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DERIVATION OF BASIC EQUATIONS FOR  
RIGOROUS DYNAMIC SIMULATION OF  
CRYOGENIC DISTILLATION COLUMN FOR  
HYDROGEN ISOTOPE SEPARATION

August 1981

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Derivation of Basic Equations for Rigorous Dynamic  
Simulation of Cryogenic Distillation Column for  
Hydrogen Isotope Separation

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( Received August 14, 1981 )

The basic equations are derived for rigorous dynamic simulation of cryogenic distillation columns for hydrogen isotope separation.

The model accounts for such factors as differences in latent heat of vaporization among the six isotopic species of molecular hydrogen, decay heat of tritium, heat transfer through the column wall and nonideality of the solutions. Provision is also made for simulation of columns with multiple feeds and multiple sidestreams.

KEYWORDS : Dynamic Simulation, Isotope Separation, Hydrogen Isotope,  
Cryogenic Distillation, Multicomponent Distillation,  
Latent Heat of Vaporization, Decay Heat, Tritium,  
Heat Transfer, Nonideality, Basic Equations

水素同位体分離用深冷蒸留塔の厳密な動特性解析  
のための基本式の導出

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水素同位体分離用深冷蒸留塔の厳密な動特性解析を行うのに先立ち、解析に必要なモデル式の導出を行った。

モデルは、各成分間の蒸発潜熱の相違、トリチウムの崩壊熱、塔壁を通しての熱移動、圧力損失、溶液の非理想性、多段フィード及び多段サイドカットなどのあらゆるファクターを考慮しており、液ホールドアップをも可変として取り扱うものである。

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## 1. Introduction

It is very important to grasp dynamic behavior of a cryogenic distillation column for assurance of its smooth operation. The author has been studying dynamics and control of cryogenic distillation columns, but the model used neglected such factors as differences in latent heat of vaporization among the six isotopic species of molecular hydrogen (  $H_2$ , HD, HT,  $D_2$ , DT and  $T_2$  ), decay heat of tritium, heat transfer through the column wall and nonideality of the hydrogen isotope solutions. If these factors have only minor effects on dynamic behavior of columns, they should not be incorporated in the dynamic simulation model to avoid unnecessarily complicated procedure and long computational time.

However, there is no guarantee that these factors' effects are minor. For this reason, the author is now moving on development of rigorous dynamic simulation model incorporating all the factors. This can be done by adaptation of mathematical model developed by Howard<sup>(2)</sup> to hydrogen isotope separation column by cryogenic distillation. The author's model has the special feature that it accounts for multiple feeds and multiple sidestreams and nonideality of the solutions.

The present report describes the basic equations for rigorous dynamic simulation of cryogenic distillation columns.

## 2. Derivation of Basic Equations for Rigorous Dynamic Simulation

### 2.1 Material and Energy Balances around j-th Stage ( j=2,...,N-1 )

The component material balances are expressed by the following ordinary differential equations where the gas holdups are neglected ( Fig. 1 ) :

$$\begin{aligned} d(R_j x_{i,j})/dt = & V_{j+1} y_{i,j+1} + L_{j-1} x_{i,j-1} - (V_j + W_j) y_{i,j} \\ & - (L_j + U_j) x_{i,j} + F_j z_{Fi,j} . \end{aligned} \quad (1)$$

The total material balance is expressed by

$$dR_j/dt = V_{j+1} + L_{j-1} - (V_j + W_j) - (L_j + U_j) + F_j . \quad (2)$$

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

$$\begin{aligned} dx_{i,j}/dt = & \{ V_{j+1}(y_{i,j+1} - x_{i,j}) + L_{j-1}(x_{i,j-1} - x_{i,j}) \\ & - (V_j + W_j)(y_{i,j} - x_{i,j}) + F_j(z_{Fi,j} - x_{i,j}) \} / R_j . \end{aligned} \quad (3)$$

The energy balance is expressed by

$$\begin{aligned} d(R_j h_j)/dt = & V_{j+1} H_{j+1} + L_{j-1} h_{j-1} - (V_j + W_j) H_j - (L_j + U_j) h_j \\ & - Q_j + q_j + H_{Fj} + \sum_{i=1}^m R_j x_{i,j} \lambda_i . \end{aligned} \quad (4)$$

If the heat of mixing is negligible, the enthalpy  $h_j$  is given by

$$h_j = \sum_{i=1}^m x_{i,j} h_{i,j}^{\circ} . \quad (5)$$

Differentiation of Eq.(5) yields

$$\begin{aligned}
 dh_j/dt &= \sum_{i=1}^m x_{i,j} (dh_{i,j}^{\circ}/dt) + \sum_{i=1}^m h_{i,j}^{\circ} (dx_{i,j}/dt) \\
 &= \sum_{i=1}^m x_{i,j} (dh_{i,j}^{\circ}/dT_j) \cdot \sum_{i=1}^m (\partial T_j / \partial x_{i,j}) (dx_{i,j}/dt) + \sum_{i=1}^m h_{i,j}^{\circ} (dx_{i,j}/dt).
 \end{aligned}
 \tag{6}$$

Substituting Eq.(2) into Eq.(4) yields

$$\begin{aligned}
 dh_j/dt &= \{ V_{j+1}(H_{j+1} - h_j) + L_{j-1}(h_{j-1} - h_j) - (V_j + W_j)(H_j - h_j) \\
 &\quad - Q_j + q_j + H_{Fj} - F_j h_j + \sum_{i=1}^m R_j x_{i,j} \lambda_i \} / R_j.
 \end{aligned}
 \tag{7}$$

From Eq.(3), Eq.(6) and Eq.(7), we obtain the following energy balance equation :

$$\begin{aligned}
 V_{j+1}(A_j + h_j - H_{j+1}) + L_{j-1}(B_j + h_j - h_{j-1}) - (V_j + W_j)(C_j + h_j - H_j) \\
 + F_j(E_j + h_j) = D_j - Q_j + q_j + H_{Fj},
 \end{aligned}
 \tag{8}$$

where

$$\begin{aligned}
 A_j = \sum_{i=1}^m h_{i,j}^{\circ} (y_{i,j+1} - x_{i,j}) + \sum_{i=1}^m x_{i,j} (dh_{i,j}^{\circ}/dT_j) \cdot \sum_{i=1}^m (\partial T_j / \partial x_{i,j}) (y_{i,j+1} \\
 - x_{i,j}),
 \end{aligned}
 \tag{9}$$

$$\begin{aligned}
 B_j = \sum_{i=1}^m h_{i,j}^{\circ} (x_{i,j-1} - x_{i,j}) + \sum_{i=1}^m x_{i,j} (dh_{i,j}^{\circ}/dT_j) \cdot \sum_{i=1}^m (\partial T_j / \partial x_{i,j}) (x_{i,j-1} \\
 - x_{i,j}),
 \end{aligned}
 \tag{10}$$

$$\begin{aligned}
 C_j = \sum_{i=1}^m h_{i,j}^{\circ} (y_{i,j} - x_{i,j}) + \sum_{i=1}^m x_{i,j} (dh_{i,j}^{\circ}/dT_j) \cdot \sum_{i=1}^m (\partial T_j / \partial x_{i,j}) (y_{i,j} \\
 - x_{i,j}),
 \end{aligned}
 \tag{11}$$



$$D_j = \sum_{i=1}^m R_j x_{i,j} \lambda_i \quad , \quad (12)$$

$$E_j = \sum_{i=1}^m h_{i,j}^o (z_{Fi,j} - x_{i,j}) + \sum_{i=1}^m x_{i,j} (dh_{i,j}^o / dT_j) \cdot \sum_{i=1}^m (\partial T_j / \partial x_{i,j}) (z_{Fi,j} - x_{i,j}) \quad . \quad (13)$$

In the present model, constant volume holdup is assumed because this is probably the constant holdup specification which is closest to the real behavior of a stage. The molal holdup does not remain constant along the time trajectory, and it is given by

$$R_j = V_j^* \sum_{i=1}^m x_{i,j} v_{i,j}(T_j) \quad . \quad (14)$$

Differentiation of Eq.(14) yields

$$dR_j/dt = \left\{ \sum_{i=1}^m x_{i,j} (dv_{i,j}/dT_j) \sum_{i=1}^m (\partial T_j / \partial x_{i,j}) (dx_{i,j}/dt) + \sum_{i=1}^m (dx_{i,j}/dt) v_{i,j} \right\} V_j^* \quad . \quad (15)$$

From Eq.(2), Eq.(3) and Eq.(15), we obtain the following total material balance equation :

$$V_{j+1}(G_j - 1) + L_{j-1}(I_j - 1) + (V_j + W_j)(1 - J_j) + F_j(K_j - 1) + (L_j + U_j) = 0 \quad , \quad (16)$$

where

$$G_j = \sum_{i=1}^m x_{i,j} (dv_{i,j}/dT_j) \cdot \sum_{i=1}^m (\partial T_j / \partial x_{i,j}) (y_{i,j+1} - x_{i,j}) V_j^* / R_j + \sum_{i=1}^m v_{i,j} (y_{i,j+1} - x_{i,j}) V_j^* / R_j \quad , \quad (17)$$

$$I_j = \sum_{i=1}^m x_{i,j} (dv_{i,j}/dT_j) \cdot \sum_{i=1}^m (\partial T_j / \partial x_{i,j}) (x_{i,j-1} - x_{i,j}) V_j^* / R_j \\ + \sum_{i=1}^m v_{i,j} (x_{i,j-1} - x_{i,j}) V_j^* / R_j, \quad (18)$$

$$J_j = \sum_{i=1}^m x_{i,j} (dv_{i,j}/dT_j) \cdot \sum_{i=1}^m (\partial T_j / \partial x_{i,j}) (y_{i,j} - x_{i,j}) V_j^* / R_j \\ + \sum_{i=1}^m v_{i,j} (y_{i,j} - x_{i,j}) V_j^* / R_j, \quad (19)$$

$$K_j = \sum_{i=1}^m x_{i,j} (dv_{i,j}/dT_j) \cdot \sum_{i=1}^m (\partial T_j / \partial x_{i,j}) (z_{Fi,j} - x_{i,j}) V_j^* / R_j \\ + \sum_{i=1}^m v_{i,j} (z_{Fi,j} - x_{i,j}) V_j^* / R_j. \quad (20)$$

## 2.2 Material and Energy Balances around Condenser and Reboiler

The material and energy balance equations can be derived in a similar manner. It is postulated that  $V_1$  (the flow rate of the top product) and  $Q_C$  (the condenser load) are specified. The condenser is the partial one (Fig. 2). The basic equations are

$$dx_{i,1}/dt = \{ V_2(y_{i,2} - x_{i,1}) + V_1(x_{i,1} - y_{i,1}) \} / R_1, \quad (21)$$

$$V_2(A_1 + h_1 - H_2) - V_1(C_1 + h_1 - H_1) = D_1 + q_1 - Q_C, \quad (22)$$

$$V_2(G_1 - 1) + V_1(1 - J_1) + L_1 = 0, \quad (23)$$

where  $V_1$  and  $Q_C$  are specified :

$$V_1 = V_1^0(t) , Q_C = Q_C^0(t) . \quad (24)$$

The basic equations around the reboiler are ( Fig. 3 )

$$dx_{i,N}/dt = \{ - V_N(y_{i,N} - x_{i,N}) + L_{N-1}(x_{i,N-1} - x_{i,N}) \} / R_N , \quad (25)$$

$$L_{N-1}(B_N + h_N - h_{N-1}) - V_N(C_N + h_N - H_N) = D_N + q_N + Q_R , \quad (26)$$

$$V_N(1 - J_N) + L_{N-1}(I_N - 1) + L_N = 0 . \quad (27)$$

There are 2N material and energy balance equations ( Eq.(8), Eq.(16), Eq.(22), Eq.(23), Eq.(26) and Eq.(27) ) and 2N variables (  $L_1, \dots, L_N, V_2, \dots, V_N, Q_R$  ). Hence, all the phase flows and the reboiler load can be calculated as functions of  $x_{i,j}$ .

### 2.3 Computational Procedure of $\partial T_j / \partial x_{i,j}$

The stage equilibrium is expressed by

$$y_{i,j} = K_{eqi,j} x_{i,j} . \quad (28)$$

This equation is subject to the condition that the sum of all the component mole fractions be equal to unity :

$$\sum_{i=1}^m y_{i,j} = \sum_{i=1}^m K_{eqi,j} x_{i,j} = 1 . \quad (29)$$

We define  $Z_j$  by

$$Z_j = \sum_{i=1}^m K_{eqi,j} x_{i,j} = Z_j( T_j, x_{1,j}, \dots, x_{m,j} ), \quad (30)$$

where  $T_j$  is a function of  $x_{i,j}$ .

Then,

$$\begin{aligned}
 dz_j &= \sum_{i=1}^m (\partial Z_j / \partial x_{i,j}) dx_{i,j} + (\partial Z_j / \partial T_j) \sum_{i=1}^m (\partial T_j / \partial x_{i,j}) dx_{i,j} \\
 &= \sum_{i=1}^m \{ \partial Z_j / \partial x_{i,j} + (\partial Z_j / \partial T_j) (\partial T_j / \partial x_{i,j}) \} dx_{i,j} \\
 &= 0 .
 \end{aligned} \tag{31}$$

Accordingly,

$$\partial Z_j / \partial x_{i,j} = - (\partial Z_j / \partial T_j) (\partial T_j / \partial x_{i,j}) . \tag{32}$$

Since the sum of the liquid mole fractions must equal unity, only  $m-1$  of the compositions are independent. Therefore, Eq.(30) should be written by

$$Z_j = \sum_{i=1}^{m-1} K_{eqi,j} x_{i,j} + K_{eqm,j} (1 - \sum_{i=1}^{m-1} x_{i,j}) = 1 . \tag{33}$$

Then,

$$\begin{aligned}
 \partial Z_j / \partial x_{i,j} &= K_{eqi,j} - K_{eqm,j} + \sum_{k=1}^{m-1} x_{k,j} (\partial K_{eqk,j} / \partial x_{i,j} - \partial K_{eqm,j} / \partial x_{i,j}) \\
 &\quad + \partial K_{eqm,j} / \partial x_{i,j} , \quad (i=1, \dots, m-1) ,
 \end{aligned} \tag{34}$$

$$\partial Z_j / \partial T_j = \sum_{i=1}^{m-1} x_{i,j} (\partial K_{eqi,j} / \partial T_j - \partial K_{eqm,j} / \partial T_j) + \partial K_{eqm,j} / \partial T_j . \tag{35}$$

It has been shown that  $T_j / x_{i,j}$  can be calculated from Eq.(32), Eq.(33) and Eq.(34).

Because of nonideality of the hydrogen isotope solutions,  $K_{eqi,j}$  is a function of  $T_j$  and  $x_{i,j}$ . Souers<sup>(3)</sup> proposed an expression for the vapor-liquid equilibrium ratio accounting for nonideality :

$$K_{eqk,j} = (p_{k,j}^{\circ}/P_j) \exp(\Delta G_{k,j}^{\bar{E}}/R_g/T_j) \exp\{B_{k,j}^* (p_{k,j}^{\circ} - P_j)/R_g/T_j\}, \quad (36)$$

$$\Delta G_{k,j}^{\bar{E}} = \sum_{l=1}^m x_{l,j} A_{k-1,j}^* - (1/2) \sum_{l=1}^m \sum_{n=1}^m x_{l,j} x_{n,j} A_{l-n,j}^*, \quad (37)$$

$$A_{l-n,j}^* = A_{n-1,j}^* = A_{n-1}^0 T_j^{-1.33}, \quad A_{n-n,j}^* = 0, \quad (38)$$

$$B_{k,j}^* = B_{kT_j}^{0b}. \quad (39)$$

From Eq.(37),

$$\begin{aligned} \Delta G_{k,j}^{\bar{E}} &= \sum_{l=1}^{m-1} x_{l,j} A_{k-1,j}^* + (1 - \sum_{l=1}^{m-1} x_{l,j}) A_{k-m,j}^* \\ &\quad - (1/2) \sum_{l=1}^{m-1} \sum_{n=1}^{m-1} x_{l,j} x_{n,j} A_{l-n,j}^* - \sum_{l=1}^{m-1} x_{l,j} (1 - \sum_{n=1}^{m-1} x_{n,j}) A_{l-m,j}^*. \end{aligned} \quad (40)$$

Hence,

$$\partial \Delta G_{k,j}^{\bar{E}} / \partial x_{i,j} = A_{k-i,j}^* - A_{k-m,j}^* + \sum_{n=1}^m x_{n,j} (A_{n-m,j}^* - A_{n-i,j}^*). \quad (41)$$

Accordingly, the derivative of  $K_{eqk,j}$  can be calculated from

$$\partial K_{eqk,j} / \partial x_{i,j} = K_{eqk,j} (\partial \Delta G_{k,j}^{\bar{E}} / \partial x_{i,j}) / R_g/T_j. \quad (42)$$

From Eq.(36),

$$\begin{aligned} \partial K_{eqi,j} / \partial T_j &= [ \ln p_{i,j}^{\circ} / dT_j + (\partial \Delta G_{i,j}^{\bar{E}} / \partial T_j - \Delta G_{i,j}^{\bar{E}} / T_j) / R_g/T_j \\ &\quad + \{ dB_{i,j}^* / dT_j + B_{i,j}^* (dp_{i,j}^{\circ} / dT_j) / (p_{i,j}^{\circ} - P_j) \\ &\quad - B_{i,j}^* / T_j \} (p_{i,j}^{\circ} - P_j) / R_g/T_j ] K_{eqi,j}. \end{aligned} \quad (43)$$

## 2.4 Summary

The key equations are Eq.(3), Eq.(21) and Eq.(25). These  $mN$ -dimensional ordinary differential equations are solved by application of a numerical analysis technique ( e.g. Improved Euler Method ). All the other variables, such as phase flows, liquid holdups and vapor compositions, are calculated from the other basic equations, because they are given as functions of liquid compositions.

## 3. Conclusions

Basic equations are derived for rigorous dynamic simulation study of a cryogenic distillation column. The model accounts for differences in latent heat of vaporization among the six isotopic species of molecular hydrogen, decay heat of tritium, heat transfer through the column wall and nonideality of the solutions. Provision is made for simulation of a column with multiple feeds and multiple sidestreams.

The author is now ready to perform rigorous dynamic simulation study. The attention will be focused on effects of the above factors on dynamic behavior of a cryogenic distillation column for hydrogen isotope separation.

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## NOMENCLATURE

- $A_{k-i,j}^*$  = Binary-mixing parameter for isotopes k and i at  $T_j$   
 (cal/g-mol)
- $B_{i,j}^*$  = 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)
- $\Delta G_{i,j}^E$  = Partial molal-excess Gibbs free energy of mixing (cal/g-mol)
- $H_j$  = Molal enthalpy of vapor stream leaving j-th stage (cal/g-mol)
- $H_{Fj}$  = Total enthalpy of feed stream supplied to j-th stage (cal/h)
- $h_j$  = Molal enthalpy of liquid stream leaving j-th stage (cal/g-mol)
- $h_{i,j}^o$  = Molal enthalpy of component i at  $T_j$  (cal/g-mol)
- $K_{eqi,j}$  = Vapor-liquid equilibrium ratio of component i at  $T_j$
- $L_j$  = Flow rate of 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)
- $p_{i,j}^o$  = Vapor pressure of component i at  $T_j$  (atm)
- $Q_C$  = Condenser load (cal/h)
- $Q_R$  = Reboiler load (cal/h)
- $Q_j$  = 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)
- $V_j$  = Flow rate of vapor stream leaving j-th stage (g-mol/h)



$v_{i,j}$  = Molal density of liquid of component  $i$  at  $T_j$  (g-mol/cm<sup>3</sup>)

$V_j^*$  = Volume holdup of liquid in  $j$ -th stage (cm<sup>3</sup>)

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

$x_{i,j}$  = Mole fraction of component  $i$  in liquid stream leaving  
 $j$ -th stage

$y_{i,j}$  = Mole fraction of component  $i$  in vapor stream leaving  
 $j$ -th stage

$z_{Fi,j}$  = Mole fraction of component  $i$  in feed stream supplied  
to  $j$ -th stage

$\lambda_i$  = 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),

$\Lambda$  = Decay heat of tritium (cal/h/g).

(Subscript)

$j$  : Stage number

$i$  : Component number, 1 : H<sub>2</sub>, 2 : HD, 3 : HT, 4 : D<sub>2</sub>, 5 : DT,

6 : T<sub>2</sub>

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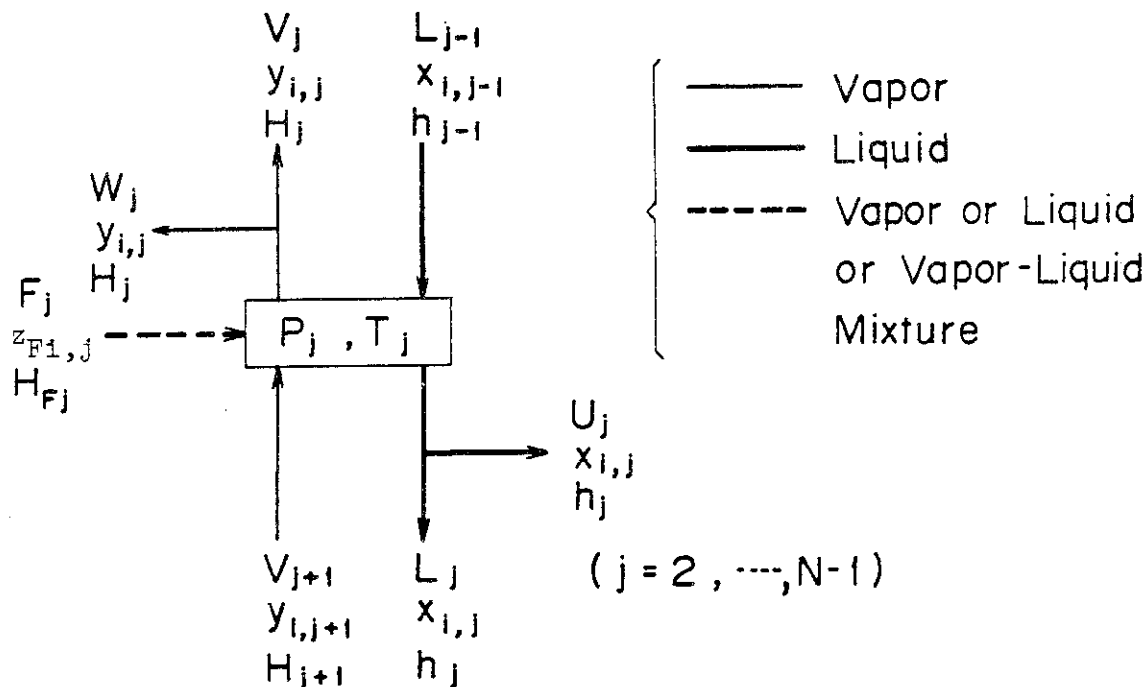


Fig. 1 Model Stage for Mathematical Simulation

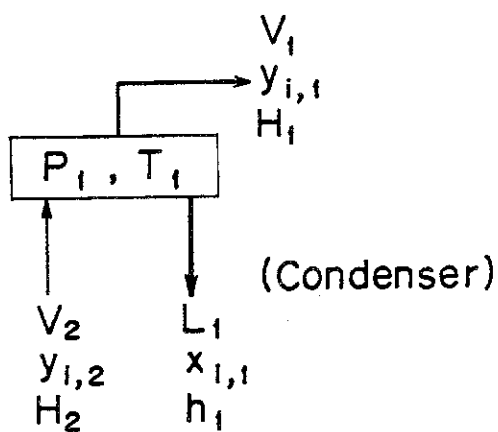


Fig. 2 Model Condenser for Mathematical Simulation

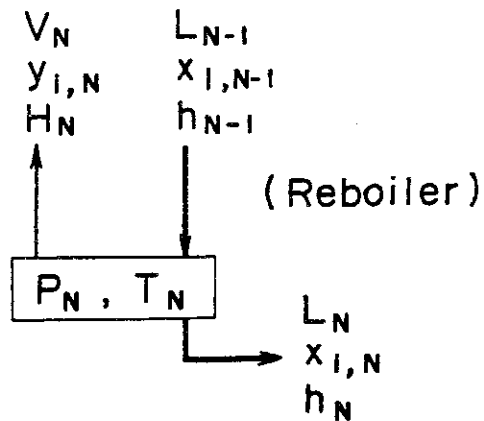


Fig. 3 Model Reboiler for Mathematical Simulation