and higher flow rates, for which adsorption rate limitations may become significant. Second, unless the permeability is constant, the optimal timing of the control sequence will vary and unless corrections for this are made in the control, suboptimal operation will result. To avoid this problem, adsorbent particles that resist abrasion and maintain a constant flow resistance should be used. The round particles used in this research were found to be satisfactory.

Another factor that affects the operation of the adsorber is the length of the column. Unlike most chemical process equipment, decreased length increases the capacity of this system (within certain limits). To achieve the same product composition for shorter lengths, higher frequencies are required. Since the optimal frequency increases as the inverse of the square of the length (Kowler, 1969), shorter lengths would require faster operation. This result is valid for flow rates for which adsorption rate limitations can be neglected. In fact, since the optimal frequency increases so quickly as length decreases, the performance of the controlling solenoid valves may limit the achievement of the optimal frequency for shorter lengths. Despite these limitations, it is clear that attempts should be made to use shorter lengths of column to increase capacity and decrease equipment costs at the same time.

Having found (theoretically) the optimal feed boundary cyclic control of (maximum pressure, zero flow, minimum pressure) and having gained a better understanding of the design parameters, a cyclic adsorption system can now be more properly designed. For the separation of gas mixtures for which there exists an adsorbent with a high relative volatility, the cyclic adsorption process may well be of commercial value.

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Part II. Theory

The fixed bed binary gas adsorber, when alternately fed and exhausted at one end, produces a purified product from the other end. Coupled partial differential equations in pressure and composition, representing total mass and component balances with local equilibrium, describe the operation. The Maximum Principle is applied to determine the optimal cyclic unsteadv feed policy for the balanced objectives of product purity and quantity. The sequence (maximum feed, no flow, maximum exhaust) is optimal. The experimental optimum is close to the calculated optimum. Dimensional analysis is used to determine parametric effects.

This cyclic adsorption device represents an example of a distributed parameter feed-driven unsteady process. Experimental work has shown that there exist both an optimal frequency and an optimal feed pressure program for achieving the goal of separation of a binary gas mixture.

This paper will concern itself with the development of the model for the system, application of optimal control theory, and the numerical solution for the optimal feed boundary pressure cycle. The details and results of the experimental study of this system have been presented in Part I.

MATHEMATICAL MODEL

We consider first the development of a mathematical model of the molecular sieve adsorber and the formulation of necessary conditions for optimal control. The state variables of the system and the adjoint variables of the control problem are governed by partial differential equations. Because of the complexity of these equations and of the computational procedures involved, the computer cost for a finely spaced finite difference solution of these equations is excessive. Therefore a lumped-parameter model (cell model) is also developed. A pictorial model is shown in Figure 1. Feed and exhaust can either alternate or they can alternate with intermediate shut-off periods.

In establishing the bases for a model for the molecular sieve bed, Turnock and Kadlec (1971) made the following assumptions and approximations:

1. Ideal gas behavior
2. Darcy's Law representation of the gas flow
3. Viscosity of the gas phase is composition invariant
4. Plug flow conditions
5. At any instant, equilibrium exists between the gas phase and the adsorbed phase.
6. The effect of the heat of adsorption on the temperature profile will be neglected; isothermal operation is assumed.

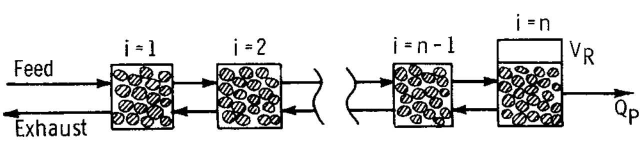

Fig. 1. Model for the cell system.

TABLE 1. MATHEMATICAL MODEL FOR THE ADSORPTION SYSTEM

In the following equations,

z = (absolute pressure)^2 in adsorption bed
y = mole fraction N_2 in adsorption bed
z_R = (absolute pressure)^2 in flow regulator
y_R = mole fraction N_2 in flow regulator

State equations:

Adsorption bed:

$$\dot{z} = F_1 = \frac{a_2 z^{1/2} z_{\lambda\lambda}}{(a_1 + a_3 z^{(\gamma-1)/2})}$$

$$\dot{y} = F_2 = \frac{a_2 z_\lambda y_\lambda - \dfrac{a_2 z_\lambda y_\lambda \, [a_3 z^{(\gamma-1)/2} y(1 - y(1-\gamma))]}{a_1 + a_3 z^{(\gamma-1)/2})\,(y + \alpha(1-y))}}{2z^{1/2} D}$$

Flow regulator at $\lambda = L$:

$$\dot{z}_R = G_1 = \frac{z_R^{1/2}}{V_R}(a_2 z_\lambda + a_4 Q_P)$$

when $z_\lambda(L, t) < 0$, $\quad \dot{y}_R = G_2 = \dfrac{-a_2 z_\lambda (y - y_R)}{2z^{1/2} V_R}$

when $z_\lambda(L, t) \geqq 0$, $\quad \dot{y}_R = 0$

Time considerations:

$$z(\lambda, t_0) = z(\lambda, t_0 + \tau)$$
$$y(\lambda, t_0) = y(\lambda, t_0 + \tau)$$
$$z_R(t_0) = z_R(t_0 + \tau)$$
$$y_R(t_0) = y_R(t_0 + \tau)$$

Boundary conditions:

$$\lambda = 0$$

when $z_\lambda(0, t) < 0$, $\quad y(0, t) = y_F$
$z(0, t)$ controlled

$$\lambda = L$$

when $z_\lambda(L, t) < 0$, $\quad y(L, t) = y_R(t)$
$z(L, t) = z_R(t)$

For the above equations,

$$a_1 = \epsilon A, \quad a_2 = \frac{AK}{\mu} \quad \text{and} \quad a_3 = \frac{WRTk\gamma}{L}$$

$$a_4 = 2RT$$

$$D = \left(a_1 + \frac{\alpha a_3 z^{(\gamma-)1/2}}{\gamma(y + \alpha(1-y))^2} \right)$$

Based upon Lederman's (1961) studies of methane-nitrogen adsorption on Linde molecular sieve type 5A, the following observations were made:

7. The total equilibrium amount adsorbed is independent of composition.

8. The equilibrium adsorption isotherms are fit well by the Freundlich relationship:

$$N = kWP^\gamma \tag{1}$$

9. The relative volatility α relates the relative adsorption of the two gases components and does not vary with composition.

$$\alpha = \frac{y/x}{(1-y)/(1-x)} \tag{2}$$

Before deriving the state equations for the system, a brief look into the validity and importance of some of the above relations will be made.

Since Davison 5A molecular sieve was used in this study, the adsorption equilibria of methane and nitrogen on this sieve were investigated experimentally. It was found that relation 7 is not strictly valid. However, the composition during operation varies over a composition range for which it is a reasonable approximation to use an average total adsorption independent of composition. This approximation is an important factor in uncoupling the pressure equation from the composition equation. This greatly simplifies the numerical solutions of the system equations and the optimal control problems, and was found to produce reasonable results.

Relations 3 and 9 are also needed to uncouple the pressure equation. The relative volatility was found experimentally to be constant over the range of operating conditions. The gas viscosity does vary with composition, but the range is not wide so that an average will be used.

Since the rate of adsorption is rapid, instantaneous equilibrium is assumed as in relation 5. This then eliminates rate considerations from the model and places an attendant restriction on its region of validity.

For any given feed boundary pressure control and product flow rate, the preceding model bases yield the differential equations, time conditions, and boundary conditions which are compiled in Table 1. For a desired product capacity, the performance of the adsorption system is directly related to this feed boundary pressure control. It is then necessary to establish the conditions necessary to find the control that optimizes the performance of the adsorber.

In the formulation of the cell model, the column and adsorbent are equally divided into n segments, each one of which behaves like an ideally mixed stage, as shown in Figure 1. To describe this cell system, the following additional assumptions are made:

1. Within each ideally mixed stage the gas and adsorbed phases are in equilibrium.
2. The pressure drop between cells is caused by a Darcy's Law pressure drop across the adsorbent.
3. The pressure of the gas flowing between cells is taken as the average of the cell pressures.
4. The last cell includes the volume of the product line preceeding the constant product flow rate controller.

THEORY

With ordinary differential equations entering the boundary conditions at $\lambda = L$ and the control entering at the $\lambda = 0$ boundary, the method for setting up the necessary conditions as proposed by Katz (1964) or the conditions presented by Egorov (1964, 1966) are inadequate. Following the variational approach used by Denn (1966), the necessary conditions for optimality in the adsorption system can be formulated.

To simplify the development, the state vectors $\vec{w}$ and $\vec{r}$ will be defined.

$$\vec{w}(\lambda, t) = \begin{bmatrix} z(\lambda, t) \\ y(\lambda, t) \end{bmatrix} \quad \vec{r}(t) = \begin{bmatrix} z_R(t) \\ y_R(t) \end{bmatrix} \tag{3}$$

Thus, the state equations presented in Table 1 take the form

$$\begin{aligned} \dot{\vec{w}} &= F(\vec{w}, \vec{w}_\lambda, \vec{w}_{\lambda\lambda}) \qquad 0 < \lambda < L \\ \dot{\vec{r}} &= G(\vec{r}, \vec{w}, \vec{w}_\lambda) \qquad \lambda = L \end{aligned} \tag{4}$$

The distributed system, containing a boundary control u, is optimized by maximizing the value of a performance index I. This index is an average, over a steady state*

* The words "steady state" are here taken to mean a repetitive process which does not change from cycle to cycle.

cycle, of functions of the state and control variables. For this adsorption system, the values taken by the state variables within the bed have no direct significance. The only terms of interest in the performance index will depend upon the product composition and the rate of exhaust from the system. Thus, the following performance index will be considered:

$$I = \frac{1}{\tau} \int_{t_0}^{t_0+\tau} [m_1(z_\lambda)_{\lambda=0} + m_2(y_R)]\, dt \qquad (5)$$

The value that the above index can achieve is constrained by Equation (4). The control u, applied at the $\lambda = 0$ boundary, is constrained by the maximum available feed gas pressure and atmospheric pressure so that

$$\text{At } \lambda = 0, \quad 1 \leq z \leq z_{\max} \qquad (6)$$

In addition, the control is required to be periodic piecewise continuous with discontinuities only at a finite number of points.

The differential equation constraints (4) are introduced into the performance index with the use of the adjoint variables, that is, the set of Lagrange multipliers. The existence of these adjoint variables, although not guaranteed, will be assumed and they will be denoted by

$$\vec{\rho}(\lambda, t) = \begin{bmatrix} \rho_1(\lambda, t) \\ \rho_2(\lambda, t) \end{bmatrix} \text{ and } \vec{\eta}(t) = \begin{bmatrix} \eta_1(t) \\ \eta_2(t) \end{bmatrix} \qquad (7)$$

Entering the differential equation constraints into the performance index for an arbitrary value of B,

$$I = \frac{1}{\tau} \int_{t_0}^{t_0+\tau} \Big\{ [m_1(z_\lambda)]_{\lambda=0} - \eta^T (\vec{r} - G(\vec{r}, [\vec{w}, \vec{w}_\lambda]_{\lambda=L})) + m_2(y_R) - B\int_0^L \vec{\rho}^{\,T} (\vec{w} - F(\vec{w}, \vec{w}_\lambda, \vec{w}_{\lambda\lambda}))\, d\lambda \Big\}\, dt \qquad (8)$$

The control is chosen as

$$u = [z_\lambda]_{\lambda=0} \qquad (9)$$

Application of a variational procedure, with τ constant, produces a set of differential equations which govern the behavior of the adjoint variables. These necessary conditions may be found elsewhere (Kowler, 1969). Major simplification of these complex equations was possible by letting $\lambda = 1.0$. Since a narrow pressure range is used in this work and the experimental value for γ is 0.87, the approximation using $\lambda = 1.0$ is reasonable.

For convenience, the function H_0 is defined such that

$$H_0 = m_1(u) - \left[B\rho^T \frac{\partial F}{\partial \vec{w}_{\lambda\lambda}} \right]_{\lambda=0} \cdot u \qquad (10)$$

The optimal choice of $u(t)$, which is denoted by u^*, is composed of subarcs which will hereafter be referred to as the optimal control components. For an optimal system where the adjoint variables are described by the necessary conditions, the following maximum principle must be satisfied:

> Except at a finite number of points, the function H_0, the feed boundary Hamiltonian, is made stationary with respect to the components of u^* which lie in the interior of its admissible region. The Hamiltonian is made a maximum with respect to the components of u^* which lie at the boundary of the admissible region.

Note that the optimal control satisfying the above maximum principle is not necessarily a unique extremal nor is the existence of such an optimal control assured.

The maximum principle can be used to predict possible forms of the optimal control through examination of the behavior of the Hamiltonian. Written in terms of ρ_1, ρ_2, a, and y,

$$H_0 = m_1(u) - u \left[\frac{B\, a_2}{(a_1 + a_3)} (\rho_1 z^{1/2} - \rho_2) \frac{a_3 y(1 - y(1-\alpha))}{2z^{1/2} D(y + \alpha(1-y))} \right]_{\lambda=0} \qquad (11)$$

For gas entering the system $u < 0$, the behavior of ρ_2 is described by a hyperbolic partial differential equation. When flow direction changes and leaves the system at $\lambda = 0$ ($u > 0$), there is no boundary condition for composition which requires that ρ_2 be immediately constrained by the boundary condition $\rho_2 = 0$. This would appear to lead to a discontinuous behavior for $\partial H_0/\partial u$ and would occur only in switching from $u < 0$ to $u > 0$.

In switching from $u > 0$ to $u < 0$, initially $\rho_2 = 0$. This adjoint variable must then change value continuously as described by its partial differential equation and thus no discontinuity in ρ_2 would result from a switch in this direction. For the case where exhaust flow is to be minimized, $\partial m_1/\partial u$ has a value only when flow leaves the system, $u > 0$. This appears to increase still further any discontinuous behavior of $\partial H_0/\partial u$ in switching from $u < 0$ to $u > 0$ and would create a discontinuity in switching from $u > 0$ to $u < 0$.

However, the Hamiltonian cannot exhibit discontinuities in its behavior. To fully examine its behavior, a specific performance index and the resulting Hamiltonian are introduced.

$$I = \frac{1}{\tau} \int_{t}^{t_0+\tau} (y_R - Cu)\, dt \qquad (12)$$

where $C = 0$ for $u < 0$.

With the exhaust rate proportional to u when $u > 0$, the magnitude of C determines the relative composition maximization. This leads to

$$H_0 = -u \left[\frac{Ba_2 z^{1/2}}{(a_1 + a_3)} \right] \left[\rho_1 + \begin{Bmatrix} M_0\rho_2, & u \leq 0 \\ C', & u > 0 \end{Bmatrix} \right] \qquad (13)$$

where

$$C' = \left(\frac{C(a_1 + a_3)}{Ba_2 z^{1/2}} \right)_{\lambda=0} \qquad (14)$$

$$M_0 = -\left[\frac{a_3 y(1-y)(1-\alpha)}{2zD\,(y+\alpha)(1-y)} \right]_{\lambda=0} \qquad (15)$$

Applying the maximum principle to the above Hamiltonian shows that u^* can be composed not only of the extreme values of the control ($u_{\max}$ and $u_{\min}$) but also of the control $u = 0$, as well as possible singular controls from the interior of the admissible region. In other words, the possible discontinuities in the Hamiltonian resulting from switches from $u > 0$ to $u < 0$ or from $u < 0$ to $u > 0$ give rise to the optimal control of $u^* = 0$ which eliminates the discontinuities. This control corresponds to one for which there is zero flow at the feed boundary. A graphical representation of the regions for applying these optimal components for values of $\rho(0, t)$ is presented in Figure 2.

For any situation where $\partial H_0/\partial u = 0$, it is possible that a control from the interior of the admissible region maximizes the Hamiltonian. This will arise if, for an interior control,

$$\rho_1 = -M_0\,\rho_2 \qquad (16)$$

occurs at more than a finite number of points. The optimal control is then referred to as a singular control.

At some time in the operation of this adsorption system the situation must exist when $u > 0$. Otherwise, with no exhaust flow, there could be no separation in steady state operation. From Figure 2 it can be seen that $u^* = u_{\max}$ is the only possible optimal control component for $u > 0$ unless $\rho_1(0, t) = 0$ for a singular control component. It is also noted that when $C' = 0$ (exhaust minimization is unimportant compared to product composition maximization), H_0 would exhibit the discontinuous behavior only in the switch from $u < 0$ to $u > 0$. Thus the component $u^* = 0$ can only occur, if at all, after $u^* < 0$ has been applied. Then if there are no singular controls, the form of the optimal sequence would be $(u_{\min}, 0, u_{\max})$ or (full feed pressure, zero flow, exhaust to minimum pressure).

Although at this point in the mathematical development the uniqueness of such an optimal control is not assured, knowledge of the behavior of the adsorption system leads to the conclusion that the existence of at least one such practical extremal is guaranteed. Specifically, if no control switching is made, there is no steady state separation. If infinitely fast switching is made between $u_{\max}$ and $u_{\min}$, the feed gas would almost completely bypass the bed and exhaust, except for a small unseparated product flow rate. Since it is known that steady state cyclic separations are possible, there must exist some maximum in product composition for a control switching sequence between no switching and infinite switching.

The model for the adsorption system previously derived leads to a very complex adjoint system. Therefore, in turning to the numerical solution of the problem, a simplified cell model approximation of the system was used.

Butkovskii (1961) has suggested the discretization of the distributed-parameter model so that Pontryagin's Maximum Principle can be used to derive the necessary conditions for optimal control (Pontryagin, 1962). These necessary conditions are well presented and applied in the text by Athans and Falb (1966). The following formulation will concern itself only with the system of interest.

For the cell model, the behavior of the system is described by

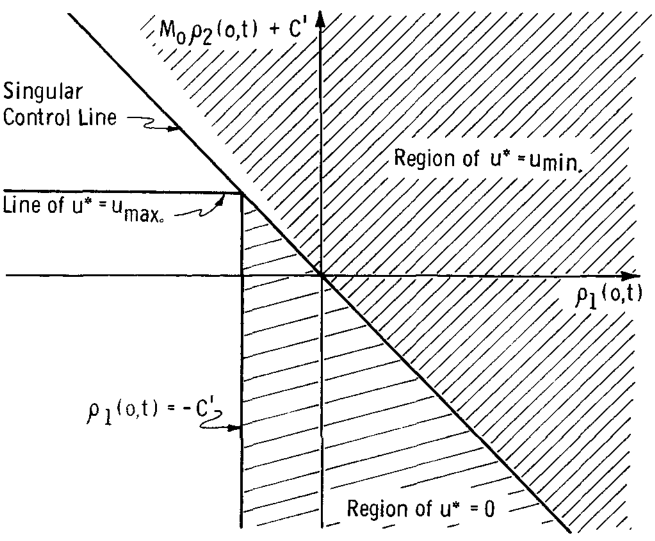

Fig. 2. Relation of optimal control components and adjoint variables at the control boundary for product composition maximization and exhaust minimization.

$$\dot{z}_i = f_i(z, u)$$
$$\dot{y}_i = g_i(z, y, u) \qquad (17)$$

where f_i and g_i are derived using the cell model approximations presented above. These state equations are the dynamical restraints on the performance index

$$I = \frac{1}{\tau}\int_{t_0}^{t_0+\tau} m_3(z_1, y_n, u)\,dt \qquad (18)$$

As in the variational approach, adjoint variables must be used to introduce these dynamical constraints into the performance index. Thus, $p_i(t)$ will be the adjoint variables associated with z_i and $q_i(t)$ will be the adjoint variables associated with y_i.

The Hamiltonian function is defined as

$$H = \sum_{i=1}^{n} (p_i f_i + q_i g_i) + m_3(z_1, y_n u) \qquad (19)$$

The maximum principle states that for an optimal control trajectory, the above Hamiltonian is maximized with respect to the control at all times provided that the adjoint variables are defined by

$$\dot{p}_i = -\sum_{j=1}^{n}\left(p_j\frac{\partial f_j}{\partial z_i} + q_i\frac{\partial g_i}{\partial z_i}\right) - \frac{\partial m_3}{\partial z_i} \qquad (20)$$

$$\dot{q}_i = -\sum_{j=1}^{n}\left(q_i\frac{\partial g_i}{\partial y_i}\right) - \frac{\partial m_3}{\partial y_i} \qquad (21)$$

The adjoint variables are also constrained by the time condition for periodic processes.

$$p_i(t_0) = p_i(t_0 + \tau) \text{ and } q_i(t_0) = q_i(t_0 + \tau) \qquad (22)$$

The maximized Hamiltonian, for the optimal system using the above definitions for the adjoint system, will be constant over the time period when the variable z_0 is treated as the control u'.

It has been shown (Kowler, 1969) that as the number of cells in the model increases, the state and adjoint equations will approach the mathematical description of the state and adjoint variables for the distributed-parameter model.

The preceding necessary conditions were based on a constant period τ. A necessary condition for the optimal period is now needed. Following the approach used by Horn and Lin (1967) the necessary condition for the optimum τ can be written for a time invariant Hamiltonian:

$$\frac{dI}{d\tau} = \frac{1}{\tau}\left[m_3(z_1, y_n, u) + \sum_{i=1}^{n}(p_i f_i + q_i g_i)\right]_{t_0+\tau} - \frac{1}{\tau} I \qquad (23)$$

With the use of the necessary conditions developed for optimality of the cell model, numerical computations for the optimal control can be made.

COMPUTATION OF OPTIMAL CONTROLS

In order to compute the optimal controls, a method suitable for use on the digital computer must be developed to find the solution to the necessary conditions for optimality. This method must not only provide a means for converging to the optimal control function from an initial assumed control function but must also provide solutions to the state and adjoint equations.

Because the state and adjoint variable time conditions require matching, a technique with minimum computation is needed to find the necessary set of conditions. As the mathematical properties of the equations describing these two systems of variables are different, separate techniques are used for the two systems. Horn and Lin (1967) have suggested the techniques to handle both. A general computational procedure is presented in Figure 3. Details of the computational methods within this procedure may be found elsewhere (Kowler, 1969).

The equation relating a change in performance index with a change in the control function is given by

$$\delta I = \frac{1}{\tau}\int_0^{t_0+\tau}\left[\frac{\partial H_0}{\partial u}\,\delta u\right]dt \tag{24}$$

This equation can be used to converge from an initial arbitrary control to the optimal control by improving the performance index until no further improvement is possible. Then where $u^+(t)$ is used as the initial control and $u^{++}(t)$ is the improved control

$$u^{++}(t) = u^+(t) + \phi\frac{\partial H_0}{\partial u}, \quad \phi > 0 \tag{25}$$

unless this violates the inequality constraint (6). In this case, $u^{++}(t)$ takes the value of the corresponding boundary of the admissible set of controls. Since second order and higher terms were not considered in deriving Equation (24), ϕ should be chosen small enough so that these neglected higher order terms will not be significant. With such a value of ϕ the integrand in Equation (24) becomes $\phi\left(\frac{\partial H_0}{\partial u}\right)^2$ which is always positive and thus improves the performance index I.

The number of equations to be integrated, per cycle of computation, is approximately four times the square of the number of cells used in the model. This rapid increase in computation and computer storage requirements was an important factor that led to the formulation of the cell model rather than directly discretizing the necessary conditions for the distributed-parameter model. Computations were made with a small number of cells. For the case where exhaust minimization was not considered ($C = 0$), the effect of the number of cells on the computed optimal control function was examined.

It was found that for this performance index, the cyclic control sequence of $[u'_{max}, z_1, u'_{min}]$ maximizes product composition. These controls are physically brought about with two valves: a feed valve and an exhaust valve. u'_{max} corresponds to opening only the feed valve, u'_{min} to opening only the exhaust valve, and $u = z_1$ to opening neither valve. The fractions of the period spent in the regions of $u'^* = z_1$, and $u'^* = u'_{max}$ are presented in Figures 4 and 5. It is evident from these figures that although the application of $u'^* = z_1$ decreased to a short interval, the fraction of the period for $u'^* = u'_{max}$, *FFVO*, does not change much for increasing n.

The number of cells used in the model has a great effect on the optimal period length τ^*. Figure 6 shows the

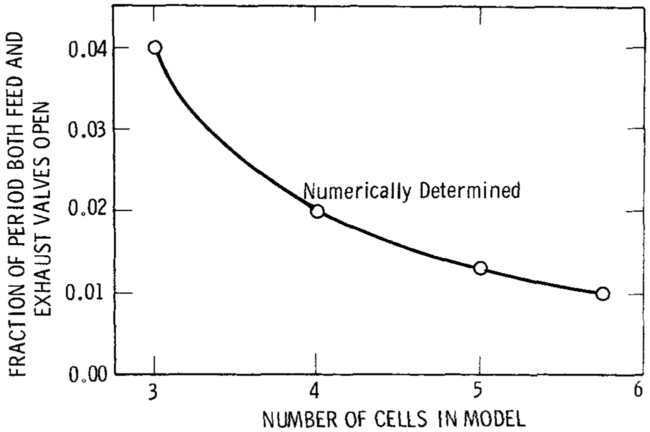

Fig. 4. Change of fraction of period with both valves closed as number of cells in model increases.

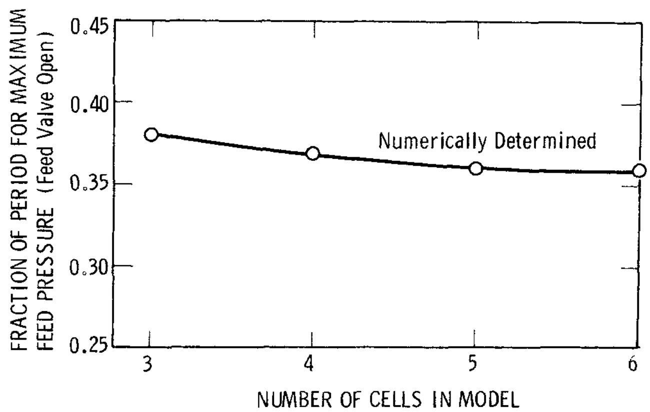

Fig. 5. Fraction of period with feed valve open versus number of cells in model.

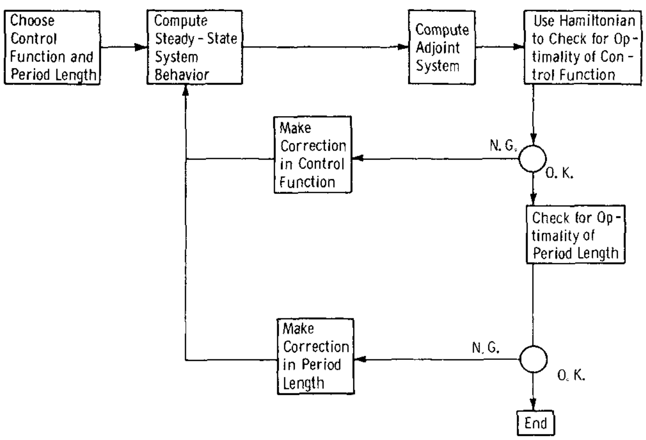

Fig. 3. Computational procedure.

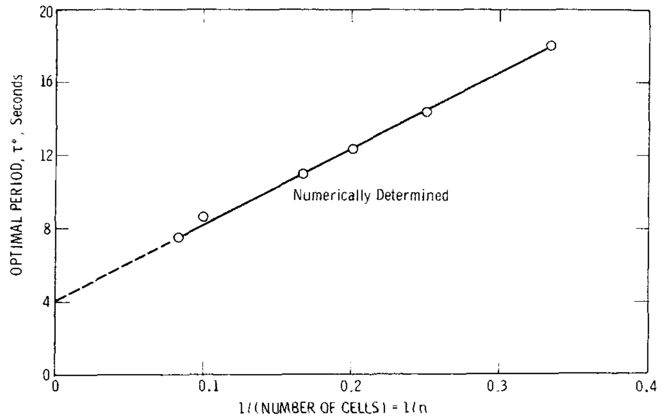

Fig. 6. Convergence of optimal period τ^* as number of cells in model increases.

change of τ^* with increasing n. By plotting τ^* versus $1/n$, as in Figure 6, the τ^* for $n = \infty$ can be predicted by extrapolation. The number of cells also greatly affects the

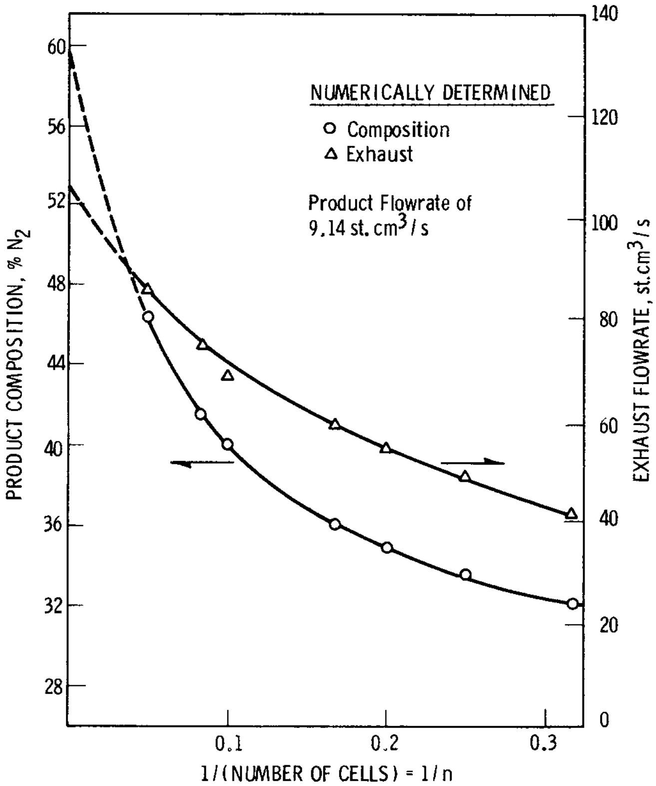

Fig. 7. Convergence of product composition and exhaust rate as number of cells in model increases.

computed system outputs, product composition, and exhaust rate. From Figure 7 it can be seen that a significantly larger number of cells is required for convergence of the system outputs than is required for the convergence of the form of the optimal control function. Thus, if interest is centered on the form of the optimal control function rather than the system outputs, a computational investigation can be limited to a small number of cells.

The influence on the optimal control form of the system constants and operating variables was investigated. These computations were carried out for the 4-cell model and the performance index of maximization of product composition. The results are presented in Table 2. Unless otherwise noted, the system constants shown at the top of the table were used with a product flowrate of 9.14 st.cm³/s and a maximum feed pressure of 168 kN/m².

A performance index which included both product composition and exhaust rate was then considered. The optimal control was computed for a few values of C. The effect on the optimal pressure control form of increased importance of the exhaust rate term was then demonstrated. These computed optimal controls for a 4-cell model are shown in Figure 8, along with the corresponding system outputs.

In each case the optimal pressure wave forms are computed to be the sequence $[u'_{max}, z_1, u'_{min}]$.

INTERPRETATION OF RESULTS

The computational results indicated that the optimal feed boundary control is the cyclic sequence (maximum pressure, zero flow, minimum pressure). These results were based on models containing 6 cells or less. Before it can be concluded that this control sequence would be optimal for a model with a large number of cells, the optimal trajectories should be examined further.

Table 2. Changes in the Computed Optimal Control Timing with Variations in the System Parameters for $n = 4$

Maximum pressure: 168 kN/m^2 (10 psig)

A = 3.45 cm³ $\quad W$ = .440 kg $\quad Q_P$ = 9.14 st. cm³/s (1.16 SCFH)

V_R = 40.0 cm³ $\quad Y_F$ = 28.6% $\quad L$ = 1.52 m

K = 100 μm² $\quad \mu$ = $1.75 \times 10^{-5}\ N \cdot s/m^2$ $\quad k = 0.155 \dfrac{\text{gmol adsorbed}}{\text{kg adsorbent}}$

α = 2.3 $\quad \epsilon = 0.623$

Unless otherwise noted, the parameters shown above were used.

Parameter varied	New value	Optimal control % of Period to be applied: Feed valve open	Optimal control % of Period to be applied: Both valves closed	Period τ^*	Product composition % N_2	Computed system outputs: Exhaust rate, st. cm³/s
None	—	37.0	2	14.3	33.57	48.7
α	1.65	36.5	2	14.5	31.33	48.5
α	2.95	37.5	2	14.3	35.37	48.9
k	0.169	37.0	2	15.2	33.66	48.0
ϵ	0.700	37.0	2	14.6	33.43	48.2
K	75.0	41.0	2	19.0	33.13	36.0
μ	1.27×10^{-5}	34.0	2	10.4	33.98	66.8
Y_F	0.200	37.5	2	14.3	24.00	48.9
V_R	70.0	37.0	2	14.5	33.61	48.5
V_R	500.0	41.0	2	>20.0	34.23	40.7
V_R**	5000.0	46.0	2	25.0	35.53	39.9
Q_P	4.72	29.0	2	14.8	34.28	45.7
Q_P	14.16	43.0	2	14.3	32.85	48.0
Q_P	18.88	47.0	2	14.3	32.39	46.3
P_{max}	138	21.0	1	15.0***	37.74	94.6

** Nonoptimal computation made to determine system outputs.
*** Nonoptimal τ.

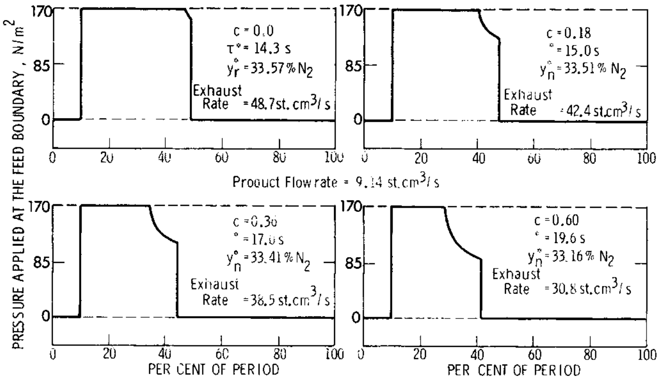

Fig. 8. Effect on 4-cell model optimal control form of minimizing exhaust as well as maximizing product composition.

For the 3, 4, 5, and 6 cell models, the optimal trajectory for product composition maximization passes through the optimal control regions for u'_{max}, z_1, and u'_{min}. From Figure 4 it appears that the fraction of the period that the trajectory spends in each of the control regions for large n can be predicted by extrapolation. However, the path that the trajectory takes between these control regions may exhibit different behavior as the number of cells is increased; a singular control subarc may become optimal. Although such a singular control component cannot be ruled out absolutely, it can be neglected practically; if the singular control does exist, it would be applied for such a small fraction of the period that its effect would be insignificant.

Experimental results presented in Part I have shown, as do the computational results in Table 2, that the optimal frequency does not noticeably vary with the product rate. $FFVO^*$ was determined and is compared with the computed results in Figure 9. Although the trend of an increase in $FFVO^*$ with increasing product flow rate is evident with both sets of data, there is a significant difference between absolute values. In addition, the experimentally determined optimal frequency of 0.35 cycles/second is significantly different from the value of 0.25 cycles/second based upon computational results. A dimensional analysis of this system is presented in the Appendix.

Exact agreement of results is not expected because of the many simplifying assumptions made in the construction of the distributed-parameter model and because of the approximate numerical solution of it.

CONCLUSIONS

With the use of a maximum principle for nonsingular controls, the optimal boundary control components are maximum pressure, zero flow rate, and minimum pressure. Without computational work, the optimal timing and sequence for the control components are unknown.

Because of the complexity of the distributed state and adjoint system, a cell approximation of the adsorption system was formulated. This model, made up of ordinary differential equations, may be used in conjunction with Pontryagin's Maximum Principle to locate the optimal feed boundary pressure controls.

Although it was found that the number of cells used greatly influences the optimal frequency, the timing of the optimal switching sequence within a dedimensionalized period length does not change significantly for cell models larger than the 4-cell model.

From experimental work it appears that applying the zero flow control for a short interval (<6% of the period) does not noticeably decrease the product composition although it does significantly reduce the exhaust rate and hence improves the performance of the adsorption system. Thus, it has been established that an optimal control other than bang-bang (maximum control-minimal control only) does not exist for this separation process.

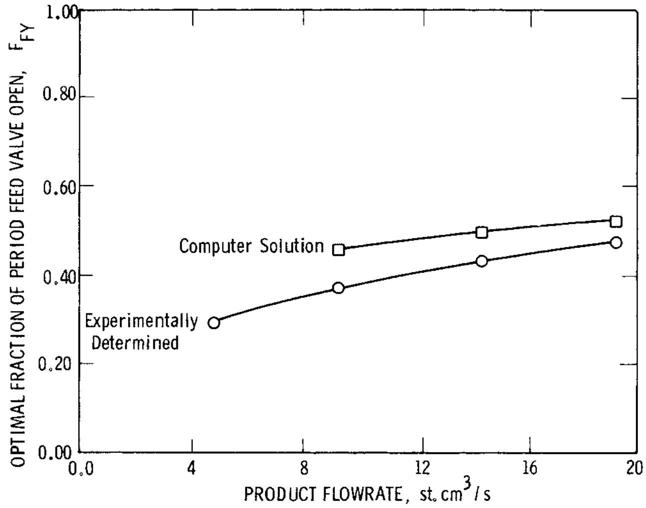

Fig. 9. Variation of optimal fraction of period feed valve open as product flowrate changes.

NOTATION

a_i = constants defined in Table 1
A = cross-sectional area of the adsorption column, cm^3
B = arbitrary factor in the variational equation
C = weighting factor for exhaust minimization
D = constant defined in Table 1
f_i = functions for z_i defined by Equation (17)
F_i = functions defined in Table 1
$FFVO$ = fraction of period that feed valve is open
g_i = functions for y_i defined by Equation (17)
G_i = functions defined in Table 1
H = Hamiltonian function
I = performance index
k = constant in the Freundlich adsorption isotherm
K = permeability of the packed adsorption column, μm^2
L = length of the adsorption column, m
m_i = functions making up the integral performance index
M_0 = group of terms defined by Equation (15)
n = number of stages in the cell model
N = amount of gas adsorbed, gmol
p_i = adjoint variable associated with the pressure variables
P = pressure, N/m^2
q_i = adjoint variable associated with the composition variables
Q_P = product flow rate, st.cm^3/s
r_i = state variables in the product line volume
R = ideal gas low constant
t = time variable, s
t_0 = initial time, s
T = temperature, K
u = controlled variable
V_R = volume of the product line preceding the pressure regulator, cm^3
w_i = state variables in the adsorption column
W = weight of adsorbent in column, g

x = composition of nitrogen in the adsorbed phase
y = composition of nitrogen in the gas phase
z = pressure^2 $(N/m^2)^2$

Greek Letters

α = relative volatility
γ = power constant in the Freundlich adsorption isotherm
δ = small variation of a state or control variable
ϵ = porosity of adsorption column
η = adjoint variables associated with pressure regulator variables
λ = distance variable, m
μ = average viscosity of gas flow stream, $N \cdot s/m^2$
ρ_i = adjoint variables associated with adsorption column distributed variables
τ = length of operating period, s
ϕ = constant which determines the magnitude of the correction of the control

Subscripts

F = relating to a property of the feed gas
i = one of a set
j = one of a set
max = upper limit of possible values for variable
min = lower limit of possible values for variable
0 = applicable at feed boundary of the column
R = relating to the product line between end of adsorption column and pressure regulator
λ = first partial derivative with respect to distance
$\lambda\lambda$ = second partial derivative with respect to distance

Superscripts

0 = first partial derivative with respect to time
$\rightarrow$ = vector quantity
T = transpose of a vector
$*$ = optimal value
$'$ = rescaled variable
$+$ = starting value
$++$ = corrected value

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APPENDIX. DIMENSIONAL ANALYSIS

A dimensional analysis of the state Equations (4) shows that if the dimensionless groups

$$\left[\frac{K\tau z_{\max}^{1/2}}{\mu L^2}\right], \left[\frac{2RTL\, Q_P \mu}{z_{\max} AK}\right], \left[\frac{V_R}{AL}\right],$$
$$\left[\frac{wRTk\gamma z_{\max}^{(\gamma-1)/2}}{AL}\right], \left[\frac{z_{\min}}{z_{\max}}\right]$$

and the dimensionless parameters ϵ, α, γ, and y_F are not changed, the solution to the dedimensionalized state equations will not change. This means that the optimal control which maximizes the product composition of the dimensionless state equations will not change form, other than the optimal period length τ^*, if the above dimensionless groups are kept constant. Thus, if (K/μ) is changed to $(K/\mu)'$, the optimal solution will be maintained if the optimal time constant τ^* changes by a factor of $(K/\mu)/(K/\mu)'$. In addition, Q_P must be changed by the inverse of this factor. This is confirmed by the computed results in Table 2, where the values of K and μ were varied. Since Q_P was not changed, the τ^* computed in both cases was changed by the factor $(K/\mu)/(K/\mu)'$ but the *FFVQ** corresponded to that of a product flow rate which was changed by the same factor $(K/\mu)/(K/\mu)'$. For any such solution of the dimensionless state equations where the above dimensionless groups are kept constant, the exhaust rate will change by the factor

$$\left[\frac{A\, L\, z_{\max}^{1/2}}{2\tau RT}\right]$$

Thus, a variation in K or μ by changing the optimal period length τ^* will change the exhaust rate. Since τ^* is changed by the factor $(K/\mu)/(K/\mu)'$, the exhaust rate will change by the inverse factor $(K/\mu)'/(K/\mu)$.

If the separation accomplished and the fraction of feed gas recovered as product were the only important factors in the operation of the system, it would appear that operation at lower product flow rate would result in the best performance. However, a factor not yet discussed is the capacity of the system. For example, in obtaining a recovered fraction of 0.09, a 56.6% N_2 product would be achieved for a product flow rate of 9.14 st.cm^3/5 whereas a product composition of only 55.8% N_2 would be achieved for a product flow rate of 14.16 st.cm^3/5. Although a smaller separation would be obtained for the latter operating condition, a 55% increase in capacity would result. Thus, in order to find the best operating conditions for the adsorption system, the capacity, as well as the fraction of feed recovered as product, needs to be considered.

Variation in permeability from K to K' will result in a change of τ^* by the factor (K/K'). If the product flow rate is also altered by this factor, the same separation will result and because the exhaust rate changes by the same factor, the fraction of the feed gas recovered as product will remain unchanged. It is then clear that the permeability directly affects the capacity of the system but not the relationship between the fraction of feed gas recovered and the separation achieved.

An increase in the length L of the adsorption column, unlike the behavior of most chemical process equipment, will result in a decrease in the capacity of the system. A change in length from L to L' will change τ^* by a factor of $(L'/L)^2$ and the same optimal control form and separation will result if the product flow rate is changed by a factor of (L/L'). Since the exhaust rate is also altered by the factor (L/L'), the fraction of feed gas recovered as product does not change.

Caution must be used in extending the above results. It appears that shorter lengths or higher permeabilities will favorably affect the performance of the system. However, since the frequency of the control is proportional to L^{-2} and K, the optimal frequency for shorter lengths or higher permeabilities may require faster operation than is attainable with solenoid valves. In addition, the increased flow rates will increase the significance of the rate limiting factors which have been neglected.

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