Supporting Information

Theoretical Study on the Selective Adsorption of SF_6/N_2 by Covalent Triazine Frameworks

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Table S1 LJ parameters and atomic partial charges for SF_6, N_2.

Atom type	ε (K)	σ	Q(e)
SF_6(S)	165.14	3.228	0.66
SF_6(F)	27.02	2.947	-0.11
N_2(N)	38.298	3.306	-0.405
N_2(N_COM)	0	0	0.81

Table S2 The summary of the crystal stacking patterns of CTF-FUM, CTF-1, CTF-2 and FCTF-1, including AA stacking, stepped stacking and AB stacking.

l **: the distance of layer spacing**
d **: the distance of interlayer slipping**

Name	AA stacking	Staircase-like stacking	AB stacking
CTF-FUM	Side view / Top view *l* = 3.615 *d* = 0.00	*l* = 3.778 *d* = 1.892	*l* = 3.176 *d* = 6.413
CTF-1	*l* = 3.640 *d* = 0.00	*l* = 3.780 *d* = 1.960	*l* = 3.115 *d* = 8.425
CTF-2	*l* = 3.652 *d* = 0.00	*l* = 4.101 *d* = 3.331	*l* = 3.048 *d* = 10.632
FCTF-1	*l* = 3.505 *d* = 0.00	*l* = 3.805 *d* = 1.892	*l* = 3.182 *d* = 8.555

Table S3 Performance for SF_6 adsorption and separation of CTFs.

Name	$N_{SF6,298K}$ (mmol g^{-1}) 0.1bar	$N_{SF6,298K}$ (mmol g^{-1}) 1bar	$N_{SF6,0.1bar}$ / $N_{SF6,1bar}$	$Q_{st,100KPa}$ (KJ mol^{-1}) SF_6	$Q_{st,100KPa}$ (KJ mol^{-1}) N_2	K_H (mmol g^{-1} bar^{-1})	$S_{SF6/N2,298K}$ 0.1bar	$S_{SF6/N2,298K}$ 1bar
FUM1AA	2.47	2.71	0.91	41.10	13.89	428.21	1283.03	948.92
CTF-1AA	0.87	1.79	0.48	31.01	10.84	13.81	95.30	85.79
CTF-2AA	0.19	2.50	0.08	27.48	9.17	1.87	18.56	19.35
FCTF-1AA	0.81	1.04	0.78	33.63	12.13	29.58	285.74	235.00
FUM1SS	2.90	3.14	0.92	52.00	16.54	1828.58	3178.19	2321.74
CTF-1SS	1.50	1.87	0.80	36.31	12.49	74.48	388.02	268.66
CTF-2SS	0.86	2.43	0.36	33.34	10.75	9.25	72.37	81.46
FCTF-1SS	1.08	1.22	0.89	43.37	14.23	197.68	1521.02	939.60
FUM1AB	-	-	-	-	-	-	-	-
CTF-1AB	-	-	-	-	-	-	-	-
CTF-2AB	-	-	-	-	-	-	-	-
FCTF-1AB	-	-	-	-	-	-	-	-

TableS4 The calculated binding energy (E_b) and total charge transfer (ΔQ_{SF6}) during the adsorption of SF_6 on CTFs.

CTFs	E_b (kJ/mol)	ΔQ_{SF6} (e)
CTF-FUMAA	35.0521	0.0256
CTF-FUMSS	41.7822	0.0416
CTF-1AA	24.3697	0.0155
CTF-1SS	26.3296	0.0126
CTF-2AA	23.5886	0.0207
CTF-2SS	28.2184	0.0244
FCTF-1AA	23.5770	-0.0005
FCTF-1SS	29.7552	-0.0008

Table S5 The calculated binding energy (E_b) and total charge transfer (ΔQ_{N2}) during the adsorption of N_2 on different CTFs.

CTFs	E_b (kJ/mol)	ΔQ_{N2} (e)
CTF-FUMAA	14.8417	0.0082
CTF-FUMSS	16.5362	0.0065
CTF-1AA	14.0720	0.0052
CTF-1SS	14.0659	0.0038
CTF-2AA	15.3158	0.0093
CTF-2SS	10.5713	0.0043
FCTF-1AA	10.7523	-0.0057
FCTF-1SS	12.9704	-0.0066

Table S6 The SF_6 separation performance of advanced porous material (APM) adsorbents at 298 K and 0.1bar or1 bar.

APM	Adsorbents	SF_6 uptake at 0.1 bar (mmol/g)	IAST selectivity (10/90)at 1bar 298K	Refs.
1	Zeolite-13X	0.99	56.5	1
2	Activated carbon	1.01	40.10	2
3	PC-CaCit	1.05	30	3
4	RCOF-1	1.5	83	4
5	3D-TMTAPB-COF	2.72	335	5
6	MIL-53(Al)	2.4	426.5	6
7	HBU-21	0.35	184	7
8	UiO-66	0.82	127.8	8
9	$Ni(pba)_2$	1.69	200.6	9
10	$Ni(ina)_2$	2.39	375.1	9
11	$Ni(3\text{-}mpba)_2$	1.79	221	10
12	$Zn(TMBDC)(DABCO)_{0.5}$	2.48	239	11
13	CAU-10-Py	1.13	203.6	12
14	$Co_3(HCOO)_6$	1.63	120	13
15	Cu-MOF-NH_2	3.39	266.2	14
16	TKL-107	1.26	180	15
17	$Ni(ndc)(ted)_{0.5}$	2.78	750	16
18	$Ni(adc)(dabco)_{0.5}$	2.23	919.4	17
19	BUT-53	2.82	2485	18
20	Co(BPZ)	2.47	748	19
This work	CTF-FUMAA	2.47	948.92	-
	CTF-FUMSS	2.90	2321.74	-
	CTF-1AA	0.87	85.79	-
	CTF-1SS	1.50	268.66	-
	CTF-2AA	0.19	19.35	-
	CTF-2SS	0.86	81.46	-
	FCTF-1AA	0.81	235.00	-
	FCTF-1SS	1.08	939.60	-

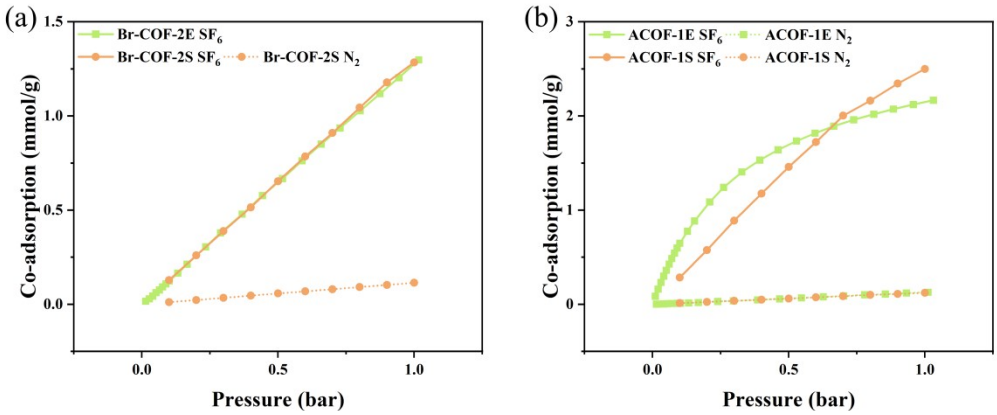

Fig. S1 Comparison of the adsorption isotherms between the simulation and the experiment for (a) BrCOF-2, (b) ACOF-1.

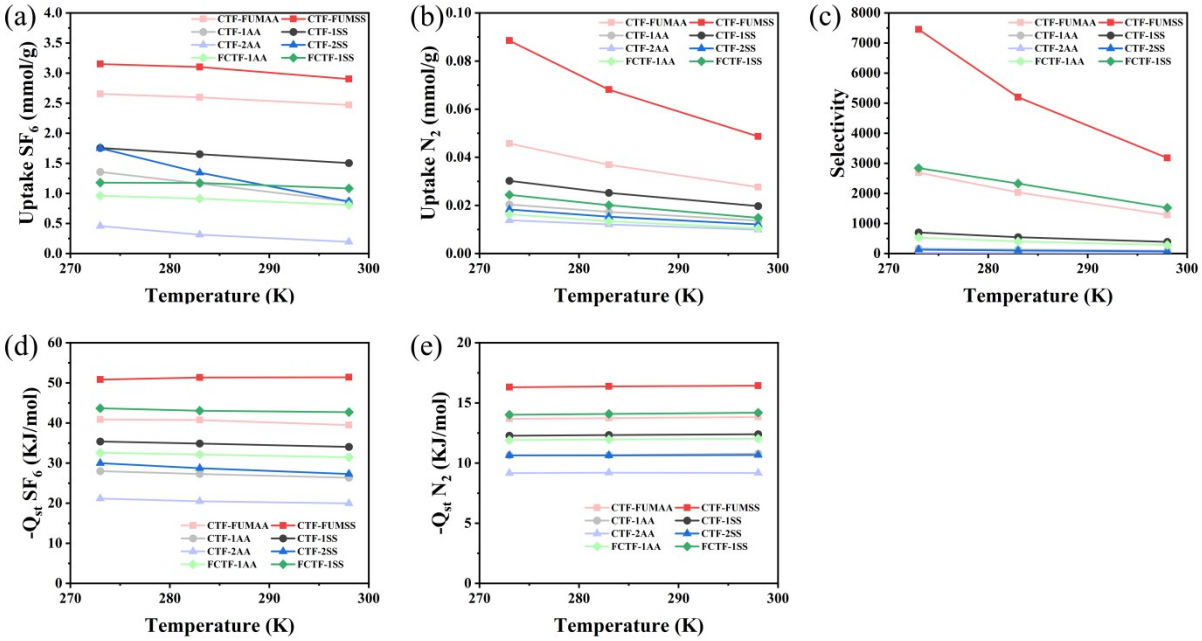

Fig. S2 Separation performance of SF_6 and N_2 for CTFs at different temperatures (273, 283 and 298 K) at 0.1 bar.

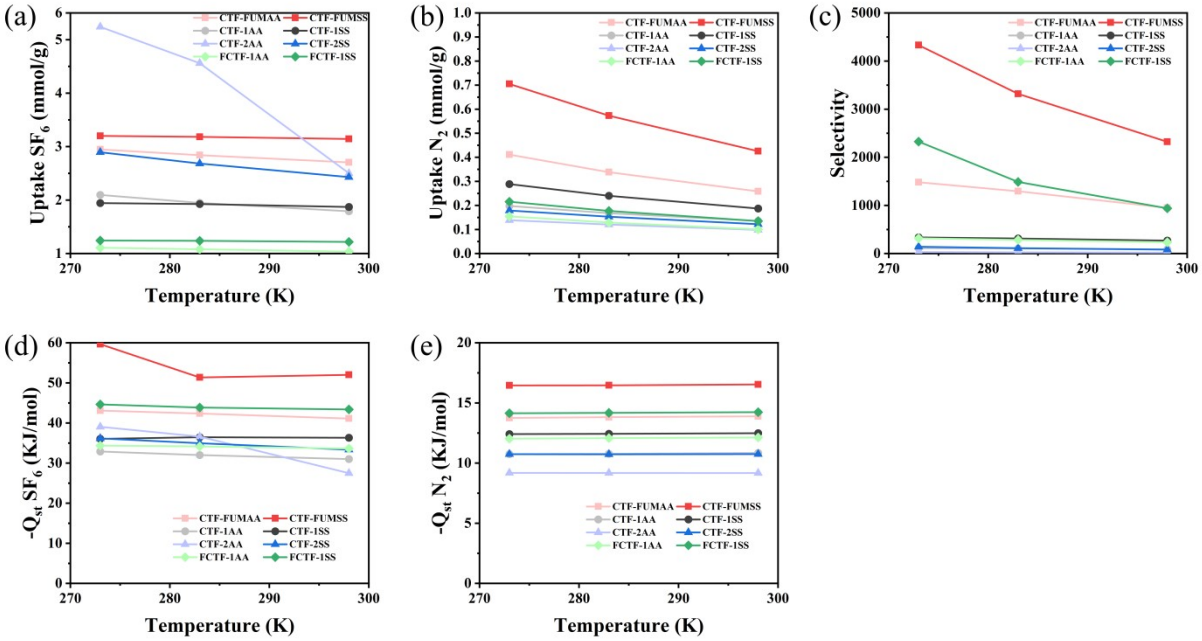

Fig. S3 Separation performance of SF_6 and N_2 for CTFs at different temperatures (273, 283 and 298 K) at 1 bar.

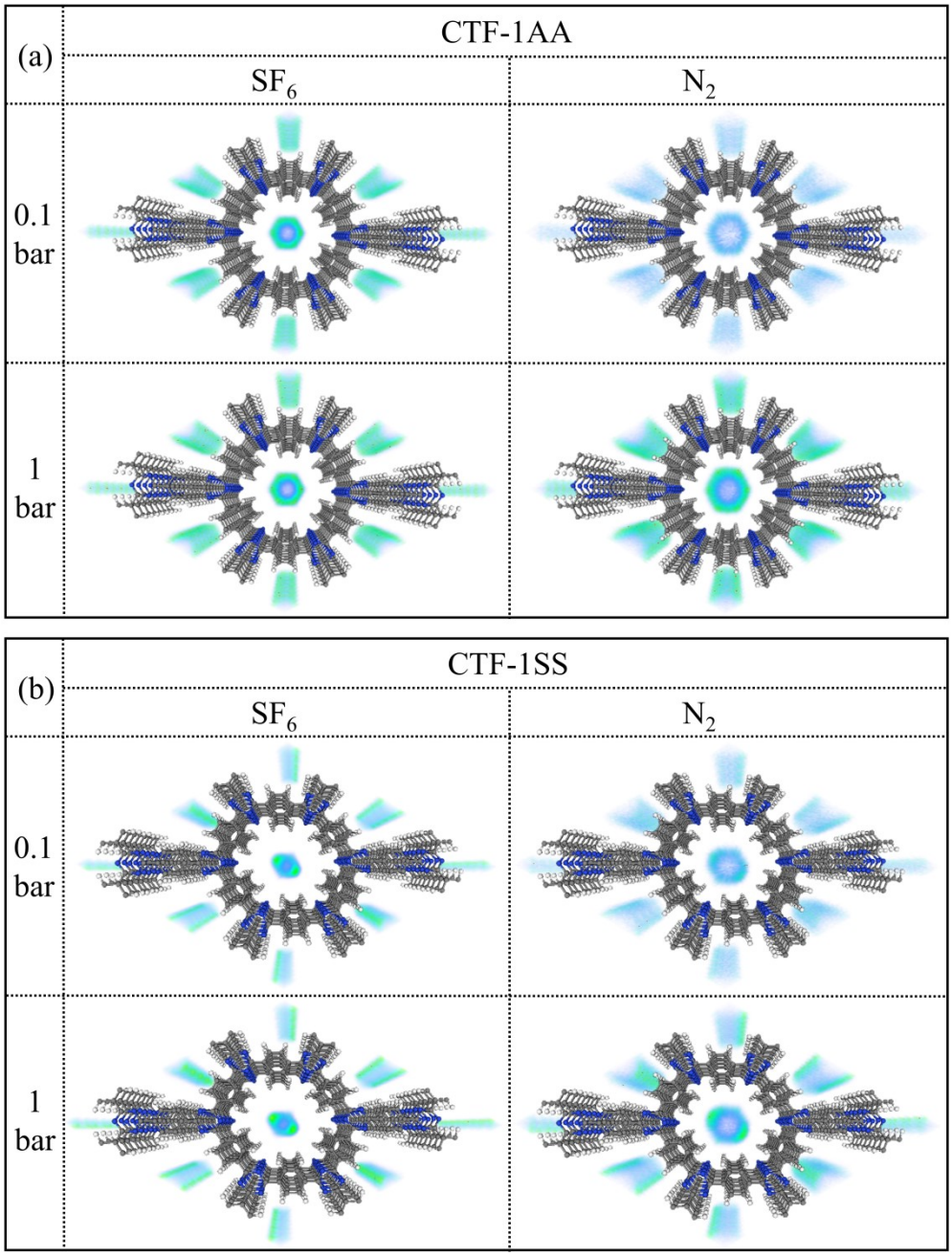

Fig. S4 COM probability distribution of (a) CTF-1AA and (b) CTF-1SS with an SF_6/N_2 ratio of 1:9 .

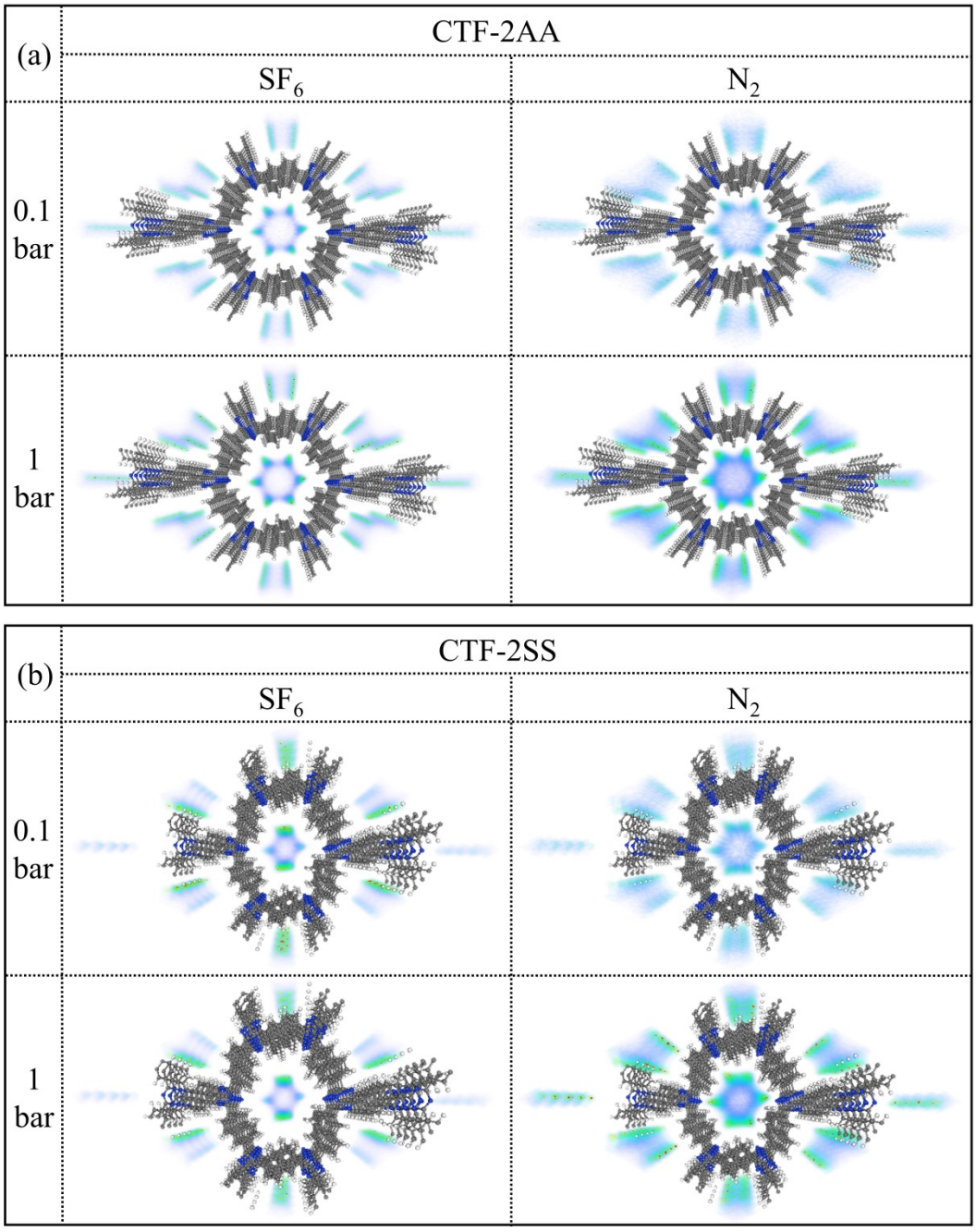

Fig. S5 COM probability distribution of (a) CTF-2AA and (b) CTF-2SS with an SF_6/N_2 ratio of 1:9 .

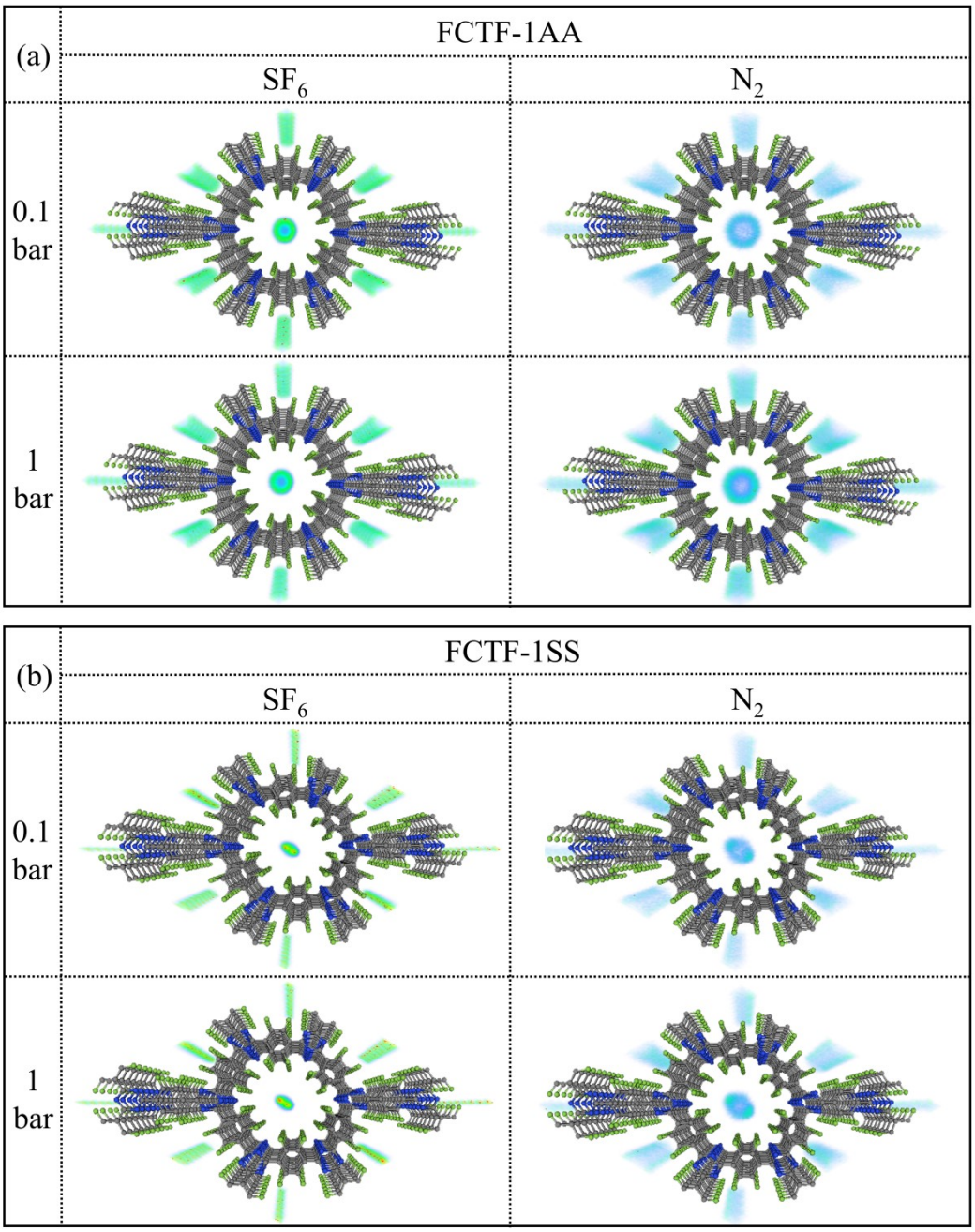

Fig. S6 COM probability distribution of (a) FCTF-1AA and (b) FCTF-1SS with an SF_6/N_2 ratio of 1:9 .

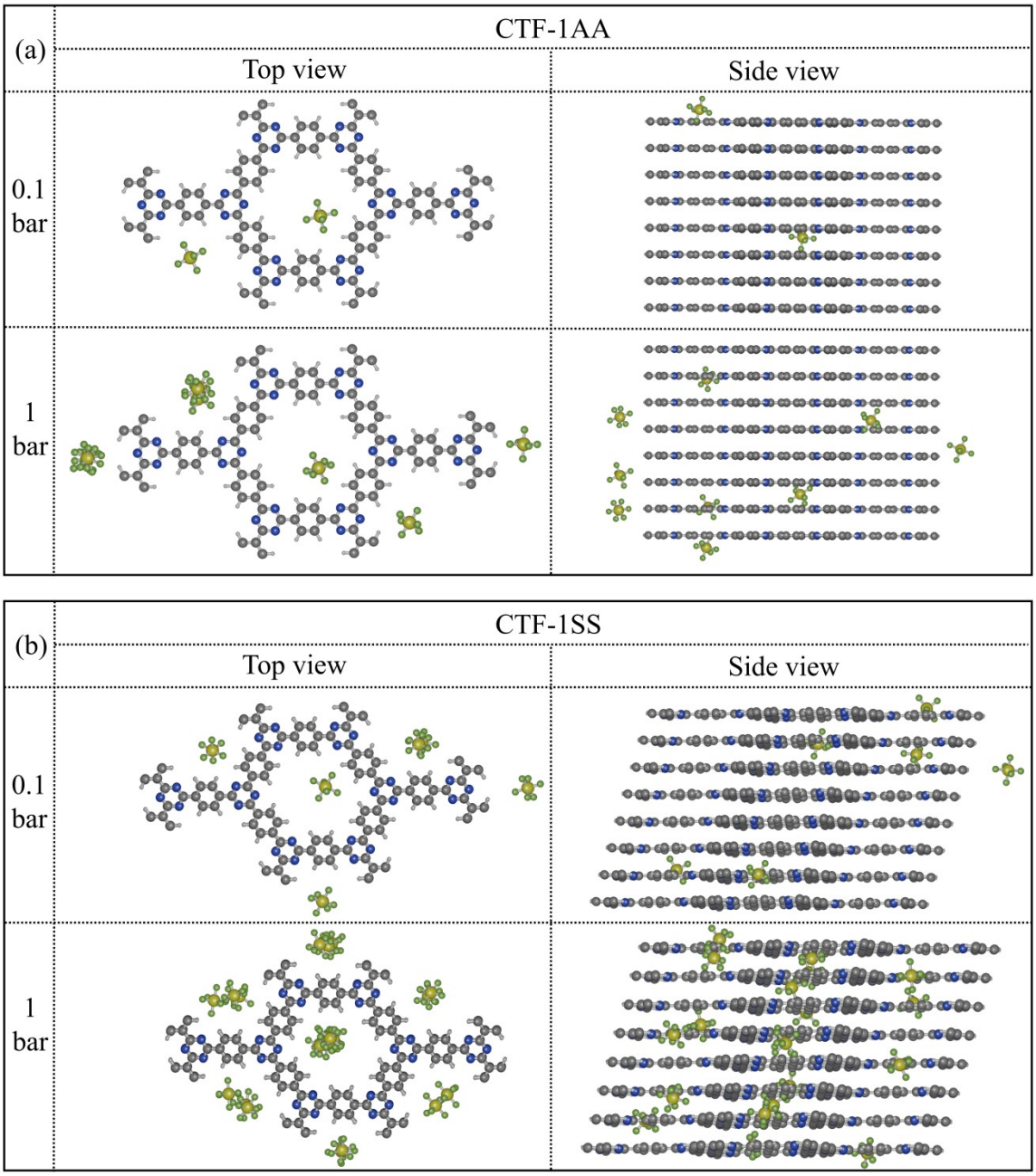

Fig. S7 Snapshots of (a) CTF-1AA and (b) CTF-1SS with an SF_6/N_2 ratio of 1:9.

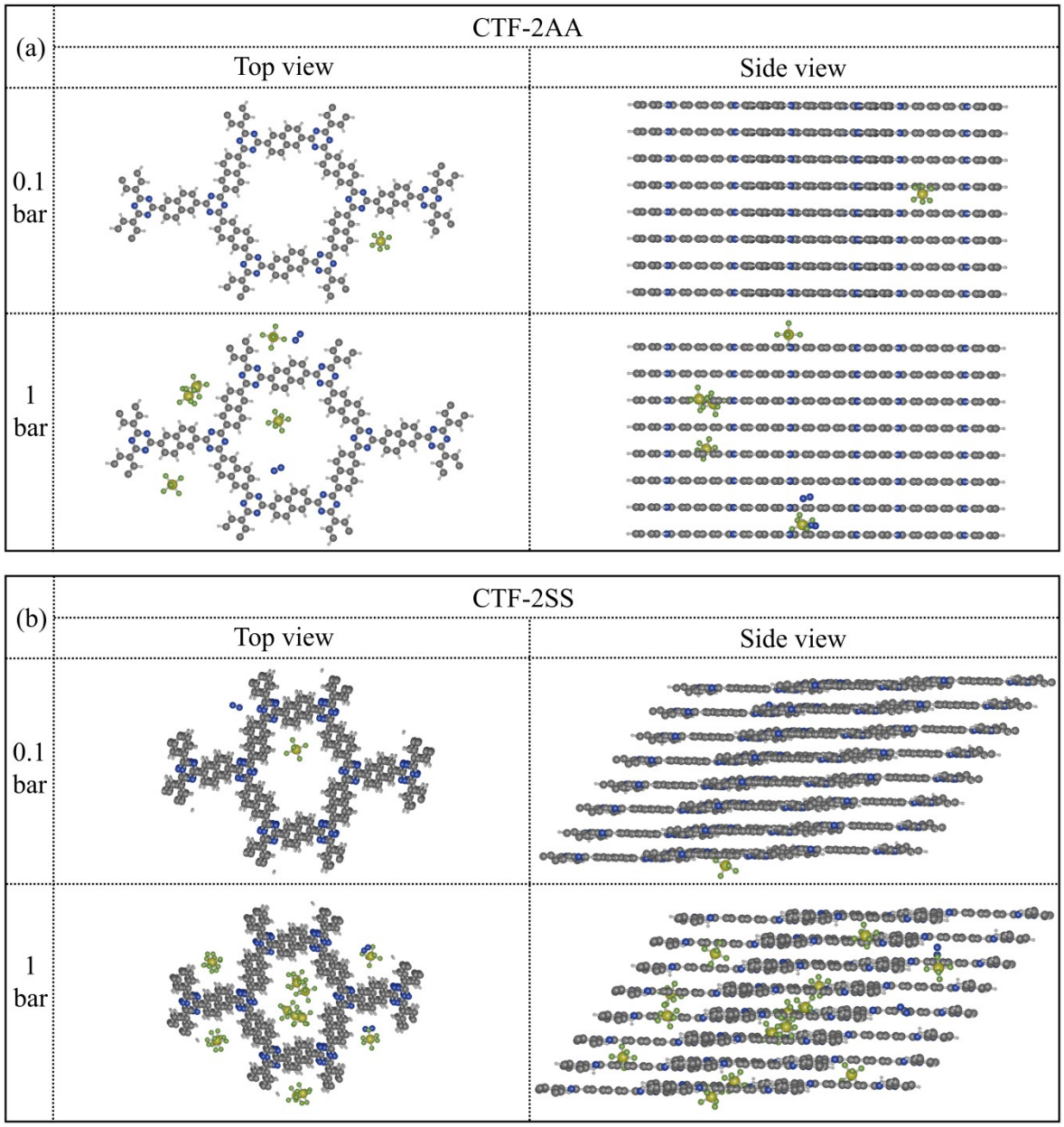

Fig. S8 Snapshots of (a) CTF-2AA and (b) CTF-2SS with an SF_6/N_2 ratio of 1:9.

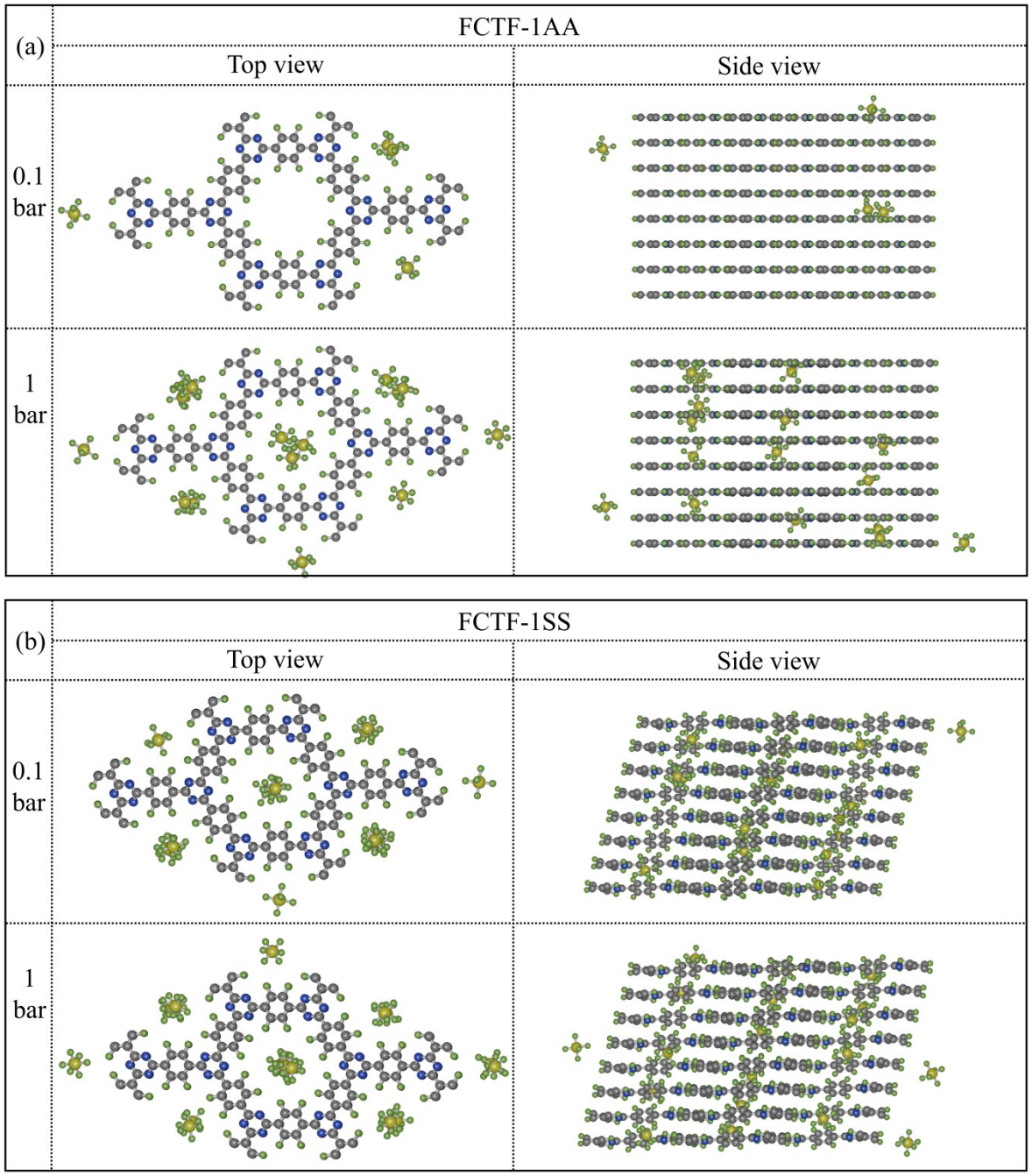

Fig. S9 Snapshots of (a) FCTF-1AA and (b) FCTF-1SS with an SF_6/N_2 ratio of 1:9.

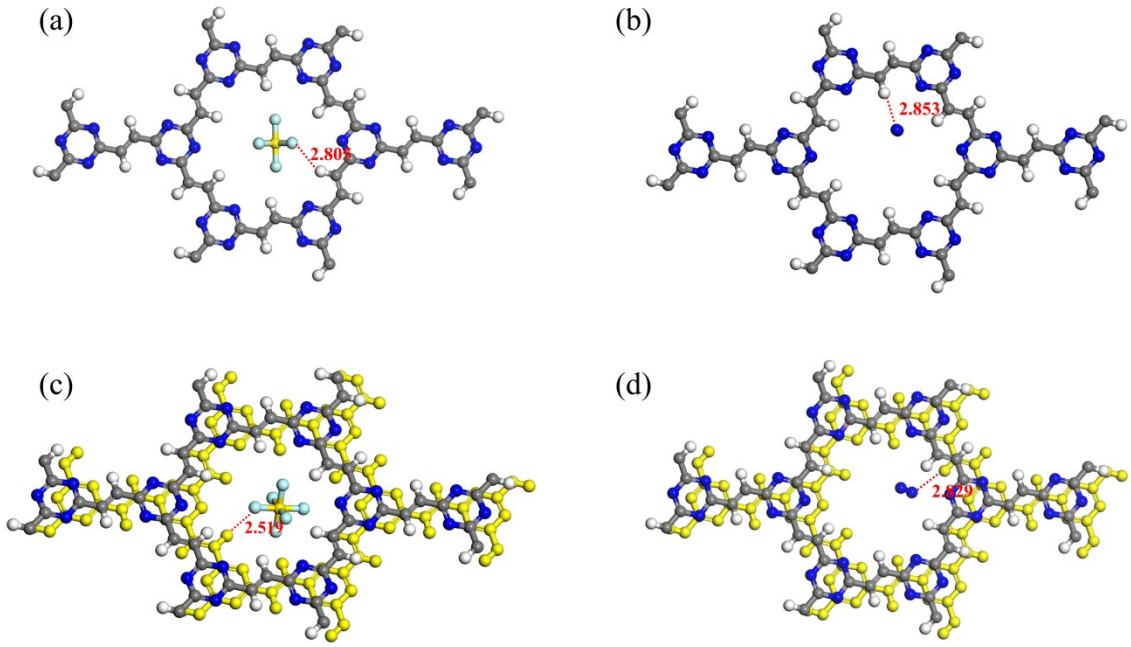

Fig. S10 Stable SF_6 adsorption configurations in (a) CTF-FUMAA and (c) CTF-FUMSS. Stable N_2 adsorption configurations in (b) CTF-FUMAA and (d) CTF-FUMSS.

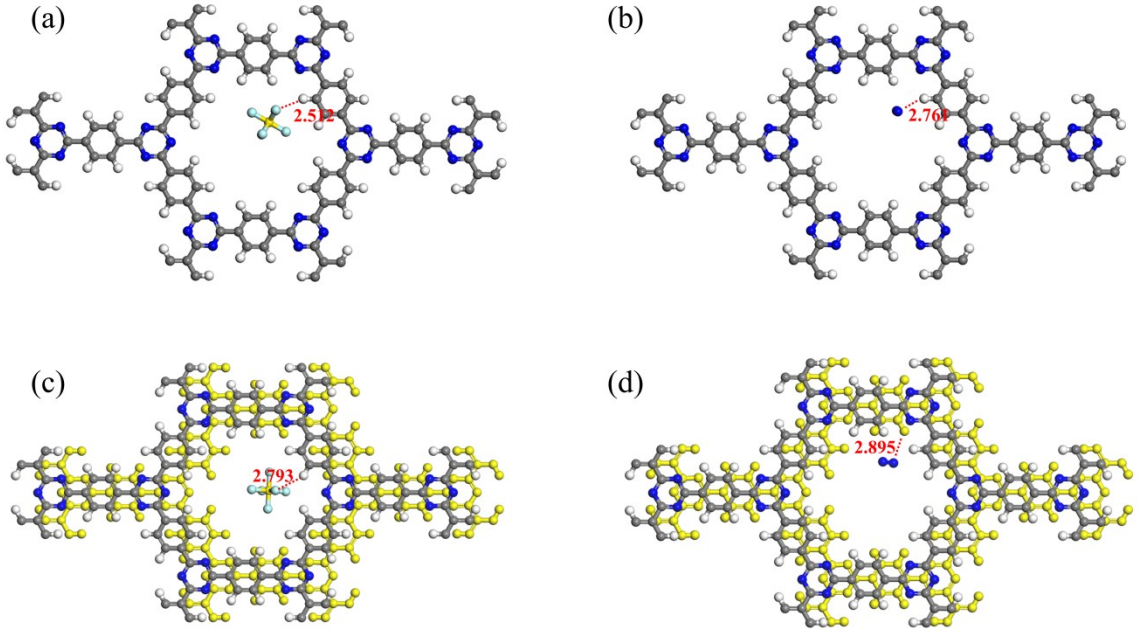

Fig. S11 Stable SF_6 adsorption configurations in (a) CTF-1AA and (c) CTF-1SS. Stable N_2 adsorption configurations in (b) CTF-1AA and (d) CTF-1SS.

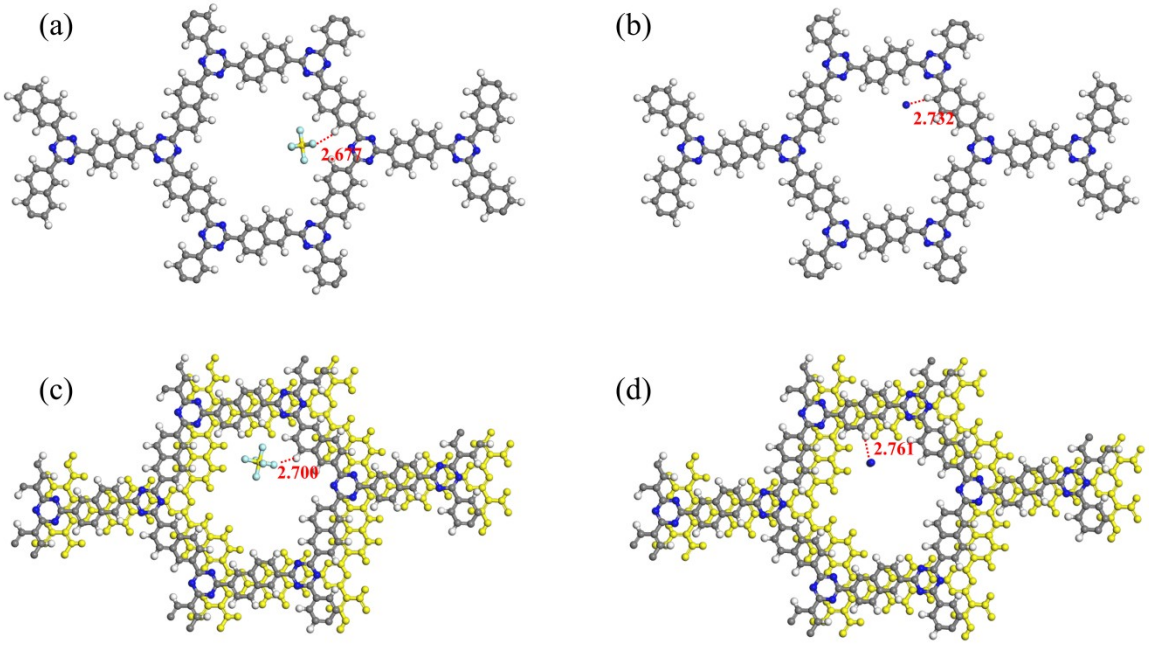

Fig. S12 Stable SF_6 adsorption configurations in (a) CTF-2AA and (c) CTF-2SS. Stable N_2 adsorption configurations in (b) CTF-2AA and (d) CTF-2SS.

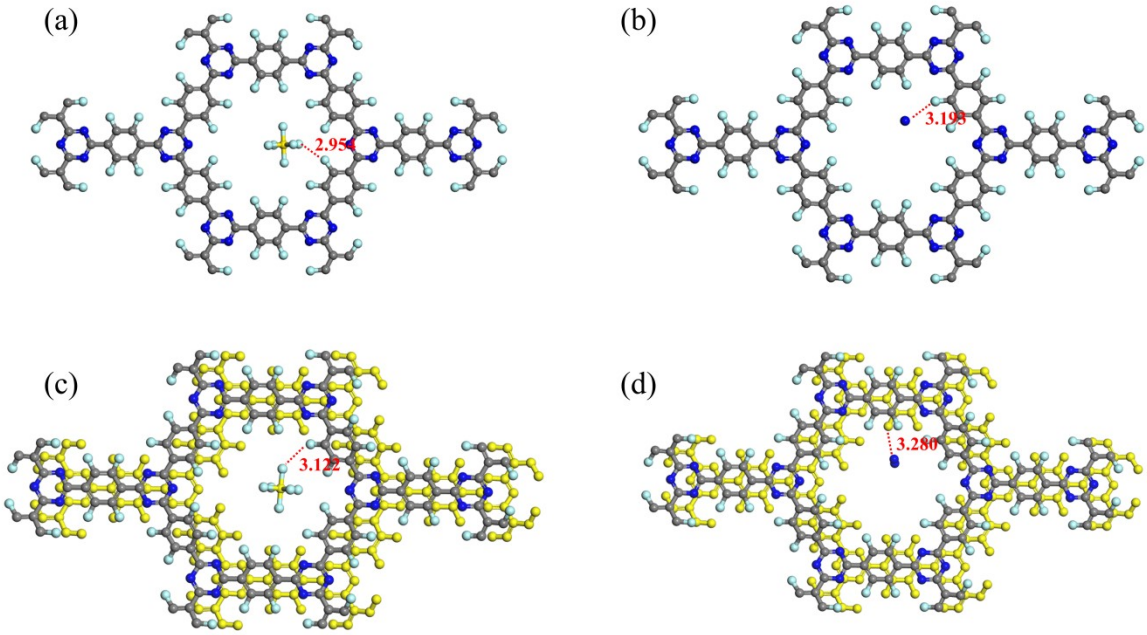

Fig. S13 Stable SF_6 adsorption configurations in (a) FCTF-1AA and (c) FCTF-1SS. Stable N_2 adsorption configurations in (b) FCTF-1AA and (d) FCTF-1SS.

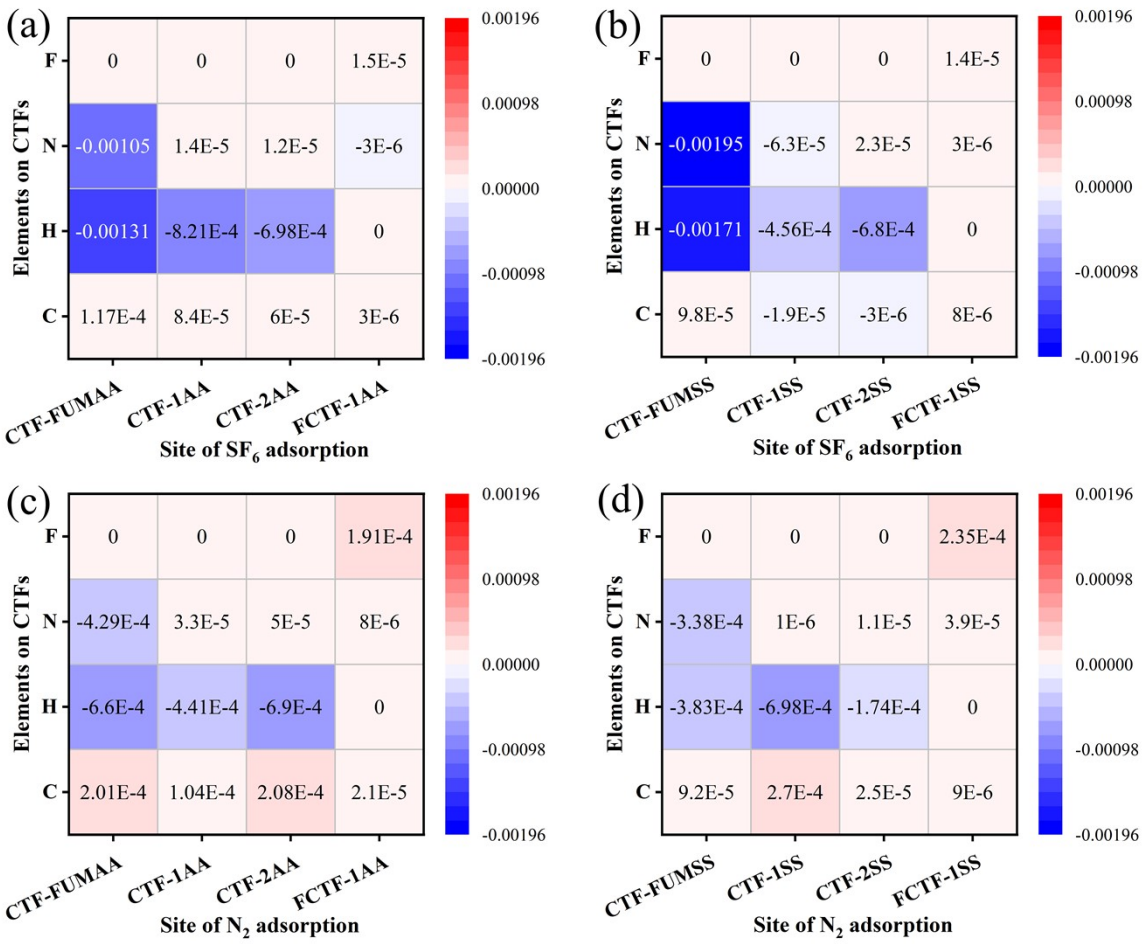

Fig. S14 The Bader charge transfer (ΔQ_F) heatmap of CTFs after adsorbing are (b) SF_6 for AA stacking, (c) SF_6 for SS stacking, (e) N_2 for AA stacking and (f) N_2 for SS stacking, respectively. Each square represents the cumulative Bader charge transfer of each type of atom. Each square represents the Bader charge transfer of each single atom (C atom, H atoms, N atoms and F atoms) in the each CTF.

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