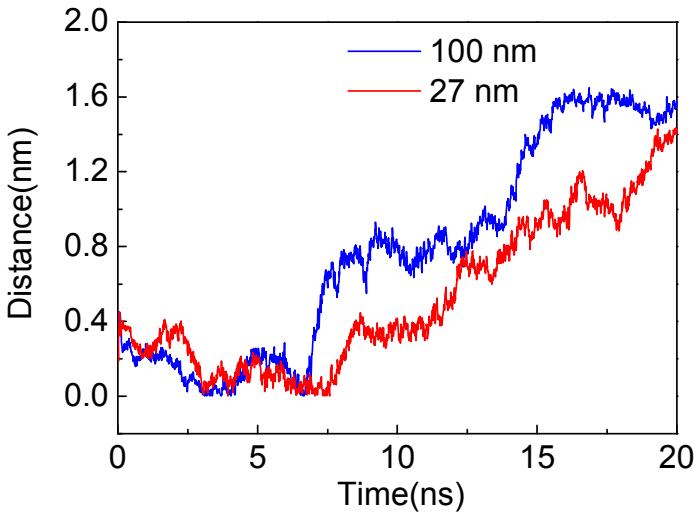

Fig. S12 Distance between saturates and other oil components of oil droplet (red) z-axis dimension = 27 nm; (blue) z-axis dimension = 100 nm