Single-phase covalent organic frameworks membranes make CO2-selective separation possible

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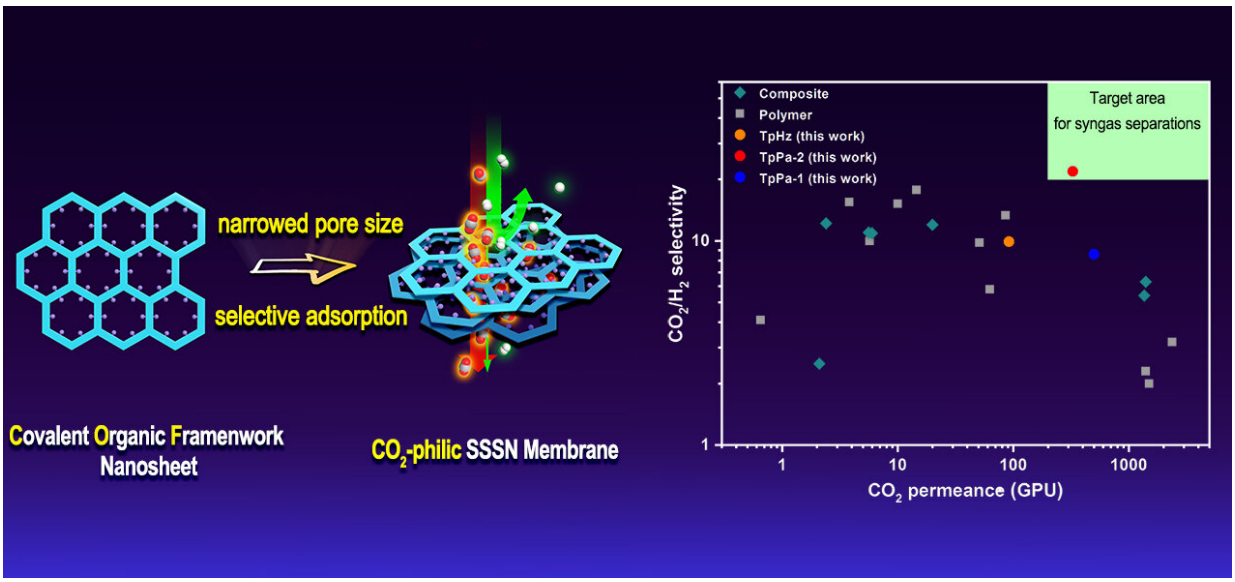

Illustration of superior CO2-selective COF nanosheet membrane for high-performance CO2/H2 separation. Credit: Peng Yuan

A research group led by Prof. Yang Weishen and Dr. Peng Yuan from the Dalian Institute of Chemical Physics (DICP) of the Chinese Academy of Sciences (CAS) developed a novel strategy to fabricate single-phase CO_2-selective covalent organic frameworks (COFs) nanosheet membranes featured with both high CO_2/H_2 separation factors

and high CO^2 permeance.

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Two-dimensional (2D) COFs, a class of crystalline organic skeletons possessing permanent one-dimensional pores, are promising to serve as high-quality membrane materials in CO_2 capture field. However, their inherent pore sizes (>0.8 nm) are too large to provide desired molecular sieve capability for gas separations currently.

The researchers exfoliated three kinds of 2D COFs, that is, TpPa-1, TpPa-2 and TpHz, with an average particle size of 3 um and different pore sizes into 2 nm-thick nanosheets. Intact crystallinities and functionalities were well-retained after exfoliation treatment.

They found that even the TpHz with relatively smallest inherent pore size could not exhibit desired gas separation performance in a form of conventional bulk membrane. With ultrathin nanosheets served as membrane building units, they developed novel COF nanosheet membranes exhibiting distinctive staggered stacking micro-structures.

"Just like the fishing net, you can irregularly pile up several nets to catch tiny fish," said Dr. Peng.

The intriguing membrane structures in collaboration with inherent CO_2-selective adsorption capacities of COF frameworks endowed the COF nanosheet membranes with permeation priority of large CO_2 molecules from mixed CO_2/H_2 gas based on a surface diffusion mechanism.

Among the membranes, TpPa-2 nanosheet membranes with medium pore size showed the highest CO_2/H_2 separation factor and CO_2

permeance, which reached the target with commercial feasibility for syngas separations.

The researchers identified that the TpPa-2 nanosheet membranes exhibited proper narrowed pores derived from the staggered stacking patterns, which allowed exactly two columns of CO_2 molecules to pass through and maximized the blocking effect for H2 passage.

This study provides a new pore engineering strategy for the design and manufacture of high-performance COF membranes in gas separation realm.

"Taking full advantage of this pore-engineering strategy, different categories of COFs with diversified pore structures can enable specified membrane-based gas separations," said Prof. Yang.

More information: Pengyuan Wang et al, Single-Phase Covalent Organic Framework Staggered Stacking Nanosheet Membrane for CO 2 -Selective Separation, *Angewandte Chemie International Edition* (2021). DOI: 10.1002/anie.202106346

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