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Multi-Objective Optimization of CO_2 Capture from Ambient Air via TVSA Process Modeling

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Summary

Direct Air Capture (DAC)- a negative emissions strategy that involves removing carbon dioxide directly from the atmosphere- is one of the key technological approaches to meeting global climate targets. Among the various sorbent materials explored for CO2 capture, amine-functionalised metal organic frameworks (MOFS) have attracted considerable attention due to their high selectivity and tunable adsorption properties, particularly for post-combustion CO2 capture from flue gas streams. In this study, the performance of mmen-Mg2(dobpdc) for DAC under atmospheric conditions (~400 ppm CO2) is evaluated through detailed process simulation using a temperature vacuum swing adsorption (TVSA) cycle. The simulation model is integrated with the NSGA-II algorithm to perform multi-objective optimisation, targeting the trade-off between CO2 recovery and specific energy consumption. Results indicate that this sorbent can achieve CO2 recovery above 90% under feed and process conditions with purity exceeding 94%. The corresponding specific energy consumption varies with the selected operating point, ranging from 3 to 7.5 MJ/Kg CO2, highlighting the importance of balanced process design.

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Introduction

The global surface concentration of CO_2 averaged 419.3 ppm in 2023, reflecting an annual increase of 2.8 ppm(Theo Stein, 2024). To effectively address climate change, it is crucial to capture emissions from diffuse sources like transportation and agriculture. Direct air capture (DAC) by extraction CO_2 directly from the atmosphere has been proposed as a viable option for stabilizing global CO_2 concentrations. Despite process modelling is crucial for evaluating the industrial potential of sorbents and enables detailed analysis of process performance, only a few of sorbents which are experimentally tested under DAC conditions, have been explored through process modelling. Previous exploration of mmen-M_2(dobpdc)- (M = Mg, Mn, Fe, Co, or Zn)- through process modelling have primarily focused on post combustion CO_2 capture from flue gas and demonstrated the ability of these sorbent to achieve high CO_2 purity and recovery (Joss et al., 2017). (Pai et al., 2019). Sinha A. (Sinha et al., 2017) evaluate the application of two metal organic frameworks, MIL-101(Cr)-PEI-800 and mmen-Mg_2(dobpdc), under DAC conditions using the monolith structure bed. through numerical modelling and concluded that mmen-Mg_2(dobpdc) outperforms MIL-101(Cr)-PEI-800 in energy requirements due to its higher CO_2 capture capacity and nonlinear isotherm behaviors. Recently the application of mmen-Mg_2(dobpdc) in indoor environment with CO_2 concentrations exceeding 1000 ppm was evaluated and concluded the efficiency of this adsorbent in terms of CO_2 productivity and purity for indoor air quality management (Shi et al., 2023). This study evaluate the application of mmen-Mg_2(dobpdc), for DAC at atmospheric concentrations (~ 400 PPM) using a temperature vacuum swing adsorption (TVSA) process model. The process was optimized with respect to CO_2 recovery and energy consumption, and a Pareto front was developed to illustrate the trade-off between these objectives, providing a set of optimal solutions for different operational preferences.

Method

Model Development

A detailed TVSA process model was developed to evaluate the performance of mmen-Mg_2(dobpdc) for DAC. The model simulates the full adsorption–desorption cycle and accounts for mass and energy balances, adsorbent properties, and process dynamics. The established isothermal model and sorbent properties proposed by (Darunte et al., 2019) was adapted for this simulation. The performance effectiveness of the DAC system was assessed using key performance indicators, including the CO_2 recovery, CO_2 purity, and specific energy consumption, as defined in equations 1-3. These metrics were determined following the process simulation.

$$Purity = \int_0^{t_{cycle}} F_{product} y_{CO2} dt \Big/ \sum_{i=1}^{m} \int_0^{t_{cycle}} F_{product} y_i dt \quad (1)$$

$$Recovery = \int_0^{tcycle} (y_{product,CO2}\, F_{product}\big|_{z=L}) dt \Big/ \int_0^{tcycle} (y_{feed,CO2}\, F_{feed}\big|_{z=0}) dt \quad (2)$$

$$Pump\ Efficiency = \int_0^{t_{cycle}} \frac{F_{vac} P_{vac} \gamma}{\eta(\gamma - 1)} \left| \left(\frac{P_{feed}}{P_{vac}}\right)^{1-\frac{1}{\gamma}} - 1 \right| dt \Big/ \int_0^{t_{cycle}} F_{product}\, y_{product,CO2} dt \quad (3)$$

To guide the optimization phase, following a sensitivity analysis process, parameters—adsorption time, desorption time, desorption temperature, vacuum pressure, and feed flow rate—was selected.

Optimization

The optimization was carried out by coupling the TVSA process simulator with the NSGA-II algorithm, a well-established multi-objective evolutionary algorithm designed to efficiently generate a diverse and high-quality Pareto front. Each candidate solution, defined by a set of decision variables, was evaluated through dynamic simulations of the process model to calculate performance metrics which are introduced in equations 1 to 3. The algorithm iteratively improved the population by ranking solutions based on non-domination and applying a crowding-distance metric to preserve diversity, enabling the identification of optimal trade-offs between competing objectives(Bagheri et al., 2025).

Results

Sensitivity Analysis and Selection of Decision Variables

Based on the sensitivity analysis conducted after model validation, six process variables - adsorption time, desorption time, vacuum pressure , feed flow rate , feed temperature , and heating temperature – were identified as having the most significant impact on system performance. Theses variables were selected as the decision parameters for the NSGA-II optimization, which was used to explore trade-off between CO_2 recovery and specific energy consumption. *Figure 1* illustrates the impact of different process parameters on key performance indicators, including energy consumption (SEC), CO_2 purity, and recovery.

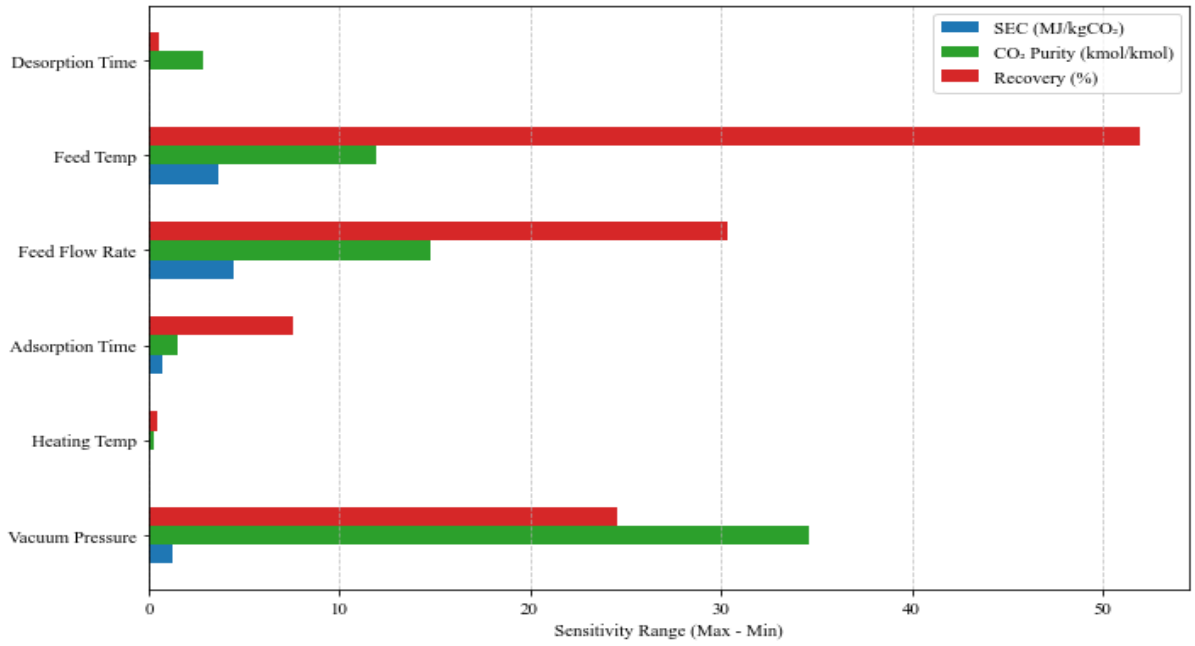

Figure 1 *Effect of process parameters and feed conditions on specific energy consumption, CO_2 purity and recovery.*

Optimization

The optimization produced a Pareto front comprising a set of non-dominated solutions, where no single solution is universally superior across all objectives. Each point on the Pareto front represents a unique combination of decision variables and cycle durations that offers a trade-off between CO_2 recovery and specific energy consumption (SEC) which is illustrated in *Figure 2*. These solutions provide flexible design options, allowing stakeholders to select operating conditions based on specific performance priorities in industrial applications.

Figure 2 shows the direct relationship between Recovery and specific energy consumption. The Pareto front can be divided into three regions. In region 1, where CO_2 recovery is below 90% , the specific energy consumption remain relatively low, under 3.5 MJ/Kg CO_2. In region 2, as recovery increase to between 90% and 91%, specific energy consumption begins to rise gradually. In region 3, pushing recovery beyond 91% lead to a steeper increase in energy demand, indicating a significant trade-off between achieving higher recovery and maintaining energy efficiency.

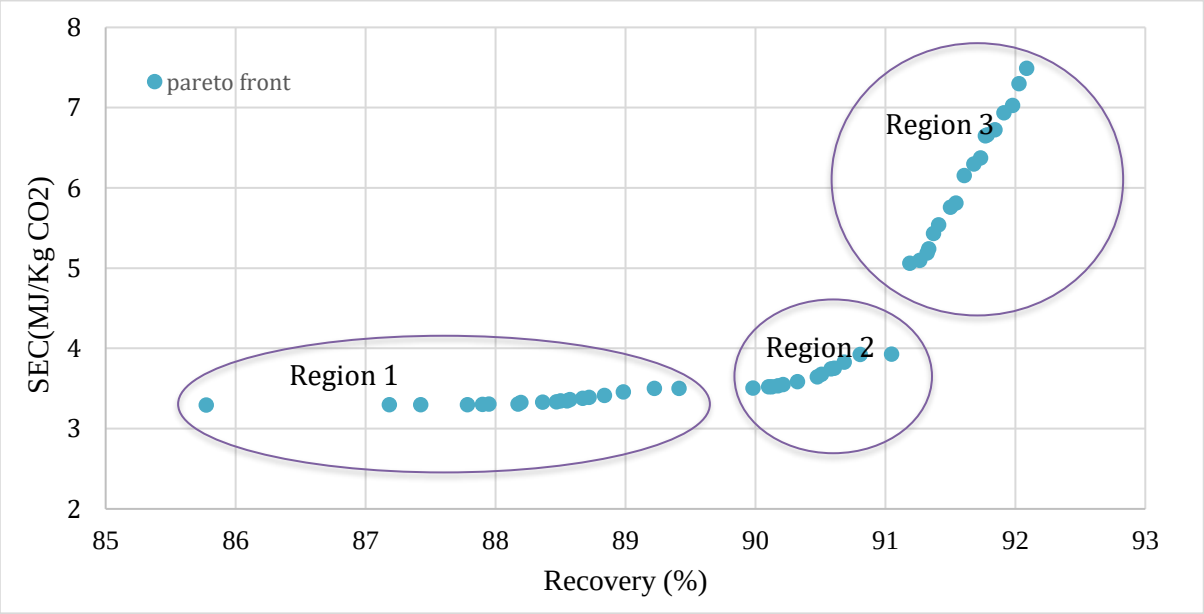

Figure 2 *Two -dimensional pareto front for CO_2 recovery and specific energy consumption.*

Conclusions

This study demonstrates the potential of the TVSA process using mmen-Mg_2(dobpdc) for capturing CO_2 directly from the atmosphere. By integrating a dynamic process simulator with NSGA-II algorithm, a multi-objective optimization was conducted, generating a pareto front that highlights the trade-off between CO_2 recovery and specific energy consumption. The results indicate that this sorbent is capable of achieving CO_2 recovery rates exceeding 91%, although doing so requires a substantial increase in energy consumption. In contrast, maintaining recovery rates between 85% and 90% can be achieved with specific energy consumption below 4 MJ/kg CO_2, offering a more energy-efficient operating window. These finding highlight the importance of balancing energy use with recovery targets, and provide a valuable basis for the design and operation of future DAC systems using advanced solid sorbents.

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