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Optimisation of carbon dioxide recovery from flue gas in a TPSA system

Vinay Mulgundmath · F. Handan Tezel

Abstract Power stations and industrial processes burning fossil fuels account for the largest percentage of carbon dioxide emissions. Carbon capture and sequestration has received enormous global attention to reduce the carbon footprint and combat global warming. Adsorption has become an alternative technique to the conventional absorption process for capturing carbon dioxide due to its low operating and capital costs.

In this study, Pressure Swing Adsorption (PSA) process has been compared with Thermal Pressure Swing Adsorption (TPSA) process for CO_2 recovery from a flue gas composition of 10% CO_2 (by vol) in N_2 using Ceca 13X adsorbent. A factorial design set of experiments was performed to optimise the carbon dioxide recovery and study the effects and interaction of four control parameters namely, purge/feed flow ratio, purge time, purge gas temperature and adsorption column pressure.

Results indicated that better regeneration conditions used in a TPSA cycle was essential over a PSA cycle for regaining maximum adsorption capacity of the used Ceca 13X adsorbent. It was found that Purge time had a significant effect on the CO_2 recovery followed by Column pressure, purge/feed flow ratio and purge temperature. A Minitab® statistical software was used to analyse the data. It was found that the test of significance for lack of fit showed the fitted model to be an adequate representation of the experimental data. The results showed that to maximise the CO_2 recovery, highest values of the control parameters have to be used.

Keywords Carbon dioxide recovery from flue gas · Carbon capture · Carbon dioxide sequestration · Recycle of carbon dioxide · 13X zeolite · Pressure swing adsorption (PSA) · Temperature pressure swing adsorption (TPSA)

Symbols

A	Purge temperature control parameter
B	Purge time control parameter
C	Purge/Feed flow ratio control parameter
D	Column pressure control parameter
F	"*F*" distribution
k	Control parameter
L	Parameter (dimensionless)
LF	Lack of fit
MS	Mean of squares
PE	Pure error
Q	Flow rate of gas in a cycle step (L/min)
R	Carbon dioxide recovery function
X	Composition of carbon dioxide in a cycle step (vol %)
Time	Time required to complete a cycle step (min)

Abbreviations

CO_2	Carbon dioxide
GC	Gas Chromatograph
GHG	Greenhouse gas
min	Minute
N_2	Nitrogen
NDIR	Non Dispersive Infrared analyser
PSA	Pressure Swing Adsorption
SL	Standard Litre
TPSA	Thermal Pressure Swing Adsorption
TSA	Temperature Swing Adsorption
UHP	Ultra high pure
vol	Volume
VPSA	Vacuum Pressure Swing Adsorption

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Greek letters

α Level of significance (0–1)
β Coefficient of model fit equation (dimensionless)

1 Introduction

Accumulation of greenhouse gases (GHGs) in the atmosphere due to carbon dioxide (CO_2) emissions is a global environmental issue. The most important sources of CO_2 emissions are fossil fuel fired plants for electricity generation (Grande and Rodrigues 2008). The solution prevails in point source reduction and recovery in order to meet the present and future constraints on the allowable emissions of CO_2. Several technologies are being developed to capture CO_2 in post combustion, pre-combustion or oxyfuel combustion processes (IEA WPFF 2002). However, these novel technologies come with economic penalties which require retrofits for existing power plants.

The most common approach appears to be with liquid phase absorption. However, this process has been more widely adopted by large scale CO_2 emitters which are economically feasible leaving the small scale CO_2 emitters with little economic viable options. Recently, adsorption processes have become an alternative to the conventional absorption process for low to medium CO_2 emitters. Most of the research in adsorption is focussed on pressure/vacuum swing adsorption for separating CO_2 (15% by vol) from flue gas in power stations that use coal as fuel (Zhang et al. 2008; Reynolds et al. 2008, 2006, 2005; Franchi et al. 2005; Golden et al. 2003; Ko et al. 2003; Gomes and Yee 2002; Takamura et al. 2001a, 2001b; Zhang et al. 1998; Ishibashi et al. 1996; Chue et al. 1995; Ruthven et al. 1994; Kikkinides and Yang 1993). Kim et al. 1994 reported that PSA process was uneconomical for the removal of CO_2 in a low concentration range (5–15 vol%) from power stations that use oil or natural gas as fuels. For this CO_2 concentration range, higher economic penalties may be needed to attain lower adsorbent regeneration pressures as in a Vacuum Pressure Swing Adsorption (VPSA) system. Thermal Swing Adsorption (TSA) process too is energy intensive because of the need to supply heat to the regenerating gas. Thermal Pressure Swing Adsorption (TPSA) system could be a promising option if waste heat which is readily available in power stations could be used to regenerate the adsorbents to offset the large amount of regenerative purge gas needed and thus reduce the power consumption for vacuum pumps (Ishibashi et al. 1996). TPSA set up works very closely to TSA, by supplying external heat through inline gas heaters or heating tapes to the purge gas under a low pressure. However, the amount of external heat required will be less since the warmer gas from the column product end can be used as a purge gas to desorb all the impurities from the column bed. By suitable adjustment of the operating conditions, it is possible to utilise only a fraction of the heat of desorption being supplied to the purge gas (Wright et al. 2005). The addition of a temperature swing increases the regenerative capacity and reduces the power consumption for the vacuum pump. Another interesting alternative could be to compress the feed equivalent to the power consumed by the vacuum pump, perform adsorption at higher pressures and regenerate the adsorbent with waste heat and atmospheric pressure. Wright et al. (2005) report that a significant saving of thermal energy in the order of 20–60% could be realised without requiring a significant decrease in cycle time.

The most widely used adsorbents for CO_2 removal in flue gas applications are zeolite 13X and activated carbon (Harlick and Tezel 2004; Yon and Sherman 2003; Birbara et al. 2002; Kanazirev and Latus 2003). Kikkinides and Yang (1993) reported that CO_2 adsorption on zeolites is too strong, leading to a higher heat of adsorption which makes desorption difficult under VSA cycles. Chue et al. (1995) reported that zeolite 13X performed better than activated carbon in a PSA/VSA CO_2 capture process.

Previously, pure gas isotherms of CO_2 and N_2 on Ceca 13X, Alcan AA320-AP and Alcan 650PC composite adsorbent indicated that Ceca 13X was the preferred adsorbent for CO_2 removal (Mulgundmath et al. 2010a). Ceca 13X exhibited a higher capacity for CO_2 over N_2 than Alcan 650PC and Alcan AA320-AP. Therefore, in this study Ceca 13X zeolite was chosen as the suitable adsorbent for the lab scale TPSA system.

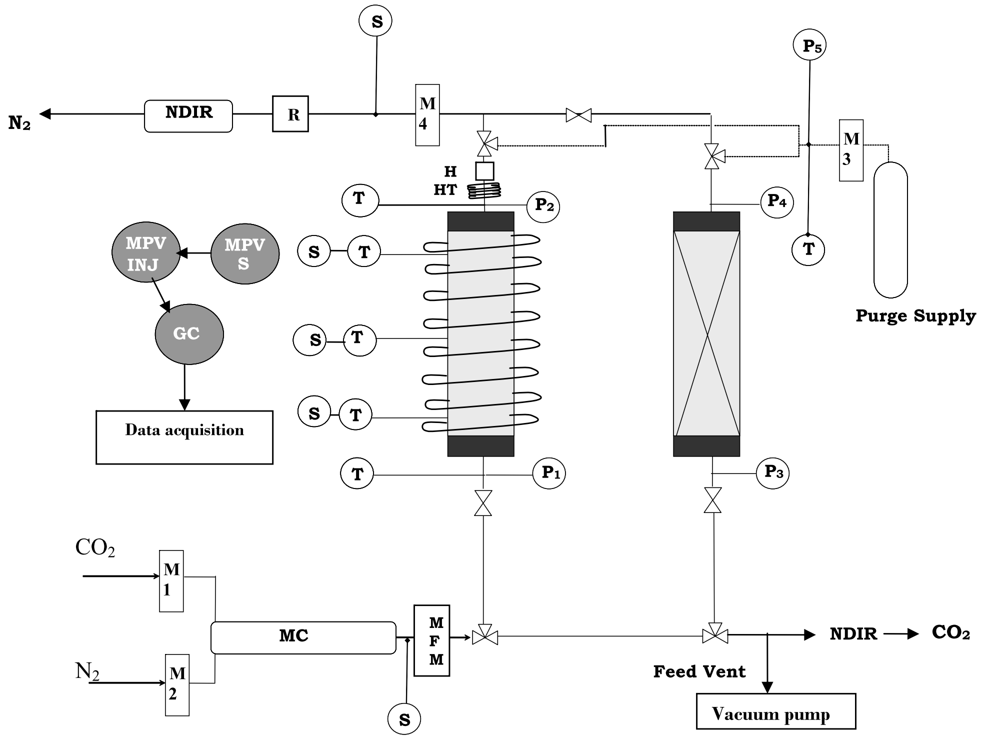
2 Experimental section

A schematic diagram of the Thermal Pressure Swing Adsorption (TPSA) system designed and built in-house is shown in Fig. 1 while the properties of the adsorbent and the fixed bed are given in Table 1.

For the experiments in this study, one column was used to analyse the breakthrough concentration and temperature profiles. Zeolite Ceca 13X supplied by CECA, from France was used to pack the adsorbent column (Length: 61 cm, Diameter: 4.4 cm). Uniform distribution of the Ceca 13X pellets inside the column was ensured by continuous tapping of the sides of the column. The adsorbent was regenerated in Fisher Scientific electric oven at 200 °C with UHP N_2

Table 1 Properties of the adsorbent and fixed bed

Bed length (m)	0.61
Column internal diameter (m)	0.044
Column wall thickness (m)	0.0029
Ceca 13X pellet radius (m)	0.00103
Ceca 13 X bulk density ($kg\,m^{-3}$)	705



GC—Gas Chromatograph; H—Heater; HT—Electrical heating tape; INJ—Injection valve; NDIR—Non Dispersive Infrared analyser; M—Mass flow controller; MC—Mixing chamber; MFM—Mass flow meter; MPV—Multiposition valve; P—Pressure transducer; R—Rotameter; S—Sample port; T—Thermocouple

Fig. 1 Schematic diagram of the two bed TPSA system

purge gas in order to remove any impurities present followed by cooling it overnight. The flue gas composition (10% CO_2 by vol. in N_2) was prepared in the gas mixing chamber using two pre-calibrated CO_2 and N_2 mass flow controllers (M1 and M2) which were connected to CO_2 and N_2 gas cylinders. A combined flow rate of 7 SL/min was prepared and used for all the experimental runs. Analysis of the breakthrough concentration and temperature profiles were obtained at three sample ports (at positions 10.2 cm, 30.5 cm and 50.7 cm from column inlet) at column centre axis. These sample ports were connected to a VALCO model SD 32-port micro-electric actuated multiposition valve. This multiposition valve had 32 ports (16 inlet and 16 outlet ports) and had the capability of connecting to 16 different sample ports. A 6-port micro-electric actuated injector valve housed the sample gas cell (500 μL) that connected the 32-port micro-electric actuated multiposition valve to a Varian 3400 Series Gas Chromatograph (GC). Temperature measurements were done using Omega type-K exposed tip thermocouples while CO_2 concentration measurements at the sample ports were obtained by using a Varian 3400 Series Gas Chromatograph (GC). A Non Dispersive Infrared Analyser (NDIR) was also used for continuous CO_2 gas detection at the adsorber column exit. A data acquisition system in conjunction with LABVIEW software obtained from National Instruments Inc. was used to monitor and record the pressure, temperature, CO_2 concentration and flow rate data continuously in an MS-Excel worksheet. The detailed experimental procedure has been explained in the earlier paper (Mulgundmath et al. 2010b).

The cycle steps used in the Thermal Pressure Swing Adsorption (TPSA) process were: pressurisation, adsorption at a constant high pressure, depressurisation and heated purge gas flow under low pressure as shown in Fig. 2. Before the beginning of each experimental run, the regenerated column was pressurised to the desired adsorption pressure using UHP N_2. When the desired pressure was attained, the flue gas composition (10% CO_2 by vol. in N_2) was prepared in the gas mixing chamber and introduced at the column entrance. Temperature and CO_2 concentrations were analysed as a function of time at three different ports along the length of the column.

When the gas concentration at the column exit reached the breakthrough CO_2 concentration in air (400 ppm—Kumar et al. 2003), the feed flow was stopped. At that point, the column was switched from the adsorption mode to counter current depressurisation mode to recover pure CO_2. The gas in the column was allowed to bleed at constant flowrate to atmospheric pressure slowly through a calibrated Rotameter (R) while the exit gas concentration was measured by a Non Dispersive Infrared (NDIR) analyser. These measured flow rate and concentration data were used in (1) to

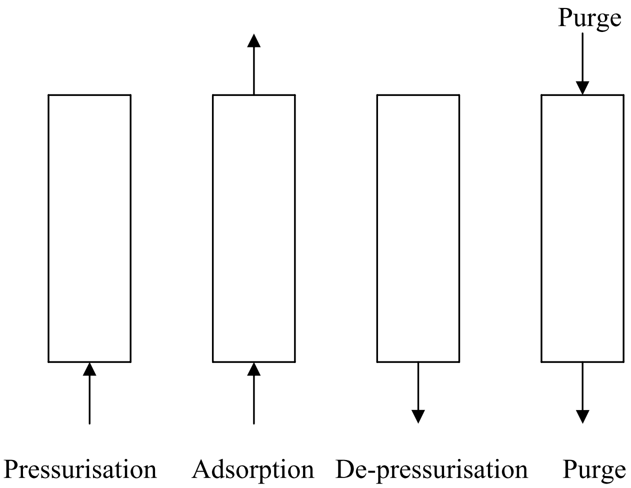


Fig. 2 Schematic diagram of the step cycles in the TPSA system

Table 2 The 2^4 factorial design for the TPSA experiment

Run	Purge/Feed ratio	Purge time	Purge gas temperature	Column pressure
1	−1	−1	−1	−1
2	1	−1	−1	−1
3	−1	1	−1	−1
4	1	1	−1	−1
5	−1	−1	1	−1
6	1	−1	1	−1
7	−1	1	1	−1
8	1	1	1	−1
9	−1	−1	−1	1
10	1	−1	−1	1
11	−1	1	−1	1
12	1	1	−1	1
13	−1	−1	1	1
14	1	−1	1	1
15	−1	1	1	1
16	1	1	1	1

calculate the CO_2 recovery. When the flow neared zero, the cycle was switched to counter-current purge mode. A constant purge pressure of 2 atm (absolute) was maintained to obtain the GC readings. Regeneration was carried out by a heated purge gas inside the insulated column. An inline gas heater, a thermostat controlled heating tape at the purge entrance of the column contributed to attaining higher regeneration temperatures (80–105 °C) within a short interval of time. When the desired cycle time was reached, the flow of heated purge gas was stopped. The recorded Thermal Pressure Swing Adsorption (TPSA) cycle data of flow rate, column pressure, temperatures and CO_2 concentration data were used to calculate the amount of carbon dioxide recovered during this cycle.

Complete regeneration of the adsorbent between different experimental runs was achieved on-site by a combination of high temperature (by using a thermostat controlled 50:50 (vol/vol) ethylene glycol water mixture at 95 °C circulating inside copper coils around the column and a heated purge gas to achieve a temperature of 120 °C in the column) and high vacuum (using Edwards dual stage high vacuum pump). During the regeneration cycle, UHP N_2 gas flowed through the column. The completion of this step was checked by stopping the vacuum and analysing the desorbed gas from the column for CO_2 concentration using the NDIR analyser. Complete regeneration of the adsorbent was achieved when the desorbed gas contained no traces of CO_2. The column was then allowed to cool under UHP N_2 purge followed by isolating it from the rest of the setup by closing the column inlet and the outlet valves. Prior to the next experimental run, the column was re-pressurised to the desired pressure and the TPSA cycle steps were repeated.

For each experimental run, the recorded data was analysed to determine the amount of CO_2 recovered in a TPSA cycle, which is given by (1):

$$\mathrm{CO_2RECOVERY} = \frac{(Q_{\mathrm{depress}} \times X_{\mathrm{CO_2\,depress}} \times \mathrm{Time}_{\mathrm{depress}})}{(Q_{\mathrm{ads}} \times X_{\mathrm{CO_2\,ads}} \times \mathrm{Time}_{\mathrm{ads}})} + \frac{(Q_{\mathrm{purge}} \times X_{\mathrm{CO_2\,purge}} \times \mathrm{Time}_{\mathrm{purge}})}{(Q_{\mathrm{ads}} \times X_{\mathrm{CO_2\,ads}} \times \mathrm{Time}_{\mathrm{ads}})} \tag{1}$$

where Q is the flow rate of gas in a cycle step (L/min) and assumed to be constant, $X_{\mathrm{CO_2\,ads}}$ is the composition of CO2 during the adsorption step (vol %) which is equal to the feed concentration. $X_{\mathrm{CO_2\,depress}}$ and $X_{\mathrm{CO_2\,purge}}$ are the time averaged compositions of CO_2 during the depressurisation and purge steps (vol %), respectively. Time is the time required to complete a cycle step (min) whereas the subscripts depress, purge and ads refer to depressurisation step, purge step and adsorption step, respectively.

3 Factorial design

A 2^k factorial design experiment was conducted to optimise the recovery for carbon dioxide from flue gas composition (10% CO_2 by vol. in N_2) in a TPSA cycle. The four control parameters as operating variables chosen for this study were purge/feed flow ratio, purge time, purge gas temperature and adsorption column pressure. A two level factorial (high and low) were considered for these control parameters in the experiments. Table 2 shows all the sixteen experimental TPSA cycle runs that correspond to the chosen control parameters.

Table 3 Range of the control parameters for the 2-level factorial design

	Purge/Feed ratio	Purge time (h)	Purge gas temperature (°C)	Column pressure (atm)
Low level	0.75	1	80	5.08
Centre point	0.875	1.25	92.5	5.76
High level	1	1.5	105	6.44

This factorial design has four main effect interactions, six two-factor interactions, four three-factor interactions and one four-factor interaction. The CO_2 recovery determined by these runs was analysed for process optimisation. Table 3 details the two level factorial (high and low) range used for the control parameters.

Along with the above sixteen TPSA cycle experimental runs, four random TPSA cycle runs corresponding to centre point replicates (purge/feed ratio = 0.875, purge time = 1.25 h, purge gas temperature = 92.5 °C and column pressure = 5.76 atm) were performed to fit the data for statistical analysis.

4 Results and discussions

4.1 PSA cycle

Initial concentration breakthrough experiments at 7 SL/min feed flow rate were performed for CO_2 (10% by vol.) and N_2 (90% by vol) feed gas mixture to see if high temperature was necessary for complete regeneration of the adsorbent. This Pressure Swing Adsorption (PSA) cycle was followed without the addition of heat during the purge and it consisted of pressurising the column with UHP N_2 and carrying out the adsorption by introducing the feed mixture at desired column pressure (6.44 atm). When the breakthrough CO_2 concentration of 400 ppm was reached, the column was switched to counter current depressurisation mode in order to recover pure CO_2. The quantity and concentration of CO_2 recovered in the depressurisation mode was determined using a Rotameter at the column exit and a Non Dispersive Infrared (NDIR) analyser, respectively, between the operating pressures of 6.44 atm to 1 atm (absolute). When the flow rate of the depressurised gas neared zero, the column was re-pressurised to the adsorption pressure using UHP N_2 and the cycle was repeated. However, the adsorbent capacity recovered during this cycle amounted to only 15% of the total capacity of Ceca 13X adsorbent. This indicated that lowering the pressure alone during regeneration was not sufficient to completely regenerate the Ceca 13X adsorbent. This observation suggested that a combination of high temperature and lower pressure during the regeneration could be a viable option to recover maximum capacity of the adsorbent. Hence, a TPSA cycle which also exhibits a larger working capacity when compared to a PSA cycle alone was chosen to optimise the CO_2 recovery in this study.

Table 4 CO_2 recovery for the TPSA factorial design experiments (highlighted numbers indicate center point replicates)

Purge/Feed ratio	Purge time (h)	Purge gas temperature (°C)	Column pressure (atm)	% CO_2 recovery
0.75	1.5	105	5.08	56.66
0.75	1.5	80	5.08	55.42
1	1.5	105	5.08	57.27
0.75	1.5	80	6.44	55.83
1	1.5	80	6.44	59.79
0.75	1.5	105	6.44	57.05
1	1.5	105	6.44	60.04
0.75	1	80	5.08	47.47
1	1	80	5.08	52.78
1	1.5	80	5.08	55.55
1	1	105	5.08	52.92
0.75	1	80	6.44	49.72
1	1	80	6.44	57.89
0.875	**1.25**	**92.5**	**5.76**	**55.18**
0.75	1	105	6.44	56.44
0.875	**1.25**	**92.5**	**5.76**	**55.45**
1	1	105	6.44	58.84
0.875	**1.25**	**92.5**	**5.76**	**55.95**
0.75	1	105	5.08	51.4
0.875	**1.25**	**92.5**	**5.76**	**55.81**

4.2 TPSA cycle

TPSA cycle experiments were performed according to the procedure described in the experimental section and CO_2 recovery was determined for each 16 TPSA cycle run shown in Table 2, plus the 4 random runs corresponding to center point replicates. A feed flow rate of 7 SL/min (10% CO_2 by vol in N_2) was maintained for all the experimental runs. Earlier attempts have been made to operate commercial TPSA cycles to maximise CO_2 recovery in a flue gas application (Ishibashi et al. 1996). However, the disadvantage lies in its long cycle time when compared to PSA and the CO_2 recovery varies until the process reaches cyclic steady state. Table 4 shows results obtained for % CO_2 recovery for each run along with the values of the control parameters while Figs. 3 to 6 explain the effect of control parameters on the CO_2 recovery in the form of 3D-Surface plots.

It was observed that CO_2 recovery ranged from 47.47% to 60.04%, for the chosen range of control parameters.

Fig. 3 Effect of purge time and purge/feed on the CO_2 recovery

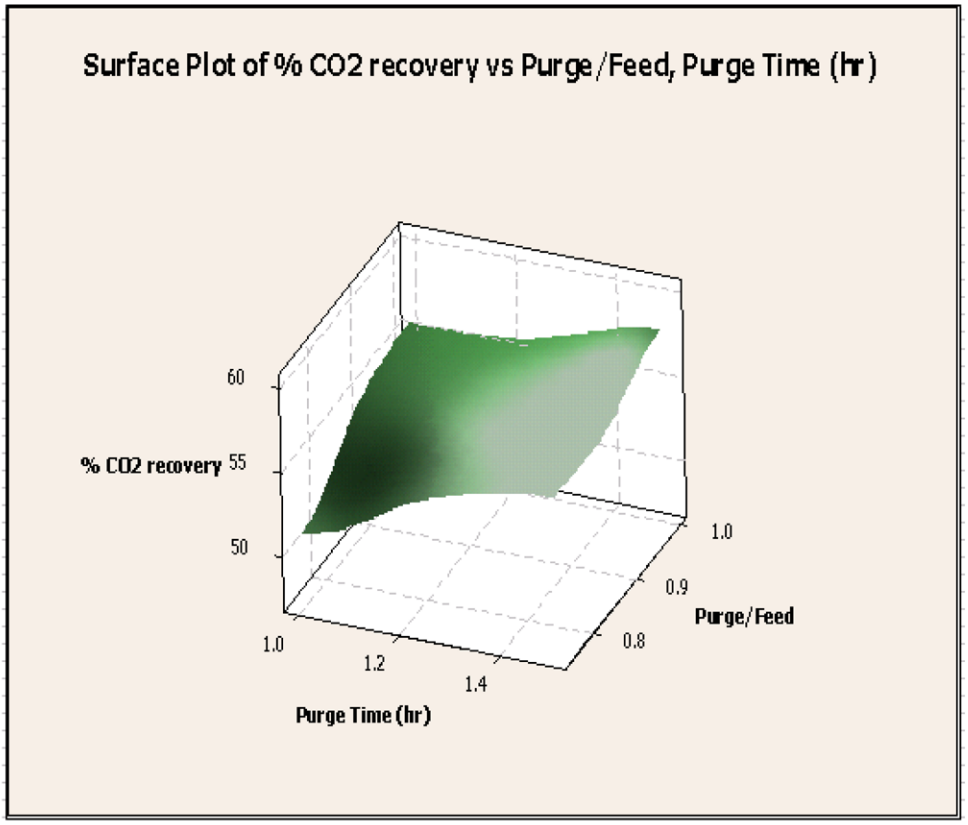
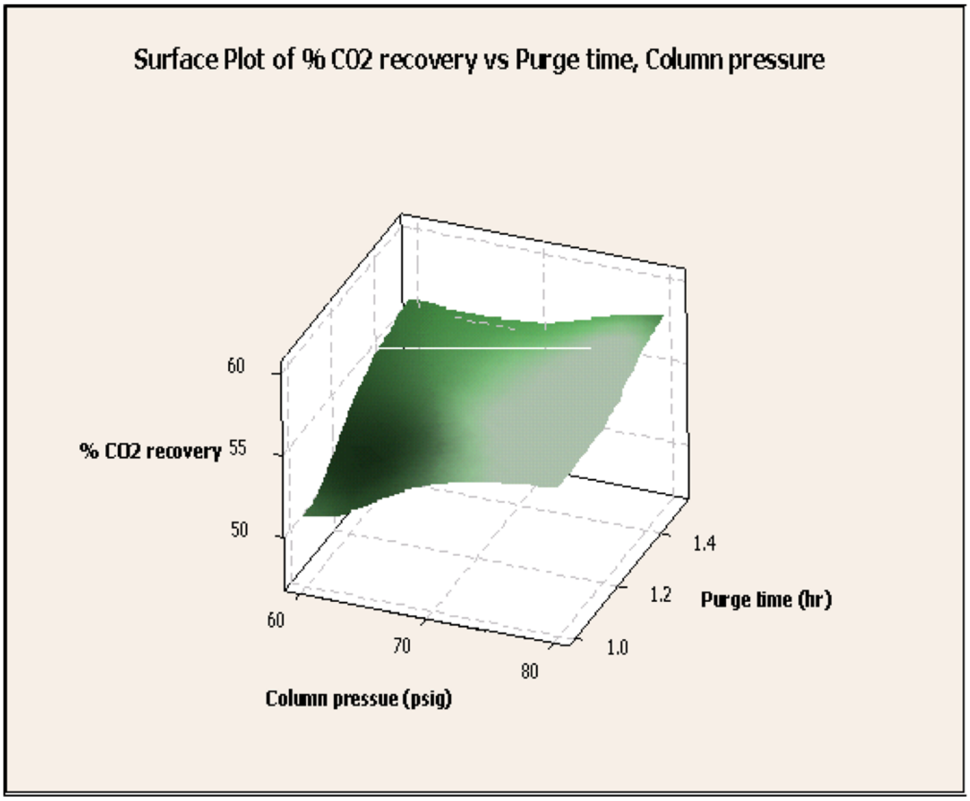


Fig. 4 Effect of purge time and column pressure on the CO_2 recovery



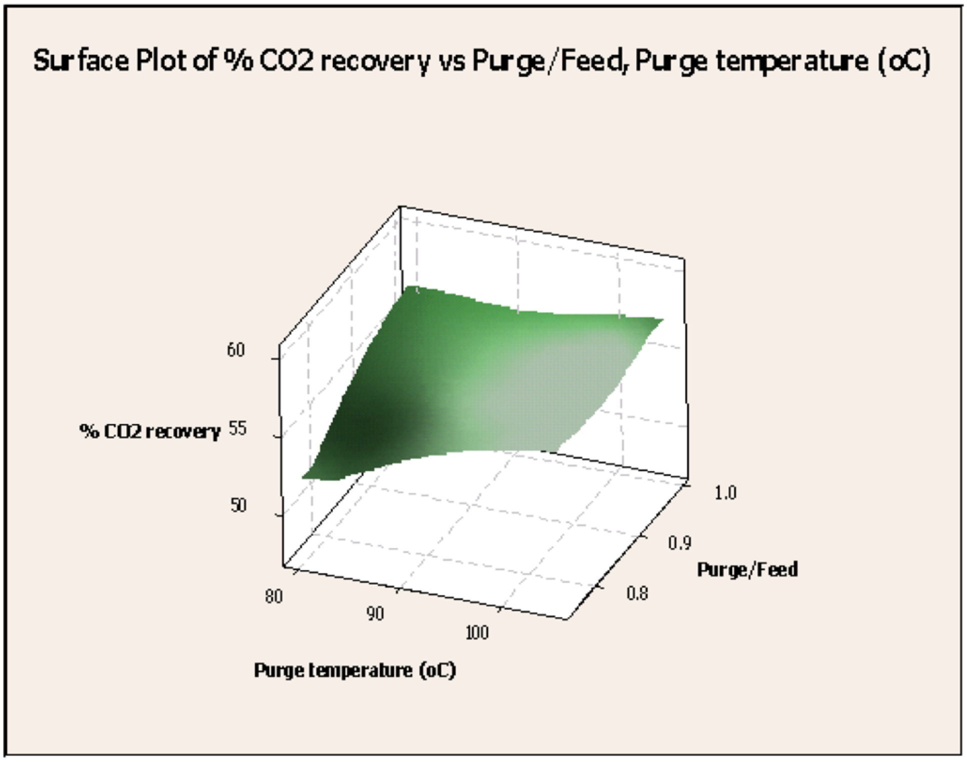


Fig. 5 Effect of purge temperature and purge/feed on the CO_2 recovery

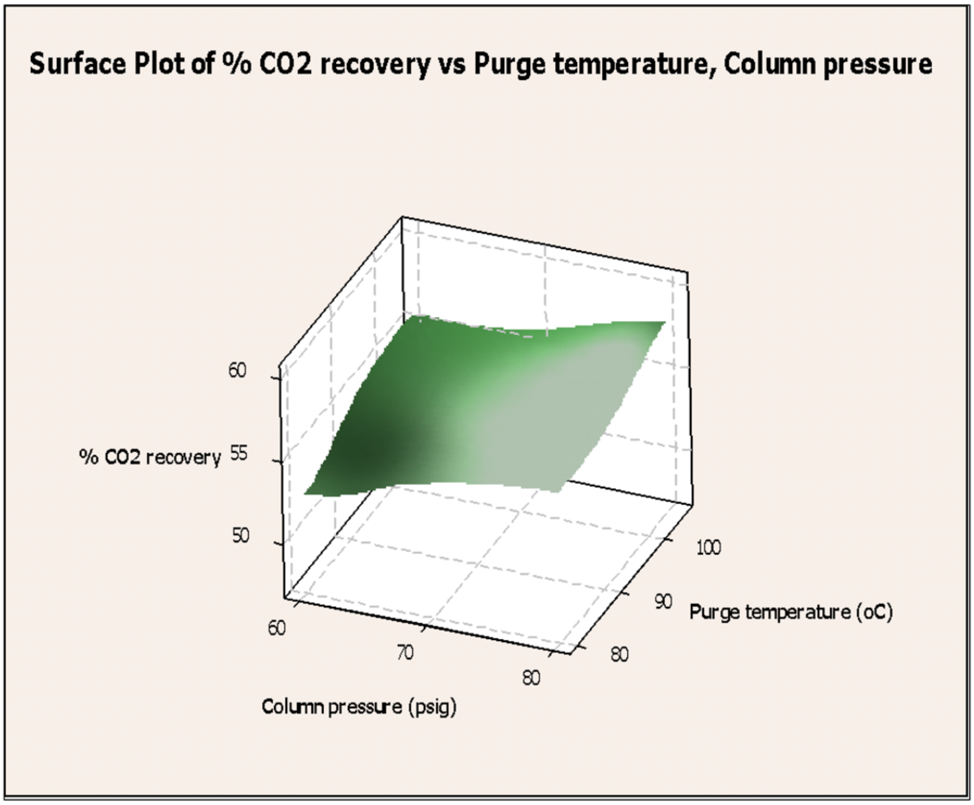


Fig. 6 Effect of purge temperature and column pressure on the CO_2 recovery

The lowest recovery (47.47%) was obtained for the lowest range of control parameters (purge/feed = 0.75, purge time = 1 h, purge temperature = 80 °C and column pressure = 5.08 atm) while the highest recovery (60.04%) was

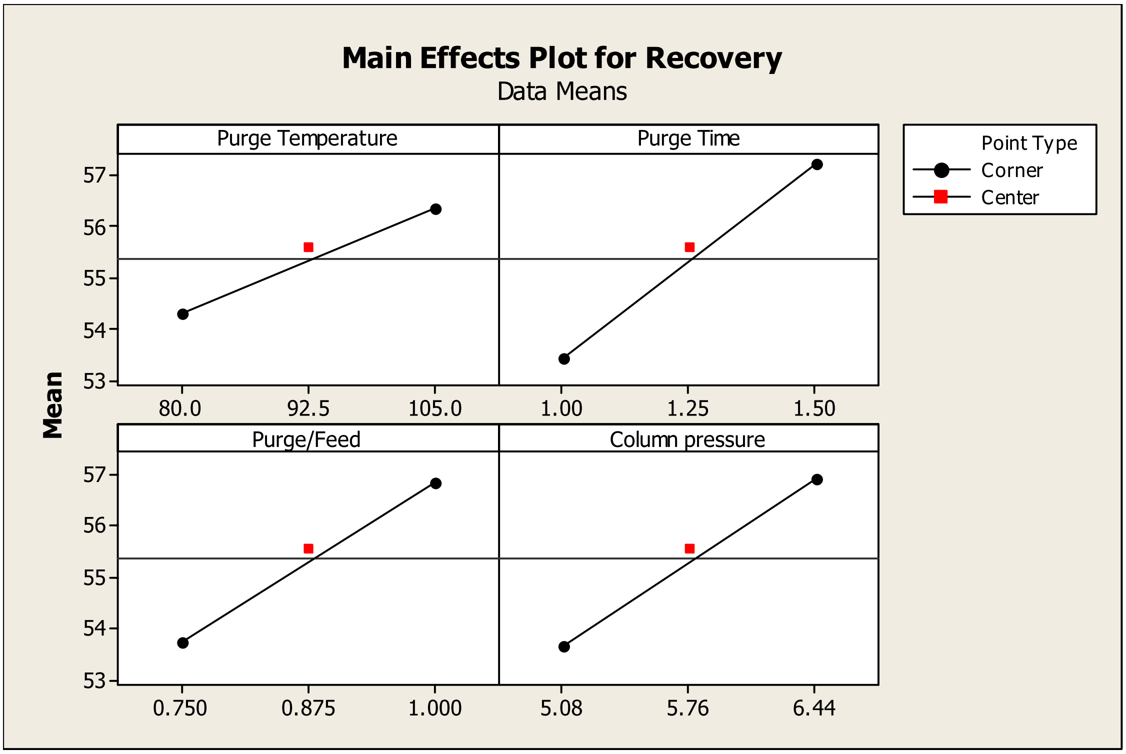


Fig. 7 Main effects plot for CO_2 recovery

obtained for the highest range of the control parameters (purge/feed = 1, purge time = 1.5 h, purge temperature = 105 °C and column pressure = 6.44 atm). The control parameters at the highest range contributed to better regeneration conditions and higher capacity, and therefore more CO_2 recovery. Higher CO_2 recoveries were observed at higher adsorption column pressures. A 100% CO_2 recovery is achievable by using higher values for control parameters for the TPSA cycle but the CO_2 concentrations obtained could be very low. This observation indicates that CO_2 concentrations along with CO_2 recovery are important parameters to measure the TPSA cycle performance. The remaining factorial design experimental runs were performed to study the effect of interactions between the control parameters. The four random center-point replicate runs indicated a similar CO_2 recovery (55.18% to 55.95%) which was within acceptable experimental error.

4.3 Optimisation

Minitab® statistical software was used to analyse the factorial design experimental data. This software provides tools to build a factorial design set of experiments, interpret and analyse the experimental data. Based on the experimental results, this software has the ability to analyse and estimate the main effects, as well as the interactions between parameters affecting the CO_2 recovery. From these estimated effects, the software determines the parameter coefficients as well as the analysis of variance for CO_2 recovery. These results help to build a model that is representative of the experimental results. Figure 7 (Main Effects Plot for Recovery) shows the statistically significant contributors “main effects” to the response mean. These plots compare the relative magnitude of the main effect parameters. Analysis shows that all four factors (purge time, column pressure, purge/feed flow ratio and purge temperature) have a significant effect on the “control” (CO_2 recovery in this case) since the lines are not parallel to the reference line (which represents the overall mean). The plots indicate that the CO_2 recovery increases when these control parameters move from the low level to the high level. “Purge time” control factor has the highest magnitude of the main effect (CO_2 recovery), since it had the highest slope, followed by column pressure, purge/feed flow ratio and purge temperature.

One common objective in a process investigation is the development of an adequate mathematical model for the behaviour of a process within a defined operating region. Another common objective is the improvement of process performance, perhaps leading to the location of optimum operating conditions. In this study, four control factors (purge/feed ratio, purge time, purge gas temperature and adsorption column pressure) were selected to study their effects on the amount of CO_2 recovery in the defined operating region.

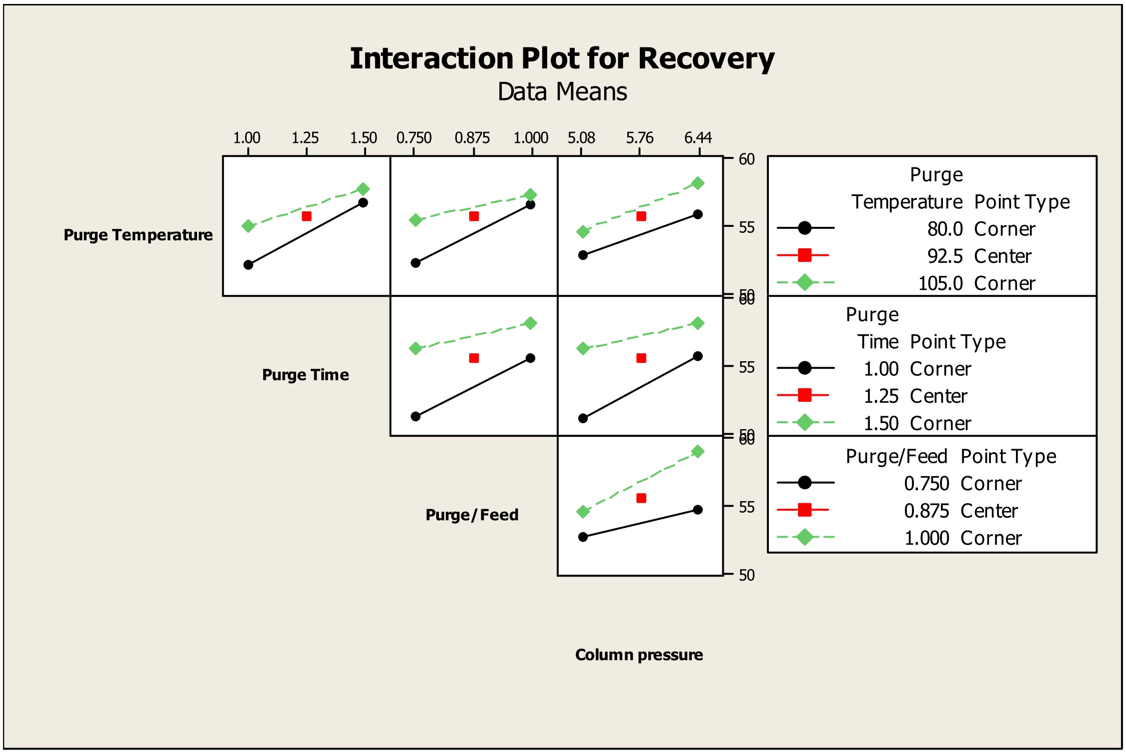


Fig. 8 Interactions plot for CO_2 recovery

Figure 8 represents the interaction plots for CO_2 recovery based on the effects of each control factor studied. Perfect parallel lines in these plots indicate no interaction between parameters considered. A greater departure from parallel lines indicates a higher degree of interaction between the control factors. From the plot in Fig. 8, it was observed that purge temperature and column pressure plots were close to parallel indicating a smaller degree of interaction between these control parameters. However when other control parameters are compared (Purge temperature vs. Purge time, Purge temperature vs. purge/feed, Purge time vs. purge/feed, Purge time vs. column pressure and Purge/feed vs. column pressure) the plots are unparallel indicating a higher degree of interaction between them.

Figure 9 gives the normal plot of the standardised effects. In this plot, points that are further from the line indicate important effects. It can be seen from this figure that the most significant factor effects in descending order are the purge time (B), column pressure (D), purge/feed ratio (C), and purge temperature (A) for a maximum acceptable level of risk for rejecting a true null hypothesis, $\alpha = 0.05$ or 95% confidence interval. The insignificant effects tend to be smaller and are centred on zero. In addition, there are two-factor and three-factor interaction effects that are also significant to note. Of most significance is the two-factor purge/feed-column pressure (denoted as CD) followed by the three-factor purge temperature-purge time-purge/feed (denoted as ABC).

Figure 10 shows the Pareto chart of the standardised control factor effects. These effects determine the magnitude and importance of an effect. The plot displays the absolute value of the effects and draws a reference line on the chart which represents the “T” distribution value (for 95% confidence interval). All control factors that extend beyond the reference line are considered to be potentially important. From Fig. 10, the most significant factors contributing to the increase in CO_2 recovery in descending order are the purge time, column pressure, purge/feed ratio, and purge temperature. These observed results are consistent with results obtained from Fig. 9.

4.4 Testing the adequacy of a fitted model

Based on the Factorial fit, the model was fitted to the data from the 20 runs of this central composite design using the same coded values of the four control factors (denoted by the variables A, B, C and D) as denoted in the general equation (2)

$$\begin{aligned} R = {} & \beta_o + \beta_A A + \beta_B B + \beta_C C + \beta_D D + \beta_{AB} AB \\ & + \beta_{AC} AC + \beta_{AD} AD + \beta_{BC} BC \\ & + \beta_{BD} BD + \beta_{CD} CD + \beta_{ABC} ABC + \beta_{BCD} BCD \\ & + \beta_{ACD} ACD + \beta_{ABD} ABD + \beta_{ABCD} ABCD \end{aligned} \quad (2)$$

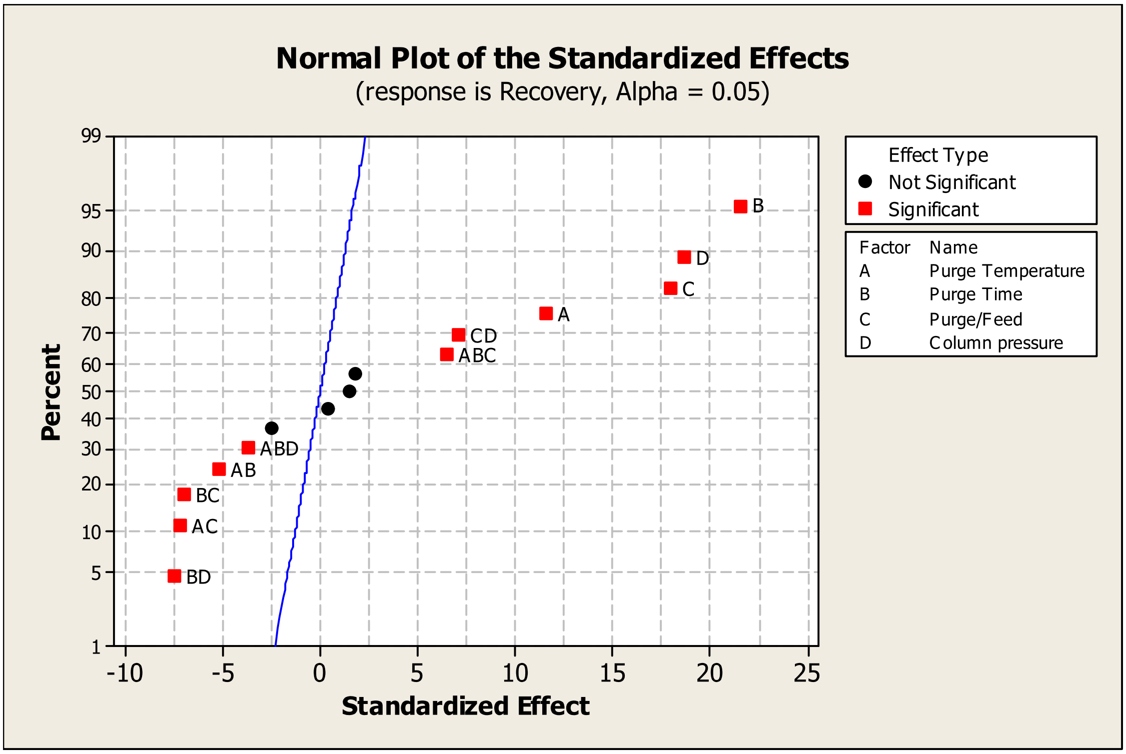


Fig. 9 Normal plot of the standardised effects

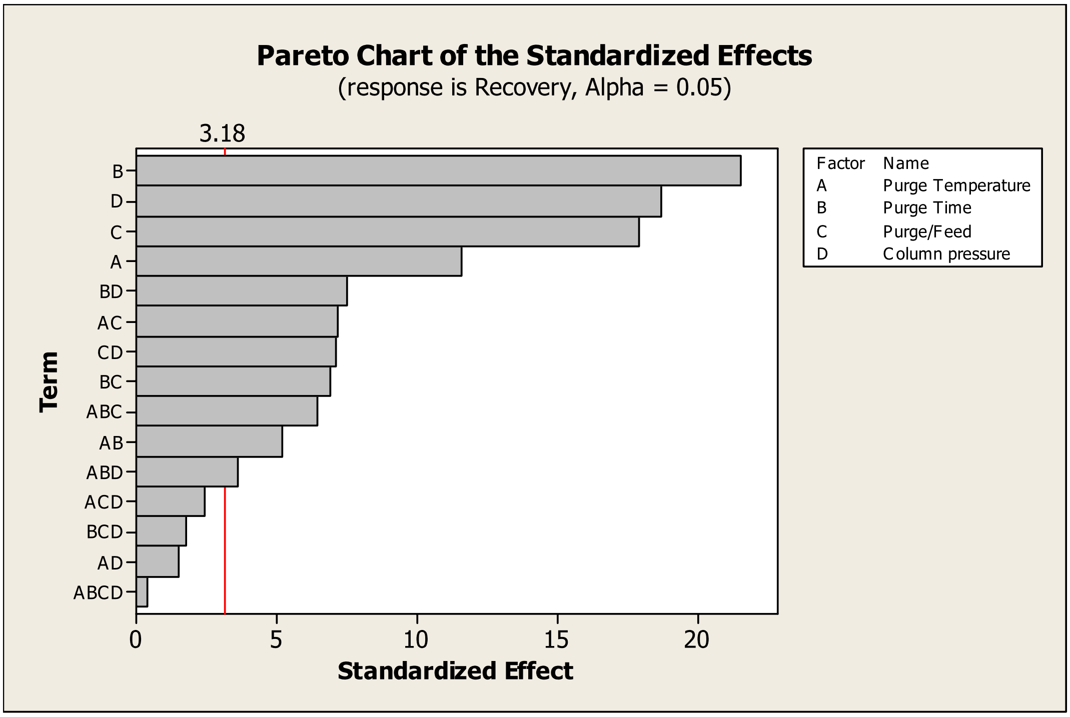


Fig. 10 Pareto chart of the standardised effects

where A, B, C and D represent purge temperature, purge time, purge/feed flow ratio and column pressure control parameters, respectively. Based on the estimated parameter coefficients by Minitab® software for a 95% confidence interval, the following fitted model was obtained as denoted by (3):

$$\begin{aligned} R = {} & 55.3169 + 1.0106A + 1.8844B + 1.5681C \\ & + 1.6331D - 0.4569AB - 0.6281AC - 0.6069BC \\ & - 0.6569BD + 0.6219CD + 0.5669ABC \\ & - 0.3181ABD \end{aligned} \tag{3}$$

Coefficients from (3) indicate that main effects contribute to a higher CO_2 recovery when compared to 2-way and 3-way interaction parameters, with higher coefficients in this equation. Purge time (B) had a significant effect on the CO_2 recovery followed by column pressure (D), purge/feed flow ratio (C) and purge temperature (A) control parameters, respectively, with coefficients decreasing in this order. Analysis of variance for recovery (P-values $< \alpha = 0.05$ or 95% confidence interval for main effects, 2-way interactions and 3-way interactions) showed this model to be an adequate representation of this data set. A test of significance of lack of fit can be determined by comparing the values obtained for:

$$L = \frac{MS_{LF}}{MS_{PE}} \quad \text{to } F_{\nu_{LF}, \nu_{PE}, \alpha} \tag{4}$$

where MS refers to the mean of squares and subscripts LF, and PE refer to lack of fit error (or residual error) and pure error, respectively while ν_{LF} and ν_{PE} refer to the degree of freedom with respect to lack of fit error (LF) and pure error (PE), respectively. So, if $L > F_{\nu_{LF}, \nu_{PE}, \alpha}$ there is no bias due to model inadequacy and the null hypothesis could be rejected. This condition concludes that there is a significant lack of fit in the fitted model. Since $L = \frac{MS_{LF}}{MS_{PE}} = \frac{0.33}{0.33} = 1$ which is less than $F_{3,3,0.05} = 9.28$, there is no significant lack of fit in the fitted model and the null hypothesis is not rejected, thus signifying that the model is a good fit. Therefore, by using such a model, one can deduce the main interaction effects to be concentrated upon and minimise the usage of less prominent interaction effects. The next step would be to increase the range of the main interaction effects in order to achieve a higher CO_2 recovery.

5 Conclusions

For the recovery of CO_2 from flue gas composition (10% CO_2 by vol. in N_2) by Ceca 13X adsorbent, it was found that better regeneration conditions used in a TPSA cycle was essential over a PSA cycle. The stronger interaction between CO_2 and Ceca 13X leading to a high heat of adsorption was the reason to substantiate the choice of the TPSA cycle. A 2^k factorial design set of experiments was conducted to optimise the recovery of carbon dioxide with respect to the effects of four control parameters (purge/feed ratio, purge time, purge gas temperature and adsorption column pressure) within a desired range in order to understand the interaction effects. It was found that each control parameter had a significant effect. Purge time had the most significant effect, followed by column pressure, purge/feed flow ratio and purge temperature on the CO_2 recovery, followed by 2-way and 3-way interactions. A Minitab®statistical software was used to analyse the data. It was found that the test of significance for lack of fit showed the fitted model to be an adequate representation of the experimental data. The results showed that to maximise the CO_2 recovery, highest values of the control parameters have to be used.

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