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Optimization of a hybrid CO_2 capture process combining a VPSA using 13X/MOFs and CPU

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Abstract

Carbon capture is one of the key technologies to drastically reduce CO_2 emissions to reach the target of zero emission by 2050 established by the Paris Agreement. Numerous carbon capture technologies have been developed in the past few years but most of them failed to reach both high CO_2 purity (>95%) and recovery (>95%) especially for low CO_2 concentration flue gas (below 10% vol.). Moreover, the strict conditions imposed by CO_2 transport (liquefaction or supercritical pipeline) require very high purity levels and low impurities level (for example less than 10 ppm O_2 and 30 ppm H_2O), requiring the use of a purification unit at the outlet of the CO_2 capture unit. To overcome this problem, hybrid technologies are being considered to mix the advantages of two capture technologies allowing to reach the target for CO_2 transport and high recovery of CO_2.

In this work, a hybrid system combining a vacuum pressure swing adsorption (VPSA) unit and a cryogenic carbon purification unit (CPU) has been investigated to treat a flue gas of 70 000 Nm³/h corresponding to around 1000 t/day of clinker production. 5% to 20% CO_2 in the flue gas was considered in order to reach a recovery of CO_2 higher than 95% while complying with the specifications required for transporting CO_2 (purity higher than 99%) [1]. In this configuration, the VPSA unit acts as a CO_2 preconcentration unit allowing to reach a purity higher than 50% with a recovery as high as possible. Then, the CPU allows the CO_2 concentration to a value higher than 99.999% while removing the impurities such as O_2. The waste stream of the CPU is recycled to the inlet of the VPSA unit to avoid a loss of CO_2 (Figure 1). First, water is removed from the flue gas before the VPSA unit by a temperature swing adsorption unit (TSA) to avoid the degradation of the zeolite 13X performances [2].

In the initial phase, the VPSA cycle used is the well-established Skarstrom cycle, incorporating a pressure equalization step [3]. This cycle is widely utilized in commercial applications, such as hydrogen purification from SMR or oxygen production–. While it enables high CO_2 recovery, it faces challenges in achieving high levels of purity. Zeolite 13X was used for the VPSA due to its commercial availability and its performance for the CO_2/N_2 separation. The simulated VPSA is composed of 5 parallel unit with a bed volume of 100 m³. Several variables were studied for the optimization of this unit: adsorption time [60-1500s], light reflux time [10-1490s], pressure equalization time [10-50s], light reflux flow rate [100 – 20,000 Nm³/h], adsorption [1-3 bar] and evacuation pressure [0.01 – 0.5 bar]. In addition, the feed flow rate [14,000 – 25,000 Nm³/h] and inlet CO_2 concentration [5 – 27%]

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were also varied to consider the reflux flow from the CPU unit. The wide bounds of the operating conditions are due to the range of flow rates and CO_2 concentrations which must be treated by the VPSA unit.

The CPU employed, previously studied by Costa et al. [4], consists of a multi-stage compressor that raises the gas to high pressure, followed by a cooling stage to reduce the temperature to -49.5°C. Under these pressure and temperature conditions, a liquid phase is formed, which is separated from the gas phase in a flash unit. The liquid CO_2 is then purified in a desorption column by stripping with pure gaseous CO_2 to eliminate dissolved impurities. The purified liquid CO_2 is subsequently compressed to high pressure for transportation. The required cold energy is supplied through the Joule-Thompson expansion of CO_2, with full energy integration achieved via a multi-stream brazed aluminum heat exchanger.

For the hybrid process investigation, a surrogate modeling approach was employed to reduce simulation time. A design of experiments with the various variables for each unit was generated, and both units were independently simulated using Aspen V14 software. The results from these experiments, including CO_2 recovery, energy consumption, and cost, were used to construct and validate the surrogate models. The simulation of the hybrid unit was achieved by coupling the two surrogate models.

The study results demonstrate that the hybrid system achieves a CO_2 recovery of over 90% for the flue gas concentration range considered, while maintaining high-purity CO_2 production (>99.99%), suitable for transportation. A trade-off analysis revealed a balance between recovery, electricity consumption, and economic feasibility. Sensitivity analysis identified key parameters influencing recovery and energy consumption, offering insights for future optimization. Techno-economic analysis underscored the effects of electricity prices and carbon taxes on total costs, with an optimal shift towards higher recovery rates observed as carbon taxes increase. Additionally, the economic viability of the system was found to be dependent on CO_2 concentrations above 10% proving more favorable compared to other technologies that require higher concentrations. At an electricity price of 75 €/MWh and a carbon tax of 100 €/tCO_2, the total cost of CO_2 capture for concentrations ranging from 10% to 20% was found to range between 123 and 80 €/tCO_2. The analysis further highlighted the significance of a low-carbon energy mix for achieving optimal carbon emission reductions. A first optimization step of the process was performed by changing the volume of the adsorption beds of the VPSA units whith a constant L/D of 4. This supplementary parameter allows to reduce the energy consumption of the VPSA unit up to 10% compared to the initial volume of 100m³. To improve the energy consumption of the VPSA unit, zeolite 13X was replaced by two promising MOFs as adsorbents, MIL-160(Al) and MIL-120(Al), to evaluate the performance of these materials in a coupling with CPU unit. Previous study shows that MIL-160(Al) outperformed zeolite 13X with the 3-bed 6-step cycle [5] making this MOF an interesting candidate for this work. MIL-120(Al) exhibits numerous interesting properties (high CO2 capacity, high CO_2/N_2 selectivity, moderate heat of adsorption, easily scalable at tons scale) which could reduce the OPEX and CAPEX of the VPSA. In addition, an alternative VPSA cycle, the 3-bed 6-step cycle developed by Khurana and Farooq (adsorption, heavy reflux, co-current evacuation, counter-current evacuation, light reflux, light product pressurization) [6], was also studied for the coupling of the two units. This cycle shows excellent results in terms of recovery and purity at moderate vacuum pressure (0.1 bar) but at the cost of higher energy consumption. The optimization of material and cycle used allows to further reduce the cost of the capture for the different CO_2 concentrations studied.

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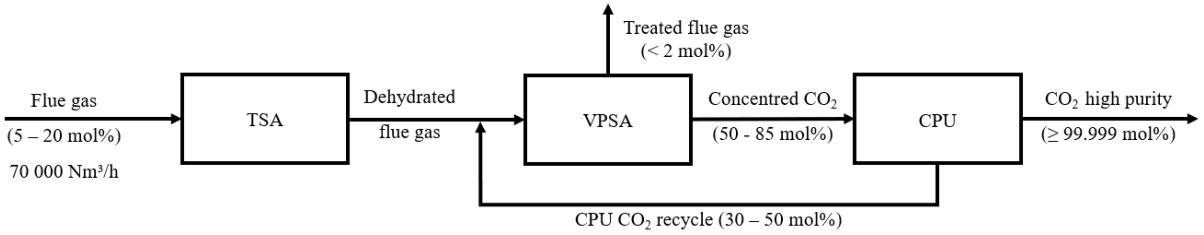

Figure 1: Hybrid VPSA-CPU carbon capture process.