Modeling-based study of monolith contactors for direct air capture

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Abstract

The evidence that the anthropogenic alteration of the earth’s carbon balance is leading to climate change has indicated the necessity of bringing global CO\textsubscript{2} emissions to net-zero by 2050 to keep global warming at a 1.5°C level [1]. This implies adopting negative emissions technologies (NET), which are technologies that result in the net carbon removal from the atmosphere. One NET option is direct air capture and storage (DACCS) via temperature-vacuum swing adsorption (TVSA) cycles on amine-functionalized sorbents.

Adsorption-based direct air capture (DAC) of CO\textsubscript{2} from the atmosphere has been proven to be technically feasible though much more expensive than post-combustion CO\textsubscript{2} capture. In fact, DAC offers advantages such as location flexibility (provided the energy), but its low concentration of CO\textsubscript{2} in the air entails that it also comes with large energy consumptions. This is because the high volumetric flow rates of air that must be processed to capture such amounts of CO\textsubscript{2} during adsorption entail that large pressure drops are expected in the capture step. Optimizing the air-solid contactor to reduce the pressure drop to a limit is crucial to reduce the overall energy consumption.

Indeed, more and more studies have shown a shift in contactor designs for DAC with respect to conventional CO\textsubscript{2} capture technologies, which are mainly in the form of packed beds in long and narrow columns [3]. Industrial and academic studies on direct air capture have indeed shown that thin packed beds with wide cross sections or, even, structured sorbents, exhibit much smaller pressure drops with respect to conventional packed beds [4-7]. Although honeycomb monoliths have been proven to indeed reduce pressure drop, studies have shown drawbacks due to their low sorbent loadings [8-10], high mass transfer resistances in the gas film forming on the support wall, and flow maldistribution [11-12], the latter resulting in flow channeling, which lead to uneven uptake among the channels. Traditional packed bed configurations using pellets, on the other hand, offer high mass transfer rates and volumetric loadings but suffer from higher pressure drops especially at high flow rates. Both the pressure drop and the mass transfer rates are a function of the geometrical properties of the pellets and monoliths at hand.

In this work, we intend to study the aforementioned phenomena and assess the trade-offs that can arise in DAC contactors with variable sorbent loadings and pressure drops. To this end, we intend to discuss the following aspects:
(i) a detailed theoretical analysis of an amine-functionalized honeycomb monolith as a novel DAC contactor, including the advantages and disadvantages of such a structured sorbent with respect to traditional packed beds;

(ii) the determination of the trade-offs between low pressure drops, high mass transfer rates and high volumetric loadings in terms of monolith and packed bed geometry in the adsorption step for different operating conditions;

(iii) the validation of the model using experimental data from a lab-scale setup that allows for the measurement breakthrough curves for packed bed configurations and for honeycomb monoliths;

(iv) first-principles modeling of temperature vacuum swing adsorption cycles of honeycomb monoliths;

(v) a comparative assessment of the process performance using TVSA cycles on honeycomb monoliths and packed beds in the context of direct air capture.

Keywords: CO₂ capture; direct air capture; structured sorbents; monoliths

1. Intergovernmental Panel on Climate Change, “Global Warming of 1.5°C. Summary for Policymakers,” 2018.