Supporting Information

A Bio-inspired CO_2-philic Network Membrane for Enhanced Sustainable Gas Separation

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■ S1. Digital photos of PEO-526 treated at various stages. (a) original PEO-526, (b) treatment at 80°C for 6 hours, (c) and (d) treatment at 160 °C for 2 hours

To determine the possibility of PEO-526 forming cross-linked structure without DA, 8 g PEO-526 was heated at 80°C for more than 6 hours with stirring and the temperature was raised to 120 °C for another 6 hours, and then heated at 160 °C for 2 hours. The digital photos are shown below. Obviously, the pure PEO-526 cannot be cross-linked simply by thermal treatment without DA

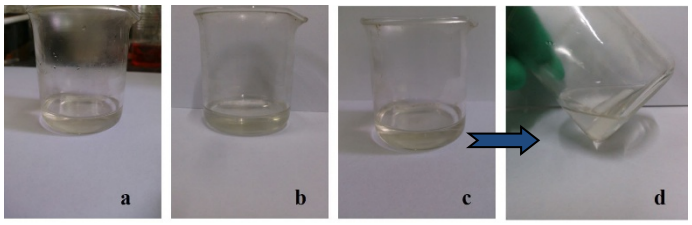

Fig. S1 Digital photos of PEO-526 treated at various stages. (a) original PEO-526, (b) treatment at 80°C for 6 hours, (c) and (d) treatment at 160 °C for 2 hours

■ S2. Digital photos of DA/PEO mixture treated (a) at 120 °C for 6 hours, (b) at 160 °C for 2 hours

To clarify the effect of oxygen environment on the reaction, PEO-526 and DA were mixed directly and then the mixture was treated under vacuum at 80 °C for 6 hours, 120 °C for another 6 hours, and 160 °C for 2 hours. Finally, canary yellow, hyaline solid was obtained, shown as below. It is clear that the reaction between DA and PEO-526 can readily happen without any oxygen (under vacuum).

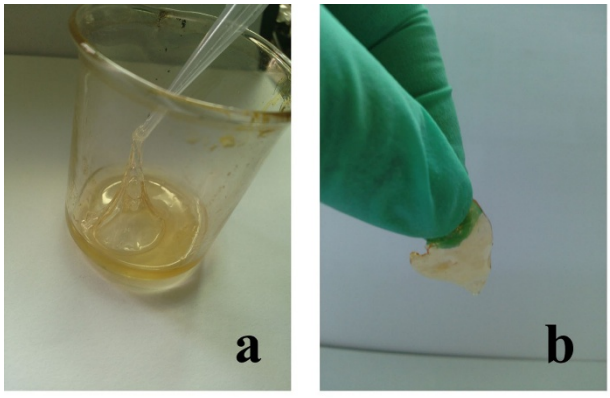

Fig. S2 Digital photos of DA/PEO mixture treated (a) at 120 °C for 6 hours, (b) at 160 °C for 2 hours

■ S3. Calculated activation energy of D-PEO and D-PEO-I

Table S1. Calculated activation energy (E_P, E_D and ΔH) of D-PEO and D-PEO-I

	D-PEO				D-PEO-I			
	H_2	N_2	CH_4	CO_2	H_2	N_2	CH_4	CO_2
E_P (kJ/mol)	39.4±0.5	50.2±0.8	58.6±0.6	34.0±0.5	28.5±0.3	33.8±0.5	31.2±0.5	18.1±0.2
E_D (kJ/mol)	-	24.8±0.8	45.3±1.5	54.5±0.8	-	31.0±0.3	29.7±0.3	34.4±0.1
ΔH (kJ/mol)	-	25.7±0.5	13.6±1.0	-22.2±0.3	-	2.8±0.3	1.5±0.2	-16.3±0.2

■ S4. Pure gas permeability of D-PEO and D-PEO-I compared with other PEO-based membranes.

Table S2. Pure gas permeability of D-PEO and D-PEO-I compared with other PEO-based membranes

	H_2	N_2	CH_4	CO_2
D-PEO*	**13.7**	**1.1**	**2.5**	**56**
D-PEO-I*	**40.9**	**8.3**	**22.1**	**309**
LCM[1]	**21.5**	**2.6**	**9.8**	**170**
Am PEO[2]	**21.0**	**3.0**	**7.1**	**143**
XLPEGDA[2]	**15.0**	**2.2**	**5.8**	**110**

* This work

■ S5. Pure gas solubility (a) and diffusivity (b) of D-PEO-I compared with LCM at 35 °C and 10 atm.

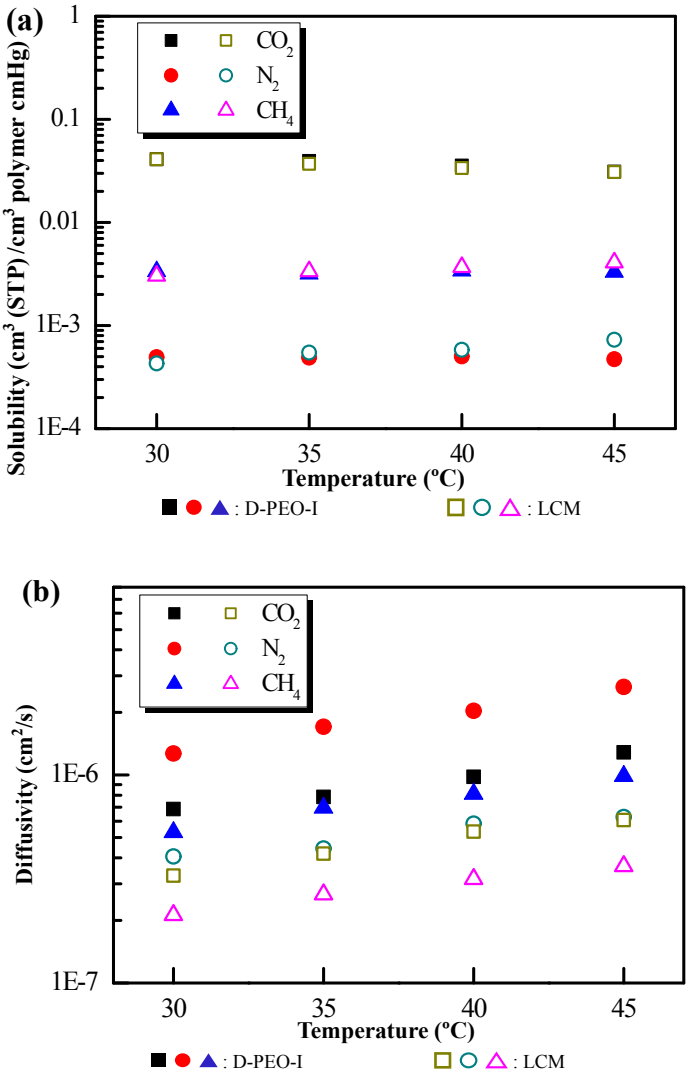

Fig. S3 Pure gas solubility (a) and diffusivity (b) of D-PEO-I compared with LCM [1] at 10 atm.

References:

[1] L. Shao, S. Quan, X. Q. Cheng, X. J. Chang, H. G. Sun and R. G. Wang, *Int J Hydrogen Energ*, 2013, **38**, 5122-5132.

[2] H. Lin and B. D. Freeman, *J Membr Sci*, 2004, **239**, 105-117.