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Dual-reflux pressure swing adsorption process for carbon dioxide capture from dry flue gas

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ABSTRACT

This work presents a detailed experimental and simulation study for capturing and recovering high purity carbon dioxide from a dry flue gas (15%CO_2/85%N_2) via a laboratory-scale dual-reflux pressure swing adsorption apparatus using silica gel as an adsorbent. To achieve a favorable carbon capture performance, parameter study was implemented to obtain a series of optimized settings of process operating variables by investigating the effects of flowrate of light product reflux, flowrate of heavy product and feed inlet position on the separation performance in terms of product purity, product recovery, and adsorbent productivity, as well as specific power-consumption. Under the optimal manipulation parameter configuration, a high-purity CO_2 product with a concentration of 95.55% and a higher CO_2 product recovery of 96.81% with a competitive specific power consumption of 110.58 kJ $mol^{-1}CO_2$ can be obtained. This finding demonstrated an efficient and economical method in the field of carbon capture and storage. In addition, a good consistency between simulation results and experimental data confirmed the reliability of the mathematical model of dual-reflux pressure swing adsorption process, and the dynamic distribution behaviors of pressure, temperature and gas-solid concentration were obtained from the simulation results to illuminate and validate the process separation performance in depth under different operating conditions.

1. Introduction

The carbon capture and storage (CCS) technology is expected to play a key role in future energy matrix because fossil fuels will remain the dominate energy source for decades into the future (Leeson et al., 2017). Recent scientific research developments have demonstrated that adsorption technology is a competitive option for capturing carbon dioxide from fossil fuel emission sources and to mitigate the emission of carbon dioxide, especially the vacuum pressure swing adsorption (VPSA) technology due to its ease of applicability over a relatively wide range of temperature and pressure conditions, its low energy requirements and low capital investment costs (Agarwal et al., 2010; Wang et al., 2013a, 2013b; Krishnamurthy et al., 2014; Webley, 2014).

Numerous articles that focus on carbon dioxide capture using the VPSA process have been extensively reported. Xiao et al. (2008) studied a 3-bed-9-step VPSA cycle using zeolite 13X for CO_2 capture from a flue gas containing 12% carbon dioxide to obtain a CO_2 product of 90% purity with a recovery greater than 70% under evacuated pressure lower than 4 kPa. Choi et al. (2003) investigated the optimal selection of operating parameters both theoretically and experimentally for maximizing carbon dioxide recovery from flue gas using a 3-bed-7-step VPSA cycle. These researchers' optimization results showed that 13% of CO_2 in the feed gas can be enriched to a high CO_2 purity value of 95% with a recovery of 70%. Reynolds et al. (2008) evaluated several stripping PSA configurations for concentrating carbon dioxide from flue gas at a high temperature level using a K-promoted HTIc adsorbent. A 5-bed-5-step stripping PSA cycle configured with a light reflux step and a heavy reflux step achieved the best separation performance of a 98.7% CO_2 purity and recovery at a feed throughput of 5.8 LSTP/hr/kg. Krishnamurthy et al. (2014) constructed a pilot-scale two-bed VPSA process using Zeochem zeolite 13X for CO_2 capture from flue gas. Using a simple four-step cycle configuration, CO_2 can be concentrated to 94.8 ± 1% with a recovery of 89.7 ± 5.6%, while the productivities can achieve 0.87-1.4 ± 0.07 t CO_2 m^{-3} adsorbent day^{-1} with a specific power consumption of 339–583 ± 36.7 kWh $tonne^{-1}$ CO_2. In their follow-up study, a rigorous mathematical model of the VPSA process was developed in a framework of MATLAB, which was validated using data from the pilot plant. Meanwhile, a systematic optimization work for the VPSA cycle using the GA-based multi-objective optimization algorithm was also performed to capture CO_2 from dry

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Nomenclature

P	Pressure (bar)
P_H	Pressure of high pressure column (bar)
P_L	Pressure of low pressure column (bar)
P_E	Pressure of equalization column (bar)
Q_F	Standard volumetric flow rate of feed stream (slpm)
Q_{HP}	Standard volumetric flow rate of heavy product stream (slpm)
Q_{HR}	Standard volumetric flow rate of heavy product reflux stream (slpm)
Q_{LP}	Standard volumetric flow rate of light product stream (slpm)
Q_{LR}	Standard volumetric flow rate of light product reflux stream (slpm)
q_i	Adsorbed phase concentration of component i (mol·kg^{-1})
R_{HP}	Recovery of component in a heavy product
R_{LP}	Recovery of component in a light product
$Time$	Time (s)
T	Temperature (K)
y_i	Gas phase mole fraction of component i
y_{HP}	Mole fraction of a component in a heavy product
y_{LP}	Mole fraction of a component in a light product
Z	Axial coordinate (m)

flue gas (Haghpanah et al., 2013a, 2013b). These researchers' optimization results also revealed that a 6-step VPSA cycle configured with a light product pressurization step and a light reflux and a heavy reflux step is capable of meeting a 95%–90% purity-recovery target under an evacuation pressure of up to 0.2 bar (Khurana and Farooq, 2016). Recently, Yan et al. (2016) employed a 2-bed-6-step VPSA cycle using silica gel as an adsorbent to study the process of power consumption optimization for capturing CO_2 from dry flue gas. Optimization results indicated that energy consumption can be reduced to an economic level of 419.99 kWh t^{-1} CO_2 from a high value of 623.64 kWh t^{-1} CO_2 when the target product purity was restricted to no less than 90%.

To recover CO_2 from flue gas with both high purity and high recovery, a two-stage pressure swing adsorption process has been developed by relevant researchers. Cho et al. (2004) experimentally established a pilot scale two-stage PSA process for carbon dioxide recovery from a flue gas containing 10.5% of CO_2. A CO_2 product with a 99% purity at 80% recovery can be obtained at a cost of theoretical power consumption of 0.28 kWh/Nm^3 CO_2. Liu et al. (2011) performed a two-stage VPSA process using 5A zeolite as an adsorbent for capturing CO_2 from flue gas via numerical simulation, where a 3-bed-5-step cycle was used for the first stage, and a 2-bed-6-step cycle was used for the second stage. First, a 15% CO_2 feed gas was enriched to 69.15% during the first stage, and the CO_2 purity can subsequently be increased to 96.05% during the second stage. Meanwhile, the total recovery was 91.97%, and the overall energy consumption was 645.7 kWh/tonne. Wang et al. (2013a, 2013b) have also constructed an industrial scale two-stage VPSA process using 13X APG (first stage) and activated carbon (second stage) as adsorbents for the CO_2 concentrate from a dry flue gas, which is released from coal-fired power plants. A good separation performance of 95.6% of CO_2 purity and 90.2% recovery with a 677.78 kWh t^{-1} CO_2 power consumption can be achieved. Finally, a comparison of operating conditions and process performances employing different processes for CO_2 capture from dry flue gas is presented in Table 1. It can be seen from Table 1 that the ultra-deep evacuation pressure is always necessary for producing high purity CO_2 product with high recovery. Currently, the specific power consumption of the VPSA process for CO_2 capture is likely to remain at approximately 2 MJ/kg CO_2, which is determined experimentally. This value is significantly lower than that for the amine scrubbing process [3–4.6 MJ/kg CO_2] (Abu-Zahra et al., 2007).

It is worth noting that the heavy product purge step and higher evacuation pressure are indispensable configurations to be applied to capture CO_2 targeting at high purity and recovery for the single stage VPSA process, while the two-stage VPSA process will notably increase the process complexities and investment costs. Recently, the dual-reflux pressure swing adsorption (DR-PSA) process has attracted significantly more attention because of its higher separation capacity and a significantly simpler process configuration for gas sharp separation. The DR-PSA process can be divided into an asymmetric two-stage unit in series and operated with an intermediate feed inlet position, as well as a dual reflux policy. This approach is a logical extension of stripping PSA and rectifying PSA, which combines them into a single two-bed system, which is analogous to distillation. This type of PSA process allows both the light and heavy products to be produced at high purity simultaneously, with neither product's purity being constrained by thermodynamic limitations (Diagne et al., 1994; Bhatt et al., 2013).

Gas separation process that is based on DR-PSA configuration has also been extensively studied in the literature. Ebner and Ritter (2004) first applied the linear isotherm equilibrium theory model to design a DR-PSA process for a binary feed separation. The subsequent parameter study, which considered feed concentration, light and heavy product reflux ratio, pressure ratio, and feed inlet position, indicated that an excellent separation performance can be achieved to produce two 100% pure product streams simultaneously. Kearns and Webley (2006a, 2006b) further evaluated four configurations of DR-PSA separately to produce two high-purity product streams from a binary feed based on the equilibrium theory model, which provided a valuable insight into the characteristics of dual-reflux PSA process and assessed its process performance on the relative energy consumption and productivity. McIntyre et al. (2010) and Bhatt et al. (2015) explored a DR-PSA process experimentally and theoretically, and employed a cycle configuration of low-pressure bed feeding and pressurization/blowdown with

Table 1
Comparison of operating conditions and performances for different processes for CO_2 capture from flue gas.

Process	Adsorbent	y_{Feed}/%	Vacuum pressure	CO_2 purity/%	CO_2 recovery/%	Power Consumption	Literature	type
PSA	13X	12.6	5–6 kPa	90–95	60–70	6–10 kW/TPDc	Zhang et al. (2008)	Exp
VPSA	13X	15	10 kPa	95	80	2.29 MJ/kg CO_2	Agarwal et al. (2010)	Sim
VPSA	5A	15	5.5 kPa	71–81	79–91	2.64-3.12 MJ/kgCO_2	Liu et al. (2012)	Exp
VPSA	13XAPG	15.5	7–8 kPa	73–82	85–95	1.79-2.14 MJ/kg CO_2	Wang et al. (2013a, 2013b)	Exp.
VPSA	13X	15	2 kPa	94–96	84–95	1.2-2.1 MJ/kg	Krishnamurthy et al. (2014)	Exp
Two-stage PSA	13X-13X	10.5	7 kPa	99	80	2.3-2.8 MJ/kg CO_2	Cho et al. (2004)	Exp.
Two-stage VPSA	AC-AC	15	5 kPa	96.40	80.42	0.83 MJ/kg CO_2	Shen et al. (2012)	Sim.
Two-stage VPSA	13XAPG-AC	16		95.6	90.2	2.4 MJ/kg CO_2	Wang et al. (2013a)	Exp.
Two-stage VPSA	CMS-CMS	15	4 kPa	90	90	0.96 MJ/kg CO_2	Haghpanah et al. (2013a, 2013b)	Sim
Two-stage VPSA	13X-13X	15	10 kPa	95.46	90.12	0.63 MJ/kg CO_2	Nikolaidis et al. (2017)	Sim
DR-PSA	Silica gel	15	20 kPa	90–99	90–97.8	1.67–2.86 MJ/kg CO_2	This study	Sim