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Zoom: https://gtiit.zoom.us/meeting/register/5zHF--pBQDKKPUjyfH-AaQ

Metal-Organic Framework-based Mixed Matrix Membranes for Gas Separation

Zhihong Lin

PhD Final-Seminar

Advisor: Prof. Moris S. Eisen
Prof. Xuezhong He
The Schulich Faculty of Chemistry, Technion – Israel Institute of Technology
The Wolfson Department of Chemical Engineering, Technion – Israel Institute of Technology
Department of Chemical Engineering, Guangdong Technion – Israel Institute of Technology

Abstract: Membrane-based post-combustion CO_2 capture relies on solution–diffusion, where separation is governed by the coupled effects of gas solubility (sorption selectivity) and diffusivity (molecular discrimination). MOF-based mixed-matrix membranes (MMMs) can enhance both, yet practical outcomes are often limited by two bottlenecks: (i) poor filler dispersion and interfacial continuity, leading to aggregation/sedimentation and nonselective voids; and (ii) insufficient intrinsic CO_2 affinity or sieving in the filler–polymer microstructure to generate strong solubility–diffusivity synergies. To overcome these barriers, we develop covalently grafted polymer of intrinsic microporosity (PIM)-based polymer-brush MOF fillers (50%cPIM/cPIM@UiO-66-NH_2) to enable a continuous, void-free interface. Tunable brush chemistry (–CN/–COOH) and geometry (thickness, grafting density) with the preserved MOF pore network enable filler-polymer wetting and suppress filler–filler coagulation. A parameterized brush-interaction model unifies steric repulsion, van der Waals attraction, and a normalized wetting preference, capturing dispersion/continuity and linking them to enhanced CO_2 separation performance. Building on the principle that interfacial cohesion and continuity are prerequisites for realizing microstructural advantages, we then apply thermal cyclotrimerization in PIM-1/MAF-stu-1 MMMs to simultaneously tailor microporosity and strengthen MOF–polymer interactions via triazine formation. The continuity-enhanced interface boosts accessible microporosity and discrimination, achieving CO_2 permeabilities >10,000 Barrer with CO_2/N_2 selectivity increasing from 18 to 46, and reducing specific CO_2 capture cost by 43.9%. Finally, as a complementary route to strengthen the intrinsic CO_2 affinity/transport contribution once a cohesive interface is secured, Ag^+ is introduced (double-solvent doping) to add CO_2-binding sites, enabling synergistic facilitated transport and pore-regulated sieving beyond established CO_2/N_2 upper bounds. Overall, these studies establish a continuity-driven design principle: a highly compatible and continuous filler–polymer interphase is essential to reliably translate filler chemistry and microporosity into high-performance CO_2 separations.