

Supporting Information

# 6FDA-DAM:DABA co-polyimide mixed matrix membranes with ZIF-8 and GO mixtures for effective CO<sub>2</sub>/CH<sub>4</sub> separation

Anand Jain <sup>1,2</sup>, Mohd Zamidi Ahmad <sup>1,3,\*</sup>, Audrey Linkès <sup>1</sup>, Violeta Martin-Gil <sup>1</sup>, Roberto Castro-Muñoz <sup>1,4</sup>, Werner Hintz <sup>2</sup>, Pavel Izak <sup>5</sup>, Zdeněk Sofer <sup>6</sup> and Vlastimil Fila <sup>1,\*</sup>

<sup>1</sup> Department of Inorganic Technology, University of Chemistry and Technology Prague, Technická 5, 166 28 Prague 6, Czech Republic; A.L. ([adv.jain92@gmail.com](mailto:adv.jain92@gmail.com)), V.M.G. ([violeta.m.gil@gmail.com](mailto:violeta.m.gil@gmail.com))

<sup>2</sup> Faculty of Process and Systems Engineering, Otto-von-Guericke-University, Magdeburg Universitätsplatz 2, Magdeburg-39106, Germany; A.J. ([adv.jain92@gmail.com](mailto:adv.jain92@gmail.com)), W.H. ([werner.hintz@ovgu.de](mailto:werner.hintz@ovgu.de))

<sup>3</sup> Organic Materials Innovation Center (OMIC), Department of Chemistry, University of Manchester, Oxford Road, Manchester M13 9PL, UK

<sup>4</sup> Tecnológico de Monterrey, Campus Toluca, Avenida Eduardo Monroy Cárdenas 2000 San Antonio Buenavista, 50110 Toluca de Lerdo, Mexico; R.C.M. ([food.biotechnology88@gmail.com](mailto:food.biotechnology88@gmail.com); [castromr@tec.mx](mailto:castromr@tec.mx))

<sup>5</sup> Department of Membrane Separation Processes, Institute of Chemical Process Fundamentals of the CAS, v. v. i., Rozvojova 2/135, 165 02 Prague 6 – Suchbát, Czech Republic; P.I. ([izak@icpf.cas.cz](mailto:izak@icpf.cas.cz))

<sup>6</sup> Department of Inorganic Chemistry, University of Chemistry and Technology Prague, Technická 5, 166 28 Prague 6, Czech Republic; Z.S. ([Zdenek.Sofer@vscht.cz](mailto:Zdenek.Sofer@vscht.cz))

\* Correspondence: V.F. ([Vlastimil.Fila@vscht.cz](mailto:Vlastimil.Fila@vscht.cz)); M.Z.A. ([mohdzamidi.ahmad@manchester.ac.uk](mailto:mohdzamidi.ahmad@manchester.ac.uk))

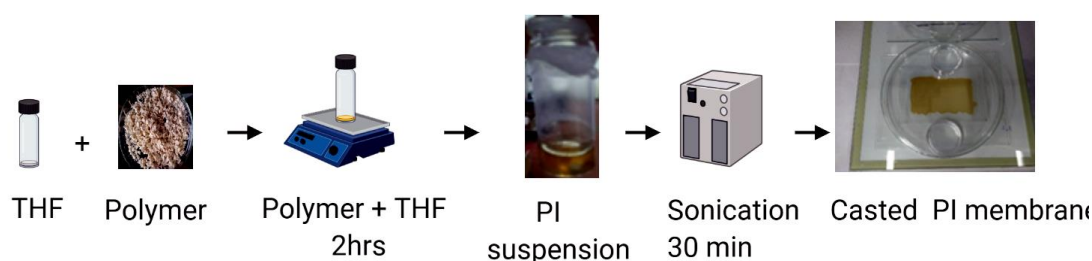


Figure S1. The illustration of preparation procedure of co-polyimide (co-PI) membranes.

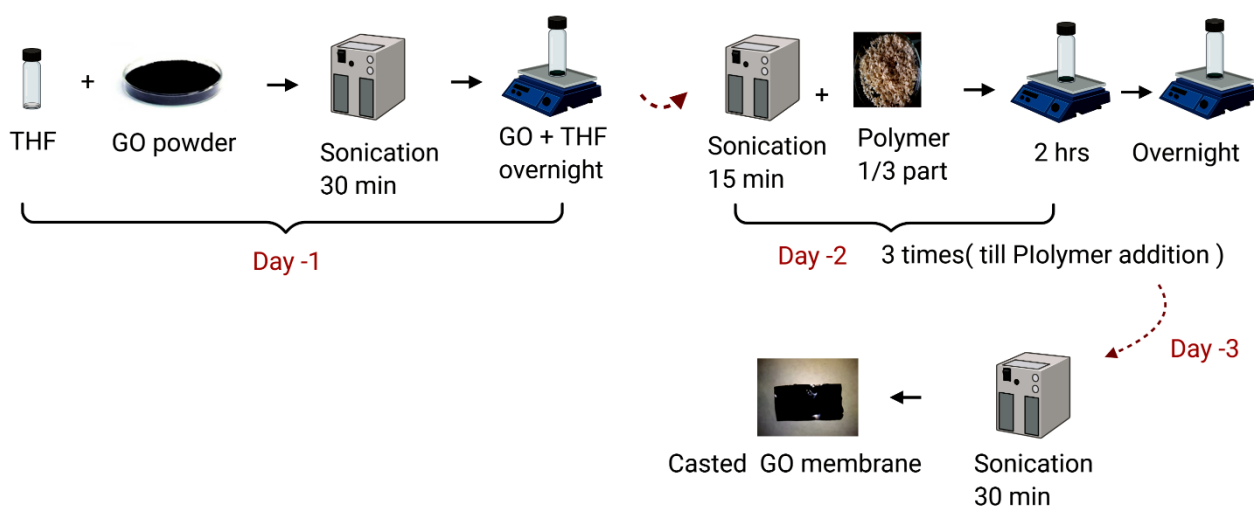


Figure S2. The illustration of preparation procedure of co-PI with GOMMMs.

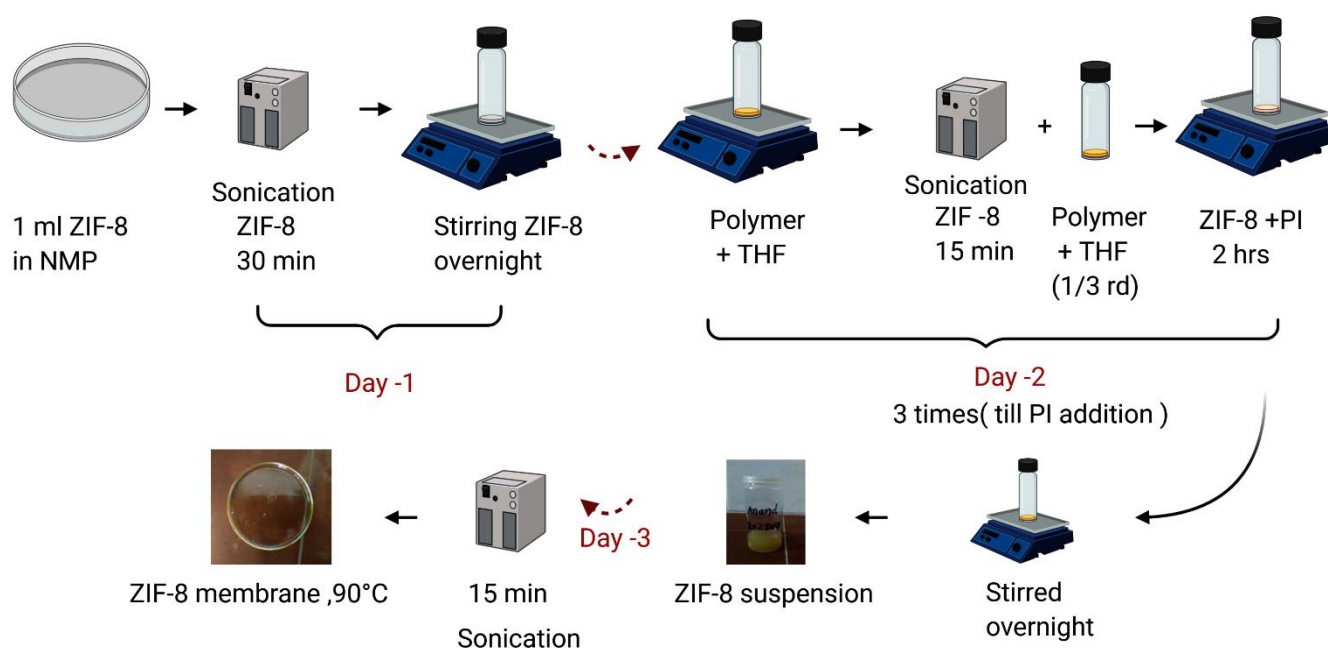


Figure S3. The illustration of preparation procedure of ZIF-8/co-PI MMMs.

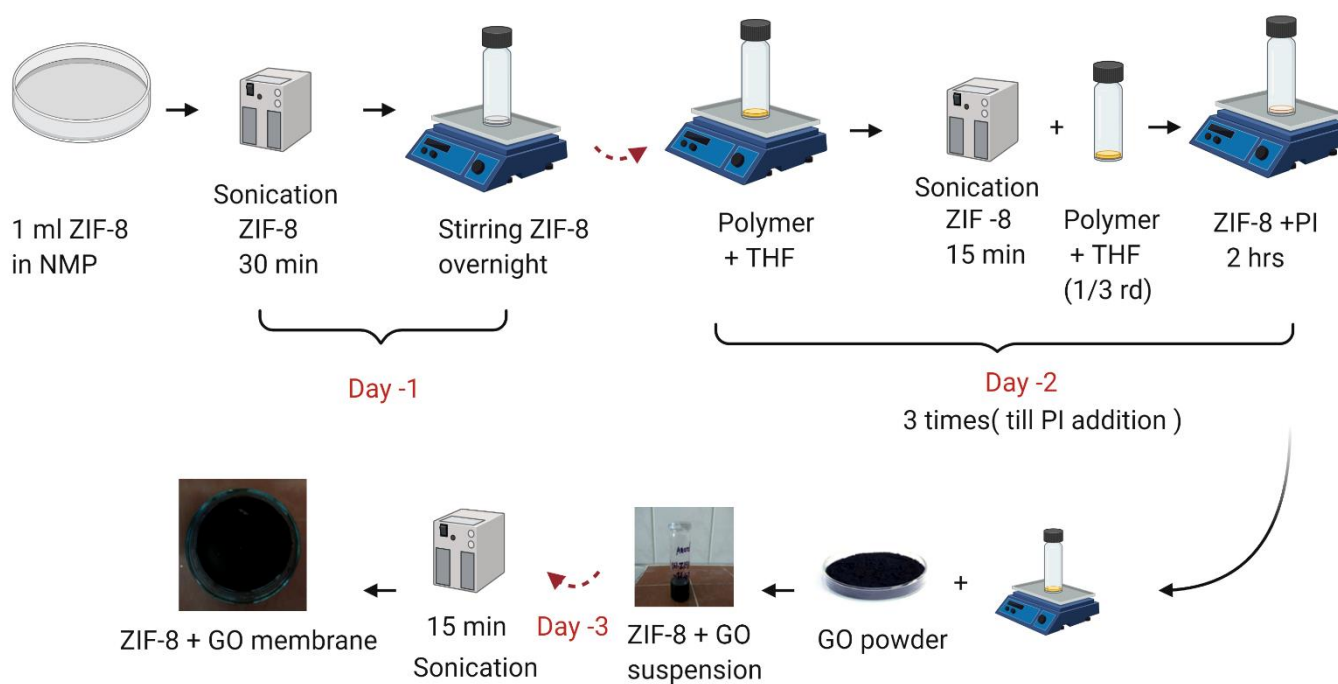


Figure S4. The illustration of preparation procedure of ZIF-8/GO/co-PI MMMs.

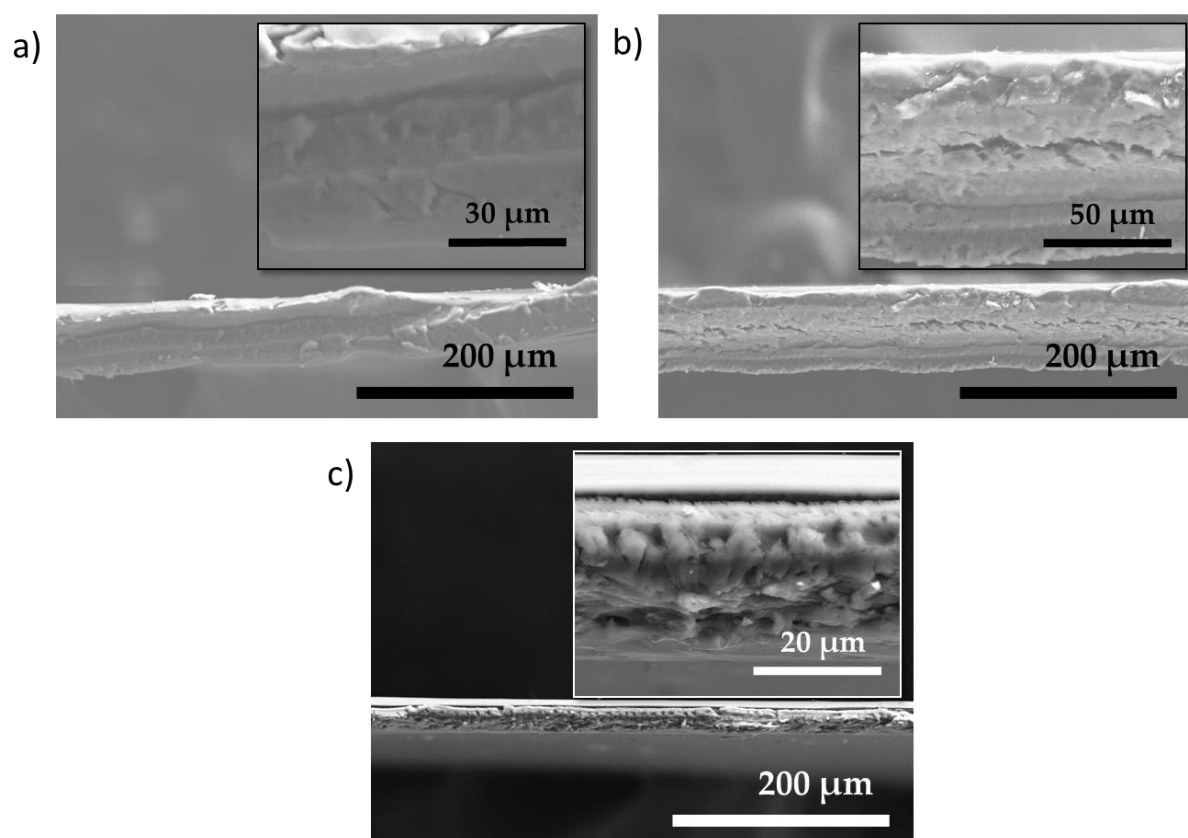


Figure S5. FESEM images of 6FDA-DAM:DABA (3:1) with (a) 1 wt.%, (b) 2 wt.% and (c) 4 wt.% GO nanosheets.

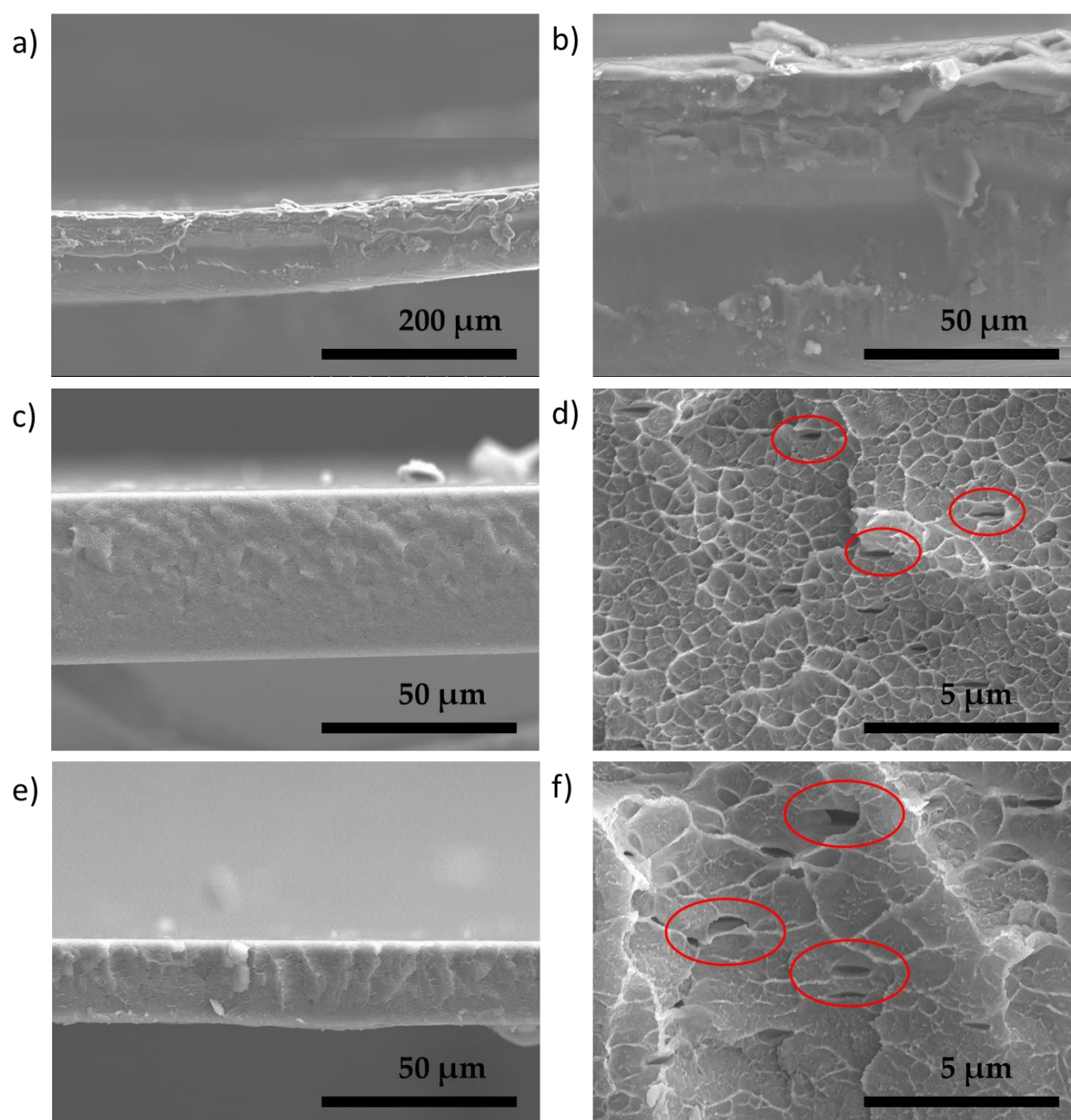


Figure S6. FESEM images of 6FDA-DAM:DABA (3:1) with (a-b) 5 wt.%, (c-d) 10 wt.% and (e-f) 15 wt.% ZIF-8 nanoparticles. Red circles highlight the defective interfaces, a result of poor filler-polymer interface interactions.

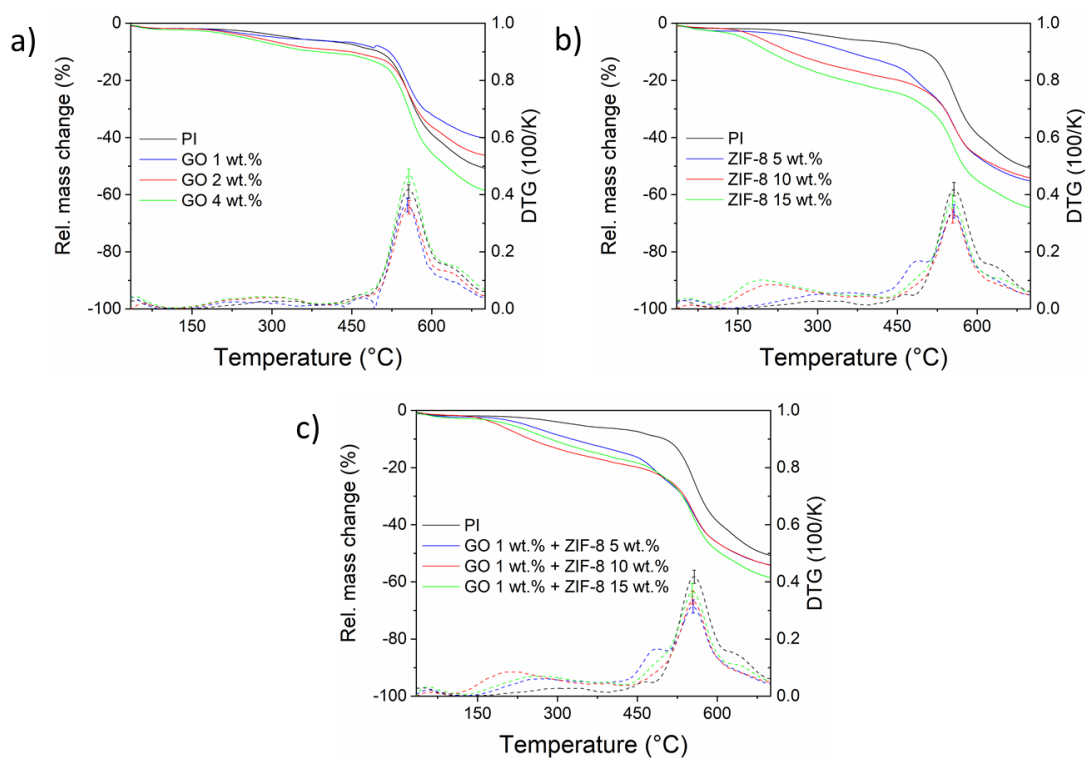


Figure S7. Thermal decomposition profiles of 6FDA-DAM:DABA (3:1) and its MMMs with (a) GO, (b) ZIF-8, and (c) GO and ZIF-8 mixtures. Included are their weight loss first derivatives.

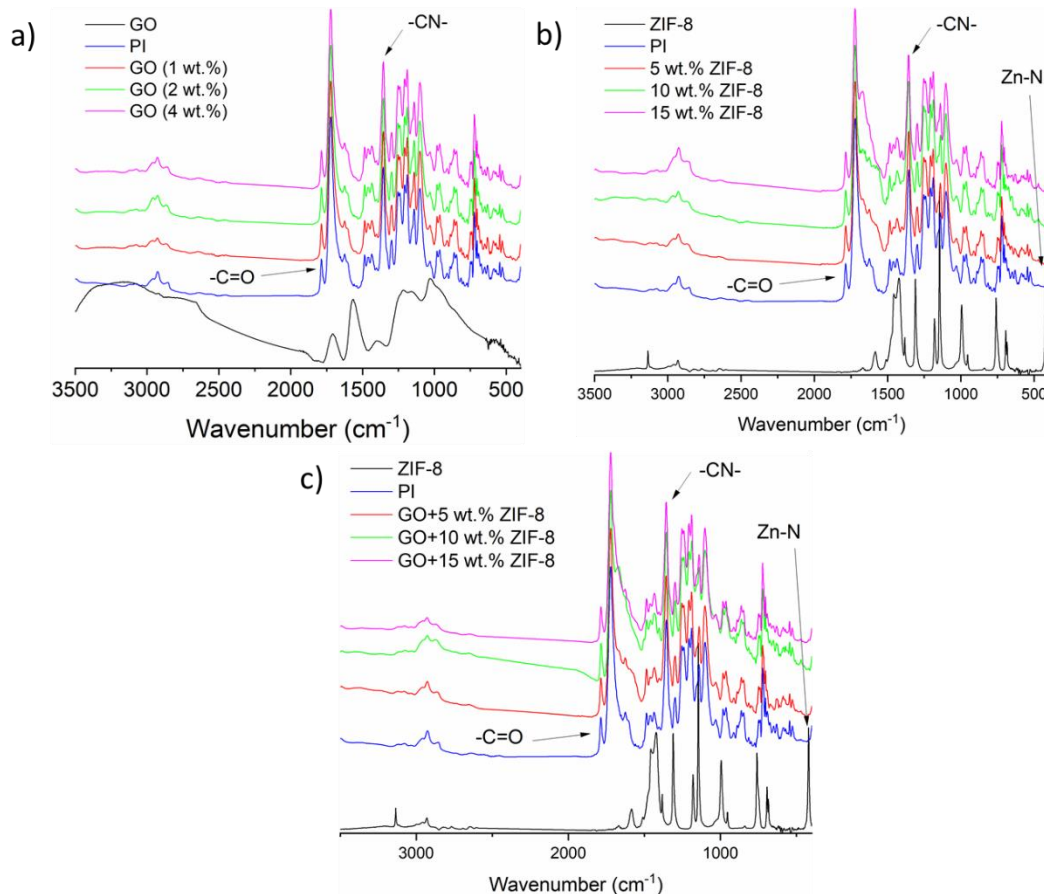


Figure S8. FTIR spectra of 6FDA-DAM:DABA (3:1) and its MMMs with (a) GO, (b) ZIF-8, and (c) GO and ZIF-8.

Table S1. Gas permeabilities ( $P$ ) and  $\text{CO}_2/\text{CH}_4$  selectivity ( $\alpha$ ) of neat 6FDA-DAM:DABA (3:1) and its respective MMMs with GO nanosheets, ZIF-8 nanoparticles, and GO/ZIF-8 mixtures. All membranes were evaluated with 50:50  $\text{CO}_2/\text{CH}_4$  binary mixtures at 25 °C, at a feed pressure of 2 bar.

| Membrane                  | Gas permeability (Barrer) |                | $\text{CO}_2/\text{CH}_4$ selectivity |
|---------------------------|---------------------------|----------------|---------------------------------------|
|                           | $\text{CO}_2$             | $\text{CH}_4$  |                                       |
| 6FDA-DAM:DABA (3:1)       | $147 \pm 6.1$             | $3.1 \pm 0.1$  | $47.5 \pm 4.0$                        |
| GOMMMs                    |                           |                |                                       |
| 1 wt.% GO                 | $179 \pm 3.6$             | $3.5 \pm 0.2$  | $50.8 \pm 3.6$                        |
| 2 wt.% GO                 | $147 \pm 3.5$             | $3.8 \pm 0.2$  | $38.5 \pm 2.5$                        |
| 4 wt.% GO                 | $104 \pm 3.9$             | $2.5 \pm 0.2$  | $42.0 \pm 3.0$                        |
| ZIF-8 MMMs                |                           |                |                                       |
| 5 wt.% ZIF-8              | $337 \pm 7.2$             | $15.1 \pm 1.2$ | $22.4 \pm 1.6$                        |
| 10 wt.% ZIF-8             | $846 \pm 28.7$            | $29.7 \pm 2.4$ | $28.7 \pm 3.3$                        |
| 15 wt.% ZIF-8             | $1,109.8 \pm 29.5$        | $43.5 \pm 3.4$ | $25.7 \pm 2.5$                        |
| GO + ZIF-8 MMMs           |                           |                |                                       |
| 1 wt.% GO + 5 wt.% ZIF-8  | $1,607 \pm 17.6$          | $40.8 \pm 1.0$ | $39.4 \pm 1.3$                        |
| 1 wt.% GO + 10 wt.% ZIF-8 | $1,800 \pm 61.8$          | $85.1 \pm 3.0$ | $21.2 \pm 1.4$                        |
| 1 wt.% GO + 15 wt.% ZIF-8 | $1,550 \pm 25.1$          | $54.9 \pm 0.8$ | $28.2 \pm 0.8$                        |

Table S2. Gas permeabilities ( $P$ ) and  $\text{CO}_2/\text{CH}_4$  selectivity ( $\alpha$ ) of neat 6FDA-DAM:DABA (3:1) and GO/ZIF-8 MMMs, tested at different feed pressure (2, 4, 6, 8 Bar). All membranes were evaluated with 50:50  $\text{CO}_2/\text{CH}_4$  binary mixtures at 25 °C.

| Membrane                 | Pressure (bar) | Gas permeability (Barrer) |                 | CO <sub>2</sub> /CH <sub>4</sub> selectivity |
|--------------------------|----------------|---------------------------|-----------------|----------------------------------------------|
|                          |                | CO <sub>2</sub>           | CH <sub>4</sub> |                                              |
| 6FDA-DAM:DABA (3:1)      |                |                           |                 |                                              |
|                          | 2              | 147 ± 6.1                 | 3.1 ± 0.1       | 47.5 ± 4.0                                   |
|                          | 4              | 111 ± 4.0                 | 2.5 ± 0.1       | 44.0 ± 3.4                                   |
|                          | 6              | 98 ± 3.5                  | 2.3 ± 0.1       | 43.1 ± 3.2                                   |
|                          | 8              | 91 ± 1.9                  | 2.2 ± 0.1       | 41.9 ± 1.5                                   |
| 1 wt.% GO+5 wt.% ZIF-8   |                |                           |                 |                                              |
|                          | 2              | 1,607 ± 17.6              | 40.8 ± 1.0      | 39.4 ± 1.3                                   |
|                          | 4              | 1,205 ± 21.0              | 37.1 ± 1.5      | 32.6 ± 1.8                                   |
|                          | 6              | 1,108 ± 44.6              | 50.6 ± 2.5      | 22.0 ± 2.0                                   |
|                          | 8              | 1,036 ± 5.3               | 59.0 ± 0.8      | 17.6 ± 0.3                                   |
| 1 wt.% GO+ 10 wt.% ZIF-8 |                |                           |                 |                                              |
|                          | 2              | 1,800 ± 61.8              | 85.1 ± 3.0      | 21.2 ± 1.4                                   |
|                          | 4              | 1,432 ± 74.3              | 75.5 ± 2.1      | 19.0 ± 1.3                                   |
|                          | 6              | 1,262 ± 9.1               | 77.5 ± 1.4      | 16.3 ± 0.4                                   |
|                          | 8              | 1,176 ± 14.8              | 77.6 ± 0.8      | 15.2 ± 0.3                                   |
| 1 wt.% GO+ 15 wt.% ZIF-8 |                |                           |                 |                                              |
|                          | 2              | 1,550 ± 25.1              | 54.9 ± 0.8      | 28.2 ± 0.8                                   |
|                          | 4              | 1,192 ± 31.7              | 50.1 ± 1.2      | 23.8 ± 1.2                                   |
|                          | 6              | 1,021 ± 4.5               | 52.2 ± 0.4      | 19.6 ± 0.1                                   |
|                          | 8              | 1,025 ± 12.2              | 85.1 ± 2.2      | 12.1 ± 0.4                                   |