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DERIVATION OF WORKING EQUATIONS FOR DYNAMIC SIMULATION OF CRYOGENIC DISTILLATION COLUMN INCORPORATING VAPOR HOLDUPS ON STAGES WITHIN COLUMN

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(Received December 20, 1982)

If a distillation column has a packed section whose inner diameter is considerably small, a small number of the total theoretical stages, and a condenser whose volume is very large, the vapor holdup in the condenser could be significant in dynamic simulation of the column. This problem is often posed for an experimental cryogenic distillation column used at a laboratory.

In the present report, working equations are derived incorporating the vapor holdups on stages within the column. The simulation procedure as well as the working equations are far more complex than in cases where the vapor holdups are neglected. The present report shows how the numerical integration techniques such as the Modified Euler method and the Runge-Kutta-Gill method can be applied to the simulation.

KEYWORDS : Distillation Column, Cryogenic Distillation, Packed Section, Total Theoretical Stages, Condenser, Vapor Holdup, Dynamic Simulation, Numerical Integration

蒸気ホールドアップを考慮した深冷蒸留塔の動特性解析のための基本式の導出

日本原子力研究所東海研究所核融合研究部
木下　正弘

（1982年12月20日受理）

蒸留塔の充填部の内径が小さく，充填高さも比較的低く，それでいてコンデンサーの容積がかなり大きい場合には，塔の動特性解析において，コンデンサー内の蒸気ホールドアップが重要となり得る。このような問題は，実験室で基礎研究に用いる深冷蒸留塔でよく見受けられる。

本報では，各段における蒸気ホールドアップを考慮した動特性解析のための基本式の導出が行われている。基本式及び解析手順は，蒸気ホールドアップを無視した場合に比べるとはるかに複雑となる。修正オイラー法やルンゲ・クッタ・ギル法などの手法がどのようにして適用できるかが示されている。

CONTENT

1. INTRODUCTION 1
2. WORKING EQUATIONS 1
3. CALCULATIONAL PROCEDURE OF $\partial T_j / \partial x_{i,j}$ 5
4. CONCLUSION 7
NOMENCLATURE 7
ACKNOWLEDGMENT 8
REFERENCES 9

目　　次

1. 序　論 1
2. 基本式 1
3. $\partial T_j / \partial x_{i,j}$ の計算手順 5
4. 結　論 7
記号表 7
謝　辞 8
参考文献 9

1. INTRODUCTION

In dynamic simulation of a distillation column, the vapor holdups are neglected in most cases. This neglection seems quite reasonable, because liquid holdups are considerably more than vapor holdups under usual conditions. Furthermore, the neglection eliminates the mathematical complication caused by presence of $dy_{i,j}/dt$. However, if the column has a packed section whose inner diameter is considerably small, a relatively small number of the total theoretical stages and a condenser whose volume is very large, the vapor holdup in the condenser could be significant in the dymanic behavior of the column.

The author et al. have been engaged in a preliminary experimental study using a cryogenic distillation column which separates N_2 and Ar.[1][2] The experimental column has only ~ 10 theoretical stages, and the vapor holdup in the condenser is approximately half as much as the liquid holdup in the condenser.

The purpose of the present study is to derive the working equations for dynamic simulation work incorporating vapor holdups on stages within the column.

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For simplicity, the heat balances are neglected in the present model. Additionally, the molal holdups are assumed to remain constant along the time trajectory. The model column for

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mathematical simulation is illustrated in Fig. 1.

The component material balances are expressed by

$$H_{L1}(dx_{i,1}/dt) + H_{V1}(dy_{i,1}/dt) = V_2 y_{i,2} - V_1 y_{i,1} - L_1 x_{i,1} , \quad (1)$$

$$H_{Lj}(dx_{i,j}/dt) + H_{Vj}(dy_{i,j}/dt) = F_j z_{Fi,j} + L_{j-1} x_{i,j-1} + V_{j+1} y_{i,j+1} - (L_j + U_j) x_{i,j} - (V_j + W_j) y_{i,j} , \quad (2)$$

$$H_{LN}(dx_{i,N}/dt) + H_{VN}(dy_{i,N}/dt) = F_N z_{Fi,N} + L_{N-1} x_{i,N-1} - V_N y_{i,N} - L_N x_{i,N} . \quad (3)$$

A mathematical complication is caused by the presence of $dy_{i,j}/dt$. By using the vapor-liquid equilibrium expressed by

$$y_{i,j} = K_{i,j} x_{i,j} , \quad (4)$$

$$dy_{i,j}/dt = K_{i,j}(dx_{i,j}/dt) + (dK_{i,j}/dt) x_{i,j} . \quad (5)$$

Since $K_{i,j}$ can be considered as a function of $x_{1,j}$, ..., $x_{m-1,j}$ and T_j,

$$dK_{i,j}/dt = \sum_{k=1}^{m-1} (\partial K_{i,j}/\partial x_{k,j})(dx_{k,j}/dt) + (\partial K_{i,j}/\partial T_j)(dT_j/dt). \quad (6)$$

The temperature T_j is a function of $x_{1,j}$, ..., $x_{m-1,j}$, so we obtain

$$dT_j/dt = \sum_{k=1}^{m-1} (\partial T_j/\partial x_{k,j})(dx_{k,j}/dt) \ . \tag{7}$$

Substituting Eq.(7) in Eq.(6) yields

$$dK_{i,j}/dt = \sum_{k=1}^{m-1} \{\partial K_{i,j}/\partial x_{k,j} + (\partial K_{i,j}/\partial T_j)(\partial T_j/\partial x_{k,j})\} \cdot (dx_{k,j}/dt) \ . \tag{8}$$

From Eqs.(5) and (8), we obtain

$$dy_{i,j}/dt = K_{i,j}(dx_{i,j}/dt) + x_{i,j} \sum_{k=1}^{m-1} \eta_{i,k,j}(dx_{k,j}/dt) \ , \tag{9}$$

where

$$\eta_{i,k,j} = \partial K_{i,j}/\partial x_{k,j} + (\partial K_{i,j}/\partial T_j)(\partial T_j/\partial x_{k,j}) \ . \tag{10}$$

Substituting Eq.(9) in Eq.(2) yields

$$(H_{Lj} + K_{i,j}H_{Vj})(dx_{i,j}/dt) + x_{i,j}H_{Vj} \sum_{k=1}^{m-1} \eta_{i,k,j}(dx_{k,j}/dt)$$
$$= F_j z_{Fi,j} + L_{j-1}x_{i,j-1} + V_{j+1}y_{i,j+1} - (L_j + U_j)x_{i,j}$$
$$- (V_j + W_j)y_{i,j} \ . \tag{11}$$

Similar equations are obtained by substituting Eq.(9) in Eq.(1) and in Eq.(3). These equations can be written by the following matrix equations :

$$\begin{Vmatrix} A_{11,j} & \cdots & A_{1m,j} \\ \vdots & & \vdots \\ A_{m1,j} & \cdots & A_{mm,j} \end{Vmatrix} \begin{Vmatrix} dx_{1,j}/dt \\ \vdots \\ dx_{m,j}/dt \end{Vmatrix} = \begin{Vmatrix} D_{1,j} \\ \vdots \\ D_{m,j} \end{Vmatrix} , \tag{12}$$

where

$$A_{kk,j} = H_{Lj} + K_{k,j}H_{Vj} + x_{k,j}H_{Vj}\eta_{k,k,j} \ , \ (k = 1, \ldots, m-1),$$

$$A_{kn,j} = x_{k,j}H_{Vj}\eta_{k,n,j} \ , \ (n = 1, \ldots, m-1), (k = 1, \ldots, m-1),$$

$$A_{km,j} = 0 \ , \ (k = 1, \ldots, m-1),$$

$$A_{mk,j} = x_{m,j}H_{Vj}\eta_{m,k,j} \ , \ (k = 1, \ldots, m-1),$$

$$A_{mm,j} = H_{Lj} + K_{m,j}H_{Vj} \ , \tag{13}$$

and

$$D_{i,1} = V_2 y_{i,2} - V_1 y_{i,1} - L_1 x_{i,1} \ ,$$

$$D_{i,j} = F_j z_{Fi,j} + L_{j-1}x_{i,j-1} + V_{j+1}y_{i,j+1} - (L_j + U_j)x_{i,j}$$

$$- (V_j + W_j)y_{i,j} \ , \ (j = 2, \ldots N-1),$$

$$D_{i,N} = F_N z_{Fi,N} + L_{N-1}x_{i,N-1} - V_N y_{i,N} - L_N x_{i,N} \ . \tag{14}$$

By solving Eq.(12), $dx_{i,j}/dt$ can be calculated : $dx_{i,j}/dt$ can be expressed by

$$dx_{i,j}/dt = f_{i,j}(t, x_{1,j}, \ldots, x_{m,j}), \ (i = 1, \ldots, m),$$

$$(j = 1, \ldots, N). \tag{15}$$

Therefore, a numerical integration technique (e.g. Modified Euler method and Runge-Kutta-Gill method) can directly be applied to Eq.(15).

If the heat balances and the assumption of constant volume holdup specification need to be incorporated, some additional considerations are required.[3]

3. CALCULATIONAL PROCEDURE OF $\partial T_j/\partial x_{i,j}$

Equation (4) is subject to the condition that the sum of all the vapor mole fractions be equal to unity :

$$\sum_{i=1}^{m} K_{i,j} x_{i,j} = 1 \ . \tag{16}$$

We define Z_j by

$$Z_j = \sum_{i=1}^{m} K_{i,j} x_{i,j} = Z_j(T_j, \ x_{1,j}, \ \cdots, \ x_{m-1,j}) \ . \tag{17}$$

Since T_j is a function of $x_{i,j}$ ($i = 1, \ \cdots, \ m-1$),

$$\begin{aligned} dZ_j &= \sum_{i=1}^{m-1} (\partial Z_j/\partial x_{i,j}) dx_{i,j} + (\partial Z_j/\partial T_j) \sum_{i=1}^{m-1} (\partial T_j/\partial x_{i,j}) dx_{i,j} \\ &= \sum_{i=1}^{m-1} \{\partial Z_j/\partial x_{i,j} + (\partial Z_j/\partial T_j)(\partial T_j/\partial x_{i,j})\} dx_{i,j} \\ &= 0 \ . \end{aligned} \tag{18}$$

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Since $x_{i,j}$'s (i = 1, ..., m-1) are the independent variables, we obtain

$$\partial Z_j/\partial x_{i,j} = - (\partial Z_j/\partial T_j)(\partial T_j/\partial x_{i,j}) \ . \tag{19}$$

Since only m-1 of the liquid mole fractions are independent (the sum of the liquid mole fractions must be equal to unity), Eq.(17) should be written by

$$Z_j = \sum_{i=1}^{m-1} K_{i,j}x_{i,j} + K_{m,j}(1 - \sum_{i=1}^{m-1} x_{i,j}) = 1 \ . \tag{20}$$

Then,

$$\partial Z_j/\partial x_{i,j} = K_{i,j} - K_{m,j} + \sum_{k=1}^{m-1} x_{k,j}(\partial K_{k,j}/\partial x_{i,j} - \partial K_{m,j}/\partial x_{i,j}) + \partial K_{m,j}/\partial x_{i,j} \ , \quad (i = 1, \ldots, m-1), \tag{21}$$

$$\partial Z_j/\partial T_j = \sum_{i=1}^{m-1} x_{i,j}(\partial K_{i,j}/\partial T_j - \partial K_{m,j}/\partial T_j) + \partial K_{m,j}/\partial T_j \ . \tag{22}$$

It has been shown that $\partial T_j/\partial x_{i,j}$ can be calculated from Eqs.(19), (21) and (22).

4. CONCLUSION

Working equations have been derived for dynamic simulation of a cryogenic distillation column incorporating vapor holdups on stages within the column. The present report shows how the numerical integration techniques such as the Modified Euler method and the Runge-Kutta-Gill method can be applied to the simulation.

If the column has a packed section whose inner diameter is considerably small, a relatively small number of the total theoretical stages, and a condenser whose volume is very large, the vapor holdup in the condenser could be significant in analysis of the dynamic column behavior. In these cases, the effect of the vapor holdup in the condenser needs to be studied in further work.

NOMENCLATURE

F_j = flow rate of feed stream supplied to j-th stage (g-mol/h)

H_{Lj} = liquid holdup on j-th stage (g-mol)

H_{Vj} = vapor holdup on j-th stage (g-mol)

$K_{i,j}$ = vapor-liquid equilibrium ratio for component i on j-th stage

L_j = flow rate of liquid stream leaving j-th stage (g-mol/h)

m = total number of components

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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

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)

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

(Subscript)

i : component index

j : stage index

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The author wishes to express his sincere thanks to Mr. Yuji Naruse and Mr. Toshihiko Yamanishi for their valuable comments. Acknowledgment is also due to Dr. Yukio Obata and Dr. Kichizo Tanaka for their continuous encouragements.

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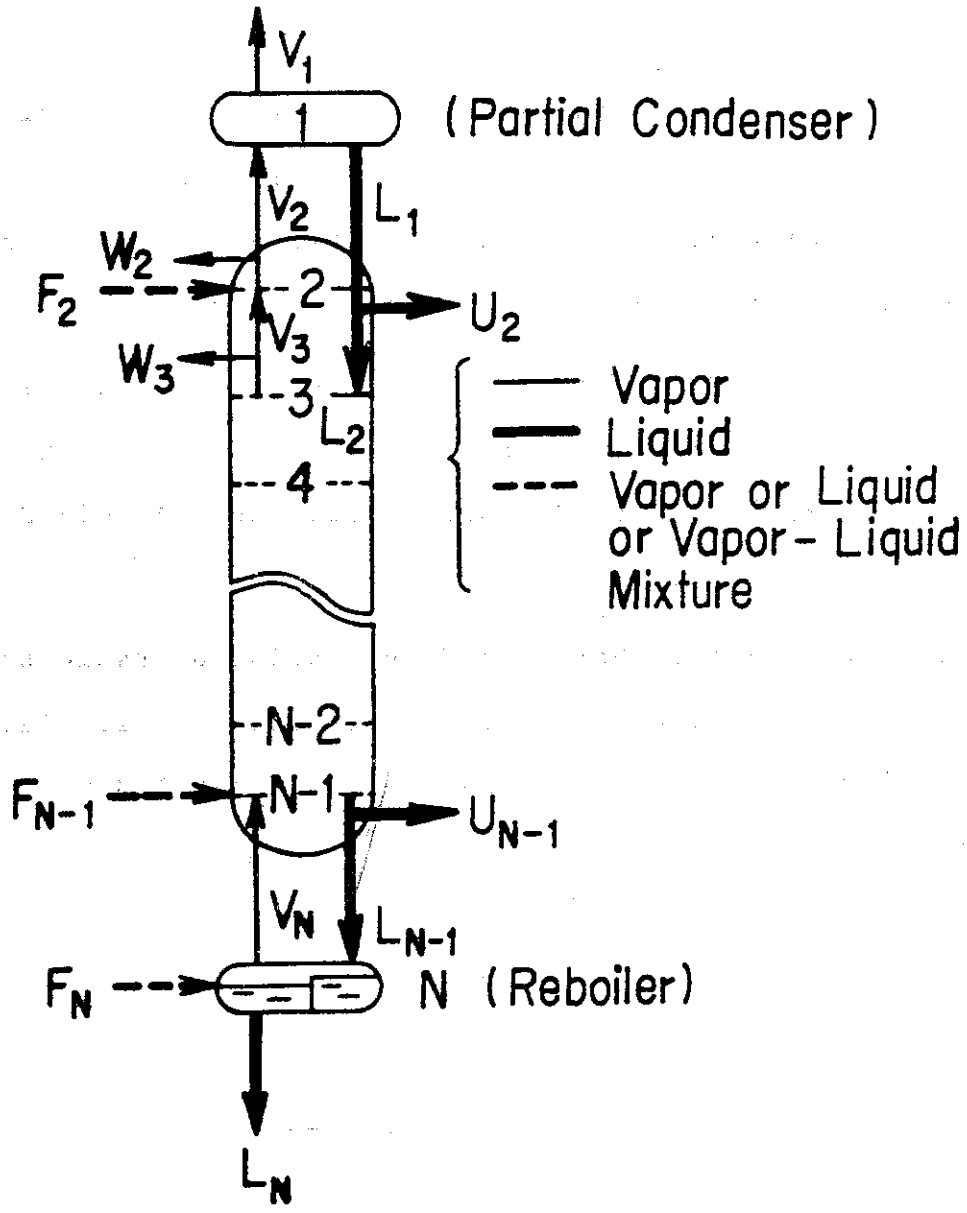

Fig. 1 Model Column for Mathematical Modeling