Separation Science and Technology
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713708471

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To cite this Article Lu, Xiaochun , Rothstein, Daniel , Madey, Richard and Huang, Jan-Chan(1987) 'Pressure Swing Adsorption for a System with a Freundlich Isotherm', Separation Science and Technology, 22: 6, 1547 — 1556
To link to this Article: DOI: 10.1080/01496398708058417
URL: http://dx.doi.org/10.1080/01496398708058417

SEPARATION SCIENCE AND TECHNOLOGY, 22(6), pp. 1547-1556, 1987

Pressure Swing Adsorption for a System with a Freundlich Isotherm

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Abstract

An analytical expression is obtained for the equilibrium relationship between the enrichment factor of the product stream and the ratio of the pressure in the feed stream to that in the product stream for a pressure-swing-adsorption system with a Freundlich isotherm. The enrichment factor is the ratio of the mole fraction of the adsorbate in the product gas stream to that in the feed gas stream. The enrichment factor increases with increasing pressure ratio in a manner similar to that for a system with a linear isotherm. The nonlinearity of the Freundlich isotherm does not result in a significant reduction in the enrichment factor.

INTRODUCTION

Since Skarstrom (*1*) introduced the heatless adsorption cycle in 1959, several modifications on pressure-swing-adsorption (PSA) processes have been developed and commercialized. Many studies of the PSA process in the literature (*2–6*) assume that the system obeys a linear isotherm. Recently some computer simulation studies were carried out for Langmuir (*7, 8*) and Freundlich (*5, 9*) systems.

The purpose of this study is to examine the PSA process with a nonlinear Freundlich isotherm, and to compare the relationship between the enrichment factor and the ratio of the pressure in the feed stream to that in the product stream for this nonlinear system with that for a linear system. The enrichment factor is defined as the ratio of the mole fraction of the adsorbate in the product gas stream to that in the feed gas stream. An analytical expression for this relationship is derived on the basis of an equilibrium model and the mass-balance equation.

PROCESS DESCRIPTION

The basic pressure-swing-adsorption cycle involves four distinct steps, as shown in Fig. 1 for a PSA system with two packed beds. During Step 1,

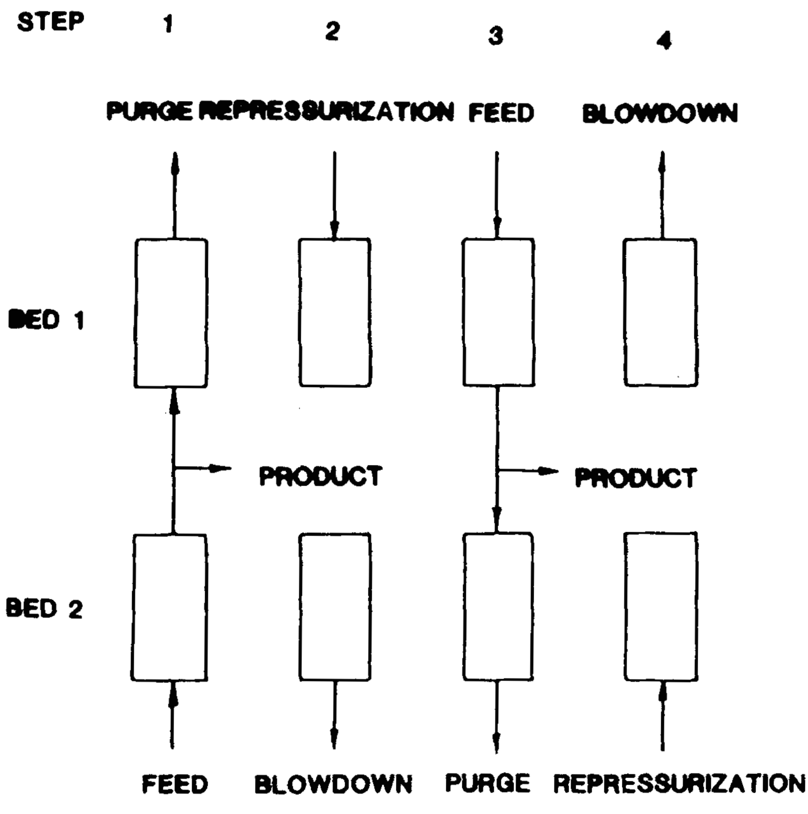

FIG. 1. Steps involved in a pressure-swing adsorption cycle.

a high-pressure feed gas is introduced into Bed 2, where adsorption takes place. A small fraction of the product gas is reduced in pressure and used to purge Bed 1. In Step 2, Bed 1 is pressurized with feed gas while Bed 2 is subjected to a pressure reduction (blowdown). The blowdown causes the desorption of some of the adsorbate which is removed during the purge step. The same cycle is repeated in Steps 3 and 4 with high-pressure flow and adsorption occurring in Bed 1 and purging occurring in Bed 2.

MATHEMATICAL MODEL

When the packed bed is held at constant temperature, and the longitudinal and radial dispersion and solid-phase diffusion are negligible, the mass-balance equation for the adsorbate flowing through a packed bed is (*2*)

$$\varepsilon[\partial C/\partial t + \partial(uC)/\partial z] + (1 - \varepsilon)\partial q/\partial t = 0 \tag{1}$$

where C is the concentration of the adsorbate in the gas phase, q is the concentration of the adsorbate in the solid phase, u is the interstitial velocity, ε is the external void fraction of the bed, t is time, and z is position along the bed.

For an ideal gas undergoing an isothermal process, the continuity equation for the carrier gas may be written (*2*)

$$\partial P/\partial t + \partial(uP)/\partial z = 0 \tag{2}$$

where P is the total gas pressure in the bed. Equation (2) assumes that the concentration of the adsorbate gas is small compared to that of the carrier.

The Freundlich isotherm for the adsorbate gas relates the adsorbed phase concentration q to the adsorbate partial pressure p in the gas phase:

$$q = kp^m \qquad (0 \leqslant m \leqslant 1) \tag{3}$$

Here the Freundlich coefficient k is proportional to the dimensionless adsorption capacity when $m = 1$.

The partial differential Equation (1) may be rewritten in terms of the Freundlich isotherm and the mole fraction ($y = p/P$) of the adsorbate in the gas phase:

$$\varepsilon[\partial(yP)/\partial t + \partial(uyP)/\partial z] + kRT(1 - \varepsilon)\partial(yP)^m/\partial t = 0 \tag{4}$$

SOLUTION OF THE PARTIAL DIFFERENTIAL EQUATION

Carrying out the differentiation in Eq. (4) and employing Eq. (2), we obtain

$$\varepsilon P[\partial y/\partial t + u\partial y/\partial z] + kRT(1 - \varepsilon)m(yP)^{m-1}\partial(yP)/\partial t = 0 \tag{5}$$

or

$$[\varepsilon + kRT(1 - \varepsilon)m(yP)^{m-1}]\partial y/\partial t + \varepsilon u\partial y/\partial z + [ykRT(1 - \varepsilon)m(yP)^{m-1}]\partial \ln P/\partial t = 0 \tag{6}$$

Equation (6) is a quasi-linear first-order partial-differential equation which can be solved by the method of characteristics (*10*). This method yields the following pairs of ordinary differential equations:

$$dz/[\varepsilon u] = dt/[\varepsilon + kRT(1 - \varepsilon)m(yP)^{m-1}] = -dy/\{ykRT(1 - \varepsilon)m(yP)^{m-1}[d \ln P/dt]\} \tag{7}$$

In order to obtain a relationship between the adsorbate gas mole fraction y and the total pressure P in the PSA process, we consider the equation given by the second equality in Eq. (7):

$$-dy/dt = (d \ln P/dt)ykRT(1 - \varepsilon)m(yP)^{m-1}/[\varepsilon + kRT(1 - \varepsilon)m(yP)^{m-1}] \tag{8}$$

or

$$-d \ln P/d \ln y = 1 + \varepsilon(yP)^{1-m}/kRT(1 - \varepsilon)m \tag{9}$$

Introduce dimensionless variables:

$$P^* = P/P_H \tag{10}$$

$$Y = y/y_H \tag{11}$$

Here the subscript H refers to quantities at high pressure in the feed stream. Similarly, let the subscript L denote quantities at low pressure in the product stream; then when $P = P_L$, $P^* = P_L/P_H = P_0^*$ and $Y = y_L/y_H = Y_0$ since $y = y_L$. The quantity P_0^* is the reciprocal of the pressure ratio and Y_0 is the enrichment factor of the product stream. Equation (9) can be rewritten in terms of these dimensionless variables:

$$-d \ln P^*/d \ln Y = 1 + \varepsilon(yP^*)^{1-m}(y_H P_H)^{1-m}/(1 - \varepsilon)kRTm \quad (12)$$

or

$$-d \ln P^*/d \ln Y = 1 + \beta_m(YP^*)^{1-m} \quad (13)$$

where

$$\beta_m = \varepsilon(y_H P_H)^{1-m}/(1 - \varepsilon)kRTm = \varepsilon(y_H P_H/q_H)/(1 - \varepsilon)RTm \quad (14)$$

The right-hand equality follows from the Freundlich isotherm, Eq. (3). Let

$$\alpha = \varepsilon/(1 - \varepsilon) \quad (15)$$

Then,

$$\beta_m = \alpha/K_H m \quad (16)$$

where K_H is the dimensionless capacity evaluated at the high pressure of the feed stream. The dimensionless adsorption capacity K is the ratio of the adsorbed-phase concentration to the gas-phase concentration at equilibrium.

Integrating Eq. (13) between steps of high-pressure feed and low-pressure purge, we can obtain a relationship between the enrichment factor and the pressure ratio for the following two cases.

(a) Linear Case (m = 1)

Equation (13) becomes

$$-d \ln P^*/d \ln Y = 1 + \beta_m \quad (17)$$

The solution is:

$$P_0^* = Y_0^{-(1+\beta_m)} = Y_0^{-[1+\alpha/K_H]} \tag{18}$$

Equation (18) is the result of Shendalman and Mitchell (*2*) for a linear isotherm.

(b) Nonlinear Case ($m < 1$)

In order to solve Eq. (13), let

$$V = P^* Y \tag{19}$$

Then Eq. (13) becomes

$$-dV/V^{2-m} = \beta_m d \ln Y \tag{20}$$

Integrate Eq. (20) to obtain

$$-V/(m-1)^{m-1} = \beta_m \ln Y + c \tag{21}$$

where c is an integration constant. Since $P = 1$ when $Y = 1$, the integration constant must be $1/(1 - m)$; hence, Eq. (21) becomes

$$(1 - m)\beta_m \ln Y = (YP^*)^{m-1} - 1 \tag{22}$$

Also, since $Y = Y_0$ when $P^* = P_0^*$, Eq. (22) yields the following relation between Y_0 and P_0^* for a PSA system with a Freundlich isotherm:

$$(1 - m)\beta_m \ln Y_0 = (Y_0 P_0^*)^{m-1} - 1 \tag{23}$$

DISCUSSION

The enrichment factor versus the pressure ratio is plotted in Fig. 2 for five values of β_m (viz., β_m = 0, 0.01, 0.05, 0.1, and 1.0) for a system with a linear isotherm (i.e., m = 1.0). For β_m = 0, the linear isotherm gives an enrichment factor of 10 for a pressure ratio of 10. The enrichment factor versus the pressure ratio is plotted in Fig. 3 for the same values of β_m but with a Freundlich exponent of m = 0.5. For all five curves the enrichment factor increases monotonically as the pressure ratio increases. The effect of the Freundlich isotherm in the PSA process is to reduce the enrichment factor in comparison with that for a linear isotherm. The

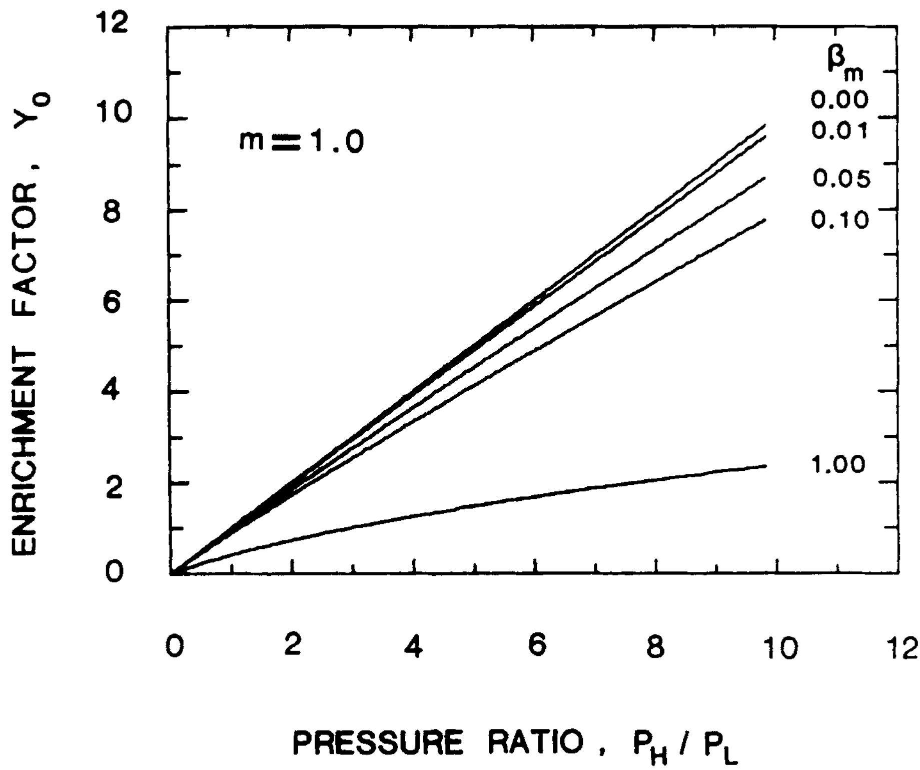

FIG. 2. The enrichment factor Y_0 versus the pressure ratio P_0^* in a pressure-swing-adsorption system with a linear isotherm (i.e., $m = 1$) for five values of the parameter β_m.

enrichment factor decreases as the value of the Freundlich exponent m decreases. The decrease in the enrichment factor becomes larger with increasing values of the parameter β_m. The enrichment factor is decreased at a large value of β_m for both $m = 1$ and $m = 0.5$; however, in order to see a significant reduction, the value of β_m has to be higher than 0.1. The magnitude of the parameter β_m is on the order of $1/K_H$, which is the volume of the bed divided by the volume of the gas processed. Since it is unlikely for a bed to process less than 10 times the bed volume before the desorption cycle, the enrichment factor is hardly effected by the values of β_m or m. Listed in Table 1 are the enrichment factors for the adsorption of ethane, propane, and n-butane on a bed packed with porous polystyrene absorbent at 298 K calculated with the parameters from Rothstein et al. (*11*).

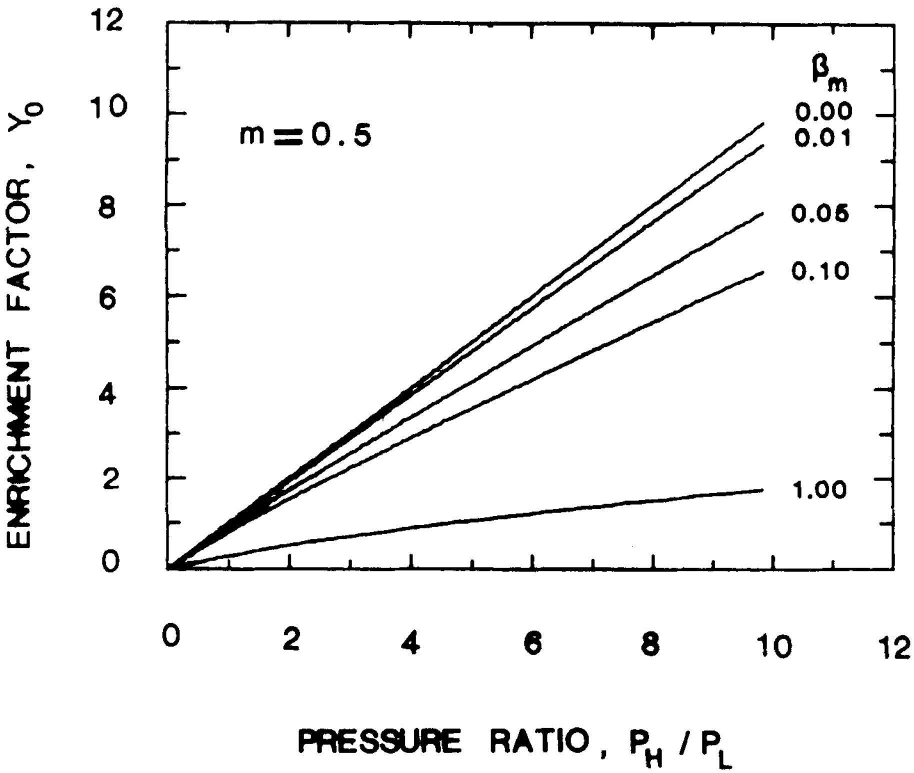

FIG. 3. The enrichment factor Y_0 versus the pressure ratio P_0^* in a pressure-swing-adsorption system with a Freundlich isotherm with an exponent $m = 0.5$ for five values of the parameter β_m.

TABLE 1
Enrichment Factors and Freundlich Isotherm Parameters for Three Gases Adsorbed at 298 K on a Porous Polystyrene Adsorber Bed with a Void Fraction $\varepsilon = 0.35$

Gas	Dimensionless adsorption capacity, K	Freundlich exponent, m	Enrichment factor Y_0 at $P_H/P_L = 10$
Ethane	57.6	0.983	9.68
Propane	135	0.864	9.91
n-Butane	434	0.652	9.96

CONCLUSION

We examined the pressure-swing-adsorption process for systems with Freundlich isotherms and obtained an analytical relation between the enrichment factor of the product stream and the ratio of the pressure in the feed stream to that in the purge stream. The nonlinearity of the isotherm does not result in a significant reduction of the enrichment factor.

SYMBOLS

C	gas-phase concentration of adsorbate (mol/cm^3)
k	Freundlich coefficient
K	dimensionless adsorption capacity ($= q/C$)
m	Freundlich exponent
p	partial pressure of adsorbate gas (mmHg)
P	total pressure of the gas (mmHg)
P_0^*	($= P_L/P_H$), inverse of pressure ratio
P_H	high pressure, in feed gas stream (mmHg)
P_L	low pressure, in product stream (mmHg)
q	solid-phase concentration of adsorbate (mol/cm^3)
t	time (s)
u	interstitial velocity (cm/s)
y	gas-phase mole fraction of adsorbate
Y	$= y/y_H$
y_H	gas-phase mole fraction of adsorbate in the high-pressure feed stream
y_L	gas-phase mole fraction of adsorbate in the low-pressure product stream
Y_0	enrichment factor of product stream ($= y_L/y_H$)
z	axial position in bed (cm)
α	constant defined by Eq. (15)
β_m	constant defined by Eq. (16)
ε	void fraction of adsorber bed

Acknowledgment

This work was supported in part by the Division of Chemical Sciences, Office of Basic Energy Sciences, Department of Energy.

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Received by editor September 15, 1986
Revised March 26, 1987