Design and Energy Evaluation of a Multi-Stage CO2 Separation Process Using Surrogate Model

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Abstract

In recent years, CCS (Carbon dioxide Capture and Storage) has been promoted worldwide, and among CCS processes, the adsorption and membrane separation processes require lower energy than gas absorption using amine but have lower separation performance. The combination of adsorption and membrane is one way to improve the separation performance, but studies on the combination and configuration of multi-stage processes are still insufficient. In addition, membrane separation is operated in a steady state and adsorption separation is operated in an unsteady state, which makes thedesign of a hybrid process complex. Therefore, to design CO2 capture processes for both adsorption and membrane separation, we applied statistical model with machine learning. For each process, parameters such as compressor and vacuum pump operating pressures, CO2 composition in the feed gas, and flow rate were set within predefined ranges, and data were collected from these ranges. Using this data, a statistical model was developed using a neural network, and multi-objective optimization was performed using NSGA-II (Non-dominated Sorting Genetic Algorithms II). The results showed that the required performance of the CCS could not be achieved with a single stage separation. The Pareto solution obtained by optimization indicated that a high compression ratio and low vacuum pressure were required. The prediction accuracy of the physical and statistical models in the Pareto solution was about 0.99 for both processes. Based on these results, a multi-stage separation process was synthesized using surrogate models. Multi-objective optimization was performed on this process, and it was found that the combined membrane and adsorption process not only met the CCS requirements, but also reduced the operating pressures of the compressor and vacuum pump compared to the single-stage process. These results highlight the effectiveness of using statistical models for process optimization.

**Keywords**: CO2 separation, Machine learning, Process design

* 1. Introduction

The Industrial Revolution occurred between the 18th and 19th centuries and brought revolutionary changes to human life and the economy. In recent years, there has been a consensus that greenhouse gas emissions from human activities are the main cause of global warming (L. Al-Ghussain, 2019). As a solution to the current situation, CCS technologies have gained much attention. Three main processes are considered for CO2 separation technology: amine absorption, membrane separation and gas adsorption separation. Among the three processes, amine absorption is currently the most widely used CO2 separation process. However, the reaction heat between the absorbed solvent and CO2 is high, and the CO2 capture energy is high (4 - 6 MJ/kg-CO2 (M. Mondal et al., 2012)), because more energy than the reaction heat is required for CO2 stripping. On the other hand, the energy consumption of adsorption and membrane separation processes, which are alternative technologies to the amine absorption process, are reported to be 2 - 3 MJ/kg-CO2 and 0.5 - 6 MJ/kg-CO2, respectively (M. Mondal et al., 2012). However, adsorption and membrane separation processes have the problem of low separation efficiency (Y. Fujii et al., 2023). One solution is to combine these processes into a multi-stage process. In the design of multi-stage processes, membrane separation is operated in a steady state and adsorption separation is operated in an unsteady state, which makes the calculation complex to design processes that combine these processes. Recently, it has been reported that this complexity can be reduced by using a surrogate model based on machine learning (M. Rahimi et al., 2021). In this study, we perform efficient membrane-adsorption multi-stage process synthesis using machine learning.

* 1. Modelling

**Figure 1** shows a schematic of the trained adsorption and membrane separation process. First, a physical model of the adsorption and membrane separation processes was developed. The mass balance and energy balance equations for component i in this process model are expressed by Eq. (1) and (2) respectively. The mass transfer to the adsorbent is represented by the Linear Driving Force (LDF) model in Eq. (3). The adsorption process assumes pressure swing adsorption; initially, the inlet gas is pressurized by a compressor and fed into the column for adsorption. After adsorption is completed, the column is depressurized by a vacuum pump for desorption. The solution was obtained by a dynamic simulation in which these steps are repeated alternately.

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| --- | --- |
| $$-ε\_{b}D\_{ax,i}\frac{∂^{2}c\_{i}}{∂z^{2}}+\frac{∂\left(v\_{g}c\_{i}\right)}{∂z}+\left(ε\_{b}+\left(1-ε\_{b}\right)ε\_{p}\right)\frac{∂c\_{i}}{∂t}+ρ\_{s}\left(1-ε\_{b}\right)\frac{∂q\_{i}}{∂t}=0$$ | (1) |
| $$-k\_{g}ε\_{b}\frac{∂^{2}T\_{g}}{∂z^{2}}+C\_{vg}v\_{g}ρ\_{g}\frac{∂T\_{g}}{∂z}+ε\_{b}C\_{vg}ρ\_{g}\frac{∂T\_{g}}{∂t}+P\frac{∂v\_{g}}{∂z}+h\_{f}α\_{p}\left(T\_{g}-T\_{s}\right)+\frac{4h\_{w}}{D\_{b}}\left(T\_{g}-T\_{0}\right)=0$$ | (2) |
| $$\frac{∂q\_{i}}{∂t}=k\_{LDF,i}\left(q\_{i}^{\*}-q\_{i}\right)$$ | (3) |

The membrane separation is based on the cross-plug flow model expressed in Eq. (4), and the pressure difference caused by pressurizing the inlet gas and depressurizing the drop side was used as the driving force for the separation in the simulation.

|  |  |
| --- | --- |
| $$\frac{dF\_{i}}{dA}=-Q\_{i}\left(p\_{h}x-p\_{l}y\right)$$ | (4) |



**Figure 1** Schematic diagram of (a) adsorption and (b) membrane process

The material of the two processes was fixed, assuming Zeolite13x (N. Jiang et al., 2020) as adsorbent and polymer membrane (B. Ghalei et al., 2017) as membrane material, 1000 GPU (=3.3×10-10 [mol/(m2 s Pa)]), CO2/N2 selectivity 50. The sampling ranges for the training data are shown in **Table 1**. The neural network was trained with about 1000 data samples. The data were collected randomly in these ranges. As input parameters, the inlet flow information, adsorption time for adsorption, and membrane area for membrane were trained. The range of adsorption time was 250 s - 2500 s, and the range of membrane area was 0.5 - 30 m2. Inlet gas is assumed to be two components, CO2 and N2. The information of outlet flow and energy consumption were trained as output parameters. The neural network was tuned for the number of hidden layers, the number of neurons in each layer, and the number of epochs, and the most accurate model was used. These learned statistical models were used as alternative models for optimization. A genetic algorithm, NSGA-II, was used for multi-objective optimization. Surrogate models were combined when constructing a multi-stage process, and multi-objective optimization was performed for the entire system.

**Table 1** Operating range

|  |  |  |
| --- | --- | --- |
| Parameter |  | unit |
| Inlet CO2 mole fraction | 15 - 99 | mol% |
| Inlet gas flowrate | 0.01 – 0.20 | mol/s |
| Compressor | 200 - 500 | kPa |
| Vacuum pump | 5 - 20 | kPa |

* 1. Result and Discussion



**Figure 2** Pareto solutions of multi-objective optimization in single-stage process

To optimize product composition and recovery in a single-stage process, a surrogate model trained on adsorption and membrane models was used. **Figure 2**(a) shows the Pareto solutions obtained from the multi-objective optimization. At 90% recovery, the composition is 0.92 for adsorption and 0.74 for membrane, which does not meet CCS requirements (product composition 0.95 [-], recovery 90%). In addition, it was shown that the membrane gives a higher recovery if the product composition is less than 0.70. Figure 2(b) shows the pressure conditions for the compressor and vacuum pump in the Pareto solutions. It is clearly seen that both adsorption and membrane separation processes operate with a high compression ratio for the compressor and vacuum pressure. Adsorption requires a significantly low vacuum pump operating pressure, around 5 kPa, despite its separation performance close to CCS requirements. This indicates that while adsorption provides superior separation performance compared to membrane separation, it also requires a higher operating pressure. The coefficients of determination between the physical and statistical models at this point showed high accuracy, with product composition at 0.993 [-] and 0.998 [-] for adsorption and membrane, respectively, and recovery at 0.978 [-] and 0.997 [-].

Next, **Figure 3(a)** shows an example of a multi-stage process constructed in this study, the membrane adsorption process. This process was combined with a surrogate model to create a system. The optimization was performed. **Figure 3(b)** shows the Pareto solutions obtained through multi-objective optimization of the membrane adsorption process. This result maximizes product composition and recovery and minimizes energy consumption. The CCS requirement (product composition 0.95 [-], recovery 90%) was achieved by the multi-stage process; the minimum energy to achieve the CCS requirement was 1.15 MJ/kg-CO2. The operating pressures of the first stage compressor and vacuum pump were 221 kPa and 9.8 kPa, respectively, and those of the second stage compressor and vacuum pump were 336 kPa and 19.2 kPa, respectively. Thus, the use of a multi-stage process avoids higher compressor and vacuum pump operating pressures compared to a single-stage process. Other similar two-stage processes considered in this study were the membrane-membrane, adsorption-adsorption, and adsorption-membrane processes, and the minimum energy consumption after achieving the respective CCS requirements was 1.26, 1.37, and 1.41 MJ/kg-CO2 for the membrane-adsorption process, respectively.

In two-stage processes such as those considered in this study, the flow entering the compressor or vacuum pump in the first stage is lower than that in the second stage. Therefore, increasing the CO2 concentration in the membrane process, which has a low energy consumption, followed by separation by the adsorption process with high separation efficiency, is considered optimal, as concluded from the optimization of the single-stage process.



**Figure 3** (a)Schematic diagram of membrane-adsorption process and (b) Result of multi-objective optimization of membrane-adsorption process



**Figure 4** Accuracy of (a) purity, (b) product flow rate, and (c) recovery in Pareto solution of membrane-adsorption process

**Figure 4** shows the model accuracy of the two-stage process in the Pareto solutions of the membrane adsorption process, which was a best case in this study. The coefficients of determination for product composition and product flow show a high accuracy of 0.97, while the accuracy for recovery is relatively low at 0.90. This is probably because recovery was not trained directly, but calculated from the trained product composition and product flow. Therefore, due to some error in the recovery, it is expected that there may be some error in the optimization results of this study. However, if models with improved accuracy can be developed, they could be applied to more complex explorations, such as the search for complicated multi-step process structures. This could make it a powerful method for process synthesis.

* 1. Conclusions

In this study, a surrogate model for adsorption and membrane processes was created using machine learning. Using this model, the design of single and multi-stage processes was performed, leading to the following conclusions:

* The use of machine learning surrogate models allowed for the avoidance of complex calculations in conventional models, providing efficient optimization of both single-stage and multi-stage processes.
* The multi-stage system was able to achieve the separation performance required for CCS. In addition, the compressor and vacuum pump pressures, which were operating at high levels in the single stage, could be reduced.
* Among the cases considered in this study, it was found that the membrane-adsorption process had the lowest energy consumption.
* Since recovery was not directly trained, the accuracy of the statistical model decreased when applied to multi-stage processes.
* This study has shown that a statistical model can be used to efficiently synthesize a multi-stage process. If the accuracy of the model is further improved, it can be applied to more complex structures, such as those that consider recycling.

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