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Production of High-Purity Nitrogen from Air by Pressure Swing Adsorption on Zeolite X

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ABSTRACT

A PSA air separation process for the production of high purity nitrogen is studied experimentally and theoretically. The experimental apparatus is composed of three columns filled with zeolite X. The PSA cycle consists of six steps: pressurization with air, adsorption, recovery, null, high pressure purge with product nitrogen, and desorption. The nitrogen purity analyzed between 95.0 and 99.992%. The effects of adsorption pressure and reflux ratio on the PSA performance are studied experimentally. The productivity is 2.8 NL/(kg·min) and the recovery is 55% at a nitrogen purity of 99.99% and an adsorption pressure of 800 mmHg. The experimental results are compared with the theoretical model, and the effects of the recovery step and the operating variables on the performance are studied theoretically. The recovery step makes the PSA performance favorable, especially in the high product purity region above 99%.

INTRODUCTION

In the production of nitrogen from air by pressure swing adsorption (PSA), there are two proven commercial processes operating on different adsorbents (carbon molecular sieve, zeolite molecular sieve) and principles.

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The process using carbon molecular sieves (CMS) was first developed by Bergbau Forschung GmbH (1, 2) and generally produces nitrogen of 97–99.9% concentration. A deoxidizing unit, composed of a catalytic reactor and a dryer, is used for further purification of product nitrogen gas. The principle of separation is based on the difference of adsorption rates of oxygen and nitrogen on CMS. Oxygen, which is smaller in kinetic diameter than nitrogen, diffuses much faster in CMS than nitrogen, and hence the weak adsorbate, nitrogen, is enriched in the effluent from a CMS bed with air feed.

In nitrogen PSA using zeolite molecular sieves (ZMS), separation is achieved by the difference of the equilibrium adsorbed amount of nitrogen and oxygen. The more strongly adsorbed component, nitrogen, is obtained by desorption of the bed of ZMS. The cycle includes a high pressure product purge step to improve the product purity and is operated in vacuum swing adsorption (VSA). The unit comprises the pretreatment beds to remove moisture and carbon dioxide, and the separation beds of oxygen and nitrogen in the second stage. Up to now, three different processes have been developed.

The process of Air Products and Chemicals (3–5) consists of two parallel trains of adsorbent columns. Each train is comprised of the pretreatment bed and the separation bed connected in series. The cycle is operated at a pressure between slightly above 1 atm and 30–100 mmHg for the separation bed and 10–50 mmHg for the pretreatment bed. The cycle steps are (I) adsorption, (II) cocurrent nitrogen purge, (III) countercurrent desorption, and (IV) countercurrent oxygen repressurization. The product nitrogen purity is in the range between 98 and 99.4%, and oxygen of 90% purity is generated as a byproduct. The nitrogen recovery is approximately 50%. This process competes favorably with the cryogenic process in the 280 to 849 Nm^3/h range. Recently this company developed the fractionated vacuum swing adsorption (FVSA) process which eliminates the nitrogen purge step of the former process (6). The FVSA process has two desorption steps, and product nitrogen of 98+% purity is collected at the low pressure levels during the latter desorption step.

The Toray Industries process (7), which was introduced by Nippon Sanso Corp. (8), consists of two pretreatment beds followed by four separation beds. The operating pressure of the pretreatment beds is between 4.5 and 1 atm, and that of the separation beds is between 4 atm and less than 0.3 atm. The cycle steps of the separation bed are (I) pressurization by feed air, (II) cocurrent nitrogen purge, (III) countercurrent blowdown, and (IV) countercurrent evacuation. The effluent in the latter period of nitrogen purge step, in which the nitrogen concentration is higher than that of air, is used as a part of the feed air. This process is advantageous for the production of nitrogen purity of more than 99.9%. They commercialized the process which has three separation beds and produces 99.995% nitrogen in the production range of 30 to 500 Nm^3/h (9).

Kobe Steel has developed a nitrogen PSA which can generate product nitrogen with less than 50 ppm oxygen (10–12). The process consists of two pretreatment beds operating in the pressure range of 1 to 5 atm and three separation beds which are operated in the range between a pressure close to 1 atm and 100–150 mmHg. The cycle steps of the separation bed are (I) pressurization by feed air, (II) cocurrent recovery by the effluent of the nitrogen purge step, (III) cocurrent nitrogen purge, and (IV) countercurrent desorption.

For the recovery of the strongly adsorbed component as the desired product, a cocurrent depressurization step or a high pressure purge step by product is introduced in a PSA process. If a high purity strong adsorptive gas is the only desired product, the product purge process is better than the cocurrent depressurization process (13, 14). Baksh et al. suggested that a process involving cocurrent depressurization and the product purge step can reduce the pressurizing cost of the purge gas. The Kobe Steel process includes a new cycle in which the high pressure purge is accomplished by two steps (recovery and purge step). In the recovery step, the strong adsorptive gas in the effluent of the high pressure purge step can be recovered.

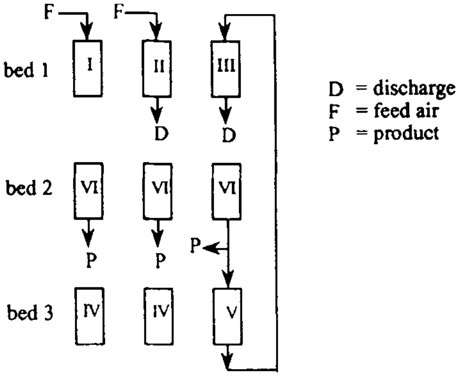
In the present paper, a nitrogen PSA using zeolite X is tested experimentally to produce high purity nitrogen. The performance of a PSA process is examined through the adsorption pressure and the reflux ratio. The experimental data are compared with the simulation results. The effects of the recovery step and the operating variables on the performance are studied theoretically.

PROCESS DESCRIPTION

The PSA process used in this work consists of two pretreatment beds and three separation beds. The process of the separation beds consists of the following six steps in each cycle: (I) pressurization, (II) adsorption, (III) recovery, (IV) null, (V) purge, and (VI) desorption. The cycle sequence and durations of steps for the three beds are shown in Fig. 1.

(I) Pressurization Step

The bed is pressurized to nearly atmospheric pressure by introducing the feed air removed H_2O and CO_2 in the pretreatment beds.



bed	process configuration								
1	I	II	III	IV		V	VI		
2	VI			I	II	III	IV		V
3	IV		V	VI			I	II	III

Steps: I) pressurization, 5 s; II) adsorption, 10 s;
III) recovery, 50 s; IV) null, 15 s; V) nitrogen purge, 50 s;
VI) desorption, 65 s

FIG. 1 Cycle sequence and step durations for a three-bed N_2 PSA process.

(II) Adsorption Step

The feed air is passed through the bed containing the nitrogen selective adsorbent. The pressure inside the column increases to a required adsorption pressure. The effluent of oxygen-enriched gas is used as the purge gas of the pretreatment bed.

(III) Recovery Step

Air feed is discontinued and the effluent of a nitrogen purge step is used to pre-purge the bed which finished the adsorption step in the same direction as feed air flow (cocurrent) at about the adsorption pressure. Therefore, the nitrogen contained in the effluent of purge step where the nitrogen concentration is higher than that of feed air is recovered in this step. The effluent of recovery step is sent to the pretreatment bed.

(IV) Null Step

Null step is used to fill the extra time present in run.

(V) Nitrogen Purge Step

The bed is purged cocurrently with a stream of high purity nitrogen at about the adsorption pressure. The effluent during this step is sent to the other bed which is in the recovery step. This step is continued until the bed is saturated with high purity nitrogen.

(VI) Desorption Step

The nitrogen-saturated bed is evacuated in the direction of feed air flow and high-purity nitrogen is produced. A part of this gas is used as the nitrogen purge of the other bed and the remainder is withdrawn as the product.

EXPERIMENTAL

The PSA unit consisted of a pretreatment section to remove moisture and carbon dioxide from ambient air and a following separation section to recover nitrogen from the purified air passed through the pretreatment section. The moisture and carbon dioxide concentrations of the feed air after the pretreatment section are −50°C dew point and 1 ppm, respectively. The separation section is composed of three nitrogen selective beds (150 cm in length, 5.45 cm in i.d.) filled with zeolite X. The control of solenoid valve switching and the data acquisition of measuring variables were done by a PLC (Texas Instruments, series 505/model 545) linked to a personal computer which was installed with FIX DMACS software. The flow rates of feed air, purge, discharge to the pretreatment bed, and product streams were measured by Sierra mass flow meters. The pressures within the beds were measured by Valcom pressure transducers. The oxygen concentration in the product stream was monitored continuously by an Ametek TM-1B oxygen analyzer.

The adsorption pressures were regulated to a pressure between 800 and 1200 mmHg with a pressure regulator, and the final evacuation pressure was between 90 and 110 mmHg. The experimental data were gathered after a cyclic steady state was attained in each run which usually took 20–30 cycles of operation.

MATHEMATICAL MODEL

The mathematical model proposed to simulate the experimental results makes the following assumptions:

1. The bed is isothermal.
2. The bulk gas phase follows ideal gas behavior.
3. The pressure drop through the bed is negligible.
4. The flow pattern is described by plug flow model. Axial dispersion is neglected.
5. The mass transfer rate is expressed by a linear driving force (LDF) model.

The feed air to the separation beds contains three components: N_2 (78.07 vol%), O_2 (21.00 vol%), and Ar (0.93 vol%). Argon adsorption on zeolite X is essentially identical to that of oxygen. The O_2–Ar pair is therefore treated as a single component. Oxygen concentration is determined by multiplying the calculated concentration of the O_2–Ar pair by (21.00/21.93).

Subject to these assumptions, the overall material balance in the bed can be written as

$$\frac{\partial C}{\partial t} + \frac{\partial uC}{\partial z} + \frac{\rho_b}{\epsilon} \sum_{k=1}^{N} \frac{\partial q_k}{dt} = 0 \qquad (1)$$

The mass balances for Component i is

$$\frac{\partial C_i}{\partial t} + \frac{\partial uC_i}{\partial z} + \frac{\rho_b}{\epsilon} \frac{\partial q_i}{dt} = 0 \qquad (2)$$

Introducing the ideal gas law and assuming that the bed pressure drop is negligible, the velocity profile is obtained from the overall mass balance:

$$\frac{\partial u}{\partial z} = -\frac{1}{P}\frac{\partial P}{\partial t} - \frac{\rho_b RT}{\epsilon P} \sum_{k=1}^{N} \frac{\partial q_k}{\partial t} \qquad (3)$$

Substituting Eq. (3) into Eq. (2) gives

$$\frac{\partial y_i}{\partial t} = -u\frac{\partial y_i}{\partial z} + \frac{\rho_b RT}{\epsilon P}\left(y_i \sum_{k=1}^{N} \frac{\partial q_k}{\partial t} - \frac{\partial q_i}{\partial t}\right) \qquad (4)$$

In the LDF model we have

$$\partial q_i/\partial t = k_i(q_i^* - q_i) \qquad (5)$$

where k_i is obtained from isobaric breakthrough curves and q_i^* is the amount adsorbed in equilibrium with the gas-phase concentration. The Langmuir adsorption isotherms are used to represent the pure component adsorption of oxygen and nitrogen.

$$q_i^* = \frac{a_i P y_i}{1 + b_i P y_i} \tag{6}$$

The boundary conditions for the subsequent cyclic steps are:

Pressurization (I):	$y_{\mathrm{I}.i}(t, 0) = y_{\mathrm{F}.i}$;	$u_{\mathrm{I}}(t, L) = 0$
Adsorption (II):	$y_{\mathrm{II}.i}(t, 0) = y_{\mathrm{F}.i}$;	$u_{\mathrm{II}}(t, 0) = u_{\mathrm{F}}$
Recovery (III):	$y_{\mathrm{III}.i}(t, 0) = y_{\mathrm{V}.i}(t, L)$;	$u_{\mathrm{III}}(t, 0) = u_{\mathrm{V}}(t, \mathrm{L}) \cdot P_{\mathrm{V}}/P_{\mathrm{III}}$
Null (IV):	no calculation	
Purge (V):	$y_{\mathrm{V}.i}(t, 0) = \bar{y}_{\mathrm{p}.i}$;	$u_{\mathrm{V}}(t, 0) = u_{\mathrm{purge}}$
Desorption (VI):	$\partial y_{\mathrm{VI}.i}(t, 0)/\partial z = 0$;	$u_{\mathrm{VI}}(t, 0) = 0$

where $\bar{y}_{\mathrm{p}.i}$ is the average effluent mole fraction of Species i during Step VI.

The initial conditions for each step are the conditions at the end of the preceding step. The pressure history is an input in PSA and is approximated in a linear dependence of pressure with time. The pressures of the recovery and purge steps are approximated to be same as that of the adsorption step (P_{H}). The pressure of the pressurization step varies from the desorption pressure (P_{L}) to the final pressurization pressure (P_{R}). In the desorption step, the pressure decreases from P_{H} to P_{L}.

An implicit backward finite difference scheme was employed to solve Eqs. (3) and (4) combined with Eqs. (5) and (6). Details of the computation method are given elsewhere (15, 16). The iteration scheme was continued until y_i was within 10^{-6} of the assumed value. The computation was carried out by using 50 space steps and time intervals as small as 0.01 second. It took approximately 4 minutes for each cycle on PC 486DX (33MHz). Depending on the parameter values, 12–40 cycles were required to attain a cyclic steady state.

Parameter Values Used in the Simulation

The equilibrium and kinetic parameters together with adsorber characteristics are listed in Table 1.

The adsorption isotherms of pure component for two different temperatures, 20 and 30°C, are shown in Fig. 2.

The adsorption rates were obtained by matching the experimental breakthrough curves with those from the numerical simulation. This method was described by Farooq and Ruthven (17) and Liow and Kenney (18). Ruthven and Xu (19) reported that intracrystalline diffusion of O_2 and N_2 is rapid in zeolite 5A so that the sorption kinetics in the commercial

TABLE 1
Characteristics of Adsorption Bed and Parameter Values Used in the Simulation

Bed diameter (d_b)	5.45 cm
Bed height (L)	150 cm
Adsorbent amount (W)	2.45 kg
Bed density (ρ_b)	0.7 g/cm^3
Bed porosity (ϵ)	0.364
Temperature (T)	303 K
Adsorbent	Zeolite X
Adsorbent diameter (d_p)	0.25 cm
Langmuir constant (a_i) for O_2, N_2	1.56E-4, 7.03E-4 mol/(kg·mmHg)
Langmuir constant (b_i) for O_2, N_2	4.67E-5, 4.18E-4 mmHg^{-1}
LDF constant for O_2	4 s^{-1}
LDF constant for N_2	1 s^{-1}
Operating pressure (mmHg): $P_L/P_R/P_H$	100/770/930, 110/1000/1200

adsorbent are controlled by macropore diffusion. The ratio of the LDF rate constants, k_{O_2}/k_{N_2}, can be approximated as inversely proportional to Henry's constant. In our zeolite X the ratio of Henry's constants ($O_2:N_2$) is about 4.5:1. Figure 3 shows the experimental and theoretical results for the adsorption and desorption of a 25.3% O_2 in N_2 mixture with pure

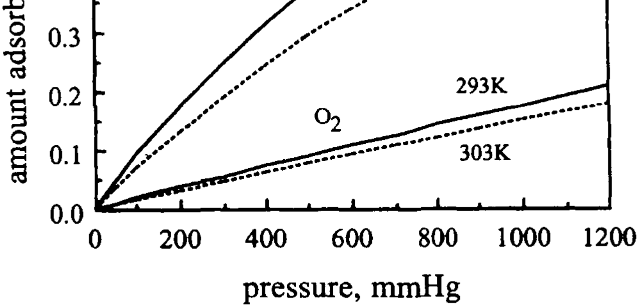


FIG. 2 Equilibrium isotherms of N_2 and O_2 on zeolite X.

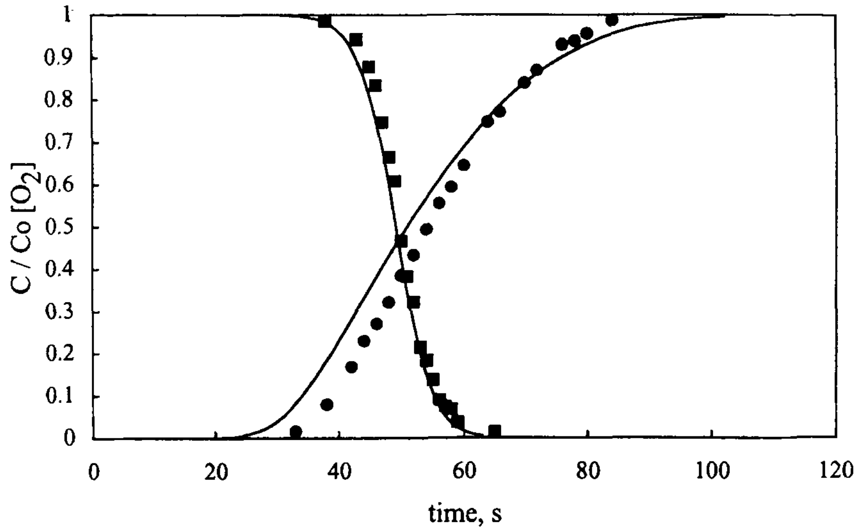


FIG. 3 Theoretical (line) and experimental breakthrough curves for the adsorption and desorption of 25.3% O_2–74.7% N_2 mixture and pure nitrogen.

nitrogen. The adsorption rate constants obtained were $k_{O_2} = 4\ s^{-1}$ and $k_{N_2} = 1\ s^{-1}$.

RESULTS AND DISCUSSION

The experimental goal of this study was to produce high-purity N_2 product (99.99% by volume) from air and to examine the effects of operating variables on the performance of this PSA process using zeolite X. The experimental results are compared with a theoretical model, and the effects of the recovery step and the operating variables on the performance are studied theoretically.

PSA performance can be described in terms of product recovery and productivity at a desired product purity. The product purities are evaluated as the volume-averaged concentration of desorbed gas during Step VI. The product recovery and the productivity are defined as

$$N_2 \text{ recovery} = \frac{(N_2 \text{ desorbed in Step VI}) - (N_2 \text{ used in Step V})}{(N_2 \text{ fed in Steps I and II})}$$

$$\text{Productivity} = \frac{(\text{gas desorbed in Step VI}) - (\text{gas used in Step V})}{\left(\text{mass of adsorbent} \times \frac{1}{3} \text{ cycle time}\right)}$$

The main operating variables are pressure (adsorption and desorption pressures), reflux ratio, and feed rate. The reflux ratio is the ratio of the reflux portion to the amount of desorbed gas during the desorption step.

$$\text{Reflux ratio} = \frac{\text{gas used in Step V}}{\text{gas desorbed in Step VI}}$$

$$\text{Feed rate} = \text{amount of feed introduced in Steps I and II}$$

The strongly adsorbed component, nitrogen, is the desired product in this PSA process. Oxygen is contained in the adsorbents and in the voids in the bed which finished the adsorption step. In order to produce high-purity nitrogen, the oxygen has to be replaced by the reflux nitrogen which is a portion of the desorbed nitrogen. Therefore, the most important step to improve the product purity is the nitrogen purge step. The reflux ratio is one of the key factors of the process economics, and the smaller the better.

Experimental Results

The oxygen concentration of the product stream was varied by changing the reflux ratio and the feed air pressure.

The effects of the reflux ratio and the adsorption pressure of feed air on the product purity and recovery are shown in Fig. 4. The figure shows that the product purity is a strong function of the reflux ratio. The product purity of N_2 for the constant adsorption pressure increases rapidly and then slowly with increasing reflux ratio. The reflux ratio of the turning point is around 0.6. In the range of the reflux ratio above 0.6, the lower the adsorption pressure becomes, the higher the product purity obtained. To produce high-purity nitrogen of more than 99.99%, the reflux ratio needs to be above 0.7 and a lower adsorption pressure is desirable. The recovery of nitrogen decreases gradually with increasing reflux ratio.

Figure 5 shows nitrogen recovery and productivity as a function of the product purity. The nitrogen recovery and productivity increases rapidly and then slowly as its purity decreases. The productivity for a given product purity increases as the adsorption pressure increases below a nitrogen purity of 99.95% (0.05% O_2), and the tendency is reversed in the other region. However, the recovery decreases with increasing adsorption pressure in these experimental conditions. The recovery is affected significantly by the feed rate (or bed utilization) more than by the adsorption pressure (analyzed in a later section). The feed rate in this experiment was varied with the adsorption pressure. The productivity is 2.8 NL/(kg·min) and the recovery of nitrogen is about 55% at a product purity of 99.99% and an adsorption pressure of 800 mmHg.

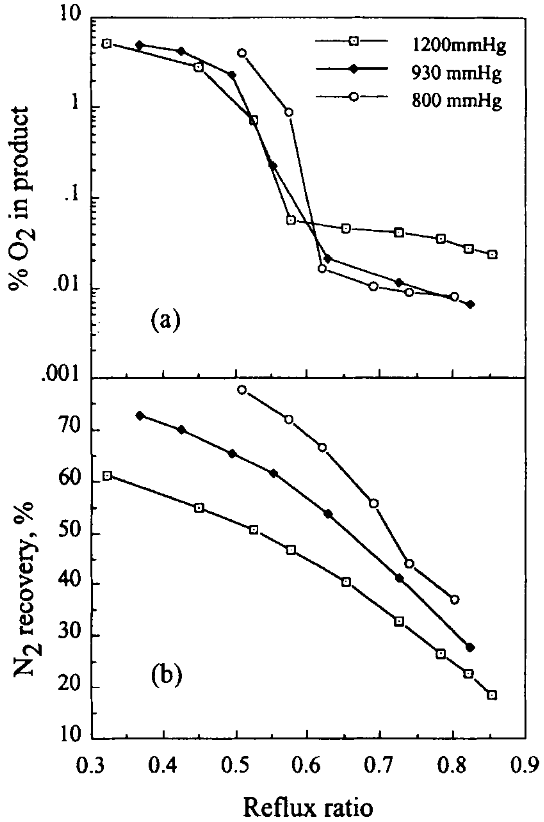


FIG. 4 Effects of reflux ratio and adsorption pressure on PSA performance (experimental results): (a) product purity, (b) recovery.

Comparison of Experimental and Theoretical Results

Figure 6 shows the comparison of experimental and theoretical results. The product purity and recovery are plotted as a function of the reflux ratio. The theory satisfactorily predicts the recovery of nitrogen and the product purity in the lower range (higher O_2 content). However, the predicted purity of product deviates severely from the experimental results at higher nitrogen purities. The major reason for this deviation is attributed to an imperfect mechanical design of the PSA apparatus or to the simplified model equations which do not consider the pore diffusion mechanism exactly within the adsorbent. The leakage of oxygen during the desorption step and the residue of oxygen in dead volume after the purge step can

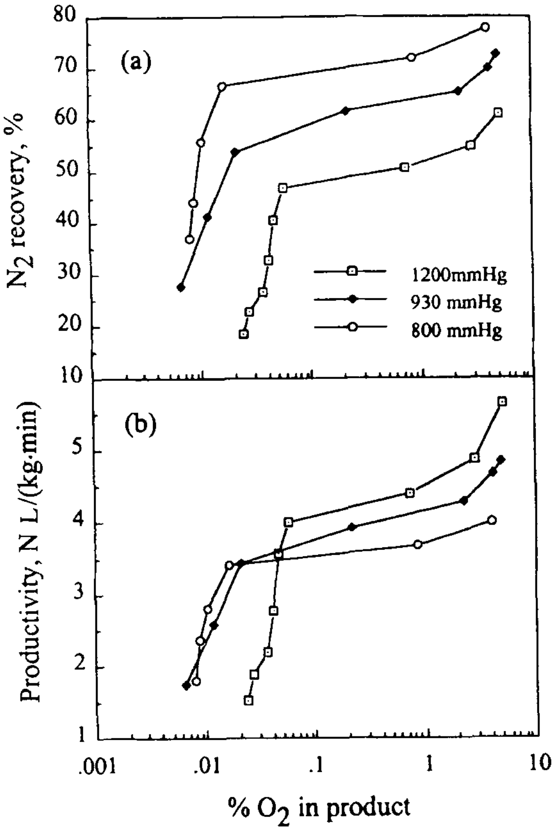


FIG. 5 Performance of N_2 PSA with adsorption pressure (experimental results): (a) recovery vs product purity, (b) productivity vs product purity.

greatly affect the high purity of the product. If 1 mL/min of oxygen is included in the product stream, the oxygen content in the product is 0.014%.

Parametric Studies by Model

Effects of Recovery Step. Figure 7 shows the gas phase nitrogen concentration profiles in the bed at the end of the pressurization (I), adsorption (II), recovery (III), and purge (V) steps for three different reflux ratios. The average concentration of the desorption step (VI) effluent is used as the concentration of N_2 purge. The bed profiles at the end of Steps

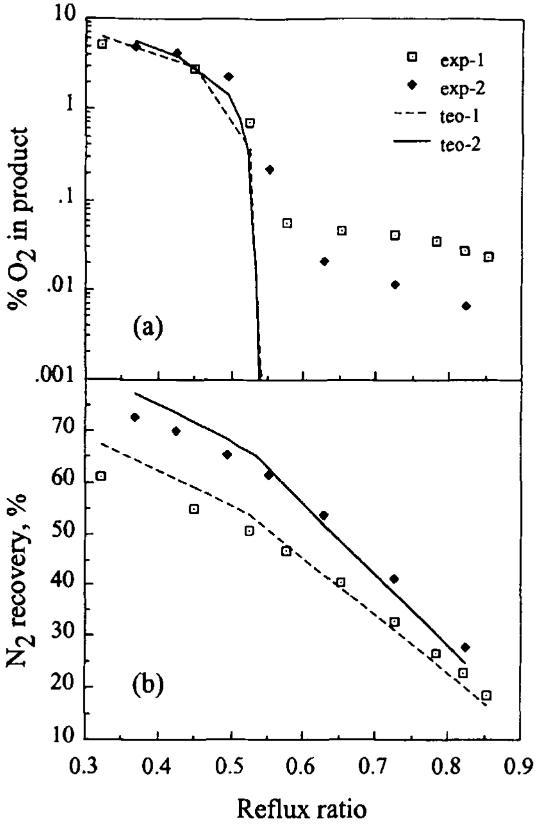


FIG. 6 Comparison of experimental and theoretical (line) results on PSA performance as a function of reflux ratio: (a) product purity, (b) recovery [adsorption pressure, (1) 1200 mmHg, (2) 930 mmHg].

I and II are nearly identical in all three runs. Even though bed utilization (the fraction of bed covered by strongly adsorbed component in adsorption Step II) is low, the profiles at the end of recovery Step III cover the profiles saturated with feed air composition ($y_{N_2} = 0.78$). Thus, much of feed air can be saved by introducing a recovery step to attain high bed utilization.

Figure 8 shows the PSA performance between the cycle with a recovery step and the cycle without a recovery step. In the region of high product purity (below 1% O_2 in product), the recovery and productivity of the cycle with a recovery step are higher than the cycle without a recovery

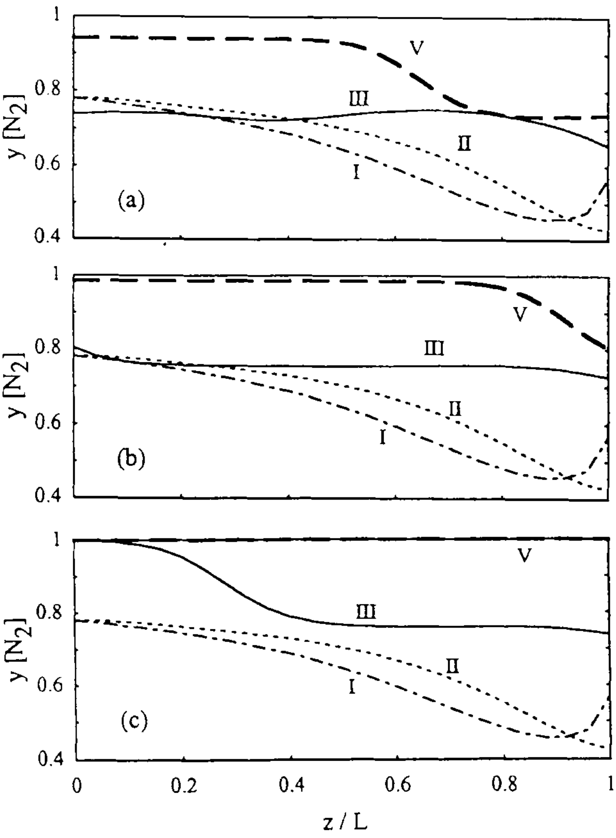


FIG. 7 Bed concentration profiles at the end of Steps I, II, III, and V: (a) reflux ratio = 0.369, (b) reflux ratio = 0.494, (c) reflux ratio = 0.535.

step. But in the other regions the performances of the two cycles are identical.

In a PSA process involving a purge step with product gas to obtain the strongly adsorbed component as the desired product, the amount of product is the difference between the amount desorbed during the desorption step and the amount used in the purge step. The amount of purge gas is the sum of the net purge gas requirement and the loss of strongly adsorbed component in the effluent during the purge step. The net purge gas requirement is the difference between the adsorbed amount of strongly adsorbed component at the end of the purge step and that at the beginning of the purge step. For a given product purity, the net purge gas requirement of

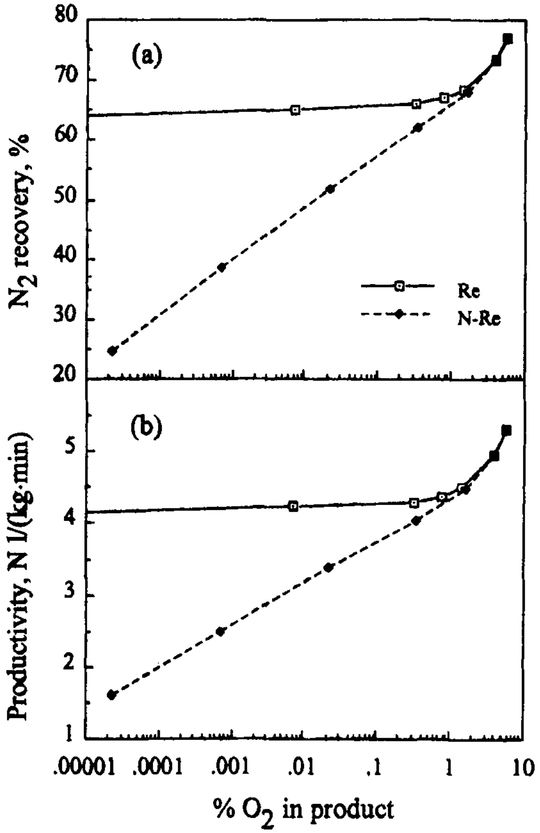


FIG. 8 Effects of the recovery step on PSA performance (Re = the cycle with recovery step, N-Re = the cycle without recovery step): (a) recovery vs product purity, (b) productivity vs product purity.

the cycle with the recovery step is smaller than that of the cycle without the recovery step because the bed profile of Step III is higher than that of Step II as shown in Fig. 7. The loss in the purge step depends on the bed concentration profiles at the beginning of the purge step. In the higher O_2 content region of product (i.e., lower reflux ratio as shown in Fig. 7a), the nitrogen concentration of the recovery gas, which is the effluent of the purge step, is slightly lower than that of the feed air, so the recovery step would not be helpful. In this case the N_2 loss of the cycle with a recovery step would be higher than that of the cycle without a recovery step, thus the smaller net purge gas requirement and the higher N_2 loss of the cycle with a recovery step compared with the cycle without a recovery step would result in an identical performance of the two cycles.

Effects of Feed Rate. The effects of feed rate on the performance are shown in Fig. 9. Feed rate is the amount of feed introduced in Steps I and II. The feed rate in Run 1 was approximately 22% lower than in Run 2. The other process conditions were kept the same in the two runs. For a given product purity, the recovery decreases with increasing feed rate. The productivity is not significantly affected by a change in feed rate. The bed concentration profiles in Fig. 10 show that the wavefront

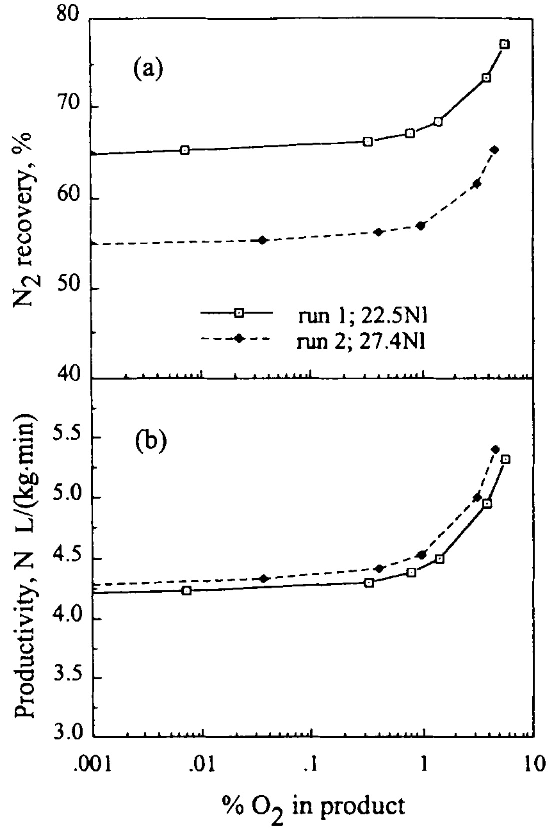


FIG. 9 Effects of feed rate on PSA performance: (a) recovery vs product purity, (b) productivity vs product purity.

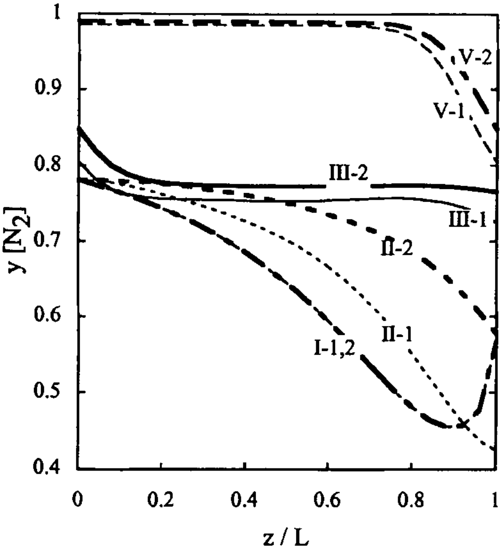


FIG. 10 Bed concentration profiles at the end of Steps I, II, III, and V with feed rate (1 = 22.5 NL, 2 = 27.4 NL).

at the end of the adsorption step moves to the bed end with increasing feed rate, i.e., the bed utilization increases. This results in an early breakthrough in Steps II and III, thus decreasing the recovery, and improves the product purity. Since the bed concentration profiles for two runs are close at the end of the Steps III and V, respectively, the productivity and the purity are not much different.

Effects of Adsorption Pressure. A change in adsorption pressure at the conditions with nearly identical bed utilization (Runs 3 and 4) does not affect the product purity and recovery, as shown in Fig. 11. The higher feed rate and the identical recovery at a higher adsorption pressure increase the product purity. The disadvantage of high adsorption pressure is the extra compression required for the high purge gas pressure.

An increase of adsorption pressure with the same feed rate (Runs 4 and 5) decreases the bed utilization, giving a late breakthrough in Steps II and III, and thus it improves both the recovery and the productivity.

Effects of Desorption Pressure. Figure 12 shows the effects of desorption pressure (final pressure of the desorption step) on performance. Both the productivity and the recovery for a given product purity increase with decreasing desorption pressure. Since the feed rates with decreasing desorption pressure are not much different (20.8, 22.5, 23.9 NL) and the adsorption pressures are identical, the bed profiles at the end of Step III are close, as shown in Fig. 10. Figure 13 is the theoretical results of a desorption step initially saturated with air. It can be seen that the average N_2 concentration and the amount of desorbed gas increase with decreasing desorption pressure. Similar effects were reported by Sircar and Hanley

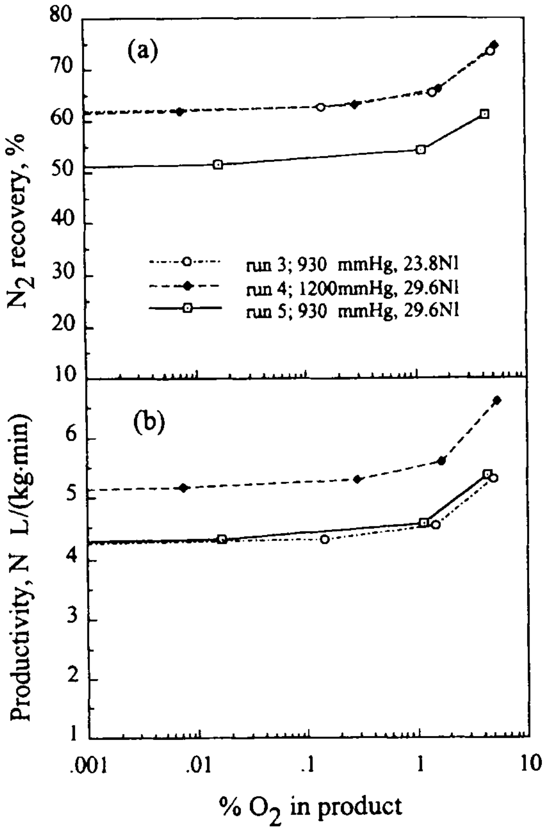


FIG. 11 Effects of adsorption pressure on PSA performance: (a) recovery vs product purity, (b) productivity vs product purity.

(6). Both the higher N_2 purity and the larger amount of desorbed gas at a lower desorption pressure can reduce the amount of purge gas required for a given product purity and result in more favorable PSA performances (high productivity and recovery). Desorption pressure, however, is related to the operating cost of a vacuum pump, so an optimal desorption pressure should be determined by considering the production cost.

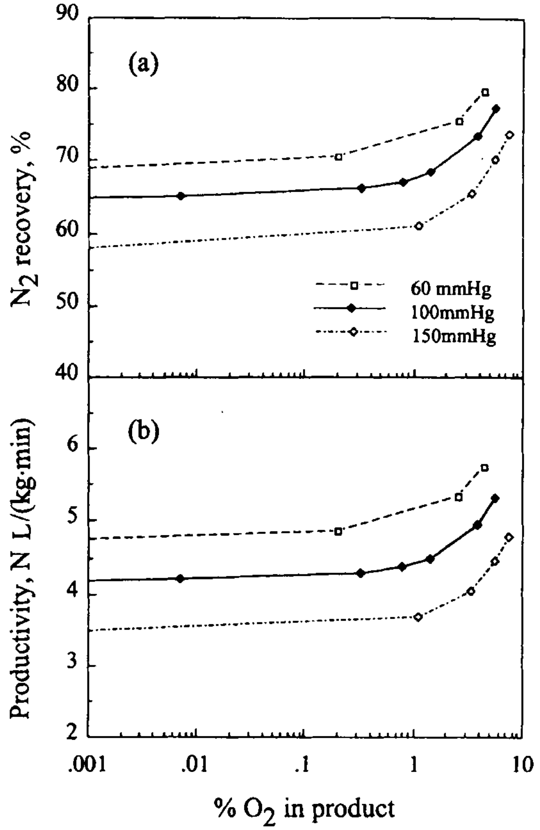


FIG. 12 Effects of desorption pressure on PSA performance: (a) recovery vs product purity, (b) productivity vs product purity.

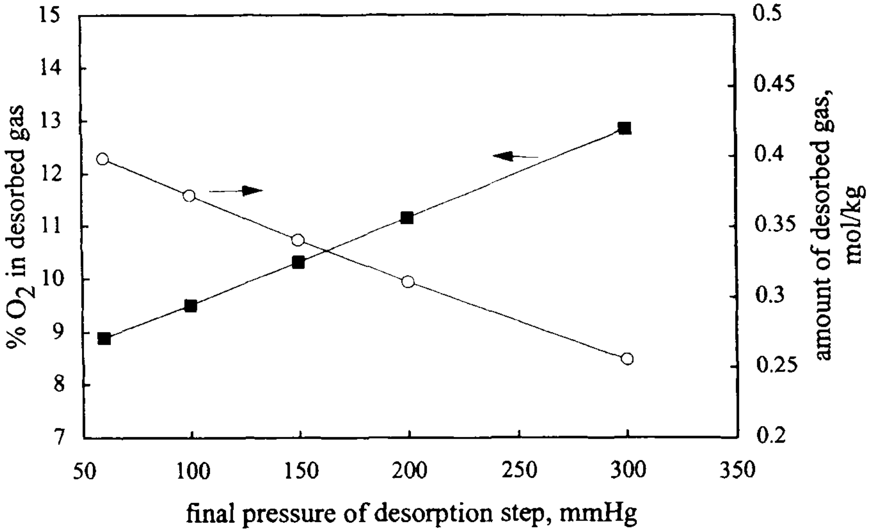


FIG. 13 Average concentration and amount of desorbed gas with final pressure during the desorption step (initial condition: 930 mmHg, saturated with air).

CONCLUSIONS

The use of the PSA air separation process to produce a high purity nitrogen product using zeolite X was studied experimentally and theoretically.

1. A PSA cycle involving a high pressure product purge step and a recovery step can produce high purity nitrogen. In a typical run, the productivity is 2.8 NL/(kg·min) and the recovery is 55% at a nitrogen purity of 99.99% and an adsorption pressure of 800 mmHg.

2. The product purity of N_2 increases rapidly and then slowly with an increasing reflux ratio. The reflux ratio of the turning point is around 0.6. For a reflux ratio in the range above 0.6, the lower the adsorption pressure becomes, the higher the product purity obtained. The recovery of nitrogen decreases gradually with an increasing reflux ratio. The productivity for a given product purity increases as the adsorption pressure increases below the nitrogen purity of 99.95% (0.05% O_2), and the tendency is reversed in the other region.

3. The theory predicts well the recovery of nitrogen and the product purity of the lower range (higher O_2 content). However, the predicted purity of the product deviate severely from the experimental results at a

higher nitrogen purity. The major reason for this deviation is attributed to an imperfect mechanical design of the PSA apparatus or to the simplified model equations which did not consider the exact pore diffusion mechanism within the adsorbent.

4. The following conclusions were drawn from parametric studies with the model. The recovery step makes the PSA performance favorable, especially in the high product purity region above 99%. The recovery is affected significantly more by the feed rate than by the adsorption pressure, while the productivity is strongly affected by the adsorption pressure.

NOMENCLATURE

a_i	Langmuir constant [mol/(kg·mmHg)]
b_i	Langmuir constant (mmHg^{-1})
C	total adsorbate concentration (mol/cm^3)
C_i	concentration of *i*th component
d_b	bed diameter (cm)
d_p	adsorbent diameter (cm)
k_i	overall mass transfer coefficient of *i*th component (s^{-1})
L	bed height (cm)
N	total number of adsorbates in adsorption system (—)
NL	units of volume, liter at 20°C and 1 atm
P	total pressure (mmHg)
P_H	pressure of adsorption step (mmHg)
P_L	end pressure of desorption step (mmHg)
P_R	final pressure of pressurization step (mmHg)
R	ideal gas constant
q_i^*	amount adsorbed in equilibrium with the gas phase concentration C_i (mol/kg)
T	temperature (K)
t	time (s)
u	interstitial flow velocity of gas (cm/s)
W	adsorbent amount (kg)
y_i	mole fraction of *i*th component in gas phase (—)
$\bar{y}_{p\cdot i}$	product purity (—)
z	axial distance coordinate (cm)

Greek Letters

ϵ	bed porosity (—)
ρ_b	bed density (g/cm^3)

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