**Supporting Information**

**Ordered Mesoporous Carbon with Enhanced Porosity to Support Organic Amine: Efficient Nanocomposites for the Selective Capture of CO$_2$**

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Gas adsorption

Before measurement, the samples were pretreated at 180 °C in a $N_2$ flow for 12 h.
The $CO_2$ and $N_2$ adsorption isotherms at 0, 30, 50 and 75 °C were measured using the
Micromeritics Tristar II 3020 system. Correspondingly, adsorption selectivity in cases
of $CO_2/N_2$ were calculated according to the Ideal Adsorption Solution Theory (IAST).

**Table S1** The element (C, N and O) content of prepared samples.

| Samples | Contents (wt.%) [a] | C (wt %) [b] | N (wt %) | O (wt %) |
|---|---|---|---|---|
| OMC | --- | 93.36 | --- | 6.64 |
| 0.29PEI@OMC | 28.2 | 80.35 | 11.29 | 4.23 |
| AOMC | --- | 92.13 | 2.85 | 5.02 |
| 0.26PEI@AOMC | 25.7 | 80.06 | 12.21 | 4.08 |
| 0.44PEI@ AOMC | 44.4 | 72.05 | 18.56 | 3.01 |
| 0.52PEI@ AOMC | 51.6 | 67.89 | 21.58 | 2.47 |
| 0.60PEI@ AOMC | 60.2 | 64.53 | 24.67 | 2.03 |

[a] The loading contents of PEI estimated by the thermogravimetric analysis. [b] Determined from elemental analysis.

**Table S2** A comparison of $CO_2$ adsorption capacities of reported porous materials.

| Samples | Temperature (°C) | Pressure (bar) | $CO_2$ capacities (mmol/g) | Refs. |
|---|---|---|---|---|
| AOMC | 30 | 0.15 | 0.52 | This work |
| 0.44PEI@AOMC | 30 | 0.15 | 0.72 | This work |
| 0.52PEI@AOMC | 30 | 0.15 | 0.69 | This work |
| COP-19 | 25 | 0.15 | 0.40 | S1 |
| Azo-COP-2 | 25 | 0.15 | 0.41 | S2 |
| PPN-101 | 25 | 0.15 | 0.39 | S3 |
| MAPOP-4 | 25 | 0.15 | 0.45 | S4 |
| PCP-BF$_4$ | 25 | 0.15 | 0.34 | S5 |
| DA-CMP-1 | 25 | 0.15 | 0.30 | S6 |
| TNCMP-2 | 25 | 0.15 | 0.30 | S7 |
| TBMID | 25 | 0.15 | 0.50 | S8 |

**Table S3** A comparison of $CO_2/N_2$ selectivities of porous materials reported in the literature.

| Porous materials | Temperature (°C) | Method | $CO_2/N_2$ selectivities | Refs. |
|---|---|---|---|---|
| AOMC | 30 | IAST[a] | 43 | This work |
| 0.44PEI@AOMC | 30 | IAST[a] | 58 | This work |
| 0.52PEI@AOMC | 30 | IAST[a] | 64 | This work |
| PFPOP-3 | 25 | IAST | 57 | S9 |
| DA-CMP-1 | 25 | IAST | 60 | S6 |
| SNW-1 | 25 | IAST | 50 | S10 |
| Mg-MOF-74 | 30 | IAST | 44 | S11 |
| Cu-BTC | 10 | IAST | 34 | S12 |
| MIL-101 | 25 | IAST | 12 | S13 |

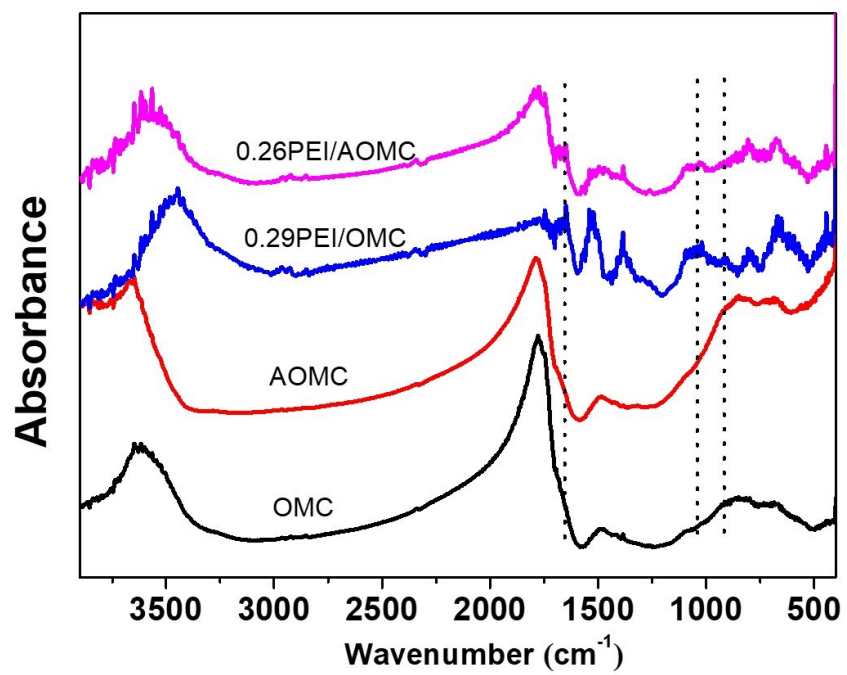[a] IAST CO2/N2 (0.1:0.9 v/v) selectivity at 1 bar over prepared samples.

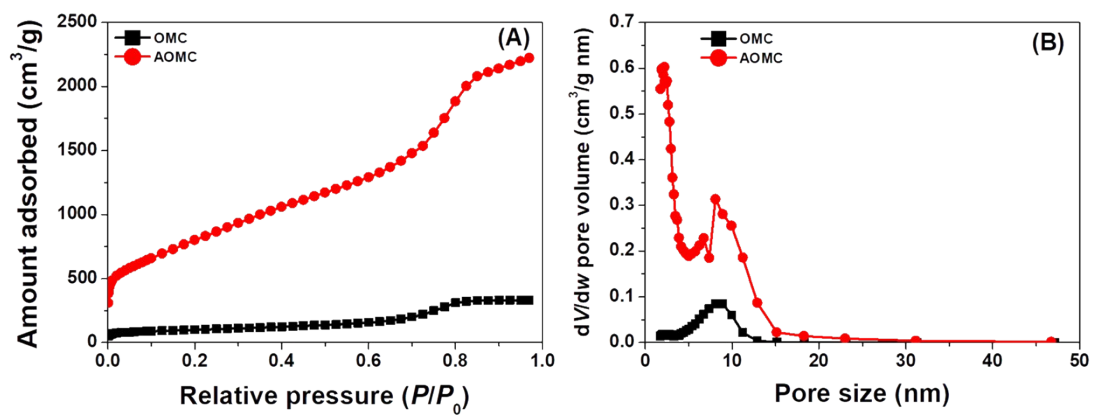**Figure S1** FTIR spectrum of prepared samples.

**Figure S2** $N_2$ adsorption isotherms at -196 ℃ (A) and BJH pore size distributions (B) of OMC and AOMC.
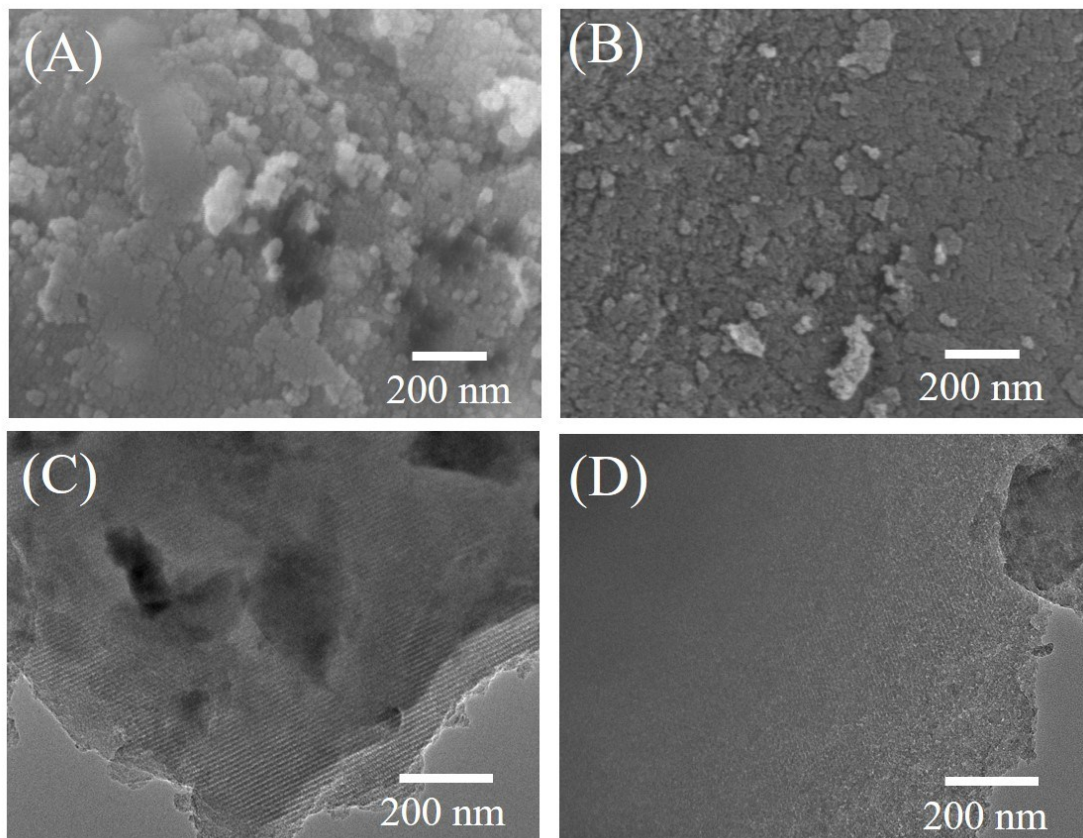
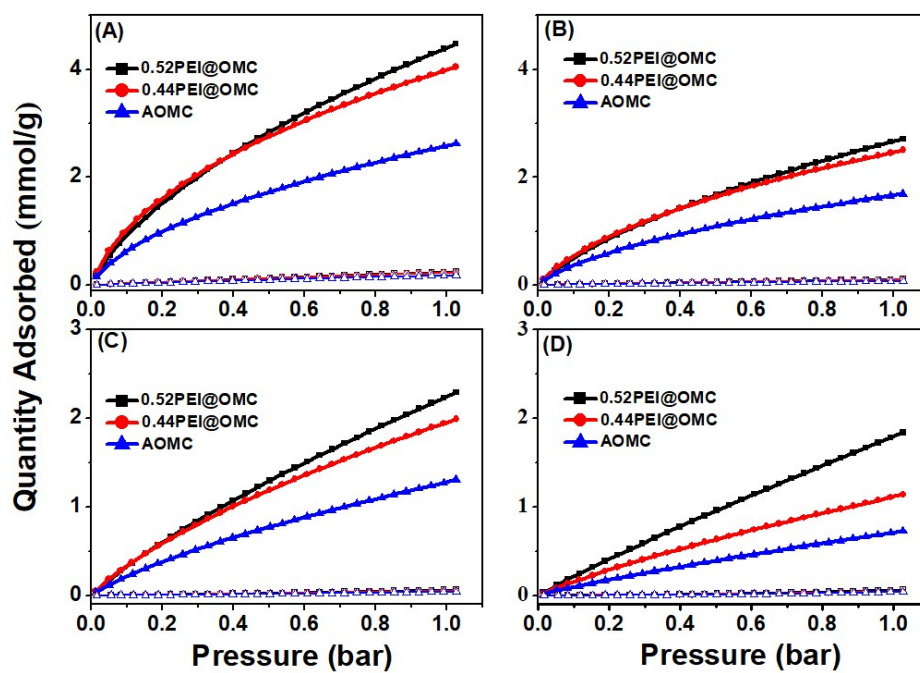**Figure S3** High resolved SEM and TEM images of pristine (A,C) 0.29PEI@OMC and (B,D) 0.26PEI@AOMC samples.

**Figure S4** $CO_2$ (solid symbols) and $N_2$ (hollow symbols) adsorption isotherms of prepared samples at (A) 0 °C, (B) 30 °C, (C) 50 °C and (D) 75 °C.

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