Supporting Information

Accurate DDEC-Charge-Guided Screening of High-Performance MOFs for SF_6/N_2 Separation

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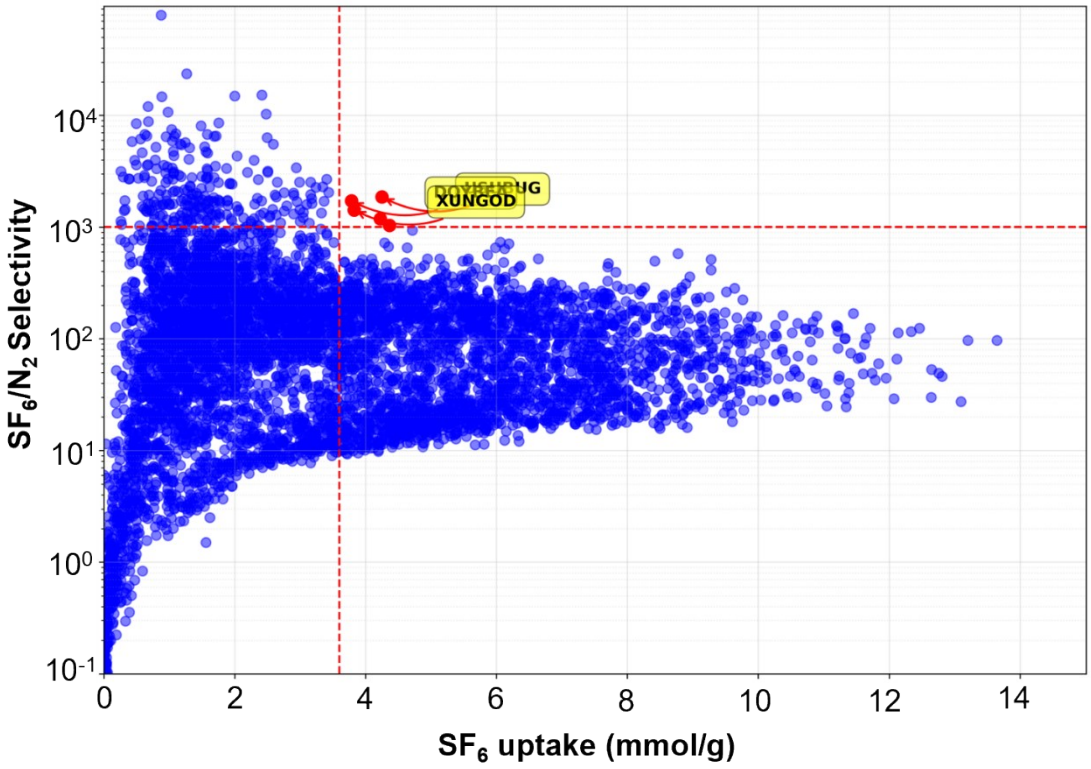

Figure S1. (a) High-throughput GCMC screening of the QMOF dataset based on SF_6/N_2 selectivity and SF_6 uptake at 298 K and 1.0 bar.

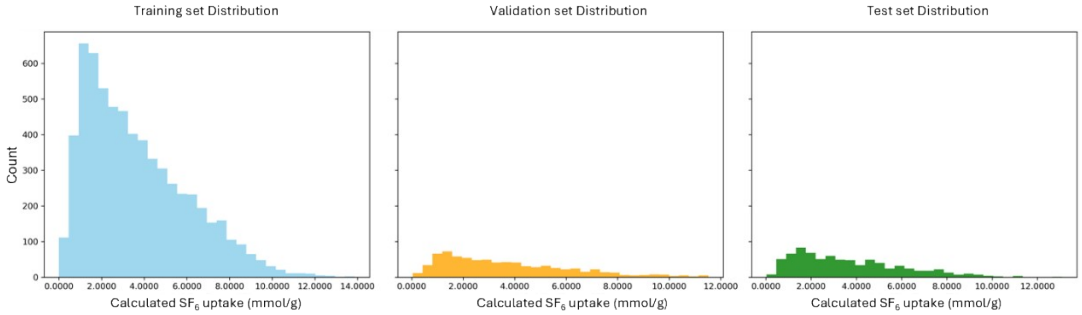

Figure S2. Distribution of SF_6 adsorption uptakes in the training, validation, and test sets after label-based binning.

Table S1. Lennard-Jones parameters of elements employed in this work.

Element	σ (Å)	ε/k_B (K)	Element	σ (Å)	ε/k_B (K)	Element	σ (Å)	ε/k_B (K)
Ac	3.10	16.6	Ge	3.81	190.69	Po	4.20	163.52
Ag	2.80	18.11	Gd	3.00	4.53	Pr	3.21	5.03
Al	4.01	254.09	H	2.57	22.14	Pt	2.45	40.25
Am	3.01	7.04	Hf	2.80	36.23	Pu	3.05	8.05
Ar	3.45	93.08	Hg	2.41	193.71	Ra	3.28	203.27
As	3.77	155.47	Ho	3.04	3.52	Rb	3.67	20.13
At	4.23	142.89	I	4.01	170.57	Re	2.63	33.21
Au	2.93	19.62	In	3.98	301.39	Rh	2.61	26.67
B	3.64	90.57	Ir	2.53	36.73	Rn	4.25	124.78
Ba	3.30	183.15	K	3.40	17.61	Ru	2.64	28.18
Be	2.45	42.77	Kr	3.69	110.69	S	3.59	137.86
Bi	3.89	260.63	La	3.14	8.55	Sb	3.94	225.91
Bk	2.97	6.54	Li	2.18	12.58	Sc	2.94	9.56
Br	3.73	126.29	Lu	3.24	20.63	Se	3.75	146.42
C	3.43	52.83	Lr	2.88	5.53	Si	3.83	202.27
Ca	3.03	119.75	Md	2.92	5.53	Sm	3.14	4.03
Cd	2.54	114.72	Mg	2.69	55.85	Sn	3.91	285.28
Ce	3.17	6.54	Mn	2.64	6.54	Sr	3.24	118.24
Cf	2.95	6.54	Mo	2.72	28.18	Ta	2.82	40.75
Cl	3.52	114.21	N	3.26	34.72	Tb	3.07	3.52
Cm	2.96	6.54	Na	2.66	15.09	Tc	2.67	24.15
Co	2.56	7.04	Ne	2.66	21.13	Te	3.98	200.25
Cr	2.69	7.55	Nb	2.82	29.69	Th	3.03	13.08
Cu	3.11	2.52	Nd	3.18	5.03	Ti	2.83	8.55
Cs	4.02	22.64	No	2.89	5.53	TI	3.87	342.14
Dy	3.05	3.52	Ni	2.52	7.55	Tm	3.01	3.02
Eu	3.11	4.03	Np	3.05	9.56	U	3.02	11.07
Er	3.02	3.52	O	3.12	30.19	V	2.80	8.05
Es	2.94	6.04	Os	2.78	18.62	W	2.73	33.71
F	3.00	25.16	P	3.69	153.46	Xe	3.92	167.04
Fe	2.59	6.54	Pa	3.05	11.07	Y	2.98	36.23
Fm	2.93	6.04	Pb	3.83	333.59	Yb	2.99	114.72
Fr	4.37	25.16	Pd	2.58	24.15	Zn	2.46	62.39
Ga	3.90	208.81	Pm	3.16	4.53	Zr	2.78	34.72

Table S2. Lennard-Jones parameters of SF_6.

Atom	ε/k_B (K)	σ (Å)	Charge (e)
S	165.14	3.228	0.66
F	27.02	2.947	-0.11

Table S3. Hyperparameters used in MOF-CGCNN model in our work.

Hyperparameters	value	range
Number of convolutional layers	5	[4-6]
Number of neurons in the hidden layer	580	[10-1000]
Number of hidden layers	1	[1-2]
Dropout	0.15	[0.1-0.6]
Learning rate	0.002	0.001,0.002
Weight Decay	1e-7	1e-6,1e-7,1e-8

Table S4. Top-performing MOFs for SF_6 adsorption, defined as those exhibiting SF_6 uptakes greater than 3.60 mmol g^{-1} and SF_6/N_2 selectivity exceeding 1000 at 298 K and 1 bar. All listed structures have passed structural validation using the MOSAEC algorithm.

MOF Name	SF_6 uptake	SF_6/N_2 Selectivity
USUBUG	4.25	1860.3
DOYBEA	3.79	1943.9
XUNGOD	3.83	1428.6
Al_2O_6-fum_B-irmof10_B_No12	4.23	1190.6
Al_2O_6-fum_B-irmof10_B_No41	4.36	1037.9

Table S5. Top-performing hypothetical MOFs for SF_6 adsorption, defined as those exhibiting SF_6 uptakes greater than 8.03 mmol g^{-1} and SF_6/N_2 selectivity exceeding 271.9 at 298 K and 1 bar.

Hypothetical MOF Name	**SF_6 uptake**	**SF_6/N_2 Selectivity**
hMOF-5060764	8.41	310.98
hMOF-5035619	8.26	387.76
hMOF-5035596	8.97	279.53