Flue Gas CO_2 Capture using Electrochemically Mediated Amine Regeneration

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To sustain global energy demand while preventing a 2°C global temperature increase, it is imperative that mitigation technologies such as carbon capture are deployed to reduce carbon footprint of the continued use of fossil fuel. This thesis describes the current status of the electrochemically mediated amine regeneration (EMAR) process for the capture of CO_2 from flue gas. The absorption step in the EMAR process is the same as in the widely used amine process, but the desorption step is accomplished electrochemically as opposed to a thermal swing operation. The EMAR process has several potential advantages over today's amine process. In terms of energy use, it has the potential to match the energy requirements of the amine process. Further, unlike thermal swing operation, EMAR uses just electricity, eliminating the need of steam. Another advantage is that theoretically CO_2 can be released at higher pressures in the EMAR process, reducing the compression energy requirement.

Although the EMAR process has many unique advantages, its practical implementation is hindered by several challenges. Firstly, few large-scale electrochemical operations can serve as guides to scale up this process. Moreover, the EMAR system requires materials (e.g., plastics, metals, and membranes) that are very different from a packed column and can be expensive to manufacture due to a smaller market demand. Motivated by these challenges, we began by investigating the thermodynamics of the mediated separation and developed process flowsheets to study the impacts of various operation parameters (e.g. desorption pressure). The net energetics for a base-case process requires up to 52 kJe/molCO_2 for flue gas treatment, which is comparable to those of thermal swing processes. Furthermore, the EMAR process can desorb CO_2 at high pressure with minimal energy penalty to further reduce the size of the compression train.

Additionally, we integrated an EMAR desorption unit into a lab-scale system for flue gas capture with online electrochemical and gas separation monitoring. With 15% CO_2, the EMAR separation scheme can continuously operate for up to 200 hours with flexible modulation of the regeneration capacity of between 0.12 to 0.62 CO_2/mol amine while consuming up to 80 kJ/mol of electrical energy. With periodic switching of the polarity, the system achieved a reproducible amine regeneration for 130 cycles, demonstrating significant improvement over prior development.

Lastly, we sized the electrochemical separation stacks incorporating kinetics and mass transfer effects to facilitate the system design. Changing system components (e.g. membranes) and operation parameters (desorption pressure, temperature, solvent capacity, and applied potentials) led to additional variation in system size and energy consumption. We identified the membrane cost as the dominant capital cost for the electrochemical separation train. The CO_2 avoided cost can be reduced to below \$60/tCO_2 with optimized process conditions (e.g. utilization of waste heat).

Thermal processes have a long history of operation at the large scales that are needed for carbon capture. The modular nature of electrochemical cells may alleviate the concern of scale to some extent, providing a viable pathway to "plug-and-play" EMAR system. The development described in this thesis indicates that the EMAR process has the potential to be a viable option for flue gas CO_2 capture.

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