Invariant Imbedding, Iterative Linearization, and Multistage Countercurrent Processes

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The equations for multistage countercurrent operations are difference equations of the boundary-value type, which are generally solved by trial and error or iterative procedures. The invariant imbedding concept is used to solve these boundary-value problems as initial-value problems. It is shown that by the combined use of invariant imbedding and iterative linearization, a sequence of estimator equations for the missing terminal conditions is obtained. These estimator equations can be used to obtain the desired terminal conditions and the number of stages required to perform a specified separation.

In a recent paper (*1*), the invariant imbedding concept has been applied to various boundary-value problems in chemical engineering. However, only boundary-value problems represented by differential equations, or the continuous boundary-value problems, were discussed. The purpose of this paper is to apply the invariant imbedding concept to countercurrent stagewise diffusional separation processes.

Continuous diffusional separation processes can be represented by differential equations of the boundary-value type, while stagewise processes can be represented by finite difference equations of the boundary-value type. Since most of these boundary-value problems, such as the equations representing a multicomponent distillation column, cannot be solved analytically, the numerical solution of these problems is very tedious and generally involves a trial-and-error iterative procedure. It will be shown that by the use of the concept of invariant imbedding, the two-point boundary-value problem can be reduced to initial-value problems in partial difference equations. Further reduction of the partial difference equations into a system of ordinary difference equations of the initial-value type can be obtained if the original equations are linear difference equations. Thus, an iterative procedure can be formulated by iteratively linearizing the nonlinear equations by the Newton-Raphson type of formula. Owing to the rapid convergence rate of the Newton-Raphson iteration scheme, the present approach appears to be a useful tool for solving diffusional separation processes.

DISCRETE INVARIANT IMBEDDING

The invariant imbedding concept has been applied to continuous problems represented by differential equations (*1, 2*). We shall show that this same concept can also be applied to discrete or stagewise problems represented by finite difference equations. Consider the system represented by the finite difference equations

$$x(k+1) = f[x(k), y(k), k] \tag{1}$$

$$y(k+1) = g[x(k), y(k), k] \tag{2}$$

with boundary conditions

$$x(0) = b, \quad y(N) = c \tag{3}$$

with $0 \leq k \leq N-1$.

The system represented by Equations (1) to (3) constitutes a two-point, nonlinear, boundary-value problem. To avoid computationally difficulties, we wish to find the missing final condition $x(N)$. The missing initial condition can be obtained in essentially the same manner.

From Equations (1) and (2), we have

$$\Delta x(k) = x(k+1) - x(k) = f[x(k), y(k), k] - x(k) = F[x(k), y(k), k] \tag{4}$$

$$\Delta y(k) = y(k+1) - y(k) = g[x(k), y(k), k] - y(k) = G[x(k), y(k), k] \tag{5}$$

Instead of the one problem represented by Equations (3) to (5), consider a family of problems represented by Equations (4) and (5) and the more general boundary condition

$$x(0) = b, \quad y(n) = c \tag{6}$$

with $0 \leq k \leq n-1$. In other words, we wish to consider a family of problems. Thus, n and c are considered as variables. Notice that in this family of processes the missing final condition $x(n)$ is a function only of two parameters c and n. Define $r(c, n)$ as the missing final condition for the system represented by Equations (4) to (6), where the process ends with stage n with the given final condition $y(n) = c$. From this definition, we have

$$r(c, n) = x(n) \tag{7}$$

Following the same procedure for the continuous case (*1*), we shall consider a neighboring process which ends at stage $n+1$. Obviously

$$x(n+1) = x(n) + F[x(n), y(n), n]$$
$$= r(c, n) + F[r(c, n), c, n] \quad (8)$$

On the other hand, the following expression can be obtained for this missing final condition by considering the definition of $r(c, n)$

$$x(n+1) = r[y(n+1), n+1] \quad (9)$$

where

$$y(n+1) = y(n) + G[x(n), y(n), n]$$

Thus, Equation (9) becomes

$$x(n+1) = r[y(n) + G\{x(n), y(n), n\}, n+1]$$
$$= r[c + G\{r(c, n), c, n\}, n+1] \quad (10)$$

Equating Equations (8) and (10), we have

$$r(c, n) + F[r(c, n)], c, n]$$
$$= r[c + G\{r(c, n), c, n\}, n+1] \quad (11)$$

To simplify the expression, the right-hand side of Equation (11) can be approximated by

$$r[c+G, n+1] = r(c, n) + \frac{\delta r}{\delta c} G + \frac{\delta r}{\delta n} + \frac{\delta^2 r}{\delta c \delta n} G \quad (12)$$

For some problems, other second-order terms and higher-order terms may also be needed to obtain a fairly accurate result. However, the manipulations may become very tedious. Another way to overcome this difficulty is to use the iterative scheme to be discussed later. The expressions $\delta r/\delta c$ and $\delta r/\delta n$ are the partial differences

$$\frac{\delta r}{\delta c} = \frac{r(c+G, n) - r(c, n)}{(c+G) - c} = \frac{r(c+\Delta c, n) - r(c, n)}{\Delta c} \quad (13)$$

$$\frac{\delta r}{\delta n} = \frac{r(c, n+1) - r(c, n)}{(n+1) - n} = \frac{r(c, n+1) - r(c, n)}{\Delta n} \quad (14)$$

Substituting Equation (12) into Equation (11), we have

$$\frac{\delta r(c, n)}{\delta n} + \left[\frac{\delta r}{\delta c} + \frac{\delta^2 r}{\delta c \delta n}\right] G[r(c, n), c, n]$$
$$= F[r(c, n), c, n] \quad (15)$$

which is the desired invariant imbedding equation. If the process had zero stages, the missing final condition would be equal to the given initial condition. Thus, the initial condition for Equation (15) is

$$r(c, 0) = x(0) = b \quad (16)$$

for all possible values of c. Equation (15) can also be written as

$$\frac{\delta r(c, n)}{\delta n} + \left[\frac{\delta r(c, n)}{\delta c} + \frac{\delta^2 r(c, n)}{\delta c \delta n}\right]$$
$$[g[r(c, n), c, n] - c] = f[r(c, n), c, n] - r(c, n) \quad (15a)$$

The relationships in Equations (4) and (5) have been used to obtain Equation (15*a*).

The missing final condition r can be obtained by solving either Equation (11) or Equation (15). However, the numerical solution of Equation (11) is generally much more tedious than the numerical solution of Equation (15) (*2*). The $(n+1)$ stage process is imbedded into the n stage process on the right-hand side of Equation (11). Thus, in order to solve Equation (11), we must have a table which contains the value of r of the n stage process for all possible values of c. If the problem has a fairly large number of the parameter c, say c is three dimensional, a computer with a fairly large memory may be required. Thus, we encounter the dimensionality difficulty which has severely limited the usefulness of dynamic programming.

COUNTERCURRENT MULTISTAGE PROCESS WITH CONSTANT FLOW RATES

The equations encountered in countercurrent stagewise separation processes are difference equations of the boundary-value type. For example, the equations representing a multicomponent distillation column or a liquid-liquid extraction column are generally nonlinear difference equations, which are fairly difficult to solve. The invariant imbedding approach can be used to overcome some of the computational difficulties. This is especially true in view of the fact that in the design of multistage countercurrent processes, we are more interested in the terminal or end conditions. The conditions inside a column is generally of less interest to the designer. Since the invariant imbedding procedure calculates directly the end conditions, it is ideally suited for countercurrent processes. Notice that Equation (15) is an expression for the missing final condition or the desired terminal condition in terms of the given terminal condition and the total number of stages n.

A multistage countercurrent separation process is shown in Figure 1, which may represent any simple diffusional separation processes such as extraction or absorption. The flow rates of the two phases are assumed constant and are represented by L and V. The concentrations of the transferable component in the L and V phases are x and y, respectively. Notice that the numbering of the concentration y is different from the literature (*3, 4*). As we shall see, this numbering system results in two first-order simultaneous difference equations.

In the design of multistage separation processes, the inlet conditions of both streams are completely given. Thus, L, $x(0)$, V, and $y(N)$ are known values. The problem is to find the number of stages N so that $x(N)$ equals to a specified value. Another design problem is that given the separation equipment or the number N, we wish to find the terminal value $x(N)$. Notice that in either design problem we are only interested in the values of N and $x(N)$. By the use of invariant imbedding, an expression for $x(N)$ in terms of N and the given terminal conditions can be obtained.

Instead of only one transferable component, let us consider a system with m components. The material balance equations are

$$Lx_i(k-1) + Vy_i(k) = Lx_i(k) + Vy_i(k-1)$$
$$i = 1, 2, \ldots, (m-1) \quad (17)$$

where x_i and y_i represent the concentrations of component i in the L and V phases, respectively. Since the streams leaving stage k are in equilibrium, the following equilibrium relationships can be established under constant temperature and pressure:

$$x_i(k) = f_i[y_1(k-1), y_2(k-1), \ldots y_{m-1}(k-1)], \quad i = 1, 2, \ldots, (m-1) \quad (18)$$

The boundary conditions for these difference equations are

$$x_i(0) = b_i, \quad y_i(N) = c_i, \quad i = 1, 2, \ldots, (m-1) \quad (19)$$

For simplicity, this system can be represented by

$$x_i(k) = f_i$$

$$y_i(k) = g_i[x_i(k-1), y_1(k-1), y_2(k-1), \ldots, y_{m-1}(k-1)] = g_i \quad (20)$$

If we define

$$r_i(c_1, c_2, \ldots, c_{m-1}, n) = x_i(n) \quad (21)$$

the invariant imbedding equations are

$$\frac{\delta r_i(\mathbf{c}, n)}{\delta n} = \sum_{j=1}^{m-1} \left[\frac{\delta r_i(\mathbf{c}, n)}{\delta c_j} + \frac{\delta^2 r_i(\mathbf{c}, n)}{\delta c_j \delta n} \right] (g_j - c_j) = f_i - r_i(\mathbf{c}, n), \quad i = 1, 2, \ldots, (m-1) \quad (22)$$

The initial conditions for Equation (22) are

$$r_i(\mathbf{c}, 0) = x_i(0) = b_i, \quad i = 1, 2, \ldots, (m-1) \quad (23)$$

for all values of $\mathbf{c}$. The symbol $\mathbf{c}$ represents a vector with $(m-1)$ components.

COMPUTATIONAL CONSIDERATION

The discrete imbedding equation, Equation (22), can be solved by various techniques. Since it is a partial difference equation, the various techniques developed for solving partial differential equations such as the method of characteristics can be used. As has been discussed in an earlier paper (*1*), another way to solve this equation is to solve the original discrete equation, Equation (11). It should be noted that the amount of computation involved is fairly large to solve Equation (11) or the partial difference equations. This is especially true when the problem involves several given and missing final conditions, and, thus, a system of partial difference equations in more than two independent variables such as Equation (22) must be solved. To simplify the computations, the following approximation schemes can be used.

Linear Approximation

Let us assume that the invariant imbedding equation, Equation (15*a*), has a solution of the following form:

$$r(c, n) = \eta(n) + \zeta(n)c + \nu_1(n)c^2 + \nu_2(n)c^3 \ldots \quad (24)$$

If either the value of c is a small number, say in the neighborhood of $c = 0$, or the original system equations are linear difference equations, the higher-order terms in Equation (24) can be neglected, and Equation (24) reduces to

$$r(c, n) = \eta(n) + \zeta(n)c \quad (25)$$

Equation (25) is an exact expression for all possible values of c if Equations (1) and (2) are linear. On the other hand, if Equations (1) and (2) are nonlinear, Equation (25) is an approximate expression in the neighborhood of small c. Although Equation (15*a*) is an expression for all possible values of c, it should be noted that we are only interested in the value of c where $y(N) = c$. If $y(N)$ is much less than 1, Equation (25) is a fairly good approximation of the problem. Furthermore, since $y(N)$ in a countercurrent process is a concentration term which is less than unity, Equation (25) can generally be used as a first approximation. Substituting Equation (25) into (22) and assuming that the system has one transferable component, we have

$$\eta(n+1) - \eta(n) + c[\zeta(n+1) - \zeta(n)] + \frac{L}{V}\zeta(n+1) [f(c) - \eta(n) - \zeta(n)c] = f(c) - \eta(n) - \zeta(n)c \quad (26)$$

For any given problem, the function $f(c)$ is a known function. Thus, Equation (26) can be reduced to a system of simultaneous difference equations. For example, if

$$f(c) = a_1 + a_2 c \quad (27)$$

then Equation (26) can be reduced to the following two simultaneous first-order difference equations by equating terms of like powers in c:

$$\eta(n+1) = \frac{L}{V}\eta(n)\ \zeta(n+1) - \frac{La_1}{V}\zeta(n+1) + a_1 \quad (28)$$

$$\zeta(n+1) = \frac{L}{V}\zeta(n)\ \zeta(n+1) - \frac{La_2}{V}\zeta(n+1) + a_2 \quad (29)$$

For a system with one transferable component, Equation (23) reduces to Equation (16). To find the boundary conditions for Equations (28) and (29), compare Equations (16) and (25) at $n = 0$ and recall that Equation (16) must be true for any value of c. We obtain

$$\eta(0) = b, \quad \zeta(0) = 0 \quad (30)$$

Equations (28) and (29) can now be solved recursively by using the initial conditions given by Equation (30). Once these equations are solved, the missing final condition $x(N)$ can be obtained from Equations (21) and (25). It should be noted that the numerical solution of the initial value problem represented by Equations (28) to (30) is much easier than the numerical solution of the boundary-value problem represented by Equations (17) to (19).

Suppose that the function $f(c)$ is of form

$$f(c) = a_1 + a_2 c + a_3 c^2 + a_4 c^3 \quad (31)$$

and the value of c is not very small such that the term $a_4 c^3$ is not negligible, then we can consider approximate the solution of the invariant imbedding equation by the cubic equation, Equation (24). After substituting this cubic equation into the imbedding equation and equating terms of like powers in c, we obtain four simultaneous difference equations. The initial conditions for these four simultaneous difference equations are

$$\eta(0) = b, \quad \zeta(0) = \nu_1(0) = \nu_2(0) = 0 \quad (32)$$

The numerical solution of these four first-order difference equations is still much easier than the numerical solution of the original boundary-value problem represented by Equations (17) to (19).

Since the original equations are linear except for the function $f(c)$, and also since Equation (27) or (31) is generally adequate to represent the equilibrium relationship, the technique discussed above is generally adequate to solve a large number of diffusional separation problems with constant flow rates. Furthermore, even if the equilibrium relation is very nonlinear and thus the above proce-

dure can only give an approximate solution, this approximate solution can still be improved by techniques to be discussed in later sections.

COUNTERCURRENT PROCESSES WITH VARIABLE FLOW RATES

Consider the multistage process shown in Figure 2, which may represent any simple diffusional separation processes. However, for concrete illustration, let us consider it as an extraction process. A mixture containing solvent A and solute C is to be extracted by a second solvent B. We shall assume that the solvents are miscible. The concentrations of C and A in the original mixture and raffinate are represented by x_1 and x_2, respectively. The corresponding components in the extract stream are represented by y_1 and y_2. The flow rates of the raffinate and extract streams are represented by L and V, respectively. We wish to reduce the concentration of component C in the raffinate or L stream. For systems with variable flow rates, it is convenient to introduce the following nomenclature:

$$l_i(k) = L(k)x_i(k), \quad v_i(k) = V(k)y_i(k), \quad i = 1, 2, 3 \tag{33}$$

Mass balances on each component give

$$l_i(k-1) + v_i(k) = l_i(k) + v_i(k-1), \quad i = 1, 2, 3 \tag{34}$$

There are six unknowns, l_i, v_i, $i = 1, 2, 3$. In addition to the above three equations, three more equations can be obtained from the equilibrium relationship. Typical equilibrium data for liquid-liquid extraction of ternary systems are represented by triangular coordinates (3, 4). For multicomponent systems, equilibrium ratios, or K values are used. Equilibrium ratios will be used in this work. However, in order to consider nonideal systems, K will be considered as a function of the compositions. Since the streams $L(k)$ and $V(k-1)$ are in equilibrium, we have

$$l_i(k) = \frac{\sum_{j=1}^{3} l_j(k)}{K_i \sum_{j=1}^{3} v_j(k-1)} v_i(k-1), \quad i = 1, 2, 3 \qquad k = 1, 2, \ldots, N \tag{35}$$

With fixed temperature and pressure, the functions K_i are functions of the composition

$$K_i = K_i(y_1(k-1)) = K_i\left[\frac{v_1(k-1)}{\sum_{j=1}^{3} v_j(k-1)}\right] \tag{36}$$

Since we only have three components in two phases, according to the phase rule only the concentration of one component is needed to specify the equilibrium. The boundary conditions for Equations (34) and (35) are

$$l_i(0) = b_i, \quad v_i(N) = c_i, \quad i = 1, 2, 3 \tag{37}$$

which are the feed compositions of the L and V streams.

Equations (34), (35), and (37) represent a two-point boundary-value problem. In a design problem we frequently specify the desired concentration of a certain component in the outlet streams. Suppose that the value of $x_1(N)$ must be below a certain specified value. The problem is to find N so that the specified $x_1(N)$ is satisfied. In the invariant imbedding approach, a series of values of $x_1(N)$ will be obtained. Thus, instead of considering the one problem with a fixed N, let us consider a series of problems with N and c_i as parameters. The boundary condition for this general problem is

$$l_i(0) = b_i, \quad v_i(n) = c_i, \quad i = 1, 2, 3 \tag{38}$$

Equations (34) and (35) can be rewritten as

$$l_i(k+1) - l_i(k) = \frac{\sum_{j=1}^{3} l_j(k+1)}{K_1 \sum_{j=1}^{3} v_j(k)} v_i(k) - l_i(k)$$

$$= f_i(l_1(k+1), l_2(k+1), l_3(k+1), l_i(k), v_1(k), v_2(k), v_3(k)) \tag{39}$$

$$v_i(k+1) - v_i(k) = l_i(k+1) - l_i(k)$$

$$= g_i[l_i(k+1), l_i(k)] \tag{40}$$

where Equation (36) was used. To obtain the invariant imbedding equation, we define

$$r_i(c_1, c_2, c_3, n) = l_i(n) \tag{41}$$

with $v_i(n) = c_i$, $i = 1, 2, 3$. The invariant imbedding equations can be obtained in essentially the same manner as discussed earlier. However, since the variables of the $(k+1)$ stage, $l(k+1)$, are present implicit in the functions f and g, the resulting invariant imbedding equations are much more complicated. These invariant imbedding equations are

$$\frac{\delta r_i(\mathbf{c}, n)}{\delta n} + \sum_{j=1}^{3}\left[\frac{\delta r_i(\mathbf{c}, n)}{\delta c_j} + \frac{\delta r_i^2(\mathbf{c}, n)}{\delta c_j\, \delta n}\right] g_j[l_j(n+1), r_j(\mathbf{c}, n)] = f_i[\mathbf{l}(n+1), r_i(\mathbf{c}, n), \mathbf{c}], \quad i = 1, 2, 3 \tag{42}$$

The initial conditions for Equation (42) are

$$r_i(\mathbf{c}, 0) = l_i(0) = b_i, \quad i = 1, 2, 3 \tag{43}$$

where $\mathbf{c}$ and $\mathbf{l}$ are three-dimensional vectors with components c_1, c_2, c_3 and l_1, l_2, l_3, respectively.

Equations (42) and (43) are the desired equations. Notice that the functions g and f are not only functions of the variables of the n stage process but also functions of the variables of the $(n+1)$ stage process. Expressions for the $(n+1)$ stage variables, $\mathbf{l}(n+1)$, must be obtained. The variable $l(n+1)$ can be approximated by

$$l_i(n+1) = r_i[\mathbf{v}(n+1), n+1] = r_i[\mathbf{v}(n) + \mathbf{g}, n+1]$$

$$= r_i(\mathbf{c}, n) + \sum_{j=1}^{3} \frac{\delta r_i}{\delta c_j} g_j[l_j(n+1, r_j] + \frac{\delta r_i}{\delta n} + \sum_{j=1}^{3} \frac{\delta^2 r_i}{\delta c_j\, \delta n} g_i[l_j(n+1), r_j] \tag{44}$$

with higher-order terms neglected. By using the above equation, Equation (42) becomes

$$\frac{\delta r_i}{\delta n} + \sum_{j=1}^{3}\left[\frac{\delta r_i}{\delta c_j} + \frac{\delta r_i^2}{\delta c_j\, \delta n}\right] g_j\left\{ r_j + \sum_{k=1}^{3} \frac{\delta r_j}{\delta c_k} g_k[l_k(n+1), r_k]\right.$$

$$+\frac{\delta r_j}{\delta n}+\sum_{k=1}^{3}\frac{\delta^2 r_j}{\delta c_k\,\delta n}\,g_k[l_k(n+1),r_k],r_j\Big\}$$

$$=f_i\Big\{\ r_1+\sum_{j=1}^{3}\frac{\delta r_1}{\delta c_j}\,g_j[l_j(n+1),r_j]+\ldots,r_2+$$

$$\sum_{j=1}^{3}\frac{\delta r_2}{\delta c_j}\,g_j[l_j(n+1),r_j]+\ldots,r_3+$$

$$\sum_{j=1}^{3}\frac{\delta r_3}{\delta c_j}\,g_j[l_j(n+1),r_j]+\ldots,r_i,\mathbf{c}\Big\} \quad (45)$$

The function g in Equation (45) is again a function of $l(n+1)$. Equation (44) can again be used to eliminate $l(n+1)$ in Equation (45). This elimination process can be continued indefinitely. However, if we neglect all the higher-order terms in the partial differences in Equation (45), only the expressions $l(n+1)$ in the function f_i on the right-hand side of Equation (45) need to be eliminated by using Equation (44). Any further elimination results in higher-order terms and thus can be neglected. Since the expressions for the functions f and g are known for a given problem, Equation (45) can be reduced into three fairly complex nonlinear partial difference equations. With the given initial conditions, Equation (43), these nonlinear partial difference equations of the initial-value type can be solved at least in theory.

To simplify the calculations, linear approximation can again be used to obtain approximate solutions for Equation (42) or (45). Instead of Equation (25), the following approximate solution can be assumed for Equation (42) if the value of c is small:

$$r_i(\mathbf{c},n)=\eta_i(n)+\sum_{j=1}^{3}\zeta_{ij}(n)\,c_j,\quad i=1,2,3 \quad (46)$$

First, Equation (44) is substituted into the right-hand side of Equation (45), and then Equation (46) is used to eliminate the variable r_i. Neglecting higher-order terms and equating terms involving like powers of c_i, we obtain a system of twelve ordinary difference equations of the Riccati type (2). The initial conditions for this system of equations are

$$\zeta_{ij}(0)=0,\quad i,j=1,2,3$$
$$\eta_i(0)=b_i,\quad i=1,2,3 \quad (47)$$

This approximate solution may be accurate enough for a number of design problems. However, if better accuracy is needed, higher-order terms of c can be included in the assumed approximate solution. However, a fairly large number of ordinary difference equations must be solved when the higher-order terms of c are included. To avoid this difficulty, the iterative linearization scheme discussed in the next section should be used.

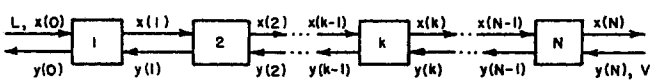

Fig. 1. Countercurrent multistage process with constant flow rates.

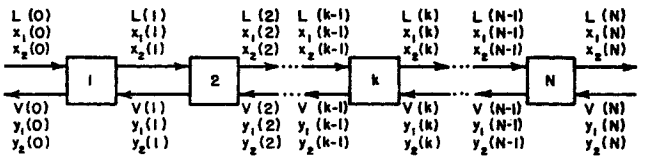

Fig. 2. Countercurrent process with variable flow rates.

ITERATIVE LINEARIZATION

It should be noted that the primary difficulty in using invariant imbedding to the separation process with variable flow rates is due to the nonlinear nature of the process equations. If Equation (39) were linear, the variable $l(k+1)$ could be solved explicitly. The functions f and g would be only functions of the variables of the k stage process and independent of $l(k+1)$. Thus, the manipulations in Equations (44) and (45) would have been unnecessary. Furthermore, if Equations (39) and (40) were linear, Equation (46) would represent the exact solution of the invariant imbedding equation. This suggests a computational scheme by the combined use of invariant imbedding and one of the iterative linearization schemes. The Newton-Raphson type of linearization, also known as *discrete quasilinearization*, will be used in the following discussions.

A general system of first-order nonlinear difference equations can be represented by

$$\mathbf{f}[\mathbf{x}(n+1),\ \mathbf{x}(n),\ n]=0 \quad (48)$$

with boundary conditions

$$x_i(0)=b_i,\quad i=1,2,\ldots,M$$
$$x_i(N)=c_i,\quad i=(M+1),\ldots,m \quad (49)$$

where $\mathbf{x}$ and $\mathbf{f}$ are m-dimensional vectors. It should be noted that an m^{th} order nonlinear difference equation can be reduced to m first order simultaneous equations (5). Thus, Equation (48) is a fairly general expression. This equation can be linearized by the use of Taylor series with orders higher than the first omitted (2). Expanding this equation around $\mathbf{x}_k(n+1)$ and $\mathbf{x}_k(n)$, we have

$$\mathbf{f}\,[\mathbf{x}_k(n+1),\ \mathbf{x}_k(n),\ n]$$
$$+\mathbf{J}_{\mathbf{x}(n+1)}\,[\mathbf{x}_k(n+1)]\,[\mathbf{x}_{k+1}(n+1)-\mathbf{x}_k(n+1)]$$
$$+\mathbf{J}_{\mathbf{x}(n)}\,[\mathbf{x}_k(n)]\,[\mathbf{x}_{k+1}\,(n)-\mathbf{x}_k(n)]=0 \quad (50)$$

where $\mathbf{J}_{\mathbf{x}(n)}\,[\mathbf{x}_k(n)]$ and $\mathbf{J}_{\mathbf{x}(n+1)}\,[\mathbf{x}_k(n+1)]$ are the Jacobian matrices with respect to $\mathbf{x}(n)$ and $\mathbf{x}(n+1)$, respectively. If we consider the variables where the subscript k are known variables and are obtained from the previous k^{th} iteration, Equation (50) represents a system of linear equations which can be rewritten as

$$\mathbf{B}_1(n+1)\mathbf{x}_{k+1}(n+1)=-\mathbf{B}_2(n)\mathbf{x}_{k+1}(n)+\mathbf{C}$$

or

$$\mathbf{x}_{k+1}(n+1)=-\mathbf{B}_1{}^{-1}(n+1)\mathbf{B}_2(n)\mathbf{x}_{k+1}(n)$$
$$+\mathbf{B}_1{}^{-1}C=\mathbf{A}(n)\mathbf{x}_{k+1}(n)+\mathbf{B}(n)=\mathbf{g} \quad (51)$$

Suppose we wish to obtain the missing final conditions $x_{1,k+1}(N), x_{2,k+1}(N), \ldots x_{M,k+1}(N)$. Let

$$r_i(c_{M+1}, c_{M+2}, \ldots, c_m, n) = x_{i,k+1}(n), \quad i = 1, 2, \ldots, M \tag{52}$$

The invariant imbedding equations are

$$\frac{\delta r_i}{\delta n} + \sum_{j=M+1}^{m} \left[\frac{\delta r_i}{\delta c_j} + \frac{\delta^2 r_i}{\delta c_j \, \delta n} \right] (g_j - c_j) = g_i - r_i, \quad i = 1, 2, \ldots, M \tag{53}$$

with the initial conditions

$$r_i(c_{M+1}, \ldots, c_m, 0) = b_i, \quad i = 1, 2, \ldots, M \tag{54}$$

Since Equation (51) is linear, the solution of Equation (53) must be of the form

$$r_i = \eta_i(n) + \sum_{j=M+1}^{m} \zeta_{ij}(n) c_j, \quad i = 1, 2, \ldots, M \tag{55}$$

By substituting Equation (55) into Equation (53) and by equating terms with like powers of c, a system of ordinary difference equations can be obtained. The initial conditions for these ordinary difference equations are

$$\zeta_{ij}(0) = 0; \quad i = 1, 2, \ldots, M; \quad j = M + 1, \ldots, m$$
$$\eta_i(0) = b_i, \quad i = 1, 2, \ldots, M \tag{56}$$

An iterative computational scheme can now be formulated (*2*). With an initial approximation $\mathbf{x}_{k=0}(n)$, $n = 1, 2, \ldots, N$ the invariant imbedding equations can be solved for the missing final conditions. Then an improved set of values $\mathbf{x}_{k=1}(n)$ can be obtained from the original equations, and an improved set of the missing final conditions can again be obtained by using $\mathbf{x}_{k=1}(n)$ and the invariant imbedding equation. This iterative procedure can be continued until the desired accuracy is obtained. Since the Newton-Raphson type of formula converges quadratically if there is convergence, computational experience has shown that in general only three to five iterations are needed to obtain a fairly high accuracy.

MULTICOMPONENT DISTILLATION

Consider a multicomponent distillation column in which the plates are numbered from the top to the bottom. The column has a total condensor, and the top plate is the first plate. Let $x_i(k)$ and $y_i(k)$ represent the concentrations of the i^{th} component leaving the k^{th} stage in the liquid and vapor streams, respectively. For simplicity, constant vapor and liquid flow rates in the rectifying and stripping sections will be assumed. This assumption eliminates the need for enthalpy balances. Thus only material balance and equilibrium relationships need be considered. The usual material balances for each component in the rectifying section are (*6, 7*)

$$Vy_i(n + 1) = Lx_i(n) + Dx_{i,D}, \quad i = 1, 2, \ldots, m - 1 \tag{57}$$

From the equilibrium ratio, the equilibrium relationships are

$$y_i(n) = K_i(n) \; x_i(n) \tag{58}$$

Combining Equations (57) and (58), we obtain

$$VK_i(n + 1)x_i(n + 1) = Lx_i(n) + Dx_{i,D}, \quad i = 1, 2, \ldots, m - 1 \tag{59}$$

where $K_i(n)$ is the equilibrium distribution ratio and is a function of the concentration and temperature.

Equation (59) represents $(m - 1)$ first-order difference equations. Let us consider the boundary conditions in distillation column design calculations. In general, the feed compositions $x_{i,f}$, feed flow rate F, and the thermal condition of the feed are completely known. The concentration of the heavy key in the distillate or overhead, x_{HD}, and the concentration of the light key in the bottom, x_{LB}, are specified. The values of V and L are determined by the specification of reflux ratio and feed. For a total condenser, $x_i(0) = y_i(1) = x_{i,D}$. Since $x_{HD} = x_H(0)$ is given and $x_{LD} = x_L(0)$ can be obtained by material balance, the two given initial conditions for Equation (59) are

$$x_H(0) = x_{HD}, \quad x_L(0) = x_{LD} \tag{60}$$

All the other initial conditions are missing. In the usual trial-and-error distillation calculations, these missing initial conditions are assumed. These assumed values are adjusted until the composition on the feed plate calculated in the rectifying section matches that calculated in the stripping section.

In the invariant imbedding approach, expressions for these missing initial conditions are obtained as functions of the given initial conditions and the number of stages in the rectifying section. Define

$$r_i(c_H, c_L, n) = x_i(n), \quad i = 3, 4, \ldots, (m - 1) \tag{61}$$

with $x_H(0) = c_H$ and $x_L(0) = c_L$. We have assumed that the first two components with $i = 1, 2$ are the two key components.

By applying the procedure discussed earlier, a system of invariant imbedding equations can be obtained. A similar set of equations can be obtained for the stripping section.

It is interesting to note that if the system is a binary mixture, or $m = 2$, Equation (59) is no longer a boundary-value problem, since specifying one concentration in the overhead product determines Equation (59) uniquely.

The above approach can be extended easily to the problem with variable flow rates. In addition to Equation (59), the enthalpy balances are also needed. These enthalpy balance equations can be obtained in the usual manner.

A SIMPLE EXAMPLE

To illustrate the technique and also to compare it with the traditional methods of approach, a simple illustrative example solved by Treybal (*8*) will be solved by the present approach. A coal gas is to be freed of its light oil by scrubbing it with wash oil. The light oil is assumed to be entirely benzene. Let l_i and v_i represent moles per hour of component i in the coal gas and wash oil streams, respectively. Let the subscripts $i = 1, 2, 3$ denote the components benzene, coal gas, and wash oil, respectively. From the notations used in Figure 2, the following feed conditions can be obtained from Treybal (*8*):

$$L(0)x_1(0) = l_1(0) = 1.612 \qquad V(0)y_1(0) = v_1(N) = 0.0675$$
$$L(0)x_2(0) = l_2(0) = 79.0 \qquad V(0)y_2(0) = v_2(N) = 0$$
$$L(0)x_3(0) = l_3(0) = 0 \qquad V(0)y_3(0) = v_3(\mathrm{N}) = 13.4 \tag{62}$$

The problem is to find the total number of stages N so that $l_1(N)$ can be reduced to 0.0791. The equilibrium ratio is $K_1(k) = 8$. Coal gas and wash oil will be assumed immiscible. Thus, the values of $l_2(k)$, $l_3(k)$, $v_2(k)$, and $v_3(k)$ remain the same throughout the column. Equation (40) reduces to

$$v_1(k+1) - v_1(k) = l_1(k+1) - l_1(k) = g_1 \quad (63)$$

and Equation (39) becomes

$$l_1(k+1) - l_1(k) = \frac{l_1(k+1) + 79}{8[v_1(k) + 13.4]} v_1(k) - l_1(k) = f_1 \quad (64)$$

The invariant imbedding equation, Equation (42), is reduced to

$$\frac{\delta r_1(c_1, n)}{\delta n} + \left[\frac{\delta r_1(c_1, n)}{\delta c_1} + \frac{\delta^2 r_1(c_1, n)}{\delta c_1 \, \delta n} \right] g_1 = f_1 \quad (65)$$

with the initial condition

$$r_1(c_1, 0) = l_1(0) = 1.612 \quad (66)$$

By using Equations (44) through (46) and the procedure discussed there, the following two simultaneous difference equations can be obtained:

$$7\eta_1(n+1) + 186.2\zeta_1(n+1) - 7\eta_1(n)\zeta_1(n+1) - 107.2\zeta_1(n)\zeta_1(n+1) - 79 = 0 \quad (67)$$

$$\eta_1(n+1) = \eta_1(n)\zeta_1(n+1) \quad (68)$$

Equation (67) is obtained by equating the terms involving c, and Equation (68) is obtained by equating the terms which do not involve c. From the above two equations, the following simplified expression can be obtained:

$$\zeta_1(n+1) = \frac{11.3}{26.6 - 15.3\zeta_1(n)} \quad (69)$$

According to Equation (47), the initial conditions are

$$\zeta_1(0) = 0, \quad \eta_1(0) = 1.612 \quad (70)$$

From these initial conditions, Equations (68) and (69) can be solved easily. Once the values of ζ_1 and η_1 are known, the value of $l_1(N)$ can be obtained from

$$l_1(N) = r_1(c_1, N) = \eta_1(N) + \zeta_1(N)c_1 \quad (71)$$

with $c_1 = v_1(N) = 0.0675$. Approximate hand calculation shows that a value of $N = 7.8$ is required to reduce $l_1(N) = 0.0791$. Treybal (8) obtained a value of $N = 7.7$.

DISCUSSION

It should be emphasized that invariant imbedding is a concept, not a technique or method. Thus, various different formulations can be used to obtain the invariant imbedding equations. Some of these different approaches are discussed in references 2 and 9. Furthermore, instead of concentrations other imbedding parameters can also be considered. For example, expressions for distillate rate as functions of other parameters and the number of stages may be desired in some calculations. Invariant imbedding expressions for this distillate rate can also be obtained.

Since the value of r of the previous stage is imbedded in the current stage on the left-hand side of Equation (11), a trial-and-error or iterative procedure is needed. In addition, we must have a table for r as a function of c for the previous stage in order to calculate the current stage. Thus, this approach is more time consuming than the usual or traditional approaches for this simple process. For more complex process such as the one shown in Figure 2, this approach results in three invariant imbedding equations for the three missing final conditions. Furthermore, the functions r_i must be functions of the three given final conditions, $V(N+1)$, $y_1(N+1)$, and $y_2(N+1)$. The table for r as functions of three variables may require a fairly large amount of fast memory in the computer. Thus, the dimensionality difficulty which has severely limited the usefulness of dynamic programming also limits the effectiveness of this approach (2).

As has been emphasized in an earlier paper (1), the main advantage of the invariant imbedding technique is its versatility. Since the concept is completely different from the usual approach, various computationally and theoretical advantages can frequently be obtained from the new formulation. Another advantage of this technique is that instead of solving one problem, a family of problems are solved. Since in design problems we frequently wish to study the neighboring processes, this approach can provide the answer for all possible values of the parameters. The reader can consult the literature for further discussions on the advantages and disadvantages of the invariant imbedding technique (2).

Obviously, the invariant imbedding approach can also be used for solving problems in continuous countercurrent processes. However, since the invariant imbedding equations for continuous processes represented by differential equations have been studied extensively (2, 10), a discussion of the application of invariant imbedding to continuous countercurrent processes will not be given here.

Since the equations representing countercurrent multistage processes are of the boundary-value type, the traditional methods for solving multistage processes are essentially trial-and-error procedures which are very tedious and time-consuming. Instead of solving the boundary-value problem directly, the present approach changes the original problem into difference equations of the initial-value type which can be solved fairly routinely and in a straightforward manner on modern computers. In addition, computational experiences have shown that the convergence rate of the quasilinearization technique is very fast and is fairly independent of the number of variables. In general, only three to five iterations are needed to obtain a five digit accuracy with a very approximate initial approximation by this technique (2). Thus, in general the present approach should require less computation time and also should be easier to use than the traditional approaches, provided fairly good initial approximations are available. This technique should be especially effective for problems with a large number of components in which a large number of the boundary conditions are missing. The quasilinearization technique has been found to be very effective for solving chemical reactor problems which are highly nonlinear and which involve the Arrhenius nonlinear exponential expression (2). This technique should be very effective for solving multistage separation processes which are much less nonlinear than the Arrhenius expression (11).

Since we have avoided the direct numerical solution of the original invariant imbedding equation such as Equation (11), the present approach requires a very small amount of computer memory compared with dynamic programming. Furthermore, since we have also avoided the direct numerical solution of the original invariant imbedding

partial difference equation such as Equation (15), (22), or (42), the memory requirement of the present approach should be about the same order of magnitude as the traditional methods. It should be noted that we encounter the dimensionality difficulty only when we solve Equation (11) directly on the computer.

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NOTATION

b = given initial condition
c = given final condition
D = distillate rate
$\mathbf{J}$ = Jacobian matrix
K = equilibrium ratio
L = flow rate of L phase
$l_i(k)$ = moles per hour of component i in the L phase from stage k
m = number of components or total number of state variables
N = total number of stages
n = stage number, a parameter
r = missing final or missing initial condition
V = flow rate of V phase
$v_i(k)$ = moles per hour of component i in the V phase entering stage k
$x_i(k)$ = concentration of i^{th} component leaving the k^{th} stage in the L phase
$y_i(k)$ = concentration of i^{th} component entering the k^{th} stage in the V phase
η, ζ, ν = functions defined by Equation (24) or (46)

Subscripts

D = distillate
f = feed
H = heavy key
i = i^{th} component
k = k^{th} iteration, assumed known
$k+1$ = $(k+1)$ iteration, assumed unknown
L = light key

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COMMUNICATIONS TO THE EDITOR

Vapor-Liquid Equilibria of the Hydrochloric Acid-Water System

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According to the criterion that the chemical potential of a component is the same in all phases at equilibrium, one writes

$$\mu_v = \mu_L \qquad (1)$$

From the definition of activity and fugacity, these are given by

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