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Supporting Information

Self-assembly of cationic surfactant n-hexadecyltrimethyl-ammonium chloride in methyltrimethoxysilaneaqueous solution: Classical and reactive molecular dy-namics simulations

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In this supporting information, the followings are available.

[Tables]

Table S1 lists equilibrium conditions of the MD systems.

Table S2-S5 list all the parameters of OPLS force field used for the MD simulations.

[Figures]

Figure S1 draws molecular structures of MTMS and CTAC.

Figure S2 draws equilibrated CTAC system. Details of simulation procedure is also noted.

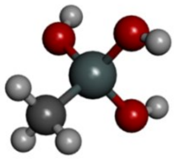
Figure S3 draws radial distribution functions relating to MTMS distance for C-0.4%, C-0.8%, and C-1.5% models.

Figure S4 shows temperature dependence of number of CTAC aggregates and the maximum CTAC cluster size measured by heating MD simulations.

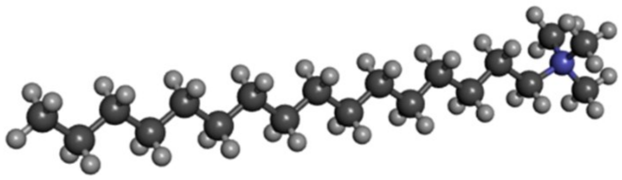
Figure S5 shows time-variation of surface/volume ration of MTMS to confirm equilibration of each system.

Table S1. Simulation time to obtain equilibrium models, and simulation cell size and density at the equilibrium conditions.

ID	Density [g/ cm^3]	MD cel size [▯]
C-0.4%	1.069	106.1
C-0.8%	1.062	108.2
C-1.5%	1.056	111.8
M-11%	1.093	98.0
M-27%	1.123	86.5
M-40%	1.129	83.3
R-3	1.080	97.2
R-6	1.076	96.5
R-10	1.059	95.8



(a) Hydrolyzed methyltrimethoxysilane



(b) n-hexadecyltrimethylammonium ion

Figure S1. Molecular structures of (a) hydrolyzed methyltrimethoxysiloane (MTMS) and (b) a cation of n-hexadecyltrimethylammonium chloride (CTAC).

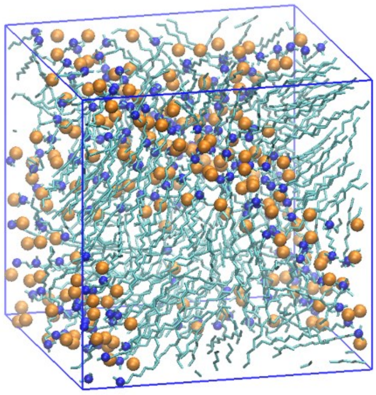


Figure S2. Final snapshot of MD simulation on n-hexadecyltrimethylammonium chloride (CTAC) at 298K. Blue and orange spheres are nitrogen atoms and chlorine ions. Lines in light blue are hydrocarbon chains. Hydrogen is invisible. First, 200 molecules were randomly put in a cubic box and the system was optimized at 298 K and 0.1 MPa by 5 ns of NPT-MD simulation. After that, the system was heated up to 1000 K and then it was cooled down to 298K with cooling rate of 1 K/ps using the quasi-slow-quenching method [see ref. 66 in the manuscript]. After additional NPT-MD run of 10 ns at 298K and 0.1 MPa, the final density is 0.905 g/cm^3 in a cubic box with side length of 4.9 nm.

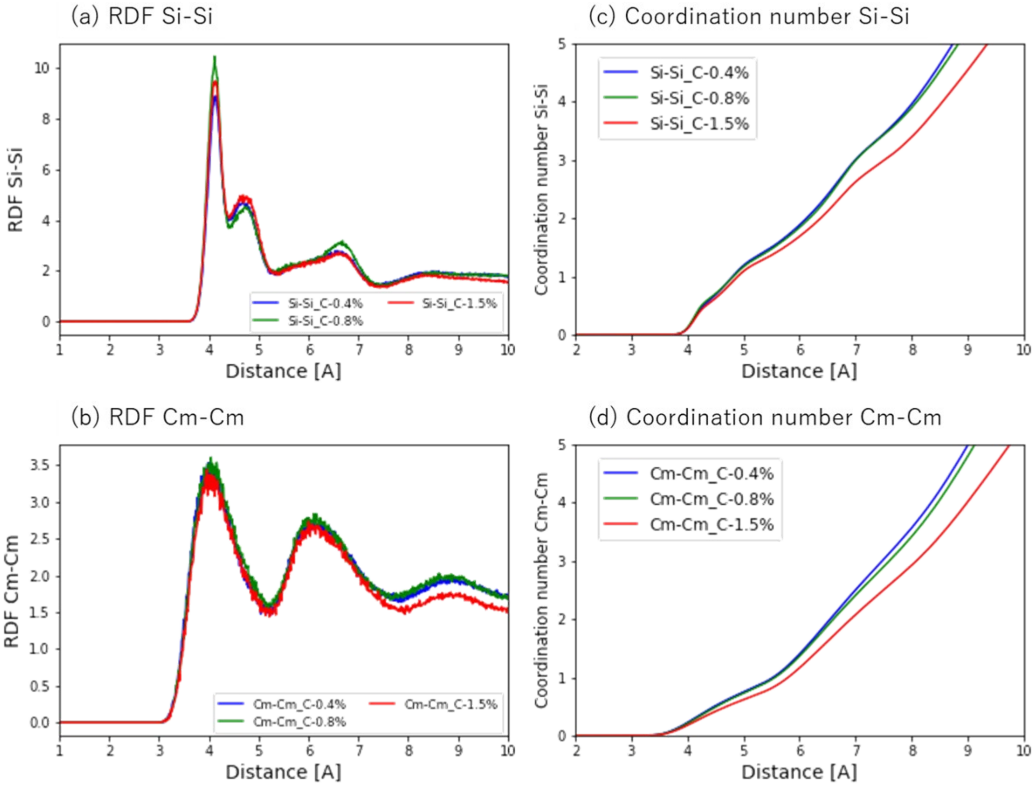


Figure S3. RDFs for (a) silicon (Si) and (b) methyl carbon (Cm) in MTMS monomers. Coordination number of (c) Si and (d) Cm at around each atom.

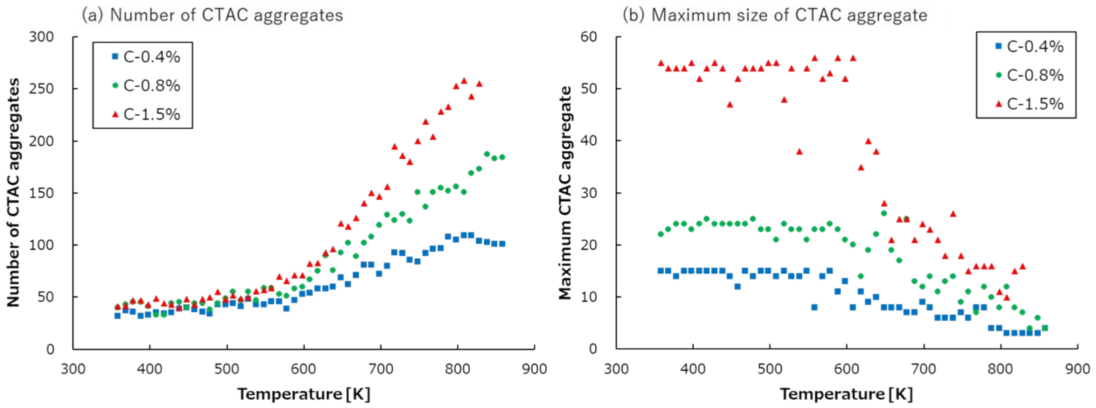


Figure S4. Temperature dependence of (a) number of CTAC aggregates and (b) maximum size of CTAC aggregate. Note that isolated CTAC molecules are also taken into account in the number of aggregates.

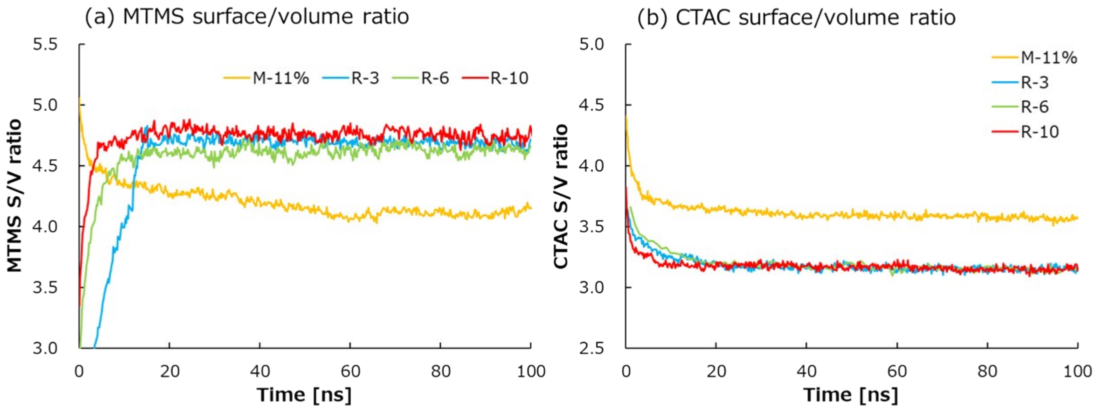


Figure S5. Time-variations of surface-volume ratio of (a) MTMS aggregates and (b) CTAC surfactant with MTMS-derived oligomers.

OPLS force field parameters.

Table S2. Parameters for Lennard-Jones interaction.

Atom kind		σ [Å]	ε [kcal/mol]	Ref.
C	alkane	3.5	0.066	[1]
H	C-H	2.5	0.03	[1]
Si	silicon	3.721	0.2381	[2]
O_{Si}	Si-O-H	2.787	0.1997	[2]
H_{Si}	Si-O-H	0	0	[1]
H_w	water	0.9	0.01	[6]
O_w	water	3.5532	0.1848	[6]
N	amine	3.3	0.17	[3]
Cl	chlorine	3.4	0.3	[5]

Table S3. Parameters for bond stretching.

Bond kind	K_B [kcal/mol]	r_0 [Å]	Ref.
C-C	268	1.529	[1]
C-H	340	1.09	[1]
Si-O_{Si}	350	1.653	[2]
O_{Si}-H_{Si}	533	0.945	[4]
C-Si	190	1.881	[2]
O_w-H_w	250	1.000	[6]
C-N	382	1.448	[3]

Table S4. Parameters for angle bending.

Angle kind	K_Θ [kcal/mol]	Θ [rad]	Ref.

C-C-C	58.35	112.7	[1]
H-C-H	33	107.8	[3]
H-C-C	37.5	110.7	[3]
C-C-N	56.2	109.47	[3]
C-N-C	51.8	107.2	[3]
H-C-N	35	109.5	[3]
Si-O_{Si}-Si	14.149	144	[2]
Si-O_{Si}-his	23.7764	122.888	[6]
O_{Si}-Si-O_{Si}	94.551	109.5	[2]
Si-C-H	28.77	111.09	[2]
C-Si-O_{Si}	50	109.5	[2]
H_w-Ow-H_w	60	109.47	[4]

Torsion kind	V_1	V_2	V_3	V_4	Ref.
H-C-C-H	0	0	0.318	0	[1]
H-C-C-C	0	0	0.366	0	[1]
C-C-C-C	1.74	-0.157	0.279	0	[3]
C-C-C-N	2.392	-0.674	0.55	0	[3]
H-C-C-N	-1.013	-0.709	0.473	0	[3]
C-C-N-C	0.416	-0.128	0.695	0	[3]
H-C-N-C	0	0	0.56	0	[3]
O_{Si}-Si-O_{Si}-H_{Si}	-2.49763	2.21179	-0.23461	0.15267	[This work]
O_{Si}-Si-C-H	-1.8378	4.2947	0.1283	0.2545	[This work]

Table S5. Parameters for dihedral angle.

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