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Optimization of a hybrid CO_2 capture process combining a vacuum pressure swing adsorption and a carbon purification unit

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Abstract

Climate change necessitates immediate measures to reduce CO_2 emissions, which constitute 80% of greenhouse gases driving global warming. This study examines a hybrid CO_2 capture system combining Vacuum Pressure Swing Adsorption (VPSA) and a Cryogenic Carbon Purification Unit (CPU), designed to process flue gases with CO_2 concentrations between 5% and 20%. The VPSA pre-concentrates CO_2, while the CPU purifies it to >99.99% purity to reach required purity for transport. The system achieves over 90% recovery across the concentration range, balancing energy consumption, cost, and recovery efficiency by using surrogate models for multi-objective optimization,. Techno-economic analysis reveals that, for an electricity price of 75 €/MWh and a carbon tax of 100 €/t_{CO_2}, total capture costs range from 123 to 80 €/t_{CO_2} for 10–20% CO_2 concentrations in flue gases respectively. The analysis highlights that low-carbon electricity sources are crucial for minimizing emissions, as higher recovery rates may not be optimal with high-emission power. These findings emphasize also the economic feasibility of concentrations above 10%, providing a competitive solution for sustainable carbon capture.

Keywords: Carbon capture process; hybrid process; vacuum pressure swing adsorption; carbon purification unit; optimization; techno-economic analysis

Nomenclature

CPU	Carbon Purification Unit
SMT	Surrogate Modeling Toolbox
TSA	Temperature Swing Adsorption
VPSA	Vacuum Pressure Swing Adsorption

1. Introduction

The critical need for immediate and effective action to mitigate climate change requires to reduce greenhouse gas emissions, particularly carbon dioxide (CO_2), which is the primary driver of global warming. CO_2 accounts for approximately 80% of all greenhouse gas emissions and contributes to more than 50% of the greenhouse effect [1]. In December 2015, the Paris Agreement was adopted at COP 21, where 196 Parties committed to limit global warming below 2°C, aiming for 1.5°C above pre-industrial levels in the best-case scenario [2]. Achieving these targets requires

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drastic reductions in CO_2 emissions, transitioning away from fossil fuels, and prioritizing renewable energies and electrification. However, certain industrial processes, such as the decarbonation in cement production, generate unavoidable CO_2 emissions, with approximately 65% of CO_2 emissions in cement plants arising from decarbonation reaction. These emissions highlight the necessity for robust Carbon Capture, Utilization, and Storage (CCUS) technologies to complement the transition to renewable energies and mitigate industrial emissions [1,3].

The transportation and storage of captured CO_2 are essential steps in CCUS processes. In Europe, offshore CO_2 storage is preferred, necessitating efficient maritime transport via pipelines or ships. The choice between these methods depends on distance and cost-effectiveness, with shipping becoming more favorable for longer distances. To ensure safe transport and storage, strict specifications are required, including low levels of oxygen (below 10 ppm mol) and water content (below 30 ppm mol) to prevent corrosion [4]. These specifications are both critical for pipeline transport and for processes involving CO_2 conversion using catalysts, where impurities can deactivate catalysts or lead to undesirable by-products. Consequently, the entire CCUS process chain must consider these factors to ensure the effectiveness and sustainability of carbon capture initiatives [5].

Numerous carbon capture technologies have been developed, including absorption, adsorption, membrane technologies, and cryogenics, each with distinct advantages and challenges [6]. Chemical absorption using amine-based solvents remains the most mature technique due to its high Technology Readiness Level (TRL). This method captures CO_2 in a solvent, which is later regenerated by temperature increase with thermal energy to release high-purity CO_2 suitable for transport. However, this method has drawbacks, such as the degradation of solvents, environmental toxicity, and high thermal energy requirements for regeneration, which can lead to additional emissions if excess heat is unavailable [7]. Adsorption, using materials like activated carbons, zeolites, or Metal Organic Frameworks (MOFs), captures CO_2 by retaining it in the material's pores. CO_2 is then released by decreasing the pressure, increasing the temperature or flushing with a different gas. While adsorption can offers high CO_2 purity or recovery rates (it's difficult to reach both), its performance can be affected by contaminants and water vapor in the flue gas, necessitating pretreatment unit [8]. Membrane technologies separate CO_2 by exploiting pressure differentials, allowing selective permeation of CO_2 molecules. Despite their scalability, achieving both high recovery (90%+) and purity (95–99%) often requires multiple units, increasing costs and footprint [9]. Cryogenic technologies, which liquefy [10] or sublimate CO_2 [11], are effective for high-purity applications but face challenges with energy consumption, especially for low-concentration CO_2 flue gases [12].

Given the limitations of individual technologies, hybrid systems combining complementary methods have emerged as a promising solution. Various configurations, such as membrane-absorption [13], membrane-adsorption [14], adsorption-membrane [14], cold membrane integrated into a cryogenic unit [15–18], or adsorption-cryogenics [19] have been explored. In this study, a hybrid system integrating a Vacuum Pressure Swing Adsorption (VPSA) process with a high-purity Cryogenic Purification Unit (CPU) is proposed, following the concept outlined by Rodrigues et al. [20,21]. VPSA is employed to pre-concentrate CO_2 while keeping a high recovery to a minimum purity of 50 mol%, the threshold at which CPU systems become energy-efficient. CPUs then purify CO_2 to 99.999 mol%, ensuring suitability for transport and catalytic utilization.

This study uses Aspen Plus® and Aspen Adsorption® V14 to model the cryogenic and VPSA units, respectively. Surrogate models, which convert simulations into mathematical representations, are employed to optimize the hybrid process at lower computational cost. These models enable the simultaneous optimization of energy consumption, cost, and CO_2 recovery, offering significant advantages over optimizing each process individually in terms of simulation time. By integrating VPSA and CPU technologies, this work aims to develop an innovative carbon capture system capable of achieving high recovery and purity levels while minimizing energy consumption. The findings provide a detailed energetic and economic analysis, contributing to the development of sustainable and efficient CCUS solutions for industrial applications.

2. Design of the process

2.1. Process configuration

The studied process combines a Vacuum Pressure Swing Adsorption process and a Carbon Purification Unit to capture CO_2 from flue gases with concentrations between 5–20 mol% and a flow rate of 70,000 Nm^3/h, equivalent to approximately 1,000 tons per day of clinker production (see Fig. 1), but it is necessary to dry the flue gases beforehand Thermal Swing Adsorption (TSA) using silica gel or alumina is generally employed for this dehydration [22,23] ; not take into account in this study. Additionally, large-scale removal of pollutants such as dust, NO_x, SO_x, and mercury may be required to protect materials, although this study focuses exclusively on nitrogen and carbon dioxide.

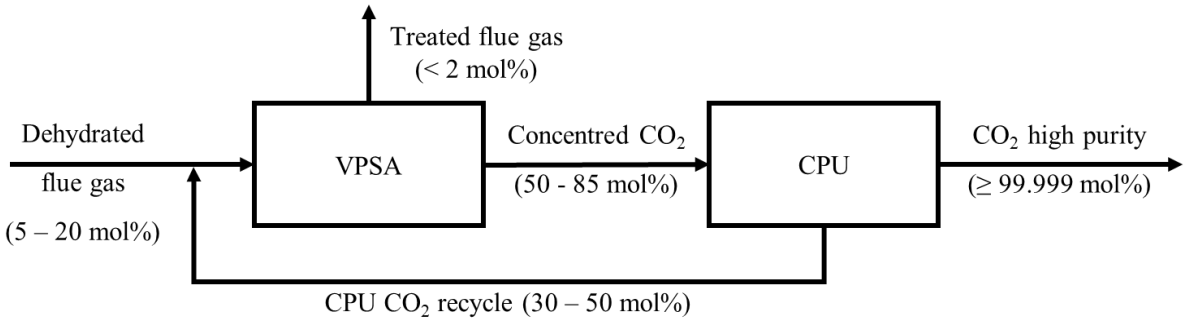

Fig. 1. Hybrid VPSA-CPU CO_2 capture process.

The VPSA unit pre-concentrates CO_2 from the flue gas to a minimum of 50 mol%, preparing it for further purification in the CPU. The CPU then purifies the CO_2 by liquefying the flue gas, separating liquid CO_2 from non-condensable gases. These non-condensable gases are recycled back to the VPSA to recover any unliquefied CO_2, maximizing recovery. The final output of the CPU achieves an ultra-high CO_2 purity of 99.999 mol%. The overall CO_2 recovery rate depends on the impact of the CPU's recycled gas on VPSA performance. While the industry-standard target for CO_2 recovery is traditionally 90%, recent studies aim for values of 95% or higher, with the possibility of exceeding these benchmarks if economically viable. As such, optimizing recovery rates is a central focus of this study.

The primary objective is to minimize the electrical energy consumption of the entire CO_2 capture process while achieving a maximum recovery rate by globally optimizing the hybrid system comprising VPSA and CPU. Traditionally, each unit is optimized to achieve specific recovery and purity targets while reducing energy costs, a major factor in overall process expenses. In the integrated system, the VPSA determines the CO_2 recovery, while the CPU finalizes the purity. This integration introduces flexibility in the purity level of the CO_2 leaving the VPSA, which directly impacts energy consumption. Higher CO_2 concentrations from the VPSA reduce the CPU's energy demand but increase the energy required for VPSA operation. Achieving a balanced trade-off between these factors is crucial to minimizing the total energy consumption.

2.2. VPSA modelling

The first stage of the process is a 2-bed VPSA unit performing the Skarstrom cycle with a pressure equalization step (2-bed 5-step). The sequence of steps for one bed, along with the corresponding pressure profile, is illustrated in Fig. 2.

The cycle begins with the adsorption step, where flue gas is introduced into the column. During this step, CO_2 is primarily adsorbed, and a nitrogen-rich gas exits the column. Following adsorption, the column is connected to a

second column at a lower pressure for pressure equalization. This step reduces the mechanical work required for compression and vacuum.

Next, during the blowdown step, the pressure is further reduced to desorb CO_2 from the adsorbent, allowing its recovery. To enhance CO_2 retrieval, a portion of the nitrogen-rich stream produced during the adsorption step is used in the purge step. This stream is introduced into the second column to flush out residual CO_2.

Finally, the two columns are reconnected for another pressure equalization step, after which the bed is repressurized with flue gas until the target adsorption pressure is reached. To maintain synchronization between the two beds, the duration of the blowdown and purge steps must match the duration of the adsorption and pressurization steps.

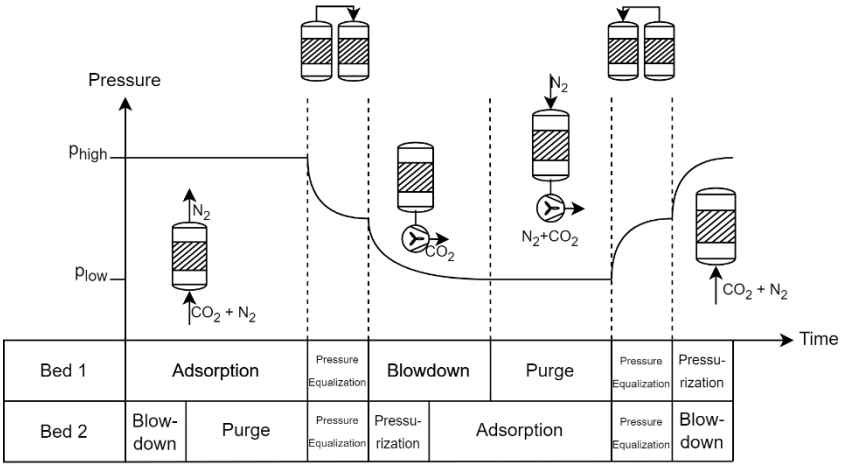

Fig. 2. Sequence of steps and pressure profile for the cycle used in the VPSA unit.[24]

The first stage of the hybrid process is composed of 5 VPSA units treating each 14,000 Nm^3/h. The volume of each adsorption bed is 80 m^3 with a diameter of 3.24 m and length of 9.71 m. The beds are filled with 3 mm beads of zeolite 13X which is a benchmark adsorbent used for CO_2 capture. The 2-bed 5-step cycle is modeled in Aspen Adsorption V14 using axially dispersed plug flow model for mass balance, Ergun equation for momentum balance, and three energy balances (solid, gas, and wall of the column). More information about the model equations and the parameters used is available in the reference [24].

2.3. CPU modelling

The second stage of the process involves the Carbon Purification Unit, inspired by the Air Liquide Callide project [25,26] and based on studies by Costa *et al.* [27]. A schematic representation of this process is shown in Fig. 3. The CPU consists of six main steps: (1) flue gas compression, (2) flue gas cooling, (3) CO_2 vapor-liquid separation, (4) CO_2 purification, (5) cold generation, and (6) CO_2 compression. This unit refines the CO_2 pre-concentrated by the five VPSA units, which supply a minimum inlet concentration of 50 mol% CO_2 to maintain energy efficiency.

The cryogenic unit plays a key role in purifying CO_2 by removing non-condensable gases. This is accomplished through cryogenic liquefaction, leveraging the unique phase behavior of CO_2. To prevent the formation of dry ice (solid CO_2), the temperature is carefully controlled to -54 °C, just above its triple point of -56.6 °C, ensuring efficient separation of phases.

The CO_2-rich stream from the VPSA units is first subjected to multi-stage compression with water intercooling (1). This prepares the flue gas for liquefaction, which is carried out using multi-stream brazed aluminum heat exchangers

(BAHX) (2). The proven effectiveness of this technology has been demonstrated in projects like the Callide project [25].

In the next step, the liquefied CO_2 is separated from non-condensable gases using a flash separator (3). This step ensures that the liquid CO_2 entering the subsequent process exceeds 95 mol% purity. To improve energy efficiency, the cold energy from the non-condensable gases is recovered using BAHX and reused within the system. These gases are then expanded to generate energy before being recycled back into the VPSA units, enhancing overall plant efficiency.

The purified liquid CO_2 is then fed into a desorption column (4), where impurities are stripped using pure CO_2 vapor. This step is critical for achieving ultra-high CO_2 purity of 99.999 mol%. A portion of the liquid CO_2 product is heated to supply pure CO_2 vapor for the column, while the remaining liquid is subjected to Joule-Thompson expansion (5). This expansion generates the cold needed for the liquefaction process, contributing to energy efficiency.

Finally, the purified CO_2 is compressed to reach the supercritical pressure of 110 bar, making it suitable for pipeline injection and distribution (6). This compression ensures the CO_2 is ready for transport, while the integration of cold recovery and energy generation throughout the process maximizes overall system efficiency.

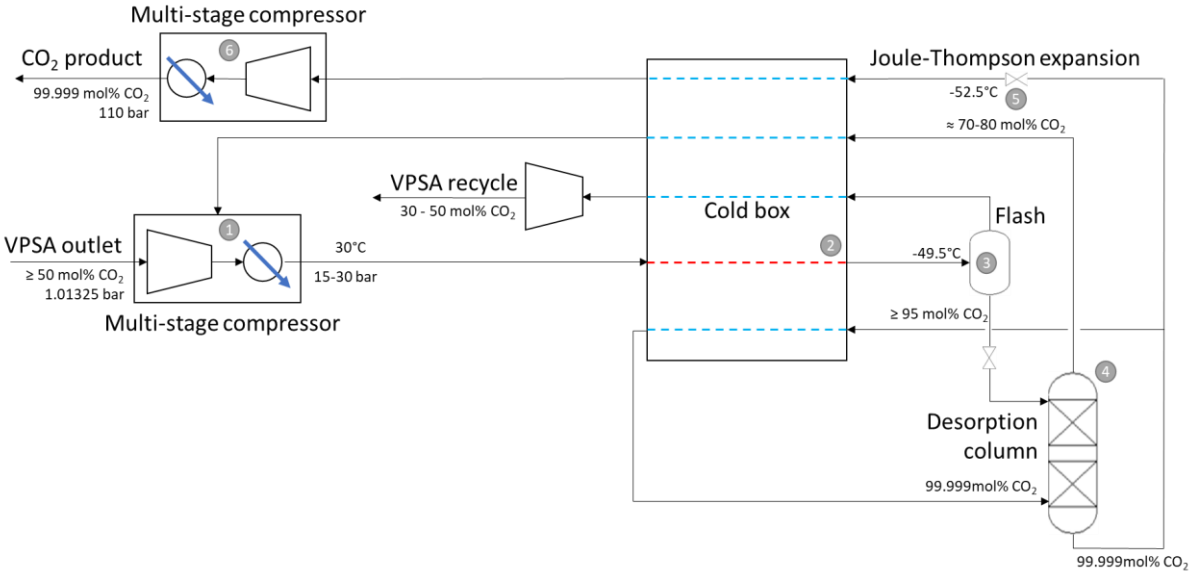

Fig. 3. Schematic plan of the CPU adapted from Costa *et al.* [27].

3. Optimization

The study focuses on optimizing a hybrid CO_2 capture process combining Vacuum Pressure Swing Adsorption and a Carbon Purification Unit (5 parameters are used for VPSA and 5 others CPU see figure 4, for the list). Simulation of these processes in Aspen software involves solving thousands of equations, leading to long computation times. To address this, surrogate models were developed for both units, significantly reducing computational effort while maintaining reasonable precision. These models were constructed using Latin hypercube sampling to generate data points, which were split for training (80%) and validation (20%). Kriging model is used to predict new results based on the training points made previously. Python's Surrogate Modeling Toolbox (SMT) was used for implementation [28].

The surrogate models allow the simulation of steady-state interactions between VPSA and CPU. The VPSA produces a CO_2-enriched stream processed by the CPU, which in turn returns a recycled stream to the VPSA. Convergence is achieved when variations in flow rate and CO_2 concentration in the recycled stream fall below a set tolerance (10^{-3}). Recovery and purity transformations were applied to ensure model robustness, and the electrical consumption of the integrated process was minimized. More information available in our previous work [24].

To optimize the coupled process with multiple objectives, such as maximizing recovery and purity while minimizing energy consumption, the U-NGSA-III genetic algorithm was used. This algorithm excels in multi-objective optimization by preserving diversity and efficiently identifying Pareto-optimal solutions. Python's Pymoo library [29] facilitated the application of this algorithm.

The study demonstrates an effective methodology for linking and optimizing VPSA and CPU systems (Fig. 4.), enabling the design of an energy-efficient CO_2 capture process. The approach balances recovery, purity, and energy consumption while reducing computational complexity.

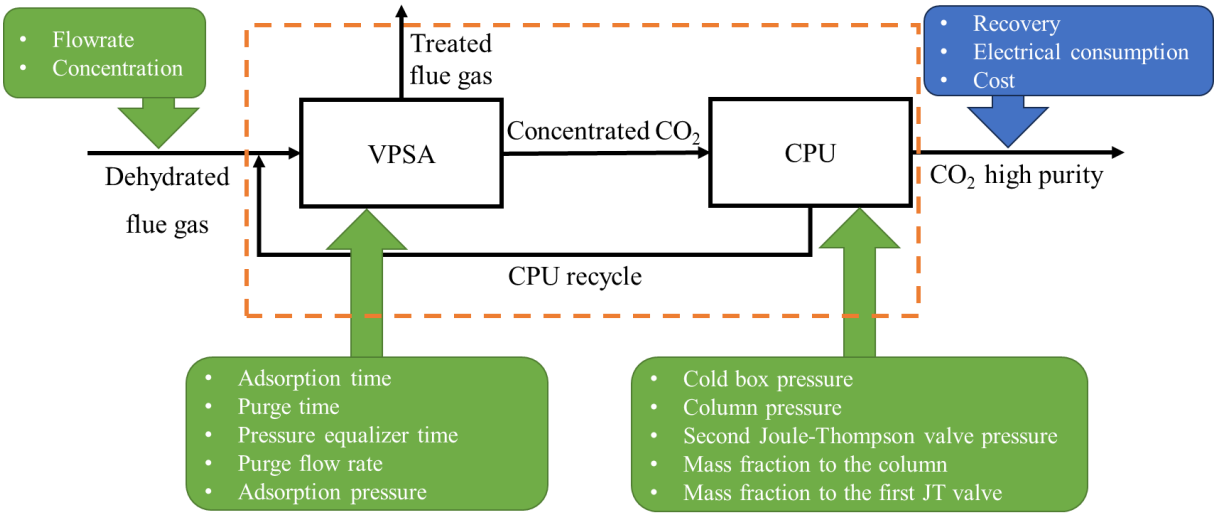

Fig. 4. Linking VPSA+CPU with inputs and outputs.

4. Results and discussion

The optimization of the VPSA and CPU coupling highlights the trade-offs between CO_2 recovery and electrical consumption, as well as how energy demand shifts between the two units depending on operating conditions. Fig. 5. provides a detailed comparison of energy consumption for various flue gas CO_2 concentrations (5, 10, 15, and 20 mol%) and recovery levels. For flue gases with a 5 mol% CO_2 concentration, the VPSA unit dominates energy consumption, accounting for more than 65% of the total. This higher share is attributed to the significant compression requirements for the incoming flue gas and the need to operate at low blowdown pressures to enhance CO_2 adsorption with the same amount of adsorbent. Additionally, achieving the required 50% CO_2 purity at the VPSA outlet imposes conditions that are not energy-optimal, further increasing the VPSA's energy consumption. At this concentration, recovery rates above 95.5% are unattainable due to these operational constraints.

As the inlet CO_2 concentration increases to 10 and 15 mol%, the energy consumption between the VPSA and CPU becomes more balanced, with each unit consuming approximately equally the total (even more in the case of 10% for VPSA). The higher concentration improves the efficiency of the VPSA by elevating the partial pressure of CO_2, which enhances the adsorption process and reduces the energy required per unit of CO_2 captured. This balance also reflects an improved interplay between the VPSA and CPU, as the CPU can handle a more concentrated stream, reducing the

recycling load. For flue gases with 20 mol% CO_2, the CPU overtakes the VPSA as the primary energy consumer. This shift occurs because the VPSA operates more efficiently at higher inlet concentrations, producing efficiently a relatively low-purity output stream that requires more extensive processing by the CPU to achieve the target purity. Additionally, as recovery rates increase, the CPU's energy consumption rises drastically compared to the VPSA, reflecting the additional work needed to purify the lower-quality VPSA output stream.

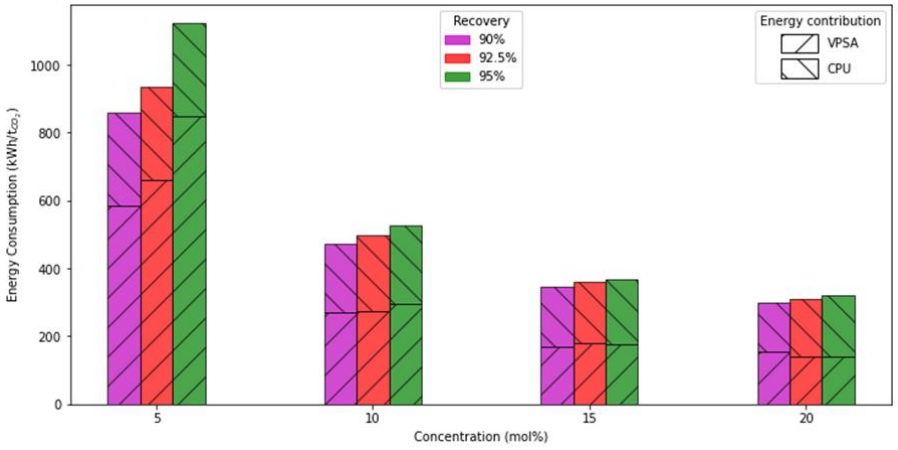

Fig. 5. Energy consumption of VPSA and CPU at several recovery for different inlet CO_2 concentrations.

Overall, the results demonstrate that energy distribution between the VPSA and CPU depends strongly on the inlet CO_2 concentration and recovery targets. At lower concentrations, the VPSA is the dominant energy consumer, while at higher concentrations and recovery rates, the CPU plays a larger role. These dynamics underscore the importance of careful optimization to balance recovery, purity, and energy consumption, tailored to specific operational scenarios.

4.1. Economics analysis

The main objective of this study is to minimize the electrical consumption of the VPSA-CPU process while evaluating its economic viability.

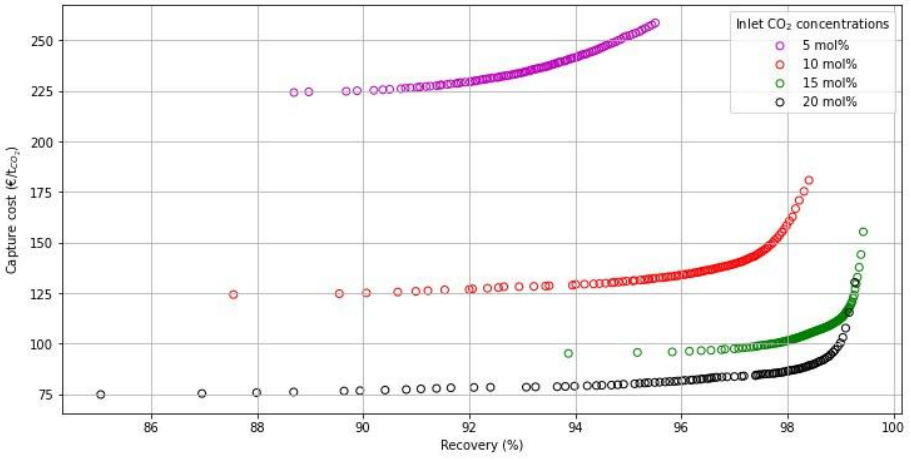

Fig. 6. CO_2 capture cost versus CO_2 recovery for different inlet CO_2 concentrations.

Since operational costs far exceed fixed annual costs, it is crucial to analyze electricity consumption and carbon taxation to identify the most cost-effective recovery rates and compare this approach with other CO_2 capture technologies methods. Electricity costs are the dominant factor, accounting for more than 90% of total expenses when electricity is priced at 100 €/MWh. As shown in Fig. 6, capture costs increase with higher CO_2 recovery rates and are significantly higher for flue gas with 5 mol% CO_2 compared to higher concentrations. This is attributed to the higher energy demand for processing low-concentration CO_2 streams, which requires additional compression and adsorption efforts. Costs rise steeply as recovery approaches 99%, driven by reduced purity at the VPSA outlet and the increased energy demand of the CPU.

Carbon taxation, based on the quantity of uncaptured CO_2, provides an additional information for determining optimal recovery rates. By balancing capture costs and carbon tax penalties, total costs can be calculated as outlined in equation (1). Fig. 7 illustrates the evolution of total costs across recovery rates and carbon tax levels for various CO_2 concentrations. At an electricity price of 100 €/MWh and carbon taxes ranging from 70 to 130 €/t_{CO_2}, the cost-optimal recovery is generally below 90%. However, as the carbon tax increases, the optimal recovery shifts toward higher rates, reflecting the economic benefit of capturing more CO_2 and minimizing emissions.

$$Total\ cost = \frac{CO_2\ recovery}{100} CO_2\ capture\ cost + \frac{(100 - CO_2\ recovery)}{100} carbon\ tax \quad (1)$$

These results emphasize the critical role of electricity prices and carbon taxation in defining the economic efficiency of the VPSA-CPU process. Optimizing the system for specific CO_2 concentrations and financial scenarios enables a more cost-effective approach to carbon capture.

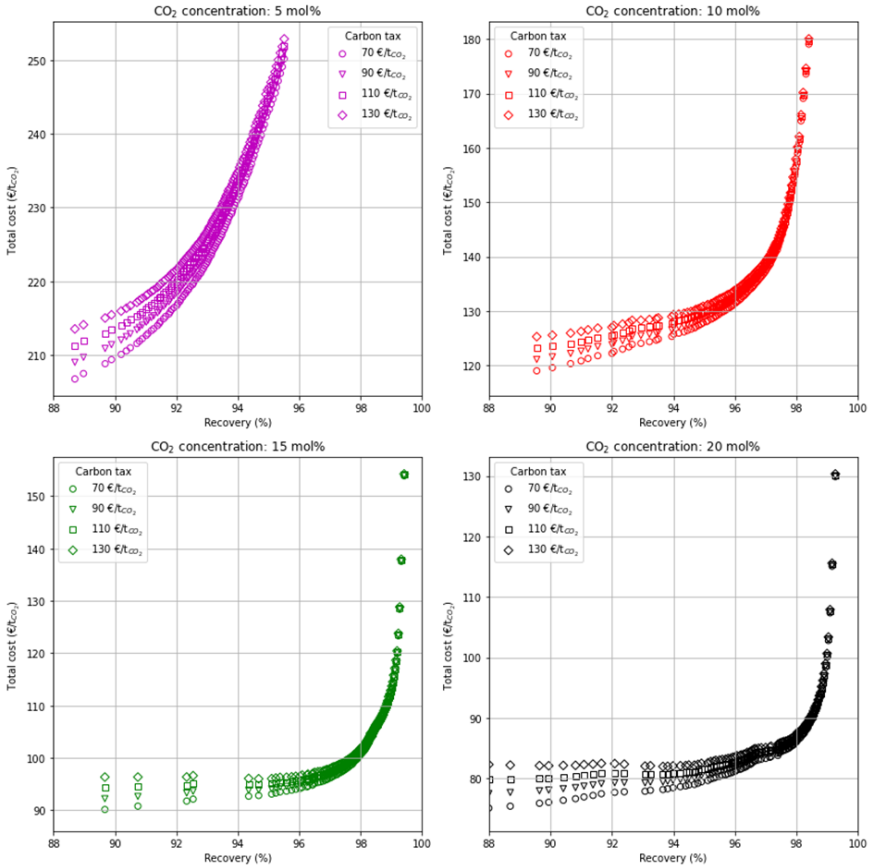

Fig. 7. CO_2 capture cost versus CO_2 recovery for different inlet CO_2 concentrations.

The environmental impact of a fully electric carbon capture unit is evaluated based on various electricity sources, highlighting the importance of sustainable energy choices. This analysis examines the environmental consequences of the power source for electric-driven carbon capture systems, emphasizing the need for renewable energy sources to enhance the overall sustainability of the process. The study considers traditional power sources such as coal and natural gas alongside renewable options like wind and solar, with the European electricity mix (ENTSO-E) included to assess the current impact on regional energy producers. Solar energy emissions are shown to be similar to those of wind energy.

Fig. 8 presents the CO_2 avoided, calculated as the captured CO_2 minus the emissions from electricity production, normalized to the total CO_2 in the initial flue gases. The formula for CO_2 avoided is as follows:

$$CO_2\ avoided = \frac{CO_2\ captured - CO_2\ emitted\ by\ power\ production}{CO_2\ flue\ gas} \quad (2)$$

When wind energy powers the system, the CO_2 avoided closely matches the recovery rates, as wind generation has minimal emissions. However, emissions from the European electricity mix are similar to those of natural gas plants, which moderately reduce the CO_2 avoided. Across all flue gas concentrations, the CO_2 avoided peaks at over 98%. At a 90% recovery rate, emissions from the European mix led to CO_2 avoided reductions of 34%, 18%, 13%, and 11% for concentrations of 5%, 10%, 15%, and 20%, respectively.

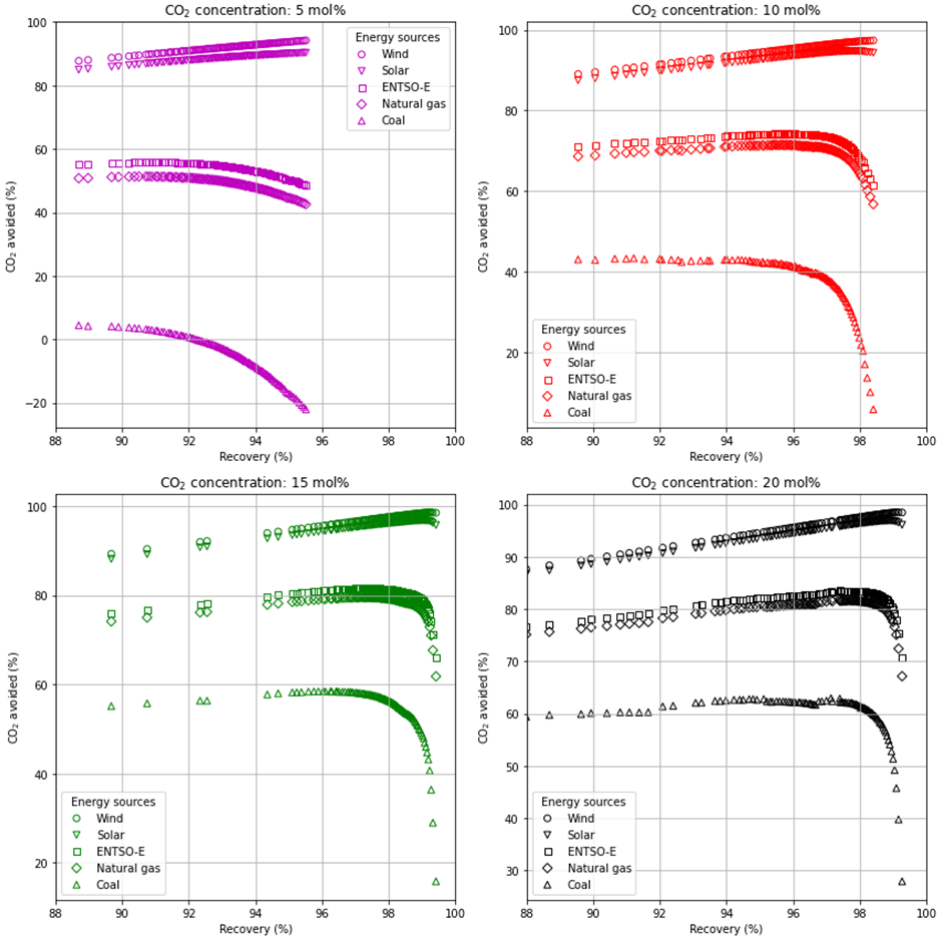

Fig. 8. CO_2 avoided versus recovery and carbon tax for different inlet CO_2 concentrations. (Emissions factor of electricity (kg_{CO2e}/kWh): Wind = 0.011; European Network of Transmission System Operators (ENTSO-E) = 0.399; Natural gas = 0.450; Coal = 1.000).

Notably, a peak in CO_2 avoided indicates that increasing recovery rates beyond a certain point, requiring higher energy consumption, may not be optimal if the energy source is carbon-intensive. For instance, at 5 mol% CO_2 concentration with coal-powered electricity, the CO_2 avoided is significantly compromised. In this case, high electricity consumption for recovery rates above 92% results in more CO_2 being emitted than captured, undermining the process's environmental benefit. This underscores the necessity of integrating low-emission energy sources for effective and sustainable carbon capture.

5. Conclusions

This study explores the integration of vacuum pressure swing adsorption and a carbon purification unit to create a hybrid CO_2 capture system capable of high recovery rates and producing CO_2 at transport-grade purity. Studied for flue gas concentrations ranging from 5% to 20%, the system relies entirely on electricity, offering a sustainable and environmentally friendly solution for carbon capture. This solution do not require chemical solvents storage and utilisation.

The use of surrogate models enabled optimization across key objectives, such as energy efficiency, cost, and CO_2 recovery. The analysis highlights a critical trade-off between recovery rates and electrical consumption, revealing that higher CO_2 concentrations in the feed gas reduce energy demands. Recovery rates above 90% are achievable, but balancing cost and environmental benefits require careful consideration of electricity prices and carbon taxes.

Techno-economic analysis identified optimal recovery points based on cost scenarios. For 100 €/MWh electricity price and carbon taxes exceeding 100 €/tCO_2, average capture costs are 123, 95, and 80 €/tCO_2 for CO_2 concentrations of 10%, 15%, and 20%, respectively, emphasizing the economic infeasibility of 5% CO_2 concentrations.

The study also examines the environmental impact of electricity sources. Low-carbon energy aligns recovery rates with CO_2 avoided, but the use of high-emission energy can make lower recovery rates more efficient.

In conclusion, this research provides a comprehensive framework for optimizing hybrid CO_2 capture systems, balancing efficiency, cost, and environmental impact. The findings contribute to advancing carbon capture technology, supporting its role in mitigating climate change and reducing greenhouse gas emissions.

Acknowledgements

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