# Supporting Information

# "All Polyimide" Mixed Matrix Membranes for High Performance Gas Separation

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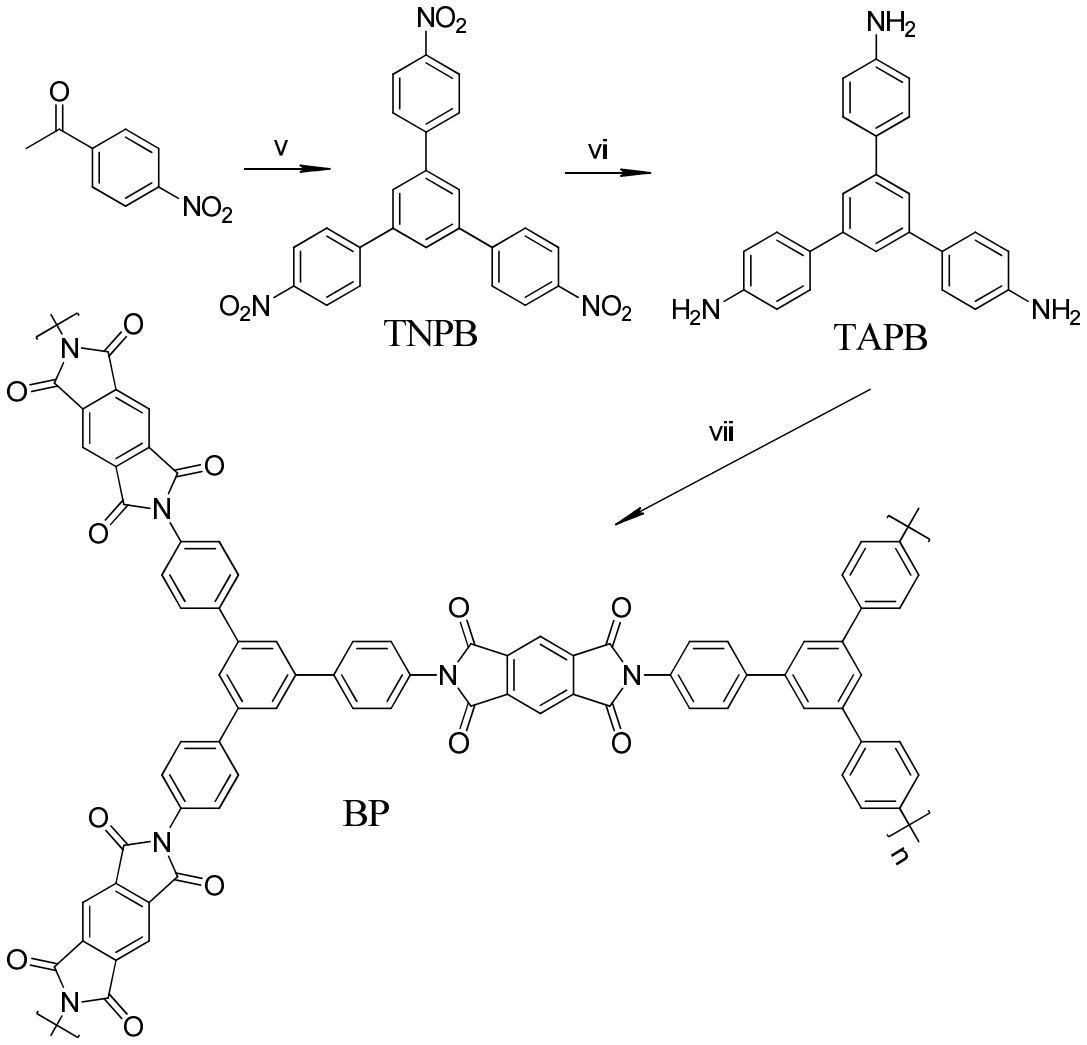
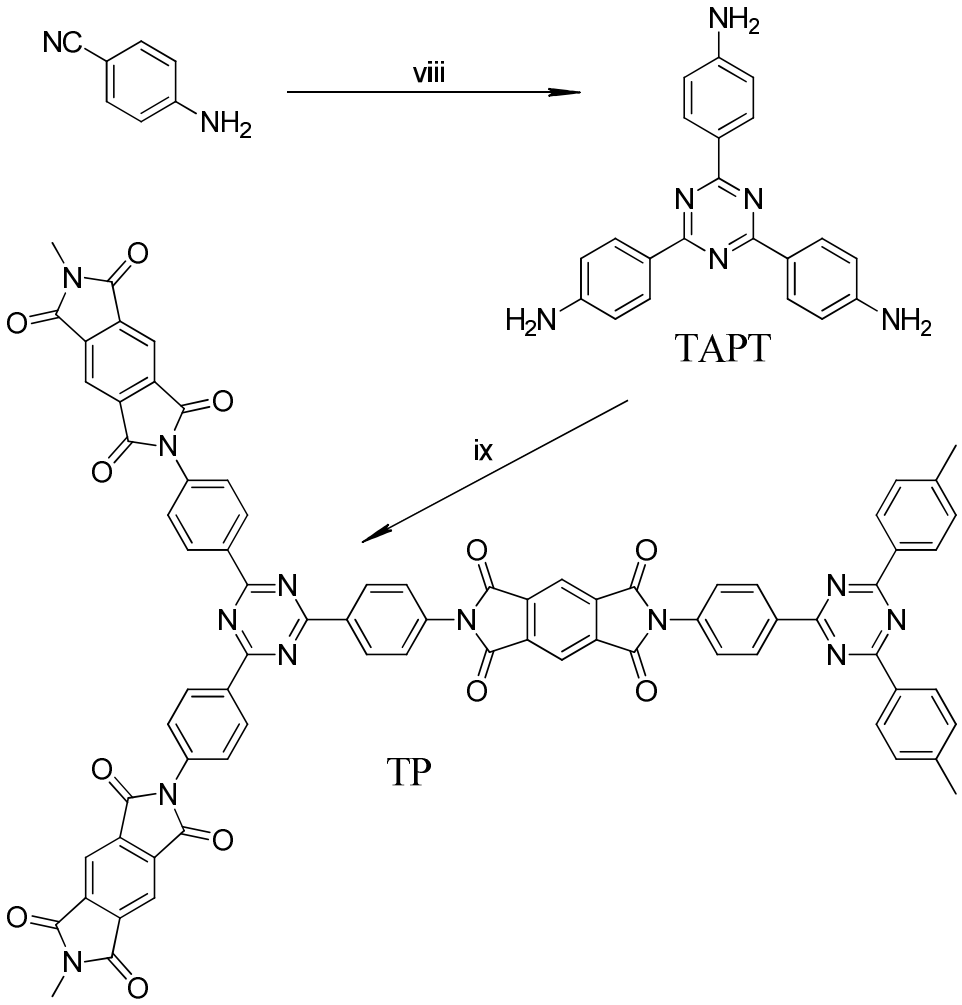



**Scheme S1.** Synthetic procedure and structures of TAPB and BP. Reagents and conditions: (v) thionyl chloride and ethanol reflux for 4 h; (vi) $H_2N$-$NH_2$·$H_2O$, Pd/C and ethanol reflux for 24 h; (vii) isoquinoline, NMP and mesitylene mix solvent, heating schedule.




**Scheme S2.** Synthetic procedure and structures of TAPT and TP. Reagents and conditions: (viii) trifluoromethanesulfonic acid and 4-aminobenzonitrile reaction in ice bath for 12 h; (ix) isoquinoline, NMP and mesitylene mix solvent, heating schedule.

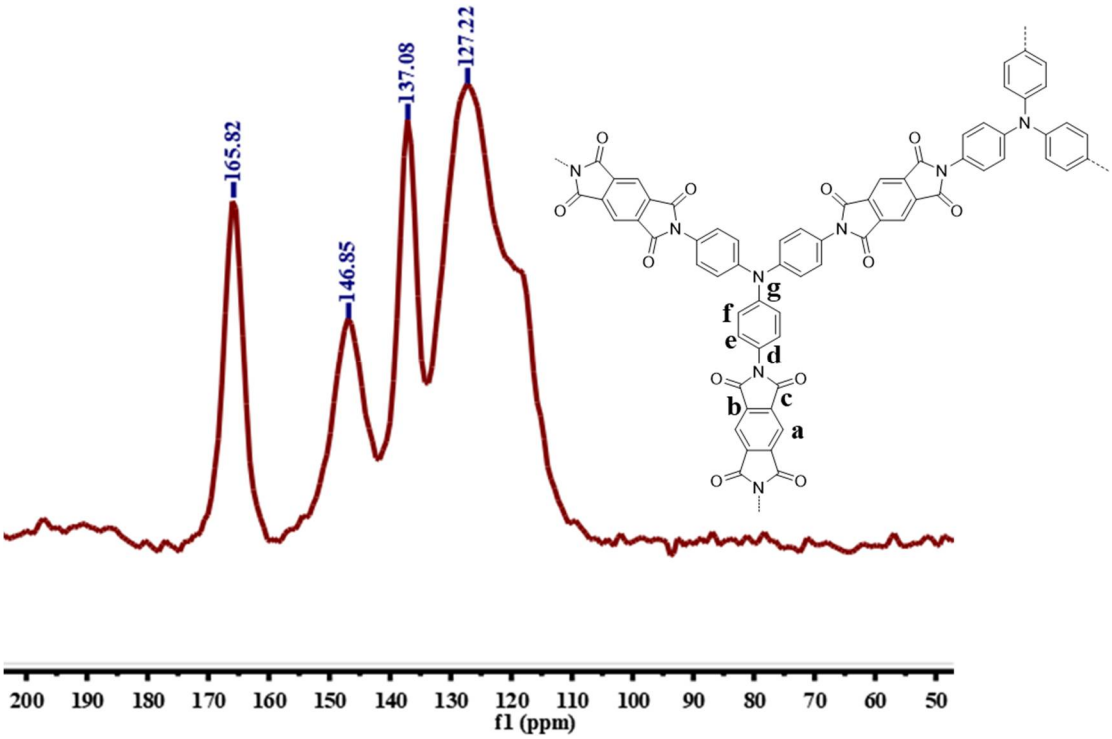



**Figure S1.** $^{13}$C CP/MAS NMR spectra of AP.

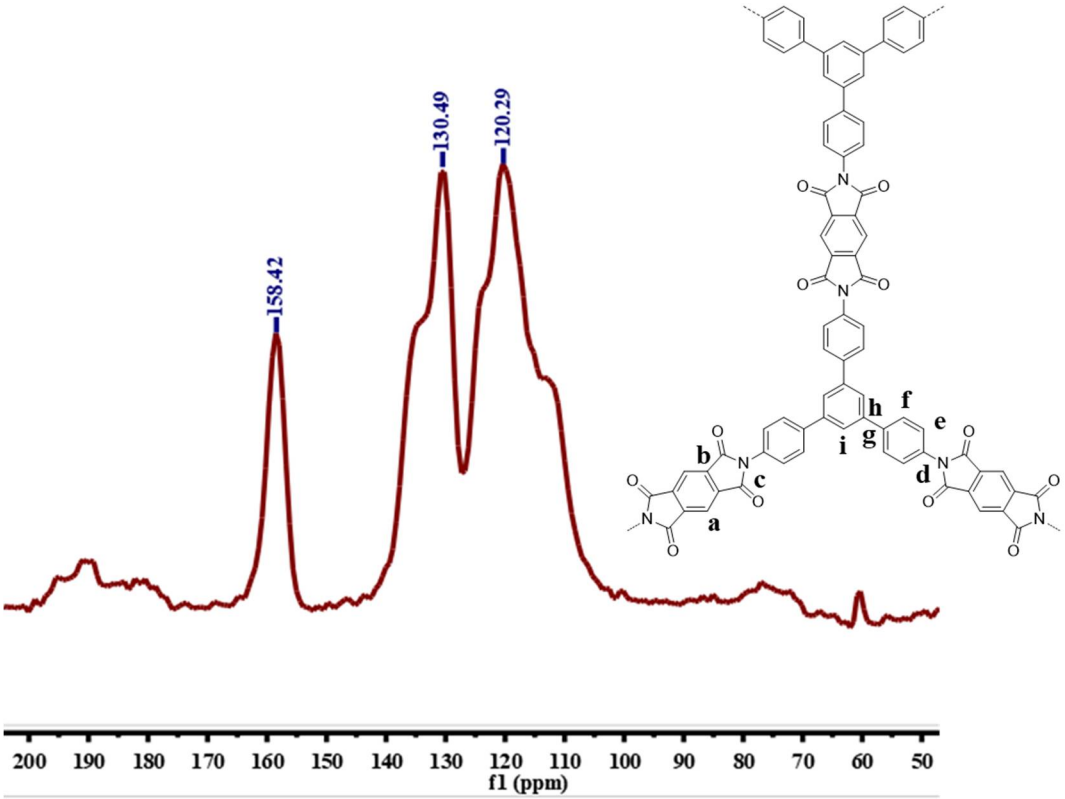



**Figure S2.** $^{13}$C CP/MAS NMR spectra of BP.

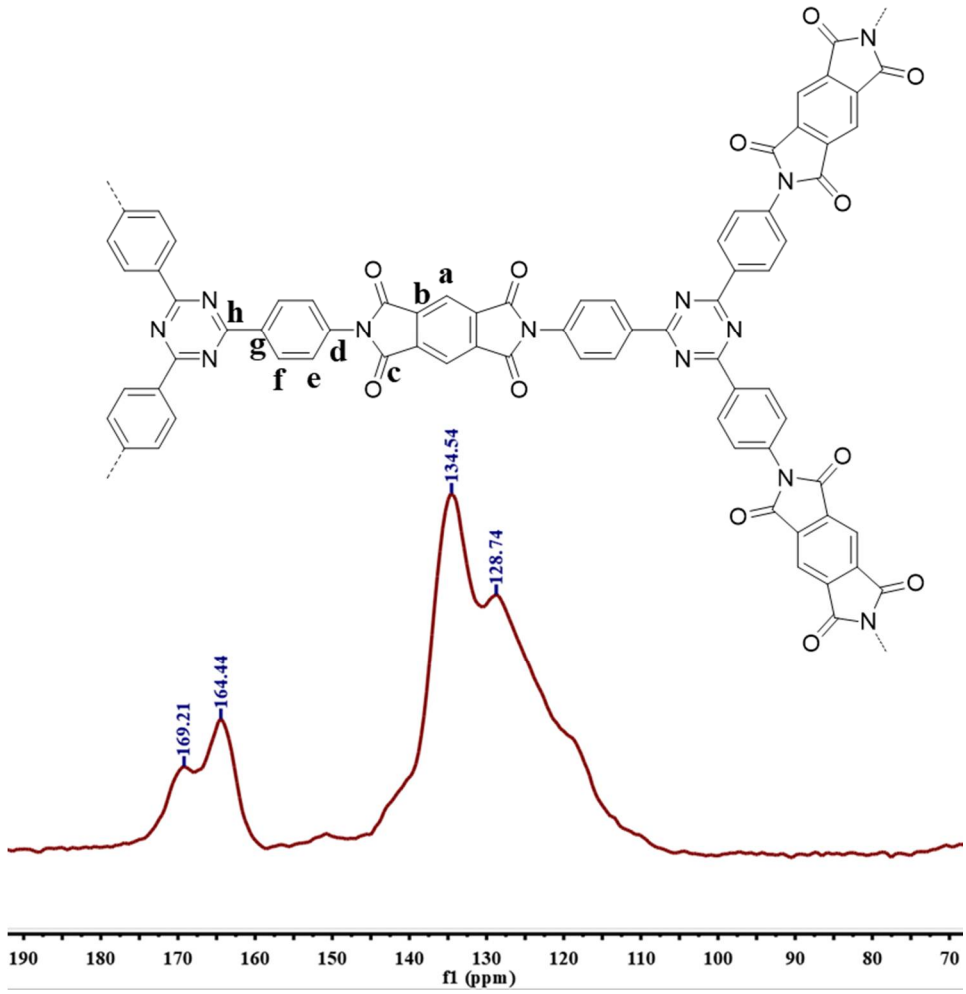



**Figure S3.** $^{13}C$ CP/MAS NMR spectra of TP.

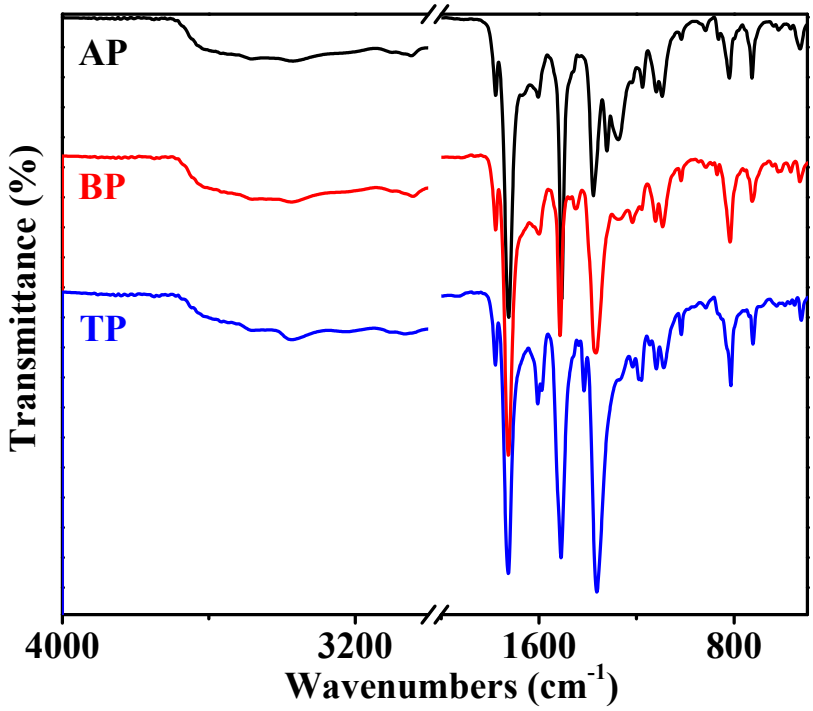


**Figure S4.** FTIR spectra of AP, BP and TP.

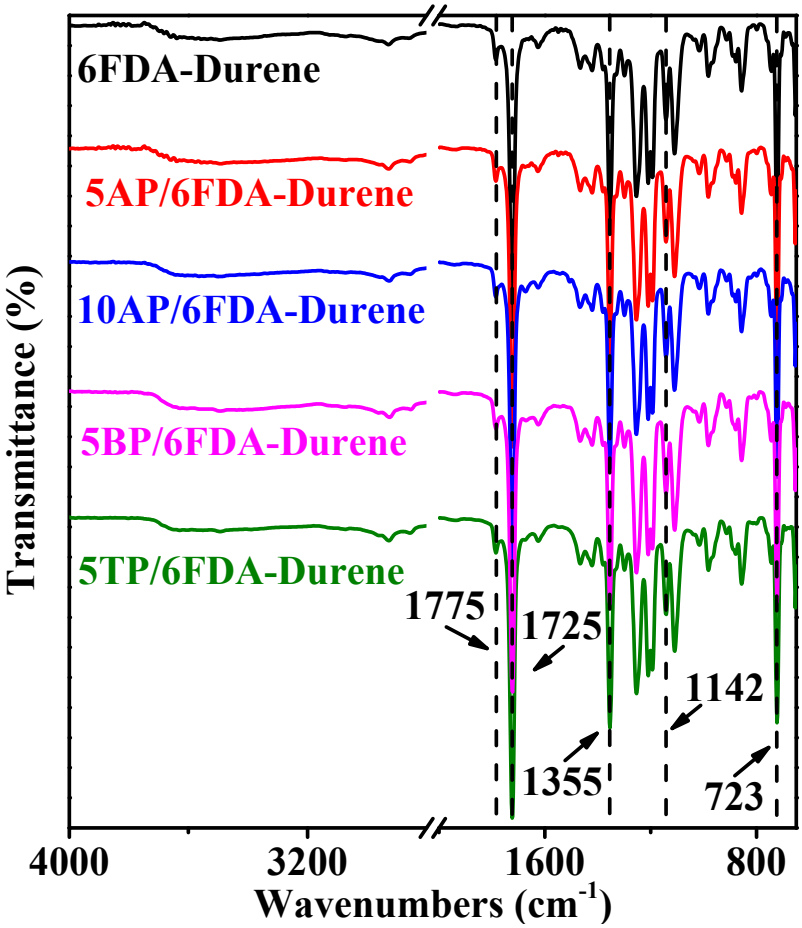



**Figure S5.** ATR-FTIR spectra of 6FDA-Durene and "all polyimide" MMMs.

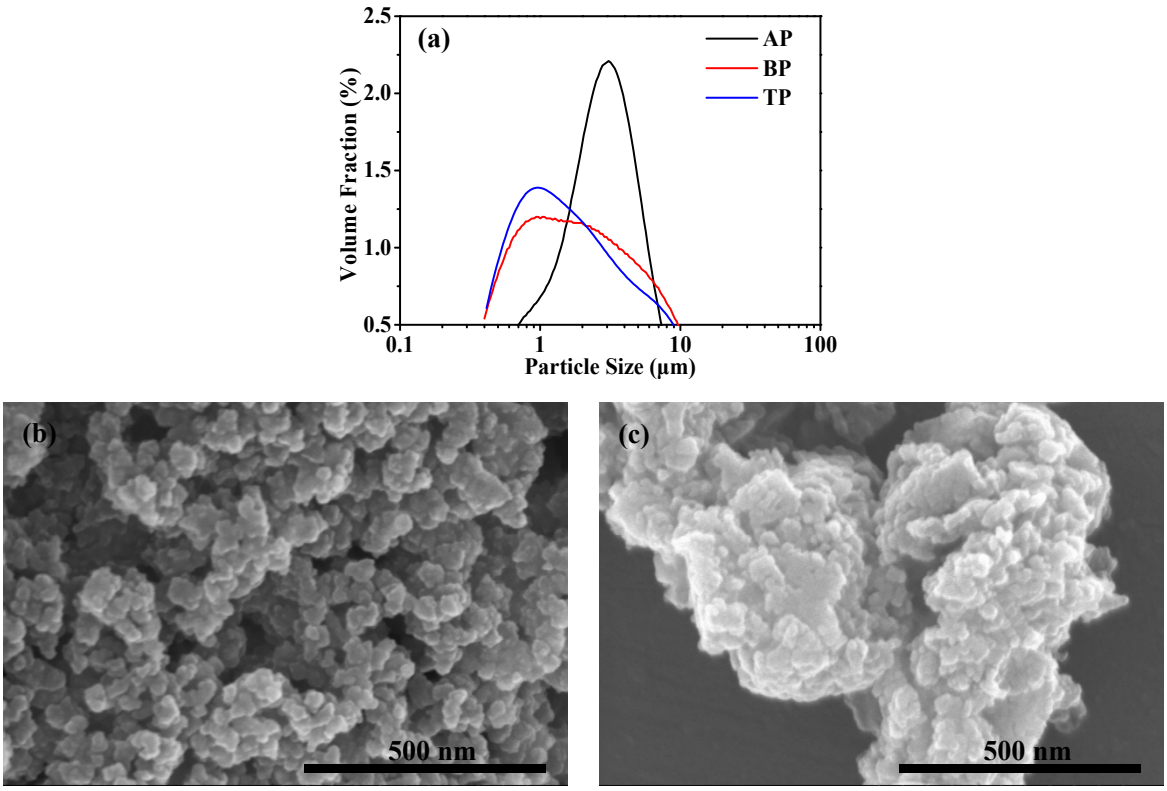



**Figure S6. (a)** DLS of AP, BP and TP; **(b)** SEM of BP; **(c)** SEM of TP .

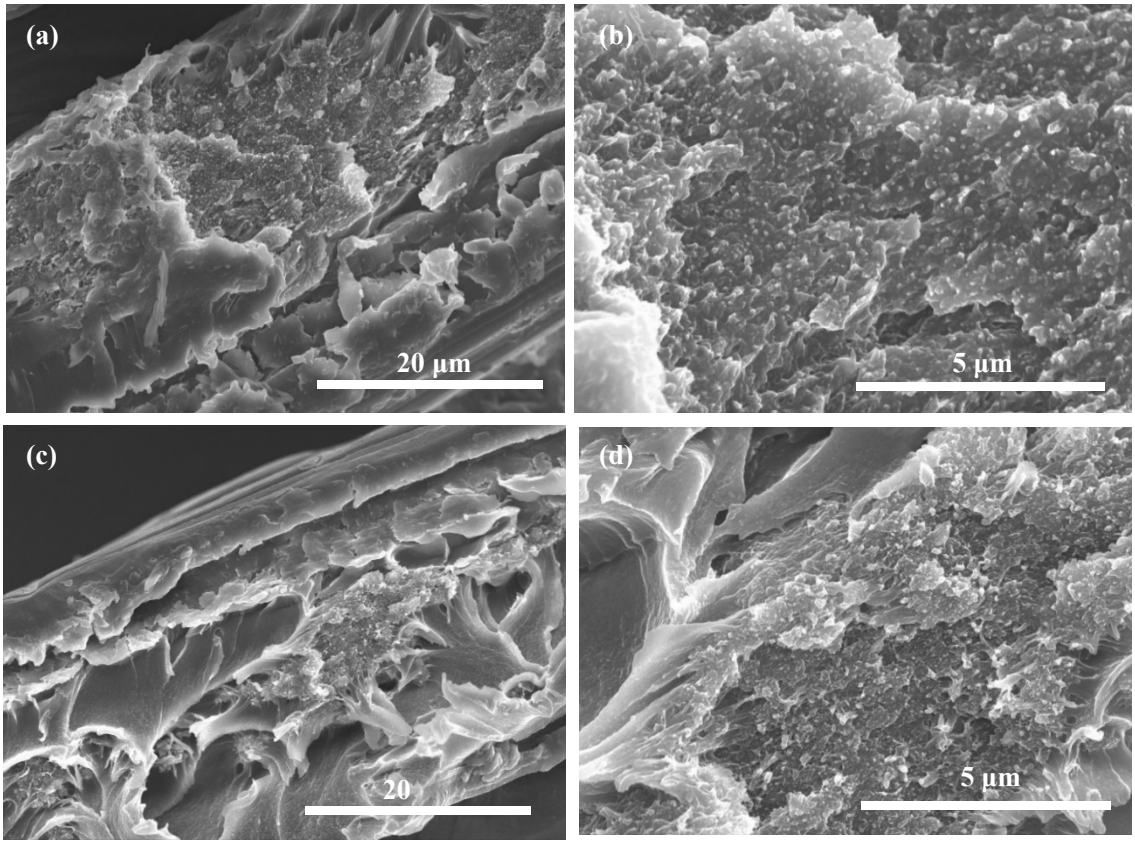



**Figure S7.** Cross-section SEM images of **(a, b)** 5BP/6FDA-Durene and amplification; **(c, d)** 5TP/6FDA-Durene and amplification

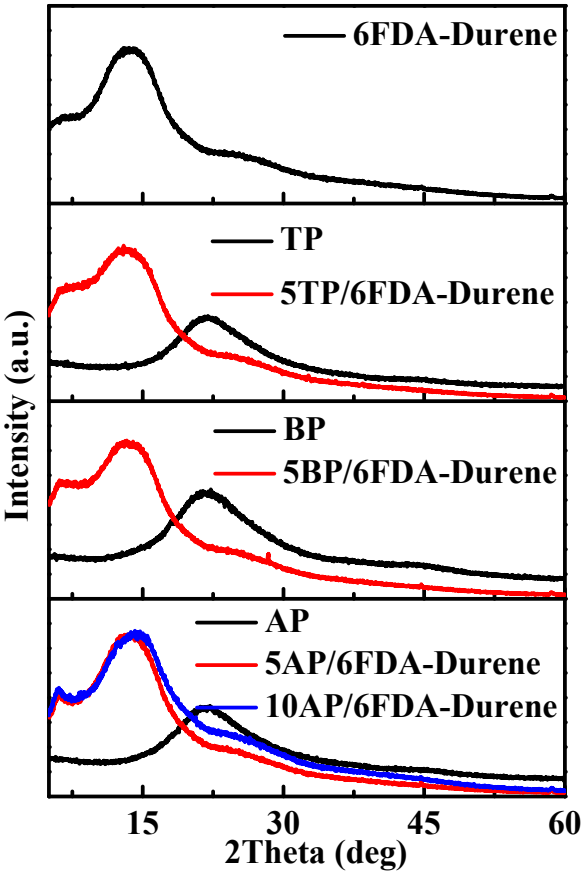



**Figure S8.** XRD patterns of microporous PIs, 6FDA-Durene and "all polyimide" MMMs.

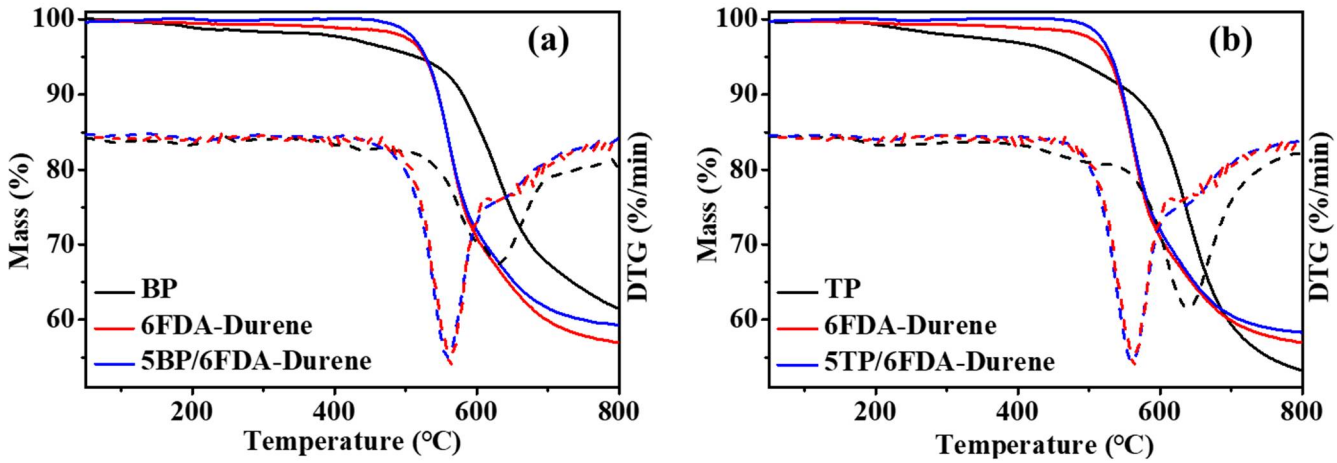



**Figure S9.** TGA (solid line) and DTG (dotted line) of **(a)** BP and the MMMs with BP loading; **(b)** TP and the MMMs with TP loading

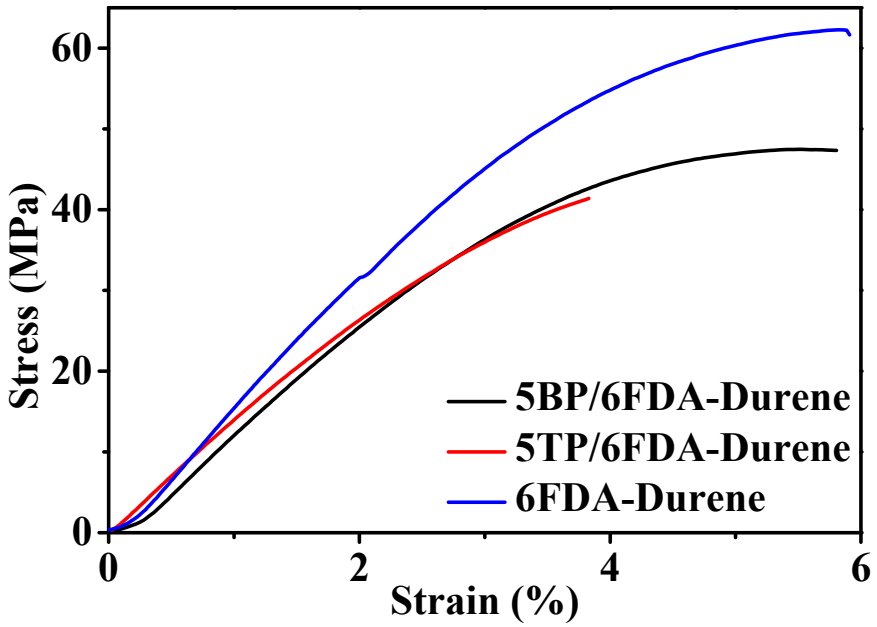



**Figure S10.** stress-strain curves of 5BP/6FDA-Durene, 5TP/6FDA-Durene and 6FDA-Durene

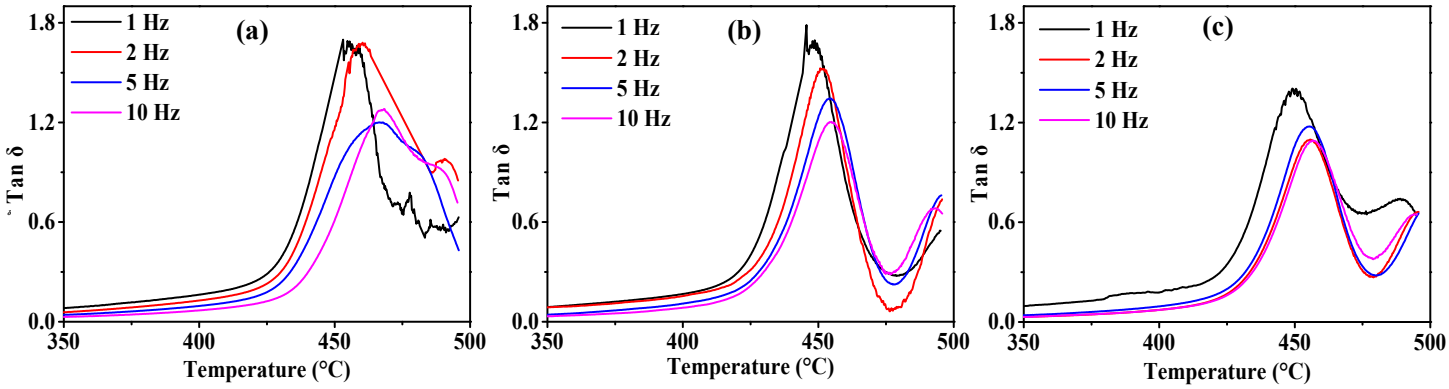



**Figure S11.** DMA curves of **(a)** 6FDA-Durene; **(b)** 5AP/6FDA-Durene; **(c)** 10AP/6FDA-Durene at 1 Hz, 2 Hz, 5 Hz, 10 Hz respectively

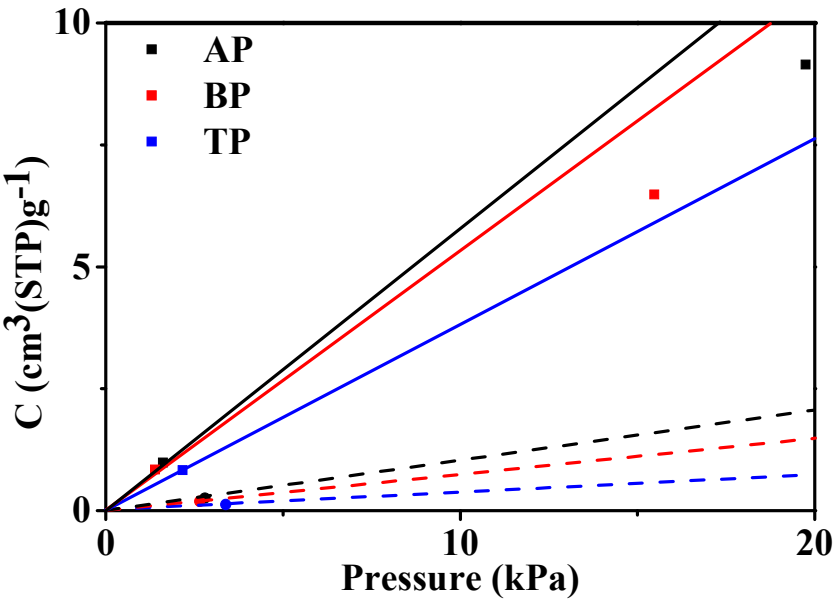



**Figure S12.** Henry fit for idea adsorption solution theory (IAST) of AP, BP and TP.

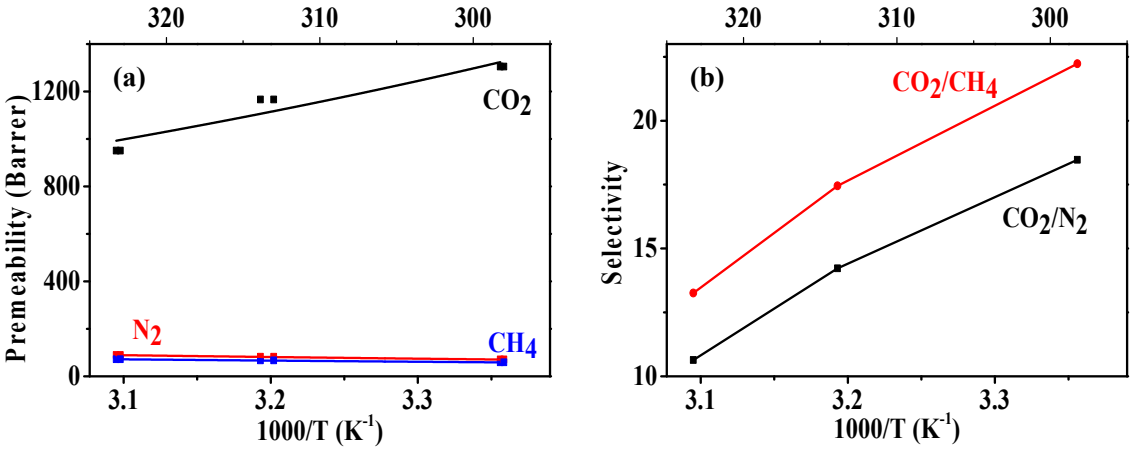



**Figure S13.** (a) Gas permeability and (b) ideal selectivity of 5AP/6FDA-Durene at 100 psi in different temperature.

**Table S1.** Assignments for **Figure. S1-S3**

| | a | b | c | d | e | f | g | h | i |
|---|---|---|---|---|---|---|---|---|---|
| AP | 125.2 | 137.1 | 165.8 | 127.2 | 122.5 | 134.2 | 146.9 | | |
| BP | 125.2 | 135.5 | 158.4 | 130.5 | 130.5 | 128.1 | 136.4 | 137.5 | 120.3 |
| TP | 125.2 | 134.5 | 164.4 | 132.7 | 128.7 | 127.7 | 130.3 | 169.2 | |

**Table S2.** The ratio of $P_{CO_2}$ of PI base MMMs to pristine PI membranes

| Matrix | Filler | Filler content (wt%) | $P_{CO_2}$ (Barrer) | $P_{CO_2}$ increment Ratio | Ref |
|---|---|---|---|---|---|
| 6FDA- | / | | 500.17 | 1 | This work |

| | | | | | |
|---|---|---|---|---|---|
| Durene | AP | 5 | 1291.13 | 2.58 | |
| | AP | 10 | 959.16 | 1.92 | |
| | BP | 5 | 1047.43 | 2.09 | |
| | TP | 5 | 753.07 | 1.51 | |
| | / | 0 | 805 | 1 | |
| 6FDA- | ZIF-71 (30 nm) | 21.2 | 2560 | 3.18 | [1] |
| Durene | ZIF-71 (200 nm) | 21.1 | 2744 | 3.41 | |
| | ZIF-71 (600 nm) | 19.3 | 1656 | 2.06 | |
| | / | 0 | 625 | 1 | |
| | NH2-MIL-101 | 10 | 1365 | 2.18 | |
| | CNT | 5 | 730 | 1.17 | |
| 6FDA- | CNT | 10 | 1100 | 1.76 | [2] |
| Durene | CNT | 15 | 1700 | 2.72 | |
| | CNT-MIL | 5 | 800 | 1.28 | |
| | CNT-MIL | 10 | 1100 | 1.76 | |
| | CNT-MIL | 15 | 1625 | 2.60 | |
| | / | 0 | 800 | 1 | |
| 6FDA- | HKUST-1 | 10 | 1300 | 1.63 | [3] |
| Durene | un-HKUST-IL | 10 | 400 | 0.5 | |
| | HKUST-IL | 10 | 1100 | 1.38 | |
| | / | 0 | 1280 | 1 | |
| | PSM-MOF | 2.5 | 1310 | 1.02 | |
| | PSM-MOF | 5 | 1340 | 1.05 | |
| | PSM-MOF | 7.5 | 1370 | 1.07 | |
| | PSM-MOF | 10 | 1400 | 1.09 | |
| | PSM-MOF | 15 | 1470 | 1.15 | |
| 6FDA- | PSM-MOF | 20 | 1540 | 1.20 | [4] |
| Durene | PSM-MOF | 25 | 1640 | 1.28 | |
| | PSM-MOF | 30 | 1710 | 1.34 | |
| | PSM-MOF | 35 | 1810 | 1.41 | |
| | PSM-MOF | 40 | 1890 | 1.48 | |
| | PSM-MOF | 100 | 3515 | 2.75 | |
| | UiO-66-NH2 | 15 | 1990 | 1.55 | |
| | UiO-66-NH2 | 30 | 3260 | 2.55 | |
| 6FDA- | / | 0 | 1143 | 1 | [5] |
| Durene | ZIF-71 | 10 | 2049 | 1.79 | |
| | / | 0 | 300 | 1 | |
| 6FDA- | PgC5Cu | 1 | 310 | 1.03 | [6] |
| Durene | PgC5Cu | 2 | 360 | 1.2 | |
| | PgC5Cu | 3 | 410 | 1.37 | |
| | / | 0 | 455.2 | 1 | |
| 6FDA- | NaX | 5 | 620 | 1.36 | [7] |
| Durene | NaX | 10 | 780 | 1.71 | |
| | NaX | 15 | 891.3 | 1.96 | |

| | | | | | |
|---|---|---|---|---|---|
| | NaX | 20 | 1000 | 2.20 | |
| | [Cu(6L)]2+@13X | 5 | 891.3 | 1.96 | |
| | [Cu(6L)]2+@13X | 10 | 960 | 2.11 | |
| | [Cu(6L)]2+@13X | 15 | 1034.1 | 2.27 | |
| | [Cu(6L)]2+@13X | 20 | 1060 | 2.33 | |
| 6FDA-Durene | / | 0 | 626 | 1 | [8] |
| | Ni(dobdc) | 21 | 1035 | 1.65 | |
| 6FDA-Durene | / | 0 | 959 | 1 | [9] |
| | ZIF-71 | 10 | 1539 | 1.60 | |
| | / | 0 | 669.12 | 1 | |
| | ZIF-67 | 10 | 779.51 | 1.16 | |
| | ZIF-67 | 20 | 1529.86 | 2.29 | |
| | ZIF-67-IL1 | 10 | 779.01 | 1.16 | |
| 6FDA-Durene | ZIF-67-IL1 | 20 | 1254.64 | 1.88 | [10] |
| | ZIF-67-IL2 | 10 | 1030.96 | 1.54 | |
| | ZIF-67-IL2 | 20 | 1426.12 | 2.13 | |
| | ZIF-67-IL3 | 10 | 672.79 | 1.01 | |
| | ZIF-67-IL3 | 20 | 889.87 | 1.33 | |
| | / | 0 | 460 | 1 | |
| | Zeolite T | 0.5 | 700 | 1.52 | |
| | Zeolite T | 1 | 843.6 | 1.83 | |
| 6FDA-Durene | Zeolite T | 3 | 700 | 1.52 | [11] |
| | Zeolite T | 5 | 620 | 1.35 | |
| | Zeolite T | 7 | 390 | 0.85 | |
| | Zeolite T | 10 | 380 | 0.83 | |
| | Zeolite T | 15 | 370 | 0.80 | |
| | / | 0 | 546 | 1 | |
| 6FDA-DAM | nano Mg2(dobpdc) | 23 | 782 | 1.43 | [12] |
| | nano mmen-Mg2(dobpdc) | 23 | 1056 | 1.93 | |
| | / | 0 | 444 | 1 | |
| 6FDA-DAM | PPN | 15 | 595 | 1.34 | [13] |
| | PPN | 30 | 710 | 1.60 | |
| | / | 0 | 970 | 1 | |
| | HKUST-1 | 10 | 1450 | 1.49 | |
| 6FDA-DAM | HKUST-1 | 20 | 1690 | 1.74 | |
| | HKUST-1 | 30 | 2360 | 2.43 | [14] |
| | branch HKUST-1 | 10 | 1600 | 1.65 | |
| | branch HKUST-1 | 20 | 1920 | 1.98 | |
| | branch HKUST-1 | 30 | 2480 | 2.56 | |
| | / | 0 | 420 | 1 | |
| 6FDA-DAM | PgC5Cu | 1 | 400 | 0.95 | [6] |
| | PgC5Cu | 2 | 580 | 1.38 | |

| | | | | | |
|---|---|---|---|---|---|
| | PgC5Cu | 3 | 480 | 1.14 | |
| 6FDA-DAM | / | 0 | 590 | 1 | [15] |
| | ns-CuBDC | 2 | 570 | 0.97 | |
| | ns-CuBDC | 4 | 430 | 0.73 | |
| 6FDA-DAM | / | 0 | 518 | 1 | [8] |
| | Ni(dobdc) | 23 | 715 | 1.38 | |
| 6FDA-DAM | / | 0 | 570 | 1 | [16] |
| | G1NP | 25 | 1050 | 1.84 | |
| | G2NP | 25 | 1920 | 3.37 | |
| 6FDA-DAM | / | 0 | 1000 | 1 | [17] |
| | ZMOF | 20 | 1600 | 1.6 | |
| 6FDA-DAM | / | 0 | 540 | 1 | [18] |
| | 3D-COF | 10 | 850 | 1.57 | |
| | 3D-COF | 15 | 1280 | 2.37 | |
| | 2D-COF | 10 | 640 | 1.19 | |
| 6FDA-DAM | / | 0 | 380 | 1 | [19] |
| | NH2-MIL-53(Al) (NP) | 5 | 440 | 1.16 | |
| | NH2-MIL-53(Al) (NP) | 10 | 480 | 1.26 | |
| | NH2-MIL-53(Al) (NP) | 15 | 600 | 1.58 | |
| | NH2-MIL-53(Al) (NP) | 20 | 700 | 1.84 | |
| Matrimid | / | 0 | 8.2 | 1 | |
| | NH2-MIL-53(Al) (NP) | 8 | 9.5 | 1.16 | |
| | NH2-MIL-53(Al) (NP) | 16 | 7.8 | 0.95 | |
| | NH2-MIL-53(Al) (NR) | 8 | 8 | 0.98 | |
| | NH2-MIL-53(Al) (NP) | 16 | 7.8 | 0.95 | |
| | NH2-MIL-53(Al) (MN) | 8 | 8 | 0.98 | |
| | NH2-MIL-53(Al) (MN) | 16 | 6.5 | 0.79 | |
| Matrimid | / | | 6.82 | 1 | [20] |
| | MMIF | 10 | 8.1 | 1.19 | |
| | MMIF | 20 | 8.65 | 1.27 | |
| matrimid | / | 0 | 8.6 | 1 | [13] |
| | PPN | 15 | 20 | 2.33 | |
| | PPN | 30 | 61 | 7.09 | |
| matrimid | / | 0 | 9.55 | 1 | [8] |

| | | | | | |
|---|---|---|---|---|---|
| | Ni(dobdc) | 23 | 9.31 | 0.97 | |
| | / | 0 | 8 | 1 | |
| | UiO-66-NH2 | 12 | 18 | 2.25 | |
| | UiO-66-NH2 | 23 | 24 | 3 | |
| | UiO-66-NH2 | 40 | 27.5 | 3.44 | |
| matrimid | UiO-66-NH2-Ipa | 12 | 20 | 2.5 | [21] |
| | UiO-66-NH2-Ipa | 23 | 29 | 3.63 | |
| | UiO-66-NH2-Ipa | 40 | 32 | 4 | |
| | UiO-66-NH2-Ic | 23 | 22.5 | 2.81 | |
| | UiO-66-NH2-Isa | 23 | 20 | 2.5 | |
| | / | 0 | 10 | 1 | |
| | CMS | 17 | 10.3 | 1.03 | |
| Matrimid | CMS | 19 | 10.6 | 1.06 | [22] |
| | CMS | 33 | 11.5 | 1.15 | |
| | CMS | 36 | 12.6 | 1.26 | |
| | / | 0 | 7.6 | 1 | |
| | PPI-1 | 20 | 10.3 | 1.36 | |
| matrimid | PPI-2 | 10 | 11.3 | 1.49 | [23] |
| | PPI-2 | 20 | 16.8 | 2.21 | |
| | PPI-3 | 20 | 20.9 | 2.75 | |
| | / | 0 | 5.78 | 1 | |
| | b-CuBDC | 7.9 | 5.21 | 0.90 | |
| matrimid | ns-CuBDC | 7.9 | 4.09 | 0.70 | [24] |
| | ns-CuBDC | 8.3 | 5.03 | 0.87 | |
| | ns-CuBDC | 1.7 | 5.38 | 0.93 | |
| | ns-Cu(2,6-NDC) | 7.6 | 6.28 | 1.09 | |
| | / | 0 | 20 | 1 | |
| 6FDA-ODA | Cd-6F | 10 | 38 | 1.9 | [25] |
| | Cd-6F | 10 | 30 | 1.5 | |
| | Cd-6F | 10 | 41 | 2.05 | |
| | / | 0 | 71 | 1 | |
| 6FDA-6FpDA | PPN | 15 | 131 | 1.85 | [13] |
| | PPN | 30 | 190 | 2.68 | |
| | / | 0 | 285 | 1 | |
| | ZIF-8 | 7 | 560 | 1.96 | |
| | ZIF-8 | 20 | 896 | 3.14 | |
| TB-polyimide | ZIF-8 | 30 | 1437 | 5.04 | [26] |
| | PDZIF-8 | 7 | 380 | 1.33 | |
| | PDZIF-8 | 20 | 702 | 2.46 | |
| | PDZIF-8 | 30 | 1056 | 3.71 | |
| | / | 0 | 55.8 | 1 | |
| 6FDA-DAT | Ni(dobdc) | 15 | 63.9 | 1.15 | [8] |
| | / | 0 | 9 | 1 | |
| 6FBI | ZIF-8 | 10 | 11.8 | 1.31 | [27] |

| | | | | | |
|---|---|---|---|---|---|
| Ulten | ZIF-8 | 20 | 20.3 | 2.26 | [22] |
| | ZIF-8 | 25 | 25.4 | 2.82 | |
| | ZIF-8 | 30 | 50.9 | 5.65 | |
| | / | 0 | 1.45 | 1 | |
| | CMS | 16 | 2.51 | 1.73 | |
| | CMS | 20 | 2.9 | 2 | |
| | CMS | 35 | 4.48 | 3.09 | |
| 6FDA-IMM | / | 0 | 224 | 1 | [23] |
| | PPI-1 | 20 | 202 | 0.90 | |
| | PPI-2 | 20 | 270 | 1.21 | |
| | PPI-3 | 20 | 289 | 1.29 | |

**Table S3.** Henry's law region fit for IAST of microporous PIs.

| | Initial slope ($CO_2$) | Initial slope ($CH_4$) | selectivity |
|---|---|---|---|
| AP | 0.335 | 0.083 | 4.02 |
| BP | 0.309 | 0.068 | 4.53 |
| TP | 0.196 | 0.036 | 5.43 |

**Table S4.** 5AP/6FDA-Durene activation energy ($E_P$) of permeation for $CO_2$, $N_2$ and $CH_4$

| | $E_P$ (kJ·mol$^{-1}$) | | |
|---|---|---|---|
| | $CO_2$ | $N_2$ | $CH_4$ |
| 5AP | -9.2 | 7.5 | 6.4 |

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