membranes

Supplementary Materials: Effect of Incorporation of Carbon-Based Particles in Enhancing CO_2/N_2 Separation Performance in Carbon Molecular Sieve Membrane

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SUMMARY

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Table S1. Fitting Parameters for CO_2 and N_2 adsorptions onto GO and YP-50F at 25 °C.

Sample	*Gas*	q_{sat}	*b*	R^2 value	Henry's Constant, k_H
GO	CO_2	0.6759	1.762	0.9942	1.191
	N_2	0.6759	0.07684	0.9943	0.05194
YP-50F	CO_2	6.087	0.7079	0.9991	4.309
	N_2	6.087	0.06686	0.9992	0.4070

Unit of P—bar; Unit of q—mmol/g

Table 2S. Fitting Parameters for CO_2 and N_2 adsorptions onto GO and YP-50F at 35 °C.

Sample	*Gas*	$q_{sat,1}$	*b*	R^2 value	Henry's Constant, k_H
GO	CO_2	0.5976	1.541	0.9959	0.9209
	N_2	0.5976	0.07229	0.9915	0.04320
YP-50F	CO_2	5.854	0.535	0.9987	3.132
	N_2	5.854	0.05791	0.9996	0.3390

Unit of P—bar; Unit of q—mmol/g

Table S3. Performance of pure CMSMs that reported in the literature for CO_2/N_2 separation.

Polymer Matrix	Carbonization Condition	Measurement Condition	CO_2 Permeability (Barrer)	CO_2/N_2 Selectivity	Ref.
BTPA-ODA	550 °C, Argon	25 °C, Pure gas	1500	32.6	[1]
BTPA-ODA-1	700 °C, Argon	25 °C, Pure gas	300	37.5	[1]
Br-Matrimid-1	550 °C, Vacuum	35 °C, 10 atm, Pure gas	2900	11.3	[2]
Kapton	550 °C, Vacuum	25 °C, Pure gas	100	8.0	[3]
Matrimid	475 °C, Vacuum	25 °C, Pure gas	11	14.0	[3]
Matrimid-1	550 °C, Vacuum	35 °C, 10 atm, Pure gas	871	14.8	[2]
Matrimid-2	550 °C, Argon	35 °C, mixed gas (20/80)	428	39.4	This work
ODPA-TMPDA	550 °C, Argon	35 °C, mixed gas (20/80)	1120	20.0	This work
Kapton-1	600 °C, Vacuum	35 °C, Pure gas	1820	22.2	[4]
ODPA-ODA	650 °C	50 °C, Pure gas	201	17.0	[5]
P84	600 °C, Nitrogen	Pure gas	276	35.4	[6]
P84-Ag	600 °C, Nitrogen	Pure gas	619	31.1	[6]
PEI	550 °C, Vacuum	25 °C, Pure gas	69	7.6	[7]
PEI-1	500 °C, Vacuum	26 °C, Pure gas	53	17.5	[8]
Polypyrrole	550 °C, Nitrogen	35 °C, Pure gas	3520	35.2	[9]
Poly(amino imide)	150 °C, Nitrogen	35 °C, Pure gas	3	20.0	[9]
PPO-2	650 °C, Vacuum	25 °C, Pure gas	218	45.4	[10]

Note: BTDA—Benzophenone tetracarboxylic dianhydride; ODA—4,4′-oxydianiline; PPO—poly(p-phenylene oxide); TMS—chlorotrimethylsilane.

Table S4. Performance of mixed-matrix CMSMs for CO_2/N_2 separation.

Membranes	Measurement Condition	CO_2 Permeability (Barrer)	CO_2/N_2 Selectivity	F_{index}	Ref.
15 wt% GO (Matrimid)	35 °C, mixed gas (21/79)	900	39.8	0.77	This work
15 wt% GO (ODPA-TMPDA)	35 °C, mixed gas (21/79)	1526	27.6	1.24	This work
15 wt% YP-50F (Matrimid)	35 °C, mixed gas (21/79)	1294	32.9	0.59	This work
15 wt% YP-50F (ODPA-TMPDA)	35 °C, mixed gas (21/79)	1833	30.4	1.70	This Work
30 wt% PS-MFI (ODPA-TMPDA)	35 °C, mixed gas (20/80)	2397	28.8	1.81	This work
30 wt% ETS-10 (ODPA-TMPDA)	35 °C, mixed gas (20/80)	1234	34.2	1.59	This work
30 wt% SAPO-34 (ODPA-TMPDA)	35 °C, mixed gas (20/80)	2615	31.7	2.15	This work
15 wt% FS (ODPA-TMPDA)	35 °C, mixed gas (20/80)	1560	30.7	1.54	This work
24 wt% SBA-15 (PAA)	25 °C, mixed gas (50/50)	1939	66	0.14	[11]
24 wt% MCM-48 (PAA)	25 °C, mixed gas (50/50)	2850	61	0.31	[11]
3 wt% Zeolite L (PFA)	25 °C, pure gas	503	20	2.97	[12]
2 wt% Zeolite T (PFA)	25 °C, pure gas	74	51.6	1.45	[13]

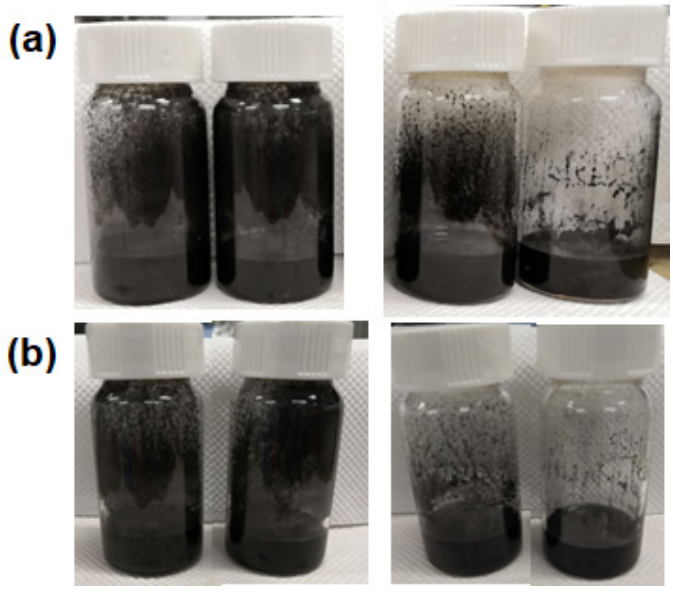

Figure S1. Photographic images upon the incorporation of GO (left) and YP-50F (right) onto Matrimid® 5218 (left) and ODPA-TMPDA (right) dope solution. The photos were taken at (**a**) 12 h and (**b**) 24 h after stopping agitation, respectively.

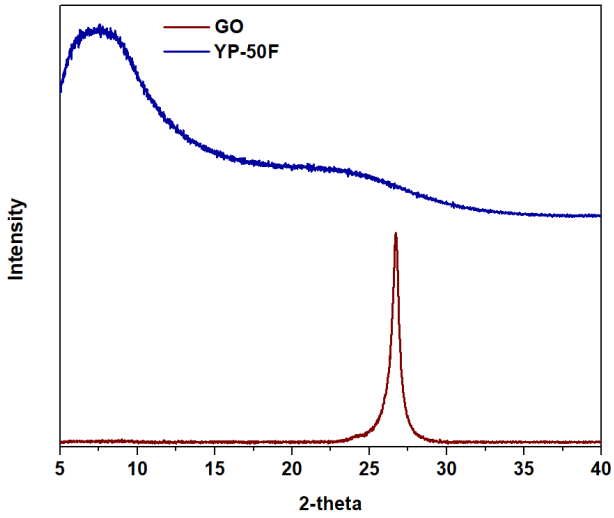

Figure S2. XRD of GO and YP-50F.

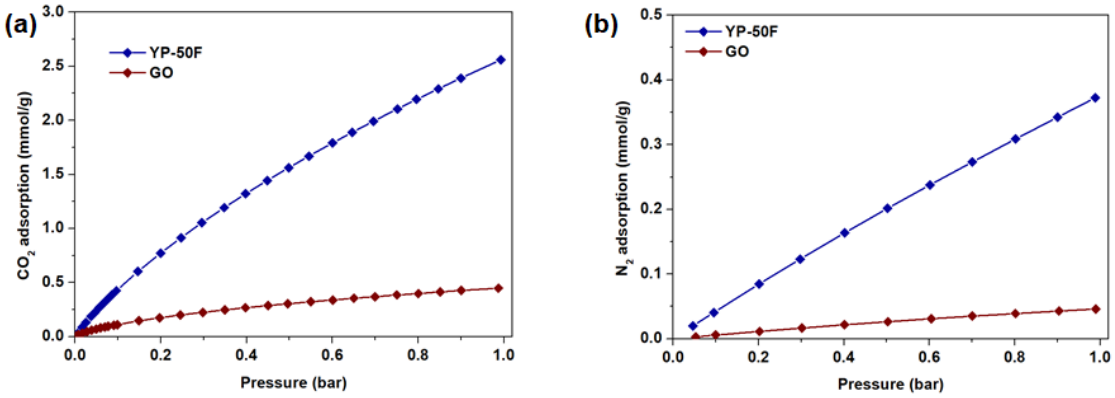

Figure S3. (**a, b**) CO_2 and N_2 adsorptions on GO and activated carbon (YP-50F) at 25 °C.

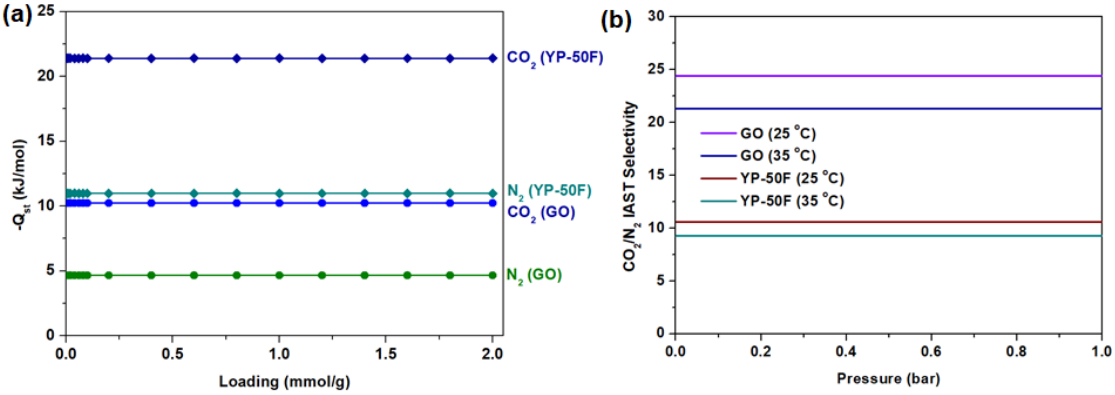

Figure S4. (a) The isosteric heats of adsorption ($-Q_{st}$) of CO_2 and N_2 and **(b)** CO_2/N_2 IAST selectivity for GO and YP-50F (the ratio of CO_2/N_2 in the feed is 20/80).

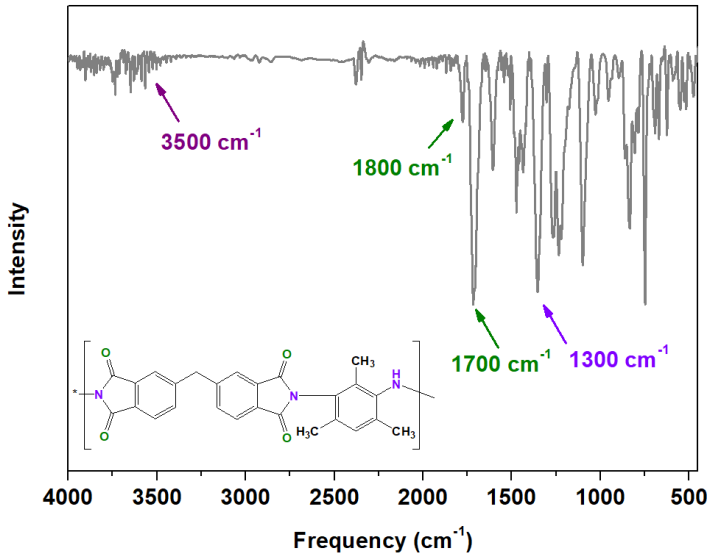

Figure S5. FT–IR spectrum of ODPA-TMPDA polymer.

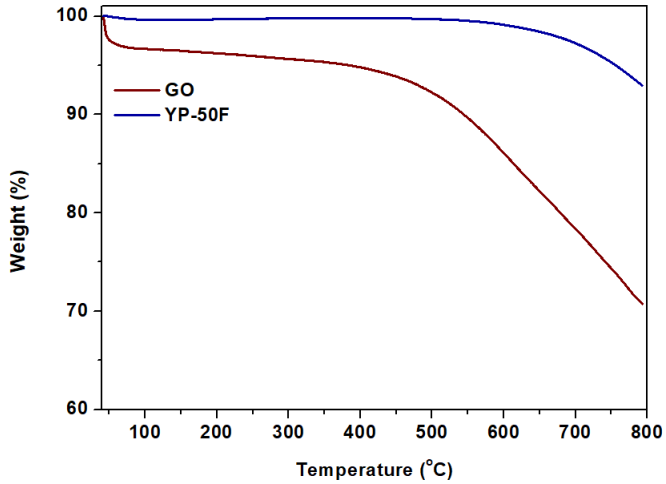

Figure S6. The TGA curves of GO and activated carbon (YP-50F).

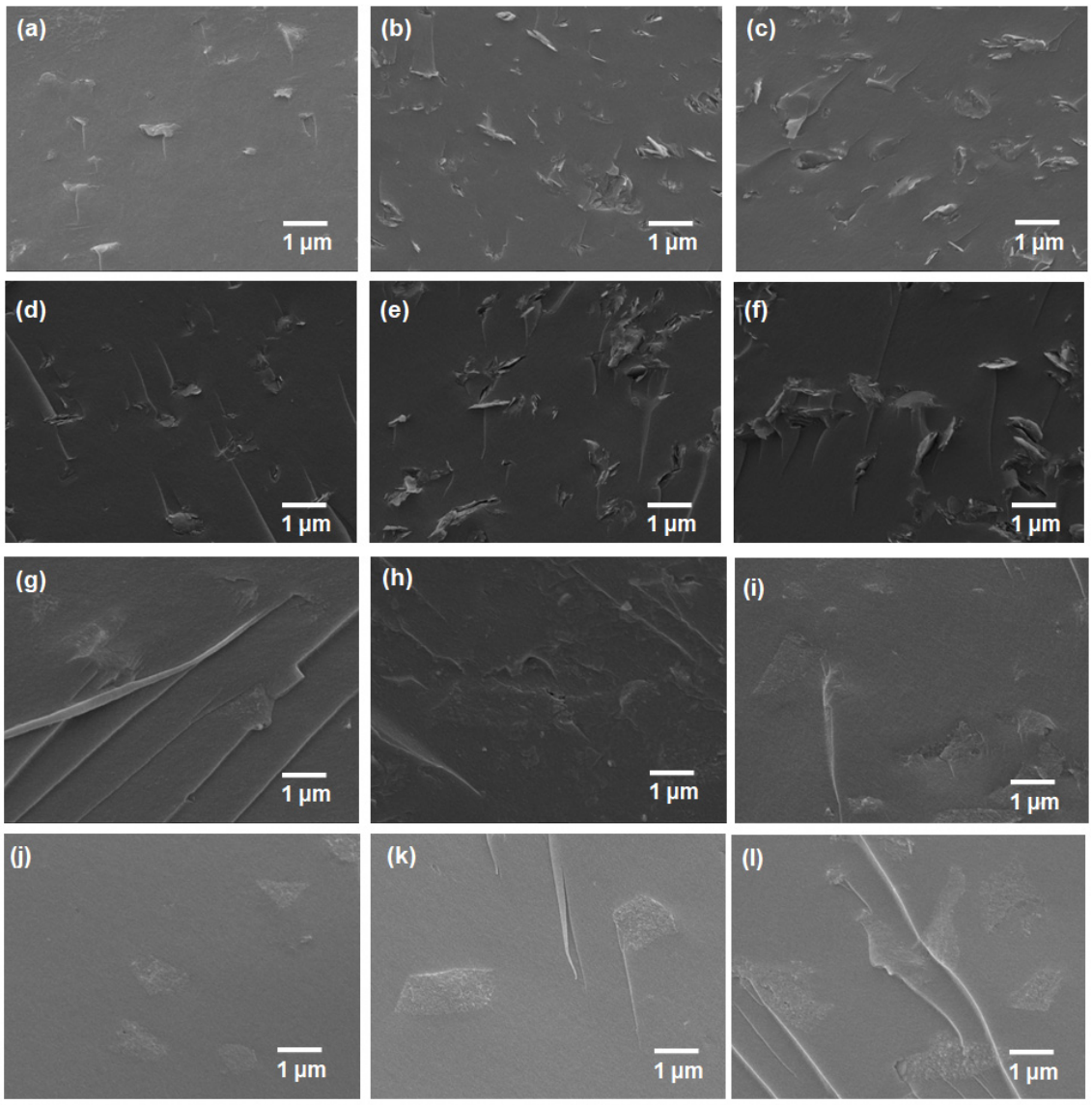

Figure S7. FESEM images of mixed-matrix carbon molecular sieve membranes at higher magnification. (**a, b, c**) 5 wt%, 10 wt% and 15 wt% GO in Matrimid® 5218; (**d, e, f**) 5 wt%, 10 wt% and 15 wt% GO in ODPA-TMPDA; (**g, h, i**b 5 wt%, 10 wt% and 15 wt% YP-50F in Matrimid® 5218; (**j, k, l**) 5 wt%, 10 wt% and 15 wt% YP-50F in ODPA-TMPDA.

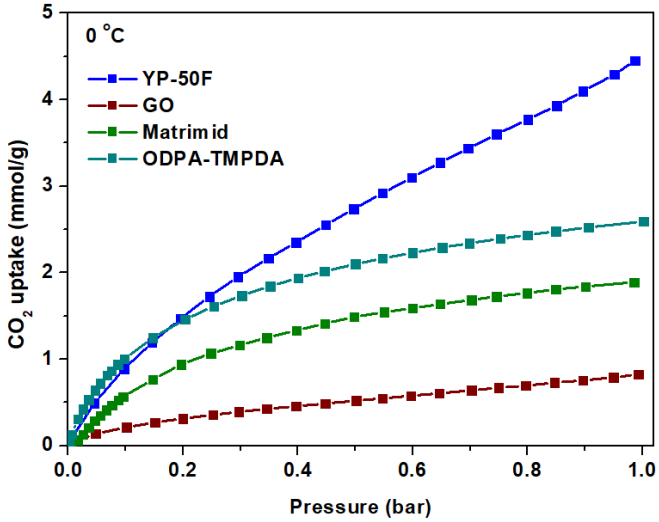

Figure S8. The CO_2 uptakes at 0 °C for YP-50F, GO, carbonized Matrimid® 5218 and carbonized ODPA-TMPDA.

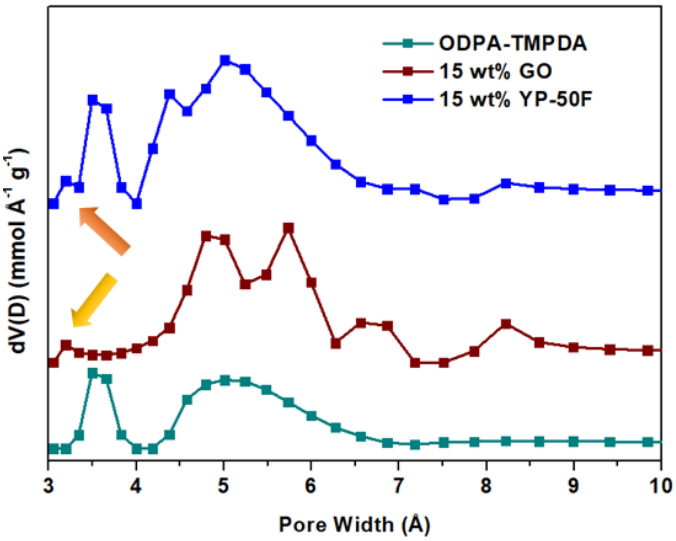

Figure S9. Pore size distribution of CMSMs and mixed-matrix CMSMs with ODPA-TMPDA as the polymer precursor. The arrowhead indicated in the figure shows the presence of smaller micropores in the mixed-matrix CMSMs.

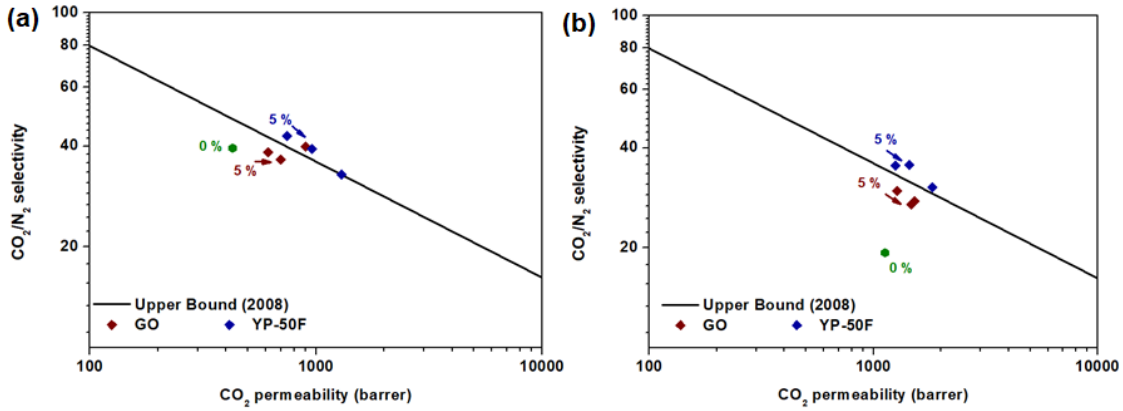

Figure S10. Robeson plot that demonstrates CO_2 permeabilities and CO_2/N_2 selectivity of CMSMs that are based on (**a**) Matrimid® 5218 and (**b**) ODPA-TMPDA polymeric precursors.

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