Supplementary Information (SI) for Nanoscale.

Supplemental Information

for

Rational design of stable carbon nitride monolayer membranes for highly controllable CO_2 capture and separation from CH_4 and C_2H_2

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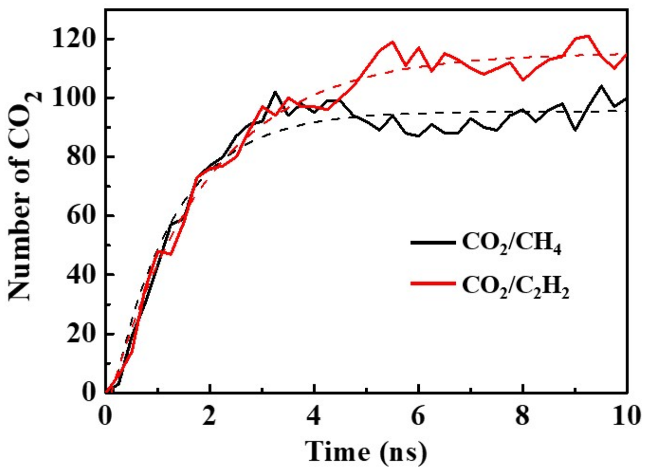

Fig. S1. The number of permeated CO_2 molecules versus the simulation time in the separation of CO_2/CH_4 and CO_2/C_2H_2 at 300 K. The corresponding dotted lines are the fitted lines.

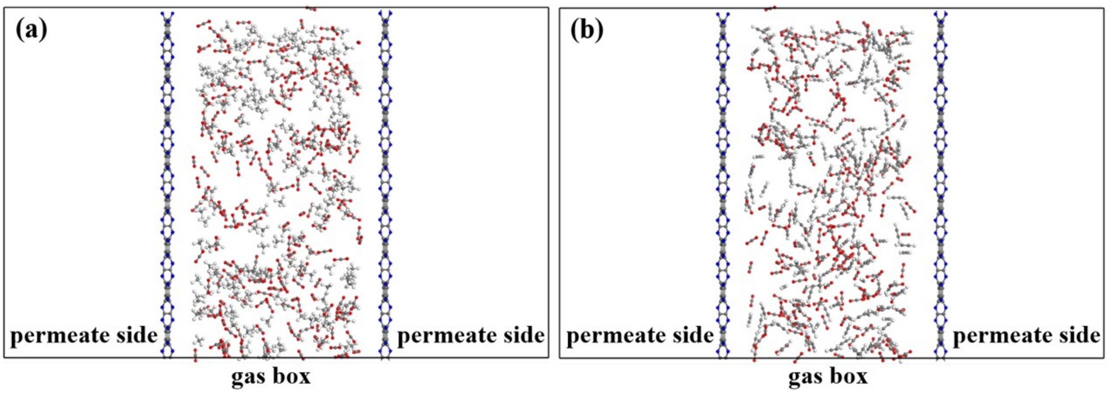

Figure S2. The initial models of (a) CO_2/CH_4 and (b) CO_2/C_2H_2 mixtures through the g-$C_{12}N_8$ membrane.

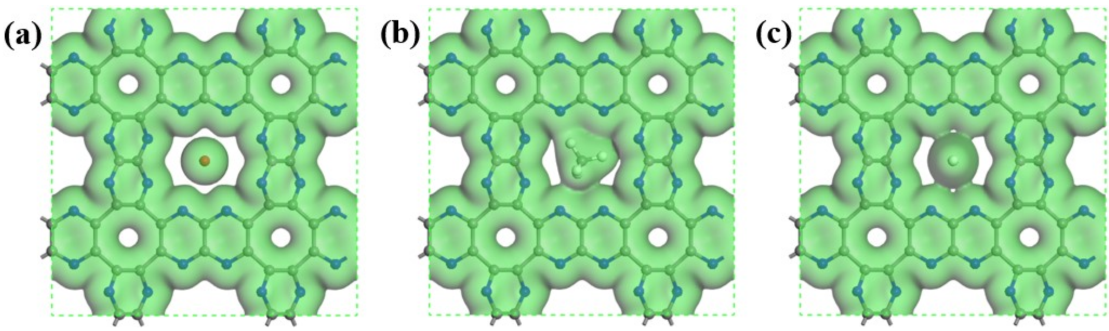

Figure S3. Electron density isosurfaces for (a) CO_2, (b) CH_4, and (c) C_2H_2 molecules permeating through the $C_{12}N_8$ monolayer (isovalue of 0.08 e·Å^{-3}).

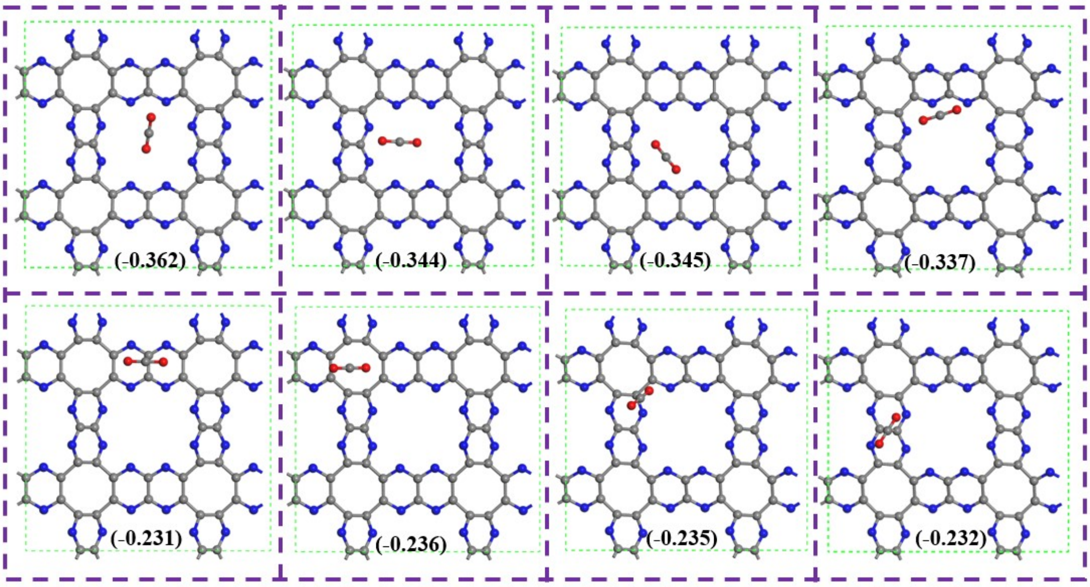

Figure S4. The stable adsorption sites for CO_2 on the g-$C_{12}N_8$ membrane. The values in parentheses are the corresponding adsorption energy (in eV).

Table S1 Atomic coordinates for the unit cell of the g-$C_{12}N_8$ monolayer.

Lattice Parameters (Å, °)	Atom	Coordinates (fractional)		
		x	*y*	*z*
a=8.01 b=8.01 c=25 α=β=γ=90	N	0.1429	0.6736	0.5057
	N	0.8571	0.3264	0.4943
	N	0.8571	0.6736	0.4943
	N	0.1429	0.3264	0.5057
	N	0.3742	0.8551	0.5374
	N	0.6258	0.1449	0.4626
	N	0.6258	0.8551	0.4626
	N	0.3742	0.1449	0.5374
	C	0.2844	0.5900	0.5119
	C	0.7156	0.4100	0.4881
	C	0.7156	0.5900	0.4881
	C	0.2844	0.4100	0.5119
	C	0.4237	0.2862	0.5151
	C	0.5763	0.7138	0.4849
	C	0.5763	0.2862	0.4849
	C	0.4237	0.7138	0.5151
	C	0.0000	0.4117	0.5000
	C	0.0000	0.5883	0.5000
	C	0.4409	0.0000	0.5214
	C	0.5591	0.0000	0.4786

Table S2 A summary of the CO_2/CH_4 separation performance of membranes in this work along with reported literature.

Membranes	Permeance (GPU)	Selectivity	Reference
g-$C_{12}N_8$	1.21×10^7	3.03×10^3	This work
MMM-0.5wt.%	154	21.4	[1]
2D-CAP	4.824×10^5	32	[2]
GO-PEGDA500	175.5	69.5	[3]
LDH membrane with 8 nm-thick	202	7	[4]
GDY monolayer(x=10%,y=20%)	1.29×10^6	5.27×10^3	[5]
ZSM-58 zeolite membranes(0.2MPa)	510	290	[6]
MMM 15%	2.74	13.30	[7]
PES CNTs	35.18	4.97	[8]
Charged NPG	5.65×10^6	42.8	[9]
Strain-controlled graphenylene	3.52×10^6	1.8×10^5	[10]
CHA (t < 500 nm)	3.82×10^4	198	[11]
Amino-functionalized SAPO-34	1.5×10^3	245	[12]
g-C_3N_4-MXene-X/Pebax	about 1.2×10^3	47.76	[13]

Table S3 A summary of the CO_2/C_2H_2 separation performance of membranes in this work along with reported literature.

Membranes	Permeance (GPU)	Selectivity	Reference
g-$C_{12}N_8$	1.39×10^7	310	This work
Strain-controlled C_2N	5.05×10^3	6	[14]
Strain-controlled C_2O	1.44×10^6	6	[14]
Porous graphene	585	17	[15]
High-silica CHA zeolite	3.28×10^3	55	[16]
ZIF-8 M-60	319.1	1.8	[17]

Reference:

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