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Optimal Design of Multistage Adsorption-Bed Systems

EDMOND SUNG

and

CHANG DAE HAN

Department of Chemical Engineering
Polytechnic Institute of New York
Brooklyn, New York 11201

and

HYUN-KU RHEE

Department of Chemical Engineering
College of Engineering
Seoul National University
Seoul, Korea

The operation of two arrangements of multistage adsorption beds, parallel and series, was analyzed, and the optimal design problem of minimizing the capital and operating costs of the multistage adsorption bed systems was formulated. Solution of the optimization problem was obtained by using the Fibonacci search scheme. For the purpose of the study, data on the adsorption of liquid on activated carbon were taken from the literature for both single solute and multisolute adsorption systems and was used to derive the expressions for adsorption rate and the equilibrium relationship. Details of the performance of the optimally designed multistage adsorption bed systems are presented in terms of the physical and design parameters.

SCOPE

In the present study, we undertook an analysis and an optimal design of multistage adsorption bed systems where the problem involves nonequilibrium adsorption. More specifically, the present study deals with formulating the optimal design problem of single and multisolute adsorption systems, developing a method for solving the system equations for an optimal design of multistage adsorption bed systems, investigating the effect of physical parameters on the optimally designed systems, determining which of the two bed arrangements (parallel or series) yields lower capital and operating costs, and investigating the effects of tight purification specifications and the size of the adsorption plant on the cost of plant operation.

Examples of wastewater treatment for single solute and multisolute adsorptions were considered, using data available in the literature. Although the specific applications of the present study have dealt with the problem of wastewater treatment, the concepts of the process design and optimization methods demonstrated here are also applicable to other liquid phase adsorption processes as well.

CONCLUSIONS AND SIGNIFICANCE

The optimal design of multistage adsorption bed systems is presented based on a theoretical consideration of transient cyclic operation. The Fibonacci search scheme has been used to solve an integer programming problem involving a nonlinear objective function. The principal conclusion from this study is that, under optimal design conditions, the serial-bed arrangement is favored over the parallel-bed arrangement, because the leading bed in the serial-bed system can achieve a higher solute saturation than the corresponding one in the parallel-bed sys-

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Correspondence concerning this paper should be addressed to Chang Dae Han. Edmond Sung is with Merck & Co., Inc., Rahway, New Jersey 07065.

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tem. Specific conclusions and significance of the present study are as follows:

1. The minimum treatment cost under optimal condition is little affected by the isotherm parameter α_i in the favorable equilibrium region ($\alpha_i > 0$), because the equilibrium isotherm does not change much as α_i varies in that region. As α_i decreases, the effluent concentration profile becomes more diffuse, resulting in the early appearance of solute in the effluent. This will result in a shorter switching time and hence a higher cost.

2. As β_i is a measure of how close to equilibrium the process is, a large value of β_i will produce a very sharp concentration profile close to the sharp discontinuity produced by the equilibrium. With a large value of β_i, the total bed volume tends to be smaller, resulting in a smaller capital cost.

3. The capacity parameter γ_i is a measure of how much solute the activated carbon is capable of adsorbing when the carbon is a total saturation. As the key component is most strongly adsorbed into the bed, the capacity parameter γ_1 affects the rate of bed exhaustion and hence the direct operating cost of the system. It is observed that the minimum total cost rises extremely rapidly as the adsorption capacity of the carbon, especially the parameter γ_1, is decreased.

4. When the purification requirement is made increasingly stringent, the allowable effluent concentration is decreased. This results in a shorter switching time and consequently a higher total cost.

In recent years, public waters have been increasingly contaminated with synthetic organic compounds. This has become a matter of widespread concern, especially since some of the pollutants, notably pesticides and chlorinated hydrocarbons, are nonbiodegradable. Many municipal waste treatment facilities are equipped only for primary treatment of waste (screening and sedimentation), but even those equipped for secondary treatment (biological process by aeration) cannot remove such compounds. When such waters serve as potable water supplies, the presence of these pollutants gives rise to a potential health hazard. Consequently, the water pollution problem not only has been of public concern, but it has also caused public demands on state and federal authorities to legislate more stringent clean water standards.

Moreover, this class of pollutants cannot be removed by some advanced treatment methods such as coagulation or chemical oxidation. On the other hand, adsorption on activated carbon has been recognized for some time as an effective means for removing organic compounds dissolved in waste streams. This is because the amount of pollutants usually present in waste streams is very minute, and the carbon used once can be reactivated for repeated use. Thus, fixed-bed adsorption on activated carbon has been considered an economical way of treating both domestic and industrial wastewaters.

Despite the long history of the use of activated carbon as an adsorbent and much study conducted on the subject of adsorption, the design of a multistage adsorption-bed system is still largely empirical, and little theoretical study on the subject has been reported in the literature. The bulk of the literature has been devoted to the analysis of single stage adsorption beds, subject to the initial condition of uniform solute concentration in the bed. This type of initial condition is valid only when the bed is either initially empty or uniformly saturated.

In the case of the multistage bed arrangement, the adsorption-bed system is characterized by a nonuniform concentration profile at the beginning of bed switching. Consequently, any analyses available in the literature provide little help for the design of multistage adsorption-bed systems. Moreover, the literature is lacking in the area of multicomponent adsorption, in which local equilibrium cannot be assumed, as is often the case in wastewater treatment.

In recent years, a few research groups (Chen et al., 1972; Svedberg, 1976; Klaus et al., 1977) carried out analyses, simulating the periodic countercurrent operation of multistage adsorption-bed systems. In view of the practical importance of multicomponent, liquid phase adsorption on activated carbon (for example, wastewater treatment), the adsorption of multicomponent systems has recently attracted the attention of some researchers (Tien et al., 1976; Hsieh et al., 1977; Klaus et al., 1977).

In the present study, we shall analyze the operation of two arrangements of multistage adsorption beds, parallel and series, and present a theoretical development of the optimal design problem of minimizing the capital and operating costs of the systems. Examples of the optimally designed systems will be presented in terms of the physical and design parameters, using data available in the literature on the adsorption of liquid on activated carbons for both single solute and multisolute adsorption systems.

THEORIES OF PHYSICAL ADSORPTION

For a theoretical understanding of physical adsorption, one needs a set of system equations consisting of a continuity equation, an equilibrium relationship, and a rate expression for adsorption.

For the isothermal, fixed-bed adsorption problem considered here, the continuity equation is given by

$$\epsilon \frac{\partial c}{\partial t} + u \frac{\partial c}{\partial z} + \rho \frac{\partial q}{\partial t} = 0 \qquad (1)$$

Note in Equation (1) that the axial dispersion in the liquid phase is neglected, which is justifiable in view of the fact that in industrial adsorption operation, the bed diameter is several orders of magnitude larger than that of the granular particles (Schwartz and Smith, 1953). When one deals with multicomponent adsorption, one has to consider Equation (1) for each component.

There are numerous equilibrium relationships proposed in the literature. The most commonly used one is the Langmuir isotherm, given by

$$q = \frac{bc^*}{1 + ac^*} \qquad (2)$$

For adsorption in a multicomponent system (or systems), the generalized form of the Langmuir isotherm may be written as (DeBoer, 1953)

$$q_i = \frac{b_i c_i^*}{1 + \sum_{p=1}^{m} a_p c_p^*}, \quad i = 1, 2, \ldots m \qquad (3)$$

There are different forms of rate expression reported in the literature, and they can be classified into three types: the film resistance model (Hougen and Marshall,

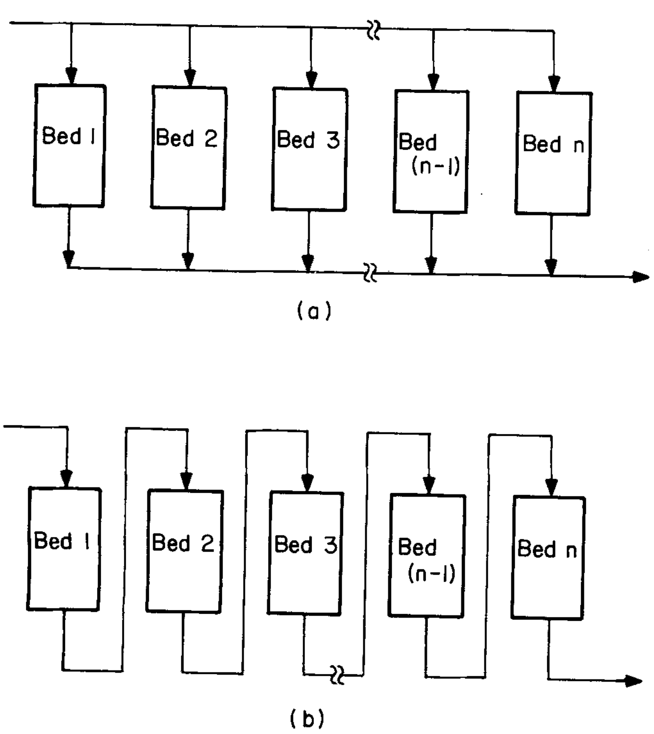

Fig. 1. Schematic of multistage adsorption-bed arrangement: *(a)* parallel-bed arrangement, *(b)* serial-bed arrangement.

1947), the combined film resistance and solid diffusion model (Rosen, 1952), and the kinetic model (Hiester and Vermeulen, 1952).

The film resistance model assumes that the film resistance is the controlling factor in adsorption where equilibrium is established between the concentrations c^* and q. The adsorption rate expression may be represented by

$$\frac{\partial q}{\partial t} = K_f(c - c^*) \tag{4}$$

The film resistance model was used by Hougen and Marshall (1947), Thomas (1944, 1948), and Glueckauf (1955) in solving the adsorption problem. More recently, Chi and Wasan (1970) extended the Hougen-Marshall (1947) solution to studying isothermal adsorption processes with nonlinear equilibrium isotherm.

A number of authors have approximated diffusion in solids obeying Fick's law by a kinetic expression similar to that of the film model. One version of such kinetic model may be expressed in terms of the solid phase concentration difference (Glueckauf, 1955; Jury, 1967; Hsieh, 1974), given by

$$\frac{\partial q}{\partial t} = K_s'(q^* - q) = K_s \frac{\rho}{\epsilon} (q^* - q) \tag{5}$$

where both K_s' and K_s are the solid-phase mass transfer coefficients, except that K_s has the same dimensional units as K_f. Hsieh (1974) uses this rate expression to describe the situation where the adsorption rate is exclusively solid diffusion controlling.

Another version of the kinetic model has the following expression (Chen et al., 1968)

$$\frac{\partial q}{\partial t} = K(Y)(c - c^*) \tag{6}$$

in which Y is the dimensionless concentration defined by

$$Y = q/q_o \tag{7}$$

and thus $K(Y)$ represents the concentration dependent mass transfer coefficient. The experimental data obtained by Magtoto (1966) and Chen et al. (1968) indicate that $K(Y)$ is a decreasing function of Y.

In the above development, we have assumed isothermal adsorption onto spherical particles. Note that the continuity equation, Equation (1), and rate expressions, Equations (4), (5), and (6), can also be used for describing the adsorption of multicomponent systems. It should be pointed out, however, that the choice of a particular rate expression must depend on experimental data. For example, the data of Magtoto (1966) for a single solute system is correlatable by Equation (6), the data of Weber and Keinath (1966) for a two solute system is correlatable by Equation (4), and the data of Hsieh (1974) for a four solute system is correlatable by Equation (5).

In studying isothermal adsorption, one has to solve a set of partial differential equations [for example, Equations (1) and (4)] with the aid of the Langmuir isotherm, Equation (2). Because of the nonlinearity of the Langmuir isotherm, one must use a numerical method for obtaining the solution of the governing system equations. For multicomponent adsorption, one has to solve the governing system equations for each component.

MODES OF OPERATION OF MULTISTAGE ADSORPTION-BED SYSTEMS

In the large scale operation of adsorption systems (for example, in wastewater treatment), the single-bed system is not normally used because of the need to replace the adsorbent at intervals. If a waste stream is very small, however, a single-bed system can be used because the cost of recharging the adsorbent may be a minor part of operating expenses.

For a large scale continuous operation, multistage adsorption beds may be arranged, either in parallel or in series, as shown schematically in Figure 1. In the parallel arrangement (see Figure 1*a*), all the beds are fed by the same feed. The beds are staggered so that they are at varying degrees of adsorbent saturation. The desired effluent (below a certain maximum cutoff effluent concentration) is made up by blending the effluents from each bed. When the cutoff concentration is reached, the bed with the longest on-stream time (most saturated) is shut down, and a standby fresh bed is then staggered into the system. The time between two successive bed replacements is called the switching time.

With the beds arranged in series (see Figure 1*b*), the stream from the last bed is the desired effluent. When the effluent reaches the cutoff concentration, the leading bed (most saturated) is shut down, and a standby fresh bed is put on stream at the end of the serial arrangement. The second bed then becomes the leading bed. Likewise, the time between two successive bed replacements is called the switching time.

Parallel-Bed Arrangement

In the parallel arrangement, each bed can be considered independent of the others throughout its entire on-stream time. This implies that each bed can be analyzed as if it were a single-bed system. A major difference between the single-bed system and the parallel-bed arrangement lies in the fact that the latter can be operated at a very high saturation level before shutdown. This is because the stream containing the highest effluent concentration from the last bed is diluted by blending the streams of lower effluent concentration from the other beds to produce a system effluent of satisfactory quality. Since the analysis of a parallel-bed system is based on that of a single bed,

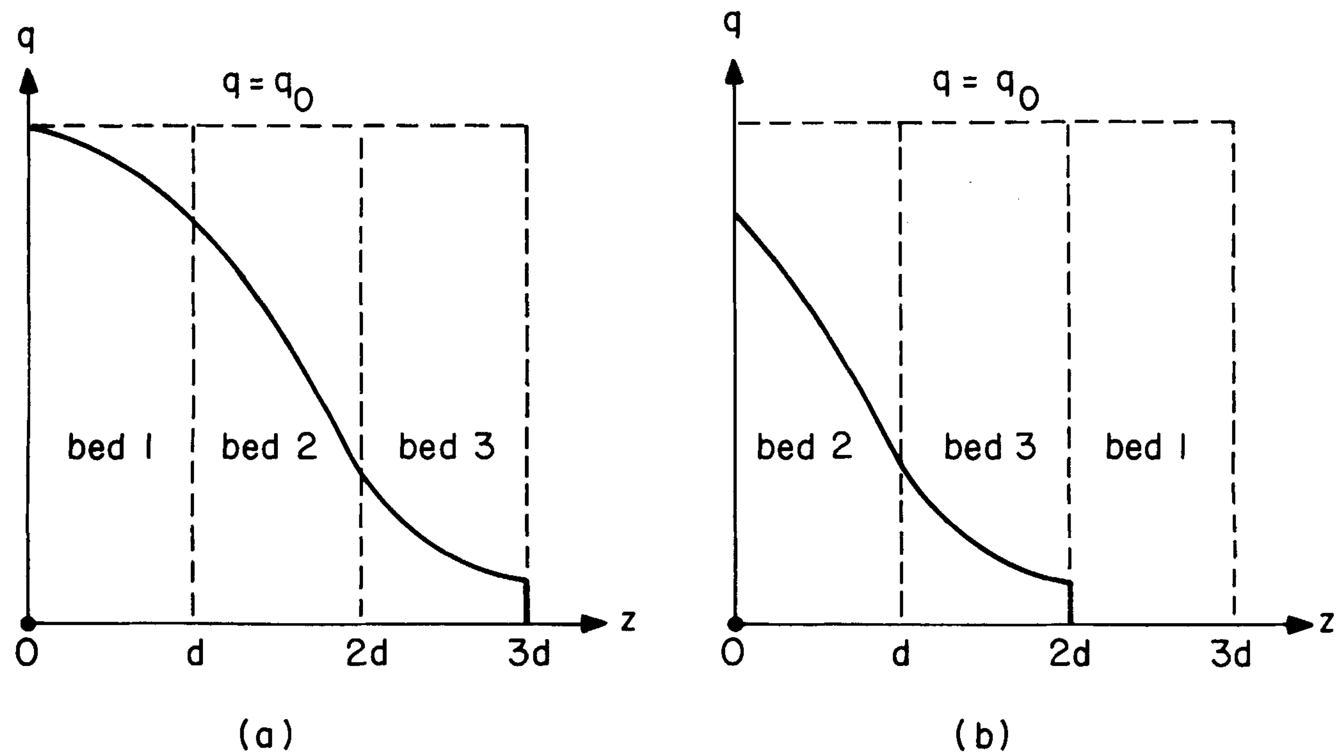

Fig. 2. Concentration profiles in the three stage adsorption beds: *(a)* $q^{(1)}[z,t_m^{(1)}]$, *(b)* $q^{(2)}(z,0)$.

the system equations for the single bed may be used to derive an expression for the effluent concentration of the multistage parallel-bed system.

The inlet and initial conditions for the single-bed operation are given by

$$c(0,t) = c_0 \quad \text{at} \quad z = 0 \tag{8}$$

and

$$q(z,t) = 0 \quad \text{for} \quad t \leq (\epsilon z/u) \tag{9}$$

respectively. At the bed exit of a single bed $(z = d)$, the variation of the effluent concentration with time may be expressed as

$$c_e = c(d,t) \tag{10}$$

Likewise, the effluent concentration in a multistage parallel-bed system may be expressed in terms of a switching time t_s as

$$\begin{aligned} &\text{first bed} && c_{e(1)} = c\,[d,\,(n-1)t_s + t] \\ &\text{second bed} && c_{e(2)} = c\,[d,\,(n-2)t_s + t] \\ &&& \vdots \\ &(n-1)^{\text{th}}\text{ bed} && c_{e(n-1)} = c(d,\,t_s + t) \\ &n^{\text{th}}\text{ bed} && c_{e(n)} = c(d,t) \\ &&& 0 < t \leq t_s \end{aligned} \tag{11}$$

For single component systems, the concentration of the stream which is blended from all the effluent streams may be expressed by

$$\bar{c} = \frac{1}{n}\sum_{j=1}^{n} c[d,\,(j-1)t_s + t], \quad 0 < t \leq t_s \tag{12}$$

Bed switching takes place when $\bar{c}$ reaches the predetermined value of cutoff concentration $\bar{c}_{\max}$ defined by

$$\bar{c}_{\max} = \frac{1}{n}\sum_{j=1}^{n} c(d,\,jt_s) \tag{13}$$

When we deal with multicomponent systems, the average concentration of the blended effluent may be expressed as

$$\bar{c}_{\text{tot}} = \frac{1}{n}\sum_{j=1}^{n}\sum_{i=1}^{m} c_i[d,\,(j-1)t_s + t] \tag{14}$$

The associated boundary and initial conditions are given by

$$c_i(0,t) = c_{i0}, \quad i = 1, 2, \ldots, m \tag{15}$$

and

$$q_i(z,t) = 0, \quad i = 1, 2, \ldots, m \quad \text{and} \quad t \leq (\epsilon z/u) \tag{16}$$

respectively. Bed switching takes place when $\bar{c}_{\text{tot}}$ reaches the cutoff concentration $\bar{c}_{\max}$:

$$\bar{c}_{\max} = \frac{1}{n}\sum_{j=1}^{n}\sum_{i=1}^{m} c_i(d,\,jt_s) \tag{17}$$

In dimensionless form, Equation (13) may be written as

$$M = \frac{1}{n}\sum_{j=1}^{n} X(L,\,jT - \theta_0) \tag{18}$$

For the multicomponent system, the general form of Equation (14) is given by

$$M = \frac{1}{n}\sum_{i=1}^{m}\sum_{j=1}^{n} f_i X_i(L,\,jT - \theta_0) \tag{19}$$

Serial-Bed Arrangement

In comparison with the single bed, the multistage serial-bed arrangement has the following three advantages: continuous operation is made possible by bed switching, a higher average bed saturation is obtainable in the leading bed before shutdown, and it provides enough total bed depth so that a highly purified effluent can be obtained.

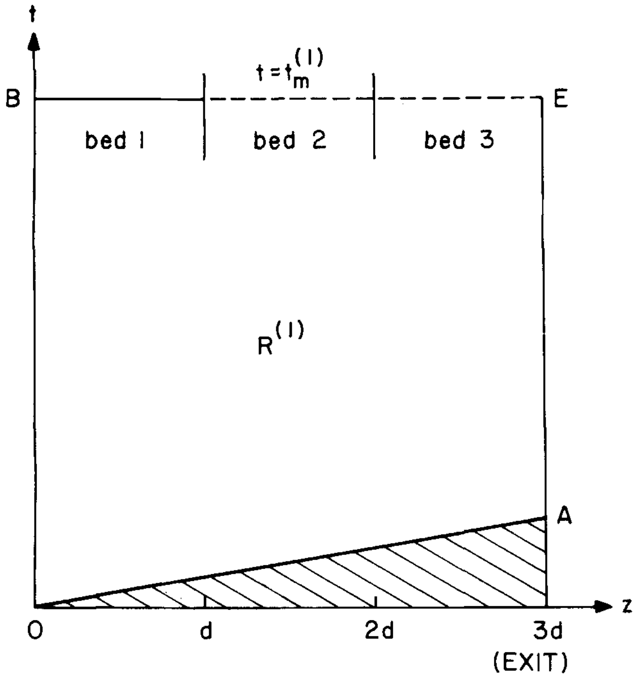

Fig. 3. Inlet and initial conditions for cycle 1 in the three stage adsorption beds.

It is important at this point to note the difference in the initial conditions between the operations of a single-bed system and that of a multistage serial-bed system. Note that in the single-bed system, the initial condition of the bed is given by Equation (9) and the inlet boundary condition is given by Equation (8). However, in the serial-bed system, the initial condition is not known a priori.

For illustration purposes, let us consider the three-stage adsorption beds arranged in series, and let the initial bed arrangement be given by the 1-2-3 sequence (see Figure 2*a*). At the time $t = t_m$ at which the effluent stream reaches the cutoff concentration, that is, $c^{(1)} = c_{\max}$, the first bed (bed 1) will be replenished with fresh adsorbent so that the new bed arrangement will have the sequence 2-3-1, as schematically shown in Figure 2*b*. It is seen that the final condition [at $t = t_m^{(1)}$] of beds 2 and 3 in cycle 1 (see Figure 2*a*) becomes the initial condition of cycle 2 (see Figure 2*b*).

To help illustrate the initial and inlet conditions of cycle 1, Figure 3 gives the $z - t$ plane for the three-stage system, in which the region $R^{(1)}$ for cycle 1 is enclosed by $0 \leq z \leq 3d$ and $0 \leq t \leq t_m^{(1)}$. The inlet condition along $\overline{OB}$ is

$$c^{(1)}(0, t) = c_0 \tag{20}$$

The shaded area enclosed by $\overline{OA}$ and the z axis represents the initial condition, and the bed is solute free ($q = 0$) at position z before the first element of inlet fluid reaches that position. By physical interpretation, $t = \epsilon z/u$ (line $\overline{OA}$) represents the trajectory of the first fluid element entering the system at $t = 0$ and moving at the velocity u. Thus, the initial condition may be represented by

$$q^{(1)}(z, t) = 0 \quad \text{for} \quad t \leq (\epsilon z/u) \tag{21}$$

At the end of cycle 1, the effluent concentration $c(3d, t)$ reaches the point E in Figure 3, the cutoff concentration, $c_{\max}$ at $t = t_m$. Therefore, the value of t_m is implicitly determined by

$$c^{(1)}[3d, t_m^{(1)}] = c_{\max} \tag{22}$$

At $t = t_m$, the bed condition is given by the line segment $\overline{BE}$ in Figure 3. Since, at the end of cycle 1, the first bed (bed 1 in the 1-2-3 sequence) ($0 \leq z \leq d$) is removed from the system, the dotted line segment of $\overline{BE}$ represents the initial condition of the first two beds in cycle 2.

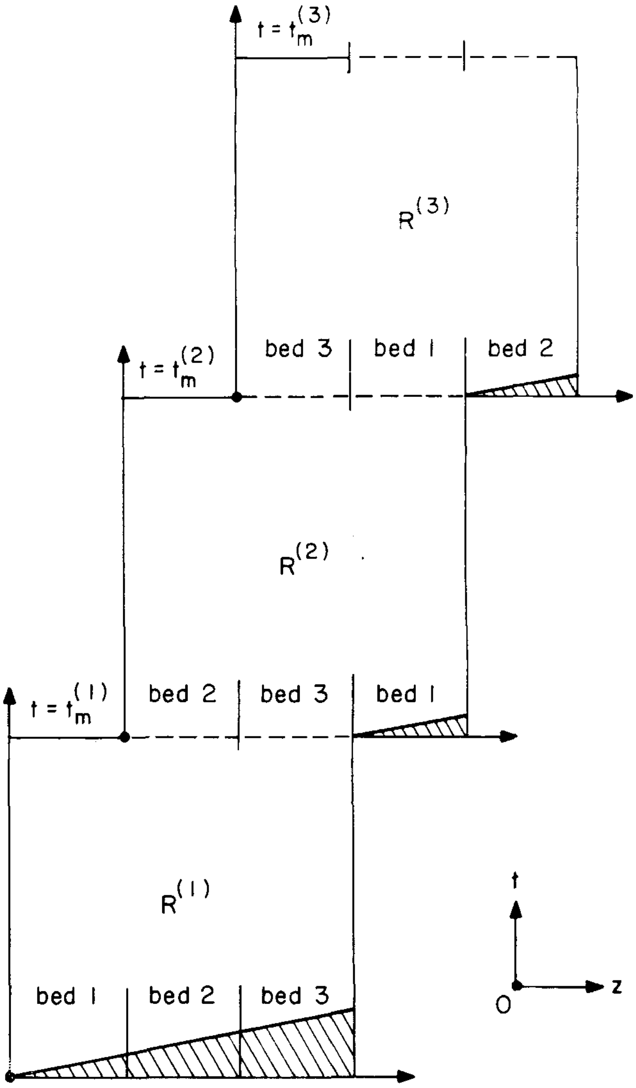

Fig. 4. Inlet and initial conditions for the first three cycles in the three stage adsorption beds.

With reference to Figure 4, the initial condition for cycle 2 may be represented by

$$q^{(2)}(z, 0) = q^{(1)}[z + d, t_m^{(1)}], \quad 0 \leq z \leq 2d \tag{23}$$

and the inlet condition is the same as that in region $R^{(1)}$, except that the feed enters bed 2:

$$c^{(2)}(0, t) = c_0 \tag{24}$$

Note that, with reference to Figure 4, the initial condition in region $R^{(2)}$ is different from that in region $R^{(1)}$ because beds 2 and 3 contain solute as the result of cycle 1 operation. This initial condition is given by Equation (23). The only bed that is solute free is the last bed, newly introduced to the series. The initial condition of this bed is

$$q^{(2)}(z + 2d, t) = 0, \quad t \leq (\epsilon z/u), \quad \text{and} \quad 0 \leq z \leq d \tag{25}$$

Cycle 2 is completed when the effluent concentration reaches the cutoff value $c_{\max}$; that is

$$c^{(2)}[3d, t_m^{(2)}] = c_{\max} \tag{26}$$

Likewise, the final condition at $t = t_m^{(2)}$ becomes the initial condition for region $R^{(3)}$ (see Figure 4); that is

$$q^{(3)}(z,0) = q^{(2)}[z + d, t_m^{(2)}] \tag{27}$$

Therefore, after the j^{th} cycle, the initial condition may be written as

$$q^{(j)}(z,0) = q^{(j-1)}[z + d, t_m^{(j-1)}] \tag{28}$$

At a large value of j, say N, $t_m^{(j)}$ will converge to some value t_s, and the functions $c(z,t)$ and $q(z,t)$ in region $R^{(j-1)}$ will converge to those in region $R^{(j)}$:

$$q^{(N)}(z,t) = q^{(N)}(z + d, t_s + t) \tag{29}$$

The above relation is valid for any given t, which includes $t = 0$ as the initial condition along the z-axis and $t = (\epsilon z/u)$ along the trajectory from the origin $q^{(j)}(0,0)$. The above condition represents the cyclic operation of the three-stage adsorption beds arranged in series, in which the same transient pattern occurs every period. The period length t_s is called the switching time.

In dimensionless form, for single component systems the cutoff concentration is given by

$$M = X(nL, T - n\theta_0) \tag{30}$$

Similarly, for the multicomponent system the dimensionless cutoff concentration is given by

$$M = \sum_{i=1}^{m} f_i X_i(nL, T - n\theta_0) \tag{31}$$

GOVERNING SYSTEM EQUATIONS

In order to facilitate analysis and computation, the governing system equations may be expressed in dimensionless form. The continuity equation may be rewritten as

$$\frac{\partial X_i}{\partial \zeta} + \mu_i \frac{\partial Y_i}{\partial \theta} = 0, \quad i = 1, 2, \ldots, m \tag{32}$$

Note that μ_i in Equation (32) becomes unity for single-component adsorption, and that for multicomponent systems, the key component is denoted by $i = 1$.

The dimensionless rate expression may be written as

For film resistance model [see Equation (4)]

$$\mu_i \frac{\partial Y_i}{\partial \theta} = \lambda_i(X_i - X_i^*); \quad i = 1, 2, \ldots, m \tag{33}$$

For kinetic model [see Equation (5)]

$$\frac{\partial Y_i}{\partial \theta} = \lambda_i(Y_i^* - Y_i); \quad i = 1, 2, \ldots, m \tag{34}$$

The dimensionless isotherm, Equation (3), may be written as

$$X_i^* = \frac{Y_i}{1 + \sum_{p=1}^{m} \alpha_p - \sum_{p=1}^{m} \alpha_p Y_p}; \quad i = 1, 2, \ldots, m \tag{35}$$

We have also the following expression when q and c^* in Equation (3) are replaced by q^* and c, respectively:

$$Y^{i*} = \left[\frac{1 + \sum_{p=1}^{m} \alpha_p}{1 + \sum_{p=1}^{m} \alpha_p X_p} \right] X_i; \quad i = 1, 2, \ldots, m \tag{36}$$

Note that Equation (35) may be used in Equation (33), and Equation (36) may be used in Equation (34). The definitions of the dimensionless variables used above are given in the Notation, and the manipulation of the various equations shown above is given by Sung (1977).

For the parallel-bed arrangement, the boundary and initial conditions, Equations (15) and (16), may be rewritten in dimensionless form by

$$X_i(0, \theta) = 1 \tag{37}$$

$$Y_i(\zeta, 0) = 0 \tag{38}$$

For the serial-bed arrangement, however, the boundary and initial conditions require, in addition to Equations (37) and (38), another relationship

$$Y_i(\zeta, 0) = Y_i(\zeta + L, T - \theta_0); \quad 0 \leqq \zeta \leqq (n-1)L \tag{39}$$

which follows from Equation (29). It is very important to note that Equation (39) is an unknown function of ζ. Because of this, the system equations cannot be directly solved using the available initial data. For this reason, design engineers often rely on lengthy and expensive pilot plant experiments to estimate an approximate size of the system, given a prespecified switching time t_s. Subsequently, the system is further oversized to ensure that in every cycle no breakthrough will occur at the exit of the system.

Clearly, a better design can be obtained if a method based on theoretical considerations is developed. What is needed, therefore, is a general method of simulating such a system (that is, the serial-bed arrangement) so that a more effective design can be obtained. As a specific objective of this study, methods for the numerical solution of the design problems described above will be developed below.

In the present study, we have devised a finite-difference scheme using predictor-corrector method for the numerical solution of the governing system equations, for example Equations (32), (33), and (35). The details of the predictor-corrector method are given in the standard textbooks (Hildebrand, 1956; Keller, 1968). A considerable number of computations were performed in order to determine acceptable step sizes in time θ and distance ζ. A check on the accuracy of the numerical procedure was made by comparing the numerical and analytical solutions for the system with the linear isotherm. Details of the numerical procedure employed, and the accuracy of the solutions obtained, are documented elsewhere (Sung, 1977).

Some representative results of the computation, namely, concentration profiles of the effluent stream, will be presented below when we discuss specific examples of both single solute and multisolute adsorption in conjunction with the performance of optimally designed multistage bed systems.

FORMULATION OF THE OPTIMAL DESIGN PROBLEM OF MULTISTAGE ADSORPTION-BED SYSTEMS

Objective Function

Given a feed composition, flow rate, and desired effluent purity, the design variables to be determined are the number of beds (n), the volume of the bed (V), and the switching time (t_s). The first two design variables (n and V) determine the capital investment, while the last two (V and t_s) determine the direct operating cost.

The equipment cost of an adsorption bed can be expressed as a linear function of the equipment volume V_b:

$$\text{Capital cost (\$/bed)} = A_0 + A_1V_b \tag{40}$$

The total capital cost for a system of $(n + 1)$ beds, including the standby bed, is expressed by

$$\text{Capital cost} = \frac{C_I}{(C_I)_0}(A_0 + A_1V_b)(n + 1)^{A_2} \tag{41}$$

Given a set of physical parameters, the objective function $I(n, V, t_s)$ (that is, the cost to be minimized) is given by (Sung, 1977)

$$I(n, V, t_s) = \frac{3.716}{G}\left(\frac{V}{t_s}\right) + \frac{0.0137}{G}(176.9 + V)(n + 1)^{0.9} \tag{42}$$

where the first term represents the direct operating cost related to the rate of replacing carbon in the bed. Note that Equation (42) must satisfy the constraint given by Equation (19) for the parallel-bed system or that given by Equation (31) for the serial-bed system. Therefore, only two of the three variables n, L, and T are independent. This means that if two variables (n, L) are fixed, the system Equations (32) to (38) for the parallel-bed arrangement or Equations (32) to (39) for the serial-bed arrangement, can be solved numerically to obtain the values of $X_i(L, \theta)$, that is, the concentration profile at a fixed bed depth L. Determination of switching time T is then equivalent to finding the root of Equation (19) or Equation (31), using the values of $X_i(L, \theta)$. Once the value of T is obtained, the value of the objective function $I(n, L, T)$ is evaluated from Equation (42).

Therefore, the process of obtaining the solution X_i and determining T with given values of n and L is actually a design procedure for a multistage bed system. And the optimization algorithm can be viewed as a procedure that strategically generates a sequence of (n, L) combinations (that is, a sequence of designs), such that the value of $I(n, L)$ will rapidly approach the minimum value

$$\underset{n,\,L}{\mathrm{MIN}}\,[I(n, L)] = I(n^*, L^*) \tag{43}$$

The Fibonacci Search Scheme

The nature of Equation (42) is such that we can employ the Fibonacci search scheme (Johnson, 1956) to find the optimal values of the variables n and L. Fibonacci search, like other direct search methods, was developed on the basis that the objective function is unimodal; that is, the function must possess a single minimum and no maximum in the region of interest, and it does not have to be continuous. The method is known to be very efficient for one directional search. The details of the search scheme can be found in optimization textbooks (Beveridge and Schecter, 1970).

We consider in our problem that the function $I(n, L)$ given in Equation (42) is also unimodal, but two dimensional. The search scheme is suitable for finding the optimal value of a real variable, such as the variable L in $I(n, L)$ if n is given. The idea that the Fibonacci scheme can be used to search for the optimal value of an integer variable seems to have been first suggested by Beveridge and Schecter (1970). In the present study, the Fibonacci scheme is extended to search for the optimal values of the integer variable n and the real variable L. Therefore, in effect, the Fibonacci scheme has been extended here to solve a mixed integer programming problem involving a nonlinear objective function and an implicit constraint. The constraint is implicit because it is imposed on the solution to a system of partial differential equations, Equations (32) and (33), or Equations (32) and (34), and therefore, generally, it has no closed form expression. This type of application has not been reported before.

Optimal Design Method for Multistage Bed Systems

The design of a parallel-bed system is based on the effluent profiles $X_i(L, \theta)$ of the single-bed system. When the solution of the parallel-bed system is represented by

$$X_i = \phi_{pi}(\zeta, \theta) \quad i = 1, 2, \ldots, m \tag{44}$$

$$Y_i = \psi_{pi}(\zeta, \theta) \quad i = 1, 2, \ldots, m \tag{45}$$

the specific design of a parallel-bed system is to determine the variables n, $\zeta = L$, and $\theta = T$ such that the constraint, Equation (19), is satisfied; that is,

$$M = \frac{1}{n}\sum_{i=1}^{m}\sum_{j=1}^{n} f_i\phi_{pi}(L, jT - \theta_0) \tag{46}$$

The associated cost minimization problem is to determine the optimal values of n, L, and T to minimize the objective function $I(n, L, T)$ in Equation (42).

To evaluate the objective function $I(n, L, T)$, a design of parallel-bed system must first be obtained. At a fixed value of $L = L_k$, the effluent profiles $\phi_{pi}(L_k, \theta)$ are now used for such a design. With a given value of n, say $n = n_l$, the root T in Equation (46) is solved by using the values of $\phi_{pi}(L_k, \theta)$:

$$M = \frac{l}{n}\sum_{i=1}^{m}\sum_{j=1}^{n_l} f_i\phi_{pi}(L_k, jT - \theta_0) \tag{47}$$

The objective function $I(n, L, T)$ in Equation (42) can now be evaluated as $I(n_l, L_k)$ using $n = n_l$, $L = L_k$, and the value of T just determined.

Essentially, the optimization procedure starts with the generation of n_l by the Fibonacci method, Then, at each n_l under consideration, a sequence of L_k is generated, and the objective function $I(n_l, L_k)$ is evaluated. The minimum value $I(n^*, L^*)$ of the objective function is obtained by comparing the results of Fibonacci searches performed at different values of n_l.

In the design of a serial-bed system, a number of numerical solutions, corresponding to the number of cycles, have to be performed until the solutions of two successive cycles are virtually identical (or converged). Actually, this corresponds to the start-up operation of a serial system; several cycles are needed to reach a routine cyclic operation.

Since the unknown in obtaining the numerical solution is the switching time T, the major loop in the numerical integration is in the θ direction. The details of the computational algorithm are given elsewhere (Sung, 1977).

When the solution of the serial-bed system is represented by

$$X_i = \phi_{si}(\zeta, \theta), \quad i = 1, 2, \ldots, m \tag{48}$$

$$Y_i = \psi_{si}(\zeta, \theta), \quad i = 1, 2, \ldots, m \tag{49}$$

the specific design of a serial-bed system is to determine the variables n, $\zeta = L$, and $\theta = T$ such that the constraint, Equation (31), is satisfied; that is

$$M = \sum_{i=1}^{m} f_i\phi_{si}(nL, T - nL/\gamma_1) \tag{50}$$

The associated cost minimization problem is to determine the optimal values of n, L, and T to minimize the objective function $I(n, L, T)$ given by Equation (42). The procedure for obtaining the optimal design of a serial-bed

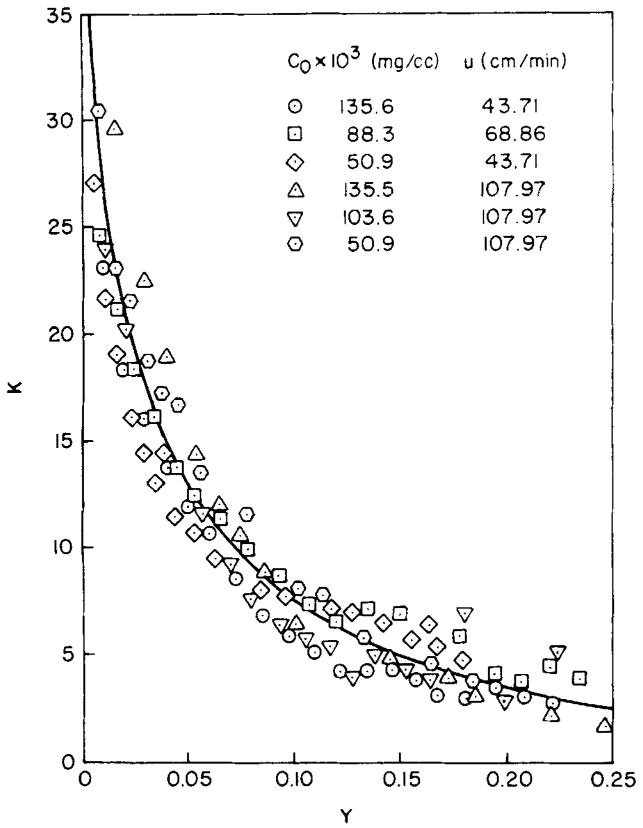

Fig. 5. Correlation of $K(Y)$ from the Magtoto's adsorption data of 2,4-D solution on activated carbon.

system is analogous to that of a parallel-bed system and will not be repeated here.

Based on our experience with 179 optimization runs made using various examples to be discussed below, the objective function seems to be unimodal in all cases. Also, it appears that the optimal value of n alawys falls within a limited range. In practice, it may be possible to apply the Fibonacci method only to locate the optimal value of L, while exhaustive search is conducted for variable n.

EXAMPLES

Having formulated the optimal design problem of multistage adsorption-bed systems in the general sense, we shall now consider some specific cases of physical adsorption systems published in the literature and discuss the performance of the optimally designed multistage adsorption-bed systems under various sets of physical and design parameters.

Data on the Adsorption of Liquid on Activated Carbon

Magtoto (1966) carried out a fixed-bed adsorption experiment for removing the herbicide 2,4-dichlorophenoxy acetic acid (2,4-D) from an aqueous solution, using activated carbon. Figure 5 gives plots of $K(Y)$ vs. Y, indicating that $K(Y)$ decreases with Y. Using the nonlinear least-squares method, we found that data to best fit the expression [see Equation (6)]

$$K(Y) = K_f e^{-\delta\sqrt{Y}} \tag{51}$$

with the numerical values of $K_f = 47.76$ ml/g-min and $\delta = 5.835$.

Figure 6 gives plots of q vs. c^* for the 2,4-D system prepared with Magtoto's experimental data (Magtoto, 1964). It is seen that the experimental data are correlatable with the Langmuir isotherm given by [see Equation (2)]

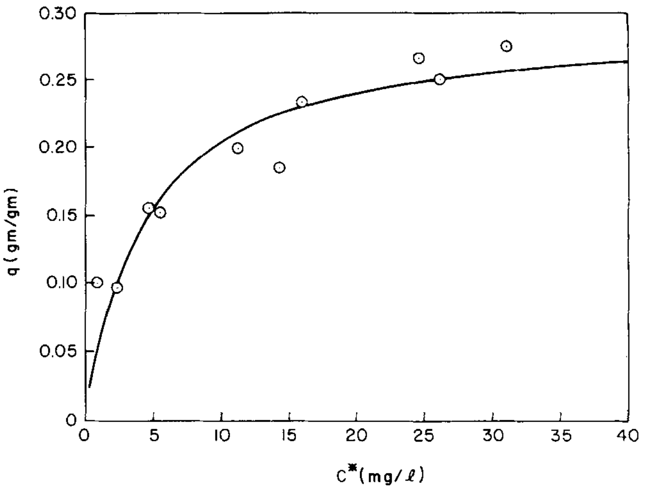

Fig. 6. Langmuir isotherm of 2,4-D solution on activated carbon (T = 30°C).

$$q = \frac{0.0666c^*}{1 + 0.2270c^*} \tag{52}$$

Weber and Keinath (1966) carried out a fixed-bed adsorption experiment to remove two solutes, sodium dodecylbenzene sulfonate (ABS) and 2,4-dinitro-o-sec-butyl phenol (DNOSBP), from aqueous solution on activated carbon. More recently, Hsieh (1974) carried out an adsorption experiment with an aqueous system containing four solutes on activated carbon to simulate treatment of municipal wastewater. The four solutes were detergents represented by ABS (1 ppm), hexoses represented by D-glucose (3 ppm), amino acids represented by DL-valine (2 ppm), and acids represented by lactic acid (6 ppm).

Tho two-component data of Weber and Keinath (1966) are correlatable by the film model [see Equation (4)]

$$\frac{\partial q_i}{\partial t} = (K_f)_i(c_i - c_i^*); \quad i = 1, 2 \tag{53}$$

According to the experimental data of Weber and Gould (1966), the equilibrium relationship for the two-component system follows the generalized Langmuir isotherm [see Equation (3)]. Table 1 gives the numerical values of the film coefficients $(K_f)_i$ and the constants in the Langmuir isotherm.

The four-component data of Hsieh (1974) are correlatable by the kinetic model [see Equation (5)]

$$\frac{\partial q_i}{\partial t} = (K_s)_i \frac{\rho}{\epsilon}(q_i^* - q_i); \quad i = 1, 2, 3, 4 \tag{54}$$

Hsieh's experimental data show that the equilibrium relationship for the four-component system follows the generalized Langmuir isotherm. Table 2 gives the numerical values of the mass transfer coefficients $(K_s)_i$ and the constants in the Langmuir isotherm.

Profiles of Effluent Concentration

Figure 7 gives the single-bed effluent curve for an aqueous solution of 2,4-D passed through a bed of activated carbon. It is seen that the effluent stream concentration of

TABLE 1. NUMERICAL VALUES OF THE FILM COEFFICIENTS AND THE CONSTANTS IN THE LANGMUIR ISOTHERM FOR THE TWO-COMPONENT SYSTEM SIMULATED

Solute	$(K_f)_i$	b_i	a_i
ABS	0.86	1.88	4.55
DNOSBP	1.12	1.64	0.17

Table 2. Numerical Values of the Mass Transfer Coefficients and the Constants in the Langmuir Isotherm for the Four-Component System Simulated

Solute	$(K_s)_i \times 10^3$	$b_i \times 10^3$	$a_i \times 10^3$
ABS	2.356	45.08	280.0
D-glucose	2.244	3.395	6.83
DL-valine	2.170	3.176	3.39
Lactic acid	1.203	2.111	1.45

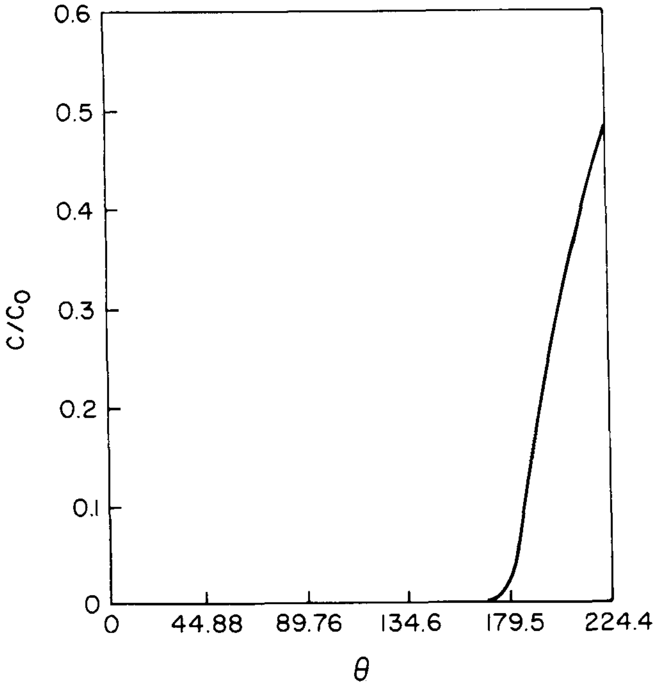

Fig. 7. Single-bed effluent profile of 2,4-D solution in a bed of activated carbon.

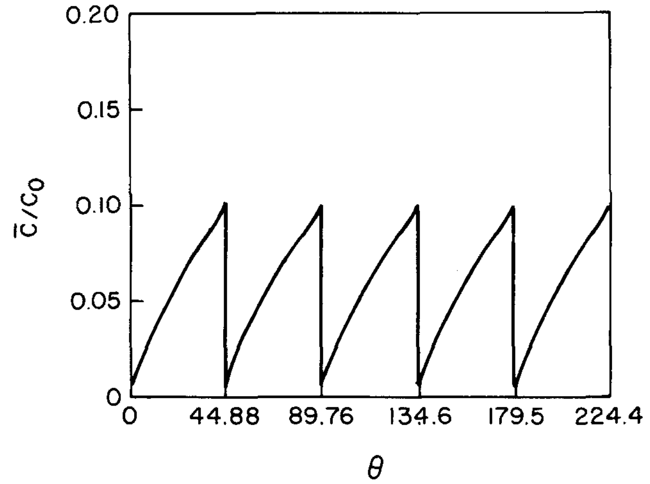

Fig. 8. Profile of 2,4-D solution in the blended effluent.

unadsorbed solute is about 50% of that in the feed stream at the dimensionless time θ of about 224.40.

Figure 8 gives the blended (that is, average) effluent concentration curve for the same solution when passed through five beds arranged in parallel, with a cutoff concentration $\bar{c}_{max}/c_0$ of 0.1. Using the criterion given by Equation (12), the dimensionless bed switching time θ_s is found to be 44.88.

In a multicomponent system, the key and nonkey components are defined according to the degree of adsorption affinity or the degree of nonlinearity in their isotherms.

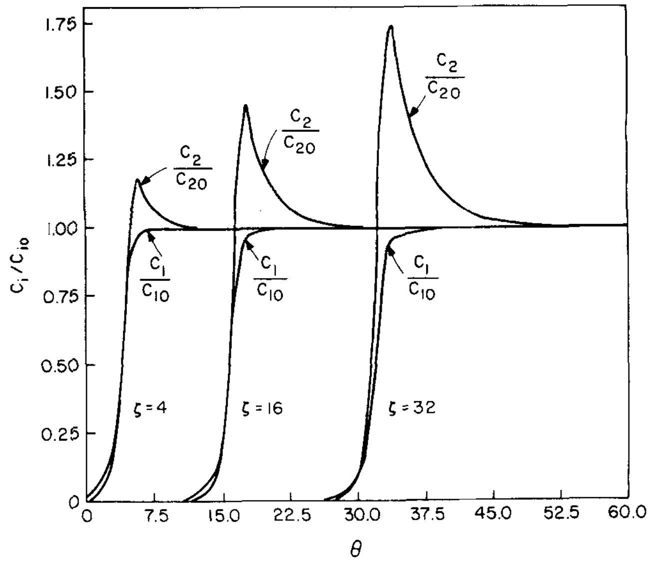

Fig. 9. Effluent profiles of two-solute system consisting of ABS(C_1) and DNOSBP (C_2).

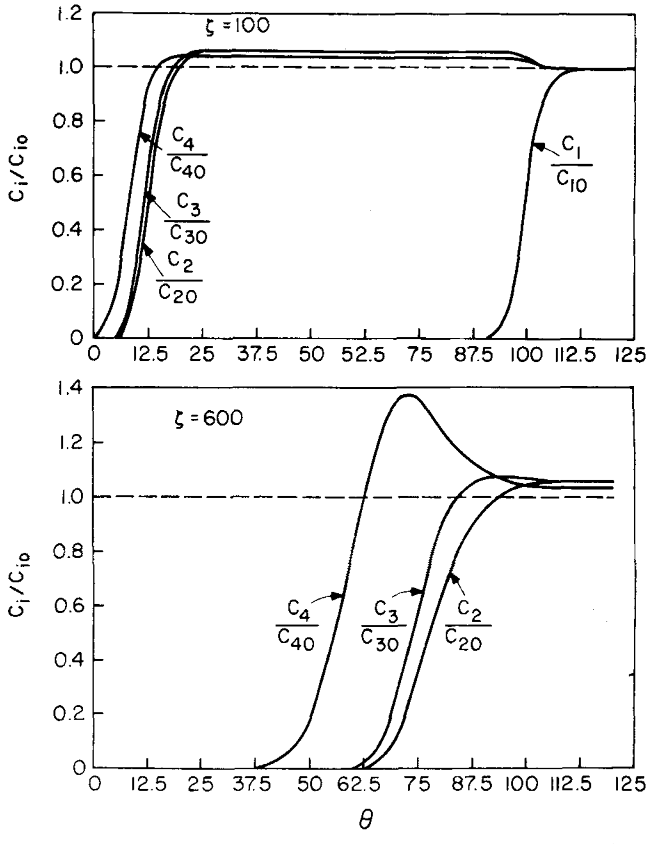

Fig. 10. Effluent profiles of four-solute system consisting of ABS(C_1), D-glucose(C_2), DL-valine(C_3), and lactic acid(C_4).

The component having the most nonlinear isotherm is defined as the key component ($i = 1$), while the others are defined as nonkey components ($i = 2, 3, \ldots m$).

The behavior of effluent profiles in a two-component system is entirely different from that in the one solute system. The effluent profiles of ABS (c_1) and DNOSBP (c_2) are plotted in Figure 9. The profiles are computed at the exit of single beds, with three different values of dimensionless bed depths: $\zeta = 4$, $\zeta = 16$, and $\zeta = 32$. In this system, the effluent profile of DNOSBP (c_2) shows a peak, indicating a concentration higher than the feed

Table 3. Examples of Optimization Runs

(a) Values of parameters used ($M = 0.1$ and $G = 1$ in the examples)

System	Solute	α_i	β_i'	$\gamma_i' \times 10^{-3}$	f_i	δ
One solute	2, 4-D	2.0	18.96	20.0	1.00	5.835
Two solute	ABS ($i = 1$)	5.0	0.3414	303.3	0.486	—
	DNOSBP ($i = 2$)	0.2	0.4448	265.2	0.514	—
Four solute	ABS ($i = 1$)	1.295	0.7351×10^{-3}	15.367	0.0188	—
	D-glucose ($i = 2$)	0.2843	0.7000×10^{-3}	1.157	0.1692	—
	DL-valine ($i = 3$)	0.1129	0.6771×10^{-3}	1.083	0.1353	—
	Lastic acid ($i = 4$)	0.2414	0.3755×10^{-3}	0.720	0.6767	—

(b) Optimal solutions

System	Bed arrangement	n	L	T	Bed volume (m^3)	Switching time (day)	Treatment cost (\$/10^6 gal)
One solute	Parallel	5	272.0	44.88	7.190	13.1	106.32
	Serial	4	69.25	61.50	9.156	17.9	100.57
Two solute	Parallel	3	2.7	0.50	6.607	123.3	27.51
	Serial	3	0.87	0.61	6.364	149.7	25.55
Four solute	Parallel	19	475.0	2.560	7.112	1.21	884.71
	Serial	17	11.59	1.202	3.297	0.57	840.02

concentration ($c_2/c_{20} > 1$). Similar observations were reported earlier by Zwiebel et al. (1974), Hsieh et al. (1977), and Klaus et al. (1977).

The peak in Figure 9 is believed to arise from competitive adsorption between the two solutes. In the earlier stages of adsorption, both ABS (c_1) and DNOSBP (c_2) are adsorbed on the activated carbon. Later, since the key component (ABS) has the stronger affinity, it replaces the nonkey component (DNOSBP), which then appears in the effluent as a concentration peak.

Note that for $\zeta = 4$ and $\zeta = 16$, the wave fronts of c_1 and c_2 cross each other (in the low concentration range). With a larger bed depth, say $\zeta = 32$, for example, one can observe that the wave fronts of c_1 and c_2 start to separate. The tendency toward front separation as ζ is increased reinforces the prediction of the asymptotic theory that with an infinite bed depth, the wave fronts are completely separated. An approximate asymptotic solution of a two-component system was given by Cooney and Strusi (1972), and a more rigorous derivation of the asymptotic theory of a two-component system was recently developed by Rhee and Amundson (1974).

Similar behavior of effluent concentration curves are observed in Figure 10 for the four-component system, where the key component is ABS (c_1) and the nonkey components are D-glucose (c_2), DL-valine (c_3), and lactic acid (c_4). With a dimensionless bed depth $\zeta = 100$, the wave front of ABS (c_1) appears at a dimensionless time θ of about 90, and the wave fronts of the other components (c_2, c_3, and c_4) all appear at about the same time, that is, $\theta < 15$. The wave fronts of c_2, c_3, and c_4 are crowded together because the values of their physical parameters (see Table 2) are close to one another. Likewise, concentration peaks occur only for the nonkey components (c_2, c_3, and c_4), which were at first adsorbed into the activated carbon in the bed. Since the key component (c_1) has the strongest adsorption affinity, it replaces other solutes already adsorbed in the activated carbon. Because of this replacement, the nonkey components are desorbed into the aqueous phase and then appear in the effluent at about $\theta < 15$. Since the key component (c_1) has by far the strongest adsorption affinity, it appears in the effluent only after a very long time, that is, when the bed has essentially exhausted its adsorption capacity. With a bed depth of $\zeta = 100$, the wave front of c_1 appears at about $\theta = 90$.

With a bed depth of $\zeta = 600$, not only do the adsorption fronts continuously move downstream, but the fronts are also further separated with time. This spatial spreading of wave fronts actually translates into a spreading of wave front arrivals at the bed exit. Consequently, the wave front of c_4 is further separated from those of c_2 and c_3, and the wave front of c_1 is so far behind that it does not even appear in Figure 10. In addition, the profile of c_4 has formed a sharp peak.

According to the theory of the asymptotic solution (Rhee and Amundson, 1974), the peaks of all nonkey components are separated in the effluent (the key component will appear last but will not appear as a peak). Since the asymptotic solution can be obtained under the condition of infinite bed depth ($\zeta = \infty$), computations were carried out to verify the tendency toward such peak separation by obtaining transient solutions at large values of ζ (at $\zeta = 2\,400$ and $\zeta = 3\,600$). The computed results (Sung, 1977) have confirmed the theoretical prediction of the asymptotic solution. However, limitation of space does not permit us to present those computational results in this paper.

Performance of the Optimally Designed Multistage Adsorption-Bed Systems

In this section, the optimal solutions under varying sets of parameter values are shown graphically. Three examples of optimization runs are given in Table 3. Complete details of optimization results including equipment sizes, operating condition, capital, and operating costs are tabulated elsewhere (Sung, 1977).

Effect of isotherm parameter. Using the parameter values $\beta' = 18.96$, $\gamma' = 20\,000$, and $\delta = 5.835$ for the single-component system, the isotherm parameter α was varied over the range of 1.0 to 10 to obtain the optimal designs of parallel-bed systems. The values of the minimum total treatment cost (objective function at the minimum value) are used to plot Figure 11.

It is seen that in the favorable region ($\alpha > 0$), where the isotherm is highly nonlinear, say $\alpha > 2$, the optimal

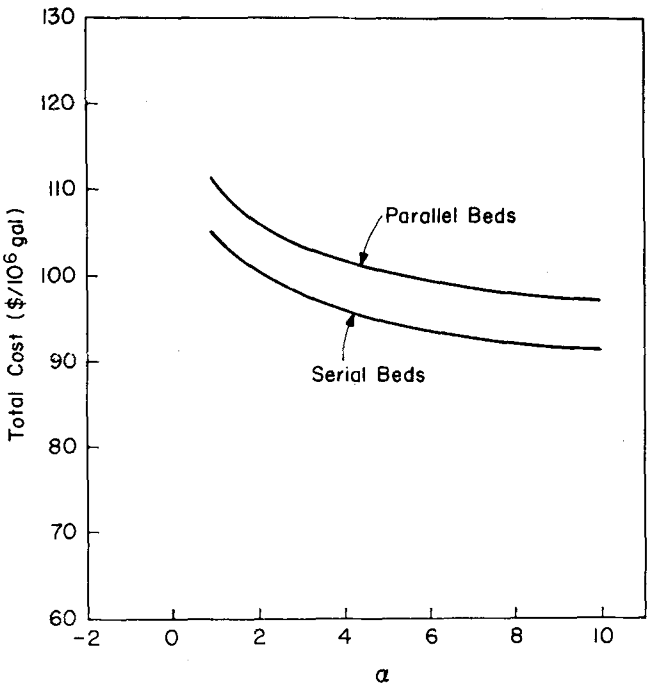

Fig. 11. Effect of isotherm parameter α (one-solute system) on total cost.

solution appeared to be less sensitive to changes in α. At low values of α, or in the region where α is close to linear isotherm ($\alpha = 0$), the minimum total cost starts to increase. Figure 11 shows that under optimal design, the serial-bed arrangement is uniformly better than the parallel-bed arrangement. Similar observations may be made in Figure 12 for the two- and four-component systems considered.

Effect of mass transfer parameter. Fixing the values $\alpha = 2$, $\gamma' = 20\,000$, and $\delta = 5.835$, the mass transfer parameter β' was varied for the system, over the range of 0.5 to 50. The results of this series of optimization runs are shown in Figure 13. Similar results are shown in Figure 14 for the two- and four-component systems. At large values of β', the optimal solution is less affected by change in β'. At the other extreme, when β' approaches zero, the minimum total cost rises extremely rapidly. This indicates that when the mass transfer is controlling, the cost of the adsorption process becomes high. Figures 13 and 14 show that under optimal design, the serial arrangement is uniformly better than the parallel arrangement.

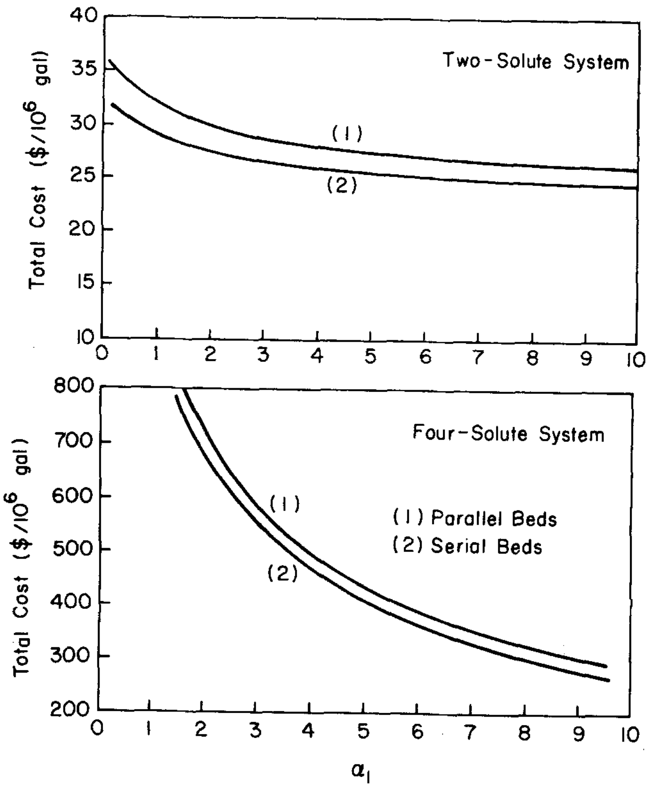

Fig. 12. Effect of isotherm parameter α_1 (two- and four-solute systems) on total cost.

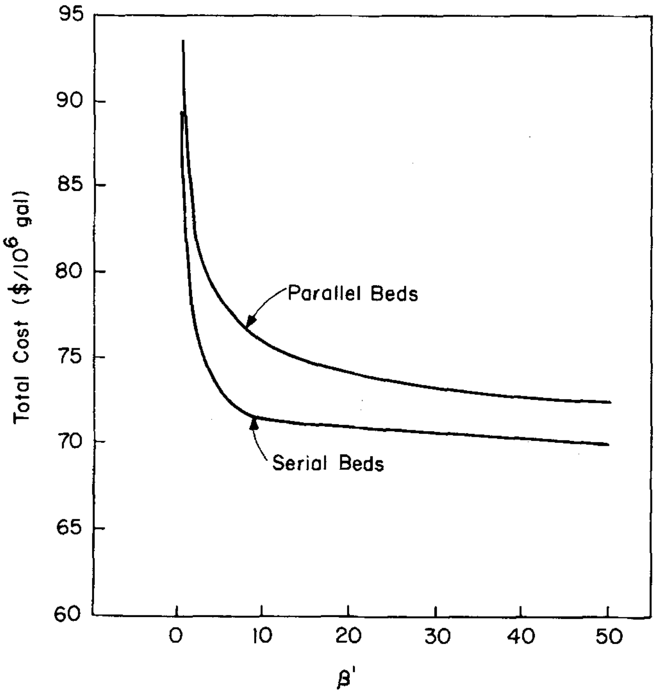

Fig. 13. Effect of mass transfer parameter β (one-solute system) on total cost.

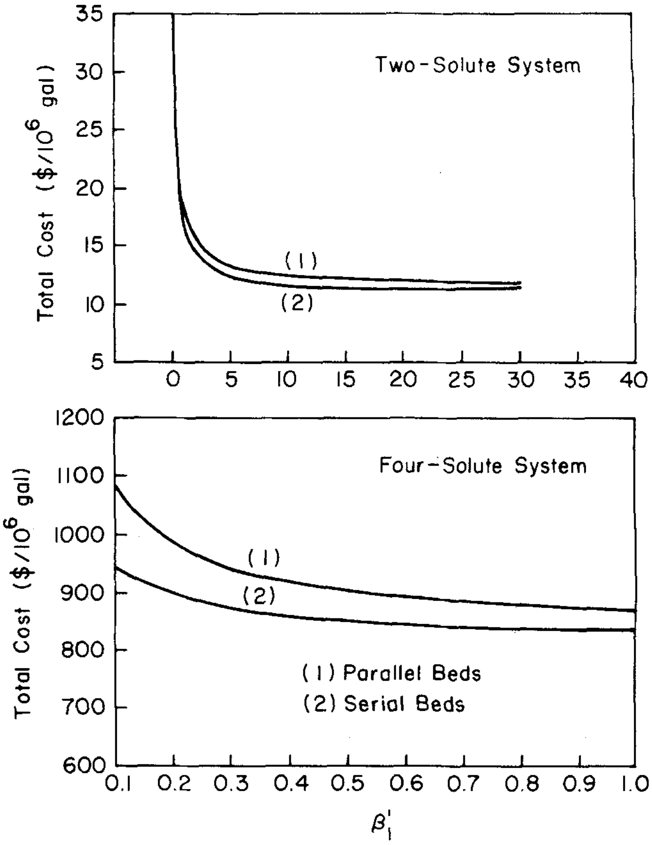

Fig. 14. Effect of mass transfer parameter β_1 (two- and four-solute systems) on total cost.

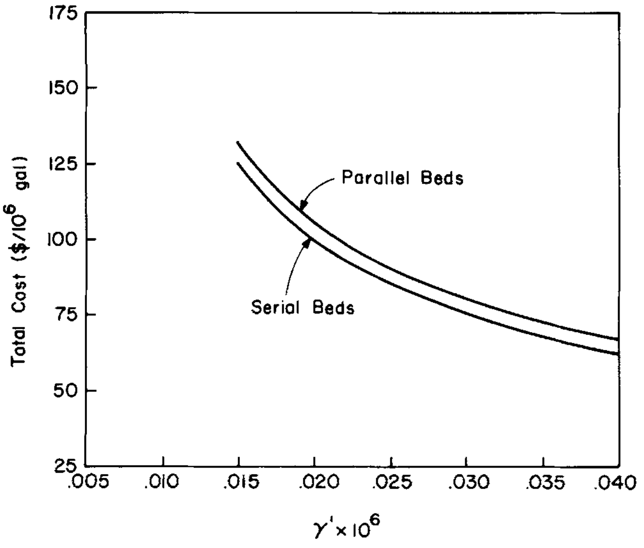

Fig. 15. Effect of capacity parameter γ' (one-solute system) on total cost.

Effect of capacity parameter. In Figure 15, the capacity parameter γ' for the single-component system was varied over the range of 0.015 to 0.04, with the other parameters held constant ($\alpha = 2$, $\beta' = 18.96$, and $\delta = 5.835$). Similar results are shown in Figure 16 for the two- and four-component systems. The results of this series of runs indicate that the optimal solution is extremely sensitive to the capacity parameter. This parameter is a measure of how much solute the carbon is capable of adsorbing, assuming that the carbon is at total saturation ($c^* = c_0$). It is expected, therefore, that the minimum total cost rises extremely rapidly as the adsorption capacity of the carbon is diminished. Again, under the optimal design, the serial arrangement appears to be uniformly better than the parallel arrangement.

Effect of purification parameter. The purification parameter M is defined as the fraction of solute from the feed remaining in the effluent. If a 95% removal of solute is required, the value of M is 0.05. Using the parameters of the single-component system ($\alpha = 2$, $\beta' = 18.96$, $\gamma' = 20\,000$, and $\delta = 5.835$), the value of M was varied over the range of 0.20 to 0.025 (80 to 97.5% solute removal). Figure 17 shows that the minimum cost increases as the

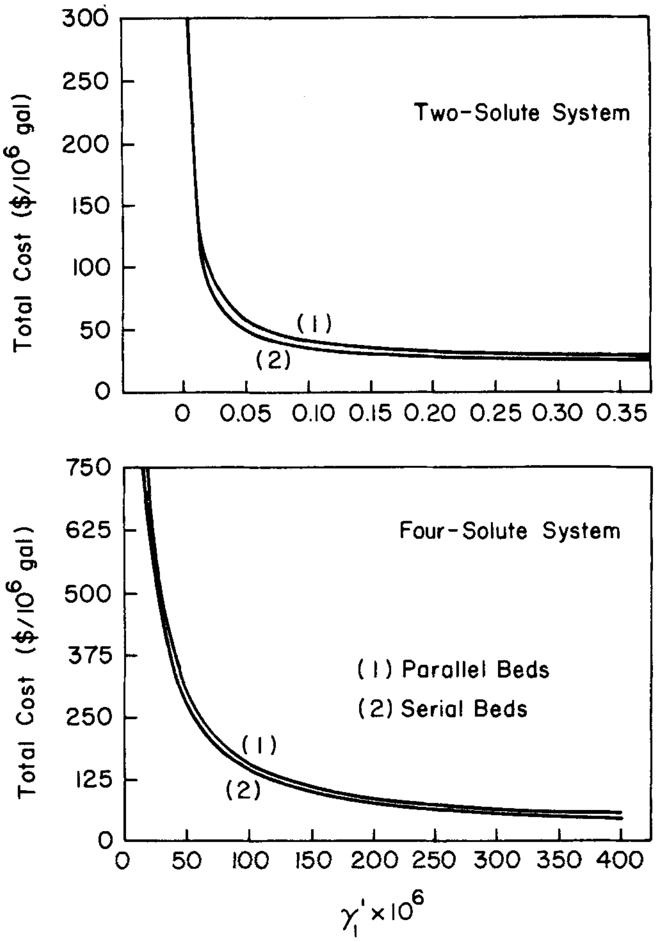

Fig. 16. Effect of capacity parameter γ'_1 (two- and four-solute systems) on total cost.

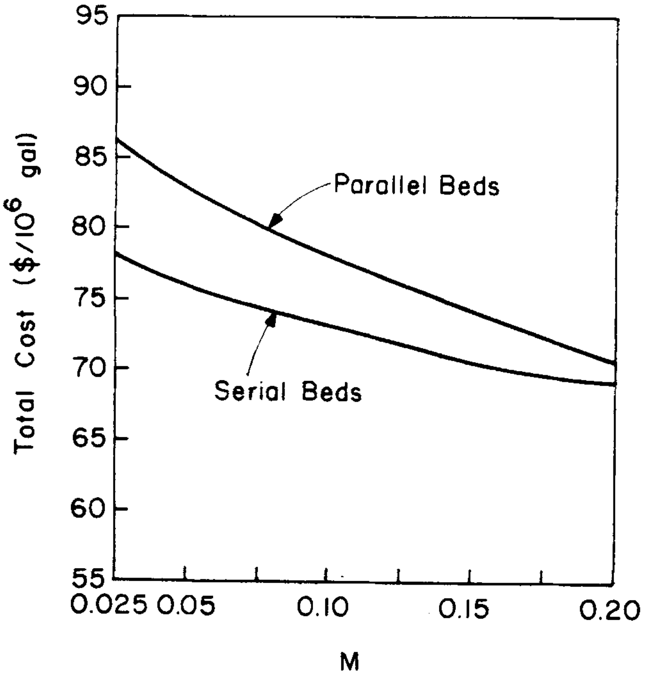

Fig. 17. Effect of purification parameter M (one-solute system on total cost.

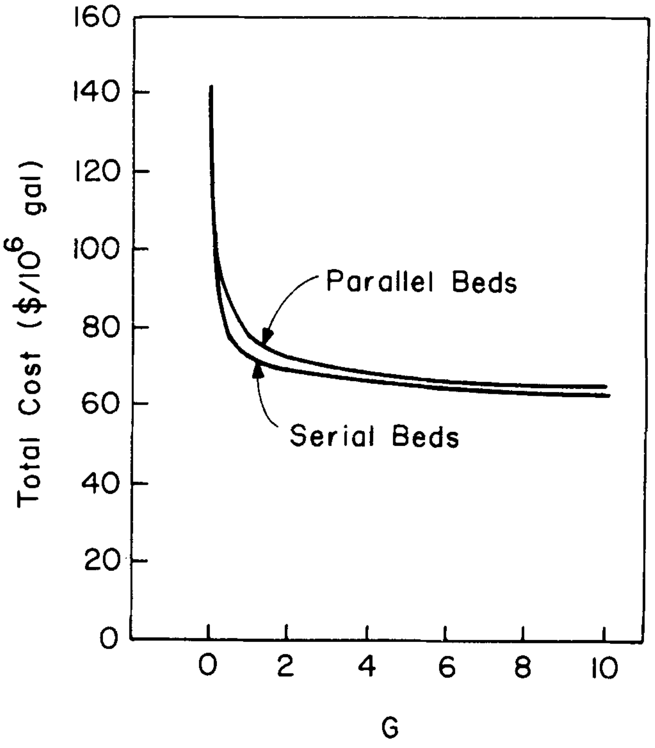

Fig. 18. Effect of stream size G (one-solute system) on total cost.

degree of purification increases. Figure 17 also shows that, under optimal design, the serial-bed arrangement is uniformly better than the parallel arrangement.

Effect of stream size. The size of input stream is a measure of adsorption plant size. Optimization runs were made in order to study whether there is any economic incentive to the design of a large plant. Using the parameter values of the single-component system, the stream size G was varied over the range of 0.1 to 10 million gal/day (MGD). The results shown in Figure 18 indicate that economy is not necessarily achieved in a large scale plant. However, when the stream size drops below 1 MGD, the cost rapidly increases. This result confirms also the general belief in the waste treatment industry that the adsorption process rapidly becomes less economically attractive when the stream size is less than 1 MGD.

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NOTATION

A_0 = fixed portion of capital cost
A_1 = incremental cost per cubic foot of equipment volume
A_2 = power factor related to number of beds in system
a = Langmuir constant (l/μ mole)
b = coefficient of equilibrium isotherm (l/mg)
C_I = cost index of year in which cost is to be estimated
$(C_I)_0$ = equipment cost index of base year
c = liquid phase concentration (μ mole/l)
$\bar{c}_{\max}$ = maximum effluent concentration of blended effluent (μ mole/l)
c_0 = feed concentration (μ mole/l)
c^* = liquid phase equilibrium concentration (μ mole/l)
d = bed depth (m)
f_i = defined as $f_i = c_{i0}/(c_{10} + c_{20} + \ldots + c_{m0})$
G = volumetric flow of input stream, million gal/day (MGD)
i = particular solute
$I(n, L)$ = objective function to be minimized (\$/million gal)
K = overall mass transfer coefficient (ml/g-min)
K_f = mass transfer coefficient in the film resistance model (ml/g-min)
K_s = solid phase mass transfer coefficient defined as $K_s' = K_s\rho/\epsilon$ (ml/g-min)
K_s' = solid phase mass transfer coefficient (l/min)
L = bed depth defined as $L = \rho kd/u$, or $L = \rho(K_f)_1 d/u$, *or* $L = \rho(K_s)_1 d/u$ (dimensionless)
L^* = optimal value of variable L
L_k = value of L at k^{th} iteration using the Fibonacci scheme (dimensionless)
M = purification parameter, the maximum effluent concentration based on $\Sigma c_i/(c_{10} + c_{20} + \ldots. + c_{m0})$
m = number of solutes
N = number of cycles
n = number of beds
n^* = optimal value of variable n
n_l = value of n at l^{th} iteration using the Fibonacci scheme
q = solid phase concentration (μ mole/mg)
q^* = interfacial solid concentration (μ mole/mg)
q_0 = solid concentration in equilibrium with c_0 (μ mole/mg)
t = time (hr)
t_s = switching time (hr)
T = switching time defined as $T = \beta T_s/\gamma$ (dimensionless)
u = superficial linear velocity (ml-fluid/cm^2/min)
V = bed volume (m^3)
V_b = equipment volume which is larger than the packed volume (m^3)
X = liquid phase concentration defined as $X = c/c_0$ (dimensionless)
Y = solid phase concentration defined as $Y = q/q_0$ (dimensionless)
z = axial position (m)

Greek Letters

α = equilibrium parameter defined as $\alpha = ac_0$ (dimensionless)
α_i = defined as $a_i c_{i0}$ (dimensionless)
β = mass transfer parameter defined as $\beta = \rho K/\epsilon$ (1/min)
β_i = defined as $\beta_i = \rho(K_f)_i/\epsilon$ or $\beta_i = \rho(K_s)_i/\epsilon = (K_s')_i$(1/min)
β', β_i' = mass transfer parameter defined as $\beta' = \beta\epsilon = \rho K$ and $\beta_i' = \beta_i\epsilon$ (ml-fluid/ml-bed/min)
γ, γ_i = capacity parameter defined as $\gamma = \rho q_0/\epsilon c_0$ and $\gamma_i = \rho q_{i0}/\epsilon c_{i0}$ (dimensionless)
γ', γ_i' = capacity parameter defined as $\gamma' = \gamma\epsilon/\rho = q_0/c_0$ and $\gamma_i' = q_{i0}/c_{i0}$ (l/mg)
δ = solid diffusion parameter (dimensionless)
ϵ = void fraction of the bed (ml-fluid/ml-bed)
ζ = axial position defined as $\zeta = \rho Kz/u$, or $\zeta = \rho(K_f)_1 z/u$, or $\zeta = \rho(K_s)_i z/u$ (dimensionless)
θ = time defined as $\theta = (\beta t - \zeta)/\gamma$ or $\theta = (\beta_1 t - \zeta)/\gamma_1$ (dimensionless)
θ_0 = constant defined as $\theta_0 = L/\gamma$ or $\theta_0 = L/\gamma_1$ (dimensionless)
θ_s = switching time defined as $\theta_s = \beta t_s/\gamma$ or $\theta_s = \beta_1 t_s/\gamma_1$ (dimensionless)
λ_i = defined as $\lambda_i = \beta_i/\beta_1$ (dimensionless)
μ_i = defined as $\mu_i = \gamma_i/\gamma_1$ (dimensionless)
ρ = bulk density of adsorbent (g/ml)
ϕ = liquid phase profile defined as $X = \phi(\zeta, \theta)$ (dimensionless)
ψ = solid phase profile defined as $Y = \psi(\zeta, \theta)$ (dimensionless)

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Effect of Flow Maldistribution on Conversion and Selectivity in Radial Flow Fixed-Bed Reactors

This paper examines the effect of flow maldistribution on the predicted conversion and selectivity in a radial flow fixed-bed reactor. A simple model was developed to relate gas maldistribution to the catalyst bed to the conversion with first- and second-order reactions and to selectivity for first-order reactions in series and parallel. For the cases considered, the conversion debit due to maldistribution of the gas phase was as much as 11%. The model was extended to cover adiabatic conditions for simple first-order reactions. For an exothermic reaction, maldistributions can enhance conversion over that obtained with a uniformly distributed gas phase.

PETER R. PONZI
and
LARRY A. KAYE
Exxon Research and Engineering Company
P.O. Box 101
Florham Park, New Jersey 07932

SCOPE

An advantage of radial flow fixed-bed reactors over axial flow units is the high flow surface area per volume of catalyst. This results in narrow, low pressure drop beds. Radial reactors with these low pressure drop configurations can be subject to uneven gas distribution along the axial length.

Fluid dynamic models of the gas distribution were developed by Dil'man et al. (1971) and Genkin et al. (1973) and extended by Kaye (1978). Lamba and Dudukovic (1975) investigated the effect of radial dispersion by the catalyst bed on conversion. The consequences of maldistributed gas flow to the catalyst bed has not been studied and is a concern in the design of low pressure drop, radial reactors. The objective of this study was, therefore, to assess the magnitude of the effects of maldistribution on reactor operation.

A derivation is presented of a simple model which relates inlet flow distribution to conversion and selectivity. The model assumes that the gas moves radially in plug flow fashion through parallel elements of the catalyst bed without axial flow by either convection or dispersion and without radial dispersion. Both isothermal and adiabatic operation are considered. For the purpose of this study, the axial velocity profiles were approximated by a simple power law expression.

Correspondence concerning this paper should be addressed to Peter R. Ponzi.