Supporting Information.

Tailoring the CO_2 selective adsorption properties of MOR zeolites by post functionalization

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Table S1. Elemental analysis results for funtionalized MOR zeolites

Sample	N content (%)	C content (%)	H content (%)
Ph_*f*_MOR	trace	1.29	1.07
$(Me)_2$Ph_*f*_MOR	0.07	1.21	1.21
MeOPh_*f*_MOR	0.15	2.52	1.19
$(MeO)_2$Ph_*f*_MOR	0.09	1.43	1.17
*i*PrPh_*f*_MOR	0.12	1.43	1.18

Table S2. Atomic parameters resulting from the Rietveld refinement of Ph_*f*_MOR [a]

Atom	Occupancy	*x*	*y*	*z*	U_{iso} (Å^2)
Si1[b]	0.8667	0.1934 (4)	0.4311 (4)	0.5428 (8)	0.0097 (14)[e]
Si2[b]	0.8667	0.1937 (4)	0.1904 (3)	0.5372 (7)	0.0097 (14)[e]
Si1[b]	0.8667	0.0865 (5)	0.3790 (5)	0.25	0.0097 (14)[e]
Si2[b]	0.8667	0.0848 (5)	0.2201 (5)	0.25	0.0112 (11)[e]
Al1[b]	0.1333	0.1934 (4)	0.4311 (4)	0.5428 (8)	0.0112 (11)[e]
Al2[b]	0.1333	0.1937 (4)	0.1904 (3)	0.5372 (7)	0.0112 (11)[e]

Al1[b]	0.1333	0.0865 (5)	0.3790 (5)	0.25	0.0112 (11)[e]
Al2[b]	0.1333	0.0848 (5)	0.2201 (5)	0.25	0.0097 (14)[e]
O1	1.0	0.1265 (7)	0.4112 (7)	0.4204 (15)	0.014 (2)[e]
O2	1.0	0.1181 (6)	0.1892 (7)	0.4316 (14)	0.014 (2)[e]
O3	1.0	0.2641 (6)	0.3865 (7)	0.5038 (18)	0.014 (2)[e]
O4	1.0	0.0878 (9)	0.2997 (5)	0.25	0.014 (2)[e]
O5	1.0	0.1740 (11)	0.1898 (12)	0.75	0.014 (2)[e]
O6	1.0	0.1677 (12)	0.4209 (11)	0.75	0.014 (2)[e]
O7	1.0	0.2302 (9)	0.5	0.5	0.014 (2)[e]
O8	1.0	0.25	0.25	0.5	0.014 (2)[e]
O9	1.0	0	0.4011 (12)	0.25	0.014 (2)[e]
O10	1.0	0	0.1944 (13)	0.25	0.014 (2)[e]
Na1	0.965 (19)	0	0.7675 (12)	0.25	0.113 (17)
Na2	0.635 (19)	0	0.4442 (15)	0.75	0.002 (14)
Ow1[c]	0.511 (10)	0.4097 (13)	0.5107 (12)	-0.174(4)	0.012 (15)
Ow2[c]	0.75 (2)	0	0.9093 (14)	0	0.068 (11)
C1[d]	0.0425	0.3110 (2)	0.4902 (4)	0.4525 (12)	0.30 (9)[e]

C2[d]	0.0425	0.3285 (2)	0.4350 (2)	0.3498 (11)	0.30 (9)[e]
C3[d]	0.0425	0.4019 (2)	0.4229 (2)	0.3010 (8)	0.30 (9)[e]
C4[d]	0.0425	0.4578 (2)	0.4663 (3)	0.3537 (12)	0.30 (9)[e]
C5[d]	0.0425	0.4401 (2)	0.5220 (2)	0.4541 (11)	0.30 (9)[e]
C6[d]	0.0425	0.3668 (3)	0.5337 (2)	0.5044 (8)	0.30 (9)[e]

a) The chemical composition of Ph_*f*_MOR is $Na_{6.4}Si_{41.6}Al_{6.4}O_{96}$ · 14.2 H_2O · 0.68 C_6H_5. Lattice constants are *a*= 18.095 (2) Å, *b*= 20.400 (2) Å, *c*= 7.5023 (7) Å, α=β= γ =90°, resulting in V= 2769.3 (6) $Å^3$. The fit parameters are GOF=2.65, R_{wp}=12.83.

b) The atomic positions of Al1-Al4 are refined as identical position of Si1-Si4, respectively.

c) Ow indicate oxygen atom of water molecule.

d) C1-C6 indicate carbon atoms of phenyl functional group.

e) The atomic displacement parameters (U_{iso}) of silicon, aluminum and oxygen atoms of the zeolite framework and carbon atoms of phenyl functional group are refined as single parameter of each element.

Table S3. Atomic parameters resulting from the Rietveld refinement of *uf*_MOR[a]

Atom	Occupancy	*x*	*y*	*z*	U_{iso} (Å^2)
Si1[b]	0.8667	0.1922 (4)	0.4277 (3)	0.5410 (8)	0.0112 (12)[d]
Si2[b]	0.8667	0.1974 (4)	0.1882 (3)	0.5360 (7)	0.0112 (12)[d]
Si1[b]	0.8667	0.0875 (5)	0.3809 (5)	0.25	0.0112 (12)[d]
Si2[b]	0.8667	0.0851 (5)	0.2223 (4)	0.25	0.0112 (12)[d]
Al1[b]	0.1333	0.1922 (4)	0.4277 (3)	0.5410 (8)	0.0131 (11)[d]
Al2[b]	0.1333	0.1974 (4)	0.1882 (3)	0.5360 (7)	0.0131 (11)[d]
Al1[b]	0.1333	0.0875 (5)	0.3809 (5)	0.25	0.0131 (11)[d]
Al2[b]	0.1333	0.0851 (5)	0.2223 (4)	0.25	0.0131 (11)[d]
O1	1.0	0.1234 (6)	0.4204 (6)	0.4151 (14)	0.026 (2)[d]
O2	1.0	0.1219 (6)	0.1944 (8)	0.4321 (13)	0.026 (2)[d]
O3	1.0	0.2612 (6)	0.3804 (7)	0.5050 (18)	0.026 (2)[d]
O4	1.0	0.0873 (9)	0.3017 (5)	0.25	0.026 (2)[d]
O5	1.0	0.1741 (10)	0.1840 (12)	0.75	0.026 (2)[d]
O6	1.0	0.1654 (10)	0.4180 (11)	0.75	0.026 (2)[d]
O7	1.0	0.2186 (12)	0.5	0.5	0.026 (2)[d]

O8	1.0	0.25	0.25	0.5	0.026 (2)[d]
O9	1.0	0	0.4002 (13)	0.25	0.026 (2)[d]
O10	1.0	0	0.1966 (14)	0.25	0.026 (2)[d]
Na1	0.970 (19)	0	0.7630 (12)	0.25	0.085 (17)
Na2	0.630 (11)	0	0.4398 (16)	0.75	0.015 (14)
Ow1[c]	0.817 (13)	0.3918 (10)	0.5132 (10)	-0.152 (2)	0.125 (14)
Ow2[c]	0.970 (3)	0	0.8948 (10)	0	0.057 (15)

a) The chemical composition of *uf*-MOR is $Na_{6.4}Si_{41.6}Al_{6.4}O_{96}$ · 20.8 H_2O. Lattice constants are *a*= 18.126 (2) Å, *b*= 20.450 (2) Å, *c*= 7.5160 (7) Å, α=β= γ =90°, resulting in V= 2785.9 (5) $Å^3$. The fit parameters are GOF=2.77, R_{wp}=13.29.

b) The atomic positions of Al1-Al4 are refined as identical position of Si1-Si4, respectively.

c) Ow indicate oxygen atom of water molecule.

d) The atomic displacement parameters (U_{iso}) of silicon, aluminum and oxygen atoms of the zeolite framework are refined as single parameter of each element.

Table S4. CO_2/N_2 and CO_2/CH_4 selectivities of prior works.

Sample	Gas Uptake (mmol g^{-1})			Selectivity		Temperature (K)	Selectivity [b]	Ref.
	CO_2	N_2	CH_4	CO_2/N_2	CO_2/CH_4			
This work								
*uf*_MOR	3.18	0.689	1.13	14[a]	6.6[a]	298	IAST	
Ph_*f*_MOR	1.85	0.288	0.611	41.3[a]	12.6[a]	298	IAST	
MeOPh_*f*_MOR	2.09	0.287	0.691	36.9[a]	12.7[a]	298	IAST	
$(Me)_2$Ph_*f*_MOR	1.96	0.299	0.619	55.4[a]	17.4[a]	298	IAST	
$(MeO)_2$Ph_*f*_MOR	1.88	0.293	0.626	47.2[a]	13.4[a]	298	IAST	
*i*Pr_*f*_MOR	1.88	0.289	0.604	59.9	18.3	298	IAST	
Zeolite (Silicate)								
Zeolite-ZK-4 (Na^+, Si/Al=1.8)	4.59	0.584		42		273	Henry's law	[1]
CLD-modified zeolite 4A	2.9	0.06	0.09	44		298	Pure component selectivity	[2]
DDR	1.02	0.134	0.344		5.7-6	298	IAST	[3]
Silicate-1	1.63	0.652	0.652		5.0-5.2	298	IAST	[3]

Beta	2.00	0.559	0.558		5.7-6	298	IAST	[3]
K-ETS-10	2.37		2.57		44.6	298	IAST	[4]
Carbon								
MOF-200/GO	1.34		0.2		18.37	298	IAST	[5]
CM2-900-1	2.64	0.246		22.3		298	IAST	[6]
CHNH1:3	2.65	0.362		69.1		298	Henry's law	[7]
Metal-Organic Framework								
SMOF-SIFSIX-1a	2.29	0.47	1.05	131.7	21.1	273	IAST	[8]
$[Zn_2(DMF)_2]$				35-40		298	IAST	[9]
BILP-10	2.52		0.688	57	9.0	298	IAST	[10]
$[Zn_3L_2(HCOO)_{1.5}][(CH_3)_2NH_2]_{1.5}$	2.57		0.399		25	273	IAST	[11]

a) The IAST selectivity of this work determined in 0.1 bar

b) Calculation method of selectivity.

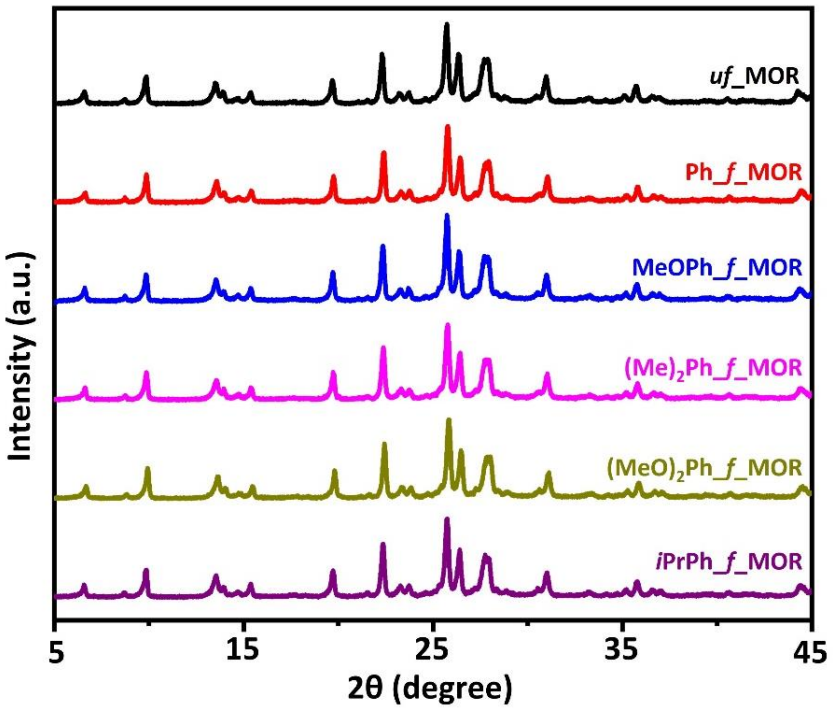

Fig. S1. Powder X-ray diffraction patterns for *uf*-MOR and *f*_MOR zeolites.

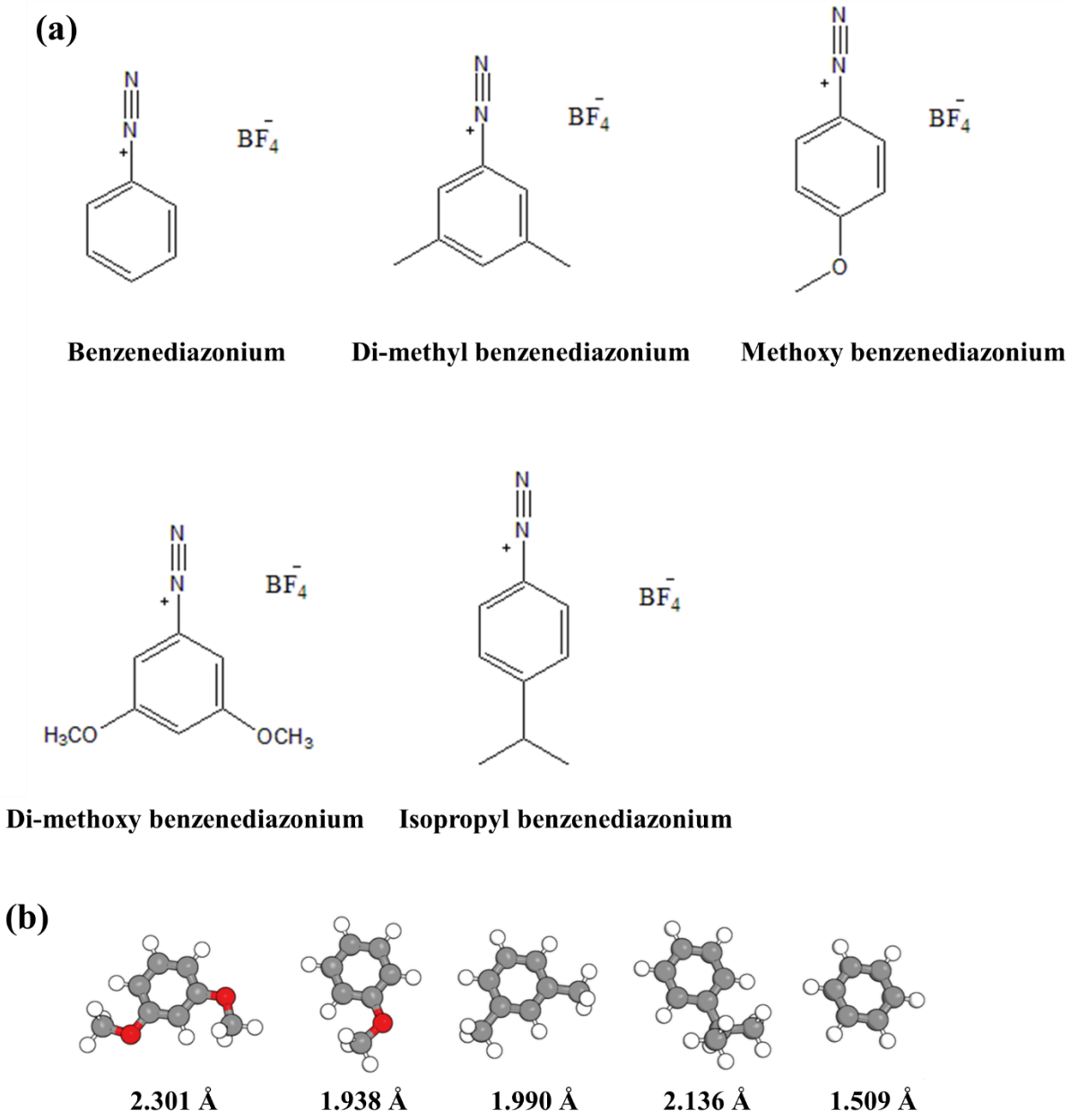

Fig. S2. (a) Gyration radii of organic functionalizing agents. (b) Chemical structures of diazonium salts used for MOR zeolite functionalization.

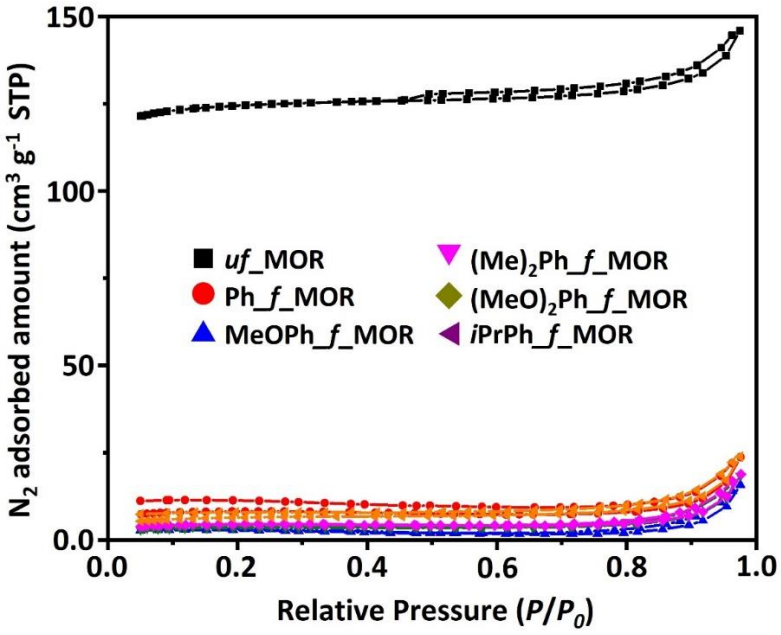

Fig. S3. N_2 adsorption–desorption isotherms of *uf*_MOR zeolite and *f*_MOR.

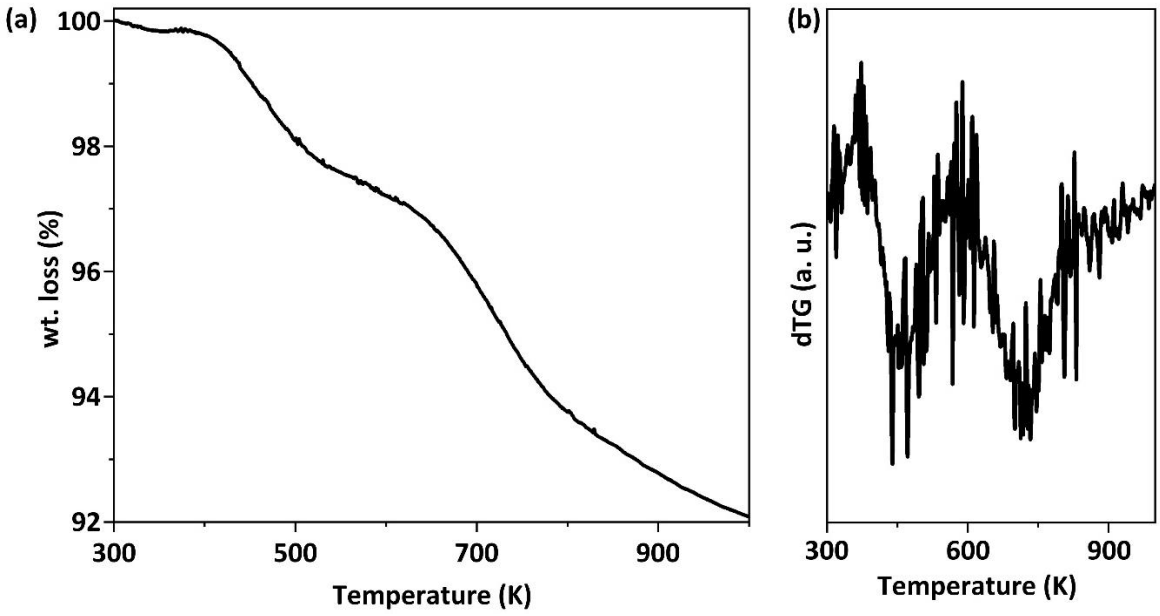

Fig. S4. TGA of Ph_*f*_MOR sample. (a) Weight curve. (b) Derivative weight curve.

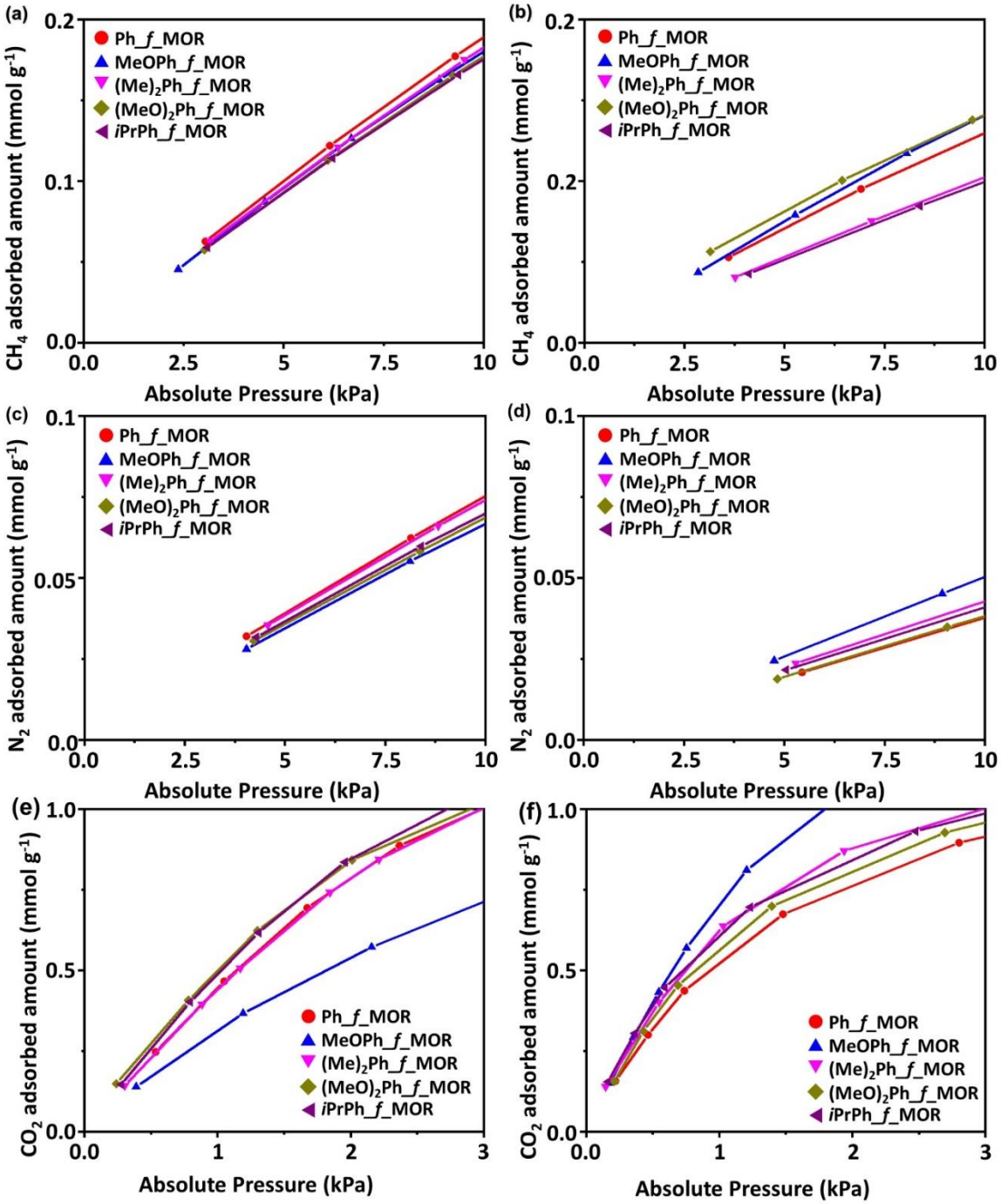

Fig. S5. Enlarged images of CH_4 (a-288 K, b-298 K), N_2 (c-288 K, d-298 K), and CO_2 (e-288 K, f-298 K) adsorption isotherms for Ph_*f*_MOR, MeOPh_*f*_MOR, *i*PrPh_*f*_MOR, $(Me)_2$Ph_*f*_MOR, and $(MeO)_2$Ph_*f*_MOR.

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