Pebax® 1041 supported membranes with carbon nanotubes prepared via phase inversion for CO_2/N_2 separation

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Membrane characterization

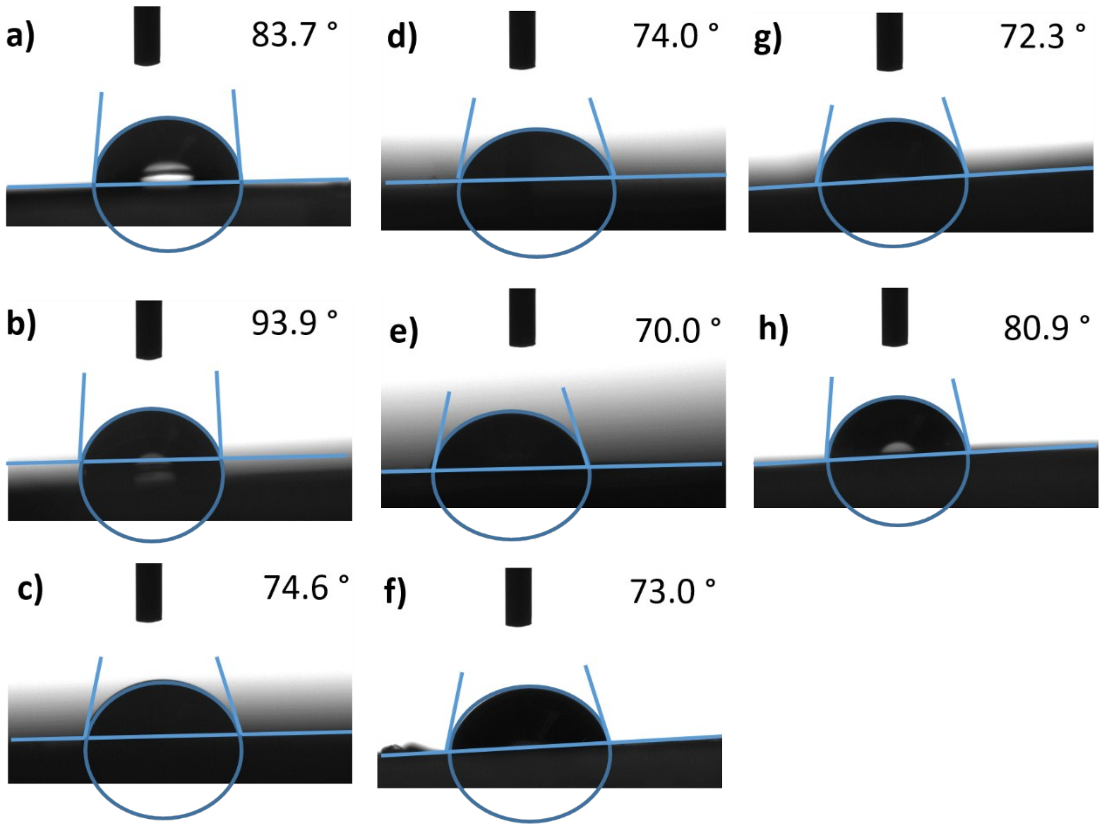

Figure S1. Contact angle measurements of PSF supports (a), supported membranes prepared with 1.5 µm (b), 2.6 µm (c), 3.6 µm (d), 4.1 µm (e) and 8.0 µm (with MWCNTs) (f) of Pebax® 1041, self-supported Pebax® 1041 membranes (g) and PSF supports coated with PTMSP (h).

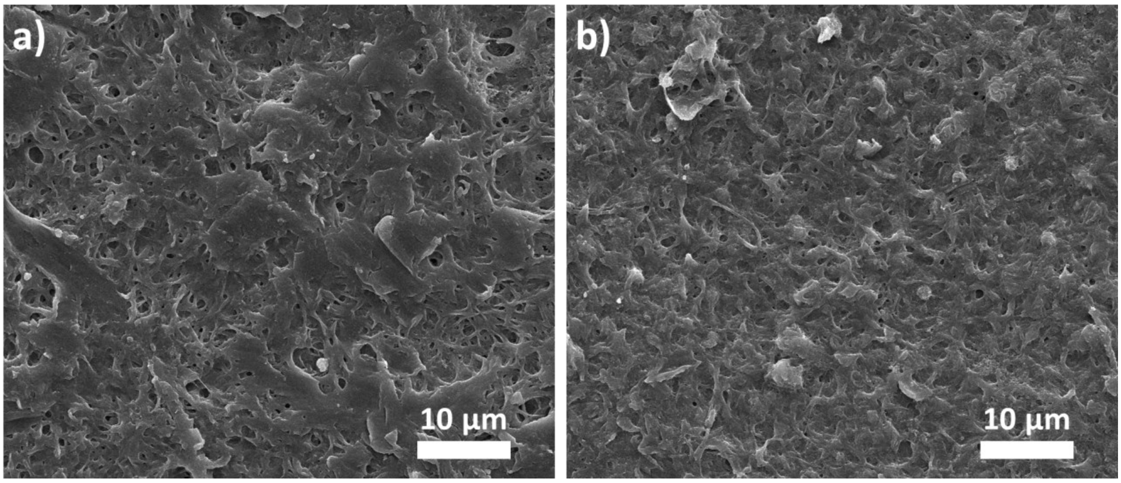

Figure S2. SEM images of the surface of supported membranes: pristine Pebax® 1041, Pebax® 1041 containing MWCNTs and MWCNTs in powder form.

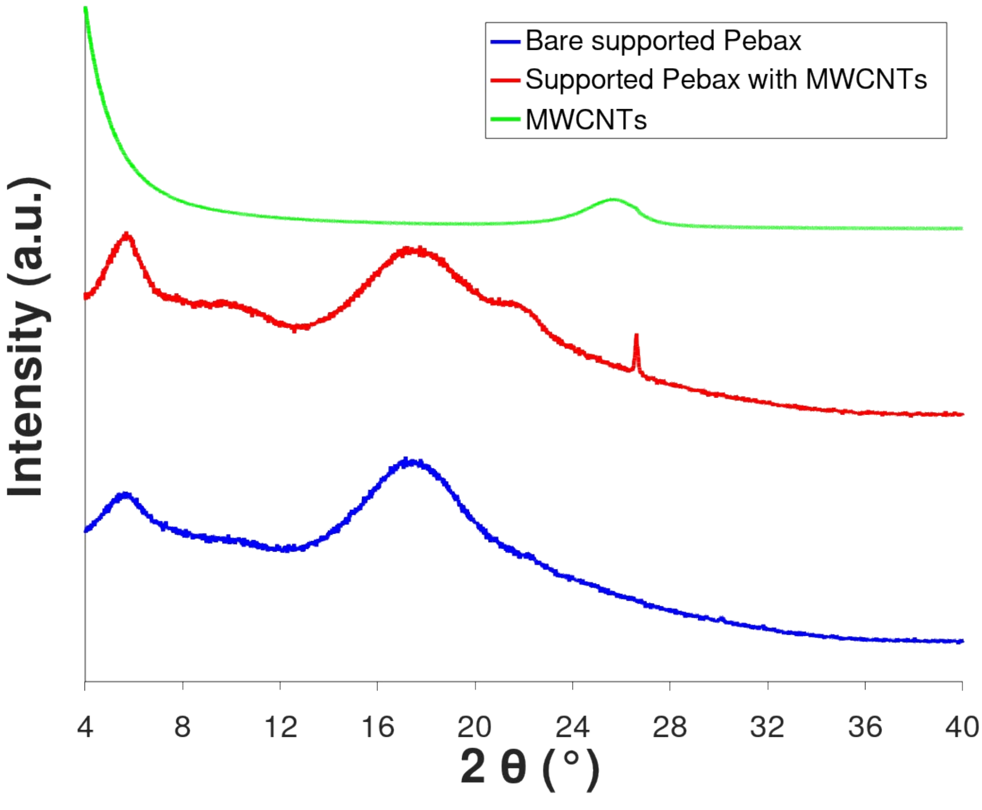

Figure S2. XRD analysis of supported membranes: pristine Pebax® 1041, Pebax® 1041 containing MWCNTs and MWCNTs in powder form.

Robeson upper-bound adapted

Table S1. Literature review with the values that defined the CO_2/N_2 upper-bound in 2008, including membrane thickness, CO_2 permeability in Barrer and CO_2/N_2 selectivity. The calculated CO_2 permeance in GPU is given.

Polymer	**Thickness (µm)**	**CO_2 permeability (Barrer)**	**CO_2 permeance (GPU)**	**α (CO_2/N_2)**	**Ref.**
Phosphazene	100	250	2.5	62.5	1

Modified poly(dimethylsiloxane)	150	2000	13.3	34.2	[2]
PIM-7	28	1100	39.3	26.2	[3]
PIM-1	46	2300	50	25	[3]

The Robeson upper-bound, revisited in 2008[4] was defined from pure component permeability data of dense membranes, allowing the determination of the state-of-the-art limits for polymeric membrane gas separation. The upper-bound relationship is expressed by $P_i = k \cdot \alpha_{ij}^{n}$, where P_i is the permeability of the more permeable gas, α is the separation factor (P_i/P_{ij}) and n is the slope of the log–log limit. It was observed that $-1/n$ vs. d_{ij} (where d_{ij} is the difference between the gas molecular diameters ($d_j - d_i$)) yielded a linear relationship. Since gas permeability was defined in Barrer, we have calculated an CO_2/N_2 upper-bound relationship in GPU, using the values from the literature that defined the original upper-bound but changing the flow values from Barrer into GPU (see Table S1). These values were represented in Figure S1 and fitted to a logarithmic equation, resulting in the following upper-bound relationship: $P_{CO_2} = 512447 \cdot \alpha_{CO_2/N_2}^{-2.967}$. A factor k of 512447 GPU was obtained and the slope n of -2.967 was near the value found in the original publication, implying that its inverse corresponds to the difference between the gas molecular diameters of CO_2 (0.330 nm) and N_2 (0.364 nm).

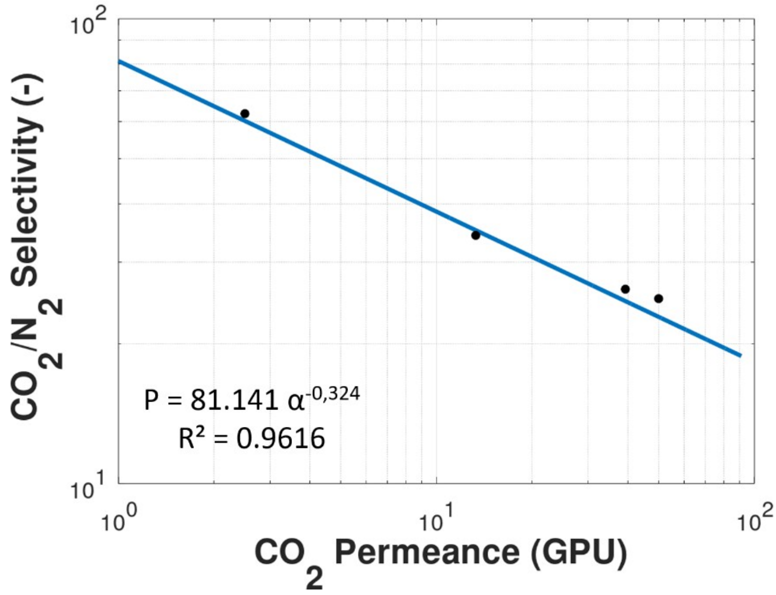

Figure S4. CO_2/N_2 bound defined in GPU at 35 ^{0}C. Squares represent the values in GPU of Table S1. The fitting equation of the linear fitting is also given with the R^2 value

Comparison with the literature

Table S2. Pebax® based membrane performances for CO_2/N_2 separation

Polymer	Filler	Operating temperature (°C)	Feed pressure (bar)	CO_2 permeability (Barrer)	CO_2/N_2 selectivity (-)	Year and reference
Pebax® 1657	-	35	3	110	36	2019[5]
Pebax® 1657	-	20	10	80.6	48	2019[6]
	20 wt% ZIF-8			156	40.5	
Pebax® 1657	-	RT	4	52.1	93.0	2019[7]
	0.2 wt% GO			40.8	107	
Pebax® 1657 /PEG-MEA	-	35	1-9	572	43	2019[8]
	0.3wt% GO			600	55.8	
Pebax® 2533	-	RT	10	202	22.8	2019[9]
Pebax® 1041	1 wt% MWCNTs	35	3	24	22.6	This work

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