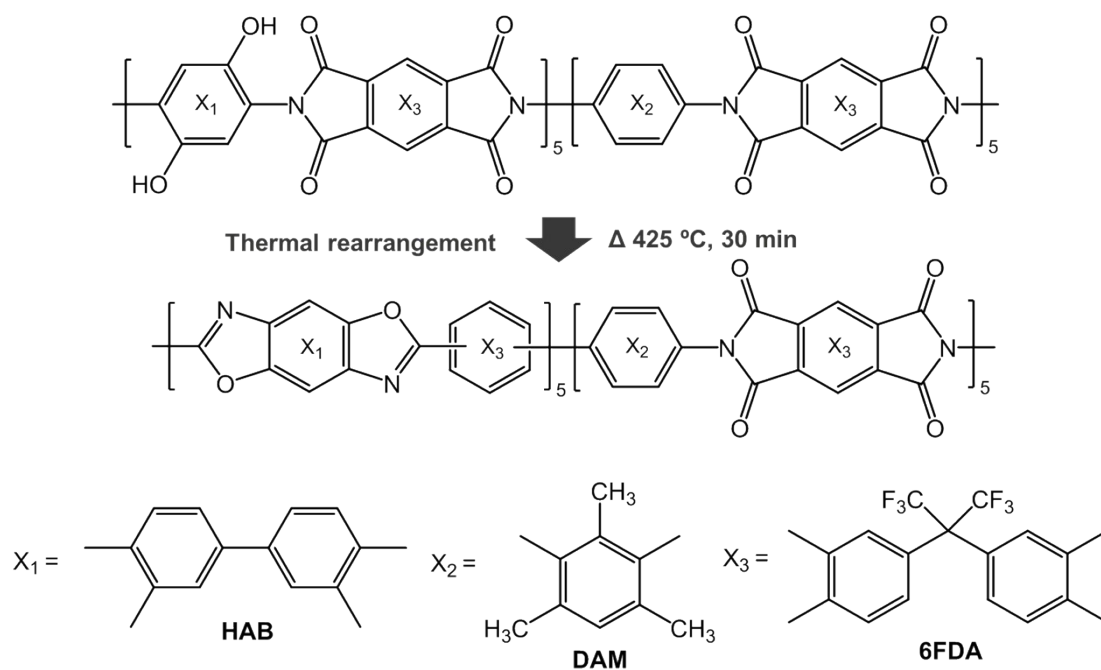


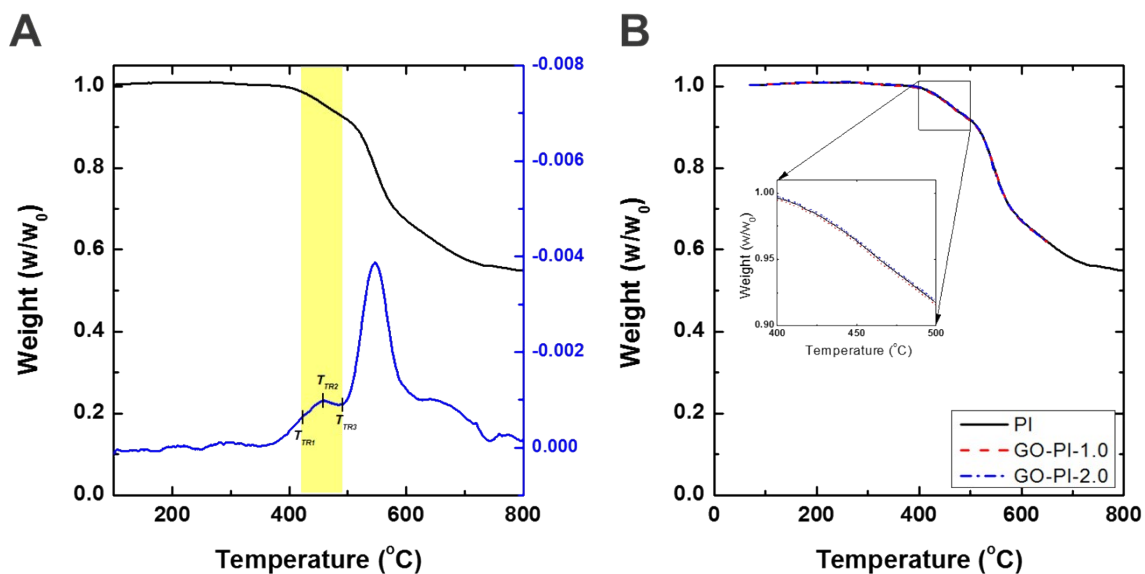
## Supporting Information

### **Highly permeable thermally rearranged polymer composite membranes with a graphene oxide scaffold for gas separation**

*Seungju Kim, Jue Hou, Yuqi Wang, Ranwen Ou, George P. Simon, Jong Geun Seong, Young Moo Lee, and Huanting Wang\**

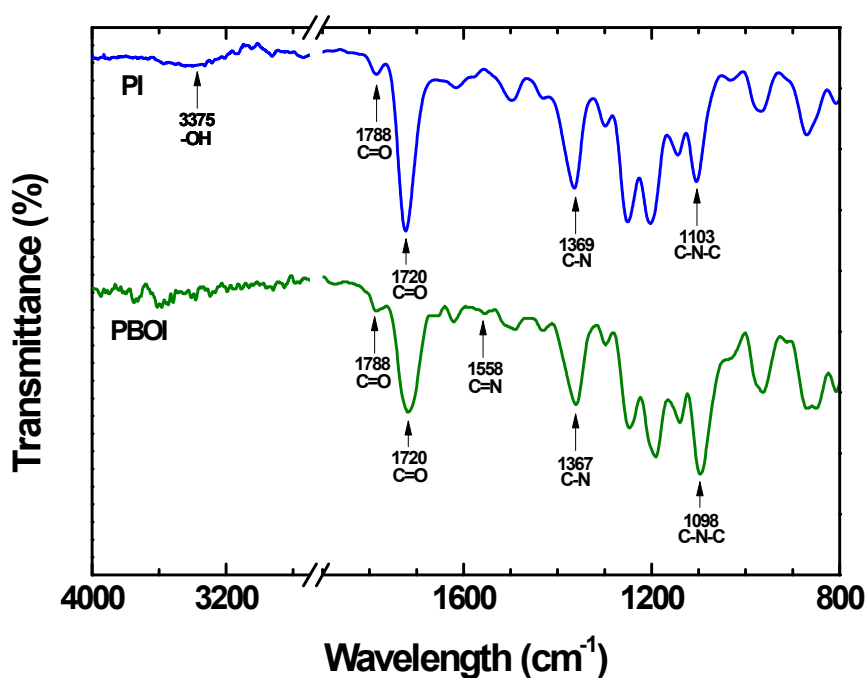


**Fig. S1** Chemical structures of the monomers, PI and TR-PBOI membranes.



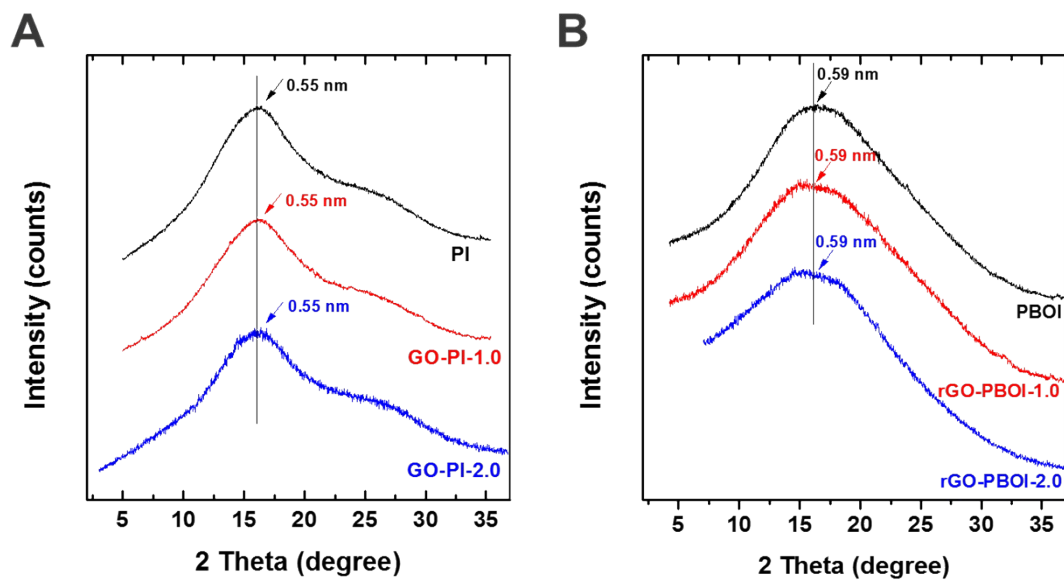
**Fig. S2** Thermogravimetric analysis (TGA) curves of PI and GO-PI freestanding membranes.

The first weight loss between 420 °C to 490 °C represents thermal conversion from PI to PBOI. Thermal rearrangement temperatures ( $T_{TR1}$ ,  $T_{TR2}$ , and  $T_{TR3}$ ) were determined from its derivation curve and a duration time at 425 °C was determined from its isotherm curve at 425 °C by comparing to theoretical and experimental weight loss of thermal conversion.

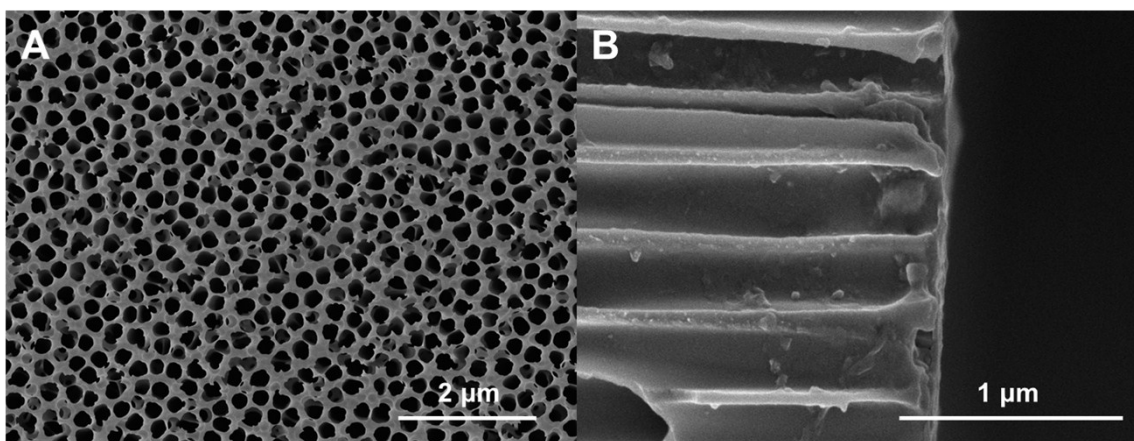


**Fig. S3** Fourier transform infrared spectroscopy (ATR-FTIR) spectra of PIs and TR-PBOIs.

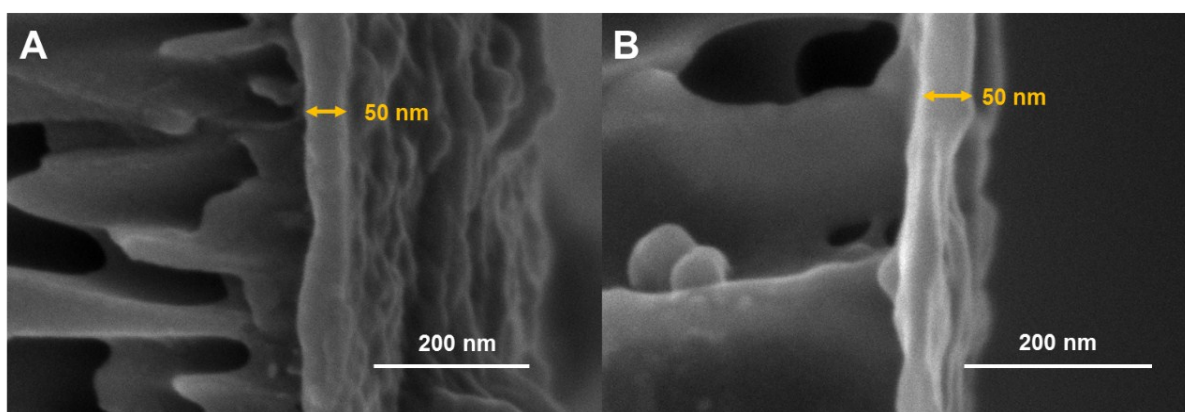
Typical bands of imide at 1788 cm<sup>-1</sup>, 1720 cm<sup>-1</sup>, 1369 cm<sup>-1</sup>, and 1103 cm<sup>-1</sup> were found in both PIs and PBOIs and a broad band of hydroxyl groups (3375 cm<sup>-1</sup>) was only observed in PIs. Hydroxyl groups in PI and GO were disappeared during thermal rearrangement and a typical band of benzoxazoles (1558 cm<sup>-1</sup>) was appeared.



**Fig. S4** X-ray diffraction (XRD) curves of PIs and TR-PBOIs.



**Fig. S5** Scanning electron microscope (SEM) images of AAO substrate (a) surface and (b) cross-section.



**Fig. S6** Scanning electron microscope (SEM) images of (a) GO-PI-2.0 and (b) rGO-PBOI-2.0 prepared from 2.0 wt% solution in THF.

**Table S1.** Single gas permeability and permeance of TR-PBOI freestanding membranes.

	H <sub>2</sub>	CO <sub>2</sub>	O <sub>2</sub>	N <sub>2</sub>	CH <sub>4</sub>	H <sub>2</sub> /N <sub>2</sub>	CO <sub>2</sub> /N <sub>2</sub>	O <sub>2</sub> /N <sub>2</sub>	CO <sub>2</sub> /CH <sub>4</sub>
Permeability (Barrer*)	203	185	32	7.5	5.3				
Permeance (GPU*)	4.1	3.7	0.64	0.15	0.11				
Ideal selectivity						27	26	4.2	35

\*1 Barrer =  $10^{-10}$  cm<sup>3</sup>(STP) cm/cm<sup>2</sup> s cmHg =  $3.35 \times 10^{-16}$  mol m/m<sup>2</sup> s Pa

\*1 GPU =  $10^{-6}$  cm<sup>3</sup>(STP)/cm<sup>2</sup> s cmHg =  $3.35 \times 10^{-10}$  mol/m<sup>2</sup> s Pa

**Table S2.** Single gas permeance of rGO-PBOI membranes.

	Gas permeance (GPU)					Ideal selectivity			
	H <sub>2</sub>	CO <sub>2</sub>	O <sub>2</sub>	N <sub>2</sub>	CH <sub>4</sub>	H <sub>2</sub> /N <sub>2</sub>	CO <sub>2</sub> /N <sub>2</sub>	O <sub>2</sub> /N <sub>2</sub>	CO <sub>2</sub> /CH <sub>4</sub>
rGO-PBOI-1.0	1847	1784	363	101	55.1	18.3	17.7	3.6	32.4
rGO-PBOI-1.5	1505	1450	280	76	41.7	19.8	19.1	3.7	34.8
rGO-PBOI-2.0	1233	1149	201	56.4	29.4	21.3	20.4	3.6	39.1
rGO-PBOI-2.2	986	872	150	41.7	20.3	23.7	20.9	3.6	43.0
rGO-PBOI-2.4	761	673	125	30.7	15.1	24.8	21.9	4.1	44.6

**Table S3.** Mixed gas permeance of rGO-PBOI-1.0 membranes.

Gas permeance (GPU)					Separation factor	
CO <sub>2</sub> - CH <sub>4</sub> pair		CO <sub>2</sub> - N <sub>2</sub> pair				
CO <sub>2</sub>	CH <sub>4</sub>	CO <sub>2</sub>	N <sub>2</sub>		CO <sub>2</sub> /CH <sub>4</sub>	CO <sub>2</sub> /N <sub>2</sub>
1780	50.7	1785	98.1		35.1	18.2