RESEARCH ARTICLE

Carbon Capture Using Amine-Functionalized Metal-Organic Framework Membranes

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Abstract: Post-combustion carbon capture from flue gas requires selective CO_2 separation under humid conditions at low partial pressures. We report amine-functionalized MOF membranes (UiO-66-NH_2-PEI) fabricated by interfacial polymerization on porous α-alumina supports, achieving CO_2/N_2 selectivity of 68.5 and CO_2 permeance of 1,420 GPU at 55°C with 15 vol% CO_2 feed mimicking coal-fired power plant flue gas. The polyethyleneimine (PEI) grafting density was optimized at 2.8 mmol/g, balancing chemisorption capacity with diffusion kinetics. Long-term permeation tests over 500 hours show <5% decline in selectivity under 85% relative humidity, attributed to the hydrophobic $Zr_6O_4(OH)_4$ backbone shielding amine sites from water competition. Techno-economic analysis indicates capture costs of $42/tonne CO_2 at 90% capture rate for a 500 MW plant.

1. Introduction

Anthropogenic CO_2 emissions from fossil fuel combustion must be drastically reduced to limit global warming to 1.5°C above pre-industrial levels. Amine-based liquid absorption remains the dominant commercial carbon capture technology, but it suffers from high regeneration energy penalties (3-4 GJ/tonne CO_2), equipment corrosion, and amine degradation. Membrane-based separation offers a compact, modular alternative, and metal-organic frameworks (MOFs) with tunable pore chemistry provide exceptional gas separation performance when fabricated as thin-film composite membranes.

2. Membrane Synthesis and Testing

UiO-66-NH_2 seed layers were deposited on polished α-alumina tubes (OD 10 mm) by secondary growth, followed by in situ PEI grafting via glutaraldehyde crosslinking at PEI concentrations of 0.5-5.0 wt%. The resulting membranes (active layer ~800 nm) were characterized by SEM, XRD, and TGA. Single-gas and mixed-gas permeation was measured in a Wicke-Kallenbach setup at 35-75°C with humidified feeds (RH 0-90%).

Table 1. Mixed-gas separation performance of amine-functionalized MOF membranes at 55°C, 15 vol% CO_2, 85% RH

Membrane	PEI Loading (mmol/g)	CO_2 Permeance (GPU)	CO_2/N_2 Selectivity	CO_2/H_2O Selectivity

Membrane	PEI Loading (mmol/g)	CO_2 Permeance (GPU)	CO_2/N_2 Selectivity	CO_2/H_2O Selectivity
UiO-66-NH_2	0	2,850	18.2	4.5
UiO-66-NH_2-PEI (low)	1.2	980	52.1	28.3
UiO-66-NH_2-PEI (opt.)	**2.8**	**1,420**	**68.5**	**45.2**
UiO-66-NH_2-PEI (high)	4.5	520	71.3	52.8

3. Results and Discussion

Amine functionalization transforms UiO-66-NH_2 from a size-sieving membrane into a facilitated transport membrane where CO_2 transport is enhanced by reversible carbamate formation at grafted amine sites. Figure 1 shows the trade-off between CO_2 permeance and selectivity as a function of PEI loading. Figure 2 presents the temperature-dependent separation performance, with optimal selectivity at 55°C where chemisorption strength and diffusivity are balanced.

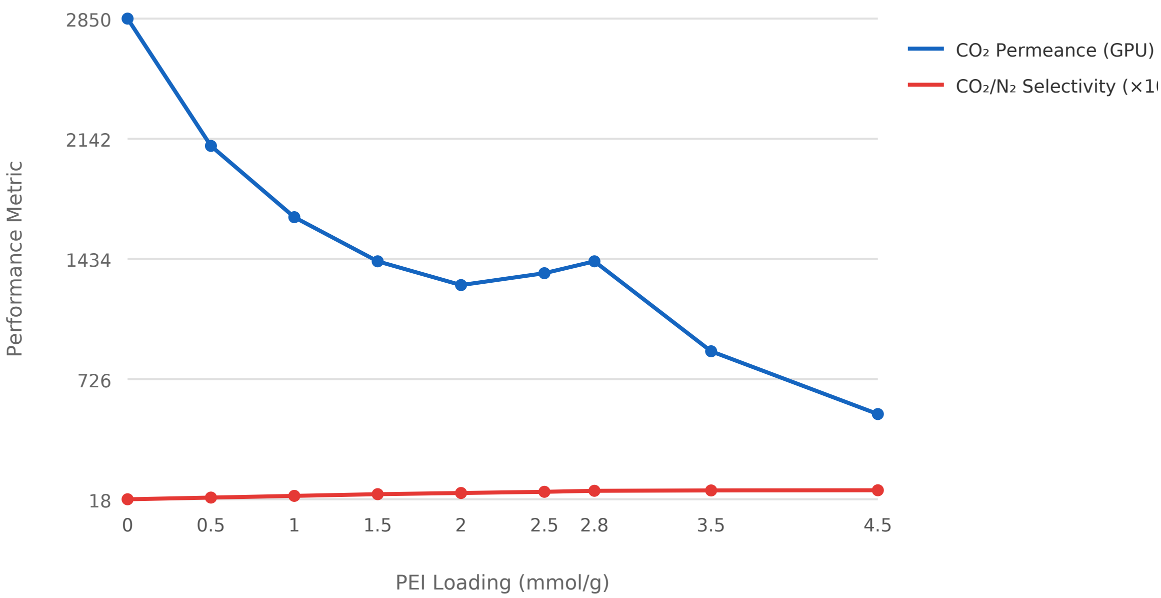

Figure 1. CO_2 permeance and CO_2/N_2 selectivity vs. PEI grafting density for UiO-66-NH_2 membranes

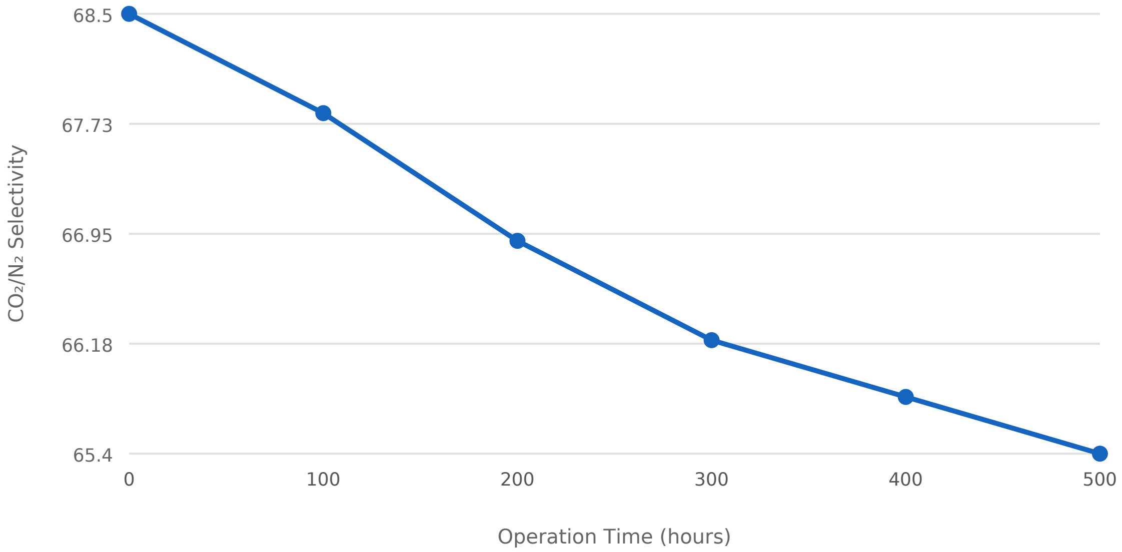

Figure 2. Long-term CO_2/N_2 selectivity stability under humid flue gas conditions (85% RH, 15 vol% CO_2, 55°C)

4. Conclusions

Amine-functionalized UiO-66-NH_2 MOF membranes achieve post-combustion CO_2 capture performance that exceeds the Robeson upper bound for polymeric membranes while maintaining stability under realistic humid flue gas conditions. The modular membrane contactor configuration enables retrofit deployment at existing power plants with reduced footprint compared to amine scrubbing columns. Future work will scale membrane area to pilot-plant modules and evaluate capture from natural gas combined-cycle flue gas.

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