

Simulation of Pressure Swing Adsorption Process of Butane/Air Separation in Carbon-Coated Ceramic Monolithic Adsorber

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The new shaped structured adsorber, carbon monolithic adsorber, are characterized by straight parallel channels separated by thin wall, high void fraction and large geometric surface area, resulting in a low pressure drop under high flow rate and large contact area. These properties make it have the advantage on adsorption application. This study simulates the adsorption separation of 30% butane and 70% air on carbon-coated ceramic monolithic adsorber for a three-step pressure swing adsorption process under isothermal condition. The parameters considered in the model include the mass transfer coefficient to the channel wall, effective diffusion within the pore structure and axial dispersion model. The model analyzes the effect of variables such as, carbon-coated wall thickness, feed pressure, etc. on the performance of pressure swing adsorption.

1. Introduction

In traditional pressure swing adsorption processes, packed beds were used due to the inexpensive and adaptable adsorbent but made high mass transfer resistance and pressure drop at high flow rate. Ruthven and Thaeon (1996) indicated that the structure adsorber with parallel passage channel is advantageous to improve the mass transfer and pressure drop for adsorption application. The monolithic adsorber was developed with straight parallel channels separated by thin wall, high void fraction and large geometric surface and contact area, resulting in a low pressure drop and high mass transfer rate under high flow rate. These properties make the monolithic adsorber have the advantage in pressure swing adsorption application. There are two types of carbon monolithic adsorbers: carbon-coated and integral carbon monolithic adsorbers. The former one is carbon coated onto either the ceramic or metallic monolithic backbone which supplies honeycomb geometric and strong mechanical properties. The coated porous carbon has to provide the adsorptive properties. The later one, integral carbon monolithic adsorber, is made of carbon completely that has to provide mechanical, geometric and adsorptive properties.

Volatile organic compounds (VOCs) are typical pollutants in the atmosphere. Adsorption is a conventional process to remove and recover VOCs from industrial emission by using activated carbon adsorbent. For the VOCs enriching purposes, the

experimental data and simulation model on carbon based monolithic adsorbent or adsorber had been reported before. Yu et al. (2002) used activated carbon monolith as an adsorbent to remove VOCs (toluene, 1-butanol and ethyl acetate) and established the adsorption isotherms of VOCs with different models. Valdés-Solís et al. (2004) studied the adsorption and breakthrough performance of n-butane on carbon-coated ceramic monoliths. The study showed the best description of the breakthrough performance was considering a parabolic gas velocity distribution over the cross-sectional area of the monoliths.

According to the characteristics and studies of carbon based monolithic adsorbent listed above, carbon monolithic adsorbents are able to be used in adsorption, and even more, with the potential application in a pressure swing adsorption (PSA) process. This study is aimed to describing a mathematical model and simulates the PSA process of butane/air separation in single carbon-coated ceramic monolithic adsorber as shown in Figure 1, where Butane is used as an example of VOCs. The simulation shows the influence of various parameters for the performance of PSA process.

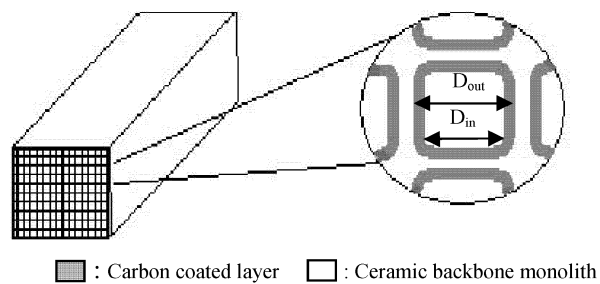


Figure 1 Schematics of the carbon coated ceramic monolithic adsorber configuration.

2. Modeling and PSA Process Description

The model used to describe the PSA process is based on single square channel with a flat carbon coating onto the ceramic backbone and is derived from the mass balances, including the following assumptions: (1) The gas behaves as an ideal gas. (2) Isothermal operation is considered. (3) The axial dispersed plug flow through the channel. (4) Effective diffusional transport and adsorption equilibrium are considered in carbon-coated layer. (5) Pressure drop and radial diffusion through the channel are neglected. (6) The adsorbed amount of air is neglected.

2.1 Overall mass balance in monolithic channel

The overall mass balance in the gas phase for the monolithic channel is

$$-\frac{\partial q}{\partial z} = \frac{\varepsilon_m A}{RT} \frac{\partial P}{\partial t} + \frac{a'(1-\varepsilon_m)Ak_f}{RT} \sum_{i=1}^n P(y_i^b - y_i^*) \quad (1)$$

where q is molar flow rate, a' is external area per unit of carbon and k_f is mass transfer

coefficient which is calculated from the Sherwood number predicted by Hawthorn correlation (Valdés-Solís et al., 2004),

$$Sh = \frac{k_f D_m}{D_m} = 2.976(1 + 0.095 \text{Re} \text{Sc} \frac{D_m}{L})^{0.45} \quad (2)$$

2.2 Mass balance for component i in monolithic channel

$$\frac{\varepsilon_m A}{RT} \frac{\partial}{\partial z} (D_{ax,i} P \frac{\partial y_i^b}{\partial z}) - \frac{\partial (q y_i^b)}{\partial z} = \frac{\varepsilon_m A}{RT} \frac{\partial}{\partial t} (P y_i^b) + \frac{a'(1 - \varepsilon_m) A k_f}{RT} P (y_i^b - y_i^*) \quad (3)$$

Where the axial dispersion coefficient D_{ax} is calculated by the Taylor-Aris relation. The flow rates at both ends of bed are estimated by valve equation that is recommended by Fluid Controls Institute Inc. (Chou et al, 1994). Where q' is flow rate in m^3 (STP)/min, P_1 is upstream pressure, P_2 is downstream pressure, and C_V is valve flow coefficient.

$$q' = 0.01605 C_V \sqrt{\frac{(P_1^2 - P_2^2)}{SG \times T}} \quad \text{for } P_2 > 0.53 P_1 \quad (4)$$

$$q' = 0.01361 C_V P_1 \sqrt{\frac{1}{SG \times T}} \quad \text{for } P_2 \leq 0.53 P_1 \quad (5)$$

2.3 Mass balance for component i in monolithic channel wall

$$\frac{\partial \bar{q}_i}{\partial t} + \varepsilon_w \frac{\partial C_i^w}{\partial t} = \frac{D_{eff}}{RT} \frac{\partial}{\partial r} (P \frac{\partial y_i^w}{\partial r}) \quad (6)$$

The adsorption starts at inner surface (gas-solid interface) of the channel diffusing through carbon coated layer towards the carbon-ceramic interface, i.e., from $r = D_{in}/2$ to $r = D_{out}/2$ in Figure 1, where r is a radial position. The amount of butane adsorbed is given by the Dubinin-Radushkevich isotherm:

$$\bar{q} = q_{\max} \exp \left[- \left(\frac{RT}{\beta E_0} \ln \frac{c_{sat}}{c_i^w} \right)^2 \right] \quad (7)$$

The parameters of Dubinin-Radushkevich isotherm and base physical parameters for carbon monolith in Table 1 were obtained from Valdés-Solís et al. (2004).

2.3 Mass balance for component i in monolithic channel wall

A three-step PSA cycle was used in this study. The steps involved in the cycle were: (I) Adsorption with the feed gas (mixture of 30% butane and 70% air on molar basis); (II) Cocurrent depressurization; (III) Countercurrent desorption and production. Figure 2 shows the schematic of the PSA cycle used and time of each step.

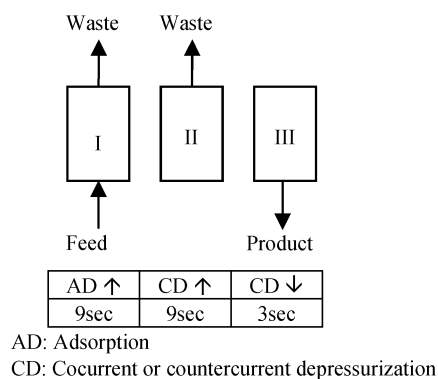


Figure 2 Schematic diagram of the three-step PSA process. ($P_F = 2\text{atm}$, $P_W = 0.1\text{atm}$, $P_P = 0.03\text{atm}$)

Table 1 Physical parameters used in the simulation for different carbon-coated cermaic monolithic adsorber.

Internal non-coated channel width, D_{out} (m)			1.320×10^{-3}
Internal coated channel width, D_{in} (m)	8×10^{-4}	1×10^{-3}	1.292×10^{-3}
Porosity of monolith, ε_m	0.37	0.59	0.96
Porosity of monolith wall, ε_w			0.69
Adsorber length, L(m)			0.5
Molecular diffusivity, D_m (m^2/s)			1.7×10^{-5}
Effective diffusivity, D_{eff} (m^2/s)			3.3×10^{-9}
Maximum amount adsorbed, q_{max} (kg/m^3)			5.0934×10^2
Saturation adsorptive concentration, c_{sat} (kg/m^3)			6.48
Affinity coefficient \times characteristic energy of adsorbent, βE_0 (J/mol)			22767

The model set of partial differential equations (PDEs) is transformed into ordinary differential equations by the method of lines. The set of ODEs are solved successfully with respect to time by using a FORTRAN program with LSODE of ODEPACK software.

3. Results and Discussion

3.1 Influence of Carbon Coated Layer Thickness

To increase the thickness of carbon coated layer decreased the void fraction of monolithic adsorber. Figure 3 shows the effect of carbon coated layer thickness on three-step PSA process with constant C_v . The results in Figure 3 show the purity and recovery increase with the thickness of carbon coated layer increase. The reason is that thicker coated carbon layer has more adsorption capacity and smaller void volume of adsorber, that make thicker one ($\varepsilon_m = 0.37$) to have lower adsorber pressure and more butane desorption than thinner one ($\varepsilon_m = 0.96$) after cocurrent depressurization step (step

II) with time and valve values fixed. The more desorbed butane enhance the purity and recovery at countercurrent production. The results for the pressure profile of adsorber with time is shown in Figure 4. In our simulation, a line representing adjusting the valve value of $\epsilon_m = 0.96$ is shown in Figure 4. The valve value for $\epsilon_m = 0.96$ is adjusted to make the profile of pressure similar to the one for $\epsilon_m = 0.37$. The simulation results show that the lower pressure after step II can improve the purity from 36.8% ($\epsilon_m = 0.96$, constant C_v) to 46.3% ($\epsilon_m = 0.96$, variable C_v), but reducing the recovery from 4.3% to 2.5%. The recovery reduction is caused by more butane exhaust for decreasing pressure of adsorber at step II.

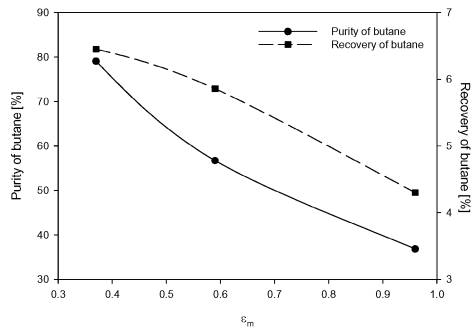


Figure 3 Effect of the carbon coated layer thickness on butane purity and recovery.

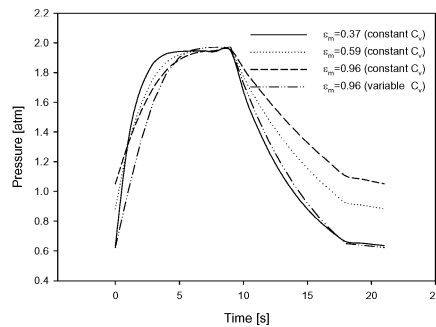


Figure 4 The pressure profile of the bed with time on different-thickness carbon coated layer.

3.2 Influence of Feed Concentration

The effect of feed molar fraction of butane on butane purity and recovery are exhibited in Figure 5. As the feed molar fraction of butane increasing, more butane flows into the adsorber and more butane is adsorbed in the carbon, which yields higher purity of butane for production. However, the recovery decreasing with feed concentration increasing, due to more adsorbed butane desorbs and vents at cocurrent depressurization for butane enrichment. Figure 6 shows the pressure profile of adsorber on different feed molar fraction of butane with time. In Figure 6, the adsorber pressure for the butane concentration of feed ,butane/air = 0.5/0.5 and butane/air = 0.25/0.75, are 0.867atm and 0.612atm at cocurrent depressurization end ($t=18\text{sec}$), respectively. The reason is that high concentration feed make more butane adsorbed on carbon and the adsorbed butane can not desorb well to depressurize the pressure of adsorber at the same step time.

3.3 Influence of Feed Pressure

The effect of the feed pressure on butane purity and recovery is shown in Figure 7. While maintaining the same operating cycle time, increasing the feed pressure will lead more butane to adsorb on carbon and flow through adsorber. Besides, the high adsorber pressure makes an inefficient depressurization step for butane desorption and production at same step time, which results in butane purity and recovery decreasing. Figure 8 shows the pressure profile of adsorber on different feed pressure with time. After adsorption step (9sec), the high feed pressure causes high adsorber pressure, which makes it inefficient to depressurize to a lower pressure than low feed pressure.

This explains the previous results of butane purity and recovery decreasing with increasing feed pressure.

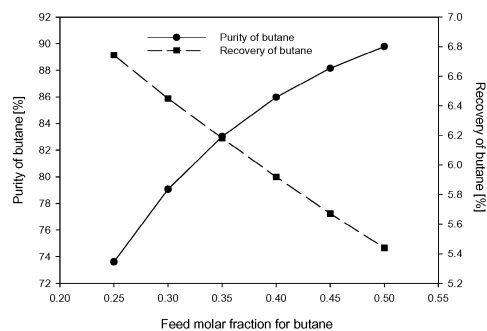


Figure 5 Effect of the feed molar fraction of butane on butane purity and recovery for $\epsilon_m = 0.37$.

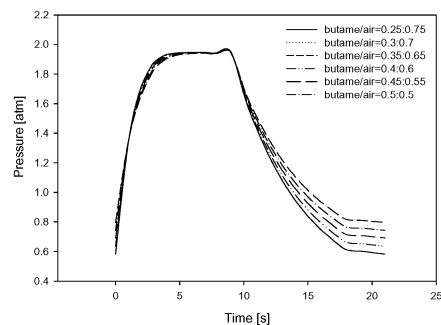


Figure 6 The pressure profile of the bed with time on different feed molar fraction of butane for $\epsilon_m = 0.37$

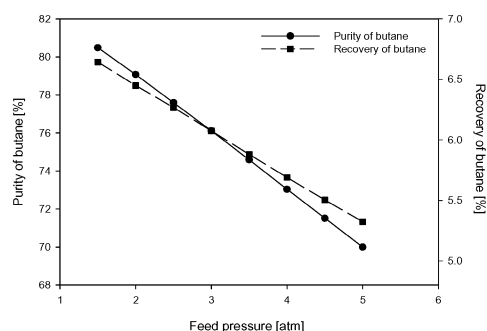


Figure 7 Effect of the feed pressure on butane purity and recovery for $\epsilon_m = 0.37$.

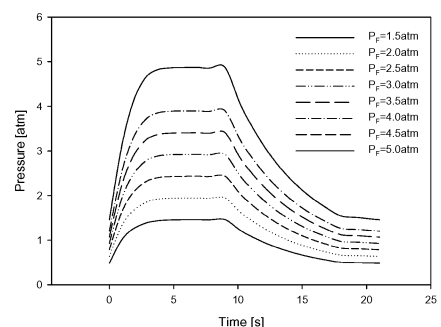


Figure 8 The pressure profile of the bed with time for different feed pressure for $\epsilon_m = 0.37$.

4. Conclusion

In our simulation study, the three-step PSA process above can be used to separate the more strongly adsorbed component butane as product from a feed gas of 30% butane and 70% air on carbon-coated ceramic monolithic adsorber. Increasing the thickness of carbon coated layer can increase the butane purity and recovery at the same valve value operation, but increasing the feed pressure will decrease the butane purity and recovery.

5. References

- Chou, C.T. and W.C. Huang, 1994, Chem. Eng. Sci., 49, 75.
 Ruthven, D.M. and C. Tharon, 1996, Gas. Sep. Purif. 10, 63.
 Valdes-Solis, T., M.J.G. Linders, F. Kapteijn, G. Marban and A.B. Fuertes, 2004, Chem. Eng. Sci., 59, 2791.
 Yu, F.D., L.A. Luo and G. Grevillot, 2002, J. Chem. Eng. Data, 47, 467.