

## EFFECTS OF PRESSURE DROP IN A PSA PROCESS

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**Abstract** – The effects of pressure drop on the dynamics of fixed-beds were theoretically studied. The system used was an H<sub>2</sub>/CO mixture (70 vol% H<sub>2</sub>, 30 vol% CO) in zeolite 5A. The pressure drop at the pressurization step affected the breakthrough time at an adsorption step in the PSA process. As a result, the combined effects of pressure drop during adsorption and pressurization steps led to earlier breakthrough compared to the case without a pressure drop. The effect of pressure drop at the adsorption step under the non-isothermal condition was slightly larger than that under the adiabatic condition. In the case of pressurization and blowdown steps with large pressure drop, the flow pattern near the open end during a short period of time had to be explained by the Ergun equation instead of Darcy's law. However, there was only a slight difference in the results of a multi-bed PSA process depending on whether or not the pressure drops at the pressurization/depressurization steps as well as at the adsorption step were considered.

Key words: PSA, Pressure Drop, Darcy's Law, Ergun Equation, Non-Isothermal Model

### INTRODUCTION

The pressure swing adsorption (PSA) process has become a widely used unit operation for gas separation or purification. To understand and develop the PSA process, many researchers have extensively studied it [Kim et al., 1995; Yang et al., 1996, 1997a, b]. Recently, the effect of pressure drop on the dynamics of gas flow in fixed-bed adsorbers has received a great deal of attention, particularly for rapid pressure swing adsorption (RPSA). In the case of a multi-bed PSA process, relatively large adsorbents and long step times are used. Therefore, it is assumed that the pressure drop in the adsorption bed is not so great and thus the axial pressure profile is ignored. However, since pressurization and depressurization steps exist even in a multi-bed PSA process and the length of the commercial adsorption bed is fairly large, it seems impossible to ignore completely the effect of pressure drop. Several investigators have studied the individual steps of a PSA process under isothermal conditions [Buzanowski et al., 1989; Hart et al., 1990; Rodrigues et al., 1991; Lu et al., 1992a]. The theoretical study of Sundaram and Wankat [1988] showed strong effects of pressure drop in the pressurization and blowdown steps in a PSA cycle. The effects of pressure drop on PSA performance under adiabatic conditions were discussed by Doong and Yang [1988]. Among these studies, Buzanowski et al. [1989] presented some experimental evidence for the effect of pressure drop during adsorption and desorption steps. Zhong et al. [1992] studied the pressurization of a single component gas using Darcy's law in a non-isothermal intraparticle diffusion model. They pointed out that

adsorption kinetics are not always a secondary effect and that they play an important role in column dynamics as well as the adsorption equilibrium properties in an activated carbon bed through experimental and theoretical studies. Kikkinides and Yang [1993] presented theoretical and experimental results of the effect of pressure drop on an adiabatic adsorber through breakthrough experiments using dilute O<sub>2</sub> in He. They pointed out that constant pattern behavior is distorted and the plateau zones can be either deflated or inflated.

To study the effect of pressure drop on separation, all the previous authors introduced a momentum equation with a form of either Darcy's law (laminar flow) or Ergun's equation (turbulent flow) in their model equations. Sereno and Rodrigues [1993] checked the validity of steady-state momentum equations in modeling pressurization of adsorption beds under the isothermal condition.

Efficient control of pressure in the adsorption bed is important in realizing the goal of a PSA process, such as high purity and/or recovery. The method of increasing/decreasing pressure in the bed especially plays a very important role in a PSA process. Therefore, more studies on the dynamics of the axial pressure drop not only in an RPSA process but also in a multi-bed PSA process need to be done in order to obtain more accurate PSA process design data [Yang, 1987; Ruthven et al., 1994].

In the present study, the effects of pressure drop on the dynamics of a zeolite 5A packed bed in H<sub>2</sub>/CO (70 vol% H<sub>2</sub>, 30 vol% CO) mixture were investigated numerically by using three different adsorption beds. Both components were considered as adsorbable gas. The simulation results of pressure drop effects for the pressurization, adsorption and blowdown steps were presented. Its effects were also examined on the performance of a multi-bed PSA process under the

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non-isothermal condition.

## THEORY

A mathematical model, including mass and energy balances, was constructed to develop a complete non-isothermal PSA model with the following assumptions: (i) The flow pattern is described by the axial dispersion plug flow model, (ii) thermal equilibrium is assumed between fluid and particles, (iii) mass transfer rate is represented by a linear driving force (LDF) model, (iv) the gas phase behaves as an ideal gas mixture, (v) the radial concentration and temperature gradients are negligible.

The mass balance equations for gas flow in the bed are for a component mass balance and for an overall mass balance, respectively:

$$-D_L \frac{\partial^2 y_i}{\partial z^2} + \frac{\partial y_i}{\partial z} + u \frac{\partial y_i}{\partial z} - \frac{RT}{P} \frac{1-\varepsilon}{\varepsilon} \rho_p \left( \frac{\partial \bar{q}_i}{\partial t} - y_i \sum_{j=1}^n \frac{\partial \bar{q}_j}{\partial t} \right) = 0 \quad (1)$$

$$-D_L \frac{\partial^2 P}{\partial z^2} + \frac{\partial P}{\partial t} + P \frac{\partial u}{\partial z} + u \frac{\partial P}{\partial z} - \frac{P}{T} \left( -D_L \frac{\partial^2 T}{\partial z^2} + \frac{\partial T}{\partial t} + u \frac{\partial T}{\partial z} \right) + \frac{1-\varepsilon}{\varepsilon} \rho_p RT \sum_{j=1}^n \frac{\partial \bar{q}_j}{\partial t} = 0 \quad (2)$$

The energy balance equation for a non-isothermal column is:

$$(\alpha \rho_g C_{p,g} + \rho_B C_{p,s}) \frac{\partial T}{\partial t} - \varepsilon K_L \frac{\partial^2 T}{\partial z^2} + \rho_g C_{p,g} \varepsilon u \frac{\partial T}{\partial z} - \rho_B \sum_i Q_i \frac{\partial \bar{q}_i}{\partial t} + \frac{2h_i}{R_B} (T - T_w) = 0 \quad (3)$$

$$\rho_w C_{p,w} A_w \frac{\partial T_w}{\partial t} = 2\pi R_{Bi} h_i (T - T_w) - 2\pi R_{Bo} h_o (T_w - T_{am})$$

$$A_w = \pi(R_{Bo}^2 - R_{Bi}^2) \quad (4)$$

For the adsorption of a binary gas mixture, the pressure drop for a column can be obtained by using Darcy's law or the Ergun equation. The steady-state momentum equation used in this study can be written as:

$$-\frac{dP}{dz} = a\mu u + b\rho u |u| \quad (5)$$

$$\text{where } a = \frac{150}{4R_p^2} \frac{(1-\varepsilon)^2}{\varepsilon^3} \quad b = 1.75 \frac{(1-\varepsilon)}{2R_p \varepsilon^3}$$

In Eq. (5), the second term on the right-hand side becomes important when the flow velocity is in the turbulent region. Eq. (5), which has only the first term on the right-hand side, is called Darcy's law.

The LDF model and the extended Langmuir-Freundlich isotherm were used as a sorption rate model and an isotherm for gas mixture, respectively. The boundary conditions used in this process and the required parameters involving the adsorption isotherms can be found in a previous work [Han et al., 1995, 1996; Yang et al., 1995, 1997]. A backward-two-step finite difference method (FDM) was used to solve the mathematical model.

**Table 1. Characteristics of adsorption bed and adsorbent**

Adsorption bed		
Bed A		
length	L=100 cm	
pellet size	R <sub>p</sub> =0.157 mm	
Bed B		
length	L=100 cm	
pellet size	R <sub>p</sub> =0.9157 mm	
Bed C		
length	L=1,000 cm	
pellet size	R <sub>p</sub> =0.5175 mm	
Bed inside radius	R <sub>Bi</sub> =2.2 cm	
Bed outside radius	R <sub>Bo</sub> =2.55 cm	
Wall heat capacity	C <sub>pw</sub> =0.12 cal/g·K	
Bulk (bed) density	ρ <sub>B</sub> =0.795 g/cm <sup>3</sup>	
External void fraction	ε=0.315	
Total void fraction	α=0.76	
Pellet density	ρ <sub>p</sub> =1.16 g/cm <sup>3</sup>	
Heat capacity	C <sub>ps</sub> =0.22 cal/g·K	
Adsorbates*	H <sub>2</sub>	CO
k <sub>1</sub> (mmole/g)	4.31	5.05
k <sub>2</sub> (mmole/g·K)	-0.01060	-0.00905
k <sub>3</sub> (atm <sup>-1</sup> )	0.002515	0.001137
k <sub>4</sub> (K)	458.2	1617.0
k <sub>5</sub> (-)	0.9860	0.5245
k <sub>6</sub> (K)	43.03	256.5
Heat of adsorption (Q, cal/mol)	2800	5300
LDF coefficient (k <sub>i</sub> , cm <sup>2</sup> /s)	0.5	0.049

\*Langmuir-Freundlich equation:

$$q_i = \frac{q_m B_i P_i^n}{1 + \sum_{j=1}^n B_j P_j^n}, \quad q_m = k_1 + k_2 T, \quad B = k_3 e^{k_4/T}, \quad n = k_5 + k_6/T$$

LDF model:

$$\frac{\partial \bar{q}_i}{\partial t} = k_i (q_i^* - \bar{q}_i)$$

In this study, three different beds were used for simulation. The first is a packed bed with small particles for an RPSA process; the others are two different beds densely packed for a conventional PSA process. For the simulation of an RPSA bed (bed A in Table 1), energy balances were excluded from the simulation model because the RPSA beds do not experience so much temperature swing as the conventional PSA beds for bulk separation. On the other hand, for conventional PSA beds (beds B and C in Table 1), energy balances of Eq. (3) and (4) were incorporated. All the bed conditions for simulation are given in Table 1.

## RESULTS AND DISCUSSION

### 1. Effect of Pressure Drop on Pressurization Step

The pressurization step was studied by using an input of a high pressure mixture of gas at 11 atm into a clean bed. Fig. 1 shows the axial profiles of the H<sub>2</sub> mole fraction during the pressurization of bed B. As can be seen in Fig. 1, the effect of pressure drop by high gas velocity in the bed was great enough to show a large difference between the predicted axial concentration profiles with the Ergun equation and those

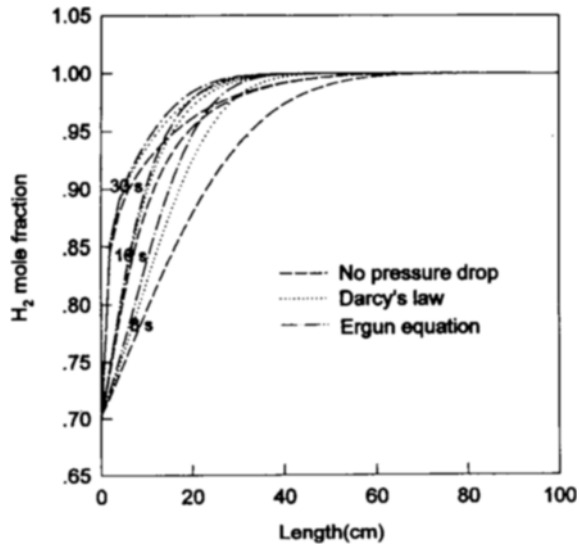


Fig. 1. Axial profiles of  $H_2$  mole fraction during pressurization step.

with Darcy's law, as mentioned in the theory section. The difference between the two axial profiles by the Ergun equation and Darcy's law was reduced with time. However, there was still some difference between the model results with/without a pressure drop. Due to high velocity at an early period of the pressurization step, a concentration front penetrated deeply into the closed end. After that short period, the concentration front moved to the open end of the bed with time in spite of continuous feeding. This is due to the adsorption of CO gas and the rapid decrease of axial velocity.

These results can be explained clearly by a bed velocity profile and a pressure profile in bed A for an RPSA process. Fig. 2 shows the axial profiles of velocity predicted by using the Ergun equation during pressurization from 1 atm to 5 atm for bed A. The corresponding axial profiles of pressure at different times are shown in Fig. 3. Since pressure pro-

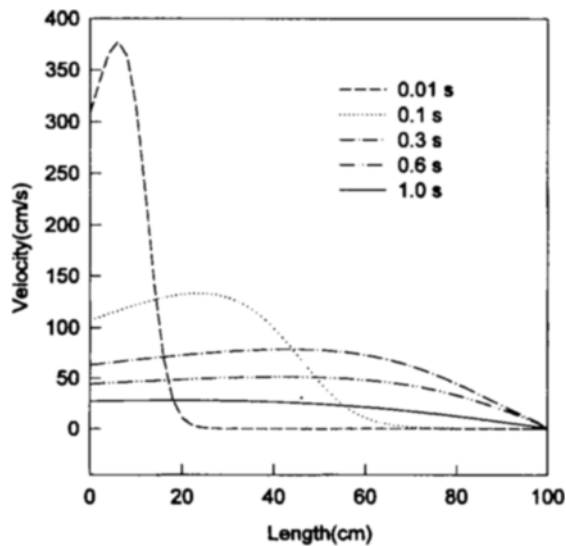


Fig. 2. Axial profiles of velocity during pressurization for bed A.

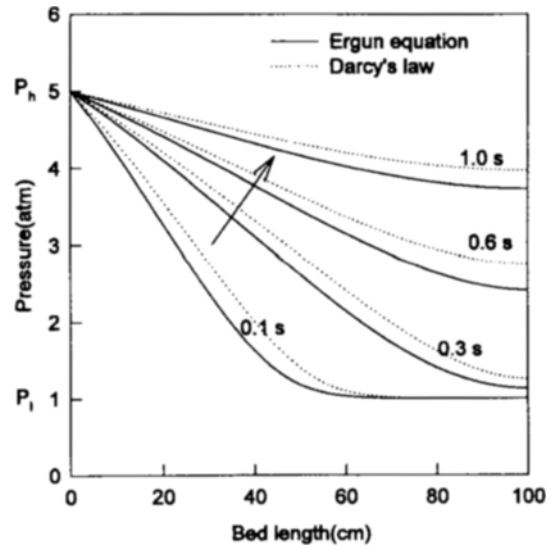


Fig. 3. Axial profiles of pressure during pressurization for bed A.

files had inflection points during a short period of time by gas expansion, the increase of velocities in Fig. 2 was much higher near the open end of the column up to these points. As a result, the axial velocity profiles have a maximum where the inflection point of pressure is present. As shown in Fig. 3, the results of pressure profiles by the Ergun equation show a larger pressure drop and take a longer time to reach a steady state than those by Darcy's law. In particular, there was a large pressure drop at the open end of the adsorption bed. However, as time passed, the pressure drop decreased.

Lu and coworkers [1992a, b] pointed out that highly turbulent flow in the open end must be considered under very high flow velocity in the pressurization and depressurization steps. This case may play an important role in kinetic separation processes [Zhong et al., 1992], because weakly adsorbing and slowly diffusing species may propagate more rapidly in the bed, which the opposite of what would be obtained from equilibrium adsorption.

## 2. Effects of Pressure Drop on Breakthrough Curves

Since the effects of pressure drop during breakthrough in short beds A and B were negligible, the adsorption step was studied by using an input of a high pressure mixture of gas at 11 atm into bed C with commercial bed length after the bed was pressurized by pure  $H_2$  up to 11 atm. In Fig. 4, the influence of pressure drop on breakthrough time under various thermal conditions in bed C was analyzed. In general, the rise of temperature due to the exothermic heat of adsorption led to earlier breakthrough because of decreased adsorption capacity. This influence became larger as the thermal condition approached an adiabatic condition. As revealed in previous studies [Doong and Yang, 1988; Buzanowski et al. 1989; Kikkinides and Yang, 1993], the pressure drop in an adsorption bed resulted in earlier breakthrough compared to the case of negligible pressure drop in every thermal condition. Such a phenomenon was explained in terms of an increase in interstitial velocity due to the expansion of gas caused by pressure drop at the adsorption step [Kikkinides and Yang,

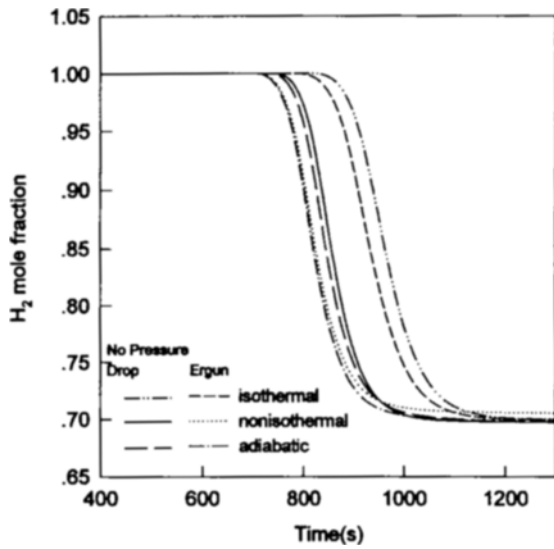


Fig. 4. Effect of thermal condition on breakthrough curve at 11 atm adsorption pressure and 2 l/min feed rate in bed C.

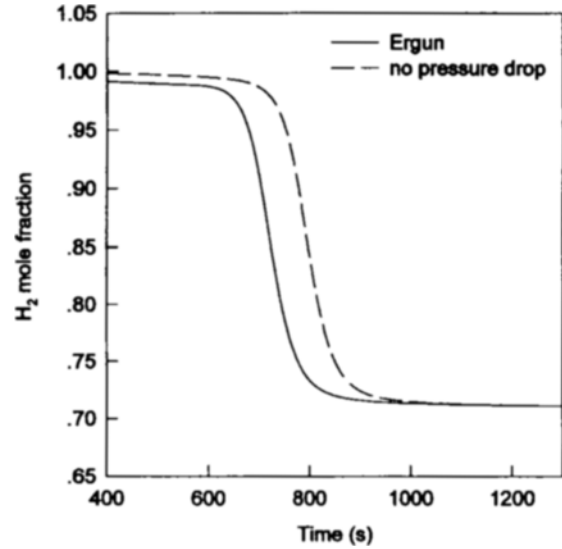


Fig. 6. Effect of pressure drop on breakthrough curve including feed pressurization step under nonisothermal condition in bed C.

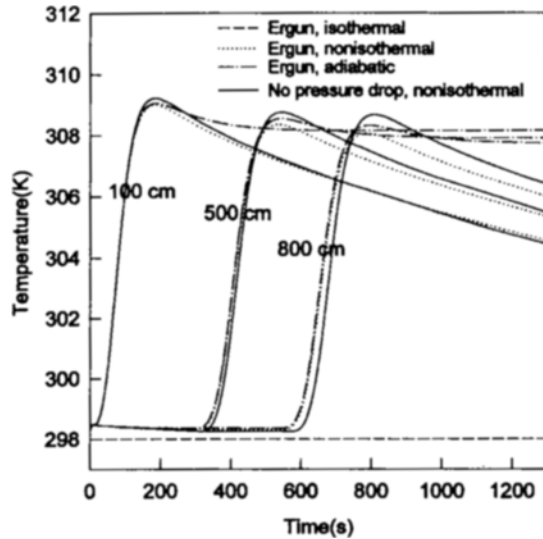


Fig. 5. Effect of thermal condition on temperature variation at three points at 11 atm adsorption pressure and 2 l/min feed rate in bed C.

1993].

The effect of pressure drop on the velocity of the thermal wave at a non-isothermal condition is also shown in Fig. 5, which plots temporal variation of temperature at three different positions in the bed. Since the continuous supply of feed gas at 298 K and the heat loss through the bed wall under the non-isothermal condition are chilling the bed, the temperature after its maximum rise inside the bed was decreased. There was no significant difference in temperature variation between the results of the Ergun equation and those of Darcy's law. Due to low velocity in the bed, the effect of the second term on the right hand side of Eq. (5) could be negligible in the adsorption step. However, the effect of pressure drop resulted in slower passing of the thermal wave and in lower temperature variation compared to the results of a neg-

ligible pressure drop.

As shown in Figs. 1-3, the pressure ratio between the two ends of the adsorption bed turned out to be very great at the beginning of pressurization. In this study, to investigate this pressure drop effect at the next step, i.e., the adsorption step, we simulated the adsorption step including a pressurization step. After the bed was pressurized by a gas mixture of up to 11 atm for 30 s, the adsorption step was started. In the case of bed B (short bed), whose pressure drop at the adsorption step was not significant, the pressure drop at the pressurization step did not affect the breakthrough time. However, as shown in Fig. 6, in the case of bed C (long bed), whose pressure drop at the adsorption step was significant, the breakthrough time was shortened by the pressure drop of the pressurization step, and the difference of breakthrough time was larger than that of the corresponding case in Fig. 4. As can be seen from the above results, even though the pressure drop at the adsorption step itself is dominant at that step, the effects of pressure drop in the pressurization step also permeate through the adsorption step.

### 3. Effect of Pressure Drop on Countercurrent Depressurization (Blowdown) Step

Countercurrent blowdown (depressurization) is the step at which a high pressure gas exhausts through one end to low pressure. This step is a very important step in an RPSA cycle as well as in a conventional PSA cycle because it performs self-purging as well as regeneration [Doong and Yang, 1988; Sundaram and Wankat, 1988].

As blowdown progresses, the changes in the velocity and pressure profiles in bed A are shown in Figs. 7 and 8. As in the pressurization step, the gas velocity and the pressure drop during the blowdown step were not constant, and the results of the Ergun equation showed a larger pressure drop than those of Darcy's law. However, the velocity profiles do not show the maximum value in the bed because gas flows to the open end and because the pressure profiles are slightly smoother

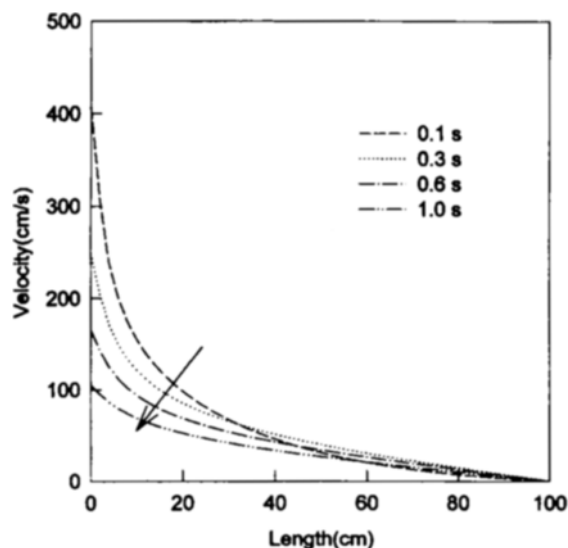


Fig. 7. Velocity profiles during depressurization from the pre-saturated bed with feed at 5 atm for bed A.

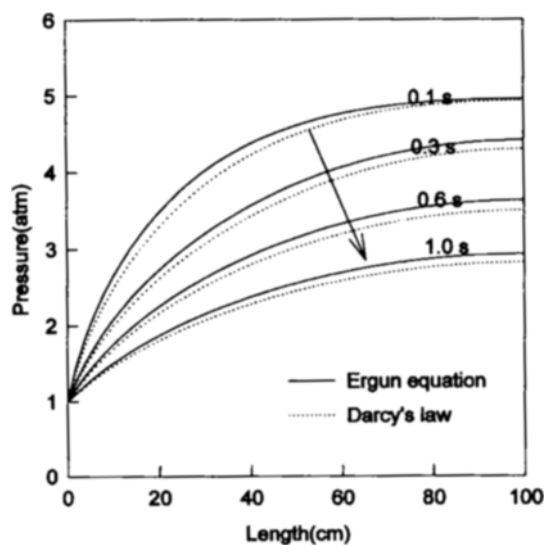


Fig. 8. Axial profiles of pressure during depressurization from the pre-saturated bed with feed at 5 atm for bed A.

than those during the pressurization step. Again, the Ergun equation would be more accurate for predicting the inside velocity and the pressure profiles than Darcy's law due to the turbulent flow. The large pressure drop during this step slowed down the blowdown rate so that it prevented the gas in the void spaces from escaping the column quickly. As a result, CO concentration in the blowdown step is high at the feed end of the bed and decreases gradually along the bed.

#### 4. Effect of Pressure Drop on PSA Performance

Fig. 9 plots the purity and recovery of H<sub>2</sub> versus a cycle number for a two-bed six-step PSA process which has the following steps: feed pressurization (30 s), adsorption (180 s), depressurizing pressure equalization (40 s), countercurrent depressurization (30 s), purge (180 s), and pressurizing pressure equalization (40 s). The details of this process are described elsewhere [Yang et al., 1997]. Using this conventional PSA

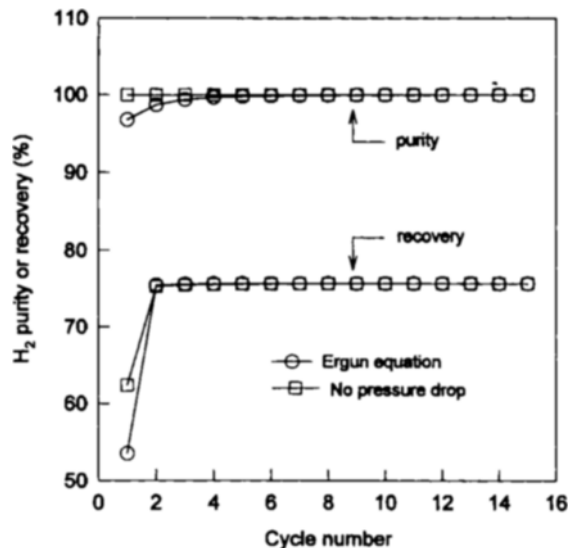


Fig. 9. Effects of pressure drop on PSA performance simulated under the nonisothermal condition for a two-bed six-step PSA process.

(Simulated condition: 2 l/min feed rate, 11 atm adsorption pressure, 0.22 l/min purge rate)

system with bed B, we tested the effect of pressure drop on PSA performance under the operating condition of 2 l/min feed rate and 11 atm adsorption pressure.

At the beginning of the PSA cycle, the H<sub>2</sub> purity and recovery predicted by simulation with pressure drop was lower than that without pressure drop. Because pressure drop resulted in a decrease of adsorption capacity of CO, the purity of the H<sub>2</sub> product was decreased by this effect. However, the discrepancy between these results by two different models disappeared gradually with a cycle number and, eventually, only a trivial difference was found under the tested condition after a cyclic steady state.

## CONCLUSIONS

A theoretical study on the effects of the pressure drop on the dynamics of three kinds of adsorption beds was presented. In addition, the influence on the performance of a conventional PSA cycle was also investigated by numerical simulation.

The pressure drop effects were significant during the pressurization and blowdown steps even when a conventional cycling time scheme was used. During pressurization and blowdown steps, the results of the Ergun equation showed a larger pressure drop, and it took longer time to reach the steady-state pressure than those of Darcy's law. However, these differences during a blowdown step were slightly smaller than those during a pressurization step.

As compared to the case without a pressure drop, the presence of a pressure drop led to an earlier breakthrough and some difference in the axial profiles of mole fraction during an adsorption step under the non-isothermal condition. The effect of pressure drop was different depending on the thermal conditions, and this effect was larger under non-isother-

mal conditions than that under isothermal and adiabatic conditions in the tested system. The pressure drop at the pressurization step affected the breakthrough time at the adsorption step in the PSA process. It seems that an increase in flow velocity, which was due to a large pressure drop at the open end of the adsorption bed during the pressurization step, resulted in shortening the breakthrough time.

For a multi-bed PSA process, there was a negligible effect of pressure drop on the separation results at a cyclic steady state. However, during a cyclic unsteady state, the effect of pressure drop on the PSA process should be considered.

#### ACKNOWLEDGMENT

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#### NOMENCLATURE

A	: cross sectional area [ $\text{cm}^2$ ]
B	: Langmuir-Freundlich isotherm parameter [ $\text{atm}^{-1}$ ]
$C_p$	: heat capacity [ $\text{cal/g}\cdot\text{K}$ ]
$D_L$	: mass axial dispersion coefficient [ $\text{cm}^2/\text{s}$ ]
$K_L$	: thermal axial dispersion coefficient [ $\text{cal/K}\cdot\text{cm}\cdot\text{s}$ ]
$k_i$	: mass transfer coefficient for LDF model [ $\text{s}^{-1}$ ]
n	: Langmuir-Freundlich isotherm parameter
P	: pressure [ $\text{atm}$ ]
q	: adsorbed amount [ $\text{mol/g}$ ]
$q_m$	: Langmuir-Freundlich isotherm parameter [ $\text{mol/g}$ ]
$\bar{q}$	: volume-averaged amount adsorbed in an adsorbent [ $\text{mol/g}$ ]
Q	: average isosteric heat of adsorption [ $\text{cal/mol}$ ]
R	: radius [ $\text{cm}$ ]
t	: time [ $\text{s}$ ]
$T_{amb}$	: ambient temperature [ $\text{K}$ ]
T	: pellet or gas temperature [ $\text{K}$ ]
u	: interstitial velocity [ $\text{cm/s}$ ]
h	: heat transfer coefficient [ $\text{cal/cm}^2\cdot\text{K}\cdot\text{s}$ ]
y	: mole fraction in bulk gas phase
z	: axial position in the bed [ $\text{cm}$ ]

#### Greek Letters

$\alpha$	: total void fraction
$\epsilon$	: interparticle void fraction
$\rho$	: density [ $\text{cm}^3/\text{g}$ ]
$\mu$	: viscosity [ $\text{cm/g}\cdot\text{s}$ ]

#### Subscripts

B	: bed
I	: species i
p	: adsorbent pellet
g	: gas phase
s	: solid phase
w	: wall

#### Superscript

*	: equilibrium
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