Interface-governed, highly efficient CO_2/N_2 gas separation in thin-film composite nanomembranes

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Gas separation membranes suitable for economically feasible post-combustion CO_2 capture should combine the moderate CO_2/N_2 selectivity (20-60) with a substantial values of CO_2 permeances (>1000 GPU). Most of membranes tailored to have these properties are so-called thin-film composite membranes (TFCM) containing different layers it structure, each having a specific function (porous support, gutter, selective, protective layer). Due to highest resistance towards gas transport selective layers in TFCM are commonly fabricated with thicknesses below 100 nm [1].

In this work, in order to achieve high CO_2 permeances in TFCM we have fabricated ultra-thin selective layers (2-20 nm) composed of hydrophilic and CO_2-selective block-copolymer (Pebax MH-1657). It was deposited on the O_2-plasma activated surface of much thicker (~400 nm) gutter layer composed of polydimethylsiloxane (PDMS). The structure was subsequently transferred on the polyacrylonitrile (PAN) microporous support to complete the TFCM (Fig.1). We have found that contrary to the theoretical predictions (resistance in series model) for the layered membrane [2], highly selective CO_2/N_2 separation membrane was achieved in the TFCM with ultra-thin selective layer. Critical role to achieve this selectivity was attributed to the specific interface formed between selective and gutter layers which was controlled by the duration of oxygen plasma treatment (PDMS activation).

Permeances of CO_2 in the developed TFCM were between 1000-3000 GPU and CO_2/N_2 selectivities between 30-100, providing the gas separation parameters that are within optimal range for cost-efficient CO_2 capture in post-combustion processes. Detailed characterization of the interface revealed the chemical structure of the outermost membrane surface suggesting the blending of the ultra-thin Pebax-1657 layer with activated surface of PDMS. This nano-thick blend layer contributed to the overall selectivity of the membrane significantly exceeding the selectivity that can be achieved by Pebax-1657 alone. Formed interface demonstrated stable gas separation with a moderate change of performance over the one year.

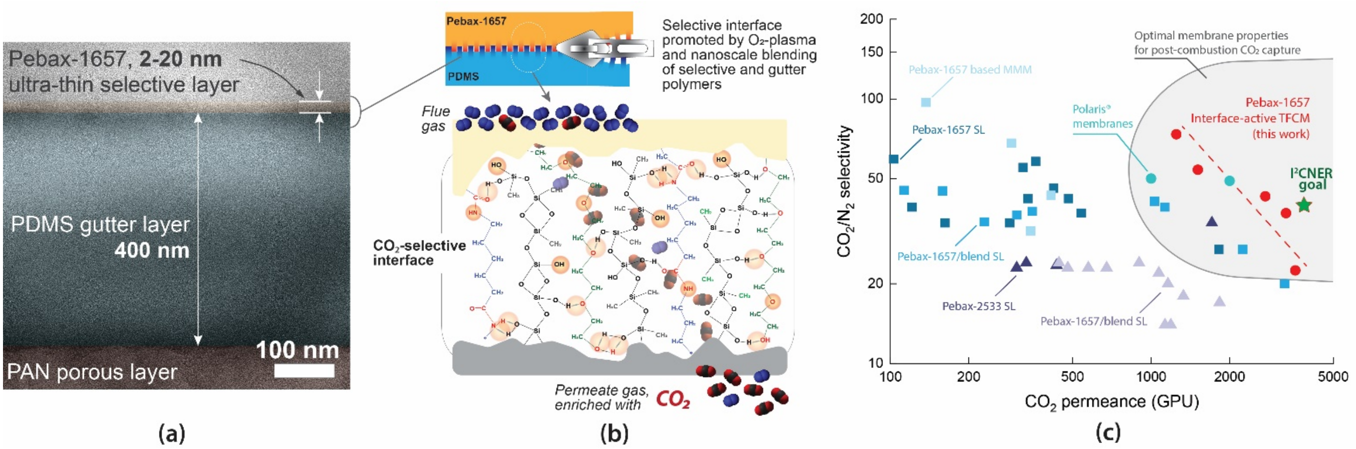

Figure 1. (a) Cross-section TEM image of the tri-layer TFCM (Pebax-1657/PDMS/PAN) with ultimately thinned selective layer; **(b)** structure of the active, CO_2-philic interface between layers; **(c)** CO_2/N_2 separation performance of series of Pebax-1657/PDMS membranes developed in I²CNER CCU in the context of the economically optimal membrane parameters (grey area) according to [3].

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