

Supplementary Information

Binary All-Nanoporous Composite Membrane Constructed via Vapor Phase Transformation for High-Permeance Gas Separation

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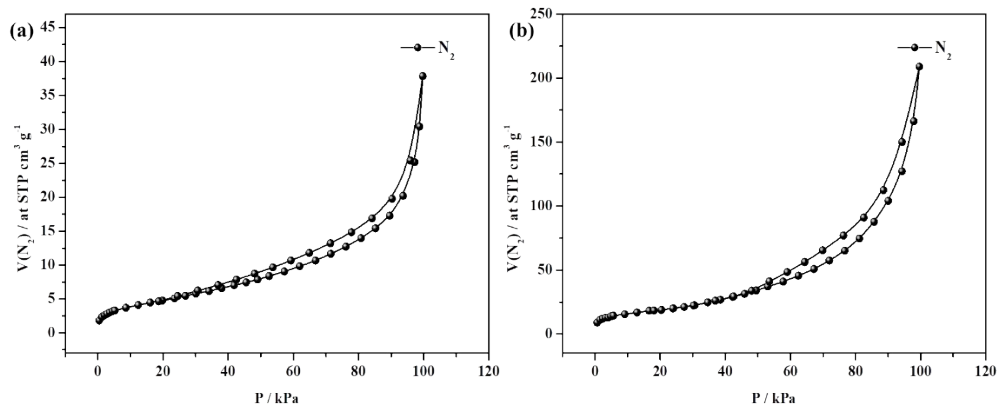


Fig. S1 N_2 adsorption-desorption isotherms of **ZIF-9** (a) and **COF-TpPa-1** (b) at 77 K.

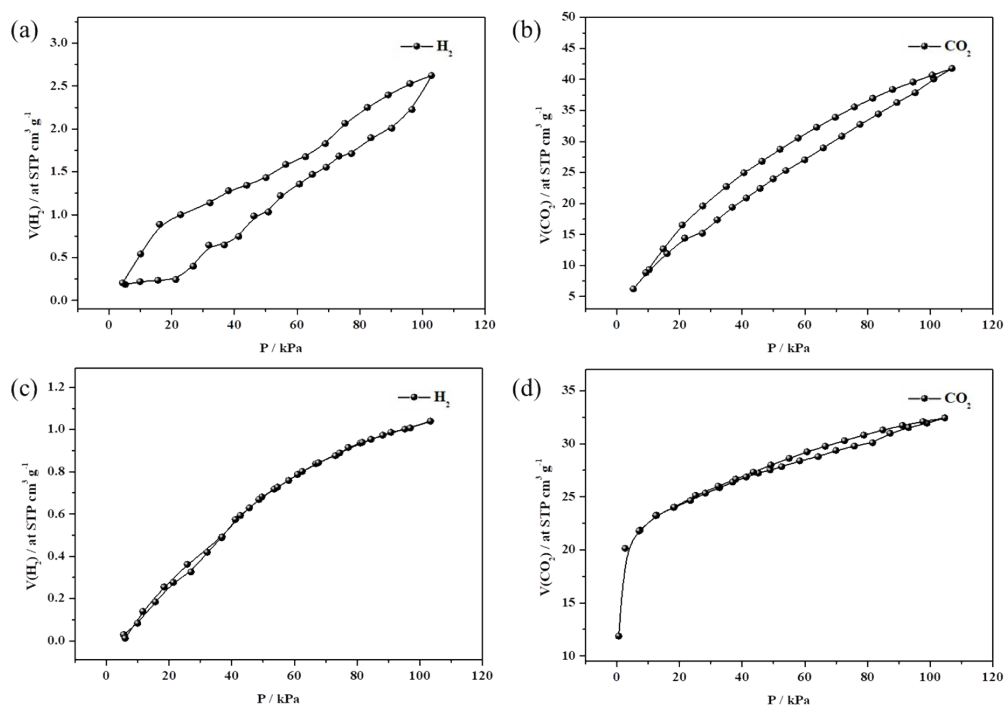


Fig. S2 H_2 adsorption-desorption isotherms of **COF-TpPa-1** (a) and **ZIF-9** (c) at 298 K; CO_2 adsorption/desorption isotherms of **COF-TpPa-1** (b) and **ZIF-9** (d) at 298 K.

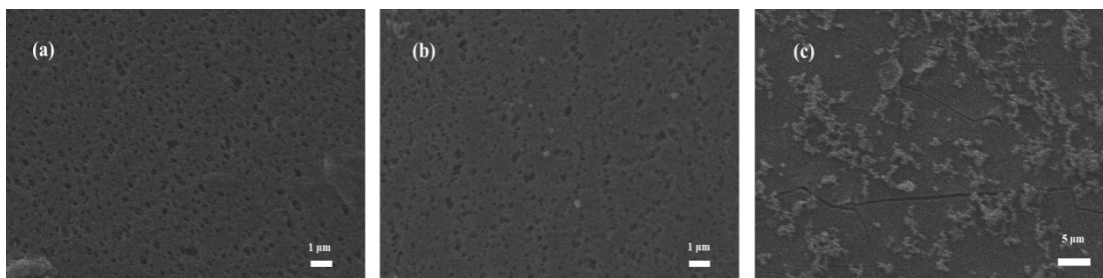


Fig. S3 Top-view images of Co gel (a), COF-TpPa-1/Co gel (b) and COF-TpPa-1 layer (c).

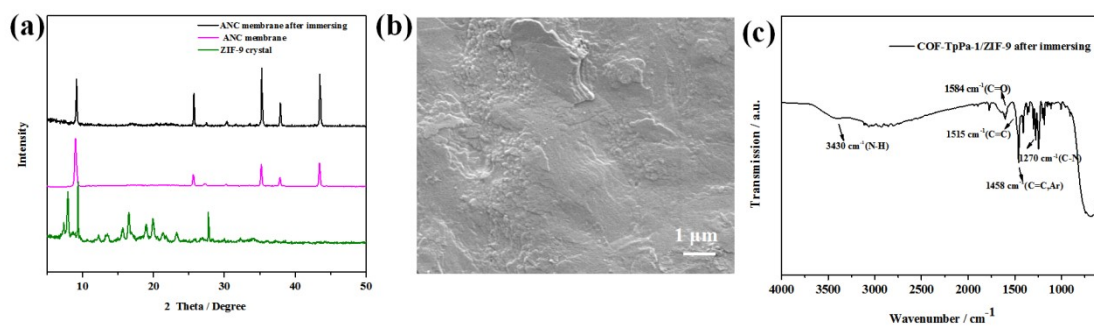


Fig. S4 PXRD pattern (a), SEM image (b) and FTIR (c) of the COF-TpPa-1/ZIF-9 ANC membranes after water treatments.

Table S1 Single gas permeance for α -Al₂O₃ substrate, COF-TpPa-1 and Co gel layer at 25 °C.

membrane	H ₂ permeance (GPU)	CO ₂ permeance (GPU)	H ₂ /CO ₂ selectivity
α -Al ₂ O ₃ substrate	5600.06±29.47	4568.50±29.50	1.23±0.005
COF-TpPa-1	5747.45±58.95	4981.14±83.36	1.15±0.003
Co gel	12.90±0.33	5.07±0.91	2.58±0.40

Table S2 Single gas permeance for **ZIF-9** and **COF-TpPa-1/ZIF-9** ANC membranes at 25 °C under the pressure of 1.6 bar.

membrane	H ₂ permeance (GPU)	CO ₂ permeance (GPU)	N ₂ permeance (GPU)	CH ₄ permeance (GPU)
ZIF-9	22.36±4.99	1.56±0.74	2.35±0.61	3.39±0.71
COF-TpPa-1/ZIF-9 ANC	551.16±25.02	72.06±6.46	79.29±10.84	150.91±7.92

Table S3 Single gas permeation properties of the **ZIF-9** membrane at 25°C and different trans-membrane pressure drops.

ZIF-9 membrane	1.2 bar	1.4 bar	1.6 bar	1.8 bar
H ₂ permeance (GPU)	23.67±4.67	21.88±4.07	22.36±4.99	26.32±6.38
CO ₂ permeance (GPU)	1.56±0.67	1.42±0.60	1.56±0.74	1.79±0.84

Table S4 Single gas permeation properties of the **COF-TpPa-1/ZIF-9** ANC membrane at 25°C and different trans-membrane pressure drops.

COF-TpPa-1/ZIF-9 ANC membrane	1.2 bar	1.4 bar	1.6 bar	1.8 bar
H ₂ permeance (GPU)	564.43 ± 31.26	529.06 ± 27.09	551.16 ± 25.02	571.80 ± 25.01
CO ₂ permeance (GPU)	77.51 ± 1.67	72.06 ± 1.88	72.06 ± 6.46	76.04 ± 4.59

Table S5 Single gas permeation properties of the **COF-TpPa-1/ZIF-9** ANC membrane at 25°C, 0.2 bar and low humidity conditions.

membrane	H ₂ permeance (GPU)	CO ₂ permeance (GPU)	H ₂ /CO ₂ selectivity
COF-TpPa-1/ZIF-9 ANC	453.7 ± 15.39	64.14 ± 8.03	7.08 ± 0.45