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A New Simulation Method for Equilibrium Stage Processes

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This paper presents a new, general method for mathematical simulation of equilibrium stage operations. The procedure solves component material balance equations with a tridiagonal matrix algorithm. Heat balances and summation equations are handled with Broyden's method. The unique feature of this procedure is that, in a mathematical sense, all equations are solved simultaneously. Therefore, the method can be used for all types of equilibrium stage processes. Additionally, the use of Broyden iteration insures solutions which are both stable and more rapid than current techniques. An exact solution for a twenty tray column with twenty components takes approximately 30 sec. on an IBM 360/65 computer. Successful simulations have been made for both absorption and distillation type of operations which have included complex columns with multiple feeds and side product streams. Design applications of the method cover a variety of equilibrium stage processes in the chemical and petroleum industries.

Countercurrent flow, mass transfer columns are primary unit operations used for separation in the chemical and petroleum industries. Methods for solving the equations simulating specific types of these column processes are known (*1* to 7). There is, however, need for a more efficient, general calculation method which will predict component distributions and temperature profiles in all types of towers, even complex units with multiple feeds and widely varying temperatures and vapor flow rates. Procedures in current use as well as those reported in the literature work well for either absorption or distillation type of problems, but in general cannot be applied to both. Although many of the column processes used are of these two types, many do not conveniently fit into either category. Rich oil demethanizers in natural gas processing plants, for example, usually have absorption characteristics over a certain part of the column and distillation characteristics over another.

This paper describes a new method which provides stable, rapid solutions to the mass and energy balance equations associated with all kinds of tower processes. The procedure is highly numerical, but with digital computers it can be applied to many tower design problems. The method is generally applicable to all countercurrent flow

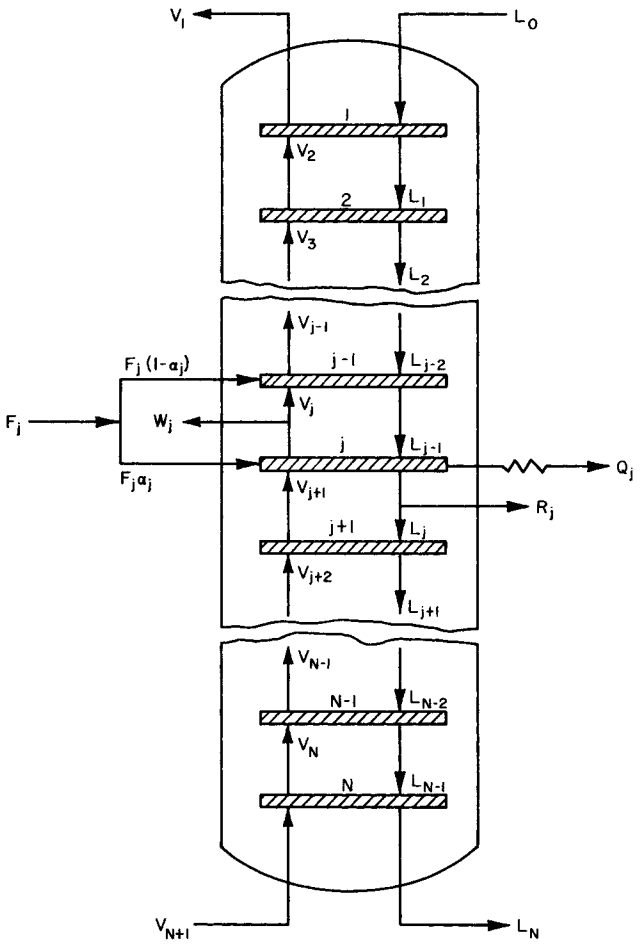

Fig. 1. Model for mathematical simulation.

equilibrium stage operations.

DESCRIPTION OF MODEL COLUMN

The model column for mathematical simulation is shown in Figure 1. The standard assumptions of constant column pressure and theoretical equilibrium trays connected by internal vapor and liquid flows are made. Because of the general form, the model represents all types of stage-by-stage separation processes. Feed streams can be introduced on each tray of the model, with the vapor portion being added to the tray above the feed tray; vapor and liquid side streams can be removed from each tray. Heat can be added to or removed from each tray to simulate reboilers, side heaters, interchillers, and condensers. For simplicity, only the feed, liquid and vapor side streams, and external heat flow for a typical tray, tray j, are shown in Figure 1.

EQUATIONS TO BE SOLVED

The basic equations for column simulation are derived by making material and heat balances around the j^{th} tray of the model. Heat and mass balances plus equilibrium relationships comprise the five sets of equations to be solved:

Overall material balances

$$L_0 + V_{j+1} + \sum_{k=1}^{j} (F_k - W_k - R_k) + F_{j+1}(1 - \alpha_{j+1}) - V_1 - L_j = 0, \quad \text{for } 1 \leq j \leq N \qquad (1)$$

Vapor-liquid equilibrium

$$y_{i,j} - K_{i,j}\, x_{i,j} = 0, \quad \text{for } 1 \leq i \leq M,\ 1 \leq j \leq N \qquad (2)$$

Component material balances

$$V_{j+1}\, y_{i,j+1} + L_{j-1}\, x_{i,j-1} + F_j\, z^l_{i,j} + F_{j+1}\, z^v_{i,j+1} - (V_j + W_j)\, y_{i,j} - (L_j + R_j)\, x_{i,j} = 0, \quad \text{for } 1 \leq i \leq M,\ 1 \leq j \leq N \qquad (3)$$

Summation equations

$$S_j = \sum_{i=1}^{M} y_{i,j} - \sum_{i=1}^{M} x_{i,j} = 0, \quad \text{for } 1 \leq j \leq N \qquad (4)$$

Heat balances

$$E_j = V_{j+1} H_{j+1} + L_{j-1} h_{j-1} + F_j \alpha_j h^F_j + F_{j+1}(1 - \alpha_{j+1}) H^F_{j+1} - (V_j + W_j) H_j - (L_j + R_j) h_j - Q_j = 0, \quad \text{for } 1 \leq j \leq N \qquad (5)$$

There are $N(2M + 3)$ equations. The $N(2M + 3)$ unknowns are vapor and liquid internal flow rates, vapor and liquid mole fractions, and tray temperatures. Quantities which must be given to obtain a solution are: rates, compositions, and temperatures of all feed streams; number of theoretical trays; rates of all side streams; amount of heat added to or removed from each tray; equilibrium and enthalpy data; and initial estimates of vapor flow rates, tray temperatures, and tray compositions (if nonideal solutions are used). Different variables may be selected as unknowns, or additional equations and unknowns may be included in the general equations to be solved. Thus, the method is considerably more versatile than as presented here.

METHOD OF SOLUTION

Equation set (2) can be substituted into the set (3) to eliminate $y_{i,j}$ and reduce the component material balance equations to the tridiagonal matrix form

$$\begin{bmatrix} B_1 C_1 & & & & \\ A_2 B_2 C_2 & & & & \\ & \ddots & & & \\ & A_j B_j C_j & & & \\ & & \ddots & & \\ & & A_{N-1} B_{N-1} C_{N-1} & & \\ & & & A_N B_N & \end{bmatrix} \begin{bmatrix} x_{i,1} \\ x_{i,2} \\ \vdots \\ x_{i,j} \\ \vdots \\ x_{i,N-1} \\ x_{i,N} \end{bmatrix} = \begin{bmatrix} D_1 \\ D_2 \\ \vdots \\ D_j \\ \vdots \\ D_{N-1} \\ D_N \end{bmatrix} \qquad (6)$$

for $1 \leq i \leq M$

where

$$\begin{aligned} B_1 &= -(L_1 + R_1) - (V_1 + W_1) K_{i,1} \\ C_1 &= V_2 K_{i,2} \\ D_1 &= -L_0 x_0 - F_1 z^l_{i,1} - F_2 z^v_{i,2} \\ A_j &= L_{j-1}, \text{ for } 2 \leq j \leq N-1 \\ B_j &= -(L_j + R_j) - (V_j + W_j) K_{i,j}, \text{ for } 2 \leq j \leq N-1 \\ C_j &= V_{j+1} K_{i,j+1}, \text{ for } 2 \leq j \leq N-1 \\ D_j &= -F_j z^l_{i,j} - F_{j+1} z^v_{i,j+1}, \text{ for } 2 \leq j \leq N-1 \\ A_N &= L_{N-1} \\ B_N &= -(L_N + R_N) - (V_N + W_N) K_{i,N} \\ D_N &= -V_{N+1} y_{i,N+1} - F_N z^l_{i,N} \end{aligned}$$

These equations are a linear set only when the equilibrium ratios, the $K_{i,j}$, are not functions of compositional changes which occur as the solution progresses. Equilibrium ratios can reflect the variation of composition with position in the column if estimates of this variation can be made prior to computation. In this case, the $K_{i,j}$'s at any position in the tower, while only functions of temperature, are different functions of temperature than those at some other position.

Equation sets (1), (2), (4), (5), and (6) are solved by using an iterative procedure which makes use of

Broyden's method (8) for obtaining the solution of simultaneous, nonlinear, algebraic equations. The procedure may be summarized as follows.

1. Some initial set of V_j and T_j is assumed.
2. The L_j are computed from Equation set (1).
3. Equation set (6) is solved by using the tridiagonal matrix algorithm (9) to give the $x_{i,j}$.
4. Equation set (2) is solved for the $y_{i,j}$.
5. These results are substituted into Equation sets (4) and (5) which are then solved simultaneously by using the procedure of Broyden for a new set of V_j and T_j.
6. By using this new set of V_j and T_j, steps 2 through 5 are repeated until an appropriate convergence criterion is satisfied.

Although it is not obvious, all equations are solved simultaneously by numerically substituting for L_j, $x_{i,j}$, and $y_{i,j}$ in Equation sets (4) and (5). These equation sets then become functions of the V_j and T_j only and can be solved simultaneously for these quantities.

The Broyden method is actually an improved Newton-Raphson iteration (10). Expanding S_J and E_J (the E_J are normalized by dividing the total heat inputs to the trays so that they are of the order of magnitude of 1) as functions of V_j and T_j using Taylor's approximation (11), and truncating after the first derivative term, we get

$$S_J^{\beta+1} = S_J^{\beta} + \sum_{j=1}^{N} \left(\frac{\partial S_J^{\beta}}{\partial V_j}\right) \Delta V_j + \sum_{j=1}^{N} \left(\frac{\partial S_J^{\beta}}{\partial T_j}\right) \Delta T_j = 0, \quad \text{for } 1 \leq J \leq N \tag{7}$$

$$E_J^{\beta+1} = E_J^{\beta} + \sum_{j=1}^{N} \left(\frac{\partial E_J^{\beta}}{\partial V_j}\right) \Delta V_j + \sum_{j=1}^{N} \left(\frac{\partial E_J^{\beta}}{\partial T_j}\right) \Delta T_j = 0, \quad \text{for } 1 \leq J \leq N \tag{8}$$

Since the S_J and E_J are errors or residuals in the summation and heat balance equations and are equal to zero when a solution is reached, these residuals are set equal to zero at the new iteration level. Equation sets (7) and (8) can also be written in matrix form:

$$\begin{bmatrix} \frac{\partial S_1^{\beta}}{\partial V_1} & \cdots & \frac{\partial S_1^{\beta}}{\partial V_N} & \frac{\partial S_1^{\beta}}{\partial T_1} & \cdots & \frac{\partial S_1^{\beta}}{\partial T_N} \\ \vdots & & \vdots & \vdots & & \vdots \\ \frac{\partial S_N^{\beta}}{\partial V_1} & \cdots & \frac{\partial S_N^{\beta}}{\partial V_N} & \frac{\partial S_N^{\beta}}{\partial T_1} & \cdots & \frac{\partial S_N^{\beta}}{\partial T_N} \\ \frac{\partial E_1^{\beta}}{\partial V_1} & \cdots & \frac{\partial E_1^{\beta}}{\partial V_N} & \frac{\partial E_1^{\beta}}{\partial T_1} & \cdots & \frac{\partial E_1^{\beta}}{\partial T_N} \\ \vdots & & \vdots & \vdots & & \vdots \\ \frac{\partial E_N^{\beta}}{\partial V_1} & \cdots & \frac{\partial E_N^{\beta}}{\partial V_N} & \frac{\partial E_N^{\beta}}{\partial T_1} & \cdots & \frac{\partial E_N^{\beta}}{\partial T_N} \end{bmatrix} \begin{bmatrix} \Delta V_1 \\ \vdots \\ \Delta V_N \\ \Delta T_1 \\ \vdots \\ \Delta T_N \end{bmatrix} = \begin{bmatrix} -S_1^{\beta} \\ \vdots \\ -S_N^{\beta} \\ -E_1^{\beta} \\ \vdots \\ -E_N^{\beta} \end{bmatrix} \tag{9}$$

If the S_J^{β} and E_J^{β} are calculated by using Equation sets (4) and (5), and if the partial derivatives are evaluated by finite difference methods, Equation (9) can be solved by matrix inversion for the ΔV_j and ΔT_j. Then, new V_j and T_j can be computed from the following equations:

$$V_j^{\beta+1} = V_j^{\beta} + t\,\Delta V_j, \quad \text{for } 1 \leq j \leq N \tag{10}$$

$$T_j^{\beta+1} = T_j^{\beta} + t\,\Delta T_j, \quad \text{for } 1 \leq j \leq N \tag{11}$$

Since

$$S_J = E_J = 0, \quad \text{for } 1 \leq J \leq N \tag{12}$$

Table 1. Reduction of Sum of Residuals with Iteration Number

$$\sum_{N}^{J=1} [(E_J)^2 + (S_J)^2]$$

Iteration No.	Absorber	Stabilizer	Demethanizer
1	0.223	0.483	0.133
2	0.353×10^{-1}	0.293	0.122
3	0.355×10^{-2}	0.129	0.109
4	0.268×10^{-2}	0.783×10^{-1}	0.927×10^{-1}
5	0.131×10^{-2}	0.297×10^{-1}	0.588×10^{-1}
6	0.333×10^{-3}	0.261×10^{-1}	0.210×10^{-1}
7	0.147×10^{-4}	0.331×10^{-2}	0.378×10^{-2}
8	0.413×10^{-5}	0.463×10^{-3}	0.108×10^{-2}
9	0.511×10^{-6}*	0.106×10^{-3}	0.129×10^{-3}
10		0.288×10^{-4}	0.284×10^{-4}
11		0.348×10^{-5}*	0.176×10^{-5}*

* Converged to prescribed tolerance.

is fulfilled when a solution is reached, the Broyden procedure (8) as used here systematically searches between -1 and $+1$ for a value of t which will meet the following condition:

$$\sum_{J=1}^{N} [(S_J^{\beta+1})^2 + (E_J^{\beta+1})^2] < \sum_{J=1}^{N} [(S_J^{\beta})^2 + (E_J^{\beta})^2] \tag{13}$$

Although this value may not be optimum from the standpoint of maximizing the reduction of the residuals, it gives assurance that the latest values of V_j and T_j result in movement toward the solution.

At this point, if

$$\sum_{J=1}^{N} [S_J^{\beta+1})^2 + (E_J^{\beta+1})^2] < \text{convergence tolerance} \tag{14}$$

the problem is considered solved. Where desired, each S_J and E_J may also be checked for convergence.

If the above test is not met, the entire procedure is repeated with the new set of V_j and T_j. However, for succeeding iterations, Broyden's procedure (8) for updating the inverse of the approximate Jacobian matrix [the matrix of partial derivatives in Equation (9)] using residuals calculated at the preceding iteration level is employed. Thus, there is no need for more than one matrix inversion per problem solution, and computation time is greatly reduced; in fact, the computation effort is not much greater than that required when the approximate Jacobian matrix is not improved.

To obtain a simultaneous solution of all the equations, new values of L_j, $x_{i,j}$, and $y_{i,j}$ must be calculated each time any of the V_j or T_j vary. This is necessary both in evaluation of partial derivatives and during the iterative solution.

ADVANTAGES OF THIS METHOD

Several types of iterative methods are available for solving column process equations. One (1, 2) uses tray-to-tray procedures, which are often numerically unstable, mainly due to buildup of truncation error (3). Another (4, 5) solves the component material balance equations for composition and then, in sequence, solves the summation equations for tray temperatures and the heat balance

equations for vapor rates. These are referred to as *bubble-point methods* and are acceptable for distillation processes, but not absorption (3). A third type (6, 7) also solves the component material balance equations for composition but sequentially solves the summation equations for vapor rates and the heat balance equations for temperature. These are called *sum rate methods* and have usually worked for absorption problems, but not distillation (3).

The present method overcomes the limitations of both the sum rate and bubble-point techniques by solving all equations simultaneously. Successful calculations have been made for absorption and distillation towers as well as complex demethanizers. Some aspects of this technique have been suggested by Friday and Smith (3) and Tierney and Bruno (12), but applications of the total concept have not been reported. Both of the suggested approaches would make use of the Newton-Raphson iteration (10) to solve the summation and heat balance equations. The recommended Broyden procedure (8), however, has two advantages over classical Newton-Raphson iteration. First, numerical stability is improved because the Broyden technique guarantees that each iterative step moves toward the solution. Second, since Broyden's procedure uses only one matrix inversion per problem, it significantly reduces the amount of computation required.

EXAMPLE CALCULATIONS

Example calculations for an absorber, stabilizer, and complex demethanizer were made. The absorber is a column of five theoretical trays with a liquid feed introduced at the top tray and a vapor stream fed to the bottom tray. The solution required 3 sec. on an IBM 360/65 computer for the nine iterations needed to converge. The stabilizer is a ten tray tower with liquid feed to the top tray and heat added to the bottom tray to simulate a reboiler. The stabilizer computation took 7 sec. and eleven iterations. The complex rich oil demethanizer column has eighteen theoretical trays. A liquid is fed to the top tray, and two-phase feed streams are introduced on trays seven and eight. There is a side heater at tray eleven, and the bottom tray serves as a reboiler. The eleven iterations used 20 sec. of computer time.

The time and number of iterations given for each solution depend upon the accuracy of the initial estimates of tray temperatures and vapor flow rates. The sum of the residuals of the heat balance and summation equations at each iteration level are shown in Table 1 and are a better indication of the method's efficiency.

CONCLUSIONS

A general method, based upon matrix operations, has been developed for giving stable, rapid solutions to column material and energy balance equations. The calculation solves all equations simultaneously with Broyden iteration methods.

This technique has the advantage of yielding solutions to all different types of tower processes, including both absorption and distillation operations. Furthermore, the iteration methods, which require only one matrix inversion for each column simulation, give numerically stable solutions with a minimum of computing time.

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NOTATION

A_j = coefficient in tridiagonal matrix equation
B_j = coefficient in tridiagonal matrix equation
C_j = coefficient in tridiagonal matrix equation
D_j = coefficient in tridiagonal matrix equation
$E_J{}^\beta$ = error in tray heat balance equation for iteration
F_j = flow rate of feed stream to tray
h_j = enthalpy of liquid stream leaving tray
$h_j{}^F$ = enthalpy of liquid portion of feed stream to tray
H_j = enthalpy of vapor stream leaving tray
$H_j{}^F$ = enthalpy of vapor portion of feed stream to tray
$K_{i,j}$ = equilibrium K value of component on tray (defined as $y_{i,j}/x_{i,j}$)
L_j = flow rate of liquid stream leaving tray
M = number of components
N = number of theoretical trays
Q_j = heat removed from tray
R_j = flow rate of liquid side stream leaving tray
$S_J{}^\beta$ = error in tray summation equation for iteration
t = weighting factor in Broyden's iterative procedure
T_j = temperature of tray
V_j = flow rate of vapor stream leaving tray
W_j = flow rate of vapor side stream leaving tray
$x_{i,j}$ = liquid mole fraction of component on tray
$y_{i,j}$ = vapor mole fraction of component on tray
$z^l{}_{i,j}$ = mole fraction of component in liquid portion of feed stream to tray
$z^v{}_{i,j}$ = mole fraction of component in vapor portion of feed stream to tray
α_j = liquid mole fraction of feed stream to tray

Subscripts

i = component number
j,J = tray number
k = dummy subscript
N = bottom tray in column

Superscripts

β = iteration number
F = feed stream
l = liquid portion of stream
v = vapor portion of stream

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