Supplementary Material (ESI) for *PCCP*

Supplementary Information:

Molecular Simulation of Swelling and Interlayer Structure for Organoclay in Supercritical CO_2

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Table S1 Atomic coordinates and effective charges in the unit cell of montmorillonite

No.	Atom	x(Å)	y(Å)	z(Å)	q_i (e)
1	AL1	–0.593	–2.708	0.000	1.575
2	H1	3.809	0.342	1.434	0.425
3	O1	2.927	0.342	1.060	-1.0808
4	H2	0.285	0.342	–1.434	0.425
5	O2	1.167	0.342	–1.060	-1.0808
6	SI1	0.287	1.862	2.730	2.1
7	O3	0.287	1.862	1.060	-1.05
8	SI2	0.287	–1.178	2.730	2.1
9	O4	0.287	–1.178	1.060	-1.1808
10	SI3	2.927	–2.708	2.730	2.1
11	O5	2.927	–4.228	3.280	-1.05
12	O6	2.927	–2.708	1.060	-1.1808
13	O7	1.607	–1.948	3.280	-1.05
14	O8	4.247	–1.948	3.280	-1.05
15	O9	0.287	0.342	3.280	-1.05
16	SI4	2.927	3.392	2.730	2.1
17	O10	2.927	3.392	1.060	-1.05
18	O11	1.607	2.622	3.280	-1.05
19	O12	4.247	2.622	3.280	-1.05
20	SI5	3.807	–1.178	–2.730	2.1
21	O13	3.807	–1.178	–1.060	-1.1808
22	O14	1.167	–2.708	–1.060	-1.1808
23	O15	–0.153	–1.938	–3.280	-1.05
24	SI6	1.167	–2.708	–2.730	2.1
25	O16	2.487	–1.938	–3.280	-1.05
26	SI7	3.807	1.862	–2.730	2.1
27	O17	3.807	1.807	–1.060	-1.05
28	O18	3.807	0.342	–3.280	-1.05
29	SI8	1.167	3.392	–2.730	2.1
30	O19	1.167	3.392	–1.060	-1.05
31	O20	–0.153	2.632	–3.280	-1.05
32	O21	1.167	4.912	–3.280	-1.05
33	O22	2.487	2.632	–3.280	-1.05
34	AL2	2.047	–1.178	0.000	1.36
35	AL3	2.047	1.862	0.000	1.575
36	H3	2.926	4.912	–1.434	0.425
37	O23	3.807	4.912	–1.060	–0.95
38	AL4	4.687	3.392	0.000	1.575
39	H4	1.169	–4.223	1.434	0.425
40	O24	0.287	–4.228	1.060	–0.95

Table S2. Potential parameters of ODTA surfactant cation

	CH_2	CH_2 [a]	CH_3	CH_3 [b]	N
Charge (e)	0	+0.407	0.0	+0.407	-0.628
ε (kcal/mol)	0.1984	0.1984	0.2500	0.2500	0.0774
σ (Å)	3.6238	3.6238	3.6993	3.6993	3.2625

Note: Charges are derived from Ref 35.
a: $–CH_2$ binded with N; b: $–CH_3$ at the end of alkylammonium chain.

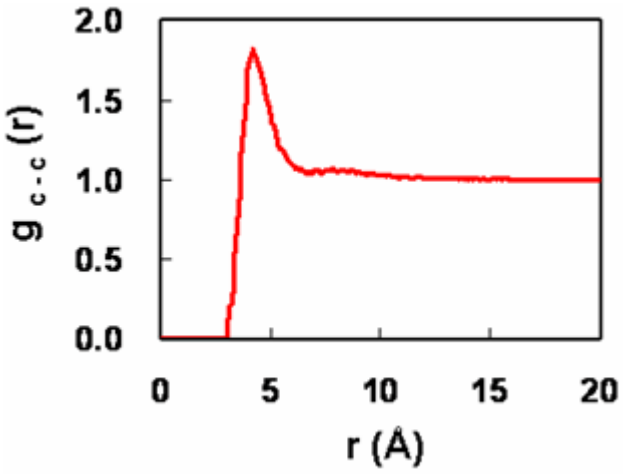

Fig. S1 The radial distribution function (RDF) of CO_2 fluid in the simulation cell2. The position of the peak in the RDF is around 4.2 Å, which is identical with the reference result.

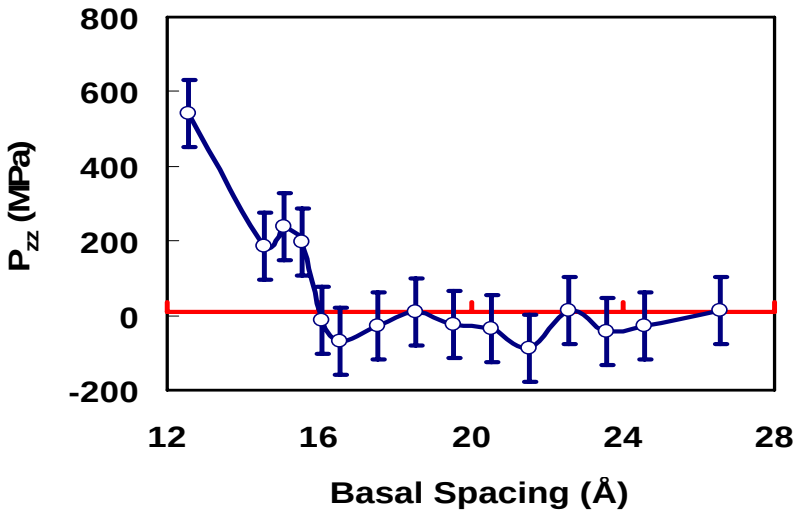

Fig. S2 Normal component of the pressure tensor as a function of the basal spacing for natural Na-montmorillonites in $scCO_2$. The horizontal solid line corresponds to the bulk pressure (8.4MPa)

Table S3 vdW interaction energies (*E*) in several stable spacings

Basal spacing (Å)	E (clay-surfactant) kcal/mol	E(Surfactant-surfactant) kcal/mol
13.56 (vacuum)	493.57	-228.38
18.46	-315.72	-221.63
21.46	-591.27	-203.69