Leveraging System-Level Analyses and Techno-Economic Modeling to Inform the Viability of Electrochemically-Mediated CO_2 Separation

by

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Technical Summary

Replacing fossil fuels with renewable energy and removing carbon dioxide (CO_2) via carbon capture, utilization, and storage (CCUS) are essential strategies for addressing the climate crisis and achieving net-zero emissions by 2050. While renewable energy is predicted to supply more than 60% of net electricity generation in the United States by mid-century, it is also predicted that coal and natural gas plants will remain operational in the near term to meet growing energy demands. This, coupled with the persistence of hard-to-decarbonize processes, requires point-source capture technologies to mitigate remaining CO_2 emissions. State-of-the-art CO_2 separation systems are typically based on low efficiency, temperature-swing cycles that exploit the natural affinity of alkanolamines for CO_2 at ambient conditions. Alternatively, electrochemical capture systems may enable CO_2 removal from flue gas streams at higher energetic efficiencies while also offering more modular and scalable designs. However, direct comparisons between the thermochemical and electrochemical approaches are scant, likely due to the nascency of the latter.

In this thesis, I develop modeling frameworks that enable system-level comparisons of two types of electrochemical CO_2 capture (eCCC) technologies and the incumbent thermochemical, amine-based capture technologies. I first start by developing a reactive absorption model to predict the absorption column sizes required in "4–stage" eCCC systems (i.e., comprising of an electrochemical reactor, absorption column, and flash tank). I use the model to inform operating conditions and molecular properties that will allow these processes to utilize columns that are comparable in size to those presently deployed in thermochemical systems. While this helps address capital cost comparisons, to couple these effects with operating costs I next combine the absorption column model with an electrochemical cell model to predict the levelized cost of capture (LCOC) of the capture platforms at a coal pilot plant facility. This techno-economic model allows for thorough investigation of the property sets, operating conditions, and target cost factors that will lead to conditions where the electrochemical systems can compete economically with amine scrubbing systems. Next, this in-house model is used to probe the effects of scale and flue gas composition on the overall LCOC to provide commentary on the conditions and costs likely for operation at commercial-scale plants. Finally, I apply my knowledge of decarbonization efforts to inform realistic pathways for decarbonizing cement production facilities in the near-term. Ultimately, the goal of this thesis is to lay the foundation for quantitative comparisons between different technologies available for point-source capture applications while also offering models that can be used to investigate the viability of promising molecules and electrolytes in eCCC.

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