Supporting Information:

3D Printed MOF-based Mixed Matrix Thin-Film Composite Membranes

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Figure S1: Schematic of the electrospray system.
Figure S2: UV-VIS absorption spectra of HKUST-1-PIM-1 mixture in chloroform compared with the individual ingredients: PIM-1 and HKUST-1.
Figure S3: SEM images for HKUST-1 nanoparticles at different magnifications (a) 300nm, and (b) 100 nm.

Figure S4: PXRD patterns for HKUST-1 nanoparticles compared to the simulated HKUST-1 from the single crystal structure.
Figure S5: N$_2$ sorption isotherm of HKUST-1 nanoparticles collected at 77 K (BET=1869 m$^2$/g).
Figure S6: PXRD patterns for 0.5 wt% PIM-1, and 0.5 wt% PIM-1/HKUST-1 TFCs compared to the simulated HKUST-1 from the single crystal structure.

Figure S7: SEM surface images of the fabricated PIM-1 TFC membrane of composition of (a) 0.1 wt% and (b) 0.5 wt%.
**Figure S8:** SEM images of the fabricated PIM-1 TFC membrane (2 cycles) of composition of 0.5 wt% (a) surface image and (b) cross-sectional image.

**Figure S9:** Cross-sectional SEM image of the fabricated PIM-1 TFC membrane (5 cycles) of composition of 0.5 wt%.
**Figure S10:** SEM surface images of the fabricated TFC MMM of composition of 0.5 wt% (a) 5 µm and (b) 50 µm.

**Figure S11:** Cross-sectional SEM surface images of the fabricated TFC MMM of composition of 0.5 wt% (a) 20 µm, (b) 10 µm, and (c) 4 µm.
Table S1: Summary of the gas transport properties of the 3D printed TFC membranes (B# denotes to the batch number).

| Sample               | Thickness (µm) | wt% | Number of cycles | CO₂ Permeance (GPU) | N₂ Permeance (GPU) | CO₂/N₂ Selectivity |
|----------------------|----------------|-----|------------------|---------------------|--------------------|---------------------|
| PIM-1                | 0.39           | 0.1 | 5                | 147                 | 57                 | 2.6                 |
| PIM-1/ HKUST-1       | 0.5            | 0.1 | 5                | 305                 | 153                | 2                   |
| PIM-1 (B#1)          | 1.8            | 0.5 | 5                | 271                 | 21                 | 13                  |
| PIM-1 (B#2)          | 2.75           | 0.5 | 5                | 159                 | 13                 | 12                  |
| PIM-1/ HKUST-1 (B#1) | 2.85           | 0.5 | 5                | 721                 | 103                | 7.0                 |
| PIM-1/ HKUST-1 (B#2) | 2.85           | 0.5 | 5                | 638                 | 116                | 5.5                 |
| PIM1-HKUST-1 (B#3)   | 2.65           | 0.5 | 5                | 730                 | 107                | 6.8                 |