Isotherm Modeling and Techno-Economic Analysis of Contactor Technologies for New Tetraamine-Appended MOF for NGCC Applications

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Abstract

Natural gas is currently a major source of electricity in the United States (Mondino et al., 2019) and fossil fuels will likely continue to play a significant role in the world’s energy mix. Therefore, new technologies and materials to reduce CO\textsubscript{2} emissions will be instrumental in meeting US goals for deep decarbonization. Flue gases from natural gas-fired power plants present a significant challenge due to the low CO\textsubscript{2} volumetric/mol composition (~4% CO\textsubscript{2}). To address this challenge, the Carbon Capture Simulation for the Industry Impact initiative has been developing models and tools that provide insight into cost effective technologies for Carbon Capture Utilization and Storage (CCUS). This work presents two rigorous first-principle models for gas/solid contactors which are applied to enable techno-economic analysis (TEA) for CCUS.

Recently, a family of tetraamine-functionalized metal-organic frameworks (MOF) has been reported as promising sorbent materials for capturing CO\textsubscript{2} from flue gas conditions relevant to NGCC applications (Kim et al., 2020). The main advantages of these materials are their two-step cooperative CO\textsubscript{2} adsorption, which gives rise to unusual two-step-shaped CO\textsubscript{2} adsorption profiles and their high thermal stability. This work presents the modeling of the two-transition isotherm of the tetraamine-appended MOF, \textit{N,N’-bis(3-aminopropyl)-1,4-diaminobutane (3-4-3)-appended Mg\textsubscript{2}(dobpdc)} (Kim et al., 2020), and the TEA of carbon capture processes utilizing this sorbent. Due to the unusual isotherm shapes of the experimental CO\textsubscript{2} adsorption data for tetraamine-appended Mg\textsubscript{2}(dobpdc) and the strong nonlinearity of CO\textsubscript{2} loading with respect to temperature and pressure, we tested two different models which use logistic functions for representing the different isotherm behaviors in the different pressure ranges.

The first model (model 1) uses the quadratic isotherm model in the low-pressure region, the Langmuir isotherm model in the middle pressure range, and the dual site Langmuir isotherm model in the high-pressure range. To model the transition between regions and ensure the model is continuous, we used the arctangent functions independent of temperature. To incorporate the temperature dependence into the isotherm model, we used the Clausius-Clapeyron relation, normalizing the data at various temperatures in the pressure direction and mapping all data points to a single curve, allowing a single model to fit all experimental data simultaneously (Ga et al., 2021). The second model (model 2) is an extension of the weighted dual-site Langmuir isotherm model presented by Hughes et al., 2021. In this

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extended model, the dual-site Langmuir isotherm is employed in the three transition regions, using temperature-dependant logistic functions to activate or deactivate the isotherm model in the low, middle, and high-pressure region. As shown in Figure 1, both models fit the experimental data quite well with root mean squared errors (RMSE’s) of 0.41 and 0.17 for model 1 and model 2, respectively. Since the model 2 resulted in a lower RMSE, it was leveraged for the development of the gas/solid contactor models used by the TEA.

Specifically, two different contactor models, an axial-flow fixed bed and moving bed contactor, were developed as part of this work. These models are dynamic, pressure-driven, and consist of mass, energy, and momentum conservation equations. A kinetic model was also developed by performing parameter estimation using experimental fixed bed breakthrough data. These models are then used to simulate CO₂ capture processes from the flue gas generated from a ~600 gross MW NGCC power plant. A cost model was developed which considers the capital cost of the reactors and the significant operating costs such as steam and electricity. Using NETL’s Framework for the Optimization and Quantification of Uncertainty and Surrogates tool (FOQUS) (Miller et al., 2016), which has the capability of linking models built using numerous modelling platforms with derivative-free optimization solvers, a techno-economic optimization of the carbon capture processes was performed which minimizes the cost of capture. This work demonstrates the utilization of the rigorous dynamic models for process optimization using the FOQUS tool.

References
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