Adsorption 4, 173–177 (1998)

R & D Note

Separation of a Nitrogen-Carbon Dioxide Mixture by Rapid Pressure Swing Adsorption

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Abstract. A N_2-CO_2 mixture is separated in a rapid pressure swing adsorption apparatus, which consists of single or double adsorbent beds filled with silica gel and operates in the sequence of adsorption, backflow and desorption. Nitrogen-rich gas is produced at the top of the bed, and carbon dioxide-rich gas at the bottom. Carbon dioxide purity of 89.5% and recovery of 70% were obtained in the single-bed apparatus, while purity of 93.5% and recovery of 72.3% were obtained in the double-bed apparatus. The feed in both cases consisted of 81% N_2 and 19% CO_2.

Keywords: rapid pressure swing adsorption, parametric pumping, carbon dioxide recovery

Introduction

Pressure swing adsorption (PSA) is an important process in the gas separation and purification industry. The cyclic operation procedure of PSA consists of the essential steps of high pressure adsorption and low pressure desorption coupled with other steps such as pressurization, depressurization, pressure equalization etc. These steps are included to improve PSA performance, which can be represented mainly by product purity, recovery and productivity. PSA systems containing more than ten beds have appeared, having complex operations of valve switching and interconnecting streams. However, a simpler process called rapid pressure swing adsorption (RPSA), or parametric pumping pressure swing adsorption (Turnock and Kadlec, 1971; Keller and Jones, 1980), utilizes a single adsorbent bed and gives several times higher productivity than the conventional PSA, at comparable purity and recovery. Its simplicity and characteristic fluid mechanism have attracted the attention of some investigators (Pritchard and Simpson, 1986; Guan and Ye, 1990a, 1993; Hart and Thomas, 1991; Alpay et al., 1993, 1994). In this paper, a revised RPSA process is used to separate a N_2-CO_2 mixture to yield the more strongly adsorbed component, CO_2, as product. A macroporous silica gel was chosen as the adsorbent.

Experiments

The macroporous silica gel, provided by Qingdao Chemicals Factory has average particle size of 0.44 mm, bulk density of 470 kg/m^3, and apparent density of 738 kg/m^3. Adsorption equilibria between N_2/CO_2 and the silica gel at 30°C were measured with the two vessel volumetric method described in Guan and Ye (1990b). The equilibrium isotherms are shown in Fig. 1. They are well linear in the pressure region experimented.

The schematic diagram of our single bed RPSA apparatus is shown as Fig. 2. The adsorbent bed is filled with macroporous silica gel as the adsorbent. CO_2 is adsorbed more strongly than N_2, while the gas mixture flows through the bed. N_2-rich product can be collected from the top of the bed, and CO_2-rich product can be obtained from the bottom when the bed is depressurized and desorbed. The flow rate of the top and bottom products, and pressure in the top and bottom tanks are controlled by two valves A and B. The feed contains 81% N_2 and 19% CO_2, and its pressure is controlled at

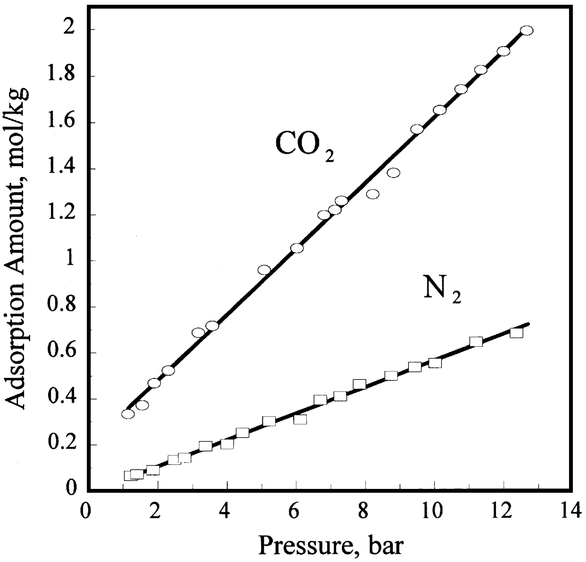

Figure 1. Equilibrium isotherms of N_2 and CO_2 on the silica gel at 30°C.

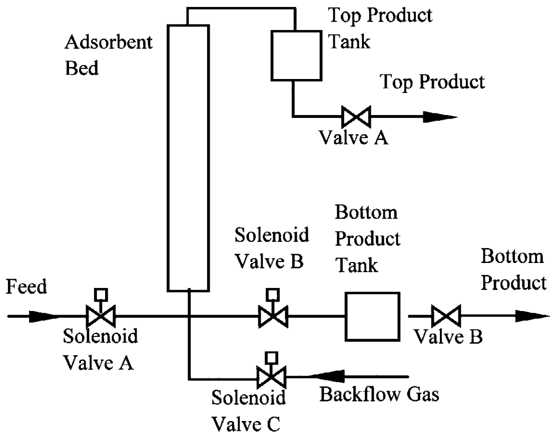

Figure 2. Schematic diagram of the single-bed RPSA apparatus.

10 bar. The equipment is operated at 30°C. The operation is controlled by three solenoid valves A, B and C plus a timer. Our cycle is composed of three steps:

(a) *Adsorption.* Solenoid valves B and C are shut off and A is opened, and thus the feed flushes into the bed, and selective adsorption occurs.
(b) *Backflow.* Solenoid valves A and B are shut off and C is opened, and backflow gas containing the same components as the bottom product flows into the bed.
(c) *Desorption.* Solenoid valves A and C are shut off and B is opened, and the gas in the bed flows out of the bed into the bottom product tank and depressurization and desorption occur.

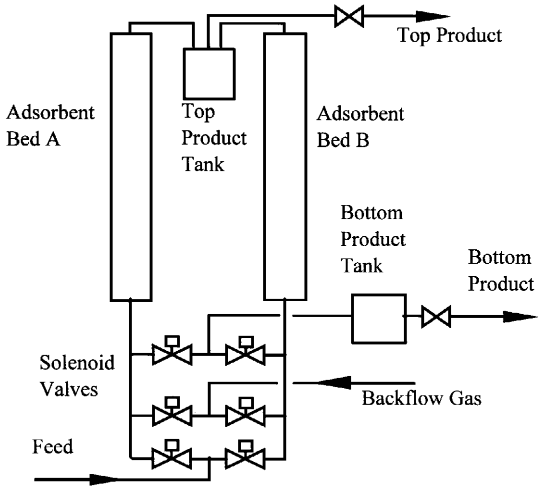

Figure 3. Schematic diagram of the double-bed RPSA apparatus.

A double-bed RPSA apparatus can be assembled by combining symmetrically two single beds, shown schematically in Fig. 3. Each bed follows the three-step cycle of the single bed described above. Bed A is in the adsorption step, and bed B is in the backflow and desorption step successively. Bed A is then in the backflow and desorption step, while bed B is in the adsorption step. The cycle is summarized as follows:

Bed A	Adsorption		Backflow	Desorption
Bed B	Backflow	Desorption	Adsorption	

The parameters for the single- and double-bed RPSA apparatus are listed in Table 1.

Table 1. Parameters for the RPSA apparatus.

Items	Values
Bed length and diameter	1.60 m, 20 mm
Top product tank volume	130 ml
Bottom product tank volume	2940 ml
Mass of adsorbent in each bed	235 g
Voidage of bed	0.356
Feed composition	81% N_2, 19% CO_2
Feed pressure	10 bar
Operating temperature	About 30°C

Discussion

An operating period of the conventional PSA is about several to tens of minutes, while that of RPSA is about several to tens of seconds. Following the operation described above, the pressure in the top product tank is still high at the end of the desorption step. At the beginning of the following adsorption step, pressurization from the top and bottom directions will occur because of the reflux gas in the top tank and the flushing feed at the bottom of the bed. The feed stream then passes into the bed towards the top tank, being accompanied with selective adsorption, and N_2-rich gas is collected in the top tank as the adsorption step proceeds. Then the adsorption step ends and backflow begins. The backflow gas contains the same components as the bottom product, i.e., enriched CO_2. The backflow gas flowing through the bed pushes the N_2-rich gas in the void of the bed towards the bed top, and on the other hand the adsorbed N_2 will be replaced by CO_2 in the backflow gas because of CO_2 being adsorbed more strongly than N_2. In this way, the purity of the desorbed CO_2 may be as high as possible. In the desorption step, depressurization occurs and the desorbed gas and the gas in the bed void flow in the reverse direction into the bottom tank as CO_2-rich product. A small amount of the top product is admitted at the top of the column to purge the desorbed gas from the bed voids at the end of desorption, as the bed is depressurized to a low pressure by rapid blowdown. Suitable pressure distribution of the bed and pressure drops in the pipes must exist for the above operation to proceed in good order. Thus, 60 ~ 80 screen mesh adsorbent was used to cause high pressure drop in the bed.

An example of experimental results and the corresponding operating conditions are shown in Table 2. It is seen from Table 2 that the pressure in the top tank is more steady and CO_2 purity, recovery and productivity are higher in the double-bed RPSA than the single-bed RPSA.

Table 2. Summary of experimental data.

Items	Single bed	Double beds
Feed pressure, bar	10	10
Period time, s	10.5	10.5
Pressure in top tank, bar	3.2 ~ 5	5.8 ~ 6.2
Pressure in bottom tank, bar	1.0	1.6
CO_2 purity of bottom product, %	89.5	93.5
CO_2 recovery, %	70	72.3
Productivity, kg 100% CO_2/kg adsorbent · day	1.95	2.07

The CO_2 recovery is calculated by

$$\mathrm{Rec\%} = \frac{(V_B - V_{\mathrm{BF}})X_B}{(V_B - V_{\mathrm{BF}})X_B + V_T X_T} \times 100 \qquad (1)$$

Figures 4–7 exhibit the influence of operating time and feed pressure on CO_2 purity, recovery and productivity of the bottom product and the pressure in the top tank. These data were measured with the single-bed RPSA apparatus.

The influence of adsorption time is shown in Fig. 4. The longer the adsorption step, the more feed flows into the bed and the more CO_2 is adsorbed in the adsorbent, which yields a higher bottom product purity

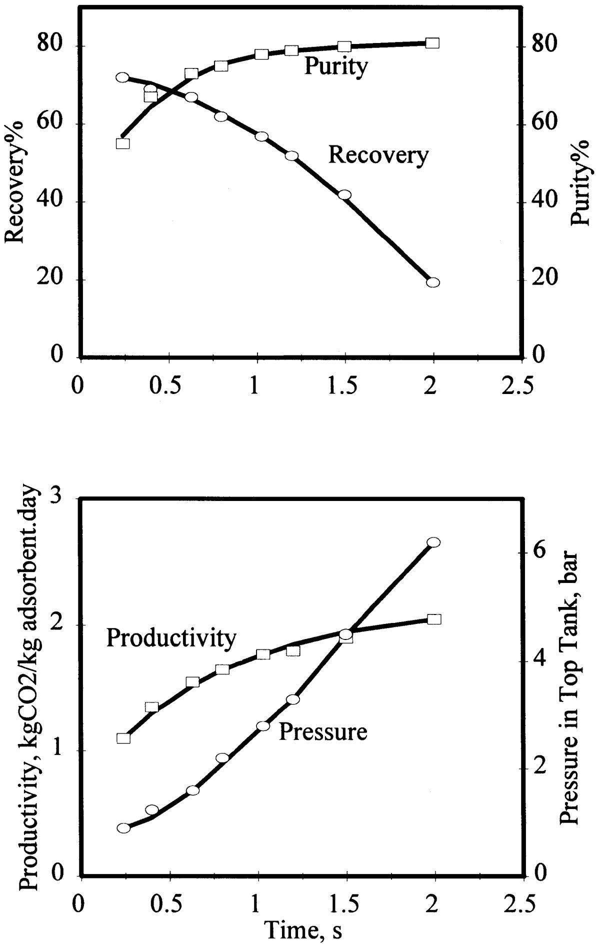

Figure 4. Influence of adsorption time. Feed pressure 10 bar, backflow time 4 s, desorption time 2 s.

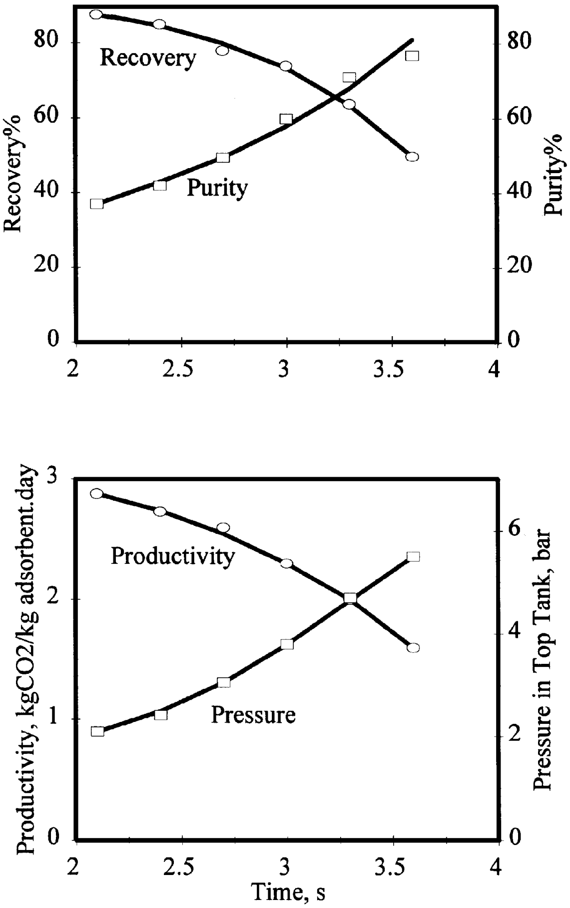

Figure 5. Influence of backflow time. Feed pressure 10 bar, adsorption time 1.5 s, desorption time 2 s.

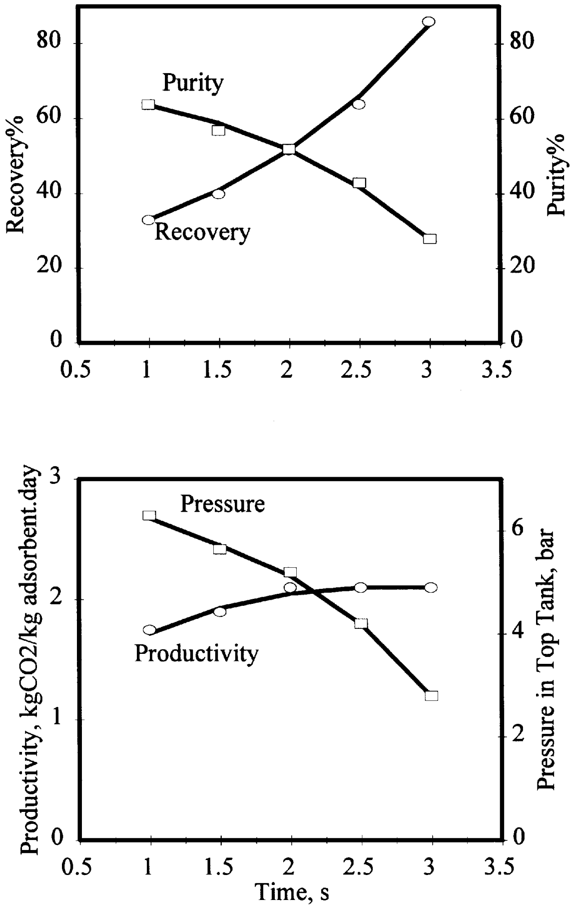

Figure 6. Influence of desorption time. Feed pressure 10 bar, adsorption time 4 s, backflow time 3 s.

and productivity and produces higher pressure in the top tank. However, the flowrate, V_T, and CO_2 mole fraction, X_T, of the top product increase rapidly as the adsorption time increases which makes the CO_2 recovery decrease rapidly, as shown as Eq. (1).

The influence of backflow time is shown in Fig. 5. As the duration of the backflow step increases, more bottom product reflows into the column and more CO_2 is adsorbed in the adsorbent and stays in the bed void. This yields higher CO_2 purity of the bottom product and increases pressure in the top tank, but lowers the CO_2 recovery and productivity because of more consumption of CO_2 in the backflow.

The influence of desorption time is shown in Fig. 6. Lengthening the desorption step will enhance depressurization and desorption of the bed into the bottom product tank, which means more CO_2 can be recovered. More top product of N_2-rich gas will reflow into the bed, though, so there is lower CO_2 purity of the bottom product and lower pressure in the top tank. Increase of recovery and decrease of purity at the same time makes the productivity change less.

The influence of feed pressure is shown in Fig. 7. If the operating time does not change, increasing the feed pressure will lead to higher pressure along the bed, which causes higher CO_2 recovery, purity, productivity and pressure in the top tank. The recovery and purity are affected less strongly than the productivity and top tank pressure.

Conclusion

The RPSA process described above can be used to yield the more strongly adsorbed component CO_2 as product from a feed gas of 81% N_2 and 19% CO_2. Higher CO_2

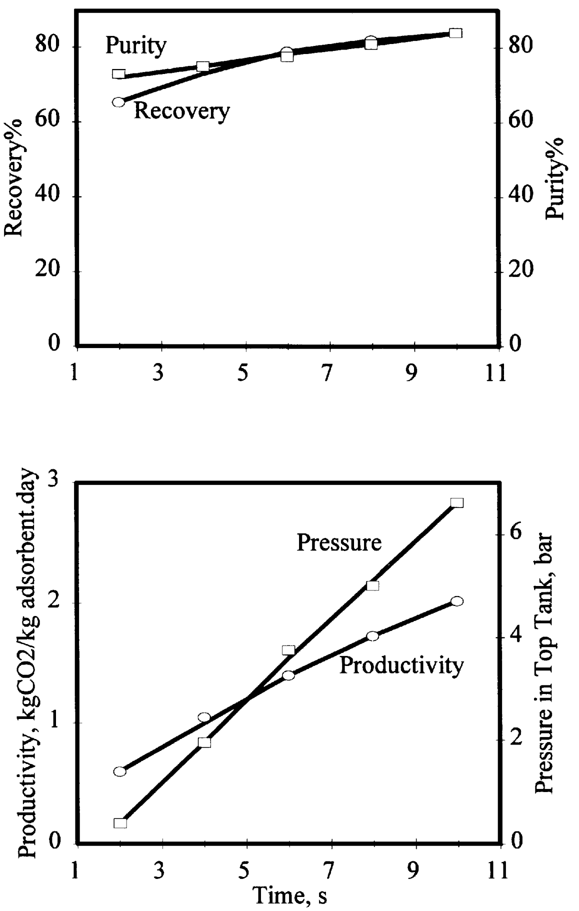

Figure 7. Influence of feed pressure. Adsorption time 4 s, backflow time 4.6 s, desorption time 2 s.

purity, recovery and productivity can be obtained with the double-bed RPSA than with the single-bed RPSA. Increasing the feed pressure can increase the CO_2 purity, recovery and productivity at the same time, but changing the operating time to increase the CO_2 purity will reduce the recovery.

Nomenclature

q	Adsorption amount	mol/kg adsorbent
P	Pressure	bar
Rec	CO_2 recovery	—
V	Volumetric flowrate,	m^3/s
X	Molar fraction of CO_2	—

Subscription

B	(of) Bottom product
BF	(of) Backflow stream
CO_2	Carbon dioxide
N_2	Nitrogen
T	(of) Top product

Acknowledgment

We are thankful to Guangdong Provincial Natural Science Foundation of China for financial support of this project. This project was supported by Guangdong Provincial Natural Science Foundation of China.

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