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ADAPTATION OF BOYNTON'S MATHEMATICAL
MODEL TO HYDROGEN ISOTOPE SEPARATION
COLUMN BY CRYOGENIC DISTILLATION

August 1981

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Adaptation of Boynton's Mathematical Model to Hydrogen
Isotope Separation Column by Cryogenic Distillation

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(Received August 14, 1981)

Boynton's mathematical simulation procedure for multi-component distillation calculations has the advantage that the Jacobian matrix is calculated analytically. The purpose of the present study is to adapt this procedure to hydrogen isotope separation columns by cryogenic distillation.

The Boynton's model is modified so that the model can incorporate decay heat of tritium, nonideality of the hydrogen isotope solutions, multiple feeds and multiple sidestreams. Basic equations are derived and the mathematical simulation procedure is briefly explained.

KEYWORDS : Multicomponent Distillation, Isotope Separation,
Mathematical Model, Tritium, Decay Heat,
Nonideality, Cryogenic Distillation, Jacobian Matrix,
Hydrogen Isotope

Boyntonの数学的モデルの深冷蒸留法による
水素同位体分離塔への拡張

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(1981年8月14日受理)

Boyntonが多成分蒸留計算用に開発したモデルは、ヤコビアン（多次の正方行列）を解析的に求めることができるという利点を有している。本研究の目的は、このモデルを水素同位体分離用深冷蒸留塔に拡張することである。

トリチウムの崩壊熱、溶液の非理想性、多段フィード及び多段サイドカットを考慮できるようにモデルを改良し、基本式の導出及び解析手順の整理を行った。

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1. Introduction

The author has been engaged in computer simulation work of hydrogen isotope separation columns by cryogenic distillation.⁽¹⁾⁽²⁾⁽³⁾ The model used incorporates such factors as decay heat of tritium, differences in latent heat of vaporization among the six isotopic species of molecular hydrogen (H_2 , HD, HT, D_2 , DT and T_2), heat transfer through the column wall, pressure drop and nonideality of the hydrogen isotope solutions. Provision is made for simulation of a column with multiple feeds and multiple sidestreams. The simulation procedure provides fast convergence in a wide range of input and output specifications. However, it has the only shortcoming that the required computational time is long. One of the main reasons for long computational time is that the Jacobian matrix ($2N$ -dimensional square matrix, where N denotes number of total theoretical stages) is calculated not analytically but numerically.

Boynton proposed an interesting procedure⁽⁴⁾ where the Jacobian matrix can be analytically calculated. However, it cannot be applied to rigorous simulation study because it accounts for neither decay heat of tritium nor nonideality. Also, it cannot treat a column with multiple feeds and multiple sidestreams.

The purpose of the present report is adaptation of Boynton's model to simulation of hydrogen isotope separation columns, incorporating all the factors described previously. The basic equations are derived and the simulation procedure is explained.

There is a possibility that this new simulation procedure provides shorter computational time. Detailed comparison between the two procedures (the one used in the author's previous studies and the new one described in the present report) is desired in further studies.

2. Derivation of Basic Equations

2.1 Material Balances and Energy Balances

The component material balances around j -th stage are expressed by (Fig. 1)

$$f_{i,j} + l_{i,j-1} + v_{i,j+1} = l_{i,j} + v_{i,j} + u_{i,j} + w_{i,j} . \quad (1)$$

The equilibrium relationships are

$$K_{i,j} = (v_{i,j}/V_j)/(l_{i,j}/L_j) . \quad (2)$$

In Eq.(1),

$$u_{i,j} = U_j l_{i,j}/L_j , \quad w_{i,j} = W_j v_{i,j}/V_j . \quad (3)$$

Substituting Eq.(3) and Eq.(2) into Eq.(1) yields

$$l_{i,j-1} - (1 + U_j/L_j)l_{i,j} - (1 + W_j/V_j)S_{i,j}l_{i,j} + S_{i,j+1}l_{i,j+1} = - f_{i,j} , \quad (4)$$

where

$$S_{i,j} = V_j K_{i,j}/L_j . \quad (5)$$

Terms containing " $j-1$ " and " $j+1$ " subscripts in Eq.(4) are zero when $j=1$ and N , respectively (Fig. 2 and Fig. 3).

Taking the total material balance around j -th stage and the bottom of the column, we obtain

$$V_j + L_N + \sum_{k=j}^N W_k + \sum_{k=j}^N U_k = \sum_{k=j}^N F_k + L_{j-1} . \quad (6)$$

Taking the energy balance around j-th stage and the bottom, we obtain

$$\begin{aligned}
 E_j = & \sum_{k=j}^N H_{Fj} + \sum_{k=j}^N q_k - \sum_{k=j}^N Q_k - \sum_{i=1}^m l_{i,N} h_{i,N}^{\circ} + \sum_{i=1}^m l_{i,j-1} h_{i,j-1}^{\circ} \\
 & - \sum_{i=1}^m v_{i,j} H_{i,j}^{\circ} - \sum_{k=j}^N \sum_{i=1}^m w_{i,k} H_{i,k}^{\circ} - \sum_{k=j}^N \sum_{i=1}^m u_{i,k} h_{i,k}^{\circ} \\
 & + \sum_{k=j}^N \sum_{i=1}^m R_k l_{i,k} \lambda_i / L_k .
 \end{aligned} \tag{7}$$

The component material balances around j-th stage and the bottom are expressed by

$$v_{i,j} + l_{i,N} + \sum_{k=j}^N w_{i,k} + \sum_{k=j}^N u_{i,k} = \sum_{k=j}^N f_{i,k} + l_{i,j-1} . \tag{8}$$

Eliminating $v_{i,j}$ from Eq.(7) by use of Eq.(8) yields

$$\begin{aligned}
 E_j = & C_j + \sum_{i=1}^m l_{i,N} (H_{i,j}^{\circ} - h_{i,N}^{\circ}) + \sum_{i=1}^m l_{i,j-1} (h_{i,j-1}^{\circ} - H_{i,j}^{\circ}) \\
 & + \sum_{k=j}^N \sum_{i=1}^m U_k l_{i,k} (H_{i,k}^{\circ} - h_{i,k}^{\circ}) / L_k - \sum_{k=j}^N \sum_{i=1}^m f_{i,k} H_{i,j}^{\circ} \\
 & + \sum_{k=j}^N \sum_{i=1}^m R_k l_{i,k} \lambda_i / L_k ,
 \end{aligned} \tag{9}$$

where

$$C_j = \sum_{k=j}^N H_{Fk} + \sum_{k=j}^N q_k - \sum_{k=j}^N Q_k . \tag{10}$$

It should be noted that both the condenser and the reboiler have neither the feed nor the sidestream, and the condenser is the partial one:

$$F_1 = F_N = W_1 = W_N = U_1 = U_N = 0 \quad . \quad (11)$$

2.2 Temperature Correction

The Boynton's mathematical procedure has two iteration loops. One of them is for finding out component flows and temperature distribution such that $\bar{L}_j = L_j$, where

$$\bar{L}_j = \sum_{i=1}^m l_{i,j} \quad . \quad (12)$$

The procedure is summarized below:

Step(1) Estimate phase flows L_j and temperature T_j .

Step(2) Correct the temperature.

Step(3) Correct the phase flows.

Repeat Step(2) and Step(3) until the energy balance equations are satisfied within convergence tolerance.

The purpose of the first loop is to solve the following N-dimensional nonlinear simultaneous equations:

$$\left. \begin{aligned} L_1 - \bar{L}_1 &= f_1(T_1, \dots, T_N) = 0 \\ &\vdots \\ L_N - \bar{L}_N &= f_N(T_1, \dots, T_N) = 0 \end{aligned} \right\} \quad . \quad (13)$$

The Jacobian matrix is expressed by

$$\begin{bmatrix}
 -(\partial \bar{L}_1 / \partial T_1)|_L \dots -(\partial \bar{L}_1 / \partial T_N)|_L \\
 \vdots \\
 -(\partial \bar{L}_N / \partial T_1)|_L \dots -(\partial \bar{L}_N / \partial T_N)|_L
 \end{bmatrix} \quad (14)$$

This is a N-dimensional square matrix and can be calculated analytically.

Equation (4) can be rewritten by the following tridiagonal matrix equations :

$$\begin{bmatrix}
 B_{i,1} & C_{i,1} & 0 & \dots & 0 \\
 A_{i,2} & B_{i,2} & C_{i,2} & 0 & \dots & 0 \\
 \dots & \dots & \dots & \dots & \dots & \dots \\
 0 & \dots & 0 & A_{i,N-1} & B_{i,N-1} & C_{i,N-1} \\
 0 & \dots & 0 & A_{i,N} & B_{i,N} & 0
 \end{bmatrix}
 \begin{bmatrix}
 l_{i,1} \\
 \vdots \\
 l_{i,N}
 \end{bmatrix}
 =
 \begin{bmatrix}
 D_{i,1} \\
 \vdots \\
 D_{i,N}
 \end{bmatrix} , \quad (15)$$

where

$$\left. \begin{aligned}
 A_{i,j} &= 1.0, \quad B_{i,j} = -(1 + U_j/L_j) - (1 + W_j/V_j)S_{i,j}, \\
 C_{i,j} &= S_{i,j+1}, \quad D_{i,j} = -f_{i,j}, \quad (j=2, \dots, N-1), \\
 B_{i,1} &= -(1 + S_{i,1}), \quad B_{i,N} = -(1 + S_{i,N}), \\
 C_{i,1} &= S_{i,2}, \quad A_{i,N} = 1.0, \quad D_{i,1} = D_{i,N} = 0.0
 \end{aligned} \right\} \quad (16)$$

\bar{L}_j can be calculated from Eq.(15), Eq.(16) and Eq.(12).

Differentiating Eq.(4) with respect to temperature at constant phase flow yields

$$\begin{aligned}
 & (\partial l_{i,j-1} / \partial T_k) \Big|_L - (1 + U_j / L_j) (\partial l_{i,j} / \partial T_k) \Big|_L \\
 & - (1 + W_j / V_j) S_{i,j} (\partial l_{i,j} / \partial T_k) \Big|_L + S_{i,j+1} (\partial l_{i,j+1} / \partial T_k) \Big|_L \\
 & = (1 + W_j / V_j) l_{i,j} (V_j / L_j) (dK_{i,j} / dT_j) \delta_{j,k} \\
 & - l_{i,j+1} (V_{j+1} / L_{j+1}) (dK_{i,j+1} / dT_{j+1}) \delta_{j+1,k} .
 \end{aligned} \tag{17}$$

By arrangement of Eq.(17), we obtain the similar tridiagonal matrix equations for calculation of $(\partial l_{i,j} / \partial T_k) \Big|_L$.

The elements of the Jacobian matrix are calculated from

$$(\partial \bar{L}_j / \partial T_k) \Big|_L = \sum_{i=1}^m (\partial l_{i,j} / \partial T_k) \Big|_L . \tag{18}$$

Hence, Eq.(13) can be solved by application of multidimensional Newton-Raphson Method.

The independent variables in the first loop are T_j and those in the second loop are L_j . Vapor flows are determined from Eq.(6).

It should be noted that the vapor-liquid equilibrium ratio, $K_{i,j}$, must be independent of concentration. Incorporation of non-ideality of hydrogen isotope solutions requires an additional iteration loop which will be discussed later.

2.3 Phase Flow Correction

The component flows and temperature calculated in the first loop do satisfy the material balance equations and equilibrium relationships, but they do not satisfy the energy balances. The second loop is for solving the following nonlinear simultaneous equations :

$$\left. \begin{aligned} E_1(L_1, \dots, L_N) &= 0 \\ &\vdots \\ E_N(L_1, \dots, L_N) &= 0 \end{aligned} \right\}, \quad (19)$$

and the phase flows are corrected by the Newton-Raphson iterative calculation at constant temperature.

The Jacobian matrix can be calculated analitically. The partial derivative $\partial E_j / \partial L_k$ is expressed by

$$\begin{aligned} \partial E_j / \partial L_k &= \sum_{i=1}^m (\partial l_{i,N} / \partial L_k) (H_{i,j}^\circ - h_{i,N}^\circ) + \sum_{i=1}^m l_{i,N} \{ (dH_{i,j}^\circ / dT_j) (\partial T_j / \partial L_k) \\ &\quad - (dh_{i,N}^\circ / dT_N) (\partial T_N / \partial L_k) \} + \sum_{i=1}^m (\partial l_{i,j-1} / \partial L_k) (h_{i,j-1}^\circ - H_{i,j}^\circ) \\ &\quad + \sum_{i=1}^m l_{i,j-1} \{ (dh_{i,j-1}^\circ / dT_{j-1}) (\partial T_{j-1} / \partial L_k) - (dH_{i,j}^\circ / dT_j) (\partial T_j / \partial L_k) \} \\ &\quad - \sum_{n=j}^N \sum_{i=1}^m f_{i,n} (dH_{i,j}^\circ / dT_j) (\partial T_j / \partial L_k) \\ &\quad + \sum_{n=j}^N \sum_{i=1}^m [(-U_n / L_n^2) l_{i,n} (H_{i,n}^\circ - h_{i,n}^\circ) (\partial L_n / \partial L_k) + \\ &\quad (U_n / L_n) (H_{i,n}^\circ - h_{i,n}^\circ) (\partial l_{i,n} / \partial L_k) + \end{aligned}$$

$$\begin{aligned}
 & (U_{n-1,n}/L_n) \{ (dh_{i,n}^\circ/dT_n)(\partial T_n/\partial L_k) - (dh_{i,n}^\circ/dT_n)(\partial T_n/\partial L_k) \} \\
 & + \sum_{n=j}^N \sum_{i=1}^m \{ R_n (-l_{i,n}/L_n^2) (\partial L_n/\partial L_k) \lambda_i + R_n (\lambda_i/L_n) (\partial l_{i,n}/\partial L_k) \}
 \end{aligned} \tag{20}$$

\bar{L}_j is a function of all the phase flows and temperature, hence

$$\partial \bar{L}_j / \partial L_k = \sum_{n=1}^N (\partial \bar{L}_j / \partial T_n) \Big|_L (\partial T_n / \partial L_k) + \sum_{n=1}^N (\partial \bar{L}_j / \partial L_n) \Big|_T (\partial L_n / \partial L_k), \tag{21}$$

where

$$\partial \bar{L}_j / \partial L_k = \delta_{j,k}, \quad \partial L_n / \partial L_k = \delta_{n,k}. \tag{22}$$

Substituting Eq.(22) into Eq.(21), we obtain the following matrix equation:

$$\begin{bmatrix}
 (\partial \bar{L}_1 / \partial T_1) \Big|_L & \dots & (\partial \bar{L}_1 / \partial T_N) \Big|_L \\
 \vdots & \ddots & \vdots \\
 (\partial \bar{L}_N / \partial T_1) \Big|_L & \dots & (\partial \bar{L}_N / \partial T_N) \Big|_L
 \end{bmatrix}
 \begin{bmatrix}
 (\partial T_1 / \partial L_k) \\
 \vdots \\
 (\partial T_N / \partial L_k)
 \end{bmatrix}
 =
 \begin{bmatrix}
 \delta_{1,k} - (\partial \bar{L}_1 / \partial L_k) \Big|_T \\
 \vdots \\
 \delta_{N,k} - (\partial \bar{L}_N / \partial L_k) \Big|_T
 \end{bmatrix}. \tag{23}$$

From Eq.(6),

$$\begin{aligned}
 \partial V_j / \partial L_n &= \partial L_{j-1} / \partial L_n - \partial L_N / \partial L_n \\
 &= \delta_{j-1,n} - \delta_{N,n}.
 \end{aligned} \tag{24}$$

Differentiating Eq.(4) with respect to L_k at constant temperature, we obtain

$$\begin{aligned}
 & (\partial l_{i,j-1}/\partial L_k)|_T - \{1 + U_j/L_j + (1 + W_j/V_j)S_{i,j}\}(\partial l_{i,j}/\partial L_k)|_T \\
 & + S_{i,j+1}(\partial l_{i,j+1}/\partial L_k)|_T \\
 & = l_{i,j} [(-U_j/L_j^2)\delta_{j,k} + S_{i,j}(-W_j/V_j^2)(\delta_{j-1,k} - \delta_{N,k}) \\
 & + (1 + W_j/V_j)\{(-V_j/L_j^2)K_{i,j}\delta_{j,k} + (K_{i,j}/L_j)(\delta_{j-1,k} - \delta_{N,k})\}] \\
 & - l_{i,j+1}\{(-V_{j+1}/L_{j+1}^2)K_{i,j+1}\delta_{j+1,k} + (K_{i,j+1}/L_{j+1})(\delta_{j,k} - \delta_{N,k})\} .
 \end{aligned} \tag{25}$$

Arrangement of Eq.(25) results in the tridiagonal matrix equations and the partial derivative, $(\partial \bar{L}_j/\partial L_k)|_T$ can be calculated from

$$(\partial \bar{L}_j/\partial L_k)|_T = \sum_{i=1}^m (\partial l_{i,j}/\partial L_k)|_T . \tag{26}$$

The elements of the N-dimensional square matrix in Eq.(23) are determined in the first loop, hence $\partial T_j/\partial L_k$ can be calculated from Eq.(23).

Finally,

$$\partial l_{i,j}/\partial L_k = \sum_{i=1}^m (\partial l_{i,j}/\partial T_n)|_L (\partial T_n/\partial L_k) + (\partial l_{i,j}/\partial L_k)|_T . \tag{27}$$

From Eq.(27), $\partial l_{i,j}/\partial L_k$ can be directly calculated.

Accordingly, all the elements of the Jacobian matrix can be calculated from Eq.(20). The phase flows are corrected by the Newton-Raphson iterative calculation.

2.4 Incorporation of Nonideality of Hydrogen Isotope Solutions

For ideal solutions, $K_{i,j}$ is expressed by

$$K_{i,j} = p_{i,j}^{\circ}/P_j \quad (28)$$

However, since the hydrogen isotope solutions do not quite obey the Raoult's Law, $K_{i,j}$ should be written by

$$K_{i,j} = \xi_{i,j} p_{i,j}^{\circ}/P_j \quad (29)$$

where $\xi_{i,j}$ denotes the deviation coefficient.

According to Souers' procedure, $\xi_{i,j}$ can be estimated from the following equation :

$$\xi_{i,j} = \text{EXP}(\Delta G_{i,j}^{\bar{E}}/R_g/T_j) \text{EXP}\{B_{i,j}^*(p_{i,j}^{\circ} - P_j)/R_g/T_j\} \quad (30)$$

One of possible methods for incorporation of nonideality is application of the following successive iteration method :

Step(1) Postulate all the deviation coefficients to equal unity.

Step(2) Find out the solutions by the adapted Boynton's procedure described previously.

Step(3) Recalculate the coefficients.

Repeat Step(2) and Step(3) until the coefficients remain unchanged within convergence tolerance.

2.5 Initial Estimates of Phase Flows and Temperature

Since the procedure uses the Newton-Raphson iterative calculations, initial estimates are expected to be vitally important. If they are far from the solutions, the calculation will fail to converge. The recommended procedures for initial estimation of phase flows and temperature distribution are described in detail in the author's previous report.⁽¹⁾

3. Conclusions

The Boynton's mathematical simulation procedure is adapted to hydrogen isotope separation columns by cryogenic distillation. The new model incorporates decay heat of tritium, nonideality of the hydrogen isotope solutions, multiple feeds and multiple sidestreams.

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NOMENCLATURE

- $B_{i,j}^*$ = Second virial coefficient in terms of density (cal/g-mol/atm)
 F_j = Flow rate of feed stream supplied to j-th stage (g-mol/h)
 $f_{i,j}$ = Flow rate of i-th component in feed stream supplied to
 j-th stage (g-mol/h)
 $\overline{\Delta G_{i,j}^E}$ = Partial molal-excess Gibbs free energy of mixing (cal/g-mol)
 $H_{i,j}^\circ$ = Molal enthalpy of i-th component in vapor state at T_j
 (cal/g-mol)
 H_{Fj} = Total enthalpy of feed stream supplied to j-th stage (cal/h)
 $h_{i,j}^\circ$ = Molal enthalpy of i-th component in liquid state at T_j
 (cal/g-mol)
 $K_{i,j}$ = Vapor-liquid equilibrium ratio of i-th component at T_j
 L_j = Flow rate of liquid stream leaving j-th stage (g-mol/h)
 $l_{i,j}$ = Flow rate of i-th component in liquid stream leaving j-th
 stage (g-mol/h)
 m = Total number of components
 N = Number of total theoretical stages
 P_j = Total pressure on j-th stage (atm)
 $p_{i,j}^\circ$ = Vapor pressure of i-th component at T_j (atm)
 Q_j = Heat subtracted from j-th stage (cal/h)
 q_j = Heat added to j-th stage (cal/h)
 R_g = Gas constant (cal/g-mol/K)
 R_j = Molal holdup of liquid in j-th stage (g-mol)
 T_j = Absolute temperature on j-th stage (K)

U_j = Flow rate of liquid sidestream from j-th stage (g-mol/h)

$u_{i,j}$ = Flow rate of i-th component in liquid sidestream from j-th stage (g-mol/h)

V_j = Flow rate of vapor stream leaving j-th stage (g-mol/h)

$v_{i,j}$ = Flow rate of i-th component in vapor stream leaving j-th stage (g-mol/h)

W_j = Flow rate of vapor sidestream from j-th stage (g-mol/h)

$w_{i,j}$ = Flow rate of i-th component in vapor sidestream from j-th stage (g-mol/h)

λ_i = Coefficient for accounting for decay heat of tritium (cal/g-mol/h), $\lambda_1 = \lambda_2 = \lambda_4 = 0$; $\lambda_3 = \lambda_5 = a\Lambda$;

$$\lambda_6 = 2a\Lambda,$$

where

a = Atomic weight of tritium (g),

Λ = Decay heat of tritium (cal/g/h).

(Subscript)

j : Stage number

i : Component number, 1 : H_2 , 2 : HD, 3 : HT, 4 : D_2 , 5 : DT,

6 : T_2

ACKNOWLEDGEMENT

The author wishes to express his sincere thanks and appreciation to Dr. Robert H. Sherman and Dr. John R. Bartlit at Los Alamos National Laboratory for giving him much useful information and valuable comments. Acknowledgement is also due to Dr. Yukio Obata for his continuous encouragement.

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ACKNOWLEDGEMENT

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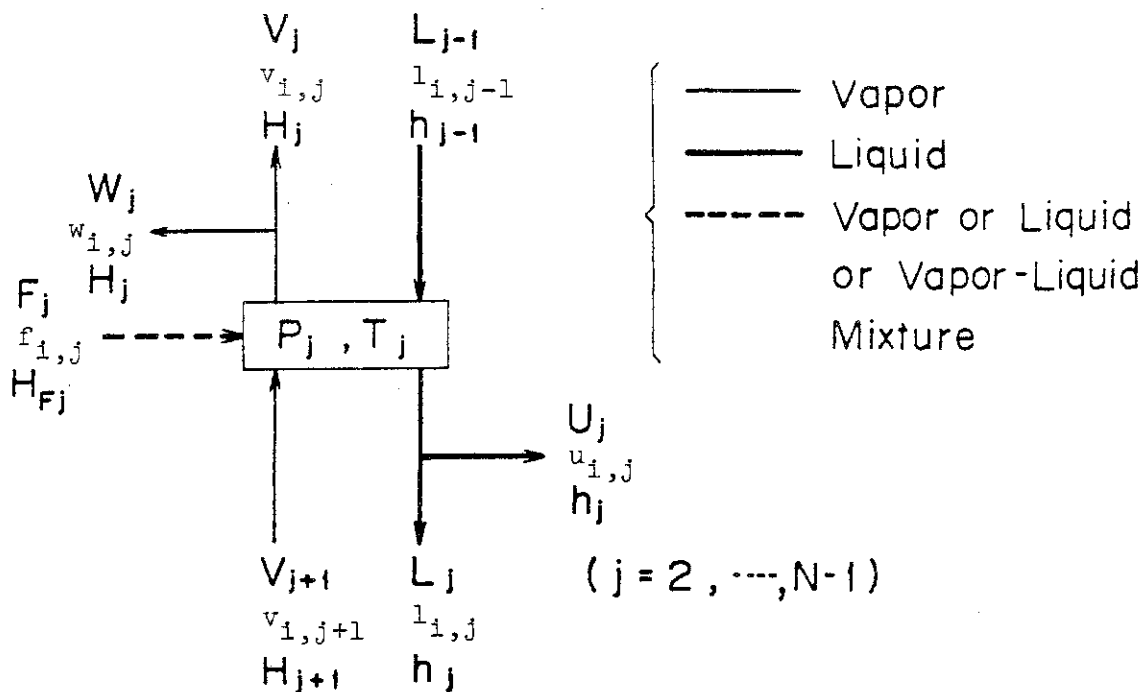


Fig.1 Model Stage for Mathematical Simulation

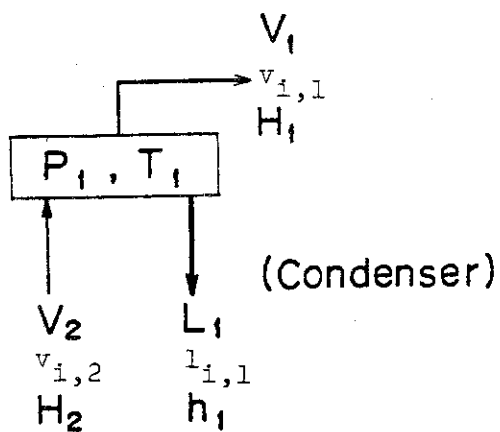


Fig.2 Model Condenser for Mathematical Simulation

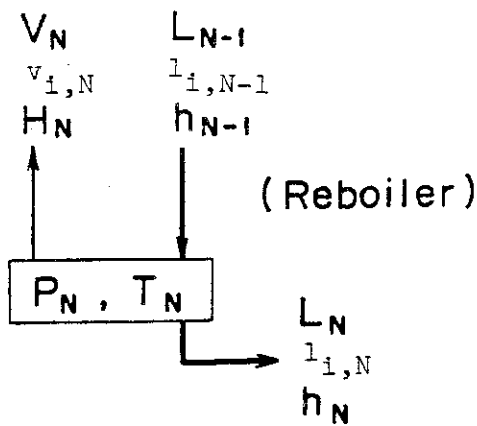


Fig.3 Model Reboiler for Mathematical Simulation