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# Iterative Methods for Solving Problems in Multicomponent Distillation at the Steady State

**D. S. BILLINGSLEY and G. W. BOYNTON**

**IBM, Houston, Texas**

**The multicomponent distillation problem is formulated so that unknown compositions do not appear explicitly either in the equilibrium and material balance equations or in the heat balance equations. Use of this formulation in absolute iteration as well as Newton-Raphson iteration is discussed. Other types of staged separation processes may be similarly expressed**

Several authors have proposed Newton-Raphson or similar schemes for solving steady state multicomponent distillation problems. Among these are Stainthorp et al. (*11, 12*), Naphtali (*7*), and Greenstadt et al. (*5*). In addition, Newman (*8*) presented a worthwhile scheme for obtaining the temperature profile corresponding to a specified phase flow profile. In these methods the unknown flow rates or mole fractions of individual components enter explicitly into the iterations. When component enthalpies and equilibrium ratios are independent of composition (heats of mixing are neglected), this explicit dependence may be removed, thereby greatly reducing the number of unknowns entering the iterative procedure.

Figure 1 illustrates indexing conventions and some notations. Component enthalpies and equilibrium ratios are assumed to be independent of composition. This assumption imposes no real limitation because nonideal mixtures can be handled as suggested in reference *3*. It will be assumed also that each feed stream is completely specified and that the heat duty $Q_q$ of each intercooler and interheater (other than the reboiler and overhead condenser) is specified. The total withdrawal rate of each side-draw, if any, will be specified, and each side-draw will be liquid. For purposes of discussion, a total condenser producing bubble point reflux is employed and any two of the following are specified: internal reflux ratio $R$, reboiler duty $Q_R$, bottom product withdrawal rate $B$, and condenser duty $Q_C$.

The special algorithm for Gaussian elimination, as applied to tridiagonal matrix equations (see reference *2* or *9*), is presumed to be used for solving all such systems otherwise numerical instability will normally result. The reader is referred to Pease (*10*) concerning differentiation of negative powers of matrices.

## ITERATION EQUATIONS

Following Ball (*1*), Equation (1) was derived previously (*2*).

$$Z_i^{-1}\ \Omega_i = \Upsilon_i \equiv (v_{iJ}, v_{iJ-1}, \ldots, v_{i1})^T \tag{1}$$

where

$$\Omega_i = (\omega_{iJ-1}, \omega_{iJ-2}, \ldots, \omega_{i0})^T$$

$$\omega_{ij} = \begin{cases} f_{ij} & \text{if } j \text{ denotes a feed stage} \\ 0 & \text{otherwise} \end{cases}$$

and the tridiagonal matrix $Z_i = (z_{mn})_i$ with

$$z_{mni} = \begin{cases} A_{iJ-n} + 1 & m = n \neq 1 \\ A_{iJ-1} + 1 - R & m = n = 1 \\ -A_{iJ-n} & m - 1 = n \neq j_s \equiv js \\ -\left[1 - \dfrac{W_s}{L^+_{J-n}}\right] A_{iJ-n} & m - 1 = n = j_s \equiv js \\ -1 & m = n - 1 \\ 0 & \text{otherwise} \end{cases}$$

The solution to Equation (1) has been given by Cullinan (*4*); his expressions, however, are not so suitable for computation as is Equation (1) in the form used here.

Summing Equation (1) over components produces

$$V \equiv (V_J, V_{J-1}, \ldots, V_1)^T = \sum_i \Upsilon_i = \sum_i Z_i^{-1}\,\Omega_i \qquad (2)$$

which is recast as

$$\Lambda(T, \mathfrak{A}) \equiv V - \sum_i \Upsilon_i = V - \sum_i Z_i^{-1}\,\Omega_i = 0 \qquad (3)$$

where

$$T^T \equiv (T_{J-1}, T_{J-2}, \ldots, T_0)$$

| $\mathfrak{A}^T \equiv$ | Specified |
|---|---|
| $(T_J, V^T)$ | $R, Q_c$ |
| $(T_J, V^T)$ | $R, Q_R$ |
| $(T_J, V_{J-1}, V_{J-2}, \ldots, V_1)$ | $R, B$ |
| $(R, T_J, V^T)$ | $Q_c, Q_R$ |
| $(T_J, V^T)$ | $Q_c, B$ |
| $(T_J, V^T)$ | $Q_R, B$ |

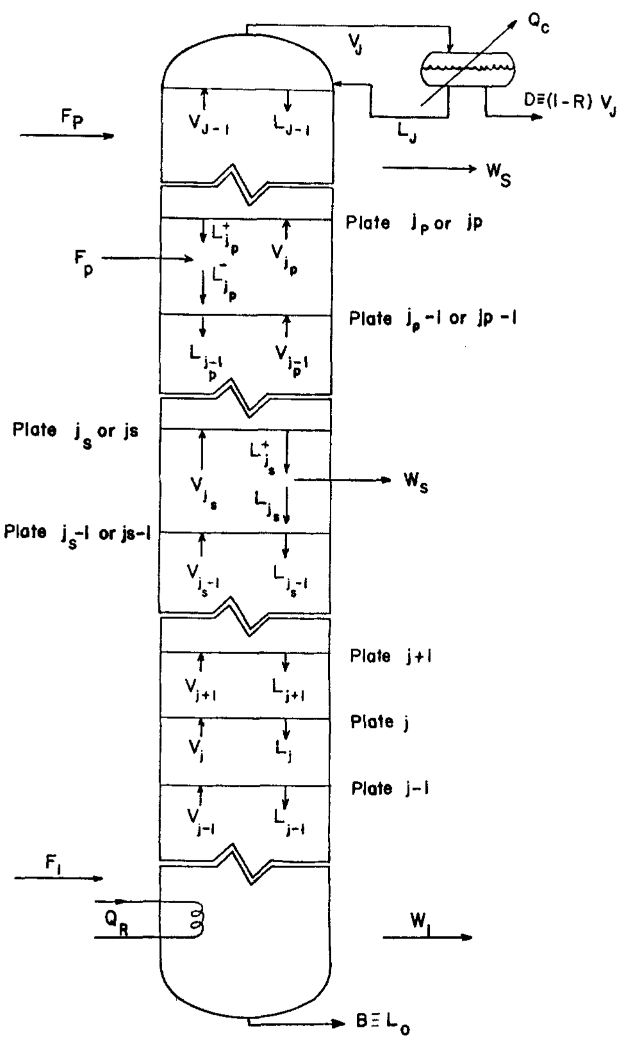

**Fig. 1. Complex distillation column.**

Note that $\Lambda$ does not explicitly involve unknown compositions because $Z_i$ does not. Note further that the reflux bubble point relation is not included in Equations (1) to (3). It would be in the case of a partial condenser which would appear simply as an additional stage.

The heat balance equations may now be placed in a form that is also independent of unknown compositions. The usual heat balances are

$$\left.\begin{array}{l} \sum\limits_i [v_{ij+1} H_{ij} + l_{ij}h_{ij} - v_{ij}H_{ij-1} - l_{ij+1}h_{ij+1}] = Q_j \\ \qquad\qquad 1 \leq j < J,\ j + 1 \neq j_s \equiv js \\ \sum\limits_i [v_{i1} H_{i0} + l_{i0}h_{i0} - l_{i1}h_{i1}] = Q_R \equiv Q_0 \\ \sum\limits_i (H_{iJ-1} - h_{iJ})v_{iJ} = Q_c \end{array}\right\} \qquad (4)$$

where

$$Q_j = \begin{cases} F_pH_p & \text{if the } p^{\text{th}} \text{ feed is on stage } j \\ Q_q & \text{if the } q^{\text{th}} \text{ heat exchanger other than the reboiler or overhead condenser is at stage } j \\ 0 & \text{otherwise} \end{cases}$$

The second of Equations (4) is used only if $Q_R$ is specified. The last of Equations (4) is employed only when $Q_c$ is specified.

If the $s^{\text{th}}$ side-draw is taken from stage $j + 1$ (that is, $j + 1 = j_s \equiv js$), then the last term on the left side of the first (or second) of Equations (4) becomes $-[1 - (W_s/L^+{}_{j+1})]l^+{}_{ij+1}\,h_{ij+1}$. In any event substitution of the material and equilibrium relations

$$l_{ij} = A_{ij}\,v_{ij+1}, \quad j \neq J$$

$$l_{ij} + v_{ij+1} - l_{ij+1} - v_{ij} = \omega_{tj}, \quad j + 1 \neq j_s, \quad j < J$$

$$l_{ij} + v_{ij+1} - [1 - (W_s/L^+{}_{j+1})]l^+{}_{ij+1} - v_{ij} = \omega_{ij}, \qquad j + 1 = j_s$$

into the first two of Equations (4) produces

$$\left.\begin{array}{l} \sum\limits_i [\{H_{ij} + A_{ij}h_{ij} - (A_{ij} + 1)h_{ij+1}\}\,v_{ij+1} \\ \qquad - (H_{ij-1} - h_{ij+1})v_{ij} + h_{ij+1}\,\omega_{ij}] = Q_j \\ \qquad\qquad 0 \leq j < J, \quad v_{i0} \equiv 0 \equiv H_{i,-1} \end{array}\right\} \qquad (5)$$

As already mentioned, the reflux bubble point relation for a total condenser is excluded from $\Lambda$. Consequently

$$\sum_i (K_{iJ} - 1)v_{iJ} = 0 \equiv Q_J$$

together with Equations (5) will constitute the vector $\Gamma$

$$\Gamma(T, \mathfrak{A}) \equiv Q - \sum_i [\Phi_i\,\Upsilon_i + \hbar_i\,\Omega_i] = 0 \qquad (6)$$

where the bidiagonal matrix $\Phi_i = (\phi_{mn})_i$ and

$$\phi_{mni} = \begin{cases} K_{iJ} - 1, \quad m = n = 1 \\ -H_{iJ-n} + h_{iJ+2-n}, \quad m = n > 1 \\ H_{iJ-n} + A_{iJ-n}h_{iJ-n} - (A_{iJ-n} + 1)h_{iJ+1-n}, \\ \qquad\qquad\qquad\qquad m = n + 1 \\ 0 \qquad \text{otherwise} \end{cases}$$

$$Q \equiv (Q_J, Q_{J-1}, \ldots, Q_1)^T \text{ if } Q_R \text{ is unspecified}$$

$$Q \equiv (Q_J, Q_{J-1}, \ldots, Q_1, Q_R)^T \text{ if } Q_R \text{ is specified}$$

Finally, $\hbar_i$ is a diagonal matrix having $h_{iJ+2-m}$ as its $m^{\text{th}}$ diagonal element and where $h_{iJ+1} \equiv h_{iJ+2} \equiv 0$.

When $Q_c$ is specified

$$\sum_i (H_{iJ-1} - h_{iJ})v_{iJ} = Q_c \equiv Q_{J+1}$$

Table 1. Maximum Discrepancy in Heat Balance at Any Stage

| Iteration | 1 | 2 | 3 | 4 | 5 | 6 |
|---|---|---|---|---|---|---|
| $10^{-4}\lvert\Gamma\rvert$* | 152 | 90.3 | 13.8 | 1.03 | 0.011 | 0.001 |

* $\lvert\Gamma\rvert$ denotes the maximum of the magnitudes of the elements of $\Gamma$.

is included in $\Gamma$.

To emphasize that $\Gamma$ is independent of unknown compositions, Equations (1) and (6) are combined to obtain

$$\Gamma(T, \mathfrak{R}) \equiv Q - \sum_i \left[\Phi_i Z_i^{-1} + \hbar_i\right] \Omega_i = 0 \qquad (7)$$

In computation, however, it is convenient to determine vector $\mathbf{T}_i$ for use whenever the product $Z_i^{-1}\,\Omega_i$ is required.

## NEWTON-RAPHSON SCHEME

Equations (3 )and (7) permit Newton-Raphson iterations using

$$\left.\begin{array}{l} \dfrac{\partial \Lambda}{\partial T}\Delta T + \dfrac{\partial \Lambda}{\partial \mathfrak{R}}\Delta \mathfrak{R} = -\,\Lambda(T, \mathfrak{R}) \\[2ex] \dfrac{\partial \Gamma}{\partial T}\Delta T + \dfrac{\partial \Gamma}{\partial \mathfrak{R}}\Delta \mathfrak{R} = -\,\Gamma(T, \mathfrak{R}) \end{array}\right\} \qquad (8)$$

where the derivative of one vector with respect to another is interpreted as the Jacobian matrix which has as the element in its $m^{\text{th}}$ row and $n^{\text{th}}$ column the partial derivative of the $m^{\text{th}}$ element of the first vector with respect to the $n^{\text{th}}$ element of the second vector. $\Delta$ of course denotes a change in the vector it precedes.

Expressions for the required partial derivatives are readily obtained. Thus, differentiating Equation (3) provides

$$\left.\begin{array}{l} \partial \Lambda/\partial T_j = \sum_i Z_i^{-1}\,(\partial Z_i/\partial T_j)\mathbf{T}_i \\[2ex] \partial \Lambda/\partial V_j = I_j + \sum_i Z_i^{-1}\,(\partial Z_i/\partial V_j)\mathbf{T}_i \end{array}\right\} \qquad (9)$$

where $I_j$ denotes the $j^{\text{th}}$ column of the identity matrix. Differentiation of Equation (7) yields

$$\left.\begin{array}{l} \partial \Gamma/\partial T_j = \sum_i \left[\{\Phi_i Z_i^{-1}\,(\partial Z_i/\partial T_j) \right. \\ \qquad \left. -\,(\partial \Phi_i/\partial T_j)\}\mathbf{T}_i - (\partial \hbar_i/\partial T_j)\Omega_i\right] \\[2ex] \partial \Gamma/\partial V_j = \sum_i \left[\Phi_i Z_i^{-1}\,(\partial Z_i/\partial V_j) - (\partial \Phi_i/\partial V_j)\right]\mathbf{T}_i \end{array}\right\} \qquad (10)$$

It is seen that each of Equations (9) and (10) is a vector corresponding to a column in one of the matrices of Equation (8). Furthermore, each derivative matrix in Equations (9) and (10) is very sparse, having in general only two or four nonzero elements if $B$ and $R$ are specified. In this connection, one notes that since

$$L_j = V_j + B + \text{constant} \qquad (11)$$

there is

$$\frac{\partial L_n}{\partial V_j} = \begin{cases} 1 & \text{if } n = j \\ 0 & \text{if } n \neq j \end{cases}$$

$$\partial L_n/\partial B = 1 \ \text{ if } B \text{ is not specified.}$$

Equations similar to (9) and (10) are obtained when $R$ or $B$ is unspecified. The relation $-\,B + \text{constant} = D = (1 - R)V_J$ is used to obtain $\partial B/\partial R$, $B$, $\partial B/\partial V_J$, or $\partial R/\partial V_J$. When $R$ and $B$ are specified, of course $L_J$ and $V_J$ are known.

A partial condenser would be treated as usual by regarding it as an additional stage with $l_{J+1} \equiv 0$. If the partial condenser duty $Q_c$ were specified, it would be treated as an intercooler.

## BOYNTON'S METHOD

The second author has devised a computational procedure that is concisely described using Equations (3) and (8) and may be expected to exhibit characteristics similar to those of the Newton-Raphson procedure, Equations (8). In Boynton's scheme:

1. $T$ and $\mathfrak{R}$ are assumed initially.
2. The corrected (or assumed) value for $\mathfrak{R}$ is used in Equation (3) which is solved for $T$ by Newton-Raphson iterations using
$$(\partial \Lambda/\partial T)\Delta T = -\,\Lambda(T)$$
as proposed by Newman (8).
3. Equation (3) is next considered as defining $T$ implicitly in terms of $\mathfrak{R}$ so that
$$\Delta T = -\,(\partial \Lambda/\partial T)^{-1}\,(\partial \Lambda/\partial \mathfrak{R})\,\Delta \mathfrak{R}$$
Substitution of this expression into the second of Equations (8) provides
$$\left[\frac{\partial \Gamma}{\partial T}\left(\frac{\partial \Lambda}{\partial T}\right)^{-1}\frac{\partial \Lambda}{\partial \mathfrak{R}} - \frac{\partial \Gamma}{\partial \mathfrak{R}}\right]\Delta \mathfrak{R} = \Gamma\,(T, \mathfrak{R})$$
which is then solved once to yield a correction, $\Delta \mathfrak{R}$ to $\mathfrak{R}$.
4. Items 2 and 3 are repeated until $\Gamma \approx 0$.

Strictly speaking, Boynton's method differs in that his heat balance equations are written around stage $j$ and the reboiler, and material balance equations are not used to eliminate one of the component flow rates from the heat balance relations. Hence, his equations would have resulted in each matrix $\Phi_i$ containing an additional nonzero element in each row. Furthermore, in the program used the required partial derivatives were approximated by difference quotients.

Results obtained using Boynton's program to solve a 25-stage, six-component reboiled absorber with one intercooler are summarized in Table 1. The feed rates were 200 moles/hr. liquid on the top stage and 150 moles/hr. vapor five stages above the reboiler. The initial assumption for liquid phase flow was 350 moles/hr. leaving each stage. The average difference between this and the correct phase flow was slightly over 65 moles/hr., correctly indicating that this initial guess was not especially good.

## ABSOLUTE ITERATION

Equations (3) and (7) may be rearranged for what has been called absolute iteration (6), which is a generalization of direction iteration. To this end write

$$\sum_i \Phi_i^1 Z_i^{-1}\,\Omega_i = Q - \sum_i \left(\Phi_i^0 Z_i^{-1} + \hbar_i\right)\Omega_i \qquad (12)$$

where $\Phi_i^1$ is the diagonal matrix having the same main diagonal as $\Phi_i$ and $\Phi_i^0 \equiv \Phi_i - \Phi_i^1$. For absolute iteration $T$ and $\mathfrak{R}$ are assumed and substituted into the right side of Equation (2). The resulting vector $V$, together with Equation (11), determines the liquid phase flows $L_j$. These phase flows together with the assumed vector $T$ are substituted into $\Phi_i^0$, $Z_i^{-1}\,\Omega_i$, and $\hbar_i$ in Equation (12). In the resulting expression,

$$\sum_i \Phi_i^1\,\mathbf{T}_i = Q - \sum_i \left(\Phi_i^0\,\mathbf{T}_i + \hbar_i\,\Omega_i\right) \qquad (13)$$

the only unevaluated quantities are the $T_j$ in $\Phi_i^1$ which is diagonal. Thus Equation (13) provides a set of scalar nonlinear equations, the first of which involves only one unknown temperature. Except for $T_{J-1}$, each succeeding equation involves only one additional unknown temperature; hence, they may be solved successively each for a single $T_j$, rather than simultaneously. $T_{J-1}$ is calculated as the dew point of the $v_{iJ}$. If repeated applications of these procedures converge, the results are a solution to the problem. Various modifications of the procedure can be obtained such as splitting $\Phi_i$ in different ways.

Absolute iteration obviously requires much less computation per iteration than does the use of Equation (8), but its convergence characteristics are unknown.

## DISCUSSION

Equations (8) constitute an iterative method in which one seeks to satisfy all the relevant equations simultaneously. This is not the case where, for example, bubble points are used to determine stage temperatures and phase flows are computed at a fixed set of compositions. These methods are normally much faster (*3*), simply by virtue of the assumptions that have been made in order to decouple the equations. Their convergence is of course dependent upon the validity of those assumptions with respect to the problem for which a solution is desired.

When the "decoupling" assumptions are not valid, all relevant equations must be solved simultaneously. In this event there is a substantial increase in the computational effort required per iteration, and it is especially important to reduce the number of iteration variables and obtain the attendant reduction in the number of iteration equations. Elimination of all unknown component flow rates and mole fractions achieves such a reduction (*7, 14, 15*).

One untried procedure is to set $\Delta$Я $= 0$ in the second of Equations (9) and iterate to obtain $T$, and to set $\Delta T = 0$ in the first of Equations (9) and iterate to obtain Я. This might be expected to work well on absorber or stripper problems as it uses the heat balance to obtain $T$, which is what Sujata (*13*) did in a different way.

Equations (3) and (7) can be regarded as multidimensional (multistage) flash equations. It is of course possible to place Equation (3) in a form in which only the ratios $L_j/V_{j+1}$ appear and to use these ratios as independent variables in lieu of the $V_j$. Where side-draws are present, this turns out to be computationally less convenient than the formulations presented here.

## NOTATION

$A_{ij}$, $A_{iJ-m}$ = absorption factor for component $i$ at stage $j$ or $J - m$ respectively; $A_{ij} \equiv L_j/(V_{j+1}K_{ij})$
$B$ = total withdrawal rate of bottoms product; $B \equiv L_0$
$D$ = total withdrawal rate of overhead product
$f_{ij}$ = feed rate of component $i$ at stage $j$; $f_{ij} \neq 0$ only if stage $j$ is a feed stage
$F_p$ = total rate of feed in the $p^{th}$ external feed where the lowest external feed is $F_1$
$h_{ij}$, $h_{iJ-m}$ = enthalpy of liquid component $i$ leaving stage $j$ or $J - m$, respectively
$\mathcal{K}_i$ = diagonal matrix defined after Equation (6)
$H_{ij}$, $H_{iJ-m}$ = enthalpy of vapor component $i$ entering stage $j$ or $J - m$, respectively
$H_p$ = enthalpy of one mole of feed $F_p$
$I_j$ = $j^{th}$ column of the identity matrix
$J$ = number of stages in the column, including the reboiler but not a total condenser; top stage number is $J - 1$ when a total condenser is used; reboiler stage number is always 0
$K_{ij}$, $K_{iJ-m}$ = equilibrium ratio of component $i$ on stage $j$ or $J - m$, respectively
$l_{ij}$, $l_{ij}^+$ = flow of liquid component $i$ leaving stage $j$; superscript + denotes flow prior to side-draw
$L_j$, $L_j^+$ = total flow of liquid phase leaving stage $j$; superscript + denotes flow prior to side-draw
$Q$ = vector having the $Q_j$ as elements
$Q_c$ = condenser duty
$Q_j$ = heat input defined after Equation (4)
$Q_q$ = heat duty for the $q^{th}$ heat exchanger other than the reboiler or condenser; $Q_1$ denotes the lowest such heat exchanger; $Q_q$ is positive for an interheater, negative otherwise
$Q_R$ = reboiler duty
$R$ = internal reflux ratio; $R \equiv L_J/V_J$
$T$ = vector defined after Equation (3)
$T_j$ = temperature of liquid on stage $j$
$v_{ij}$, $v_{iJ-m}$ = rate of flow of vapor component $i$ entering stage $j$ or $J - m$, respectively
$V$ = vector defined in Equation (2)
$V_j$ = total rate of vapor flow entering stage $j$
$W_s$ = total rate of product withdrawal in the $s^{th}$ side-draw; $W_s$ leaves stage $j_s (\equiv js)$, $W_1$ is the lowest side-draw
$z_{mni}$ = quantity defined after Equation (1)
$Z_i$ = matrix defined after Equation (1)

### Greek Letters

$\Gamma$ = vector function defined by Equation (6)
$\Delta$ = "new value minus old value"
$\Lambda$ = vector function defined by Equation (3)
$\phi_{mni}$ = quantity defined after Equation (6)
$\Phi_i$ = matrix defined after Equation (6)
$\Phi_i^1$, $\Phi_i^0$ = matrices defined after Equation (12)
$\Upsilon_i$ = vector defined in Equation (1)
$\omega_{ij}$ = quantity defined after Equation (1)
$\Omega_i$ = vector defined after Equation (1)

### Russian Letter

Я = vector defined after Equation (3)

### Superscript

$T$ = transpose

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