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An Integrated Modelling Framework for Pilot-Scale Electrochemical Regeneration of Alkaline CO_2 Capture Solvents

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Abstract

The need for sustainable industrial methods has significantly increased the demand for advanced carbon capture technologies, particularly in energy-intensive sectors such as cement production, magnesite processing, and natural gas combined heat and power (CHP) systems. These industries are major contributors to global CO_2 emissions, presenting substantial challenges in developing scalable solutions that align with international decarbonization goals. Conventional carbon capture technologies often face limitations in efficiency, scalability, and integration with existing industrial processes, necessitating innovative approaches to enhance performance and reduce operational costs. To address these challenges, this study introduces an integrated modelling framework developed using Aspen Plus and Aspen Custom Modeler (ACM) to combine CO_2 absorption with electrochemical regeneration. This computational platform enables researchers and engineers to design and optimize alkaline solvent-based carbon capture systems with high accuracy by incorporating thermodynamics, reaction kinetics, and transport phenomena. By providing a detailed simulation of pH-driven CO_2 desorption and solvent regeneration (ConsenCUS), the framework offers a comprehensive tool for analysing process dynamics and optimizing system performance. Unlike traditional modelling approaches, which often struggle to capture the complex interplay between physical and chemical processes in electrochemical carbon capture, this framework provides a more precise and adaptable solution. As a result, it enhances the fundamental understanding of electrochemical carbon capture mechanisms while facilitating the development of more efficient, cost-effective, and scalable technologies. Ultimately, this work contributes to advancing carbon capture strategies, supporting global efforts to reduce industrial CO_2 emissions and mitigate climate change.

Keywords: CO_2 capture; electrochemical regeneration; integrated modeling; pilot-scale optimization; industrial decarbonization; sustainable processes.

Methodology

The model integrates three interconnected compartments: (1) an acidifying zone for CO_2 release, (2) a membrane separator for ion migration, and (3) an alkaline regeneration zone for solvent recovery. Thermodynamic equilibria

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govern pH shifts and carbonate speciation (e.g., $CO_3^{2-} \rightleftharpoons HCO_3^- \rightleftharpoons CO_2$), while electrochemical kinetics regulate ion transport under applied currents. Transport phenomena, including bubble-induced mass transfer limitations, are explicitly modelled to account for reduced ionic conductivity at high current densities. Nonlinear equations describing charge conservation, electroneutrality, and reaction equilibria are solved iteratively using numerical methods, ensuring self-consistent predictions of system behaviour across operational scales.

Key Results and Discussion

Model validation against experimental data showed good predictive accuracy for CO_2 production rates at low-to-moderate current densities (15–50 mA/cm²), with simulated outputs closely mirroring observed trends. However, at higher currents (>75 mA/cm²), deviations emerged due to mass transport constraints and nonlinear effects commonly seen in electrochemical systems. Specific energy consumption (SEEC) trends corroborated experimental findings, though discrepancies at low currents pointed to the omission of activation overpotentials and parasitic side reactions, such as oxygen evolution, in the current model. Pilot-scale simulations tailored to industrial flue gases illuminated critical operational trade-offs. For instance, increasing the liquid-to-gas (L/G) ratio from 2 to 5 boosted CO_2 capture efficiency from 60% to near-complete removal (100%), but this enhancement came at a steep cost, escalating SEEC by 300%. This underscores the fundamental tension between maximizing capture performance and minimizing energy penalties—a key consideration for industrial adoption.

Further analysis revealed that higher flue gas CO_2 concentrations (e.g., 13.5% vs. 3.5%, typical of cement vs. natural gas CHP emissions) reduced energy demands by approximately 40%. This improvement stemmed from enhanced reaction kinetics and more efficient ion transport, which collectively lowered the electrochemical workload. Potassium ion (K^+) load ratio emerged as a critical emerged as a pivotal design parameter ($L_{K^+} = \frac{j_C.A}{C_{K^+}.Q.F}$): optimal load ratio (0.8–0.9) minimized ohmic losses. Sector-specific simulations underscored divergent industrial priorities. In the natural gas CHP case, the model achieved full CO_2 capture (100%) at an energy cost of 9 MJ/kg, prioritizing environmental performance over efficiency. Conversely, the cement plant scenario favoured high throughput (15 kg_{CO_2}/h) at a reduced energy intensity of 4 MJ/kg, reflecting a focus on operational scalability and cost-effectiveness over absolute capture rates (Figure 1).

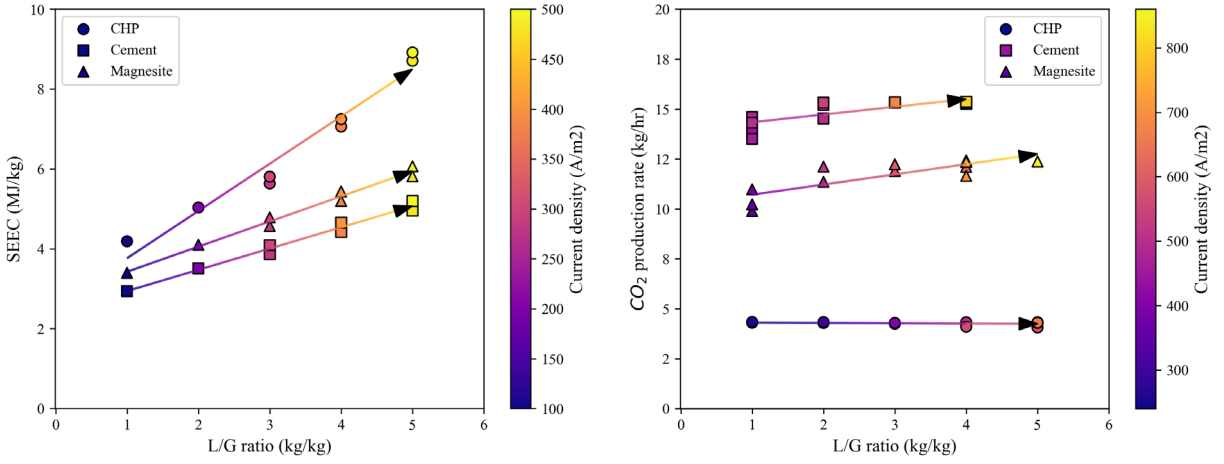

Figure 1. Simulation results for the best performance of the electrochemical cell (L_{K^+} = 0.8 − 0.9) across all case studies at different L/G ratios and current densities.

Implications for Industrial Applications

By bridging molecular-scale mechanisms with system-level engineering, this framework offers a versatile platform for optimizing critical components, including electrode materials, membrane properties, and operational parameters, while maximizing energy efficiency. Its open-access implementation fosters collaborative refinement and customization, empowering researchers and engineers to develop tailored solutions for diverse industrial contexts (https://github.com/mijndertvanderspek/ConsenCUS). By aligning with global net-zero ambitions, the framework supports the broader transition to sustainable industrial systems, offering both practical tools and scientific insights to accelerate the deployment of next-generation carbon capture technologies.