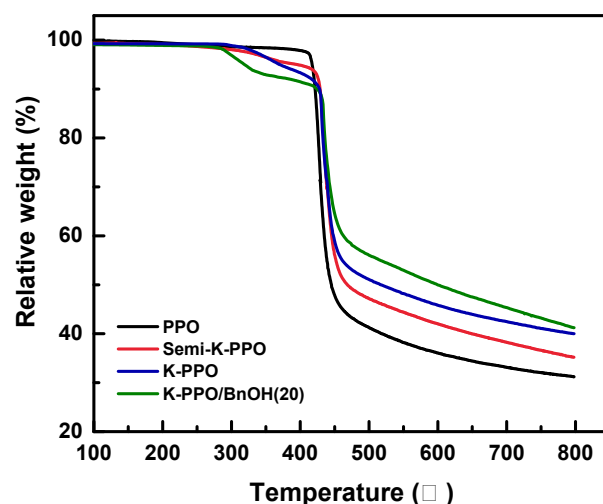


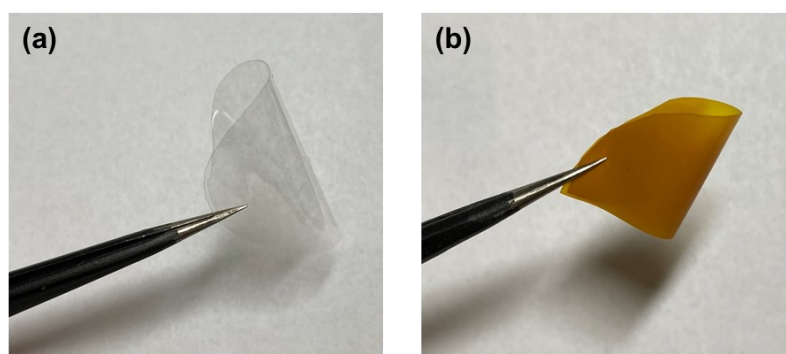
# Electronic Supplementary Information

## In situ knitted microporous polymer membranes for efficient CO<sub>2</sub> capture

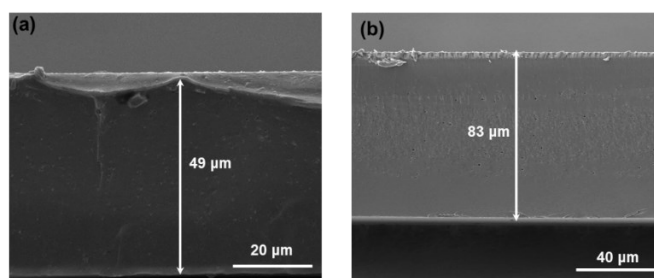
*Yingzhen Wu,<sup>a,b</sup> Na Xing,<sup>a,b</sup> Sen Li,<sup>a,b</sup> Leixin Yang,<sup>a,b</sup> Yanxiong Ren,<sup>a,b</sup> Yutao Liu,<sup>a,b</sup> Xu Liang,<sup>a,b</sup> Zheyuan Guo,<sup>a,b</sup> Hongjian Wang,<sup>a,b</sup> Hong Wu<sup>abc\*</sup> and Zhongyi Jiang<sup>abd\*</sup>*



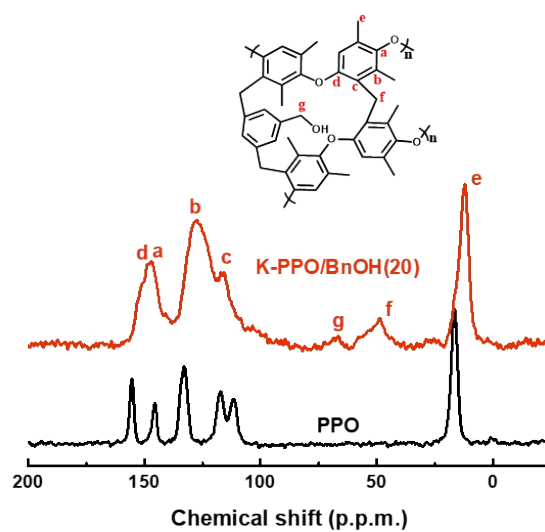
**Fig. S1.** Thermogravimetric curves for K-PPO and K-PPO/BnOH(20) membranes.



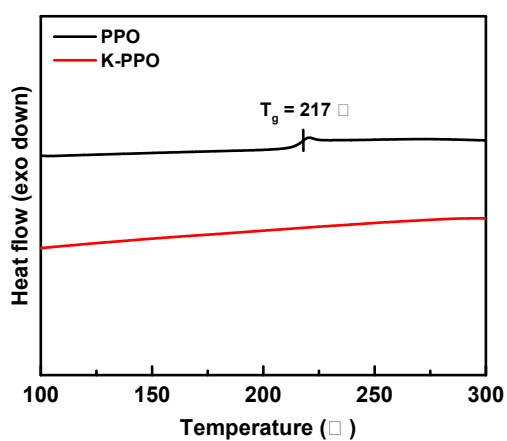
**Fig. S2.** Optical images of (a) PPO/BnOH composite membrane and (b) K-PPO/BnOH(20) membrane.



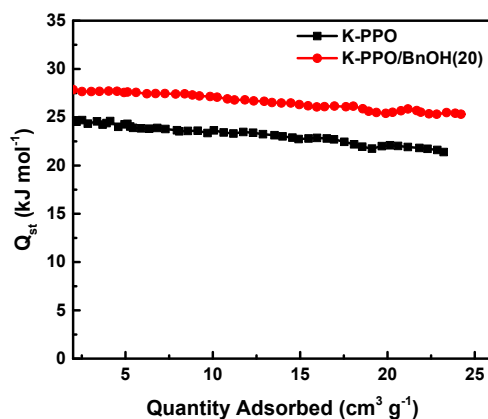
**Fig. S3.** Cross-sectional SEM images of (a) PPO/BnOH composite membrane and (b) K-PPO/BnOH(20) membrane.



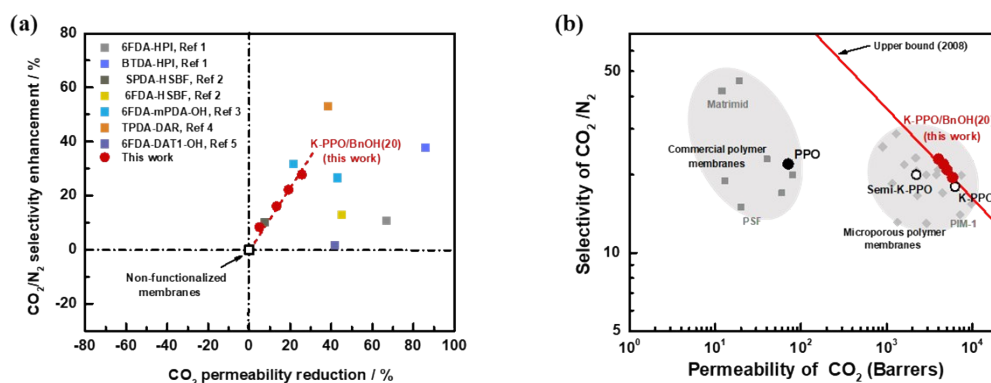
**Fig. S4.** Solid-state  $^{13}\text{C}$  CP/MAS NMR spectra of PPO and K-PPO/BnOH(20) membranes.



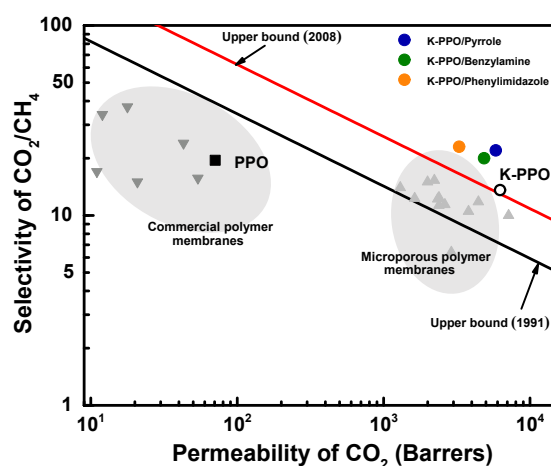
**Fig. S5.** Thermal DSC analysis of the PPO and K-PPO membranes.



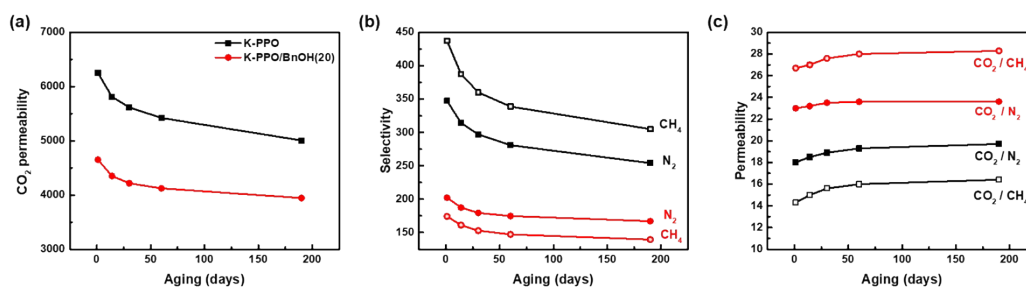
**Fig. S6.** Isosteric heats of adsorption of CO<sub>2</sub> for K-PPO and K-PPO/BnOH(20) membranes



**Fig. S7.** (a) Comparison of the CO<sub>2</sub>/N<sub>2</sub> performances enhancement for hydroxyl-functionalized membranes. The red dashed line is drawn to guide the eye. (b) Comparison of CO<sub>2</sub>/N<sub>2</sub> separation performance between our work and others<sup>1-5</sup> in Robeson plots.



**Fig. S8.** Exploratory results of gas separation performances of three other knitted microporous polymer membranes using benzylamine, pyrrole, and 1- phenylimidazole building units.



**Fig. S9.** (a)  $\text{CO}_2$  permeability, (b)  $\text{N}_2$  and  $\text{CH}_4$  permeability, and (c)  $\text{CO}_2/\text{N}_2$  and  $\text{CO}_2/\text{CH}_4$  selectivity for K-PPO and K-PPO/BnOH(20) membranes as functions of time.

**Table S1** the crosslinking degree in different membranes

Membrane	m <sub>1</sub> (g)	m <sub>2</sub> (g)	Weight gain (wt%)	Crosslinking degree %
Semi-K-PPO	0.7589	0.8146	7.35	126
K-PPO	0.7650	0.8525	11.43	196

According to the previous reports<sup>6</sup>, we calculate the molar ratio of -CH<sub>2</sub>- bridge to PPO unit by the weight gain after crosslinking reaction. Pristine membranes were weighed before crosslinking reaction to determine weight (m<sub>1</sub>). After reaction, membranes were reweighed (m<sub>2</sub>). The membrane weight was measured by an electronic balance (OHAUS, CP224C). Each membrane was tested for three times and the average value was adopted. The weight gain (wt%) was calculated based on the following equations:

$$\text{weight gain} = (m_2 - m_1) / m_1 \times 100\% \quad (\text{S1})$$

The crosslinking degree was calculated by:

$$\text{Crosslinking degree} = 2N_c/N_p \quad (\text{S2})$$

where N<sub>c</sub> refers the amount of crosslinking bridge. N<sub>p</sub> is the amount of repeat polymer units.

## References

- [1]. C. H. Jung and Y. M. Lee, *Macromol Res*, 2008, 16, 555-560.
- [2]. X. H. Ma, O. Salinas, E. Litwiller and I. Pinnau, *Polym. Chem.*, 2014, 5, 6914-6922.
- [3]. N. Alaslai, B. Ghanem, F. Alghunaimi, E. Litwiller and I. Pinnau, *J. Membr. Sci.*,

2016, 505, 100-107.

[4]. N. Alaslai, B. Ghanem, F. Alghunaimi and I. Pinnau, *Polymer*, 2016, 91, 128-135.

[5]. N. Alaslai, X. Ma, B. Ghanem, Y. Wang, F. Alghunaimi and I. Pinnau, *Macromol. Rapid. Commun.*, 2017, 38, 1700303.

[6]. H. Liu, S. Li, H. Yang, S. Liu, L. Chen, Z. Tang, R. Fu and D. Wu, *Adv. Mater.*, 2017, 29, 1700723.