

## Electronic Supporting Information

### **Fabrication of mixed-matrix membrane containing selective and facilitated CO<sub>2</sub> transport metal-organic framework composite with task-specific ionic liquid for efficient CO<sub>2</sub> separation**

Jing Ma, Yunpan Ying, Xiangyu Guo, Hongliang Huang, Dahuan Liu\* and Chongli Zhong\*

*State Key Laboratory of Organic-Inorganic Composites, Beijing University of Chemical Technology,  
Beijing 100029, China*

Correspondence to: [liudh@mail.buct.edu.cn](mailto:liudh@mail.buct.edu.cn), [zhongcl@mail.buct.edu.cn](mailto:zhongcl@mail.buct.edu.cn).

## 1. Transmission electron microscope (TEM) analysis

The TEM images were investigated using a Tecnai G<sup>2</sup>20 equipment.

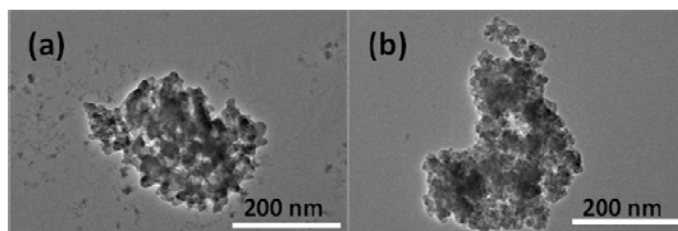


Fig. S1 TEM images of TSIL@NH<sub>2</sub>-MIL-101(Cr) (a); NH<sub>2</sub>-MIL-101(Cr) (b).

## 2. EDX-mapping

The EDX-mapping analysis was carried out using a FEI S-4800 equipment.

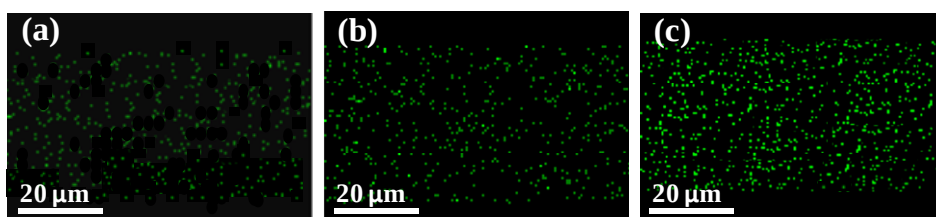


Fig. S2 Cr mapping of (a) 5% TSIL@NH<sub>2</sub>-MIL-101(Cr)/PIM-1, (b) 10% TSIL@NH<sub>2</sub>-MIL-101(Cr)/PIM-1, (c) 15% TSIL@NH<sub>2</sub>-MIL-101(Cr)/PIM-1 membranes.

## 3. Thermogravimetric (TGA) analyses

Thermal stability of NH<sub>2</sub>-MIL-101(Cr), TSIL, TSIL@NH<sub>2</sub>-MIL-101(Cr) as well as the fabricated membranes were characterized on a TGA 1/1100 SF instrument with a heating rate of 10 °C/min under nitrogen atmosphere and the resultant thermograms were shown in Fig. S3. In the first step (< 250 °C), the weight loss is due to the loss of the adsorbed water inside the pores. In the second step (250–300 °C), the weight loss is attributed to the decomposition of the NH<sub>2</sub>-MIL-101(Cr) powder and TSIL. In the whole process, the TSIL@NH<sub>2</sub>-MIL-101(Cr) showed less weight loss than NH<sub>2</sub>-MIL-101(Cr) and TSIL, which indicates a better thermal stability due to the interaction between the TSIL and NH<sub>2</sub>-MIL-101(Cr).<sup>S1</sup> In the case of membranes, the decomposition of TSIL@NH<sub>2</sub>-MIL-101(Cr)/PIM-1 membrane is started at around 400 °C, signifying the TSIL@NH<sub>2</sub>-MIL-101(Cr) decomposition. The second stage of weight loss occurs at around 500 °C, which is fully in agreement with the decomposition temperature of PIM-1 membrane. Besides, the TSIL@NH<sub>2</sub>-MIL-101(Cr)/PIM-1 membrane shows a better thermal stability than NH<sub>2</sub>-MIL-101(Cr)/PIM-1 membrane, which can also confirm the interaction between the TSIL and NH<sub>2</sub>-MIL-101(Cr) in TSIL@NH<sub>2</sub>-MIL-101(Cr).

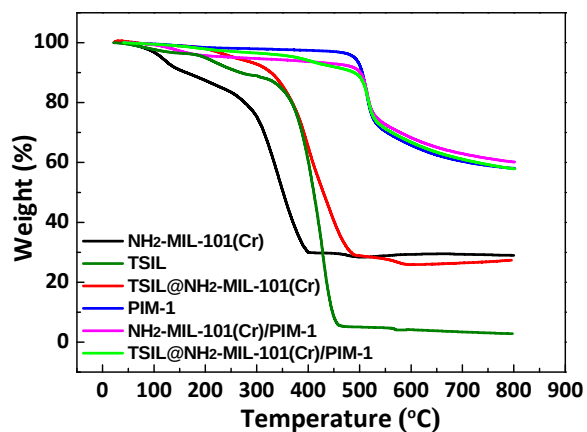


Fig. S3 Thermal stability of the samples.

#### 4. Long-term operation stability measurement

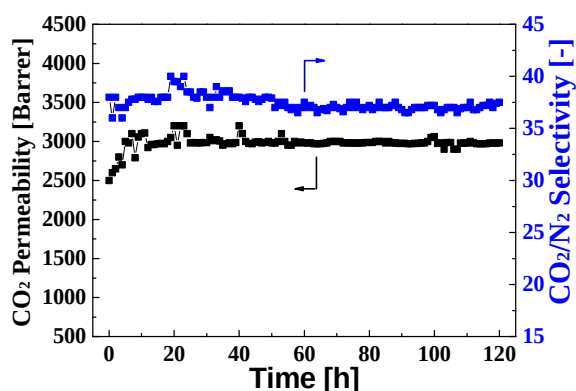


Fig. S4 Evolution of the CO<sub>2</sub> permeability (left-axis) and the CO<sub>2</sub>/N<sub>2</sub> selectivity (right-axis) of TSIL@MOF/PIM-1 membrane with 5 wt% loading.

Operation conditions: T = 298 K, ΔP = 3 bar.

#### 5. Gas separation performance comparison

Table S1 Comparison of gas separation performance of related membranes

| Filler                                                     | Polymer     | Loading<br>(wt %) | Operation conditions |             | P(CO <sub>2</sub> )<br>[Barrer] | α<br>(CO <sub>2</sub> /N <sub>2</sub> ) | Ref. |
|------------------------------------------------------------|-------------|-------------------|----------------------|-------------|---------------------------------|-----------------------------------------|------|
|                                                            |             |                   | T<br>( ° C )         | ΔP<br>(bar) |                                 |                                         |      |
| Cu 1,4-BDC                                                 | PVAc        | 15                | 30                   | 4           | 3.3                             | 35.4                                    | S2   |
| ZIF-8                                                      | Matrimd     | 50                | 25                   | 2.7         | 4.7                             | 26.2                                    | S3   |
| HKUST-1                                                    | PMDA-ODA    | 6                 | 25                   | 10          | 227.2                           | 5.5                                     | S4   |
| ZIF-8                                                      | PPEES       | 30                | 30                   | 1           | 50                              | 24.5                                    | S5   |
| ZIF-7                                                      | Pebax       | 22                | 20                   | 2.75        | 111                             | 97                                      | S6   |
| HKUST-1                                                    | PPO         | 40                | 30                   | -           | 115                             | 26                                      | S7   |
| ZIF-8                                                      | Pebax       | 35                | 25                   | 6           | 1287                            | 32.3                                    | S8   |
| [Cd <sub>2</sub> 6FDA(H <sub>2</sub> O)-5H <sub>2</sub> O] | 6FDA-ODA    | 10                | 25                   | 2           | 37.8                            | 35.1                                    | S9   |
| ZIF-8                                                      | 6FDA-durene | 30                | 25                   | 6           | 2185.5                          | 17                                      | S10  |
| MIL-53(Al)                                                 | Matrimd     | 37.5              | 35                   | 2           | 40                              | 95.2                                    | S11  |
| c-MOF-5                                                    | PEI         | 25                | 25                   | 6           | 5.4                             | 28.4                                    | S12  |

|                                   |                            |                 |    |     |        |       |           |
|-----------------------------------|----------------------------|-----------------|----|-----|--------|-------|-----------|
| HKUST-1                           | Ultem                      | 35              | 35 | 3.5 | 4.1    | 28    | S13       |
| ZIF-71                            | 6FDA-durene                | 20              | 35 | 3.5 | 4006   | 12.9  | S14       |
| PEG-PEI-GO                        | Pebax                      | 10              | 25 | 1   | 1330   | 120   | S15       |
| Mg <sub>2</sub> (bobdc)           | PI                         | 10              | 25 | 2   | 850    | 23    | S16       |
| GO                                | Pebax                      | 0.1             | 25 | 3   | 100    | 91    | S17       |
| PEI@MIL-101(Cr)                   | SPEEK                      | 40              | 25 | 1.5 | 2490   | 80    | S18       |
| NH <sub>2</sub> -MIL-101(Al)      | 6FDA-DAM                   | 20              | 25 | 1   | 737    | 26.3  | S19       |
| ZIF-8                             | PIM-1                      | 39              | 22 | 1   | 4270   | 21.89 | S20       |
| PAF-1                             | PIM-1                      | 30              | 25 | 1   | 3250   | 22    | S21       |
| Ti <sub>5</sub> UIO-66            | PIM-1                      | 5               | 25 | 2   | 5340   | 24    | S22       |
| ZIF-71                            | UV-PIM-1                   | 30              | 35 | 3.5 | 3458.6 | 26.9  | S23       |
| CC3                               | PIM-1                      | 30              | 25 | 1   | 5430   | 20    | S24       |
| MFI                               | PIM-1                      | 23.5(v%)        | 25 | 1   | 2530   | 30    | S25       |
| RTIL@ZIF-8                        | PSF                        | 6(v%)           |    |     | 312    | 117   | S26       |
| RTIL                              | PVDF                       | PVDF/RTIL (1/2) | 35 | 2   | 1778   | 41.1  | S27       |
| ZIF-8                             | P[vbim][Tf <sub>2</sub> N] | 18.9            | 35 | 3.5 | 198.8  | 19.5  | S28       |
| [emim][Tf <sub>2</sub> N]/ZIF-8   | P[vbim][Tf <sub>2</sub> N] | 23.7            |    |     | 693.6  | 19.6  |           |
| --                                | Cellulose acetate          |                 | 30 |     | 6.3    | 30    | S29       |
| --                                | Ethyl cellulose            |                 | 30 |     | 26.5   | 8.28  |           |
| --                                | Polycarbonate, brominated  |                 | 30 |     | 4.23   | 23.5  |           |
| --                                | PDMS                       |                 | 30 |     | 2700   | 10.8  |           |
| --                                | Matrimid                   |                 | 30 |     | 10.7   | 33.4  |           |
| --                                | Polymethylpentene          |                 | 30 |     | 84.6   | 12.63 |           |
| --                                | Polyphenyleneoxide         |                 | 30 |     | 75.8   | 19.9  |           |
| --                                | Polysulfone                |                 | 30 |     | 5.6    | 22.4  |           |
| --                                | Polyetherimide             |                 | 30 |     | 1.32   | 28.09 |           |
|                                   | PIM-1                      |                 | 25 | 3   | 2430   | 16.99 | This work |
| TSIL@NH <sub>2</sub> -MIL-101(Cr) | PIM-1                      | 5               | 25 | 3   | 2979   | 37.24 |           |

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