

Supporting Information

**High-Performance Ultrathin Mixed-Matrix Membranes**  
**Based on Adhesive PGMA-co -POEM Comb-like**  
**Copolymer for CO<sub>2</sub> Capture**

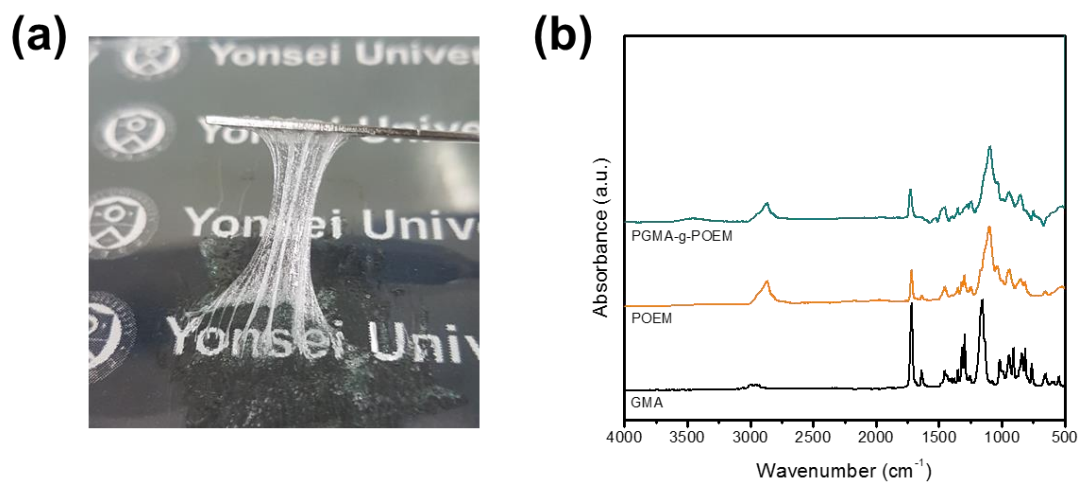
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Seodaemun-gu, Seoul 03722, South Korea*

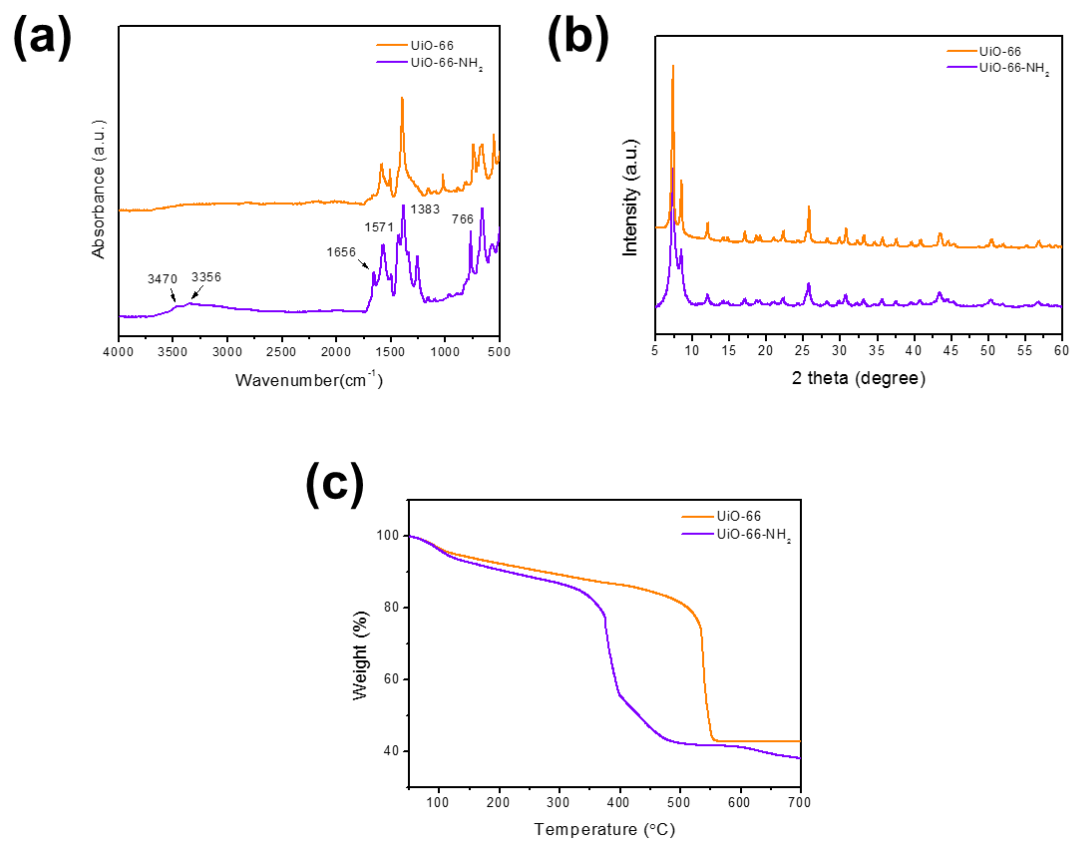
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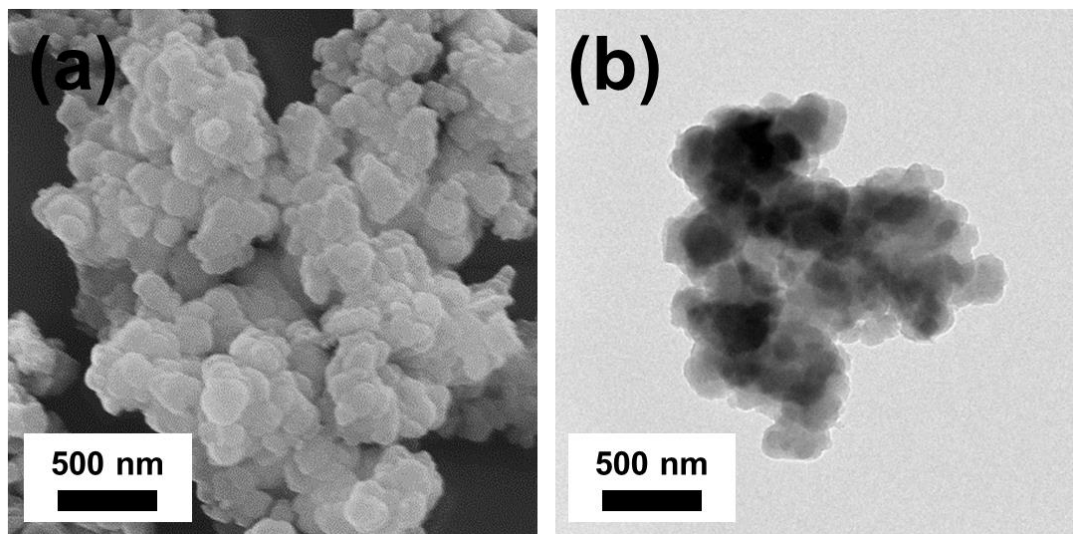
E-mail: [jonghak@yonsei.ac.kr](mailto:jonghak@yonsei.ac.kr)



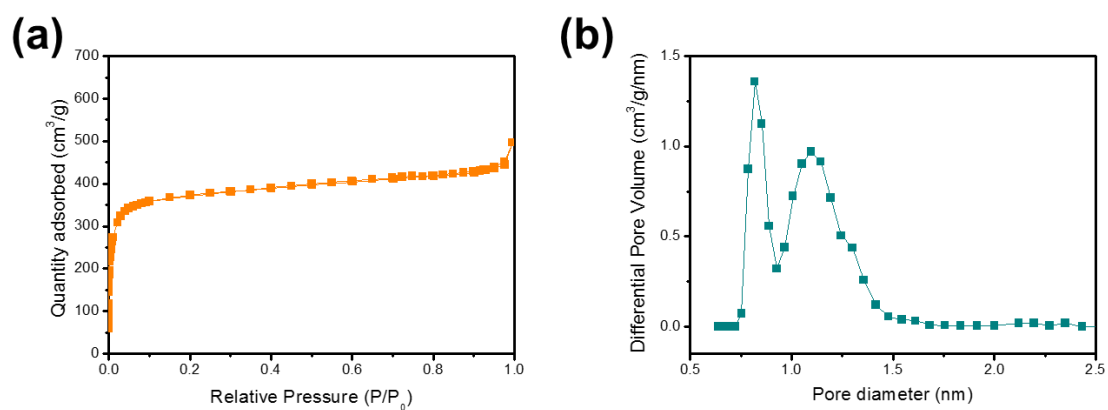
**Fig. S1.** (a) Photograph of the PGMA-co-POEM comb-like copolymer, showing its adhesive property; (b) FT-IR spectra of monomers and the PGMA-co-POEM comb-like copolymer.



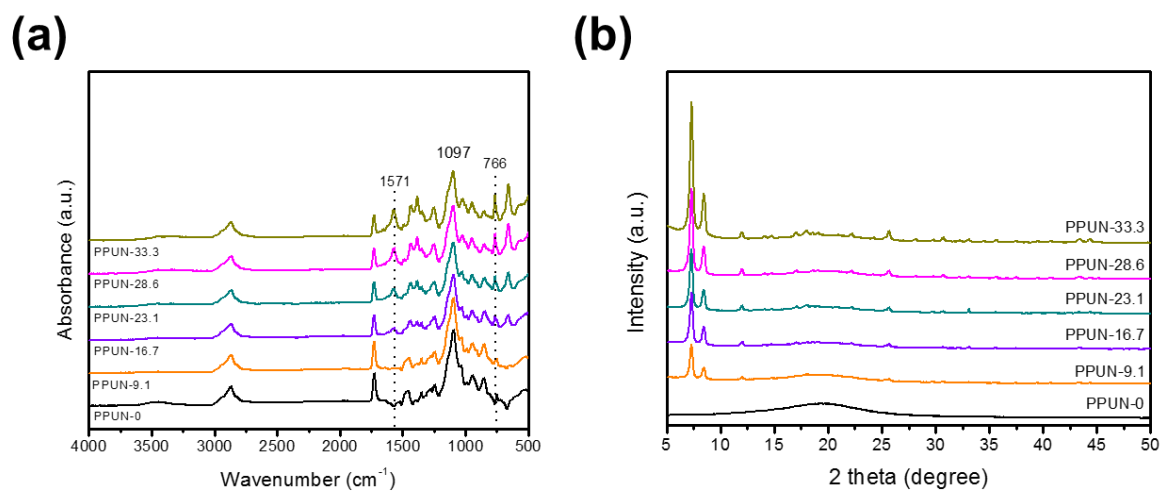
**Fig. S2.** (a) FT-IR spectra, (b) XRD patterns, and (c) TGA curves of UiO-66-NH<sub>2</sub> and pristine UiO-66 nanoparticles.



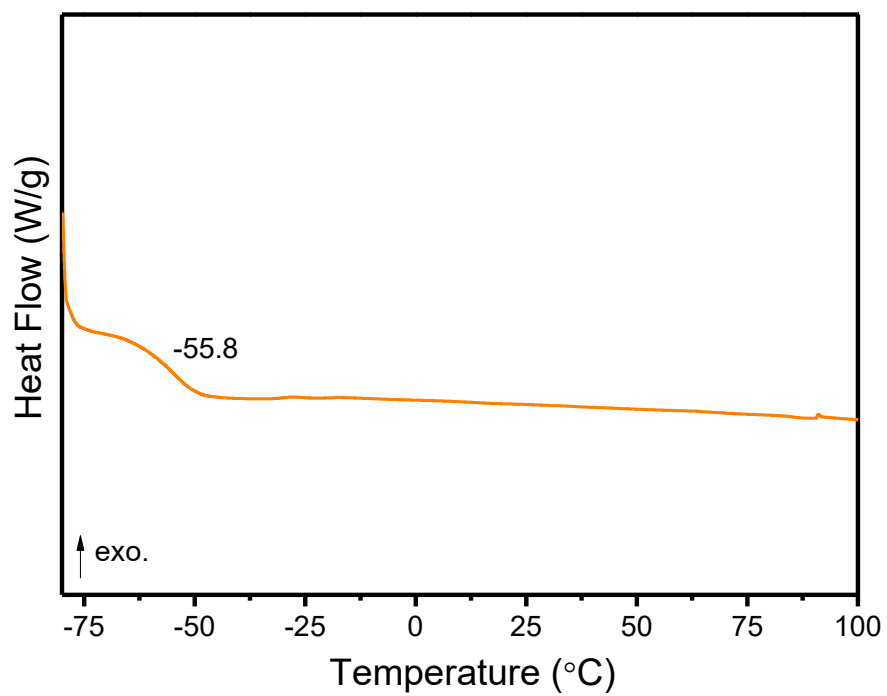
**Fig. S3.** (a) SEM and (b) TEM images of pristine UiO-66 nanoparticles.



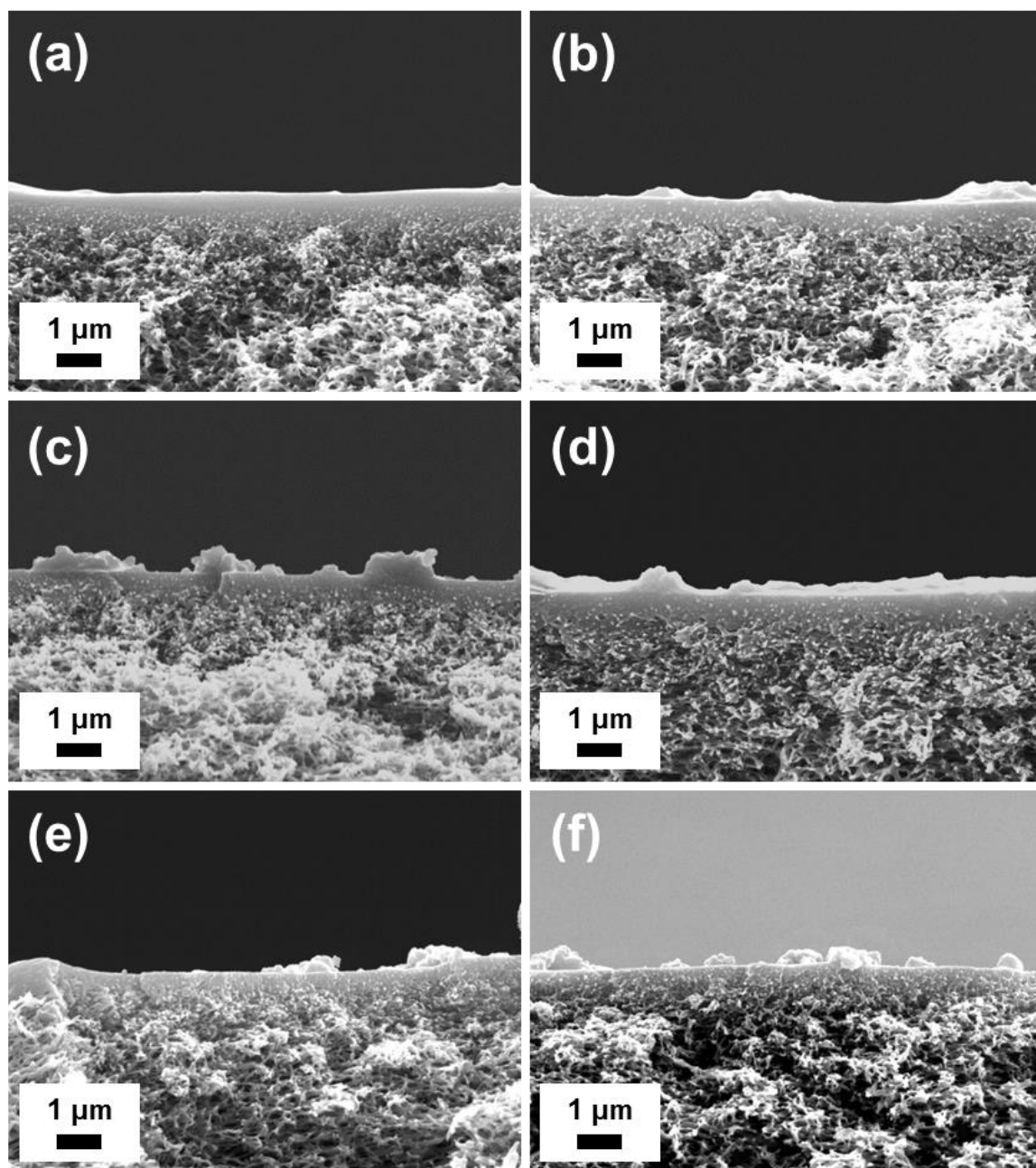
**Fig. S4.** (a) N<sub>2</sub> adsorption–desorption isotherm and (b) pore-size distribution of pristine UiO-66 nanoparticles.



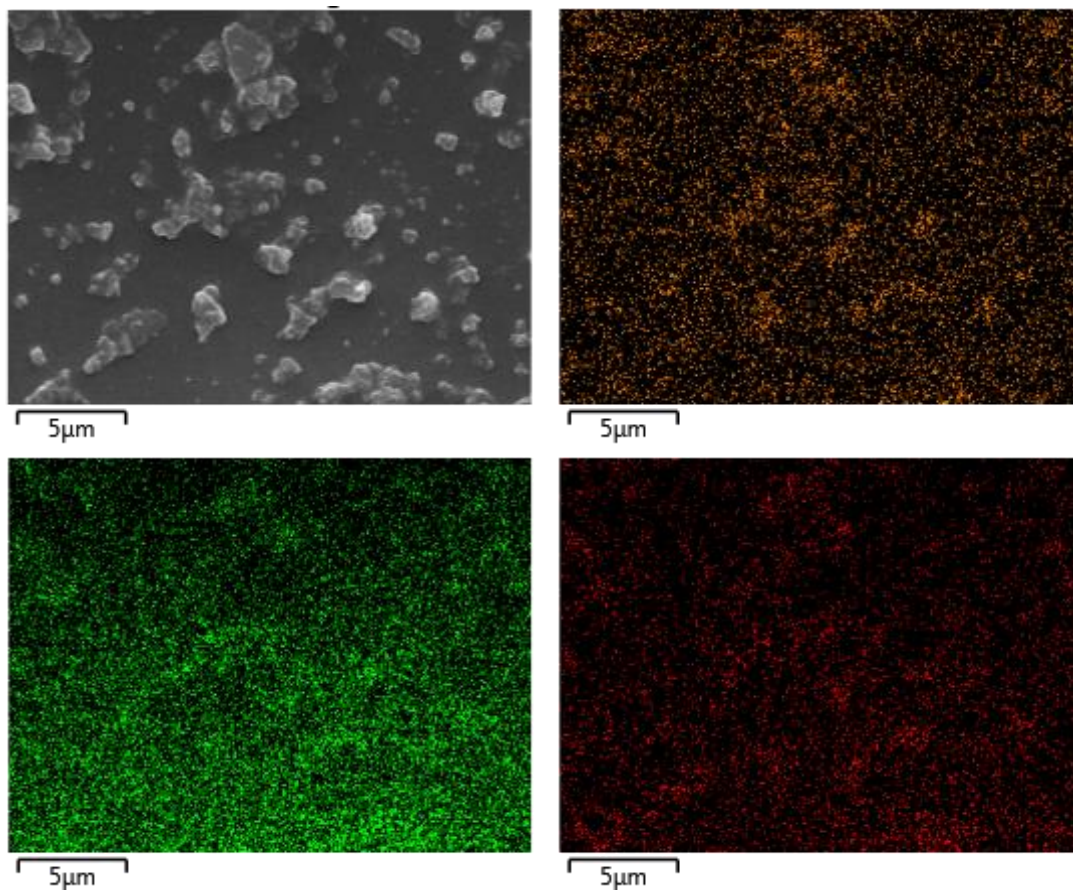
**Fig. S5.** (a) FT-IR spectra and (b) XRD patterns of PPUN membranes with different UiO-66-NH<sub>2</sub> loadings.



**Fig. S6.** DSC curve of the PPU-9.1 membrane.



**Fig. S7.** Cross-sectional SEM images of PPUN membranes: (a) PPUN-0, (b) PPUN-9.1, (c) PPUN-16.7, (d) PPUN-23.1, (e) PPUN-28.6, and (f) PPUN-33.3.



**Fig. S8.** SEM-EDX surface mapping of PPUN-28.6 membrane (Zr signal: orange, C signal: green, O signal: red).

**Table S1.** BET surface areas and micropore volumes of the MOFs studied.

|                                          | UiO-66-NH <sub>2</sub> | UiO-66 | Copolymer-grafted<br>UiO-66-NH <sub>2</sub> |
|------------------------------------------|------------------------|--------|---------------------------------------------|
| BET surface area<br>(m <sup>2</sup> /g)  | 1001                   | 1416   | 889                                         |
| Micropore volume<br>(cm <sup>3</sup> /g) | 0.354                  | 0.495  | 0.308                                       |

\* Micropore volume was calculated via the t-plot method.

**Table S2.** Pure-gas separation performance of PPUN membranes, measured at 25 °C and 1 bar.

| Membrane  | Permeance (GPU) |                | Selectivity                     |
|-----------|-----------------|----------------|---------------------------------|
|           | CO <sub>2</sub> | N <sub>2</sub> | CO <sub>2</sub> /N <sub>2</sub> |
| PPUN-0    | 51              | 1.1            | 46.3                            |
| PPUN-9.1  | 100             | 2.0            | 50.1                            |
| PPUN-16.7 | 278             | 8.3            | 33.5                            |
| PPUN-23.1 | 354             | 10.3           | 34.4                            |
| PPUN-28.6 | 488             | 15.3           | 31.9                            |
| PPUN-33.3 | 366             | 15.1           | 24.2                            |
| PPU-28.6  | 666             | 29.7           | 22.4                            |

**Table S3.** Gas separation performance of PPUN-28.6 membranes prepared with coating solutions of different concentrations.

| Membrane  | Polymer concentration | Permeance (GPU) |                | Selectivity                     |
|-----------|-----------------------|-----------------|----------------|---------------------------------|
|           |                       | CO <sub>2</sub> | N <sub>2</sub> | CO <sub>2</sub> /N <sub>2</sub> |
| PPUN-28.6 | 2.5%                  | 488             | 15.3           | 31.9                            |
|           | 2%                    | 962             | 29.1           | 33.1                            |
|           | 1.7%                  | 1320            | 42.9           | 30.8                            |

**Table S4.** Gas separation performances of various reported MMMs.

| MMM                                           | Filler<br>loading<br>(wt%) | CO <sub>2</sub><br>permeance<br>(GPU) | CO <sub>2</sub> /N <sub>2</sub><br>selectivity | Test<br>condition  | Feed gas                                                              | Ref. |
|-----------------------------------------------|----------------------------|---------------------------------------|------------------------------------------------|--------------------|-----------------------------------------------------------------------|------|
| ZIF-8<br>/PVAm                                | 13.1                       | 297                                   | 83                                             | 1.1 MPa<br>/25 °C  | Mixed gas<br>(CO <sub>2</sub> /N <sub>2</sub> = 15/85)                | [14] |
| UiO-66-NH <sub>2</sub><br>/PVAm               | 28.5                       | 1295                                  | 91                                             | 0.3 MPa<br>/25 °C  | Mixed gas<br>(CO <sub>2</sub> /N <sub>2</sub> = 15/85),<br>humidified | [22] |
| NH <sub>2</sub> -ZIF-8<br>/polyamide<br>(TFN) | N/A                        | 1572                                  | 230                                            | 0.2 MPa<br>/25 °C  | Mixed gas<br>(CO <sub>2</sub> /N <sub>2</sub> = 15/85),<br>humidified | [36] |
| PEI-g-ZIF-8<br>/PVAm                          | 16.7                       | 1990                                  | 79.9                                           | 3 bar<br>/25 °C    | Mixed gas<br>(CO <sub>2</sub> /N <sub>2</sub> = 15/85),<br>humidified | [44] |
| ZIF-7<br>/Pebax                               | 34                         | 39                                    | 105                                            | 3.75 bar<br>/20 °C | Pure gas                                                              | [45] |
| SiO <sub>2</sub><br>/PEGDMA9                  | 5                          | 1290                                  | 27                                             | 350 kPa/<br>35 °C  | Pure gas                                                              | [46] |