

Supporting Information for

## Engineering HOF-Based Mixed-Matrix Membranes for Efficient CO<sub>2</sub> Separation

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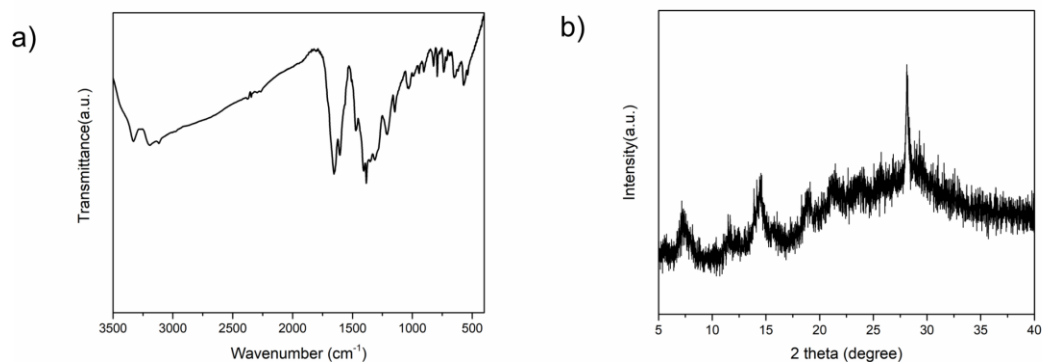
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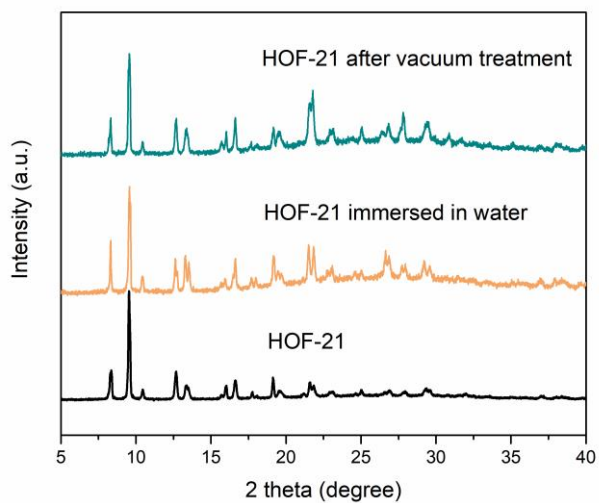
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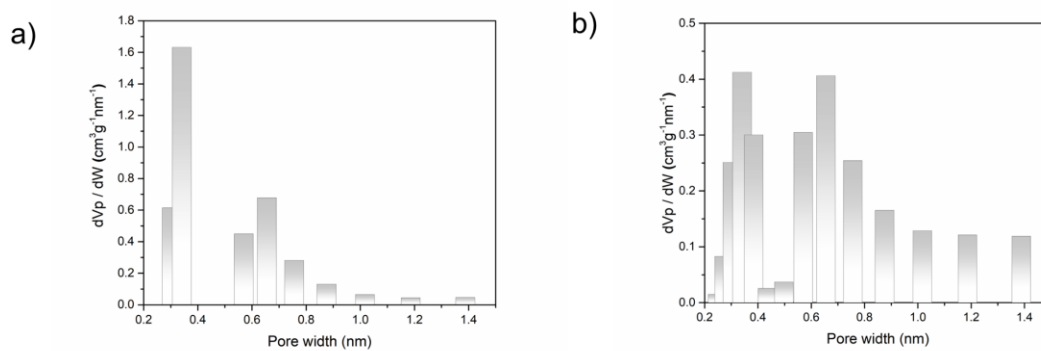
### Supplementary Figures



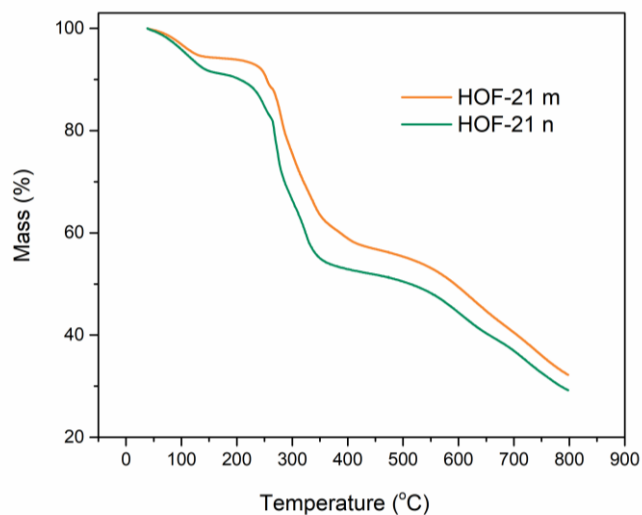
**Fig. S1** FTIR spectra and XRD pattern of Cu (ade)<sub>2</sub>



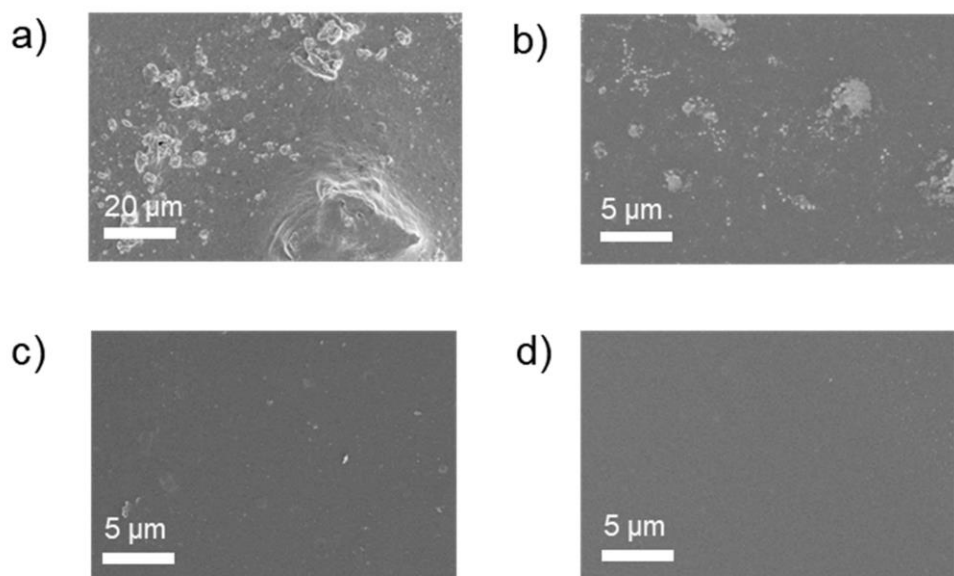
**Fig. S2** XRD patterns of initial HOF-21 sample (a) and the HOF-21 samples after immersed in water and vacuum treatment



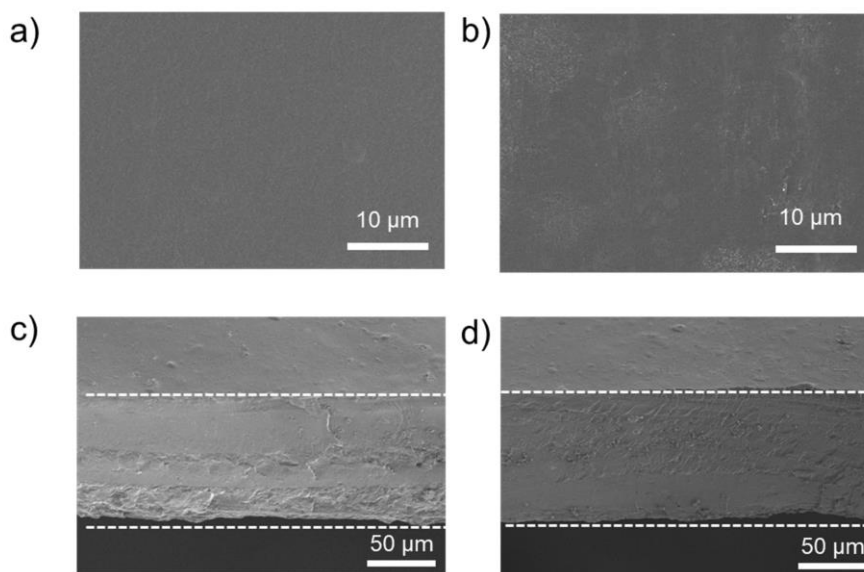
**Fig. S3** Pore size distribution of HOFm (a) and HOFn (b)



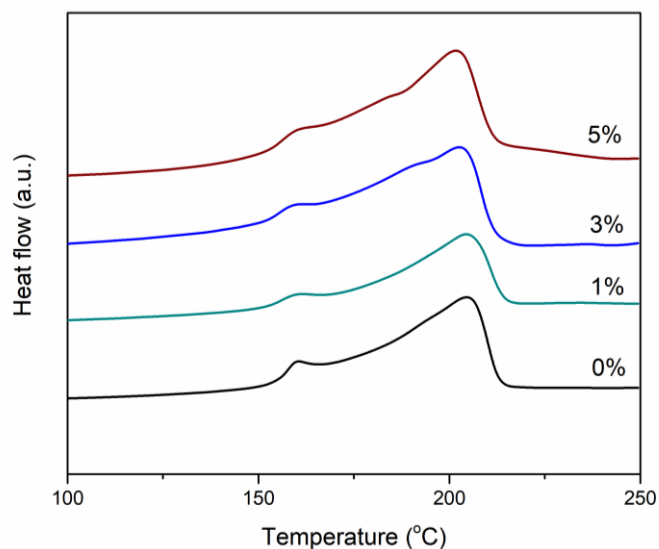
**Fig. S4** Thermogravimetric analysis of HOF-21m and HOF-21n



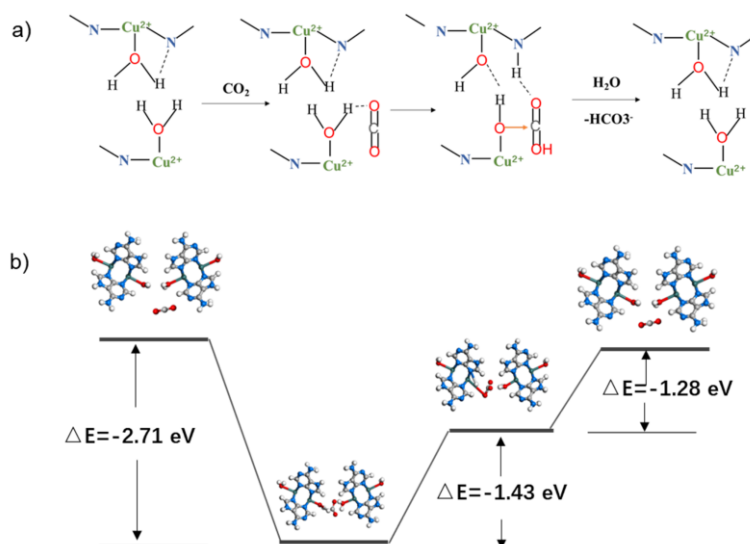
**Fig. S5** SEM images of the membranes with the fillers regulated by the ratios of  $(\text{NH}_4)_2\text{SiF}_6$ / diethylamine (**a** for the ratio of 1:0, **b** for the ratio of 1:0.5, **c** for the ratio of 1:1, **d** for the ratio of 1:2)



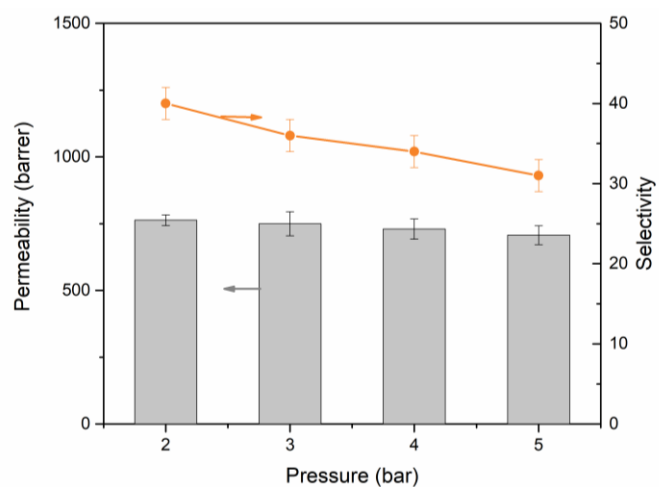
**Fig. S6** The top-section SEM images of MMMs with the mass loadings of 1 wt% (**a**) and 5 wt% (**b**), the cross-section membrane SEM images of MMMs with the mass loadings of 1 wt% (**c**) and 5 wt% (**d**)



**Fig. S7** DSC analysis of the MMMs with the filler content of 0, 1, 3, 5 wt%



**Fig. S8** Facilitated transport mechanism (a) and energy calculation (b)



**Fig. S9** Effect of pressure on the separation performance of HOF-21@Pebax-3