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The Optimal Control of a Periodic Adsorber:

DANIEL E. KOWLER and ROBERT H. KADLEC

Department of Chemical Engineering
The University of Michigan, Ann Arbor, Michigan 48104

Part I. Experiment

Cyclic pressure variations in a fixed bed adsorber can cause significant separation of gaseous mixtures. Feed pressure changes are the driving force in this parametric pumping process. The optimal feed sequence is maximum (maximum flow in) pressure, zero flow (variable pressure), maximum flow out (minimum pressure). For the nitrogen-methane feed gas at 168 k N/m^2 and a 1.22 m bed of adsorbent, the optimal cycle time is 3 seconds, and feed is sustained 50% of the time. The zero flow mode is unnecessary if product purity is the sole objective.

SCOPE

The components of a binary gas mixture exhibit different adsorption characteristics on a solid adsorbent such as a molecular sieve, the difference usually being expressed as a relative volatility. If such a mixture is steadily passed through a fixed bed of adsorbent particles, the adsorbed phase will adjust to a composition which is consistent with the equilibrium adsorption isotherms for the system for the given feed composition and the bed temperature. Under such a steady flow condition, no separation of the mixture occurs after the adsorbent is loaded.

If the feed pressure is alternated between values higher and lower than the bed entrance pressure, a feed flow will alternate with feed-end exhausting of the bed. This can be done while continuously withdrawing gas at the product end of the bed. If such cycling is continued until each cycle is like the previous, the product gas is enriched in the least strongly adsorbed component.

The system will not separate if run exclusively at the maximum feed rate or at the minimum (negative) feed rate, thus an optimum feed-exhaust policy must exist. Furthermore, if the cycle time is too short, the bed sees a mean inlet feed pressure, and no separation occurs. Likewise, an infinitely slow cycle time means steady operation, and again, no separation. Thus an optimum cycle time exists. The objectives of the optimization can be either product purity or capacity, or a combination of both.

The scope of Part I of this paper is the determination of the experimental optimum pressure wave form and cycle time for the methane-nitrogen-molecular sieve system. The theory of Part II indicates that the family of square waves encompasses the optima for a system in dynamic equilibrium. Therefore, the variables of cycle time, % of time at maximum feed pressure, no feed flow, and minimum (exhaust) feed pressure were experimentally explored. Other quantities, such as bed length and diameter, feed composition, temperature and molecular sieve parameters, were not varied.

The model of this system was previously developed for the instantaneous equilibrium assumption and is employed here. The theoretical optimization procedures were both an analysis of the model partial differential equations and an extension of the procedure of Horn and Lin, based on the Pontryagin Maximum Principle.

Hence, the scope of this work includes the experimental

Correspondence concerning this paper should be addressed to R. H. Kadlec.

and theoretical optimization of the cyclic adsorber, via the use of a simplified model. The results are compared with experiment, and the potential effects of unvaried parameters are predicted using dimensional analysis. The potential applications of the methods include the optimization of any distributed parameter cyclic process.

CONCLUSIONS AND SIGNIFICANCE

The optimum feed pressure wave form was found to be a square wave, for the objective of product purity alone. If capacity is included in the objective, a period of no feed and no exhaust must be included. The cycle time for maximum separation is approximately 3 seconds, at approximately 50% on-time, for the nitrogen-methane system in a laboratory-scale device. Thus, the optimal operating sequence is maximum feed, no feed or exhaust, maximum exhaust.

The theoretical work provided an excellent guide to the experimental by narrowing the scope of the experimental work. Computations which were very inaccurate composition predictors, due to large finite difference step size, were very accurate optimization routines. We conclude that the optima are insensitive to the process model. These calculations are exceedingly complex and should be attempted only if there are large incentives for the optimization.

The operability of this unsteady process was easily implemented with automatic on-off valves; no difficulties in control of the process were experienced. A steady flow of product was obtained. No moving parts were replaced in over a 100 hours of operation, but valve wear would be expected in longer periods of operation.

It is significant that this process has no steady state analog and therefore cannot be described against the background of a steady state theory. Joined with other parametric pumping processes, a whole new spectrum of processes appear for potential use by industry. This separation process has now been quantified and optimized for one particular equipment.

For most continuous processes within the chemical industry, it is assumed that using constant operating conditions is the best mode of operation. The recent studies of Wilhelm (1966, 1968), Douglas (1967), and Schrodt (1967) have shown, however, that cyclic or periodic operation in some processes can lead to process improvements. Since the magnitude and direction of the change in process outputs changes with cycle type, it is important to locate the best possible operating conditions. This requires both experimentation and the application of optimal control theory.

An inherently periodic process and the subject of this research is a cyclically operated molecular sieve adsorber. An outgrowth of Skarstrom's (1959) heatless adsorber, this gas separation column has practical significance due to the following advantages over some present adsorption methods:

1. No separate adsorbent regeneration process necessary
2. Continuous operation
3. No solids handling needed during operation.

In addition, its fast startup time may make it practical where the startup delays of conventional continuous mode plants are not acceptable.

This cyclic adsorption process, as illustrated in Figure 1, is operated at ambient temperature by alternating between a flow of feed gas mixture and a flow of exhaust gas at one end of the column, while regulating the product end of the column for constant flow. The manner in which the column is pressurized and depressurized at the feed end (feed boundary pressure control) greatly affects the composition of the product stream and the amount of gas exhausted. Thus, it is desirable to establish the optimal feed boundary pressure control which maximizes certain performance criteria of the adsorber. One performance index which provides for minimizing exhaust rate as well as maximizing product composition is given by

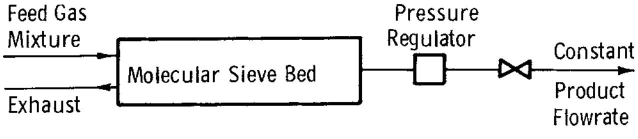

Fig. 1. Periodic adsorption process.

$$I = \frac{1}{\tau} \int_0^\tau (\text{Composition} - C\ [\text{Exhaust rate}])\, dt \quad (1)$$

where τ is the period of a cycle and where the value of the constant C determines the importance of the minimization of exhaust rate relative to the importance of the maximization of product composition.

The modeling of this adsorption process, the application of optimal control theory, and the accompanying numerical analysis will be presented in Part II of this paper. The adsorber can be described by two partial differential equations for pressure and composition, and an ordinary differential equation at the product boundary. The necessary conditions for the optimal control problem have been derived both for the distributed-parameter system and for a cell model approximation of it. Computational work using the cell model and experimental studies were used to maximize the performance index for a fixed product flow rate and a limited available pressure for the feed gas.

This article will deal with the experimental optimization of separation of the binary nitrogen-methane mixture. With unlimited possible control policies, an exhaustive experimental exploration of all process variables would be unrealistically complex. Therefore, the actual adsorber performance was investigated for variation in a general optimal pressure control form that may be established from the application of optimal control theory. These theoretical studies have shown that the general optimal pressure control form, which can also be simply described as the optimal feed boundary flow control, should be of the form [maximum flow in, zero flow, minimum flow in (which is maximum flow out)]. The experimental investigation was further simplified by considering a performance index which considers only the maximization of product composition.

THE EXPERIMENTAL ADSORPTION SYSTEM

The basic experimental adsorption system used in this research was based upon the work of Turnock and Kadlec (1971) and is shown in Figure 2. The desired cyclic pressure control forms can be physically constructed with two 2-way solenoid valves using the following cycling sequence:

1. Feed valve open, exhaust valve closed
2. Both valves closed
3. Feed valve closed, exhaust valve open

These valves are electrically controlled by relays which are activated by the square wave outputs of an analog computer.

Surge vessels are used for both the inlet and outlet flow. The nitrogen-methane mixture, pressure regulated at the feed gas cylinder, passes through a 8.2 dm^3 surge vessel, and the exhaust flows through a 36.0 dm^3 surge vessel before flow measurement. All other flow resistances are kept to a minimum. The solenoid valves were chosen with a 6.35 mm internal orifice, which presents very little resistance to flow. The only other obstacle to flow between the surge vessel and the adsorption column is one thickness of 80 mesh screen used to support the adsorbent.

In his research, Turnock found that the use of crushed molecular sieve pellets led to variable flow resistance caused by attrition of the adsorbent particles. Use of 20-50 mesh round particles of molecular sieve as the adsorbent eliminated that problem. Adsorbent is packed into a 1.9-cm diameter, 1.52-m long, steam jacketed, schedule 40 pipe. This packed bed is mounted on a 35° angle to eliminate the possibility of forming a void channel along the bed.

At the product end of the column, the exit stream passes through a pressure regulator which regulates the pressure of the downstream flow. The flow to the product flowmeters is nearly constant whereas the flow at the end of the column varies significantly. This product stream is then metered by two rotameters: one metering and controlling the flow to a thermal conductivity cell; the other measuring and controlling the remainder of the product flow.

The composition of this product stream is measured by the calibrated thermal conductivity cell. A 100% N_2 stream, also serving as the reference flow, a 50% N_2-50% CH_4 mixture, and the feed gas mixture were used as the calibration gases. The output signal from this thermal conductivity cell is displayed on a digital voltmeter.

A total material balance on the operating system can be completed when the exhaust flow is measured. The flow pulses from the column are damped by the large surge volume so that the flow is easily measured by two wet test meters.

To achieve the desired cyclic pressure control, the signals activating the relays which control the feed and exhaust valves must be properly set. Care must be taken to make certain that the closing of the exhaust valve exactly coincides with the opening of the feed valve. If there is any time that both valves are simultaneously open, gas will bypass the column and flow directly from the feed to the exhaust.

The feed pressure is limited with the pressure regulator on the feed gas cylinder and the product flow is adjusted with the product rotameters. The feed pressure was 168 kN/m^2 for this study. During the startup period, the composition is continuously metered with the output of the thermal conductivity cell. By rechecking the output of this thermal conductivity cell for the calibration gases before and after each run, the relative uncertainty of a composition is within 0.2%.

RESULTS

The initial experimental studies were run with the cyclic sequence (maximum flow, minimum flow), that is, the feed and exhaust valves alternately opening. Figure 3 presents the effect of frequency on product composition. It is noted that neither a different product flow rate, a variation in the fraction of the period that the feed valve remains open (*FFVO*), nor a small change in feed gas composition from 28.6% N_2 to 32.2% N_2 change the frequency, 0.35 cycles/second, at which the maximum in product composition occurs. Thus the variation in the timing of the valve switches was carried out at only one frequency for all product flow rate studied.

Next, the exhaust flow rate is examined as a function of the frequency of the applied control. Figure 4 shows that the faster the control is cycled, the more gas is exhausted. This behavior led to a computational study that included exhaust rate minimization in the performance index. The exhaust rates are only slightly sensitive to the changes in product flow rate; the exhaust rate decreases slightly as the product flow rate is increased.

At the constant frequency of 0.35 cycles/second, the effect of the length of time that the feed valve is open on the product composition is shown in Figure 5. As anticipated, a clear maximum in this composition is exhibited. It is clear that if the valve were either not open at all or

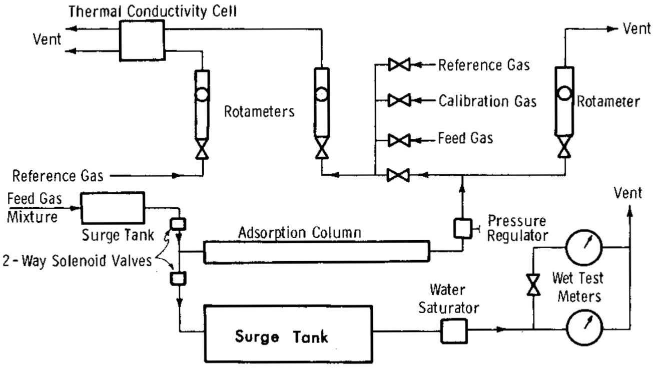

Fig. 2. Schematic diagram of experimental adsorption system.

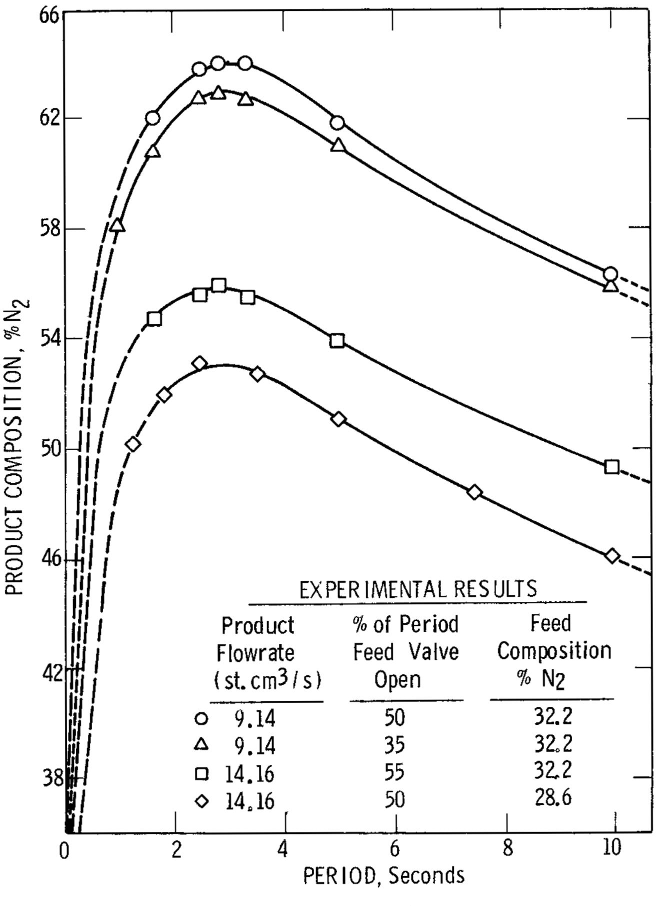

Fig. 3. Effect of frequency on product composition.

if it were open 100% of the time, no steady state separation would be possible.

For simple on-off operation of the feed and exhaust valves, the controls to maximize product composition for different product flow rates have now been experimentally specified. The effect of delaying the opening of the exhaust valve after the feed valve has been closed is shown in Figures 6 and 7.

DISCUSSION OF RESULTS

The experimental results show that the control component of zero flow is not needed in the control sequence to maximize product composition. The optimal frequency is 0.35 cycles/second and there is no noticeable effect of changing product flow rate on this frequency. In addition, it can be seen from Figure 5 that the optimal fraction of the period spent applying maximum pressure increases slightly with increasing product flow rate.

By including a separate term for exhaust minimization in the performance index, as well as product composition maximization, the control component of zero flow becomes important. This component is applied after the maximum pressure component is applied. 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 thus improves the performance of the adsorption system. To construct this control component, two valves were used at the feed boundary of the adsorption column. The timing of the optimal sequence [feed valve open (exhaust valve closed), both valves closed, exhaust valve open (feed valve closed)] will depend upon the relative importance of the exhaust minimization term compared to the term for maximization of product composition.

In the thermal parametric pumping separation process

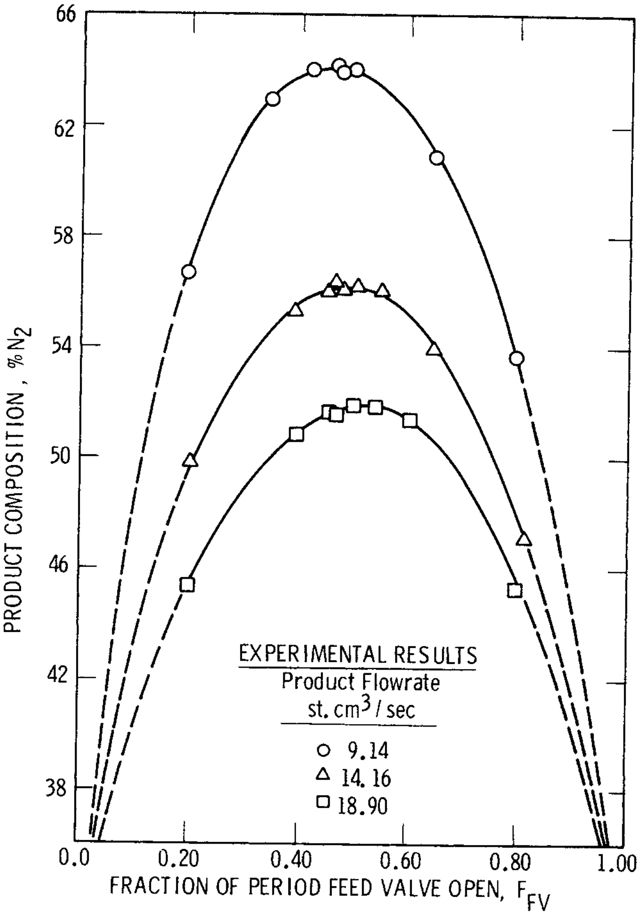

Fig. 5. Effect of fraction of period feed valve open on product composition for 0.35 cycles per second and 32.2%N$_2$ feed.

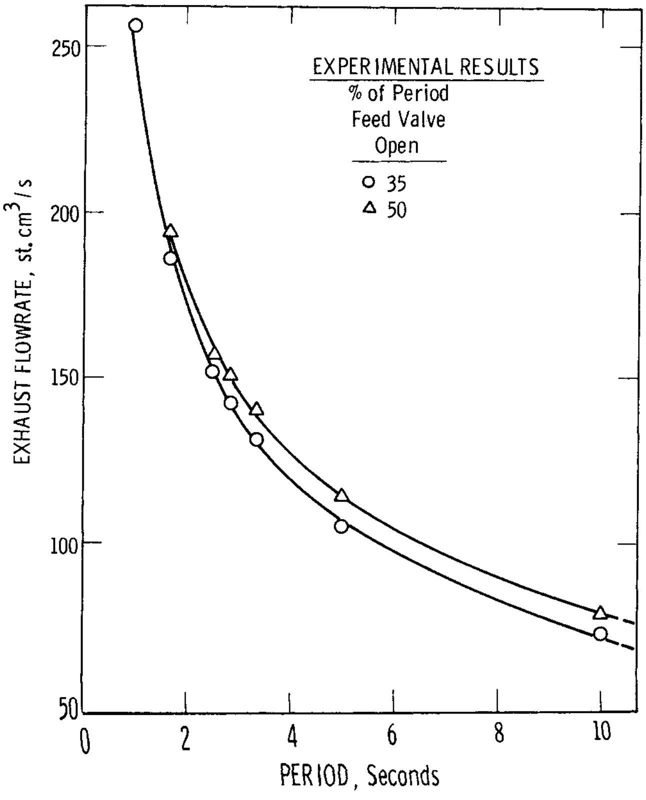

Fig. 4 Effect of frequency on exhaust flow rate for a product flow rate of 9.14 st.cm^3/s.

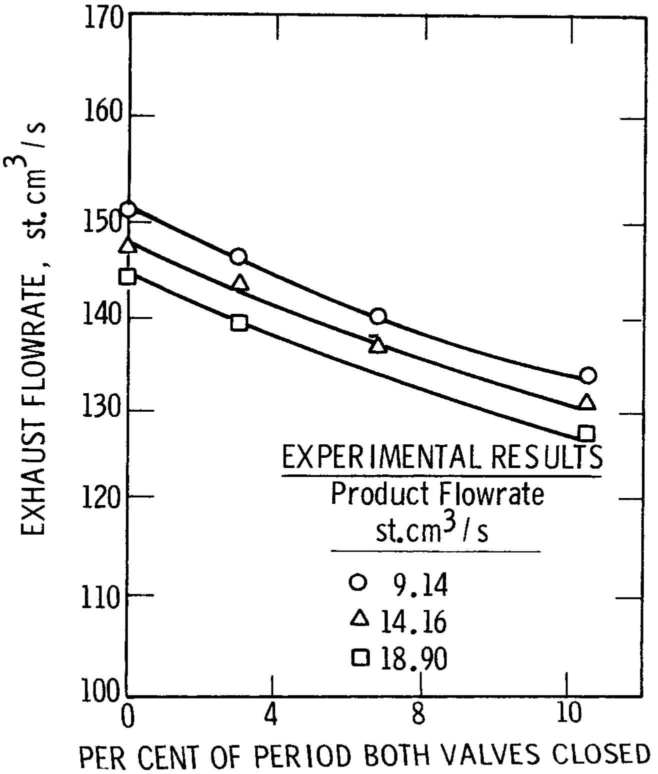

Fig. 6. Effect of closing both valves on exhaust flow rate (0.35 cps, feed 32.2% N$_2$, and feed valve open 47% of period).

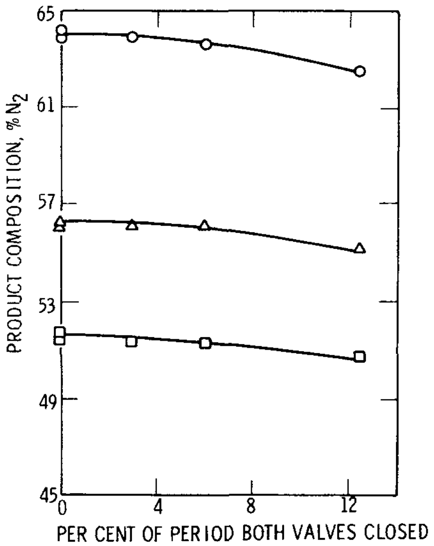

Fig. 7. Effect of closing both valves on product composition. (0.35 cps, feed 32.2% N_2, and feed valve open 47% of period.)

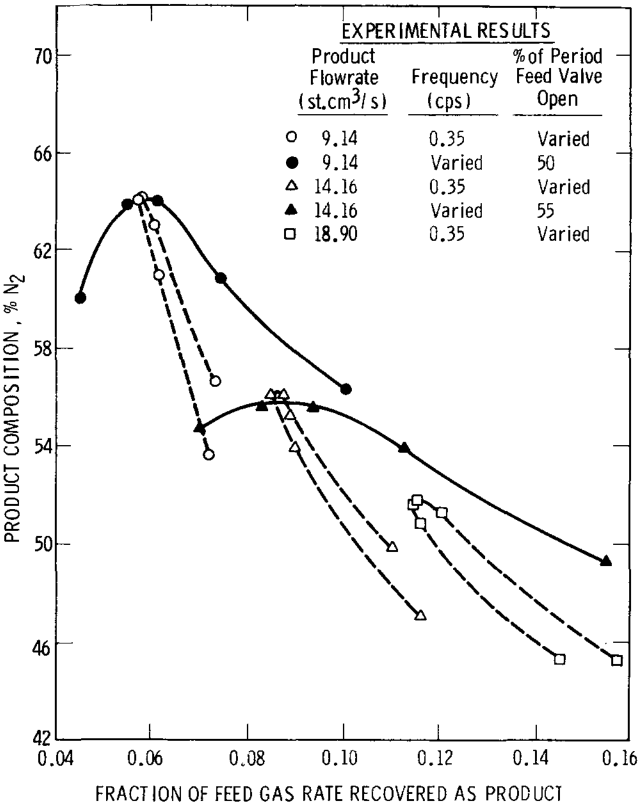

Fig. 8. Comparison of adsorption system outputs with individual variations in frequency of control or in the fraction of period that the feed valve is open, for a feed gas composition of 32.2% N_2-67.8% CH_4.

developed by Wilhelm et al. (1966, 1968), the performance depends upon alternating the direction of the flow of the solvent. It is this kind of behavior in the cyclic adsorber that gives rise to the possibility of the optimal control component of zero flow. Thus, it is anticipated that this control component may also play a role in the optimal control of the thermal parametric pumping process. A full theoretical and numerical investigation is necessary to define the actual optimal sequence.

Figure 3 shows that the optimal frequency does not noticeably vary with product rate. At a constant frequency of 0.35 cycles/second, which corresponds to the maxima in product compositions in Figure 5, *FFVO* was varied for the flow rates of 9.14, 14.16, and 18.9 st.cm³/s.

Another factor of interest, for which no experimental exploration was made, is the level of the available feed pressure. Although the separation for increased pressure markedly increases, the amount of gas exhausted increases as well. In considering the pressure level for column operations, the increased separations for higher pressure levels must be balanced by the increase in the energy required for the pressurizing of the feed gas and the decreased fraction of feed gas recovered as product. A study, either numerical or experimental, would be required to find the optimal feed pressure.

In the preceding experimental work a system of fixed dimensions was operated at different levels of product throughput for a fixed available feed pressure. The problem that should now be considered is how these operating variables and system dimensions affect the operation of the cyclic adsorption process.

From the experimental results in Figures 3 and 5 it is apparent that decreased product flow rate increases the separation accomplished. However, accompanying this increase is a decrease in the fraction of feed gas that is recovered as product. Illustrated in Figure 8 is the change of the process outputs of separation accomplished and fraction of feed gas recovered, as the frequency of operation is varied or as the fraction of the period that the feed valve is open, is varied. These changes are shown for the three different product flow rates of 9.14, 14.16, and 18.9 st. cm³/s. From this figure it is seen that decreased frequency results in an increase of the fraction of feed gas recovered while, as in Figure 3, the accomplished separation goes through a maximum. It is clear that variations of the frequency from the optimal value do not decrease the separation accomplished with respect to the fraction of feed gas recovered, nearly as much as variations of *FFVO* from its optimal value. Thus, if the system is to be run at different flow rates, the frequency of operation need not be set as carefully as the fraction of the period that the feed valve is to be open.

If the separation accomplished and the fraction of the feed gas recovered as product were the only important factors in the operation of the system, it would appear from Figure 8 that operation at lower product flow rates would result in the best performance. However, the capacity of the system must also be considered. For example, if the recovered fraction is 0.09, a 56.6% N_2 product would be achieved for a product flow rate of 9.14 st.cm³/s whereas a product composition of only 55.8% N_2 would be achieved for a product flowrate of 14.16 st.cm³/s. 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 overall operating conditions for the adsorption system, the capacity, as well as the fraction of feed gas recovered as product, needs to be considered.

The system dimensions were constant during the course of the experimental work. In addition, the bed permeability was a fixed quantity. In order to design a cyclic adsorption system, the effect on operations of these equipment specifications must be understood.

The higher the permeability, the greater the capacity of the system provided that the appropriate higher frequencies are used. Consideration of this behavior for design purposes requires two notes of caution. First, if larger adsorbent particles are used to achieve higher permeability, the optimal operation will be at higher frequencies 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 unsteady 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.

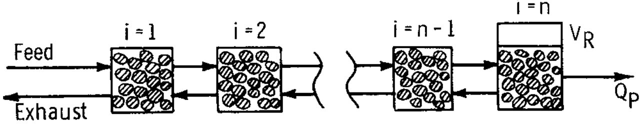

Fig. 1. Model for the cell system.