From Fundamental Modeling to Prototype: The Development of a Hydrogen-Powered Hydroxide Exchange Membrane Carbon Capture System

Marco Colin Martinez

Advisor: Yushan Yan

Committee Members: Brian P. Setzler, Raul Lobo, Ajay Prasad, and Dongxia Liu

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102 Colburn Laboratory | Teams Meeting Link | Teams Password: U7nN9py2

Direct air capture is needed to remove carbon dioxide from ambient air and support net-zero emissions goals, but incumbent technologies remain limited by high capital cost and high-temperature regeneration. To be deployed at scale, direct air capture must approach costs below \$100/tCO_2 captured. Electrochemical direct air capture offers a potential alternative because it can operate at low temperature, use modular stack designs, and integrate with renewable electricity or hydrogen. To realize this potential, low-cost electrochemical direct air capture systems must operate under realistic environmental conditions and achieve high performance, including (i) high CO_2 flux, (ii) high CO_2 removal, (iii) low device energy consumption, and (iv) low air-side pressure drop. This work focuses on an H_2-powered hydroxide exchange membrane carbon capture system, or H_2-HEMCC, in which H_2 is the energy source; thus, device energy consumption is determined by electron efficiency. Overall, this dissertation advances H_2-HEMCC by improving the four key performance metrics for low-cost electrochemical direct air capture, validating operation under realistic weather conditions, and designing, constructing, and automating a direct air capture prototype for real-air operation.

In Chapter 2, we experimentally show improved CO_2 flux, CO_2 removal, and electron efficiency guided by a 1-D through-plane physics-based model. At a fixed air flow rate, CO_2 flux and CO_2 removal are linked: increasing current density can increase both, but it decreases electron

efficiency. To illustrate the importance of electron efficiency, values under 23% would make H_2 OPEX alone exceed \$100/tCO_2, while prior membrane-based electrochemical DAC systems have typically remained below 40%. To overcome this limitation, the model was used to diagnose high-flux losses and revealed that hydroxide leaks across the membrane before fully reacting with CO_2. Guided by this insight, a thicker interlayer increased the reactive zone for CO_2 and hydroxide and increased peak electron efficiency from 38.7% to a record-high value of 63.8% at higher flux. The model also showed that carbonate and bicarbonate transport losses dominate at low CO_2 flux, which was experimentally confirmed by showing that higher membrane resistance improves electron efficiency.

Chapter 3 demonstrates that H_2-HEMCC can operate across weather-relevant conditions, with performance measured from 5-40 °C and 50-90% relative humidity, in the context of a conceptual green methanol production plant. Lower cathode relative humidity improved electron efficiency, while device temperature had a smaller effect over the tested range. External heating would add energy demand and cost, so ambient-temperature operation is important for practical DAC. Based on this performance and its relevance to realistic operating conditions, 20 °C and 50% RH were selected as the DAC design basis for the green methanol process model. Experimental results guided the design of a green methanol process model integrating H_2-HEMCC with HEM-based water electrolysis, HEM-based CO_2 electrolysis, and catalytic methanol synthesis. The base-case e-methanol cost was \$1,440/t methanol, while an alternate case reduced the cost to \$712/t methanol, below the DOE target of \$800/t methanol.

Chapter 4 demonstrates a two-order-of-magnitude reduction in air-side pressure drop through cathode-frame designs guided by Darcy's law and advances H_2-HEMCC from cell-level testing toward a fully automated prototype. A techno-economic analysis showed that low-cost air delivery requires stack pressure drop below 100 Pa and favors propellers at scale. This motivated the design of low-pressure drop 3D-printed cathode frames, reducing pressure drop from 8090 Pa

to 18 Pa at the same active-area-normalized flow rate. These frames were integrated into a scaled H_2-HEMCC stack and balance-of-system with hydrogen supply, CO_2/H_2 recycle, CO_2 separation, and fully automated PLC control. The integrated system demonstrated real-air CO_2 capture and preliminary durability data, while techno-economic analysis identified a pathway to capture costs below \$100/tCO_2.