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Separation of CO_2/H_2 Gas Mixture Using Novel 4-Bed and 3-Bed Pressure Swing Adsorption Process

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Abstract: *The CO_2 and H_2 gas mixture is obtained from various sources such as refinery off gas, and during gasification of petroleum coke and coal. Efficient separation of CO_2 is required to use this gas mixture as fuel for energy production. Various methods are available to remove CO_2 from the mixture for example cryogenic distillation, membrane separation, absorption and adsorptive separation. Pressure Swing Adsorption(PSA), an adsorptive separation method, is one of the promising methods for CO_2 removal due to variety of adsorbent material available and various combinations of PSA cycles possible to fit the process requirement. Most of the PSA cycles proposed in the literature are the variants of the Skarstrom cycles involving four basic steps namely, pressurization, adsorption, blowdown and purge. Based on the nature of adsorption equilibria the adsorptive separation of binary gas mixture has been classified as distillation-like and absorption-like processes. In the distillation like process: both components show competitive adsorption in a binary gas mixture and the separation factor is moderate, whereas in the absorption like process one component preferentially adsorbs over the other and the separation factor is very high (∞). Separation of CO_2/H_2 using activated carbon is an example of absorption-like process. In this work simulation studies have been carried out to compare the performance of a 4-bed PSA cycle having steps: stripping, enriching, blowdown and pressurization with a 3-bed PSA cycle having steps: stripping, depressurization, blowdown and pressurization, for the separation of a binary mixture of CO_2/H_2, representing a typical refinery off gas (after treatment). The salient result shows that a moderately high productivity can be achieved with 3-bed PSA compared to the 4-bed PSA for a target purity of 97 mol% of H_2, but, the former requires a deep vacuum to facilitate this high productivity using activated carbon as a solid adsorbent.*

Keywords: *PSA, Carbon dioxide (CO_2) capture, isothermal simulation*

1. INTRODUCTION

The effluent gas from steam reforming contains large fraction of H_2 and CO_2 which leaves the reformer at high pressure (above 30 bar) and high temperature. Removal of CO_2 is essential before using this gas as a source of energy and also to reduce the potential environmental damage due to release of CO_2 (IPCC 2007).

CO_2/H_2 gas mixture can be separated by various methods such as Absorption, cryogenic distillation and adsorptive separation processes. Pressure swing adsorption is a promising adsorptive separation based technology used commercially for H_2 production in the refineries. Various possible PSA cycles and the availability of wide range of adsorbent materials have made PSA a preferable choice for H_2 production. Most of the PSA cycles are the variants of Skarstrom PSA cycle having stripping, blowdown, purge and pressurization steps. Other steps such as pressure equalization are added to improve the energy efficiency of the cycles. Industrial PSA

units for H_2 production use a multitude of adsorber beds. The number of adsorber could be as large as 16 beds which make the unit bulky. A significant reduction in the size of these units are desirable which can be achieved (i) by modifying the PSA cycle (ii) by using better adsorbent media.

In this work we have selected a 4-bed and a 3-bed PSA cycle for the comparison of performance and to explore the possibility of process intensification. Activated carbon has been selected as solid adsorbent for the study. These cycles are similar to those proposed by the Sircar and co-workers but for the implementation of blowdown during extract collection, we have used counter-current blowdown whereas they have used co-current blowdown [1]. It has been seen that if the required purity is less than 99.99 mol% of both products the direction of blowdown makes significant difference in throughput and in this case the counter current blowdown is preferable [2,3]. Yang and co-workers performed experimental and theoretical studies in a single column PSA and using 5step process with 5A zeolite to recover 67.5% H_2 of purity 99.9 mol% from a binary mixture containing 70mol% H_2 and the rest CO_2 [4]. Malek and Farooq simulated the performance of a six bed dual sorbent (activated carbon and silica gel) for H_2 purification using plant data with non-isothermal and isothermal conditions. They showed that though the isothermal predicted slightly higher recovery and purity of H_2 compared to nonisothermal simulation, the deviation was not more 3% between the two results [5]. Park and co-workers analysed the effect of activated carbon to zeolite ratio on H_2 recovery and productivity for a given purity in a layered 4-bed PSA process [6]. Waldron and Sircar analysed the separation of methane – hydrogen gas mixture using the adiabatic PSA model [7]. Jiang and co-workers investigated the performance of a PSA system of 5 bed 11 step system for the separation of CO_2-H_2 mixture [8]. This survey indicates that process intensification of PSA has been not given proper attention for the production of hydrogen from the refinery off gas. Present work addresses this issue.

2. 4-BED AND 3-BED PSA PROCESS

Figure 1 and **Figure 2** show the cycle sequence of 4-bed and 3-bed respectively. The processes are briefly described here; detailed process description is presented in our previous work [2].

In 4-bed PSA each bed undergoes stripping, enriching, blowdown and pressurization while in 3-bed PSA only stripping, blowdown and pressurization takes place as shown in the figure.

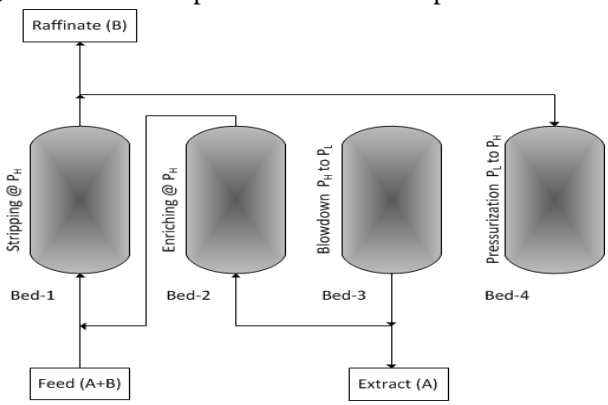

Figure 1. *Schematic diagram of 4-Bed PSA*

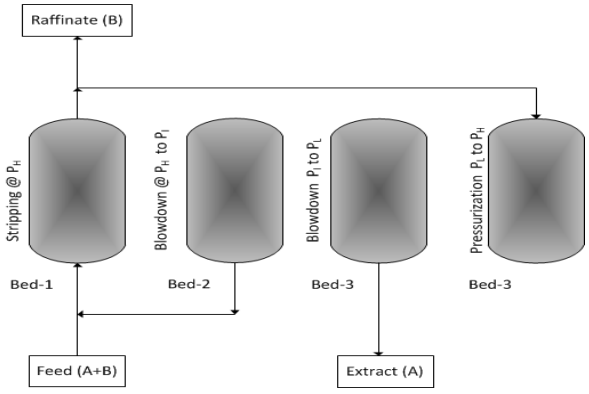

Figure 2. *Schematic diagram of 3-Bed PSA*

3. MODELING AND SIMULATION

The mathematical model described by R.S.Thakur and co-workers has been adapted here [9]. A brief outline of the model is given here. In the modelling following assumptions are made: (a) the gas obeys the ideal-gas law, (b) the process is isothermal, (c) the gas mixing in the bed is represented by a dispersed-plug flow model, (d) the interphase mass transfer is represented by the linear driving force (LDF) model and (e) the adsorption equilibrium follows the extended Langmuir isotherm model.

The process model for a bed undergoing any of the steps mentioned above is given by

Total Pressure

$$\frac{\partial P}{\partial t} = KP\frac{\partial^2 P}{\partial z^2} + K\left(\frac{\partial P}{\partial t}\right)^2 - \frac{(1-\varepsilon_B)}{\varepsilon_B}RT\sum_{i=1}^{N}\frac{\partial q_i}{\partial t} \quad (1)$$

Mass balance for gas phase

$$\frac{\partial x_i}{\partial t} = \frac{D_{L0}}{P}\frac{\partial^2 x_i}{\partial z^2} + K\left(\frac{\partial P}{\partial t}\right)\left(\frac{\partial x_i}{\partial t}\right) - \frac{(1-\varepsilon_B)}{\varepsilon_B}\frac{RT}{P}\left(x_i\sum_{i=1}^{N}\frac{\partial q_i}{\partial t} - \frac{\partial q_i}{\partial t}\right) \quad (2)$$

Where $K = -\frac{{d_p}^2{\varepsilon_B}^2}{150\mu(1-\varepsilon_B)^2}$ and i=1, N-1 (3)

and, the mass balance for the solid phase is given by:

$$\frac{\partial q}{\partial t} = ({q_i}^e - q_i) \qquad i= 1, 2 \quad (4)$$

Eq. 1-4 is solved with appropriate boundary conditions. The method of simulation is given elsewhere [3]

The parameters used in the simulation are presented in **Table 1**. In all cases the extract and raffinate product rates were set as from the overall mass balance equations. The recovery rates of the components can be calculated from the overall component mass balances

Table 1. *Parameters used in simulation*

Molar feed composition	20% CO_2 80% H_2
Particle diameter (d_p), *mm*	3
Adsorbent	Activated carbon
Bed length (L), *m*	1
Bed diameter, *mm*	25
Langmuir constant for CO_2 (b), m^3/mol	0.932E-02
Langmuir constant for H_2 (b), m^3/mol	0.217E-03
Bed voidage (ε_B)	0.74
Saturation constant for CO_2 (q_s), mol/m^3	6776.00
Saturation constant for H_2 (q_s), mol/m^3	3637.42
LDF constant for CO_2 (k_{CO2}), s^{-1}	0.1
LDF constant for H_2 (k_{H2}), s^{-1}	1.0
Operating temperature (T), *K*	298
Bulk density , kg/m^3	850

4. RESULTS AND DISCUSSION

The purity, recovery, energy requirements and productivity are the measures of the performance of a PSA process and are crucially affected by several process parameters such as feed rate, operating pressures and step time. In order to obtain the best operating condition to achieve high productivity with high purity and recovery of both products, effect of adsorption pressure, step time and the feed rate has been systematically investigated for both 3-bed and 4-Bed PSA columns. The salient results are reported below.

Table 2. *Comparison for 95 mol% purity of CO_2*

	3-Bed PSA	**4-Bed PSA**
Feed *SLPM*	19.5	19.5
P_H *atm*	20	20
P_I *atm*	0.683	-
P_L *atm*	1.55	1.43
Extract Purity *(mol%)*	~95	~95
Raffinate Purity *(mol%)*	95.82	94.29
Productivity of CO_2 *(SLPM/hr.Kg)*	0.0496	0.0370
Productivity of H_2 *(SLPM/hr.Kg)*	0.1991	0.1470

Table 3. *Comparison for 97% purity of CO_2*

	3-Bed PSA	4-Bed PSA
Feed *SLPM*	32	32.5
P_H *atm*	20	20
P_I *atm*	0.155	-
P_L *atm*	1.45	0.5
Extract Purity *(%)*	~97	~97
Raffinate Purity *(%)*	99.03	99.03
Productivity of CO_2 *(SLPM/hr.Kg)*	0.0827	0.0628
Productivity of H_2 *(SLPM/hr.Kg)*	0.3378	0.2573

The simulation results presented in above table 2 and 3 shows that a moderately high productivity can be achieved with 3-Bed PSA compared to 4-bed PSA, thereby minimizing the initial cost as well as operating cost for CO_2 separation using PSA technology.

The reason for better performance of 3-bed over 4-bed PSA can be understood by analyzing the concentration profiles shown in Figures 3-6 typically for 97 mol% purity of CO_2. This figure shows the solid and gas phase concentration profiles at the end of the steps in 3-bed and 4-bed PSA.

The shift in the concentration profiles from stripping to enriching step in Figure -5 indicates that large amount of CO_2 from the blowdown is circulated to enriching step and this limits the productivity of 4-bed PSA process. Whereas in 3-bed PSA recirculation between depressurization and stripping step is moderate as indicated by the shift in concentration profile between pressurization to stripping in Figure-3. It is observed that as purity requirement is lowered the recirculation amount reduces in both PSA process and their performance becomes almost similar.

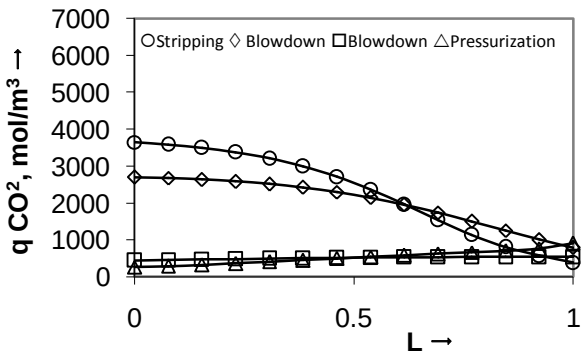

Figure 3. *Variation of amount adsorbed in solid phase with bed length for 3-Bed PSA (97% Purity)*

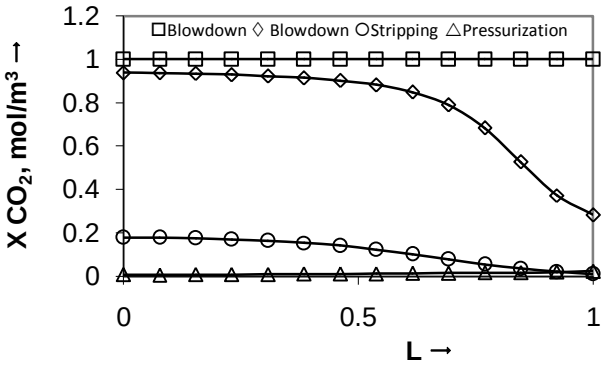

Figure 4. *Variation of concentration with bed length for 3-Bed PSA (97% Purity)*

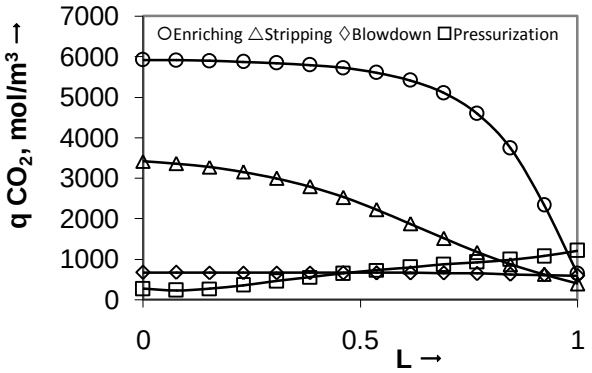

Figure 5. *Variation of amount adsorbed in solid phase with bed length for 4-Bed PSA (97% Purity)*

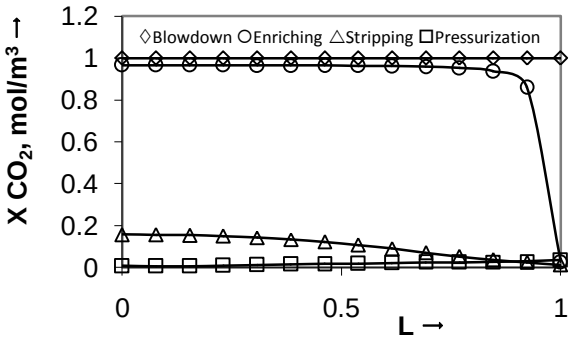

Figure 6. *Variation of concentration with bed length for 4-Bed PSA (97% Purity)*

5. CONCLUSIONS

The simulation results show that in 3-Bed PSA relatively high productivity can be achieved for high purity requirements of both products CO_2 and H_2. However, as the purity requirement is lowered the two processes give nearly same performance. The 3-bed PSA appears to be promising for CO_2-H_2 separation, however the results are only based on simulation and it warrants experimental validation.

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