System optimization of hybrid processes for CO2 capture

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Abstract

An optimization framework is developed to investigate the techno-economic potential of optimal hybrid process designs for post-combustion CO2 capture, where a hybrid process involves a combination of different CO2 capture technologies. System level optimization (i.e., system optimization) is compared with optimizing the capture technologies in sequence (i.e., cascade optimization). The technologies considered to be combined in a hybrid configuration are a vacuum pressure swing adsorption (VPSA) process and a membrane process. Fit-for-purpose models of each of these processes are developed and integrated with a detailed techno-economic analysis (TEA) model to allow CO2 avoidance cost as a key metric for optimization. The optimization results showed that both system and cascade optimization are able to approach a similar optimal hybrid process design. Cascade optimization obtained a minor improvement in terms of overall cost at the price of a significantly higher computational time. If a stream recycle is implemented, a feature that can reduce the cost of about 1%, system optimization is preferable as the iterative effort required to solve the recycle configuration would lead to an excessively high computational time for cascade optimization.

**Keywords**: CO2 capture, optimization, hybrid processes, techno-economic analysis.

* 1. Introduction

A hybrid process is a combination of different technologies selected to perform a separation process in an efficient manner. When a single technology is used to perform a specific separation process, it is often required to operate at non-ideal conditions. Hence, standalone technologies must normally trade off some efficiency to match the case-specific operating conditions and achieve the targeted separation. Conversely, hybrid processes offer increased flexibility by combining multiple technologies. The underlying idea is to put each technology in the conditions at which its performance is maximized by tailoring the hybrid solution to the given application. Among the most relevant applications for hybrid processes, there is CO2 capture. There exist several separation technologies to capture CO2. A multitude of potential hybrid configurations for CO2 capture has been proposed (Song et al., 2018). Different studies have shown that hybrid processes can fare well with respect to single technologies, for instance reducing the energy consumption (Mat et al., 2019). However, there are also expected disadvantages connected to the implementation of hybrid processes. A key disadvantage concerns the increased complexity of the system. Particularly relevant is the difficulty of identifying the optimal design of the hybrid process.

In this work, we investigate how to identify optimal designs of hybrid processes for CO2 capture. The importance of design optimization at the system level (i.e., system optimization) is investigated and compared to the more standard practice involving the independent optimization of the single steps of a hybrid configuration (i.e., cascade optimization). To investigate the impact of the different approaches to hybrid process design optimization, a hybrid concept is selected with VPSA and membrane technology.

* 1. Modelling framework
		1. Hybrid process

A schematic representation of the VPSA-membrane hybrid process is presented in Figure 1. The hybrid configuration consists of a first VPSA step for bulk CO2 separation and a second membrane step for final CO2 purification. The hybrid configuration can also involve stream recycle, where the retentate gas from the membrane stage(s) is recycled to the VPSA process.



Figure 1. Process scheme of the VPSA-membrane hybrid process.

* + 1. Machine learning model of VPSA process

The VPSA process uses an activated carbon as adsorbent material. The rigorous model of the VPSA process is based on a set of partial differential equations as outlined by (Haghpanah et al., 2013). To allow for optimization, an artificial neural networks (ANN) model was developed (the ANN architecture consists of one input layer with 8 decision variables, two hidden layers with 20 neurons each, and an output with one output), trained (using Bayesian regularization with backpropagation algorithm) and validated (using an independent dataset of 600 samples: R2>0.98). Previous studies demonstrated very good capabilities of ANNs in predicting the performance of VPSA processes (Subraveti et al., 2019). The VPSA model simulates a 5-step cycle: adsorption (ADS), high-pressure reflux (HR), blowdown (BLO), purge (PUR) and light product pressurization (LPP).

* + 1. Membrane multi-stage model

The membrane gas separation step is simulated with an established multi-stage membrane design and optimization framework (Roussanaly et al., 2017). The model represents a mature membrane technology (i.e., MTR Polaris), with the following characteristics: permeance: 5.94 Sm3/m2barh, selectivity: 50. One or two membrane stages are simulated depending on the most efficient layout to meet the gas separation requirements.

* + 1. Techno-economic analysis model

A techno-economic analysis (TEA) model is integrated into the overall modelling framework. The TEA model, based on the guidelines outlined by (Roussanaly et al., 2021), estimates capital and operational expenditures. The former with a bottom-up approach, the latter based on utilities consumption and standard factors for maintenance and labor.

* 1. Optimization of the hybrid process
		1. Optimization problem

The CO2 capture technologies must treat the industrial flue gas considered and return a product gas stream which: (i) has a CO2 concentration of at least 95%; (ii) contains at least 90% of the CO2 originally in the flue gas. Those requirements constitute the CO2 purity and capture rate constraints of the optimization problem. The objective is to minimize the CO2 avoidance cost (CAC). The CAC is defined as the ratio between the sum of the annualized cost and the annualized emissions avoided. The process design degrees of freedom are the variables listed in Table 1, for a maximum of 13 optimization variables (see also Figure 1). Variables 11 and 12, referring to the pressure levels of the second membrane stage, are relevant only in the case of a 2-stage membrane layout. Variable 13 is the recovery level of the membrane step. If stream recycle is considered, this parameter can be optimized as it affects the amount of gas that will be recirculated into the VPSA step. Conversely, if there is no stream recycle, the recovery of the membrane step contributes to the final recovery rate of the hybrid process – a process constraint – hence it is determined by the recovery achieved by the VPSA step. The optimizations used a genetic algorithm (GA).

Table 1. Optimization variables and optimization bounds

|  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- |
| Variables VPSA | Var. no. | Bounds |  | Variables Membrane | Var. no. | Bounds Stage 1 | Bounds Stage 2 |
| High pressure (bar) | 1 | 1.05 – 3.6 |  | **High pressure (bar)** | 8/11† | 1.1 – 50 | 1.1 – 50 |
| Low pressure (bar) | 2 | 0.01 – 0.5 |  | **Low pressure (bar)** | 9/12† | 0.05 – 0.9 | 0.05 – 0.9 |
| Interstitial vel. BLO (m/s) | 3 | 0.45 – 2.5 |  | **No. membr. stages** | 10 | 1 – 2 |
| Interstitial vel. PUR (m/s) | 4 | 0.2 – 1.0 |  | **Recovery membr. stages** | 13‡ | 0.4 – 0.95 |
| ADS time (s) | 5 | 100 – 500 |  |  |  |  |  |
| Factor PUR time | 6 | 0.1 – 0.99 |  |  |  |  |  |
| Column size (m) | 7 | 6 – 9 |  |  |  |  |  |
| †These variables apply only when two membrane stages are considered |
| ‡This variable applies only when recycle is considered |

* + 1. Cascade optimization

*Cascade optimization* implies that the two capture technologies are optimized in sequence. The industrial gas provides the input conditions for the optimization of the first step – the VPSA process. Optimization variables 1 to 7 are used to determine a design that minimizes the CAC associated with VPSA. A single constraint applies to this case: a target capture rate without stream recycle (different capture rate levels are tested as discussed in the following) or ≥ 90% with stream recycle (as this will make up the entire hybrid process capture rate). The CO2-enriched gas produced by VPSA is sent to the membrane stage(s). In this case, the relevant optimization variables are 8 to 13. The CO2 purity constraint (≥ 95%) applies, while a capture rate constraint (subjected to the capture rate achieved from VPSA to ensure meeting the overall requirement ≥ 90%) applies only if stream recycle is not implemented. This optimization strategy splits the optimization burden between the two process steps, decreasing the number of variables for each of them – i.e., the search space for the optimal design. On the other hand, it fails to capture the entire system perspective, with the risk that the two optimal solutions for VPSA and membrane steps result in a suboptimal solution for the hybrid process. This is particularly evident when considering VPSA optimization. If the VPSA is optimized independently, the minimum cost would most likely be achieved by designing a process performing the bare minimum separation duty, i.e., lowest allowed CO2 enrichment and capture rate. However, these conditions are not favorable for the membrane step, with the risk of providing a suboptimal system performance. To account for this, the cascade optimization routine is set up such as to test a matrix of minimum CO2 purity and capture rate levels at the outlet of the VPSA process:

* 10 VPSA CO2 purity levels tested, evenly spaced between 30% and 75%
* 4 VPSA CO2 capture rate levels tested, evenly spaced between 90.5% and 99.5%

This implies that (10X4) 40 optimizations are performed to identify the overall optimum.

* + 1. System optimization

*System optimization* sees the system as a whole and optimized it as such. This implies a single optimization process, including optimization variables 1 to 13 and the two hybrid process constraints on CO2 purity (i.e., ≥ 95%) and capture rate (i.e., ≥ 90%). This optimization strategy allows to inherently explore the mutual influences of the two capture technologies and, if successfully implemented, to seek for the system optimum. The disadvantage is that of increased complexity of the optimization problem, which must explore a space that is significantly larger than those from the cascade optimization.

* 1. Results of process design optimization

The VPSA-membrane hybrid process is designed to capture CO2 from an industrial flue gas with a volumetric CO2 concentration of 15%. The gas stream is assumed to be a binary mixture of CO2 and N2. The gas flow rate is assumed to be 200 t/h.

* + 1. Parameters for optimization

The following key parameters for the GA algorithm were selected for the *cascade optimization*:

* Population size: 30 X number of optimization variables
* Number of generations: 100

And for the *system optimization*:

* Population size: 60 X number of optimization variables
* Number of generations: 200

For *cascade optimization* the selected population size and number of generations showed fit to obtain a good approximation of the optimum. Conversely, a sensitivity analysis had to be performed for *system optimization*. The parameters reported above appear to be a convenient compromise.

* + 1. System optimization vs. cascade optimization for baseline hybrid process

Ten runs were performed for both *cascade* and *system optimization* to identify optimal designs of the baseline hybrid process, i.e., not including stream recycle. The obtained results in terms of CAC are summed up in Table 2.

*Cascade* and *system optimization* obtained similar cost numbers, with similar levels of standard deviation in the results. *Cascade* *optimization* achieved slightly lower cost figures. However, the difference is minimal (0.3% on average) and arguably negligible. On the other hand, the execution time is, on average, more than 4 times smaller for *system optimization*. The reason is the combination of CO2 purities and capture rates to be investigated by *cascade optimization*. An a priori knowledge of the system could allow reducing the range of combinations tested, consequently reducing the gap in execution time. However, such a gap will hardly be closed without sacrificing some accuracy.

Table 2. Results of cascade and system optimization for the design of the hybrid process. The results of system optimization include the possibility of stream recycle.

|  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- |
| Optimization strategy | Number of runs | CAC\* avg (€/t) | CAC\* min (€/t) | CAC\* max (€/t) | CAC\* std. dev. (€/t) | Execution time avg (s) |
| Cascade (w/o recirc.) | 10 | 56.1 | 55.4 | 56.7 | 0.4 | 9120 |
| System (w/o recirc.) | 10 | 56.3 | 55.8 | 56.9 | 0.4 | 2053 |
| System (w/ recirc.) | 10 | 55.7 | 54.9 | 56.4 | 0.5 | 6882 |
| \*Objective function (CAC\*) differs slightly from actual cost (CAC) because of penalty function |

The process designs identified by *cascade* and *system optimization* that led to the minimum CAC are outlined in Table 3 for comparison. Very similar designs of the hybrid process are identified. *Cascade* and *system optimization* basically allocated the same separation duty to VPSA, which pre-concentrated CO2 up to 45-46%. The larger differences can be noted in the pressure levels selected for the first membrane stage. However, this difference led to a minor difference in the overall cost split between the CO2 capture technologies. All in all, it can be concluded that both optimization strategies approached what is presumably the optimal VPSA-membrane hybrid process design.

Table 3. Selected characteristics of process designs for minimum CAC.

|  |  |  |  |
| --- | --- | --- | --- |
| VPSA-membrane hybrid process | Cascade opt. (w/o recirc.) | System opt. (w/o recirc.) | System opt. (w/ recirc.) |
| Optimized process |  |  |  |
| High pressure VPSA (bar) | 2.6 | 2.4 | 2.5 |
| Low pressure VPSA (bar) | 0.3 | 0.3 | 0.3 |
| High pressure memb. stg. 1 (bar) | 2.0 | 4.5 | 1.8 |
| Low pressure memb. stg. 1 (bar) | 0.2 | 0.5 | 0.2 |
| High pressure memb. stg. 2 (bar) | 2.8 | 2.5 | - |
| Low pressure memb. stg. 2 (bar) | 0.8 | 0.9 | - |
| Performances |  |  |  |
| Purity VPSA | 45 % | 46 % | 54 % |
| Recovery VPSA | 97 % | 97 % | 97 % |
| Purity memb./hybrid | 95 % | 95 % | 95 % |
| Recovery memb. | 93 % | 93 % | 81 % |
| Recovery hybrid | 90 % | 90 % | 97 % |
| Power VPSA (kW) | 7594 | 7573 | 10472 |
| Power memb. (kW) | 4181 | 4230 | 2495 |
| Power hybrid (kW) | 11774 | 11803 | 12967 |
| CAC VPSA (€/t) | 38.8 | 39.3 | 46.1 |
| CAC memb. (€/t) | 16.6 | 16.4 | 8.5 |
| CAC hybrid (€/t) | 55.4 | 55.7 | 54.7 |

* + 1. System optimization for hybrid process with recycle

Ten additional optimization runs were performed for *system optimization* to identify the optimal design of the hybrid process when implementing stream recycle. *Cascade optimization* was not used in this case as the iterative effort required to solve a recycle configuration would have led to excessive computational time. On the other hand, *system optimization* achieved similar effectiveness in finding the optimum design at reduced execution time, hence it was used. The obtained results are reported in Table 2. *System optimization* with stream recycle slightly decreases the CAC (ca. 1% on average), while at the same time achieves higher CO2 capture rate (up to 97%, meaning that in this case the related constraint does not limit the optimum). The iterative procedure to solve a system with stream recycle resulted in a more than three-fold increase in execution time.

The process designs identified by *system optimization*, with and without stream recycle, that led to the minimum CAC are outlined in Table 3. The use of stream recycle affects the inlet gas to the hybrid process and, consequently, its optimal process design. The increased CO2 concentration eases the gas separation duty of VPSA, which can efficiently concentrate CO2 at higher levels – up to 54% compared to 46% observed without stream recycle. This, together with the increased capital expenditures due to the larger inlet gas flow rate, led to a higher cost associated to the VPSA step – 46.1 against 39.3 €/t. A single stage membrane unit is shown sufficient to perform the final CO2 purification and the cost of the membrane step is almost halved – 16.4 against 8.5 €/t. Overall, a slight cost decrease (ca. 1 €/t) and higher capture rate was achieved with stream recycle.

* 1. Conclusions

Two approaches are studied to optimize a VPSA-membrane hybrid process and identify cost-efficient designs. *System optimization* considers the system as a whole and optimized it as such. *Cascade optimization* focuses on one capture technology at a time, meaning that the two are optimized in sequence.

* The two optimization strategies identified a very similar design of the hybrid process that minimizes the cost.
* *System and cascade* *optimization* achieved basically the same cost figures (0.3% difference on average), but *system optimization* isfour times faster.
* Stream recycle leads to a cost reduction of ca. 1 €/t and to a higher CO2 capture rate. In such case, *system optimization* is the most convenient optimization strategy on the ground of reasonable computational time.

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